The Development of Rubber Forming
as a Rapid Thermoforming Technique for Continuous Fibre Reinforced Thermoplastic Composites

- Quality control by process control -

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Dit proefschrift is goedgekeurd door de promotoren:
Prof.dr.ir. Th. de Jong
Prof.dr.ir. R. Marissen

De promotie commissie bestaat uit:
Prof.drs. J. Bussink
Prof.dr.ir. I. Verpoest
Prof. P.C. Powell
Prof.ir. L.B. Vogelesang
Ir. A. Beukers

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Aan Monique
## CONTENTS:

List of Symbols.

1. **Introduction**................................................................................................................. 1  
   1.1 Background of the current investigations.  ................................................................. 2  
   1.2 Goal and perspective. ................................................................................................. 6  
   1.3 Thesis overview. ......................................................................................................... 7  

2. **Deformation of fabric reinforced thermoplastics**........................................................................... 9  
   2.1 Introduction. ....................................................................................................................... 9  
   2.2 Deformation modes of fabric reinforcement. ................................................................. 10  
      2.2.1 Overview ...................................................................................................................... 10  
      2.2.2 Interply slipping, geometrical description ............................................................... 15  
      2.2.3 Intraply shearing, geometrical description .............................................................. 20  
   2.3 Interply slipping of thermoplastic fabric reinforced laminates. ....................................... 24  
      2.3.1 Interply slip modelling, previous work ................................................................... 24  
      2.3.2 Experimental verification ....................................................................................... 32  
      2.3.3 A modified interply slip model ............................................................................. 40  
   2.4 Intraply shearing of fabric reinforced thermoplastics. .................................................... 53  
      2.4.1 Theory of intraply shearing forces ......................................................................... 53  
      2.4.2 Intraply shearing of dry fabric reinforcement .......................................................... 57  
      2.4.3 Influence of thermoplastic resin ............................................................................ 62  
   2.5 Optimal processing conditions for CFRTP product shaping. ............................................ 69  

3. **The technical development of rubber forming**......................................................................... 71  
   3.1 Introduction. ..................................................................................................................... 71  
   3.2 Overview of current thermoforming techniques. ........................................................... 72  
      3.2.1 Heating methods ......................................................................................................... 72  
      3.2.2 Major thermoforming processes ............................................................................. 73  
   3.3 The rubber forming technique. ....................................................................................... 76  
      3.3.1 Description of the rubber forming setup ................................................................... 76  
      3.3.2 Consolidation features .............................................................................................. 78  
      3.3.3 Forming features ....................................................................................................... 82  
      3.3.4 Additional remarks ................................................................................................... 87  
   3.4 Automation of the rubber forming technique. ................................................................. 89
4. Optimisation of rubber forming process parameters

4.1 Introduction.
4.2 Optimisation of heat flow during rubber forming.
   4.2.1 Theory of the heat transfer in laminates
   4.2.2 Setup of the heat transfer experiments
   4.2.3 Results of the heat transfer experiments
   4.2.4 Preliminary conclusions of the heat transfer experiments
4.3 Optimisation of the pressurization.
   4.3.1 Theory of the pressurization by rubber moulds
   4.3.2 Setup of pressure experiments with U-shaped moulds
   4.3.3 Results of the pressure experiments
   4.3.4 Preliminary conclusions of the pressure experiments
4.4 The application of external forming forces.
   4.4.1 Prevention of interply slip wrinkling
   4.4.2 Prevention of intraply shear wrinkling
   4.4.3 Technical realization of forming forces
4.5 General discussion.
4.6 Conclusions.

5. Material response to rubber forming.

5.1 Introduction.
5.2 The influence of non-isothermal consolidation.
   5.2.1 Previously published work on consolidation
   5.2.2 Setup of the consolidation experiments
   5.2.3 Results and discussion
   5.2.4 Superposition of non-isothermal consolidation increments
5.3 The influence of sudden cooling.
   5.3.1 Microcracking of the matrix layers
   5.3.2 Thermally induced distortion of the product geometry
5.4 The influence of forming forces.
   5.4.1 Setup of the forming force experiments
   5.4.2 Test results
   5.4.3 Discussion
   5.4.4 Conclusions of the forming force experiments
5.5 Concluding remarks.

6. Quality of the rubber forming technique.

6.1 Introduction.
6.2 Fabrication and testing of real-size CFRTP beams.
   6.2.1 Production of the U-shaped beams
   6.2.2 Determination of material properties by coupon testing
   6.2.3 Determination of material stiffness by full-scale testing
6.3 The influence of rubber forming on the stiffness properties.

6.3.1 Effect of a rubber forming cycle on the final product quality

6.3.2 Determination of the required starting material

6.3.3 Introduction of the production-factor concept

6.4 Quality control by process control.

7. Concluding remarks

7.1 Conclusions.

7.2 Recommendations for future work.

References.

Acknowledgement.

Summary.

Samenvatting.

About the author.

Appendix A: Calculation of the boundary conditions in case of radiation heating.

Appendix B: Numerical calculation of the heat transfer through thin materials.

Appendix C1: Material data of the used CFRTP materials.

Appendix C2: Overview of rubber materials, used for rubber forming.

Appendix C3: Examples of thermoplastic composites components.

Appendix D: Theoretical determination of the modulus of elasticity of rubber.

Appendix E: Design of rubber stamps.
List of symbols.

A \quad \text{lamine surface}

a \quad \text{acceleration, height of a rectangular element}

b \quad \text{width of a rectangular element}

C \quad \text{constant}

C_p \quad \text{heat capacity}

D \quad \text{tip load}

D_{au} \quad \text{degree of autohesion}

D_{ic} \quad \text{degree of intimate contact}

d_i \quad \text{thickness of a layer}

E \quad \text{modulus of elasticity, activation energy}

E_o \quad \text{Young’s modulus}

E_c \quad \text{apparent compression modulus}

E_{oo} \quad \text{bulk compression modulus}

F \quad \text{force}

f_p \quad \text{production factor}

G \quad \text{shear modulus}

g \quad \text{heat generation}

H \quad \text{force in horizontal direction}

h \quad \text{height, thickness of a fabric layer}

I \quad \text{moment of inertia}

K \quad \text{universal gas constant, constant}

k \quad \text{thermal conductivity, constant}

L \quad \text{yarn length, segment length, specimen length}

l \quad \text{length}

M \quad \text{bending moment}

m \quad \text{mass, constant}

n \quad \text{number of layers, constant}

P \quad \text{load}

p \quad \text{pressure}

R \quad \text{bending radius, resultant force}

S \quad \text{shape factor, bond strength, static moment}

s \quad \text{slip displacement}

T \quad \text{temperature}

T_g \quad \text{glass transition temperature}

t \quad \text{time, thickness}

V \quad \text{volume, absolute velocity}

v \quad \text{slip velocity, coordinate}
<table>
<thead>
<tr>
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<tr>
<td>$W$</td>
<td>weight</td>
</tr>
<tr>
<td>$w$</td>
<td>width, distance between two rectangular elements</td>
</tr>
<tr>
<td>$x, y, z$</td>
<td>coordinates</td>
</tr>
<tr>
<td>$x_m$</td>
<td>mass fraction</td>
</tr>
<tr>
<td>$x_v$</td>
<td>volume fraction</td>
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<tr>
<td>$\alpha$</td>
<td>bending angle, thermal diffusivity</td>
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<tr>
<td>$\gamma$</td>
<td>shear strain</td>
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<td>$\Delta$</td>
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<td>coordinate, mould angle, crossover angle</td>
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<td>viscosity</td>
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<tr>
<td>$\rho$</td>
<td>density, local curvature</td>
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<td>constant</td>
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<tr>
<td>$\phi$</td>
<td>crossover angle, bending angle</td>
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<tr>
<td>$\phi$</td>
<td>change of crossover angle (shearing angle)</td>
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CHAPTER 1. Introduction

From the early days of mankind, people have been trying to shape structural materials to achieve effective structures. Wood, stone and metals have been gradually entering society since men succeeded in their effort to form the materials into useful structural shapes. Through those times, the craftsmen held key-positions in society. Nowadays, in the "age of technology", experienced craftsmen are still indispensable: aircraft manufacturers for instance, are incapable of completely automating the forming processes of aircraft materials.

With the first application of composites in military aircraft, during the Second World War, a new material concept was born. The anisotropy of those fibrous materials opened the possibility to design structures with "custom" material properties. The number of applications of composites increased during several decades, while the composite materials themselves were developed further, resulting in high performance structural materials. Most of the applications were based on thermosetting resins, which permanently harden (set) when heated under pressure.

Few of the thermoset resins, though improved and toughened$^{1-2}$, had met the combined requirements of damage tolerance and hot/wet compression strength as they exist in aerospace industries. In the early eighties, thermoplastics were introduced to compete with the traditional thermoset resins used in the composite materials. Thermoplastics do not permanently harden, but can be reheated and plastically reformed numerous times. In addition to their higher impact resistance, the newly developed thermoplastic composites were expected to offer a reduction in manufacturing costs. One of the major advantages of thermoplastic composites was the easy, industrial, processability into final products by simply heating the material until the matrix is sufficiently soft to allow fibre rearrangements$^{3-4}$.

At present however, the shaping of continuous fibre reinforced thermoplastics is still a stumbling block in the application of these promising materials. Though several manufacturing methods, such as diaphragm forming and matched-metal-die forming are available$^{5-7}$, the techniques for producing continuous fibre reinforced thermoplastic products are still far away from their envisaged production performance. Meanwhile thermoplastic composites slowly enter sporting good designs, orthopaedic prostheses and prototype automotive applications. The number of potential applications of these materials is still increasing. But as pointed out before, efficiently forming of the material into a functional product is a problem that has to be solved before thermoplastic composites can be applied on a larger scale.
1.1 Background of the current investigations.

Though the use of thermoplastic composites is gradually rising, the application of these materials is mainly restricted to aircraft interior parts and a limited number of automotive products. In load-carrying aerospace structures, thermoplastic composites are still hard to find. The replacement of traditional metal parts by fibre reinforced thermoplastics was started in the late eighties with the introduction of composite wing skin panels and access panels in the fuselage. From the introduction of these new materials, however, the development of products has been an evolution, rather than the promised revolution. This has been particularly true for aerospace product development. Product performance has to be on a high level, to ensure efficient and reliable structures. Production companies cannot allow themselves dangerous experiments with new materials. Moreover, cost aspects play a major role.

Because aircraft structures have to fulfil extreme safety demands, the development of materials with better mechanical, chemical and thermal properties has received much attention from the beginning. Since it had to be demonstrated that new materials indeed had those desired better properties, much emphasis was put on material coupon testing. This development was stimulated by the need for new high-performance materials in military aircraft projects and space applications. So, qualification and certification of new materials seemed to become the most important research goal.

Unfortunately, the desired high thermal properties of thermoplastic matrices implied high processing temperatures for thermoplastic based fibre-reinforced composites and the hot shaping of thermoplastic laminates, the so-called thermoforming, required extreme processing environments. Moreover, the product manufacturers traditionally had no experience with the shaping of materials that do not have the capability to deform locally, like metals have. So the need for research, focused on the processing of continuous fibre reinforced thermoplastics (CFRTP), became evident.

The notion that more research should be directed to thermoforming techniques has come too late for a number of material manufacturers. The recent decay of military and civil aircraft projects combined with a global economic recession forced various material companies to stop their composite development activities. Some important examples of development, commercializing and casting off again of CFRTP materials are the BASF trade family of "Ultrason" PolyEtherSulfone (PES) and PolySulfon (PSU) composites and the "Polysulf" trade family of PolyCarbonate (PC) and PolyAmide (PA) composites from BAYER. Even the more successful products suffered extreme cutbacks in the years 1991/1992. Examples are "APC-2", a PolyEtherEtherKetone (PEEK) based composite of ICI and "Ryton" PolyPhenyleneSulfide (PPS) composite of Phillips Petroleum. Yet, a turn has already set in. The manufacturing and distribution of thermoplastic composite materials is being concentrated by fewer (often smaller) companies that are more specialized in composites. Although in the short term CFRTP composites will not be the bulk materials
as was expected before, these companies still foresee a promising future in the longer term. More experience is gained with the manufacturing of thermoplastic prepregs and consequently more combinations of thermoplastic resins and fibre reinforcements can be efficiently realized. In Appendix C3, a collection of a few possible components for CFRTP composites is given.

Recognition of the fact that the development of efficient processing techniques are the key to success of thermoplastic composites have significantly increased the research effort in this direction. The problems that arise while thermoforming two- and three- dimensionally shaped products are described more intensively in the open literature. Investigations on the deformation of fabric reinforced composites are reported, including the influence of the thermoplastic resin during the thermoforming processes\textsuperscript{13,14}. Concerning the use of stampable continuous fibre reinforced thermoplastic sheets in relatively fast production processes, important research work has been described recently by several authors. In their work, a tendency can be recognized from manually controlled heating and stamping of thermoplastic laminates towards the introduction of more automated processes and even continuous production lines\textsuperscript{15-17}.

![Fig 1.1 Examples of composite products that are thermoformed at the Structures and Materials Laboratory.](image-url)
Fig. 1.2 The CFRTP main-landing gear door at a Fokker 50.

With increasing experience in thermoforming methods, some "pioneers" have been able to realize interesting composite products and structures (Fig. 1.1). Examples in the aerospace industry are the main landing gear door and the ice protection plates on the Fokker 50 (Fig. 1.2)\textsuperscript{18,19}. The next step is replacing the materials of structural aircraft parts by CFRTP materials. Preconditions, however, are substantially better control and verification of the mechanical properties of the final product. In the automotive industry, examples of CFRTP prototype parts are composite bumper- and other bars, crash cones and stiffened plates (Fig. 1.3)\textsuperscript{20-22}. Marketing techniques control the market acceptance of these composite parts in the short term, but the market demand in the longer term is highly depending on the "value-for-money" criterion\textsuperscript{23}. In this criterion, the cost effectiveness of a structure must be as high as possible. It is defined by:

\[
\text{cost effectiveness} = \frac{\text{structure effectiveness}}{\text{acquisition costs} + \text{utilisation costs}}
\]

The structure effectiveness of a thermoplastic composite part can be increased by a better control of the thermoforming technique, resulting in a higher performance and reliability of the manufactured part which are an important part of the structure effectiveness. Another influence of the thermoforming method on the cost effectiveness is directly expressed in the acquisition costs of the structure. In the present work, it is shown that the forces to shape a CFRTP product are substantially lower than those necessary in for instance injection moulding or GMT processes (Fig. 1.4).
Introduction

Fig. 1.3 Automotive applications of thermoplastic composites, realized at the Structures and Materials Laboratory.

The reduction of required forces enable the use of low force, low energy machinery for thermoforming. This considerably lowers the machine- and tool costs and thus the acquisition costs for a specific product. Therefore, by optimisation of the thermoforming technique, the cost effectiveness of a composite structure is positively influenced by both an increase of the structure effectiveness and a decrease of the acquisition costs.

Fig. 1.4 Comparison of required machine forces for the production of PBT resin based products (GE Plastics).
It is important that the cost effectiveness of a thermoformed product is more favourable than that of the original non-composite part. Apart from new product designs adapted to the new materials, a design philosophy has to be created in which the thermoforming process is taken into account in an early stage of the design process. It is therefore important to research the possibilities and limitations of thermoforming techniques.

1.2 Goal and perspective.

The principal goal of the present work is twofold:

- the understanding of the fundamental deformation processes of continuous fibre reinforced thermoplastics during a manufacturing cycle of two- or three-dimensional products made from flat laminates;

- the development of an efficient thermoforming technique for manufacturing high quality thermoplastic composite products.

The realisation of the two parts is closely related. It is impossible to design an efficient manufacturing technique without thorough understanding of the material behaviour during the product manufacturing. At present, the implementation of CFRTP products is based primarily on trial and error because of the lack of alternatives. However, in the long term, trial and error is not an economic way to introduce a production technique. On the other hand, fundamental research is only viable if the results can be transferred to real applications. Therefore the technical development of a thermoforming technique is a priority in this work as well. The technique should incorporate the results of the more fundamental investigations of the present work. An ideal thermoforming technique would combine the quality requirements for composite parts for aerospace applications and the production speed for the large production series for automotive and other applications.

The successful transformation of a flat sheet of thermoplastic resin-impregnated fabric to a high quality, three-dimensional composite product requires an optimal execution of subsequent thermoforming steps. In a highly simplified model, the following steps can be identified: heating, forming, consolidating and cooling. In this report the "forming" of the composite laminate will be defined as the shaping of the hot material. It is characterized by the deformation of the resin impregnated fibre reinforcement. At the so-called "transition point", the forming phase stops and the consolidation phase starts. At that point, the laminate is completely shaped but has to be pressurized further. "Consolidation" is the subprocess where a void-free and homogeneously fibre reinforced material is produced by the complete fusion of fully impregnated layers. A shaped and consolidated thermoplastic product has to cool down to room temperature. During cooling down the material will "solidify".
Earlier work on these different subjects can be found in the literature. Whereas heating and cooling of thermoplastic laminates often is considered as a logical expansion of earlier research work on unreinforced polymers\textsuperscript{13}, most composite researchers are aware of the novelty of forming and consolidating thermoplastic composites. Modelling work on forming and consolidation therefore is done at various universities and institutes\textsuperscript{14-24,27}. At Delft University of Technology, Bersee is working on the modelling of isothermal consolidation\textsuperscript{28,29} and Bergsma is developing the computer model "Drape" to simulate the forming phenomenon of intraply shearing of reinforcing fabrics\textsuperscript{30,31}.

In the present work, previous work on forming and consolidation has been consulted and refined where possible in order to approach the "real-world" processes. For instance, an important feature of the newly developed thermoforming technique is high processing speed, which implies the need for dynamic experiments. In most of the experimental work presented here, the thermoplastic composites that are used are glass fabric reinforced PolyEthyleneTeraphthalate (DESTEX\textsuperscript{*} supplied by DSM, the Netherlands) and glass fabric- and carbon fabric reinforced PolyEtherImide (CETEX\textsuperscript{*} supplied by TenCate, the Netherlands). See Appendix C1 for an overview of material data of these materials.

The rapid thermoforming technique is called "rubber forming" in analogy with diaphragm forming and matched-metal-die forming techniques, between which it has found its place. Rubber moulds are used to solve the forming and/or consolidation problems that occur in matched-metal-die forming (inhomogeneous pressure distribution on the deforming composite material) and in diaphragm forming (no possibility of local pressurization of the laminate).

An important part of the research work is dedicated to the determination of the material response on the rubber forming technique. The lack of knowledge about final mechanical material properties has always been a major problem for the designer of CFRTP parts. Up to now the only way to obtain reliable strength and stiffness data has been the full-scale testing of the final product. When it is possible to map the influence of the processing history, including the influence of subprocesses during a thermoforming cycle, then it is possible to hand the designer of CFRTP parts an accurate "production-factor" that allows him to calculate the final product strength and stiffness.

The perspective of this thesis work is probably best described by: "quality control by process control." Particularly with respect to composite applications in aerospace, a number of obstacles have to be overcome before this statement will be a reliable one.

### 1.3 Thesis overview.

A large part of forming problems that occur in thermoforming attempts results from misunderstanding of the fundamental deformation behaviour of the material rather than
technical process imperfection. This report, therefore, starts in Chapter 2 with the
determination of the forming forces inside fabric reinforced thermoplastics that are
necessary to successfully thermoform CFRTP laminates. Models of the fundamental
deformation behaviour of fabric reinforced thermoplastics are described and verified.
Consequently, the most important subprocesses and process parameters are pointed out.
From this, the "ideal" processing environment for thermoforming in general is abstracted.
Chapter 3 presents the technical development of the rubber forming process, a
thermoforming method that has the potential of fulfilling the processing demands posed in
the preceding chapter. Technical details will be given, together with the description of the
realisation of CFRTP parts by rubber forming. The application boundaries of the rubber
forming technique are also clarified in Chapter 3.

The next chapter (Chapter 4) focuses on investigations into the influence of process
parameters (external forces, temperature and pressure) on the material subprocesses during
a thermoforming cycle. Models for heat flow and pressurization in thermoplastic laminates
are used and compared to process measurements.

In Chapters 5 and 6, the material response (mechanical properties) in a rubber forming
cycle is investigated. Explanations for the material response on a material scale are verified
in Chapter 5, whereas in Chapter 6 a rubber formed product as a whole is considered.
Comparisons between calculated and estimated product strength and stiffness are given.
"Production factors" are introduced with which a designer, aware of the material
subprocesses, can calculate the final product properties. Finally, in Chapter 7, conclusions
and future research recommendations are summarized.

The work described in this report is strongly focused on the achievement of high quality
CFRTP products, so on "real-world" problems. To prove the viability of rubber forming
of thermoplastic composites, directly related material phenomena are exposed. An
implication of the choice for this approach is that during this research, other scientifically
interesting problems with respect to the material behaviour could not be focused on. It is
hoped, however, that the present work can provide a solid base for further development, use
and optimization of thermoforming techniques.

2.1 Introduction.

Thermoplastic polymer sheets own their capability of transformation into relatively complex shapes to the thermoplastic nature of the material. The loss or reduction of molecular coherence that occurs when the material is heated above its glass transition temperature Tg and melt temperature Tm for amorphous and semi-crystalline thermoplastics, respectively, allows the use of small forming forces. The thermoplasticity of the material will cause the constitution of the new shape after cooling.

With unreinforced thermoplastics, the overall deformation of the material above Tg or Tm is determined by the interaction between external forming forces and the molecular resistance\textsuperscript{32,33}. The allowable deformation is almost unlimited because of the flow characteristics of unreinforced polymers.

When stiff reinforcing particles like short fibres are added to the thermoplastic material, the deformation behaviour will change. In general, the reinforcing particles must be uniformly distributed in the final product. This requires a more controlled polymer flow\textsuperscript{34} and thus a better knowledge of the processing conditions.

When continuous fibres are used as reinforcement the behaviour changes even more. The fibres control the shaping of the material to a high extent\textsuperscript{1,3,5,14,27}. The continuous reinforcement will restrict any major elongation in its own direction and because of its elastic behaviour any external forming force results in small, purely elastic deformations only. Therefore the shaping of a CFRTP material in practice cannot be realized by simple material elongation. The continuity of the fibres requires actual rearrangements of the reinforcement during the transformation of a sheet into a three-dimensional product. The rearrangement of the reinforcement by external forming forces often implies distortion of the original fibre direction(s). The necessary rearrangements have to be understood and controlled to a high extent, in order to:

- make a certain CFRTP product shape producible anyhow;
- control the direction of the final reinforcement and thus obtain specific mechanical properties.

The most important rearrangements of a fabric reinforced material are characterized in the following section. Because of the resemblance of fabric deformations in composite materials with fabric deformations in the well documented textile technology, connections will be made with earlier developed experiments and deformation theories for clothing material.

Though the "overall deformation" of CFRTP materials is not primarily ruled by the behaviour of the thermoplastic matrix, the rearrangement of the fibre reinforcement has to
be supported by polymer matrix flow. Therefore this flow has to be controlled also. In Sections 2.3 and 2.4 the influence of the thermoplastic matrix will be discussed on two important deformation modes, interply slipping and intraply shearing. A theory on this influence will be derived and an experimental verification will be presented.

In the last section of this chapter, conclusions will be given to point out the optimal thermoshaping conditions of fabric reinforced thermoplastics.

2.2 Deformation modes of fabric reinforcement.

2.2.1 Overview.

During the shaping of a fabric reinforced laminate, two main deformation phenomena can be distinguished. The total deformation of the laminate can be realized by intraply deformation, by interply deformation or by a combination of both.

CFRTP products generally are formed of laminates of more than one layer. During interply deformation, the fibre reinforcement of a layer in a laminate is rearranged relative to that of the adjacent layers. The only interply deformation described in this report is the interply slipping of the layers of a laminate:

- interply slipping of the fabric layers

During intraply deformation, the reinforcement is rearranged inside one layer of the laminate. This rearrangement can consist of:

- intraply shear (or trellis effect)
- straightening of the fibres
- elongation of the fibres
- transverse fibre flow (squeezing flow)

Interply slipping.

A thermoplastic composite laminate can be presented schematically as alternatively stacked fibre rich layers and resin rich layers. If in a thermoforming process the laminate is bent in one of its fibre directions, the fibre rich layers which are relatively inextensible will slip relative to each other (Fig. 2.1). During this slipping the resin rich layer must act as a lubricant.

Interply slip takes place as soon as one of the reinforcement layers has to follow a geometrical route different from that of its neighbouring layers. The internal adjustment of a thermoplastic laminate to bending deformation will be further described in Section 2.2.2, whereas the thermoplastic matrix interaction during slipping is discussed in 2.3.
**Intraply shearing (trellis effect).**

During thermoforming, tensile forces can be generated in the fabric in directions other than the principal directions of the fabric. They result in deformations as shown in Fig. 2.2. The relative change in fibre direction of the fabric will cause a change in the laminate geometry as a whole. The fabric will lengthen in the direction of the applied force and become shorter in the direction perpendicular to that force. This way the dimensions of a fabric reinforced laminate can be adjusted to a desired product geometry. However, the fibres will rotate only if the matrix allows such a rearrangement. In Section 2.2.3, the geometrical consequences of intraply shearing are stated. In 2.4, the forces necessary for intraply shearing are discussed, together with the influence of the thermoplastic matrix on the shearing deformation.

**Straightening of the fibres.**

If the fibres in the reinforcement layers are not fully stretched before the thermoforming process starts, any tensile force applied on the heated laminate will straighten the fibre bundles in force direction (Fig. 2.3). The straightening will need small forces only. A distinction must be made between two different straightening modes.
First, the straightening of not fully aligned fibre bundles takes place. This can be considered as a favourable correction process for the material. Although curved fibres give a high deformation potential to a reinforced laminate, misaligned fibre bundles will decrease the material properties if they are not entirely straightened during the thermoforming process. Straightening of intentionally curved fibre bundles is not considered a useful tool to realize a product shape.

A second mode of straightening of fibre bundles can take place in fabrics. Due to the nature of woven textiles, interlaced fibre bundles always have some degree of plane curvature, contrary to the fibre bundles in unidirectional reinforcement layers (Fig. 2.4). The reduction of the curvature of the fibre bundles in one direction of a fabric simultaneously increases the curvature of the fibres in the direction perpendicular to it. The mechanical properties of the composite material will rise in the direction of the straightening forming force, but a decline of mechanical properties in the perpendicular direction has to be expected.

---

Fig. 2.3 Straigthening of non-aligned fibre yarns.

Fig. 2.4 Straightening of woven yarns.
Elongation of the fibres.

Once the fibres are straightened, a further increase of the tensile force in fibre direction during thermoforming will elongate the fibres. Because of their high stiffness, the resulting deformation generally will be very small, even when high tensile stresses are created in the fibre bundles. Because of the Hookian behaviour of most fibres (glass, aramid, carbon fibres), the elongations will be purely elastic. This means that when the stressed fibres are frozen in in the cooling laminate after forming, the high fibre tensile stresses have to be counteracted by compressive matrix stresses. An advantage of the tensile stresses during thermoforming is that they can be useful to reduce thermal stresses in the product. Disadvantages of the creation of high tensile stresses during thermoforming, however, are that fibre breakage can occur during a forming cycle (Fig. 2.5) and that high residual stresses can lead to premature product failure or mismatches in the shape of the product. Elastic elongation of the fibre reinforcement is therefore not a deformation that can be used to shape composite products.

![Image of a CFRP product](image)

*Fig. 2.5 Local failure of the fabric reinforcement due to high forces during the thermoforming of a CFRTP product.*

Transverse fibre flow (squeezing flow).

Another cause of rearrangement of the fibre reinforcement can be the matrix itself. Due to pressure differences over the laminate, a matrix flow will be induced. This flow forces the fibre bundles to drift, either in fibre direction or sideways to a less pressurized spot in the laminate. Fig. 2.6 shows this phenomenon.
Fig. 2.6 Schematic view of transverse fibre flow (squeezing flow) due to pressure differences in the laminate.

Fig. 2.7 Thickness adjustment of a thermoplastic laminate due to squeezing flow.
Deformation of fabric reinforced thermoplastics

If it happens in a controlled way, transverse fibre flow can have a positive effect on the product quality. Relatively small mould flaws can be compensated by matrix and fibre motion, for instance (Fig. 2.7). If the motion of the fibre reinforcement takes place in an uncontrolled way, it can result in a disturbance of the orientation and distribution of the fibres. The material rearrangement enables the layers to make intimate contact and thus improves the consolidation of the laminate\textsuperscript{25,26}. During this process, the pressure differences on the thermoplastic matrix are small inside the laminate and fibre motion is more or less controlled. For the overall deformation of fabric reinforced laminates in a thermoforming process, fibre motion induced by differences in pressure in the matrix is of less importance, however.

A fabric reinforcement yields a better dimensional stability than a unidirectional fibre reinforcement. Therefore the control of fibre rearrangement due to matrix flow will be easier, in that case.

2.2.2 Interply slipping, geometrical consideration.

As has been stated before, a CFRTP laminate can schematically be presented as a stack of fibre rich layers alternated by thermoplastic resin rich layers (Fig. 2.8).

![Schematic presentation of the stacking of a laminate.](image)

In Fig. 2.9 is shown how a particular fibre bundle in a reinforcing fabric has to bend (several times) to meet the curvatures in the product. Because of its thickness, each layer has a different bending radius at a curvature. To obtain a stress-free product, the laminate has to adjust to the different bending radii by a deformation called interply slipping. During interply slipping the layers of the laminate slide relative to each other.

In the following geometrical description of interply slipping, some assumptions are made:

1. all fibre rich layers are originally flat and identical
2. all layers have constant thickness and length
3. the matrix layer is infinitely thin
4. there is no friction between the layers
5. the layers do not debond during forming

Fig. 2.9 Cross-section of a curved, laminated product.

Bending of an n-layered laminate (Fig. 2.10) will generate a relative displacement of the layers according to the following geometrical expression:

\[ s_{0-m} = \alpha \cdot (R + m \cdot d_l) - \alpha \cdot R = m \cdot \alpha \cdot d_l \]  \hspace{1cm} \text{(2.2.1)}

where \( s_{0-m} \) is the slip between layers 0 and m,
\( \alpha \) is the bending angle,
\( R \) is the bending radius of the mould,
\( d_l \) is the thickness of one fibre rich layer.

Fig. 2.10 Relative displacement (interply slip) of a bent n-layered laminate.
If more curvatures (k) are introduced in a product, the resulting slip becomes (Fig. 2.11):

\[ s_{\text{total}} = \sum s_{i,0-m} = m \cdot d_i \sum \alpha_i \quad \text{(for } i = 0 \text{ to } k) \]

where \( s_{\text{total}} \) is the total amount of slip in the product between layers 0 and k,
\( \alpha_i \) is the local bending angle,
\( m \) is the number of layers that is considered.

Note that the total amount of slip in a thermoplastic composite laminate does not depend on the radius of the mould.

\[ \alpha \] is the bending angle

\( s \) is the amount of slip

Fig. 2.11 Resulting slip displacement of a laminate to form a k-curved product.

Fig. 2.12 Possible blankholding points that do not obstruct the necessary interply slip.
The equations have two important consequences for the shaping of a laminate in a thermoforming process.

First, equation \(<2.2.2.2>\) implies that areas of a deformed laminate with the same resulting bending angle \(\Sigma \alpha_i\) have identical slip deformations. In Fig. 2.12, this means that the resulting slip \(s(p_i) - s(p_j)\) between two laminate spots \(p_i\) and \(p_j\) is zero, because the resulting bending angle \(\Sigma \alpha_i\) between \(p_i\) and \(p_j\) is zero. The laminate can be clamped at \(p_i\) and \(p_j\), without restricting any slip deformation of the material in between. It is obvious that the material must not be clamped between the two mould surfaces during thermoforming until the necessary interply slipping between sequential clamping points has completed.

A second important implication is that the rate of slip deformation depends on the rate of change of the direction of the laminate. So, from \(<2.2.2.1>\):

\[
\frac{ds_{0-m}}{dt} = \frac{d(m \cdot \alpha \cdot d_i)}{dt} = m \cdot d_i \cdot \alpha'\quad <2.2.2.3>
\]

where \(\alpha' = \frac{d\alpha}{dt}\) is the rate of change of bending angle.

In 2.3, it will be shown that the required forming forces on the laminate are primarily determined by the slip velocity \(v_s\) between two neighbouring layers. From \(<2.2.2.3>\), the slip velocity between two neighbouring layers is given by:

\[
v_s = (\frac{ds_{0-m}}{dt})_{m=1} = d_i \cdot \alpha'\quad <2.2.2.4>
\]

The forming forces, necessary to create interply slip deformation, apparently do not depend on the product shape but on the geometrical route in which that shape is realized during the forming process. As an example of this, the forming of a hat-shaped beam is outlined shortly.

Consider two options for the thermoforming of the beam:

**option a)** The beam is shaped by two matching dies that force the laminate directly in its final shape (Fig. 2.13). The corners are formed almost instantly when the punch comes down, creating a high rate of change \(\alpha'\) of bending angle and thus a high slip velocity \(v_s\). Moreover, when a particular piece of the laminate slides over the corners \(A1\) and \(A2\), it has to bend 90°. When it slides further into the mould, it is straightened again. The resulting up and down slipping can only be accomplished by sufficiently high forming forces.

**option b)** The laminate is shaped by a flexible medium (e.g. an air pressurized diaphragm) into a stiff mould. First the middle of the bottom of the mould is touched (Fig. 2.14). The corners are fully shaped only then when the
flexible medium has completely filled the mould, while in option a) the corners A1, A2, B1 and B2 are formed almost instantly. So, the rate of change of bending angle $\alpha'$ is lower in all of the four bending areas and the corners are more gradually realized, requiring much lower forming forces.

Fig. 2.13 Matched-die forming of a U-shaped product.

Fig. 2.14 Diaphragm forming of a U-shaped product.

Equation <2.2.2.2> is a simple but convenient tool to examine interply slipping in a geometrical sense. With regard to the assumptions made at the start of this section, it yields the possibility to quantify and compare the slipping deformation of thermoplastic laminates in a thermoforming process.
2.2.3 Intrapy shearing, geometrical description.

An important rearrangement of a fabric reinforcement is the relative change of the crossover angle of the warp and weft yarns of the fabric (Fig. 2.15). The changing of this angle in fabrics is called "shearing" or "trellis effect". Because of the fact that this shearing takes place inside every single layer in a laminate, the deformation is called "intrapy shearing". In general, the changing of the crossover angle is caused by a tensile force on the fabric in a direction that does not coincide with a fibre direction. Concerning the overall deformation of the laminate, the intrapy shearing deformation implies an elongation of the composite material in the force direction and a contraction in the direction perpendicular to it.

Fig. 2.15 Shearing or trellis effect of two interlaced yarns.

During the shaping process of a fabric reinforced thermoplastic laminate, tensile forces are applied on the reinforcing fibre bundles and shearing of the fabric will take place. Theoretically, the shearing can be assisted by compression forces in the plane of the fabric which results in a shortening of the material in that direction. However, the amount of compression forces that a fibre bundle, surrounded by a fluid matrix, can transfer is negligible. Therefore it is assumed in this section that intrapy shearing takes place due to tensile forces in the fabric only.

Intrapy shearing offers a high deformation capability to a fabric reinforced laminate. In the figure is shown how two interlacing fibre bundles out of an originally square fabric have sheared. The shearing will stop when the forming forces are not sufficiently high any more to overcome the increasing shearing resistance (2.4). Then the maximum shearing angle \( \phi_{\text{max}} \) will be reached. Assuming that the maximum shearing deformation has been reached all over the fabric surface, all fibre bundles in a flat fabric are straight (Fig. 2.16).

The shortening of the fabric piece that includes a rotating yarn with length \( L \) can be given by:

\[
\Delta l = L(\cos(\phi/2 - \psi/2) - \cos(\phi/2))
\]

<2.2.3.1>
and the reduction of width of the fabric piece:

\[ \Delta w = L(\sin(\phi/2) - \sin(\phi/2 - \varphi/2)) \] <2.2.3.2>

where \( L \) is the length of the rotating fibre bundle,
\( \phi \) is the original crossover angle,
\( \varphi \) is the change of crossover angle.

Fig. 2.16 Uniform shearing of a piece of fabric that include a yarn with length \( L \).

In the mostly used square fabrics, \( \phi = \pi/2 \), then

\[ \Delta l = L/2 \cdot \sqrt{2}(-1 + \cos(\varphi/2) + \sin(\varphi/2)) \] <2.2.3.3>

\[ \Delta w = L/2 \cdot \sqrt{2}(1 - \cos(\varphi/2) + \sin(\varphi/2)) \] <2.2.3.4>

Geometrically it can be shown that the reduction in width \( \Delta w \) of a sheared fabric is always larger than the increase in length \( \Delta l \) of the same fabric (\( \phi=\pi/2 \)) because

\[ \cos(\varphi/2) < 1 = (1-\cos(\varphi/2)) > (-1+\cos(\varphi/2)) = \Delta w > \Delta l \]

for all \( 0 < \varphi < \pi \).

This means that the deformation of a fabric reinforced composite sheet due to intraply shearing is always larger in the direction perpendicular to the tensile forming forces, than it is in the direction of those forces.

Another important feature of shearing of a fabric reinforced thermoplastic laminate is the change in thickness of the sheared material. Because a laminate, impregnated with resin, has to fulfil the constant volume requirement, the thickness \( t \) of the laminate must increase during the change of crossover angle.
\[ V = l \cdot w \cdot t = \text{constant} \rightarrow (l + \Delta l) \cdot (w - \Delta w) \cdot (t + \Delta t) = l \cdot w \cdot t \]

resulting in an increase \( \Delta t \) of laminate thickness after intraply shearing:

\[
\Delta t = t \cdot \frac{(l\Delta w - w\Delta l + \Delta w\Delta l)}{(l w - \Delta w + w\Delta l - \Delta w\Delta l)} \tag{2.2.3.5}
\]

where

\[ l = L\sqrt{2}/2 \]
\[ w = L\sqrt{2}/2 \]
\[ \Delta l = L\sqrt{2}/2 \cdot (-1 + \cos(\phi/2) + \sin(\phi/2)) \]
\[ \Delta w = L\sqrt{2}/2 \cdot (1 - \cos(\phi/2) + \sin(\phi/2)) \]

This finally gives

\[
\Delta t = t \cdot \frac{(1 - \cos \phi \cdot \cos \phi)}{\cos \phi} \tag{2.2.3.6}
\]

With this relation, the local change of thickness of a sheared CFRTP laminate can be directly calculated from the local change of the crossover angle. Fig. 2.17 gives a representation of the change of laminate thickness as a function of the local shearing angle. It can be noticed that a shearing angle of 60\(^\circ\), which is not uncommon during thermoforming, requires a doubling of the material thickness.

![Graph showing the inherent increase of laminate thickness during shearing of the reinforcing fabric.](image)

Fig. 2.17 Inherent increase of laminate thickness during shearing of the reinforcing fabric.
Several computer programs have been developed to calculate the draping of a one-layered fabric cloth over a three dimensional shape\textsuperscript{39,40,41}. In general, computer software gives a geometrical prediction of the final crossover angles of an originally square fabric which completely covers the surface of a product mould. This geometrical prediction gives useful information about the possibility of forming a laminate into a three dimensional shape. In Fig. 2.18, an example is given of a shearing simulation by the computer program DRAPE of the Delft University of Technology. In this way the necessary shearing deformation can be determined beforehand in order to locate critical product details. So, optimisation of product moulds with regard to the intraply shearing capability of a fabric reinforced material is possible.

![Fig. 2.18 Output of a DRAPE shearing simulation of a fabric (right), to cover a product shape (left).](image)

With \textless 2.2.3.6\textgreater{} and known local crossover angles, it is possible to calculate the change in material thickness caused by shearing of the reinforcing fabric. This gives the mould designer a tool to adapt the local clearance in the mould, especially if rigid matching dies are to be used in a thermoforming process. The use of computer simulation of the draping can thus be very convenient when designing moulds.

Experiments\textsuperscript{42} have shown that draping simulations yield good approximations of reality for specific product shapes, but the user has to be well aware of the assumptions and boundary conditions that are used.

Some of the main discrepancies between the present software simulation and real intraply shear are the result of:
- straightening of the fibre bundles (Section 2.2.1),
- sliding of the crossover points, depending on the structure of the reinforcing fabric,
- the use of more fabric layers inside a laminate than one,
- forming forces, introduced by the mould surface and clamping devices (Chapters 4 and 5),
- the presence of a viscous thermoplastic resin.
The consequences and influences of the above mentioned items will be discussed in 2.4, where intraply shearing and the interaction with the thermoplastic matrix is treated more extensively.

### 2.3 Interply slipping of thermoplastic fabric reinforced laminates.

In Section 2.2.2, the importance of interply slip for the deformation of a thermoplastic laminate has been emphasized. The geometrical deformation as a result of the slip movements inside the laminate has been described. In this section, the interply slip phenomenon is dealt with from an experimental point of view. This allows for the estimation of the forming forces in a real thermoforming process and to quantify the interply slip deformation. The experimental results are compared to existing slip models, which are adjusted where necessary.

#### 2.3.1 Interply slip modelling, previous work.

The schematic description of a laminate in Section 2.2.2 helps to estimate the forming behaviour, but it is not sufficient to understand the internal force dynamics of the slipping laminate. The dimensions of the soft matrix interlayer, for instance, have been neglected until now. The behaviour of this matrix layer is important for the way the entire laminate is loaded during bending. Therefore the CFRTP laminate can be outlined more realistic by stacked plies, as illustrated in Fig. 2.19.

![Diagram of laminate by stacked plies](image)

*Fig. 2.19 Representation of a laminate by stacked plies.*

Each ply consists of a fibre rich layer with thickness $h$ and a matrix rich layer with thickness $\delta$. When this laminate is bent in a thermoforming process, the fibre layer and the matrix layer will behave differently. Tam$^{43,44}$, Scherer$^{45,47}$ and Talbot$^{48}$ give a theoretical description and an experimental verification of the behaviour of both the fibre rich layer and the matrix rich layer.
The linear viscoelastic ply-slip deformation model of Tam.

First the linear viscoelastic ply-slip deformation model of Tam\textsuperscript{44} and Tam and Gutowski\textsuperscript{45} is presented and discussed.

In their model it is assumed that the fibre rich layers behave linearly elastic, which implies that Hooke's law is supposed to be valid for the axial stress component:

$$\sigma_i = E_i \cdot e_i$$  \hspace{1cm} <2.3.1.1>

where \(\sigma_i\) is the axial stress in the \(i^{th}\) elastic layer, \(E_i\) is the Young's modulus, \(e_i = \delta u_i/\delta x\) is the longitudinal strain and \(u_i = u_i(x,t)\) is the longitudinal displacement. All the quantities refer to the x-direction.

The matrix rich layers are assumed to behave like a Newtonian fluid (this is valid if the strain rates are sufficiently low). For a one dimensional laminar flow between flat plates, with a linear velocity profile, the relation between the slip velocity and the shear stress in such a fluid is:

$$\tau_i = \mu_i \cdot \frac{V_{i+1} - V_i}{\delta_i}$$  \hspace{1cm} <2.3.1.2>

where \(\tau_i\) is the shear stress acting between the \(i^{th}\) and \((i+1)^{th}\) elastic layers, \(\mu_i\) is the matrix viscosity and \(V_i = \delta u_i/\delta t\) is the velocity of the \(i^{th}\) elastic layer.

A force balance on a differential element in the \(i^{th}\) elastic layer of a laminate gives, according to Tam's model (Fig. 2.20):

$$0 = \sigma_i h_i dy + \frac{\delta (\sigma_i h_i)}{\delta x} dx dy + \frac{\mu_i}{\delta_i} (\dot{u}_{i+1} - \dot{u}_i) dy dx dy - \sigma_i h_i dy - \frac{\mu_i}{\delta_i} (\dot{u}_i - \dot{u}_{i-1}) dy dx dy$$  \hspace{1cm} <2.3.1.3>

where the dot denotes differentiation with respect to time.

\[\frac{\mu_i}{\delta_i} (\dot{u}_i - \dot{u}_{i-1}) dy dx\]

\[\frac{\mu_i}{\delta_i} (\dot{u}_{i+1} - \dot{u}_i) dy dx\]

\[\sigma_i h_i dx dy\]

\[\sigma_i h_i dx dy + \frac{\delta (\sigma_i h_i)}{\delta x} dx dy\]

\[\frac{\mu_i}{\delta_i} (\dot{u}_{i+1} - \dot{u}_i) dy dx\]

Fig. 2.20 Force balance on a differential element, according to the Tam\textsuperscript{44} interply slip model.
Assuming that $h_i$ is constant, (2.3.1.3) reduces to:

$$0 = h_i \frac{\delta \sigma_i}{\delta x} + \mu_i \frac{\delta u_i}{\delta x} + \left[ \frac{\mu_i}{\delta_i} + \frac{\mu_{i-1}}{\delta_{i-1}} \right] \frac{\delta u_i}{\delta x} + \left[ \frac{\mu_{i-1}}{\delta_{i-1}} \right] \frac{\delta u_{i-1}}{\delta x}$$  \hspace{1cm} (2.3.1.4)

Assuming moreover that $E_i$ is independent of $x$, Hooke’s law can be differentiated, resulting in:

$$\frac{\delta \sigma_i}{\delta x} = E_i \frac{\delta \epsilon_i}{\delta x} = E_i \frac{\delta^2 u_i}{\delta x^2} = E_i \ddot{u}_i$$  \hspace{1cm} (2.3.1.5)

where the double accent denotes double differentiation with respect to place. (The individual bending stiffness of the elastic fibre layer is ignored.)

$\sigma_i$ can be solved from the coupled partial differential equations (2.3.1.4) and (2.3.1.5) with Fourier techniques and some simplifying assumptions; for more details see Tam et al. 43,44.

A possible deformation phenomenon in thermoforming is a three point bend strain input with a constant forming speed. The resulting stresses in the fibre rich layers can be solved, for instance as a function of the matrix viscosity $\mu_i$, or what is equivalent, the material temperature.

---

Fig. 2.21 Simulation of the stress development in a deforming isotherm laminate 44.
To evaluate the equations numerically, Tam has developed the software program **FORMSIM** that can calculate the induced stresses for a variety of forming cases and arbitrary user-definable material parameters. With this program several interesting thermoforming cases of CFRTP laminates were simulated:

- isothermal deformation (constant temperature through the thickness of the laminate)
- hotter or colder on the punch side (linear temperature profile)
- colder on both sides (parabolic temperature profile)

Conclusions were drawn with respect to the internal stress development during thermoforming. In Fig. 2.21 for example, a graph is shown in which the developing stress is calculated by **FORMSIM** during the three-point bending of an isothermal laminate.

**The yield shear stress and friction model of Scherer et al.**

Scherer, Zahn and Friedrich\(^4\) presented their first model of the interply slip process of unidirectional carbon-PEEK laminates in April 1990. This model is supported by a finite element analysis. The actual calculations are not reported however.

Each ply of the laminate is divided in a fibre rich area and a thin resin interlayer. The fibre rich layer is assumed to be relatively stiff and to behave linearly elastic. The behaviour of the hot matrix interlayer that allows for interply slip is described as follows:

- interply slip only takes place if the shear stresses in the laminate, raised by bending of the laminate exceed a certain shear yield stress \(\tau_{\text{yield}}\);
- when the \(\tau_{\text{yield}}\) is exceeded and slipping is started, the sliding resistance inside the matrix interlayer can be indicated by a coefficient of friction that represents the internal friction due to the material viscosity.

In Fig. 2.22, it is shown how the load-displacement curves of a slipping laminate are modelled.

---

Fig. 2.22 Load-displacement curves given by Scherer\(^4\) for bending of a laminate in which interply slip takes place.
In September 1990, Scherer and Friedrich\textsuperscript{46,47} presented an experimental background for the finite element analysis of the interply slip process: A pull-out experiment on unidirectional carbon fibre reinforced Polypropylene (Fig. 2.23). In this experiment a hot layer is pulled out of a laminate. The necessary force and the pull-out velocity are measured.

\begin{figure}
\centering
\includegraphics[width=\textwidth]{fig2_23.png}
\caption{Interply slip test setup of Scherer\textsuperscript{46}.}
\end{figure}

One of the main conclusions of the experimental work is that the interply sliding of a layer cannot be described as frictional slip alone. A more important role of material viscosity is proposed. A proposal to describe the interply slip resistance is given by a powerlaw:

\[ \tau = \tau_{\text{yield}} + \eta \cdot \dot{\gamma}^n \]  \hfill <2.3.1.7>

where $\eta$ is the viscosity of the hot matrix,
$n$ is a constant,
$\dot{\gamma}$ is the rate of shear deformation of the hot resin interlayer under shear traction (see Section 2.3.3).

Scherer et al. did not present modelling results however.

\textit{The linear elastic ply-slip deformation model of Talbott and Miller.}

A ply-slip deformation conception is the purely elastic ply-slip deformation model of Talbott and Miller\textsuperscript{48}, published in December 1990. In this model, each of the plies is represented by a hard, elasstical layer, representing the fibre rich layer and a soft elasstical layer representing the matrix rich layer. The laminate is divided in $N$ plies, each containing
M segments along their length. A node is placed at each end of an element, in the middle of the hard layer (Fig. 2.24). An element is identified by two subscripts, the segment number \( m \) followed by the ply number \( n \). Stress and strain are related by a linear constitutive equation:

\[
\sigma_{mn} = E \cdot \varepsilon_{mn} \tag{2.3.1.8}
\]

\[
\tau_{mn} = G \cdot \gamma_{mn} \tag{2.3.1.9}
\]

where \( E \) is the Young’s modulus of the hard material,
\( G \) is the shear modulus of the soft material,
\( \varepsilon_{mn} \) is the normal strain in the hard layer of element \( mn \),
\( \gamma_{mn} \) is the shear strain in the soft layer of element \( mn \).

![Fig. 2.24 Representation of the nodes of a laminate, used in the interply slip model of Talbott and Miller](image)

Normal stresses in the soft matrix material are neglected and the shear stress in a hard layer is assumed to be equal to the average of the shear stresses in the neighbouring soft layers.

The central equation, used in the model, states that the forces at each node must balance in the direction parallel to the laminate at that node. Individual elements of the laminate are subjected to both normal and shear stresses from the adjacent material in the same ply and shear stresses from the adjacent material above and below. For a node \( mn \) and half the element on each side thereof (Fig. 2.25):
\[
\begin{align*}
\tau_{mn} & \frac{1}{2}L_m \cos(\frac{\theta_m}{2}) + \tau_{mn}^{\text{mat}} \left( t^{\text{ply}} + \frac{1}{2} t^{\text{ply}} \right) \sin \theta_m + \tau_{m,n-1} \frac{1}{2} t^{\text{ply}} \sin \left( \frac{\theta_m}{2} \right) - \tau_{m,n-1} \frac{1}{2} t^{\text{ply}} \cos \theta_m \\
& + \tau_{m+1,n} \frac{1}{2} t^{\text{ply}} \cos \left( \frac{\theta_{m+1}}{2} \right) + \tau_{m+1,n}^{\text{mat}} \left( t^{\text{ply}} + \frac{1}{2} t^{\text{ply}} \right) \sin \theta_{m+1} + \tau_{m+1,n-1} \frac{1}{2} t^{\text{ply}} \sin \theta_{m+1} \\
& - \tau_{m+1,n-1} \frac{1}{2} t^{\text{ply}} \cos \left( \frac{\theta_{m+1}}{2} \right) - \sigma_{mn} \left( t^{\text{ply}} \cos(\theta_m) - \sigma_{mn} \right) t^{\text{ply}} \cos(\theta_m)
\end{align*}
\]

where:
- \(t^{\text{ply}}\) and \(t^{\text{mat}}\) is the thickness of the fibre and the matrix layers, respectively,
- \(\theta_m\) is the angle between the line perpendicular to the laminate at node \(mn\) and the perpendicular at the middle of segment \(m\),
- \(\theta_{m+1}\) is the angle between the perpendicular at node \(mn\) and that at the middle of segment \(m+1\),
- \(L_m\) and \(L_{m-1}\) are the lengths of segment \(m\) and segment \(m+1\), respectively.

Fig. 2.25 Stress balance on node \(mn\).

This description of the laminate is used to calculate the point-wise applied bending forces on the laminate, necessary to obtain a desired curvature distribution. Talbott and Miller use a Pascal computer program to evaluate the internal stress distributions over the segments and plies.

In their work, Talbott and Miller also present a dimensional parameter, called the Interply Sliding Parameter. This ISP indicates the ability of a laminate to relieve internal stresses by interply slip:
Deformation of fabric reinforced thermoplastics

\[ ISP = \left( \frac{E}{G} \right) \cdot \left( \frac{t_{\text{ply}}}{\text{length}} \right) \cdot \left( \frac{t_{\text{mat}}}{\text{length}} \right) \]  
\(<2.3.1.11>\)

where 'length' is the total length of the composite laminate undergoing bending. Each of the variables in the ISP affects the interply slip: the higher the ISP, the easier is sliding.

Discussion of the previously described interply slip models.

A problem with the use of Tam's linear viscoelastic deformation model is his description of the viscous behaviour of the matrix, given by equation <2.3.1.2>. The absolute value of the viscosity \( \mu \) has to be determined experimentally which is difficult for a CFRTP composite material\(^{29,49-51} \). Moreover, as with most unreinforced polymer materials, the viscosity depends on temperature and pressure.

For the use in rapid forming processes, Tam's model has another drawback: the assumption that the polymer matrix shows a Newtonian behaviour is not valid for fast flowing polymers. In the following section, experiments will be presented and discussed which show that the matrix material PEI behaves according to a power-law\(^a\). This implies that the viscous behaviour of the resin strongly depends on the deformation velocity. Especially at high deformation rates, the internal stresses in the laminate can be expected to differ substantially from those calculated with Tam's Newtonian model.

Nevertheless, the linear viscoelastic forming model of Tam and the FORMSIM software give a significant contribution to the understanding of the interply slip behaviour of thermoplastic laminates during thermoforming.

The calculation of the internal stresses by Talbott and Miller differs from the Tam and Gutowski model in the way the equations have been solved. But more than that Talbott and Miller use a completely different matrix interlayer behaviour by assuming a Hookian relation for the internal stress \( \tau_{\text{int}} \) and the shear deformation. This assumption is an oversimplification of the flow behaviour of softened thermoplastic materials. For the present work it is considered to be insufficiently accurate for the calculation of internal stresses\(^{12,33} \).

The Interply Sliding Parameter can by useful in practice, although the inclusion of the shear modulus \( G \) of the soft, interlaminar matrix material is questionable. The lack of experimental backup is also an important drawback of the model of Talbott and Miller. The linear, elastic model, however, can still be used to indicate the relative change of the internal stresses due to the application of external forming forces, both in and out of plane of the laminate.

\(^a\) Non-newtonian behaviour of liquid polymers is often represented by a power-law

\[ \tau = k \cdot (\dot{\gamma})^n \]

where \( n \) is the power-law index (\( n=1 \) for Newtonian behaviour).
In the work of Scherer et al., no insight is given in the calculation of the development of internal stresses in a laminate under bending deflection. They present a laminate as a stack of elastic beams that are fixed to each other until the imposed shear stress between the layers exceeds a typical yield shear stress. Here the classical bending theory seems to play a role in the estimation of internal stresses in the layers. More important is the way the resin rich layer is modelled. In the first paper (April 1990), a friction is suggested during interply slip raised by the matrix interlayer. A viscous drag however, cannot be represented by a friction coefficient, because friction does not depend on the slip velocity for instance. In September 1990 therefore, Scherer and Friedrich already adjusted the matrix modelling into a power-law behaviour. Unfortunately, the finite element analysis is not further discussed in the paper and therefore no theoretical results are compared to experimental ones.

2.3.2 Experimental verification.

In order to be able to quantify the influences of the processing parameters time, temperature and pressure on the interply slip behaviour of CFRTP materials, interply slip experiments have been carried out.

In the first experiment, an optimized version of the Scherer test setup is used. A small piece of the centre layer inside a glass fabric reinforced Poly Ether Imide (G/PEI) is pulled out by a constant force \( F_{\text{pull}} \). Meanwhile, the 5-layered test laminate is pressurized and heated by a hot platen press (Fontijne TP1000). The test setup has two main advantages compared to the original Scherer test setup: The first advantage is that in the new test setup, only the centre layer is allowed to slip, which enables a precise measurement of the slip displacement. The second advantage is the relatively small slip area, compared to the total specimen surface. When the centre layer is pulled out of the laminate, the pressurized centre-ply surface will decrease. Because the specimen is pressurized by a constant force of the hot platen press, the pressure on the slipping centre ply will increase during the test. This increase of surface pressure will be lower in the improved Scherer test setup because of the relatively small slip area.

The slip displacement of the centre layer is plotted as a function of time. The parameters pull-force, press-force and temperature are constant during the experiment. With the displacement/time plot, the slip velocity \( v \), and the acceleration \( a \), of the centre layer can be calculated at moment \( t \), with:
\[ v_i = \frac{x_{i+1} - x_{i-1}}{t_{i+1} - t_{i-1}} \tag{2.3.2.1} \]

\[ a_i = \frac{v_{i+1} - v_{i-1}}{t_{i+1} - t_{i-1}} \tag{2.3.2.2} \]

During a slip experiment, the pulling out of the centre layer is resisted by an interply shear stress \( \tau_i \), acting on the slip surfaces of the centre layer. Because of the decreasing slip surface during the test and thus the decreasing shear force, the constant pull force will cause an acceleration of the moving system. The force balance as presented in Fig. 2.26 is obtained by:

\[ F_{\text{pull}} - F_{\text{shear}} = m \cdot a_i \tag{2.3.2.3} \]

and

\[ F_{\text{shear}} = 2 \cdot \tau_i \cdot w \cdot (x_0 - x_i) \tag{2.3.2.4} \]

where \( m \) is the total mass of the accelerated setup parts and \( w \) is the width of the laminate.

![Diagram](image)

**Fig. 2.26** Schematic presentation of the force balance on a slipping laminate.

The interply shear stress at any moment \( t \), then can be calculated with:

\[ \tau_i = \frac{F_{\text{pull}} - m \cdot a_i}{2 \cdot w \cdot (x_0 - x_i)} \tag{2.3.2.5} \]

To facilitate the use of the experimental results in the next section, the slip data obtained from the tests are visualized in graphs, showing the interply shear stress versus the slip velocity.

In Fig. 2.27, the results are shown of interply slip at different slip temperatures. The specimen is practically without surface pressure. The figure shows that interply slip movement at lower temperatures and/or higher velocities, requires higher (shearing) forces.
Fig. 2.27 The influence of slip temperature on the shear stress in a slipping laminate, without pressure (improved Scherer test).

Fig. 2.28 gives the interply shear stress - slip velocity relation for a specimen surface pressure of about 0.11 MPa (increasing during the test to 0.17 MPa, because of the inherent reduction of the pressurized centre ply surface). This figure shows the same tendencies.

Fig. 2.28 The influence of slip temperature on the shear stress in a slipping laminate, slightly pressurized (improved Scherer test).
Fig. 2.29 Interply slip results, showing no evident influence of slight pressure differences on a slipping laminate (improved Scherer test).

Fig. 2.30 The influence of pressure on the shear stress in a slipping laminate (improved Schere test).
To visualize the effect of a pressure increase on the surface of the slipping specimen, several experiments are carried out with different pressing forces of the hot platen press. The results for these interply slip experiments are given in Fig. 2.29 for a test temperature of 360°C and relatively low press settings. The influence of the change in surface pressure on the slip behaviour of the specimen is not evident here. If the surface pressure is further increased, matrix flow during the experiments is noticed and the specimens tend to become thinner. The slip results for these specimens, slipped at a temperature of 360°C as well, are shown in Fig. 2.30. Additional tests have shown that interply slip becomes impossible when more than about 1 MPa surface pressure is applied on the centre layer of the laminate.

To overcome some of the drawbacks of the interply slip test setup as it is used in the previously described experiments, another test setup is developed, which will be referred to as the "diaphragm test setup". In the diaphragm test setup, the laminate specimen is pressurized in a pressure chamber by two Poly Imide (Upilex-R®) diaphragms that are forced against the specimen surface by air pressure. In this way, the pressure on the slip surface can be controlled better, since it does not vary with the amount of slip displacement like it does in the Scherer test setups. The specimen layers themselves are directly attached to the clamps of a 10 kN MTS test equipment with which a more precise force and displacement monitoring is possible. This test facility also offers the possibility of displacement controlled slip experiments. A displacement controlled test with a constant slip velocity of the centre layer of the test laminate implies the absence of acceleration phenomena and is closer to the interply slip deformation that takes place in real thermoforming processes.

[Diagram of a 12-layered G/PEI interply slip specimen in the diaphragm test setup]

**Fig. 2.31** Illustration of an interply slip specimen in the diaphragm test setup.

In Fig. 2.31 an illustration of a 12-layered G/PEI interply slip specimen is presented. The confined specimen is placed in a hot air chamber which provides the necessary heating of the specimen (Fig. 2.32). During heating, premature deconsolidation of the layers is prevented by a low surface pressure (0.1 MPa) provided by the diaphragms. At the slip temperature, the surface pressure is adjusted and pulling out of the outer layers of the specimen with a specified slip velocity is realized by the computer controlled test facility.
The interply shear stress is measured by a force transducer attached to the centre layer. In this way, the surface friction between the Upilex diaphragms and the moving specimen does not affect the measurement.

![Image of diaphragm test setup](image)

*Fig. 2.32 Diaphragm test setup for interply slip experiments.*

The shear stress at any moment can be calculated with:

\[
\tau_i = \frac{F_{\text{pull}}}{2 \cdot w \cdot (x_0 - \Delta x_i)}
\]

where \(\Delta x_i\) is the displacement of the layers, \(x_0\) is the initial length of the slip area and \(w\) is the specimen width.

In Fig. 2.33 an example is given of the diaphragm test output for a 6 mm/s slip experiment at 276°C and a surface pressure of 0.1 MPa. During the first 2 mm displacement, the rate of displacement is set at 0.1 mm/s to remove the slack in the setup, then the velocity is increased to \(v_{\text{slip}}\) instantaneously. The shape of the curve reveals a so-called "yield shear stress" at the start of the slipping process that has to be overcome before a constant slip velocity is realized. Once the yield shear stress has been overcome, the necessary shear stress to maintain the slip velocity is constant. The latter shear stress is the kind of interply shear stress that is also measured by the improved Scherer test setup.
Figs. 2.34 and 2.35 show the slip behaviour at 305°C. Fig. 2.35 shows that once the yield point is exceeded, a pressureless specimen does not offer slip resistance any more. The yield shear stresses and the slip shear stresses at 0.1 MPa pressure are higher at higher slip velocities. Interply slip tests at specimen surface pressures of 0.30 MPa and 0.35 MPa (at 305°C and an imposed deformation of 6 mm/s) result in necessary shear stresses of 0.20 MPa and higher, at which the present test setup will fail.
Fig. 2.35 The influence of the imposed slip velocity on the shear stress in a slipping laminate, without pressure (diaphragm test setup).

Fig. 2.36 The influence of slip temperature on the shear stress in a slipping laminate, slightly pressurized (diaphragm test setup).

In Figs. 2.36 and 2.37, the influence of slip temperatures is presented, for interply slip at a pressure of 0.1 MPa and for pressureless interply slip. The stress decreasing influence of higher temperatures is apparent, for both the yield stress and the slip stress.
Fig. 2.37 The influence of slip temperature on the shear stress in a slipping laminate, without pressure (diaphragm test setup).

The interply slip behaviour of glass fabric reinforced PEI as it was investigated with the diaphragm test setup reveals some interesting material properties. They will be implemented in the next section together with the slip data gathered by the improved Scherer test setup.

2.3.3 A modified interply slip model.

In Section 2.3.1, two flow models are mentioned to describe the interply slip behaviour of thermoplastic composites were discussed\textsuperscript{32,33}. It was already mentioned there that Tam et al. use a Newtonian flow behaviour:

\[
\tau = k \dot{\gamma}
\]

whereas Scherer et al. use a power-law (pseudo-plastic) flow behaviour:

\[
\tau = k \dot{\gamma}^n
\]

where \(\tau\) is the interply shear stress, \(k\) and \(n\) are constants and \(\dot{\gamma}\) is the rate of shear deformation.

The relations between shear stress and the rate of shear deformation can be extended with a yield value \(\tau_y\), yielding the Bingham model description:
\[ \tau = \tau_0 + k \cdot \dot{\gamma} \]  

<2.3.3.3>

and the "power-law with yield value":

\[ \tau = \tau_0 + k \cdot \dot{\gamma}^n \]  

<2.3.3.4>

which was already presented in a slightly different form as <2.3.1.7>. In Fig. 2.38, the relation between the shear stress and the deformation rate for the different flow models is schematically presented.

*Fig. 2.38 Relation between the shear stress and the deformation rate of a polymer material according to different models.*

*Fig. 2.39 Shear deformation of the matrix rich layer during interply slipping.*
The rate of shear deformation during interply slipping with a (relative) slip velocity \( v_{\text{slip}} \) is given by (Fig. 2.39):

\[
\dot{\gamma} = \frac{v_{\text{slip}}}{\delta_m}
\]  \(<2.3.3.5>\)

where \( \delta_m \) is the thickness of the matrix rich layer between the two adjacent fibre rich layers.

Unreinforced fluid polymers generally show a powerlaw behaviour. In Fig. 2.40\(^{33a} \), the relation between the apparent viscosity \( \eta \) and the shear rate is given for Ultem 1000\(^\circ\), the PEI resin that is used in the composite slip specimens. The minimum deformation rate \( \dot{\gamma}_{\text{min}} \) for pseudo-plastic behaviour (at 360°C) is about \( 10^2 \) s\(^{-1}\). Fig. 2.41 shows that although the thickness of the matrix rich layer of fabric reinforced composites hardly can be defined, \( \delta_m \) certainly is not larger than \( 10^{-2} \) mm. Using equation \(<2.3.3.5>\), the minimum \( v_{\text{slip}} \) to obtain pseudo-plastic behaviour is roughly 1 mm/s.

![VISCOSITY OF ULTEM 1000 vs. SHEAR RATE](image)

*Fig. 2.40 Apparent viscosity of Poly Ether Imide (Ultem 1000) as a function of the shear rate\(^{33} \).*

The interply slip test results of Fig. 2.28 for 360°C and slip velocity > 1 mm/s, fit well with the powerlaw \(<2.3.3.2>\). In Fig. 2.44 this is shown for \( n=0.435 \) and \( K=k/(\delta_m)^n = 0.124 \) [N s\(^n\) mm\(^{(n-2)}\)].
Fig. 2.41 Distribution of the reinforcement in a fabric reinforced PEI laminate.

curve fitting ($n=0.435$, $K=0.124$)

slip temperature 360°C
surface pressure 0.11 MPa

Fig. 2.42 Powerlaw curve fitting of interply slip results of the improved Scherer tests.
In the relation

\[ \tau = K \cdot \nu_{\text{slip}}^n \quad \text{(2.3.3.6)} \]

the constant \( K = k/(\delta_m)^n \) includes the thickness of the matrix rich layer \( \delta_m \). This thickness cannot be measured in CFRTP laminates as shown in Fig. 2.41, so it only can have a theoretical value. The constant \( k \) is strongly related to the apparent viscosity \( \eta \) of the resin\(^a\).

The constant \( K \) therefore has to be determined by interply slip tests of the appropriate slip environment. The experimental results of Section 2.3.2 show the relation between the slip shear stress and the slip temperature at a constant slip velocity. In Figs. 2.43 and 2.44 this relation is given and the relation of "constant" \( K \) with temperature is calculated from powerlaw curve fitting. The power constant \( n \) hardly changes with the slip temperature, as is demonstrated in Fig. 2.45.

![Graph showing the relation between slip shear stress and temperature](image)

*Fig. 2.43 Relation of the slip shear stress with the test temperature, according to measurements with the improved Scherer setup and the diaphragm setup.*

\(^a\)Often the temperature dependence of polymer viscosity is given by the Arrhenius relation \( \eta = A e^{B/T} \) \(^{32-34}\).
Fig. 2.44 Relation of the "constant" K with temperature during the interply slipping of a laminate.

Fig. 2.45 Powerlaw curve fitting of interply slip results for various slip temperatures, fitted with a constant power n=0.33.
It can be concluded that the general interply slip behaviour indeed can be described by the powerlaw relation <2.3.3.6> and the representation of interply slip in a laminate is valid once the interply slip deformation is started. The relation is useful to quantitatively describe the internal stresses and thus the necessary forming forces during the deformation of a thermoplastic laminate. Because of the realistic slip environment during the experiments, it is expected that the influence of the process parameters in a thermoforming process on interply slip deformation can be predicted accurately by the relation <2.3.3.6>.

Fig. 2.46 Microscope picture of the nesting of the fabric reinforcement of neighbouring layers in a PEI laminate.

The slip deformation of fabric reinforced thermoplastic layers is not a purely rheological phenomenon, however. The schematic representation of a CFRTP material by fibre rich layers alternated by matrix rich layers as shown in Fig. 2.37, is not completely accurate. This is shown by the microscope picture of Fig. 2.46. From this picture, taken before interply slip is introduced, it can be concluded that the fibre bundles of adjacent fabric layers are intimately nested. In order to start interply slip in such a laminate, the neighbouring fabric layers have to be separated first (deconsolidation) and a mechanical "friction" has to be overcome. In Fig. 2.47 a picture is shown on which the roughness of the slipped layer surface is visible. Fig. 2.48 demonstrates the appearance of a slipping laminate. Between the slipping surfaces, voids are dragged in the slip direction by the fibre bundles perpendicular to the slip direction. The voids are introduced by the deconsolidation of the layers.
Fig. 2.47 Microscope picture showing the surface roughness of a slipped layer due to deconsolidation.

Fig. 2.48 Voids in a slipped laminate, which are dragged by the slipping surfaces.
The "mechanical friction" that has to be overcome to start slipping can be denoted as the yield shear stress $\tau_{\text{yield}}$.

It must be emphasized that the yield shear stress $\tau_{\text{yield}}$ that is observed during the diaphragm slip test is considerably different from the yield shear stress $\tau_{\text{yield}}$ as introduced by Scherer in equation <2.3.1.7>. In the work of Scherer, it is suggested that the shear yield stress is the minimum required shear stress to start and continue interply slip deformation. The shear yield stress as described in <2.3.3.7>, however, is the minimum required shear stress only to start an interply slip motion.

Once the shear yield stress is exceeded, this lower limit is no longer of importance: as long as the layers continue to slip, the interply slip deformation is governed completely by the shear stress according to relation <2.3.3.6>, hence by the velocity of the slipping layers.

![Shear Stress vs Displacement](image)

*Fig. 2.49 Interpretation of the test output of interply slip experiments with the diaphragm test setup.*

The $\tau_{\text{yield}}$ is demonstrated by the interply slipping results of the diaphragm test setup in Section 2.3.2 (Fig. 2.49). As demonstrated by the slip tests, the $\tau_{\text{yield}}$ necessary to start interply slip at a certain imposed velocity can exceed the required $\tau_{\text{slip}}$ to continue interply slip at that velocity. This can cause a "bump" in the shear stress - displacement curve, as is also illustrated by Fig. 2.49. In that case, the necessary deconsolidation of the layers requires a higher shear stress than the continuation of the slipping itself. The height of the bump in Figs. 2.34 to 2.37, or, when the bumps are absent, the height of the shear stress at the same displacement, can be set against the slip temperature and the imposed slip velocity (Fig. 2.50 and Fig. 2.51).
Fig. 2.50 Influence of the imposed slip velocity on the yield shear stress in a laminate at an imposed slip velocity of 6 mm/s.

Fig. 2.51 Influence of the imposed slip velocity on the yield shear stress in a laminate at a temperature of 305°C.
The linear dependence of the yield shear stress with slip temperature and velocity can be depicted by:

\[ \tau_{\text{yield}} = C_1 \cdot v_{\text{slip}} + C_2 \cdot T_{\text{slip}} + \tau_0 \]  \hspace{1cm} \text{(2.3.3.7)}

where the constants \( C_1 > 0 \) [N·s·mm\(^{-1}\)], \( C_2 < 0 \) [N·K·mm\(^{-2}\)] and \( \tau_0 = f(\text{pressure}) \). Both the imposed slip velocity dependence and the temperature dependence of the yield shear stress is stronger than those of the slip shear stress as presented by \(<2.3.3.6>\).

From the slip results of Section 2.3.2, it can be noticed that when the slip velocity is sufficiently low and/or the temperature is sufficiently high, the slip shear stress will exceed the yield shear stress. In that case, the bump in the shear stress - displacement curve is absent (for instance see Fig. 2.36) and the deconsolidation of the layers is not the most determining resistance against slip deformation. The comparison of \( \tau_{\text{yield}} \) with \( \tau_{\text{slip}} \) is graphically depicted in Figs. 2.52a and b as a function of slip velocity and temperature. An interesting slip result is obtained when the matrix interlayer is artificially increased up to a thickness where the neighbouring fabric layers are not nested anymore. When such a laminate is forced to slip, the \( \tau_{\text{yield}} \) is nearly absent then, and \( \tau_{\text{slip}} \) is decreased (Fig. 2.52c and d).

![Graph showing the comparison between yield shear stress and slip shear stress](image_url)

**Fig. 2.52a** Comparison between yield shear stress and slip shear stress at various temperatures (imposed slip velocity is 6 mm/s).
Fig. 2.52b Comparison between yield shear stress and slip shear stress at various imposed slip velocities (at a temperature of 305°C).

Fig. 2.52c Microscope photograph of the absence of nesting in a laminate with an extra amount of resin between the fabric layers.
Fig. 2.52d The shear stress of a laminate with an extra amount of resin compared to the measured yield and slip shear stress of a usual laminate at the same slip parameters.

To conclude this section, an overview is given of the described interply slip models in Table 2.1. The inclusion of the yield shear stress in the visualization and modelling of the interply slip phenomenon is necessary to determine the history of internal stresses inside a deforming thermoplastic laminate. It has been shown in this section that a rheological approach alone is not sufficient to completely describe the interply slip phenomenon.

<table>
<thead>
<tr>
<th>Model</th>
<th>Interply Slip Description</th>
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<tbody>
<tr>
<td></td>
<td>Fabric Behaviour</td>
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<tr>
<td>Tam</td>
<td>Linear Elastic</td>
</tr>
<tr>
<td>Talbott</td>
<td>Linear Elastic</td>
</tr>
<tr>
<td>Scherer</td>
<td>Linear Elastic</td>
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<tr>
<td><strong>Present Work</strong></td>
<td>In-Extensible</td>
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</tbody>
</table>
2.4 Intraply shearing of fabric reinforced thermoplastics.

In Section 2.2.3, a geometrical description has been given of the deformation mode intraply shearing of dry fabrics. Here a more complete view on the deformation phenomenon is the goal in that sense that the forces acting on the shearing fabric and the interaction with the viscous thermoplastic resin will be considered.

2.4.1 Theory of intraply shearing forces.

Textiles are being used in clothing all over the world and it is therefore not surprising that a great deal on fabric and yarn research comes from textile industry. The results of investigations on fabrics like they have been performed on a large scale in the flourishing textile industry of the sixties and seventies are numerous. Many of the results of this research can be used to obtain a better understanding of the behaviour of fabrics in reinforced composites. Investigators like Eeg-Olofsson$^{54}$, Behre$^{55}$, Treloar$^{56}$ and others$^{57,58}$ developed techniques for the measurement of shear properties of fabrics and analyzed the shear deformation.

In Fig. 2.53, the instrument is shown that has been devised by Mörner and Eeg-Olofsson for measuring the shearing properties of a vertically clamped fabric in its own plane. By rotating a disc, a horizontal displacement of the lower clamp is imposed and shearing of the clamped fabric is the result. By the measurement of the change of reaction force $\Delta P$, the resistance against shearing of the fabric can be measured as a function of the shearing angle. The shearing angle at which wrinkling starts is also estimated. The shearing resistance is assumed to determine the drape qualities.

![Fig. 2.53 Shearing apparatus, developed by Mörner and Eeg-Olofsson\textsuperscript{54}.](image-url)
Behre analyzed the stress distribution in a fabric sample subjected to shear by a modification of Mörner’s device and subsequently constructed a non-recording shear tester. Treloar investigated the effect of test piece dimensions of the shearing fabrics and found out that especially at low fabric tension the shape of specimens has a large influence. He also introduced a simplified version of a shear apparatus, which will be used in Section 2.4.2.

In 1973, Kawabata, Niwa and Kawai published "The Finite-Deformation Theory of Plain-Weave Fabrics". It consists of three parts: the biaxial-deformation theory, the uniaxial-deformation theory and the shear-deformation theory. Kawabata et al. determined the behaviour of a fabric from its structure and the fibre and yarn properties. In their work they also considered large shear deformations (up to 30°) as well as combined tensile and shear deformations. Parts of the Kawabata shear-deformation theory are interpreted in this thesis for the use in the description of the shear deformation of fabrics in CFRTP materials. The ultimate goal is the understanding of the inherent resistance of reinforcing fabrics in thermoplastic composites to shearing deformation during rubber forming.

\[ T_r = T_{r0} + (C_1+C_2\phi) \cdot F_c + (C_3+C_4F_c) \cdot \phi \]  

Fig. 2.54 Schematic presentation of shearing fabric yarns (\(\theta\) and \(\varphi\) are the original crossover angle and the shearing angle, respectively).

Kawabata introduced a linear approximation for the torque required for a decrease \(\varphi\) of the crossover angle in an originally rectangular fabric (Fig. 2.54):

This torque is related to the value of the compressive force \(F_c\) acting on the contact surfaces of the warp and weft yarns (Fig. 2.55) and also to that of \(\varphi\). The second term in equation <2.4.1.1> represents the effect of friction between the intersecting yarns. The friction increases with decreasing crossover angle as the contact surface of the interlacing fibre bundles is enlarged (Fig. 2.54) and it is assumed to be linearly proportional with the compressive force. The third term is an elastic term, again denoting the influence of
crossover angle and compressive force. Equation \( \langle 2.4.1.1 \rangle \) is shown to be a good enough approximation for practical calculations, according to Kawabata.

Force equilibrium (Fig. 2.55) yields the relation between the contact compressive force \( F_c \), the warp yarn tension \( F_{T_1} \) and the weft yarn tension \( F_{T_2} \):

\[
F_c = 2F_{T_1}\sin\theta_1 = 2F_{T_2}\sin\theta_2 \tag{<2.4.1.2>}
\]

where \( \theta_{1,2} \) are the angles between the yarns and their principal axis.

Fig. 2.55 Force balance on two intersecting fabric yarns.

Fig. 2.56 Presentation of the resolving of the external forces in a resin impregnated fabric.
In the clothing yarns, the tensions $F_{T1,2}$ also cause stretching. However, because the fibre bundles in the composite reinforcements have a relatively high stiffness, stretching is not considered in the shearing deformation of the reinforcing fabrics in CFRTP materials.

The fibre tensions $F_{T1,2}$ increase the contact compressive force $F_c$ and therefore the required torque necessary for shearing, according to Kawabata’s equation $<2.4.1.1>$. In order to change the crossover angle during rubber forming, the torque $T_r$ has to be raised by the external imposed forming forces (stretching forces). In Fig. 2.56 is outlined how these external forces $F_{ext}$ also introduce the fibre tension force $F_{te} = \frac{1}{2} F_{ext} \cdot \cos (\phi/2)$. Together with a possible global fabric tension $F_{rg}$, the external force $F_{te}$ provides the tension force $F_T$:

$$F_T = F_{te} + F_{rg} = \frac{1}{2} F_{ext} \cdot \cos(\phi/2) + F_{rg}$$  \hspace{1cm} <2.4.1.3>

Equation of moments yields for the torque:

$$T_r = F_{T1} \cdot m = \frac{1}{2} F_{ext} \cdot \sin(\phi/2) \cdot m$$  \hspace{1cm} <2.4.1.4>

![Fig. 2.57 Bending of fabric yarns during thermoforming because of the relatively high viscosity of the resin.](image)

In practice, when impregnated with viscous thermoplastic matrix, the distance $m$ will not be equal to the mesh width, as depicted in Fig. 2.57, because of slight bending of the fibre bundles. Some important theoretical conclusions, however, can already be drawn for the prediction of the shearing capability of fabrics;

a) From equation $<2.4.1.4>$, it can be concluded that an externally applied force $F_{ext}$ becomes less effective during the shearing deformation, since the crossover angle $\phi$ is decreasing.
b) From equations <2.4.1.1>, <2.4.1.2> and <2.4.1.3>, it can be concluded that during shearing deformation the necessary torque for further shearing increases. This is not only due to the decrease ϕ of the crossover angle, but also because the fibre tension F₁ and thus the compression force Fₖ at the crossover contact point increases.

c) When a fabric is used with lower interlacing degree, for instance a 2/2 twill weave instead of a plain weave (Fig. 2.58), a lower crossover compression force Fₖ will occur, because equation <2.4.1.2> will change into Fₖ = F₁sinθ. An even lower interlacing degree (e.g. 5H satin, Fig. 2.58) will reduce the number of compressed crossover points and a lower required torque Tₑ for shearing will be the result.

![Diagram of fabric weaves: Plain weave, 2/2 twill weave, 5H satin weave](image)

Fig. 2.58 Schematic presentation of various important fabric structures.

d) It can be expected that a fabric with a higher density requires higher external forming forces Fₑₓ for shearing, because the crossover surface is larger (higher friction term in equation <2.4.1.1>) and the mesh width m in equation <2.4.1.4> is smaller, making the Fₑₓ less effective. Shearing causes a further increase in fabric density and thus will be hindered when the starting density is already high.

In Section 2.4.3 the influence of the mentioned fabric parameters is investigated experimentally and will be discussed in relation to the equations presented here.

### 2.4.2 Intrapy shearing of dry fabric reinforcements.

To verify the theoretical considerations for intrapy shearing drawn in Section 2.4.1, a test setup is constructed. With the test setup intrapy shearing tests are performed on dry (non-impregnated) glass fabrics, which are used as reinforcements in CFRTP materials.

In Fig. 2.59, a schematic illustration of the intrapy shear test is presented which is derived from the Treloar shear apparatus. The dry fabric (one layer) that is to be tested is vertically positioned in the setup by clamping of two edges (AB and DE in the figure). The upper clamp is fixed, whereas the lower clamp can freely move in the vertical plane. The lower clamp can be loaded in vertical direction by a weight W and in horizontal direction by a force F.
Fig. 2.59 Schematic presentation of the shearing test setup for non-impregnated fabrics.

If the force $F$ is increased, the fabric will be forced to shear in its plane and a shearing angle $\varphi$ (= change of crossover angle) can be established, as illustrated in the figure. As soon as $\varphi \neq 0$, an extra horizontal force $H$ is necessary to neutralize the horizontal component $W \tan \varphi$ on the shearing fabric. To expel this effect, the force $R$ is denoted to represent the force necessary to overcome the internal shearing resistance of the specimen:

$$R = F - H = F - W \cdot \tan \varphi$$

During the intraply shear test, the force $R$ will be increased until no further change in the shearing angle $\varphi$ can be noticed or wrinkles start to appear. At that point the maximum shearing angle $\varphi_{\text{max}}$ is defined. The detection of the wrinkles is facilitated by the use of light beams shining parallel to the fabric surface.

In Fig. 2.60, an example of the test output is given for a 1/3 twill fabric (Fig. 2.61). The tested fabric has a density of 12 fibre bundles per cm². Per 1 cm² of the fabric surface, six fibre bundles run in warp direction and six fibre bundles run in weft direction. Because of the friction at the crossover points, the shearing fabric shows a hysteresis effect; apparently, the relation of the shearing force $R$ with the shearing angle $\varphi$ depends on whether $\varphi$ is increasing or decreasing. The downgoing $R$-$\varphi$ curve will not be presented any further in the test results, since in general the increase of $\varphi$ is the most relevant during thermoforming.
Deformation of fabric reinforced thermoplastics

Fig. 2.60 Example of the output of the shearing test setup, showing the hysteresis effect of (dry) shearing yarns.

Fig. 2.61 Examples of the interlacing of yarns in various fabric reinforcements (from left to right: plain, twill and 5H satin weave).

From Fig. 2.60, it can also be noticed that the increase of the shearing angle $\phi$ with increasing force $R$ is reduced at higher deformations. This demonstrates the effect of equation <2.4.1.4> since the interlacing angle $\phi$ decreases with increasing shearing angle $\phi$. Beside this, due to the higher fabric tension at a higher $R$ force, the compression force $F_c$ at the crossover contact points increases. Finally, at $\phi_{\text{max}}$ an equilibrium is established.

The glass fabrics, tested in the shearing apparatus\textsuperscript{66}, are listed in Table 2.2.
Table 2.2 Properties of the investigated fabrics.

<table>
<thead>
<tr>
<th>fabric type</th>
<th>number of yarns/mm²</th>
<th>yarn weight (tex)</th>
<th>fabric thickness (mm)</th>
<th>fabric weight (g/m²)</th>
</tr>
</thead>
<tbody>
<tr>
<td>plain I</td>
<td>6</td>
<td>6</td>
<td>37</td>
<td>0.54</td>
</tr>
<tr>
<td>plain II</td>
<td>16.5</td>
<td>16.5</td>
<td>69</td>
<td>0.59</td>
</tr>
<tr>
<td>plain III</td>
<td>12</td>
<td>12</td>
<td>70</td>
<td>0.60</td>
</tr>
<tr>
<td>twill I</td>
<td>12</td>
<td>22</td>
<td>23</td>
<td>0.43</td>
</tr>
<tr>
<td>twill II</td>
<td>12</td>
<td>18</td>
<td>140</td>
<td>0.51</td>
</tr>
<tr>
<td>twill III</td>
<td>11</td>
<td>7</td>
<td>74</td>
<td>0.83</td>
</tr>
<tr>
<td>satin</td>
<td>22</td>
<td>21</td>
<td>58</td>
<td>0.52</td>
</tr>
</tbody>
</table>

In Figs. 2.62, the intraply shear results are presented of the dry fabrics. A less interlaced fabric structure (Fig. 2.61) offers a substantially higher intraply shearing potential. A higher fabric tension (higher W) hardly influences the maximum shearing angle but does increase the necessary shearing force R, as is expected from equations <2.4.1.1> and <2.4.2.1>.

In Fig. 2.62a Shearing results of the tested glass fabrics (tested at weights W=0.09 N/cm and W=0.20 N/cm).
Fig. 2.62b Shearing results of the tested glass fabrics (tested at weights \( W = 0.12 \) N/cm and \( W = 0.30 \) N/cm).

Fig. 2.63 Influence of the fabric structure and density on the maximum shearing angle.
Fig. 2.63 shows the influence of the fabric structure and density on the maximum shearing angle. The decrease of crossover surface at a lower degree of fabric interlacing and a lower fabric density enables a higher intraply shearing deformation. The conclusions from the discussion on the Kawabata theory (Section 2.4.1) are verified by the intraply shearing tests on several types of glass fabric reinforcements. The Kawabata theory, therefore, can be helpful in solving intraply shearing problems during thermoforming processes. The influence of the thermoplastic resin on the intraply shearing behaviour of a fabric reinforcement, however, has to be investigated also. This is carried out in Section 2.4.3.

2.4.3 Influence of thermoplastic resin.

A test setup is assembled\textsuperscript{61}, to determine the optimal processing environment for intraply shear deformation of fabric reinforced thermoplastics. With this intraply shear test setup the processing conditions temperature, surface pressure and applied shear forces on a laminate specimen can be varied. Basically, the test setup consists of a hot platen press (Fontijne TP1000) in which a specimen is heated and pressurized, and a frame that is used to apply the shear forces on the laminate and to monitor the intraply shear deformation (Fig. 2.64).

![Diagram of test setup](image)

**Fig. 2.64 Test setup for the measurement of intraply shearing of impregnated fabrics.**
A (4-layered) +45°/-45° oriented fabric reinforced G/PEI laminate (30x30cm²) is fixed at one short end and pulled at the other short end by a constant weight W at a specified temperature and pressure. The elongation of the specimen that is imposed this way has to be realized by intraply shearing of the fabric reinforcement (Section 2.2.3). By monitoring the displacement of the loaded specimen end, the intraply shearing angle φ inside the laminate can be determined. When the specimen elongation is stopped, the maximum shearing angle φ_{max} is obtained. It is noted that the change of the angle φ during the experiment is not homogeneously distributed over the length of the specimen and thus the measurement is not completely reliable (Fig. 2.65). However, the test gives a good indication of the influence of the process parameters.

![Diagram](image)

*Fig. 2.65 Presentation of the "bulk" deformation of the shearing specimens.*

In Fig. 2.66, an example is given of the output of an interply shear test of a 4-layered G/PEI laminate at 350°C and 0.15MPa surface pressure by a shear force of 400N. (Because of the relatively long deformation times, acceleration effects are neglected.) It can be noticed that the shearing rate diminishes with increasing shearing angle, as was expected from the Kawabata theory for dry fabrics. When the specimen cannot be elongated any further, an equilibrium is established where φ_{max} is realized. The maximum shearing angle φ_{max}, which is a measure of the intraply shearing potential of the laminate at a particular process environment, is used to determine the influence of the process parameters.

In Fig. 2.67, the increase of the maximum shearing angle with increasing fabric stretching force W is indicated. The raise in φ_{max} demonstrates that the maximum shearing deformation is determined by an internal force equilibrium (as discussed in Section 2.4.1) which can be shifted by a change in externally applied forming forces. Since the effectiveness of a forming force diminishes at higher shearing angles, the relation φ_{max} vs W is non-linear. Theoretically, the necessary shearing force W has to raise infinitely to approach the situation where the fabric is sheared to φ_{max} = 90°. Beside this, at higher W the crossover
compression force $F_c$ is increased according to $<2.4.1.2>$. This also raises the torque $T_r$, necessary for shearing (equation $<2.4.1.1>$). Because of the applied surface pressure on the laminate specimen, intraply wrinkling will be postponed compared to the shearing of free dry fabrics, since a pressurized fabric has a higher stability.

**Fig. 2.66** Example of the test output of the intraply shearing tests on impregnated fabrics.

**Fig. 2.67** Maximum shearing angles in a glass fabric reinforced PEI laminate.
The influence of the process temperature on the intraply shearing behaviour of the specimen is depicted in Figs. 2.68. An increase of process temperature enables a higher maximum shearing deformation but also raises the intraply shearing rate (Fig. 2.68b). Obviously, the rheology of the thermoplastic resin is important during intraply shearing because at higher temperatures the shearing rate is increased. The raising of $\varphi_{\text{max}}$ at increased process temperature confirms that the thermoplastic matrix in a CFRTP material acts as a lubricant. A lower viscosity of the matrix (at a higher temperature) lowers the crossover point surface friction of the interlacing fibre bundles.

![Graph showing the relationship between pressure, maximum shearing angle, and temperature.](image)

**Fig. 2.68a Influence of the process parameters temperature and pressure on the maximum shearing deformation of a laminate.**

Fig. 2.68a also points out that the maximum shearing deformation is decreased at a higher surface pressure. In Fig. 2.69a the $\varphi_{\text{max}}$ - surface pressure relation at 300°C and a shearing force of 400N is presented. It is believed that the surface pressure directly influences the crossover compression force $F_c$ of the interlacing fibre bundles in the laminate. According to relation $<$2.4.1.1$>$ a higher pressure will increase the torque necessary for intraply shearing leading to a force equilibrium at a lower shearing angle (lower $\varphi_{\text{max}}$). In Fig. 2.69b, it is shown that the deformation rate is lowered at higher pressure on the specimen. Besides the increase of friction at the fabric crossover points, a second cause can be pointed out. During the intraply shear deformation, the fibre volume fraction is unchanged (no matrix squeeze out). According to equation $<$2.2.3.6$>$, an increase of the specimen thickness must accompany the shearing deformation. The surface pressure will obstruct the thickening of the laminate and can lower the deformation rate, therefore.
Fig. 2.68b Influence of the temperature on the shearing velocity and maximum shearing deformation of a laminate.

Fig. 2.69a Influence of the surface pressure on the maximum shearing deformation of a laminate.
Deformation of fabric reinforced thermoplastics

Fig. 2.69b Influence of the surface pressure on the shearing velocity and maximum shearing deformation of a laminate.

Because of the non-linear relation between $\phi_{\text{max}}$ and $W$, and because of the influence of the surface friction between the elongating specimen and the press platens, a doubling of the number of laminate layers of a specimen will reduce the maximum shearing angle less than 50% when the shearing force is not changed (Fig. 2.70).

Another point of interest is that an increase of the matrix volume fraction by adding an extra amount of resin between the laminate layers does not affect the intra-ply shearing force behaviour, which confirms the intra-ply character of the shearing deformation (Fig. 2.71).

Fig. 2.70 Influence of the number of fabric layers in a laminate on the shearing behaviour.
Fig. 2.71 Influence of the fibre volume fraction on the shearing behaviour of a laminate.

Finally, in Fig. 2.72 a comparison is shown between the maximum shearing deformation of an impregnated satin fabric reinforced G/PEI laminate and the same fabric without thermoplastic resin. Although only a rough qualitative comparison is possible (different process parameters give different results), the figure is representative for the lubricating influence of the thermoplastic resin on intraply shearing deformation.

To conclude this section, it can be stated that in general the thermoplastic matrix has a positive influence on the intraply shear potential of a fabric. Although the Kawabata theory does not take into account the lubrication effect of the thermoplastic resin, it still can be used to qualitatively describe the fabric shearing, as presented in Section 2.4.1.

Fig. 2.72 Comparison between the shearing results of dry fabrics and resin impregnated fabric reinforced laminates.
2.5 Optimal processing conditions for CFRTP product shaping.

The goal of a thermoforming cycle is the realization of a continuous fibre reinforced thermoplastic product with a specified product quality. To deform a CFRTP sheet into a more dimensional product, forming forces are necessary, as is discussed in this chapter. The forming forces not only must be sufficiently high, they also have to be applied in the most adequate material direction to enable interply slipping of the layers and intraply shearing of the reinforcing fabric where necessary.

Because residual stresses have to be avoided (see 5.5) and because of economical considerations, the necessary forming forces must be as low as possible. Sections 2.3 and 2.4 show how the process parameters during a forming cycle influence the level of the required forming forces: The surface pressure on the deforming laminate has to be minimized, the process temperature has to be high and the fabric deformations have to take place as slowly as possible to facilitate the shaping of the composite product. Relations <2.3.3.6>, <2.3.3.7> and <2.4.1.1> between those parameters are verified by experimental results in the preceding two sections. For real thermoforming, however, these relations are not sufficient as may become clear from the following paragraphs.

![Diagram showing the effect of deformation time on slip shear force](image)

**Fig. 2.73 Illustration of the determination of an optimal deformation time of a slipping laminate.**

It is important to recognize the mutual dependence of processing parameters in a thermoforming technique. As an example, it is schematically shown in Fig. 2.73 in what way the necessary shear force for interply slip deformation depends on the time that is technically assigned for the simple bending of an originally flat sheet in a non-isothermal
thermoforming cycle: According to equation <2.3.3.6>, a slower process requires lower necessary slip shear forces because the imposed slip velocity decreases. In a non-isothermal thermoforming technique (with relatively cold moulds) the material quickly cools down during the deformation. An increase in deformation time therefore results in interply slip at lower temperature, which will raise the necessary slip shear force, as is depicted in Fig. 2.73. The result of this consideration is that an optimal deformation time (= optimal forming speed) must be defined with respect to the required slip shear force.

During the thermoforming of a more complex composite product, the imposed forming sequence of the product details has to be controlled. First, the local deformation velocity depends on the mould geometry (see the example at the end of Section 2.2.2). Especially at processing conditions where the shear stress, necessary to start interply slip \( \tau_{\text{yield}} \) (Section 2.2.3) at a certain imposed slip velocity is high, it is advantageous to increase the slip velocity as soon as the (lower) \( \tau_{\text{yield}} \) at a lower imposed slip velocity has been overcome. In other words, the forming sequence of a laminate has to be imposed in such way that deformations are introduced gradually.

The sequence of surface pressurization of local material spots during a moulding process is also determined by the shape of the thermoforming moulds. From Sections 2.3 and 2.4 it has become clear that an increase of the surface pressure lowers the deformation capability and rate. During the thermoforming cycle, therefore, the application of surface pressure has to be minimized on material spots that still have to be rearranged internally. Furthermore, the cooling rate of a laminate spot is increased when it is pressurized by the relatively cold mould, also increasing the required forming force.

To optimize CFRTP product shaping, it is necessary to include the thermoforming moulds in the definition "processing conditions". The design of the moulds and an optimal combination of the process parameters time, temperature and pressure, therefore, are equally important. Non-optimal processing conditions can easily lead to product mismatches like material wrinkling and breaking during thermoforming. Other mismatches are inhomogeneous fibre-matrix distributions and residual internal stresses in the product. In those cases, the mechanical properties of the product will be affected seriously.

In order to be able to offer the most ideal processing conditions during thermoforming, a moulding technique is necessary in which both the process parameters and the mould geometry can be adapted to the laminate deformation needs as much as possible. In the following chapters the realization of optimal processing conditions by the newly developed rubber forming technique is presented.
CHAPTER 3. The technical development of rubber forming.

3.1 Introduction.

The arrangement of the fibre reinforcement primarily determines the mechanical performance of the final product. If it is possible to arrange the fibres in the most efficient configuration in a composite product, a mechanically optimal composite part can be created. In practice, deviation from the ideal fibre configuration in the final product is often the result of the limitations of the forming process.

In manufacturing of thermoset composite products, the shaping can be close to ideal. This is possible due to the "forming forces" that can be applied directly to each fibre layer or even each fibre bundle in the mould (e.g. hand lay-up).

In thermoplastic composite product manufacturing, it is rarely possible to directly impose the locally necessary forming forces in the fibre reinforcement during the shaping of the product. In a thermoforming cycle the rearrangements of the fibres have to be controlled by indirectly working technical devices like stamp, clamping frame, etc.

The deformations of the fabric reinforcement of a CFRTP sheet, necessary to realize a contoured, aligned product have already been discussed in Chapter 2. To allow the deformation of the reinforcement the thermoplastic matrix first has to be softened. This requires a heating cycle in the process before the forming forces can be applied. When the viscosity of the thermoplastic resin has been lowered sufficiently, the deformation of the sheet can commence.

In reality, it is difficult to apply the forming forces onto the reinforcement in a theoretically ideal way. Most of the current thermoforming methods differ in their technique of imposing the necessary forming forces on the composite laminate. From the development and use of thermoforming techniques up to now, it can be concluded that the higher the required level of mechanical performance of the final products, the more complex (and the more expensive) the processing techniques. There seems to be a dilemma between "ease of production" and "product performance".

A consequence of process improvements is that a higher product performance can be realized while the production costs remain constant. Sometimes a certain product quality can be realized at lower production costs using thermoforming processes that originally were unable to cope with the quality demands.

In 3.2 an overview is given of the main current thermoforming techniques. To clarify the differences between the various techniques, special significance is given to the shaping
methods and the technical equipment to realize them.

The fast compression moulding technique, called the rubber forming technique, is introduced in a separate section (3.3), since it forms the main subject of the present work. In this technique, a rubber die-half is used to force the thermoplastic composite material into the product shape. Using rubber forming, a relatively high degree of fibre control can be realized in the shaping phase, followed by a good consolidation at the end of the forming process.

In 3.4, an example of automation of fast thermoforming processes, as it is realized at Delft University of Technology, is described. The rubber forming technique as well as matched-metal die forming are applied with this automated facility. Both processes show good perspectives for commercial applications\textsuperscript{62-64}.

3.2 Overview of current thermoforming techniques.

3.2.1 Heating methods.

Prior to the forming process, the thermoplastic composite laminate is heated. Its temperature must be sufficiently high to reduce the matrix viscosity to processing values. The choice of the heating device mainly influences the heating time rather than the final product quality. Most of the commercially available heating equipment can be used.

One possibility is the heating of the laminate by contact heat between two heated plates (\textit{conduction heating}). A disadvantage of this heating method is the fact that the laminate actually contacts the heating equipment. Proper releasing agents have to be used, to prevent the sticking of the material to the heating equipment.

In \textit{dielectric and induction heating} methods, a strong alternating magnetic field or electrical field is created in the composite material\textsuperscript{165,166}. These fields induce an electrical current and agitation of the molecules in the material which raise the material’s temperature.

\textit{Convection heating} in an oven is also possible, but is usually relatively slow. Because of the long heating times, the use of inert gas is preferable to prevent oxidation of the polymer at high temperatures.

The use of inert gas is also recommended when using \textit{infrared heating methods}, though heating times usually are short in those cases. A disadvantage is that in thick sheets temperature gradients can develop through the thickness which sometimes restrain the formability in the forming phase. The use of radiation implies the absence of surface contact with the material and yields a quick heating method in general. The heating device is well controllable in general.
3.2.2 Major thermoforming processes.

The major processing methods to shape continuous fibre reinforced thermoplastic materials that are described in the literature\textsuperscript{1-3,6,15,17,28} are:

- diaphragm forming
- hydroforming
- rubber pressing
- matched (metal) die forming
- rubber forming

The first four methods will be briefly discussed in the following paragraphs. Rubber forming, the main subject of the present work, will be discussed separately in 3.3, as mentioned before.

Diaphragm forming (Fig. 3.1)

In diaphragm forming, the laminate is placed between two plastically deformable diaphragms. The disposable diaphragms are clamped onto the mould and vacuum is applied between these foils. The whole setup is heated to the processing temperature where air pressure (in an autoclave) is used to force the diaphragms, with the laminate in between, into the mould.

![Diaphragm forming diagram](image)

Fig. 3.1 Schematic view of diaphragm forming.

During the forming process the laminate can slide within the foils. The diaphragms are stretched and create surface friction on the laminate. This friction finally introduces tensile forces in the fibre reinforcement. The forming forces can be controlled indirectly by way of the friction between the diaphragms and the laminate surface. When the sheets finally touch the mould, the friction process will proceed on a more local scale. During the deformation the diaphragms will create good out-of-plane support and suppress wrinkling. Due to the low deformation rate, internal stresses are easily relieved in the process. Because the rigid mould has to be heated to the processing temperature, extensive heating and cooling cycles are necessary. The process cycle time in diaphragm forming is relatively
high (30 min. and more) compared to other thermoforming techniques. Another disadvantage is the temperature dependent limitation of the formability of the diaphragms. Recent studies, however, give good prospects to reduce these limitations.

*Hydroforming (Fig. 3.2)*

Hydroforming is similar to diaphragm forming in that sense that a fluid medium, usually a hydraulic oil, is used to deform the laminate over or in a mould. The fluid is trapped by a rubber diaphragm, yielding one-sided contact between the diaphragm and the deforming laminate only. Hydroforming, therefore, offers less possibilities than diaphragm forming to control and induce specific forming forces on the thermoplastic laminate. On the other hand, higher hydrostatic pressures can be imposed on the surface of the laminate. In hydroforming, the laminate is heated outside the forming equipment which results in a lower cycle time. Hydroforming stems directly from the metal sheet forming industry. It usually implies the use of massive pressure systems. Only one product-dependent tool (one mould) is needed, however, which makes hydroforming a fairly flexible manufacturing technique.

![Diagram of hydroforming](image)

*Fig. 3.2 Schematic view of hydroforming.*

*Rubber pressing (Fig. 3.3)*

Rubber pressing of thermoplastic composites (sometimes called rubber block forming or rubber pad press forming) is another forming technique that was derived from the metal industry. Here a thick pad of rubber acts as a pressure medium that forces the hot laminate against a male or female tool.
The technical development of rubber forming

![Diagram of rubber pressing](image)

**Fig. 3.3 Schematic view of rubber pressing.**

The rubber will create a combination of surface pressure and surface tension during the rubber pressing deformation. The process provides high normal forming forces but they are not uniformly distributed over the laminate. The local forming forces are determined by the shape of the die only, which counteracts the closing of the press. The complexity of products that can be formed in the rubber pressing technique is limited, due to the limited local deformation of the rubber pad. Only one product-dependent mould has to be used.

**Matched-die forming (Fig. 3.4)**

Matched-die forming is the most widely used thermoforming system. Here, the hot laminate is positioned between two matching metal dies and quickly forced into a product shape. The use of matching metal dies in a forming technique often results in a proper surface quality and precise details on both sides of the thermoplastic composite part. The male and female dies usually can be easily heated internally. High (local) pressures can be applied on the laminate during forming.

![Diagram of matched metal-die forming](image)

**Fig. 3.4 Schematic view of matched metal-die forming.**
A disadvantage of the use of rigid moulds is that it does not provide the ability to compensate thickness mismatches between the deforming laminate and the mould cavity. This will dominate the pressure distribution on the material, which can result in a non-uniform consolidation and/or varying fibre-volume fractions along the product.

In matched metal die forming, it generally is difficult to control the deformation of the fibre reinforcement, because the rigid forming dies introduce only highly localized surface forces on the deforming laminate. To solve the problem of uncontrolled reinforcement deformation and non-ideal pressurization, more complicated moulds can be used equipped with moving parts inside ("living moulds"). These parts can ensure a certain forming sequence.

3.3 The rubber forming technique.

3.3.1 Description of the rubber forming setup.

The rubber forming technique has been developed to combine some of the advantages of the matched metal-die forming and the rubber pressing technique. In the rubber forming technique, one of the rigid metal dies of the matched metal-die forming method has been replaced with a flexible, rubber die. A typical rubber forming setup consequently consists of a rigid (metal) mould, a flexible (rubber) mould and eventually a clamping or sliding frame.

Fig. 3.5 Clamping frame for transport and blankholding of hot thermoplastic laminate sheets.
The thermoplastic laminate is heated to the processing temperature outside the forming equipment. This reduces the necessary heating time substantially, since only the composite sheet has to be heated. At the TUD, heating is done either by contact heating in a hot platen press or by infrared heating. When infrared heating is used, a typical heating cycle for a laminate without clamping frame can be carried out within 1-4 minutes.

When the temperature of the thermoplastic sheet equals the necessary processing temperature, the material is quickly transferred to the forming press. Cooling of the laminate during the transfer has to be minimized. A clamping frame can be used as a transferring device from heating to forming system (Fig. 3.5), but sometimes it is more convenient to integrate the clamping frame in the moulds (Fig. 3.6).

![Fig. 3.6 Clamping frame, integrated in the mould.](image)

After the sheet is positioned between the two moulds and (if necessary) the clamping or sliding device is installed, the press is closed and the product is formed. Forming times of 1-5 seconds are normal in rubber forming. When the shape of the composite part is realized and final consolidation (reconsolidation) is assured, the product must be cooled down under pressure. The cooling time can be reduced by forced water cooling of the metal die. When the temperature of the part is low enough, it can be taken out and trimmed if necessary.

As already mentioned, the forming device in rubber forming consists of one rigid die (mostly made of steel or aluminium) and a flexible rubber die. In general, the rubber die is made of PolyUrethane (PU) or a heat resistant castable silicon rubber (see Appendix C2). Different rubber hardnesses are available. Because of the flexibility of the rubber die, exact
matching of the two dies is not necessary; this is illustrated in Fig. 3.7. The special mould features of the rubber forming technique in the forming and the consolidation phase will be discussed in the following sections.

![Diagram of rubber forming technique]

**Fig. 3.7 Schematic view of the rubber forming technique.**

### 3.3.2 Consolidation features.

The results described in Chapter 2 show that for optimal deformation of the fibre reinforcement the surface pressure on the laminate has to be minimal. Until the deformation of the laminate into the product shape has been accomplished, the rearranged layers of the laminate must not be pressurized.

Apparently, an antithesis occurs during the manufacturing of thermoplastic composite parts: To create a good environment for the forming phase, surface pressure must be avoided and to assure good reconsolidation, a surface pressure is needed.

**Rigid moulds.**

In the matched (metal) die technique, the forming device is a combination of two rigid matching moulds. The forming and the final consolidation are realized by the closing of these two moulds.

Three main problems can occur:

- deformation of the laminate can be obstructed when surface pressure starts too early;
- thickness differences due to intraply shearing must be anticipated rather precisely;
- final consolidation will be different over the product due to important differences in surface pressure in the consolidation phase.
If a product shape is fairly open, like the product A, depicted in Fig. 3.8, the surface pressure requirements in both the forming and the consolidation phase can be fulfilled. During the deformation of the laminate, there is enough space between the approaching moulds to avoid surface pressure on the laminate. At the end of the forming stroke, the final product shape is already present and no internal rearrangement of the reinforcement is necessary any more. Finally, the two rigid matching moulds will close completely and the laminate will be pressurized completely, nearly at once. The required surface pressure for reconsolidation will be present everywhere.

![Various examples of product shapes](image)

*Fig. 3.8 Various examples of product shapes.*

If a product shape has steeper sides (e.g. shapes B and C in Fig. 3.8) the transition from forming to consolidation pressurization will be less evident. Unfortunately, the free space between the stamp and the bottom mould will decrease more rapidly at the sides than at the bottom (Fig. 3.9). The laminate still rearranges internally by interply slip or intraply shear until the freedom of movement disappears completely at the sides. At that moment, the bottom is not fully covered by the composite material. This means that forming of the material can be obstructed before the final product shape is realized. A low product quality is the result.

![Example of a product shape](image)

*Fig. 3.9 Example of a product shape.*
During consolidation, the pressure is not uniformly divided over the product surface. In the case of the product of Fig. 3.9, the force $P$, externally provided by the press facility, is oriented in z-direction. Assuming that $P$ is reacted uniformly over the projected mould surface, the magnitude of the normal surface pressure at the bottom of the product is proportional to $P$. The normal surface pressure at the sides of the product, however, is related to $P \cdot \cos \theta$. This implies that during the consolidation of a product, for instance product B from Fig. 3.8, the effective consolidation pressure varies from maximum at the centre line of the product to zero at the top of the sides. For a hat shaped stiffener with vertical sides the surface pressure is absent completely over the sides (product C in Fig. 3.8). In real products the consolidation is far from optimal, there.

In the case of less "sensitive" shapes, for instance for low values of $\theta$, it may be possible to find an optimum in dimensioning of the two matching moulds to assure a complete deformation of the laminate into the product on one hand, and a sufficiently good consolidation on the other hand. But it will not be possible to design the moulds in such a way that pressureless forming together with a homogeneous consolidation afterwards are likely.

During the processing of a continuous fibre reinforced thermoplastic, the material has limited ability to adjust in thickness direction by matrix flow. Even if this ability is exploited, the limited flow of thermoplastic matrix can create unwanted changes in the fibre rearrangement or the matrix volume fraction. So material flow during thermoforming of CFRTP materials is not a reliable deformation phenomenon.

It implies that, in the case of thermoforming CFRTP materials, the concept of "flexible materials with rigid moulds" as it is used for pressing unreinforced materials, with good flow characteristics, has to be changed into "flexible moulds for rigid materials". Where "rigid" means that the thickness of the deforming material cannot be influenced by the pressing force. It is only determined by the degree of deformation of the fibre reinforcement.

---

*Fig. 3.10 Example of a "living" mould, equipped with moveable parts.*
Some research has been done at Delft University of Technology on the use of "living" moulds. One rigid mould has been divided into several sub-moulds (e.g. bottom and sides) that are mechanically assembled in such a way that they can act fairly independently. In Fig. 3.10 an example of such a "flexible" mould is shown. In this mould, first the bottom of the product will be formed and "frozen" before the sides will be pressurized.

Rubber moulds.

A less complicated way of creating a flexible mould to process CFRTP sheets is the replacement of one of the rigid moulds by a rubber die. A rubber mould has the capability to adjust its shape elastically to a rigid casing under relatively low loading. The combination of this adjustment capability and a low compressibility (= constant volume), make rubber an ideal pressure medium.

Since rubber acts like a fluid, a force in "punch" direction can be transformed into forces in other directions (Fig. 3.11). If, for instance, a rubber positive mould is used, a high clearance is possible between the positive and negative moulds. Interply slip and intraply shear are easily possible inside the laminate because of the absence of any surface pressure during the forming phase.

![Diagram of rubber forming process](image)

Fig. 3.11 Illustration of the "yielding" characteristic of a loaded rubber block.

When the rubber mould touches the bottom of the rigid mould, a further punch movement downwards results in deformation of the rubber at the contact point. Due to its "fluid" properties, the rubber tends to fill the rigid mould completely. The extension of the rubber mould closes the free space between the rubber mould and the rigid mould completely and finally pressurization of the laminate at the sides of the product will start.
Fig. 3.12 Pressure measurements on the sides and bottom in a U-shaped mould (the different pressure profiles are gained during incremental increases of the top load on the stamp).

An important advantage of rubber is that in a locked situation it behaves almost hydrostatically. An increase of the punch force P on the rubber mould results in a more or less proportional increase of the surface pressure on the laminate everywhere (Fig. 3.12). Therefore, the consolidation quality is influenced less by the (part) geometry than in the case of two rigid moulds. The angle θ in Fig. 3.9 is of minor importance in the pressurization during consolidation. If the casing around the rubber mould, formed by product and punch is perfect, the pressure system is nearly a hydraulic one. Rubber forming approaches the uniform distribution of the consolidation pressure of a hydroforming system.

3.3.3 Forming features.

With the rubber forming technique it is possible to impose the forming sequence of a CFRTP laminate in the optimal way. Certain kinds of rubbers, e.g. several silicone rubbers, allow up to 600 percent elongation. The combination of a certain stiffness of the rubber with the high adjusting capability of the rubber mould to the rigid mould offers extraordinary forming possibilities. These possibilities will be demonstrated here qualitatively with four examples.
Most of the forming options of a rubber mould are based on the fact that the surface pressure on a laminate has to be minimal during its deformation. The hat-shaped stiffener of Section 3.3.2 is used as an example to show how an intentional gap between the sides of a positive mould and a negative mould can be closed in the consolidation phase, so during the increase of mould pressure. This gap is needed to avoid pressurization of the sliding laminate during the forming phase.

Fig. 3.13 Example of the use of a highly non-matching rubber stamp to form a partly "closed" beam.

An extreme of this hat-shaped beam is shown in Fig. 3.13. In the figure, it is shown how an extremely non-matching rubber stamp could be used to thermoform such a non-releasable product. During the forming phase, the laminate will be drawn into the negative mould with low surface pressure only. The rubber mould will hardly deform. When the laminate touches the bottom of the rigid die, it will be pressurized by the rubber stamp. The reacting pressure will cause a deformation of the rubber, which will proceed until the mould has been filled completely. By then, the surface pressure on the laminate at the middle of the bottom will already be high. This part of the laminate however is already formed and no sliding or other internal deformation is needed there. The optimal form of the rubber stamp has to be calculated from rubber deformation theories (see also Appendix E).

Another example is the production of a sine-wave shaped bar or a stiffened panel, of which the negative moulds are illustrated in Fig. 3.14. Manufacturing the ledged product shapes by a stamp that exactly matches can give forming problems due to clamping of the laminate before all ridges are completely formed. This clamping is schematically shown in Fig. 3.15. Forming a sine-wave by matching dies is still possible when the bar only consists of a few (say two) waves, but if more sine-shaped waves are to be realized, the cumulative friction of all the ridges will soon become too large to transport the CFRTP material to the bottom of each sine-wave.

Fig. 3.14 Illustration of a sine-wave shaped and a hat-stiffened panel.
Fig. 3.15 Illustration of the clamping of a laminate due to excessive friction during forming.

A solution to the forming problem could be a change of the forming sequence of the laminate. Instead of forming all the waves at the same time and creating friction surfaces at all those places, they can be imposed one by one, leaving the remaining surface of the laminate free of pressure and friction. A schematic view of the suggested rubber forming method is given in Fig. 3.16.

Fig. 3.16 Proposals for the shape of a rubber stamp to enable the forming of a laminate into a ledged product.

For the realisation of an aircraft beam with flanged lightening holes and flanged edges as illustrated in Fig. 3.17, a forming sequence must be determined beforehand to enable free laminate rearrangement as much as possible. Several critical surface areas can be indicated. The fibre reinforced laminate that has to cover surfaces marked A and B in Fig. 3.18, is likely to get clamped at the higher neighbouring sides when two exactly matching moulds are used. To prevent this clamping when one side of the sheet is fixed, the opposite side must be free of pressure. In that way it is possible to supply the composite material from the free side to fully cover the mould surface.
The technical development of rubber forming

Fig. 3.17 Illustration of an aircraft beam with lightening holes and flanged edges.

Fig. 3.18 Designation of the critical forming areas in the beam.

An optimal forming sequence is depicted in Fig. 3.19 by the numbers 1 to 7, accounting for the "one-side-free" forming concept. The critical areas A (sequence 1 to 5) and the areas B (6) must be totally formed before the sides (7) are pressurized. The shape of the suggested rubber mould is also sketched and again it can be noted that in order to realize the desired forming sequence, a highly non-matching rubber mould has to be used.

Fig. 3.19 Proposal for the shape of a rubber stamp to determine an optimal forming sequence.
The last example is a car rim. A negative mould is shown in Fig. 3.20. It is noted that the final shape of the rim contains a number of circumferential ridges. The rim also contains several (nearly) vertical areas, so the use of rubber for a positive mould is already justified by the need for consolidation pressure at these vertical areas. Due to the adjusting capability of the rubber it is not necessary to fully copy all details of the final product shape in the rubber mould. It is sometimes possible to use a non-detailed, only slightly preformed rubber surface to realize a detailed, curved product shape like the car rim.

![Image of a car rim with ridges](image)

*Fig. 3.20 Ledged shaped of the rigid negative mould to shape a composite car rim.*

This last example shows that the use of a rubber mould does not necessarily lead to a thermoforming process that is more complicated than a matched metal-die process. Nevertheless, the non-matching rubber approach as applied in positive rubber forming implies the calculation of the required geometry of the rubber stamp. This will be discussed later.

A change of stiffness of the mould (which is technically noted as a change of hardness) will alter the forming sequence of the laminate. A soft rubber will approach the forming behaviour of a hydroforming pressure medium and therefore will yield a uniform pressure distribution, especially on flat surfaces. A stiff (hard) rubber will behave more like a rigid mould and can therefore locally introduce higher surface pressures. It is also possible to use hybrid rubber moulds that consist of two or more kinds of rubbers to optimize the pressure profile during rubber forming (Fig. 3.21).
Fig. 3.21 Example of a hybrid rubber stamp, consisting of (differently) coloured rubbers with different hardness.

In order to estimate the optimal geometry and hardness of a rubber mould, it is important to investigate the rubber behaviour thoroughly. The optimization of pressure profiles in the rubber forming technique will be discussed in Chapter 4.

3.3.4 Additional remarks.

To complete the description of the rubber forming technique, some additional advantages and disadvantages, relative to matched-metal die forming are discussed.

The rubber moulds can be created out of PolyUrethane or Silicone rubber types, which are often castable (Appendix C2). Some Silicone rubber types are available in room temperature hardening versions (RTV-silicone rubber). The possibility of casting one of the moulds in the other one can reduce the mould manufacturing time and costs essentially. The rigid mould has to be machined or casted only. It then can be used as a casting die for the rubber mould by covering the rigid mould surface by a sheet (e.g. wax) with a specific thickness. If the rubber mould substantially differs from the rigid one, as is discussed in Section 3.3.3, this method is not possible and a separate casting die has to be assembled. The high material price of castable, high-temperature resistant rubbers compared to aluminium or steel mould material is not advantageous.

Rubber is a good insulator. An insulating mould has the advantage that it slows down the cooling of the heated laminate when the mould touches the laminate. In the case of non-isotherm thermoforming this is important, since in the forming phase the laminate has to stay hot as long as possible to facilitate the deformation of the material. A disadvantage of the rubber insulation is the resulting high temperature of the rubber surface during
thermoforming. Rubber has a limited service life due to wear, friction and disintegration at high service temperatures. A hot rubber surface will be affected more and the service life of the rubber will decrease significantly with rising temperature. The service life of a rubber mould has to be investigated, still. Furthermore, internal mould heating and forced cooling of the rubber to optimize the temperature control of the deforming and consolidating laminate is not possible due to the low heat conductivity. In case of a temperature rise of the rubber mould during processing, the high coefficient of thermal expansion (in the order of $10^{-4}$/K) should be accounted for.

Another point of attention in the rubber forming technique is the relatively low stiffness of rubber, which can give premature deformation of the mould in the forming phase of the process. Especially when narrow and deep details have to be formed in the composite product there is usually not much space left to apply a non-matching rubber mould. Moreover, a low width to height ratio of the rubber mould can cause excessive deformation of the rubber by laminate tensile forces (Fig. 3.22). A so-called "barrelling effect" of the rubber can be the result, causing clamping of the composite sheet at the sides before the details of the bottom of the product have been entirely realized.

![Diagram](image)

**Fig. 3.22 Schematic view of the barrelling effect of rubber dies during rubber forming.**

Due to a low stiffness, the rubber surface of a mould can be indented very easily by small particles like fibre bundles (especially when the rubber surface is hot and soft). These deformations are elastic, so the rubber surface itself is not permanently damaged, but the surface quality of the side of the product that contacted the rubber is not as good as the quality of the opposite side. A high quality surface can only be formed at the rigid mould side. If both surfaces must be very smooth, a matched-metal die forming process has to be considered to realize the product.
3.4 Automation of the rubber forming technique.

The development of a rubber forming technique that fulfils the manufacturing demands of both aerospace and automotive applications is difficult. Automation of the thermoforming technique is a necessity to come close to either one of these application areas. The use of automated production steps provides low cycle times necessary in car part manufacturing. It also may enhance the reproducibility and thus the mechanical quality of CFRTP products up to aerospace standards. This automation has to prove the viability of the manufacturing of the high quality CFRTP parts.

In the automotive industry, the production ranges from 100 to 1000 identical parts a day. This justifies the use of high speed machinery and relatively expensive tooling, as tooling costs are only a minor part of the total costs of a product. In aerospace industry on the contrary, it is usually not necessary to concentrate on a high production speed. There a large variety of small series of airplane parts with reliable (mechanical) quality is characteristic. The use of light-weight, low-cost tools that can be changed quickly, is preferred. In aerospace the use of flexible, multi-purpose equipment is most suitable and specific, product related tooling costs have to be limited. Nevertheless, a higher production speed at the same cost level is advantageous also in aerospace industries.

The processing advantages of CFRTP materials have rarely been exploited in practice. At the Delft University of Technology an automated thermoforming facility (NHR134) has been developed and installed in 1992 in close cooperation with the Dutch manufacturer of special machinery FONTINE HOLLAND B.V. It gives the possibility to change from laboratory scale experiments to prototype and 0-series production of thermoplastic composite parts. The technical use and working of each processing step with this thermoforming equipment will be described here.

In Section 3.3.1 a technical description of the rubber forming machinery that is needed has already been given. In the (automated) thermoforming facility, all the product-independent equipment is integrated in the standard machinery. The product-dependent moulds and clamping devices are not considered as parts of the facility and can easily be installed and removed. Due to the flexible, product-independent hardware, the machine can be used for several kinds of hot stamping processes. Only rubber forming is described here.

During a rubber forming cycle, six main steps can be distinguished:

1. feeding
2. heating
3. clamping
4. forming
5. consolidating and cooling
6. removing

These steps are materialized in the machinery, shown in Fig. 3.23.
Fig. 3.23 View of an automated rubber forming facility (realized by Fontijne Holland BV).

Fig. 3.24 Illustration of a transferring device for hot thermoplastic laminates.

Between the several steps, the composite material is transferred by a transferring device. This device consists of two pneumatic "holding arms", mounted on an electrically driven frame (Fig. 3.24). It transports the CFRTP sheet from the feeding area into the infrared oven, holds the sheet in the oven for a predetermined time and then quickly transfers it into the forming facility where it is taken over.
The transfer speed can be changed: 0.1 m/s or 1 m/s. Another feature of the transferring device, is that it does not transport the laminate in a straight line: When the laminate is heated to the processing temperature, it will tend to sag. In order to compensate for this "sagging", the transferring device will be lifted hydraulically before the hot laminate is transported into the mould. In this way it is tilted over the mould edge. There the device will be lowered again to enable the taking over of the hot laminate.

**Feeding of the prepreg sheet.**

Starting material of the rubber forming in general is a pre-consolidated sheet. It is placed manually in the transferring device which pneumatically clamps the sheet at two sides. It is also possible to use commingled fabric or separated prepreg sheets as starting material. However, the heating device consists of a radiation heater, so by using separated layers highly inhomogeneous heating may occur. Furthermore, the use of unconsolidated layers implies the need for a longer consolidation afterwards which will increase the total cycle time drastically. The maximum sheet format is 1.85 m x 1.00 m, while the maximum thickness is about 10 mm. If economically justified, the placement can be automated by installing a feedstock at the beginning of the transferring device, as is illustrated in Fig. 3.23.

---

*Fig. 3.25 Illustration of the infra-red heating panels as used in the Fontijne NHR134 thermo-forming facility.*
Heating of the material.

After the laminate has been loaded in the transferring device, it is transported between two infrared heating panels. The panels consist of medium-wave quartz heaters. The radiation power of the heaters is at full capacity within a few seconds, offering a heating device that is capable of in-situ controlling sheet temperature with a few degrees accuracy (see Section 4.3.3). The distance between the panels and the composite sheet can be varied. The radiating surface of the heating panels is divided into areas which can be activated separately, depending on the laminate area that has to be heated (Fig. 3.25).

If the thermoforming facility runs in the automatic mode, the laminate is positioned automatically between the IR-heaters and the radiation is activated during a pre-adjusted heating time. This time must be sufficient for the sheet to reach the processing temperature. Then the hot laminate is transported into the forming station.

Clamping of the hot laminate.

When the transferring device with the laminate has been lowered over the bottom mould, the hydraulic press will start to close. During the first seconds of the stroke of the press, a pneumatically driven clamping frame will take over the laminate from the transferring device (Fig. 3.26). Then the transferring device will return empty to the feeding position where it is ready to accept a new sheet of prepreg.

\[ \text{Fig. 3.26 Illustration of a pneumatically driven clamping frame.} \]
In the meantime, the edges of the hot laminate are clamped at specific points or areas around the mould. The main function of the clamping frame is the introduction of forming forces in the plane of the laminate. These forming forces are sometimes necessary to realize a wrinkle free product. This will be discussed in more detail in Chapter 4. Mechanisms can be attached on the clamping frame to impose a particular forming behaviour of a the material.

**Forming of the laminate.**

The actual forming is an interaction between the downward movement of the upper mould and the obstruction of material sliding at the edges of the laminate. During the forming stroke the press force is relatively low. The closing speed of the moulds is rather important because during the closing the thermoplastic composite is already cooling down and the deformation of the laminate has to take place while the material is above a certain temperature. The forming stroke of the hydraulic press is displacement controlled. The closing speed can be varied from 0 mm/s to approximately 180 mm/s. During a stroke two velocity steps are possible. This enables a transition from maximum closing speed to a more gentle forming speed.

**Consolidating and cooling of the product.**

When the two moulds are closed, the laminate has been shaped completely and has to consolidate again. To provide sufficient consolidation pressure, a 1600 cm$^2$ cross-section hydraulic jack can be pressurized within a few seconds to supply a total pressing force up to 4000 kN. While working with the rubber forming method, this means that a maximum sized product (approx. 0.80 m x 1.75 m projected surface) can be consolidated at a hydrostatic pressure of 3 MPa at all product surfaces. After the consolidation the pressure has been maintained for a determined period during which the product cools down; the press then opens automatically.

**Removal of the composite part.**

During the opening stroke of the press, the clamping frame will stay closed for another moment, so the cooled down product is forced to remain in the lower mould. Then it can be removed, triggering a switch that allows the following heated laminate to enter the mould. It is also possible to have the product taken out by a robotic device that can subsequently trim the edges of the product and/or realize the drilling of any desired slots or holes. This option is included in Fig. 3.23.

All of the mentioned steps can be controlled, either manually or in the automatic mode. In the automatic mode the operator only has to give a starting command after a composite sheet has been loaded. The only other actions he has to take care of are the loading of the sheet and the removal of the final products.
With the described thermoforming facility it is possible to realize thermoplastic composite products from flat sheet at room temperature to a cooled down product, in a production time of 2-5 minutes. This time depends on the thickness (and kind) of the composite sheet only and not on its size, since the bottlenecks of the thermoforming cycle are the IR-heating and the cooling in the mould of the laminate. Due to the use of heating of the composite sheet outside the mould, two sheets can be processed at the same time: while the first laminate is being shaped, reconsolidated and cooled down in the moulds, the transferring device will return, reload and take the next sheet into the IR-oven. It is therefore possible to manufacture a product every 55 seconds, as is proven with the production of the full-scale car doors of Fig. 3.28.

![Full-scale car doors, rubber formed in the automated thermoforming facility out of glassfabric reinforced thermoplastics.](image)

The short thermoforming cycles that are realized by automation make rubber forming or matched-metal die forming of continuous fibre reinforced thermoplastics products competitors, not only for traditional metal forming, but also for production methods for composite products as sheet moulding compound (SMC) and resin transfer moulding (RTM)\(^{68,69}\). In combination with a good control of the rearrangement of the continuous fibre reinforcement in thermoplastic composites, the rubber forming technique has the potential to close the gap between automotive and aerospace applications.
CHAPTER 4. Optimisation of the rubber forming process parameters.

4.1 Introduction.

The efficient application of process parameters during a thermoforming cycle is important to realize good thermoplastic composite products. In Chapter 2, it was explained what process conditions are required to ensure optimal deformation behaviour of the CFRTP material. The technical devices with which the process conditions are to be controlled during rubber forming were demonstrated in Chapter 3.

In the present chapter, it is investigated how the technical devices available during rubber forming, can be used to create an optimal processing environment. It is also an important goal to find out what theories and calculation methods are applicable to predict the history of the process parameters.

The following three sections of this chapter respectively discuss the process parameters temperature of the laminate, pressure and the application of additional forming forces during rubber forming, respectively. Calculations are compared with experimental results in each of those sections. For the calculation of the temperature history of laminates during rubber forming, a numerical computer program is used. In 4.5, the overall results of the investigations into the process parameters are discussed, whereas the most important conclusions are summarized in 4.6.

First, several rubber forming subprocesses are defined to facilitate the discussion of the process parameters. For this purpose, a typical temperature-pressure-time profile is presented in Fig. 4.1, where four process stages can be recognized:

Starting at point 0, the (preconsolidated) laminate is heated by a specific heating device until the processing temperature is reached at point 1. Next, the laminate is quickly transferred to the forming equipment.

The following subprocess is the forming phase of the laminate: At point 1, the rubber stamp is pressed against the hot laminate, forcing it into the rigid die. At point 2, the laminate touches the rigid die.

In the figure it is assumed that the mould is completely filled at point 2, the transition point, where the forming phase is supposed to end and the consolidation phase is started: The composite material is then pressurized to the consolidation pressure.

When the formed composite product is cooled below a certain temperature, the moulds are opened (point 3) and the CFRTP part is removed from the forming equipment: It will cool down further to room temperature (point 4).

This subdivision of the rubber forming cycle will be used throughout the present chapter, whenever relevant.
Optimisation of the rubber forming process parameters

Fig. 4.1 General rubber forming profile: laminate temperature and pressure during the process cycle.

4.2 Optimisation of heat flow during rubber forming.

The control of heat transfer during a thermoforming cycle is probably the most critical. In the preceding chapters, it was demonstrated that the following temperature history is ideal:

- during heating, low temperature gradients through the thickness and over the surface of the CFRTP laminate,
- deformation of the laminate sheet at a sufficiently high temperature,
- (re)consolidation of the laminate while its temperature is still sufficiently high,
- during cooling, low temperature gradients through and along the CFRTP laminate.

Furthermore, from an economic point of view, it is demanded that the heating and cooling of the laminate takes place as quickly as possible, with a minimum amount of energy.
In this section, numerical calculations of heating and cooling laminates are compared with experiments. The results give an indication of the effect on the heat transfer of technical changes in the thermoforming process, like infra-red heating intensity, material thickness, mould temperatures and insulation.

4.2.1 Theory of the heat transfer in laminates.

The composite material sheet has to be processed in a dynamic heat flow environment, which is inherent to the intention of developing the rubber forming technique as a rapid technique. There is not sufficient time to reach a steady state equilibrium inside the heating or cooling material. This implies the use of the unsteady state energy equation. When a thermoplastic laminate is presented in a system like the one in Fig. 4.2, the two-dimensional transient heat transfer can be described by the energy equation

\[
\frac{\delta}{\delta x} \left( k_x \cdot \frac{\delta T}{\delta x} \right) + \frac{\delta}{\delta y} \left( k_y \cdot \frac{\delta T}{\delta y} \right) + g = \rho \cdot C_p \cdot \frac{\delta T(x,y)}{\delta t} \tag{4.2.1.1}
\]

where:
- \( k_x \) is the thermal conductivity in the x-direction [W/mK],
- \( k_y \) is the thermal conductivity in the y-direction [W/mK],
- \( \rho \) is the density of the composite laminate [kg/m³],
- \( C_p \) is the heat capacity of the composite laminate [J/kgK],
- \( g \) is the heat generation due to chemical reactions or to crystallization [W/m³],
- \( T \) is the temperature [°C],
- \( t \) is the time [sec].

The solution of this energy equation is necessary to obtain information about the temperature profiles of the laminate during heating, forming, consolidation and cooling. Calculation of the heat transfer during each of these subprocesses requires different boundary conditions and starting conditions.

![Fig. 4.2 Schematic view of a laminate in a heat environment.](image-url)
It is assumed that the heat transfer through the laminate thickness is more important than the heat transfer in the plane of the laminate. In that case, the monitoring of the temperature can be limited to the most critical area of the laminate.

For an infinitesimal material part with no internal heat generation $g$ and constant thermal conductivity $k_x = k$, equation $<4.2.1.1>$ can be evaluated into the one-dimensional energy equation $^9$:

$$\frac{\partial^2 T(x,t)}{\partial x^2} = \frac{1}{\alpha} \frac{\partial T(x,t)}{\partial t}$$

$<4.2.1.2>$

where $\alpha = k/(\rho C_p)$ is called the thermal diffusivity $[m^2/s]$ of the composite material.

The boundary conditions (radiation, convection or conduction heat transfer) for each particular laminate part must be implemented in $<4.2.1.2>$. To obtain these boundary conditions, it must be noted that the total amount of energy at a boundary is constant:

heat flow into the composite = heat flux from the medium to the surface

As an example, for part A of a laminate during the forming phase (Fig. 4.3):

at $x = 0$, a free surface, a "convection" boundary is established:

$$-k \cdot \frac{\partial T(x,t)}{\partial x} \bigg|_{x=0} = h_A \cdot (T_A - T(0,t))$$

at $x=d$, with a constant rubber contact, a "conduction" boundary is established:

$$-k \cdot \frac{\partial T(x,t)}{\partial x} \bigg|_{x=d} = h_c \cdot (T_r(d,t) - T(d,t))$$

where $k$ is the thermal conductivity of the composite material,

$h_A$ is the heat transfer coefficient at the air-composite boundary,

$h_c$ is the interface conductance between rubber and composite,

$T_A$ is the air temperature,

$T_r(x,t)$ is the local and temporary rubber temperature,

$T(x,t)$ is the local and temporary composite temperature.

The determination of boundary conditions in case of radiation heating is discussed in Appendix A. Because of the complexity of the heat transfer and the large variety of boundary conditions, solutions of equation $<4.2.1.2>$ are obtained numerically in the present work. A so-called forward difference approximation is used, which is further explained in Appendix B.
Fig. 4.3 Change of boundary conditions on an infinitesimal part $A$ of a laminate during a rubber forming cycle.

Complications in the calculation of the heat transfer in a thermoplastic laminate are inherent to the composite nature of the material. In the literature\textsuperscript{72,73} the series model of the law of mixtures is often used to estimate the thermal conductivity $k$ of a voidless composite:

\[
k = \frac{1}{x'_{\nu_m} \frac{x'_{\nu_f}}{k_m} + \frac{x'_{\nu_f}}{k_f}}
\]

<4.2.1.3>
where \( k \) is the composite thermal conductivity through the thickness, 
\( k_m \) and \( k_f \) are matrix and fibre thermal conductivity, respectively, 
\( x_{vm} \) and \( x_{vf} \) are matrix and fibre volume fraction in the composite, respectively.

The comparison of heat flow calculations with this model for the thermal conductivity and experiments give satisfying results\(^{74,75}\).

Similarly, the heat capacity \( C_p \) of a composite material can be obtained by the mass-averaged heat capacities of the matrix and reinforcement\(^7\), that is:

\[
C_p = x_{vm} \cdot C_p_m + (1-x_{vm}) \cdot C_p_f
\]

<4.2.1.4>

where \( C_p \) is the composite’s heat capacity,
\( C_p_m \) and \( C_p_f \) are the matrix and the fibre heat capacity, respectively,
\( x_{vm} \) is the mass fraction of the matrix.

With <4.2.1.3> and <4.2.1.4>, the thermal diffusivity \( \alpha = k/(\rho_c \cdot C_p) \) of a composite material can be calculated. It is a measure for the rate of heat flow inside the composite. The higher \( \alpha \), the easier heat propagates through the material.

Other properties, which have to be measured from the material rather than calculated from theoretical predictions, are the interface conductances \( h \), between the laminate and several surrounding (mould) media and the heat transfer coefficient \( h_a \) for the calculation of heat transfer by convection.

The mentioned thermal properties are sensitive to material and processing conditions like void content of the laminate, processing pressure and also temperature. It is expected therefore, that the reliability of the numerical solutions is limited.

4.2.2 Setup of the heat transfer experiments.

Numerous temperature profiles of thermoplastic laminates have been measured during rubber forming cycles\(^{76-82}\). In this section, the most relevant temperature profiles will be discussed and compared to temperature profiles that are calculated using the software models of Appendix B. For the estimation of the most critical spots in/on the thermoplastic laminate several test setups are used. Two types of heat transfer experiments are discussed in this section: infrared heating experiments and cooling experiments.

The actual temperature measurements are carried out using thermocouples. The censored electrical signals from the thermocouples are monitored either by an in-house developed computer program, or by a LabWindows temperature registration software\(^8\). Experiments\(^8^1\) show that the response of the scanning setup is sufficiently fast for the temperature measurements in these investigations.
The CFRTP materials that are investigated in this section are glass fabric reinforced PolyEtherImide (G/PEI), carbon fabric reinforced PolyEtherImide (C/PEI) and glass fabric reinforced PolyEthyleneTerephthalate (G/PET) (Appendix C1). Their thermal properties, supplied by the material manufacturers, are listed in Table 4.1.

<table>
<thead>
<tr>
<th>Table 4.1 Thermal properties of the investigated composites:</th>
</tr>
</thead>
<tbody>
<tr>
<td>material</td>
</tr>
<tr>
<td>---------------</td>
</tr>
<tr>
<td>C/PEI</td>
</tr>
<tr>
<td>G/PEI</td>
</tr>
<tr>
<td>G/PET</td>
</tr>
</tbody>
</table>

For the infrared heating experiments, specimens are heated between the infrared heating panels of the TP1000 thermoforming facility of the laboratory. In Fig. 4.4, a schematic view is given of the heating panels, divided in specific zones. The heating zones can be activated separately.

![heating zone](image)

*Fig. 4.4 Subdivision in zones 1-4 of the IR-heating panels of the NHR134 thermoforming facility.*

In Table 4.2, an overview is given of the infra-red heating experiments that are presented in this section.
Table 4.2 Infra-red heating experiments:

<table>
<thead>
<tr>
<th>exp.</th>
<th>specimen material</th>
<th>thickness (no. layers)</th>
<th>heating zones</th>
<th>applied IR-power</th>
<th>calculation compared</th>
</tr>
</thead>
<tbody>
<tr>
<td>h1</td>
<td>C/PEI</td>
<td>2.45mm /8</td>
<td>1,2,3</td>
<td>90 %</td>
<td>yes</td>
</tr>
<tr>
<td>h2</td>
<td>C/PEI</td>
<td>2.45mm /8</td>
<td>1,2,3</td>
<td>100 %</td>
<td>yes</td>
</tr>
<tr>
<td>h3</td>
<td>G/PEI</td>
<td>1.00mm /4</td>
<td>1,3</td>
<td>90 %</td>
<td>yes</td>
</tr>
<tr>
<td>h4</td>
<td>G/PEI</td>
<td>1.00mm /4</td>
<td>1,3</td>
<td>100 %</td>
<td>yes</td>
</tr>
<tr>
<td>h5</td>
<td>G/PET</td>
<td>6.40mm</td>
<td>1,3</td>
<td>50 %</td>
<td>yes</td>
</tr>
<tr>
<td>h6</td>
<td>G/PET</td>
<td>6.40mm</td>
<td>1,2,3,4</td>
<td>50 %</td>
<td>yes</td>
</tr>
<tr>
<td>h7</td>
<td>G/PET</td>
<td>6.40mm</td>
<td>1,2,3,4</td>
<td>100 %</td>
<td>--</td>
</tr>
</tbody>
</table>

For the infrared heating measurements, thermocouples are consolidated in the centre layer of the laminate or they are evenly distributed through the thickness. The monitored section is situated in the middle of the laminate sheet. During the IR heating investigations, not only the kind of composite material is varied, but also the applied radiation power and/or the activated heating surface.

To investigate the cooling during rubber forming, two test setups are used (Fig. 4.5):

- In a test setup with a flat shaped mould, temperature measurements are carried out in hot laminate sheets, pressed by a rubber stamp against a flat mould. It is mainly used for the characterisation of the heat transfer in a laminate that is rubber formed with different mould materials or at different mould temperatures.

![Fig. 4.5 The Flat-type and the U-type test setup for heat transfer experiments.](image)
In the "U-type" test setup, the influence of the change in local boundary conditions during the various subprocesses in rubber forming is estimated. For this purpose a hot laminate is rubber formed in a U-shaped mould. Different locations in the plane of the laminate are scanned and critical material spots are pointed out. The shape of the U-shaped mould can be varied slightly.

In Table 4.3, an overview is given of the cooling experiments that are described in this section.

<table>
<thead>
<tr>
<th>exp.</th>
<th>specimen material</th>
<th>thickness (no.layers)</th>
<th>mould shape</th>
<th>mould temp. [°C]</th>
<th>insulation used</th>
<th>calculation compared</th>
</tr>
</thead>
<tbody>
<tr>
<td>c1</td>
<td>C/PEI</td>
<td>2.45mm /8</td>
<td>flat</td>
<td>variable</td>
<td>--</td>
<td>yes</td>
</tr>
<tr>
<td>c2</td>
<td>C/PEI</td>
<td>3.25mm/10</td>
<td>flat</td>
<td>20</td>
<td>variable</td>
<td>--</td>
</tr>
<tr>
<td>c3</td>
<td>G/PEI</td>
<td>2.45mm/10</td>
<td>U</td>
<td>20, 150</td>
<td>--</td>
<td>--</td>
</tr>
<tr>
<td>c4</td>
<td>G/PEI</td>
<td>1.65m/6</td>
<td>U</td>
<td>20</td>
<td>--</td>
<td>yes</td>
</tr>
</tbody>
</table>

- In the experiments c1, two thermocouples are placed between the outer layer and the adjacent layer, on both sides of the laminate (Fig. 4.6). The laminate is heated to 300°C and subsequently pressed between a PU rubber stamp and an aluminium plate. The aluminium plate is preheated to a certain "mould temperature" $T_d$, to investigate the influence of mould heating on the cooling rate of the laminate.

![thermocouples](image1)

**Fig. 4.6 Position of thermocouples in a laminate during the c1 cooling experiments.**

- In the experiments c2, the centre layer of the cooling laminates is monitored, while pressed in a flat mould. The aluminium mould is insulated, respectively with two and four PolyImide (PI) foils of 0.15 mm thickness and two and four PolyTetraFluoroEthene (PTFE) foils of the same thickness.
- In the experiments c3, the cooling is monitored of a laminate that is rubber formed into a hat shaped bar of 90 mm long, 70 mm wide and 70 mm high. In the centre layer of the laminate, nine thermocouples are placed so that they will be situated in the final product as depicted in Fig. 4.7. The thermoplastic laminate is positioned in an aluminium blankholder and heated in a hot platen press to 325°C. Then the hot blankholder with the laminate is transferred to the press unit for rubber forming the material into the U-shaped (steel) mould, which is at room temperature (20°C) or preheated to 150°C.

![laminate_with_thermocouples](image)

*Fig. 4.7 Position of thermocouples during the c3 cooling experiments.*

- In the experiments c4, a slightly different U-type mould is used as shown in Fig. 4.8. Thermocouples are consolidated between the centre layers at two different positions to investigate the influence of different boundary conditions over the surface of a laminate.
4.2.3 Results of the heat transfer experiments.

Heating results.

The results of IR heating of 2.45 mm thick C/PEI laminates are shown in Fig. 4.9 for 90% applied IR power (experiment h1) and in Fig. 4.10 for 100% IR power (experiment h2). The laminates reach an estimated process temperature of 280°C within 130 seconds and 105 seconds, respectively. In both cases, the calculated heating curves are somewhat higher than the measured ones.

The heating curves of 1.00 mm thick G/PEI laminates at 90% IR power (experiment h3) and 100% IR power (experiment h4) are shown in Fig. 4.11 and Fig. 4.12, respectively. The measurements hardly show a difference between the h3 and the h4 heating results. The rise in temperature for the G/PEI laminate that is heated at 100% power is overestimated most by the calculations.

The results of the IR heating of relatively thick (6.4 mm) G/PET laminates (experiment h5) are presented in Fig. 4.13. A low heating radiation level of 50% is applied and only two heating zones are activated. An estimated processing temperature of 255°C is not reached within 15 minutes. The comparison with the calculated heating curve shows a high discrepancy.
Fig. 4.9 Comparison between the measured and calculated temperatures of the IR-heating of a 2.45 mm thick C/PEI laminate (zones 1, 2 and 3 at 90% IR power).

Fig. 4.10 Comparison between the measured and calculated temperatures of the IR-heating of a 2.45 mm thick C/PEI laminate (zones 1, 2 and 3 at 100% IR power).
Fig. 4.11 Comparison between the measured and calculated temperatures of the IR-heating of a 1.00 mm thick G/PEI laminate (zones 1 and 3 at 90% IR power).

Fig. 4.12 Comparison between the measured and calculated temperatures of the IR-heating of a 1.00 mm thick G/PEI laminate (zones 1 and 3 at 100% IR power).
Fig. 4.13 Comparison between the measured and calculated temperatures of the IR-heating of a 6.4 mm thick G/PET laminate (zones 1 and 3 at 50% IR power).

Fig. 4.14 Comparison between the measured and calculated temperatures of the IR-heating of a 6.4 mm thick G/PET laminate (zones 1, 2, 3 and 4 at 50% and 100% IR power).
When a 6.4 mm thick G/PET laminate is heated by zones 1, 2, 3, 4 at 50% IR power (experiment h6) and 100% IR power (experiment h7), the laminate shows a behaviour as presented in Fig. 4.14. The calculated rise in temperature for the h6 laminate is an unusable overestimation. In the figure, it is also shown that the thick G/PET laminate reaches an estimated processing temperature of 255°C within 200 seconds when heated at full capacity.

Cooling results.

The output of the cooling measurements of a 2.45 mm thick C/PEI laminate (experiment c1) is presented in Figs. 4.15 and 4.16. From the figures, it can be seen that the difference in cooling rate between the rubber pressurized side and the aluminium pressurized side of the laminate is smaller if a high aluminium mould temperature is applied. However, the higher thermal conductivity of aluminium (k=200W/mK), compared to that of rubber (k≈0.012W/mK), is not completely compensated, even if the aluminium mould is preheated to 150°C.

The calculated temperatures of the rubber pressurized side and the aluminium pressurized side of a laminate, pressed in a mould at 20°C and 150°C, are shown again in Figs. 4.17a and 4.17b, respectively. Because the measured curves of both sides are closer to each other than the calculated curves, the calculation suggests a lower heat diffusion through the laminate than there is in reality.

In Fig. 4.18, the effect of insulation (experiments c2) is demonstrated. The cooling rate of a 3.25 mm thick C/PEI laminate is reduced most when the laminate is separated from the aluminium mould by a relatively thick layer of PolyImide (four foils of 0.15 mm).

From Fig. 4.19a, it can be noticed that the temperature distribution over a 2.45 mm thick G/PEI laminate is fairly non-uniform during rubber forming (experiments c3). The locally different boundary conditions of the spots in the deforming laminate impose a variation of material temperature in the "plane" of the laminate. In the figure, it can be noticed that at points 4 and 6, the local cooling rate is lowest (points 4 and 6 are located on the corners of the shaped profile). When the temperature of the aluminium mould is increased to 150°C, the variation of material temperature over the laminate is considerably reduced (Fig. 4.19b).

In Fig. 4.20, the results are shown of the cooling of a 1.65 mm thick G/PEI laminate (experiment c4). The monitored cooling curves for the two different locations in the laminate are compared with their calculated equivalents, using the proper boundary conditions. The calculated cooling rates closely approach the measured rates, for both laminate positions.
Fig. 4.15 Measured cooling curve on the rubber contacted side of 2.45 mm thick C/PEI laminates, pressed against aluminium moulds of different temperatures by a rubber stamp.

Fig. 4.16 Measured cooling curve on the aluminium contacted side of 2.45 mm thick C/PEI laminates, pressed against aluminium moulds of different temperatures by a rubber stamp.
Fig. 4.17a Measured cooling curves of 2.45 mm thick C/PEI laminates, pressed by a rubber stamp against an aluminium mould of 20°C.

Fig. 4.17b Measured cooling curves of 2.45 mm thick C/PEI laminates, pressed by a rubber stamp against an aluminium mould of 150°C.
Fig. 4.18 Measured cooling curves of 3.25 mm thick C/PEI laminates, pressed against insulated aluminium moulds by a rubber stamp.

Fig. 4.19a Temperature distribution of a 2.45 mm thick G/PEI laminate that is rubber formed into a hat shaped bar, in an aluminium mould of 20°C.
Fig. 4.19b Temperature distribution of a 2.45 mm thick G/PEI laminate that is rubber formed into a hat shaped bar, in an aluminium mould of 150°C.

Fig. 4.20 Cooling curves of a 1.65 mm thick G/PEI laminate that is rubber formed into a hat shaped bar, measured in the centre of the bar (T1) and at the side (T2).
4.2.4 Preliminary conclusions of the heat transfer experiments.

Some preliminary conclusions can be drawn from the total of the experiments to optimize the heat flow in a rubber forming cycle:

- Infrared heating can suitably provide the necessary heating rates for a fast thermoforming process like rubber forming.
- Insulation and/or low conductivity mould materials are needed to prevent high cooling rates while contacting the heated laminate.
- Increase of the mould temperature has a limited effect on the cooling rate of rubber formed laminates during the first few forming and consolidation seconds.
- Handling and forming speed have to be high to guarantee a sufficiently high temperature at the start of the consolidation phase.
- The infrared heating profile of a laminate can be calculated numerically. The predictions, however, overestimate the heating rate of the material.
- The cooling profile of laminates during a rubber forming cycle can be calculated rather accurately.

In 4.5, the results of the presented heat transfer experiments are discussed further and implicated in the general optimisation of rubber forming process parameters.

4.3 Optimisation of the pressurization.

The development of the rubber forming technique was started because of the potential of rubber moulds to impose a more favourable pressure distribution on the deforming and consolidating laminate. The pressurization requirements for a rubber forming mould are determined by the forming behaviour of the laminate and can be summarized from the preceding chapters:

- low pressure during the forming phase,
- uniform pressure distribution at the transition point,
- high pressure during the consolidation phase.

In this section, the results of measurements with in-house developed pressure sensors are presented. They are used to quantify the pressure profiles that are imposed by a rubber mould on a rigid mould at room temperature. The goal is to obtain a better control of the pressurization of the laminates by a better knowledge of the behaviour of pressurized rubber.
4.3.1 Theory of the pressurization by rubber moulds.

A vulcanized rubber is a crosslinked polymer that is in a rubbery state at room temperature. The possible elastic deformation is often extremely large (up to 600% is possible) and the resistance against deformation is relatively low. The elastic behaviour of a rubber can be described best by thermodynamic contemplations. The resistance of a rubber against deformation is a consequence of the distortion of intermolecular potential fields. This resistance results in so-called entropy elasticity. The elastic behaviour of rubber is therefore quite different from the elasticity of solid materials. However, to be able to calculate the necessary forces on a rubber for deformation, a theoretical modulus of elasticity $E$ is defined as $E = \sigma / \epsilon$. In Appendix D, a short overview is given of the theoretical determination of this modulus of elasticity.

An important drawback in using a modulus of elasticity in rubber deformation calculations is the temperature influence during thermoforming. The temperature of the rubber moulds is often above room temperature and non-uniformly distributed in the mould. The "stiffness" of rubber strongly increases with temperature. Equation $<D.1>$ from Appendix D gives an indication of the influence of temperature on the rubber deformation behaviour.

One of the properties of rubber that is often referred to is its "hardness". This hardness can be determined for instance by a Shore A measurement (ASTM D2240): A conical body is pressed against the rubber surface with a specified force and the depth of indentation after 1 second is registered. The Shore A hardness (0-100) can be measured by a small hand-held device and is accurate within 5% only. The IRHD (International Rubber Hardness Degree) is also measured by the indentation of the rubber surface (IRHD degree = Shore A point ± 1). Since the measurement of rubber hardness is really a measurement of the resistance against deformation, the hardness is directly related to the elasticity of rubber.

The mentioned drawbacks on the determination of the modulus of elasticity make calculations of the pressure in a rubber forming process extremely difficult. It is therefore necessary to obtain real time measured pressure profiles on the moulds.

The Young’s modulus $E_0$ of rubber can only be obtained from the linear part of a load-deflection curve of a pure homogenous compression of a rubber block. The real deformation behaviour of rubber moulds however, is also largely dependent on the shape of the rubber and the rigid moulds. During the compression of a rubber block that is prevented from slipping at the loading surfaces, any decrease in the thickness of the block is accompanied by "barrelling", the bulging of the rubber at the force-free surfaces (Fig. 4.21). This transforms the internal stress in the block from simple uniaxial compression to a combination of shear and compression. It will cause the rubber block to appear stiffer than the Young’s modulus suggests.
Fig. 4.21 Schematic presentation of the "barreling" of a pressurized rubber block.

The apparent compression modulus $E_c$ of bonded rubber blocks with rectangular or circular cross-section, can be described by:

$$E_c = E_0 (1 + 2kS^2) \quad \text{<4.3.2.1>}$$

For long strips of rubber under compression, with negligible strain along the length:

$$E_c = \frac{4E_0 (1 + kS^2)}{3} \quad \text{<4.3.2.2>}$$

$k$ is a numerical factor, decreasing with increasing rubber hardness ($\frac{1}{2} < k < 1$). $S$ is the so-called "shape factor" which is defined as (Fig. 4.22):

$$S = \frac{\text{area of the loaded surface}}{\text{total force free area}}$$

**S = A1/2(A2 + A3)**

Fig. 4.22 Definition of the shape-factor $S$ of a pressurized rubber block.

Concerning the use of rubber moulds in the rubber forming technique, the shape factor is especially important when non-matching rubber dies are used. The larger the force free areas of a rubber stamp, the less stiff the stamp will appear during the pressurization of a
laminate. Such a stamp will deform more before filling the rigid mould. A high restriction on the barrelling freedom of a rubber stamp during compression, gives a high apparent stiffness. The shape factor S can be useful in the design of rubber stamps.

For a rubber block with a high shape factor, $E_c$ tends towards the bulk compression modulus $E_\infty$:

$$\frac{1}{E_c} = \frac{1}{E_0(1+2kS^2)} + \frac{1}{E_\infty}$$

The bulk compression modulus of rubber $E_\infty$ (1000-2000 MN/m$^2$) is many times larger than the Young's modulus, which means that rubber hardly changes in volume compared to its geometrical deformation.

When a rubber stamp has completely filled a rigid negative mould, no stamp deformations are possible any more. In such a situation it is assumed that a further increase of pressure on top of the rubber stamp will give an additional pressure increase that is uniformly distributed over all the surfaces of the rubber stamp. This rubber feature is most important for the consolidation of a thermoplastic laminate when a sufficiently high surface pressure has to be applied on the composite material. The optimisation of a rubber stamp pressurization in a rigid mould can be carried out by designing the flexible stamp according to the shape-factor theory. The "matching" rubber stamp has to be designed in a way that the sides of the rubber stamp touch the mould sides only then when the bottom of the rubber stamp has completely contacted the bottom of the rigid mould. The design of rubber stamps is discussed in more detail in Appendix E. The expected uniform pressurization of the rubber stamp beyond the transition from forming to consolidation has to be verified by measurements.

### 4.3.2 Setup of pressure experiments with U-shaped moulds.

In this section only positive rubber moulds, i.e. stamps are considered because the use of negative rubber moulds is not advised for three main reasons. First, the pressure distribution is difficult to control in a rubber forming setup with a rigid stamp and negative rubber mould. Second, generally the surface quality of the outside of the composite product has to be high, which can only be imposed by a rigid mould. Third, the high heat conductance of a metal stamp will impose a high cooling rate on the deforming laminate, because the naturally high rubber insulation is not exploited.

Pressure distributions are presented in Section 4.3.3, measured in a U-shaped mould test setup consisting of a rectangular rubber stamp and a matching steel negative mould (Fig. 4.23)\(^{92-94}\). Pressure sensors are integrated in the surface of the steel mould (Fig. 4.24). The sensors are developed in-house and are based on the measurement of the bending deflection
of a cantilever supported small steel plate (10x5x2mm³). Because the deflection of the plate is very sensitive to its internal temperature distribution, all the pressure measurements presented in this section are carried out at room temperature without using a laminate, or using a fake laminate*. The deflection of the small steel plate is registered by a strain gage that measures the elongation of the lower side of the plate. The test setup is placed in a 100kN MTS test bench, which presses the rubber stamp into the steel mould. The sensors are scanned by a computer and transformed into pressure data, graphically presented on screen.

Fig. 4.23 Test setup for pressure measurements in a U-shaped steel mould, pressurized by a rubber stamp.

Fig. 4.24 Cross-section of a pressure sensor in the surface of a steel mould.

* The fake laminate as used in the experiments is a fabric reinforced Kelprox™ rubber sheet at room temperature.
In Table 4.4, an overview is given of the pressure experiments that are carried out.

<table>
<thead>
<tr>
<th>experiment</th>
<th>investigated parameter</th>
<th>rubber hardness [Shore A]</th>
</tr>
</thead>
<tbody>
<tr>
<td>p1</td>
<td>width to height ratio of the stamp</td>
<td>90</td>
</tr>
<tr>
<td>p2</td>
<td>tolerance of the rigid and rubber mould</td>
<td>60, 78 and 90</td>
</tr>
<tr>
<td>p3</td>
<td>laminate tension</td>
<td>78</td>
</tr>
</tbody>
</table>

- In **experiment p1**, a stamp of 90 Shore A PolyUrethane (PU) rubber is pressurized in a steel mould. The stamp has a clearance of 0.5 mm on every side in the steel mould. The width of the steel cavity and the matching rubber stamp can be varied to investigate the influence of the width to height ratio of the moulds. Cross-sections of the test setup, including the placement of the sensors, are illustrated in Fig. 4.25. The average pressure (i.e. the load applied on the top surface of the rubber stamp, divided by the top surface) is increased from 0 MPa to 10 MPa.

- In **experiment p2**, a fixed geometry of a 50 x 50 x 100 mm³ (width x height x length) silicone rubber stamp is used to form a 1.35 mm thick flexible fake laminate in a rigid steel mould with variable internal width:

  width of 52.75 mm (denoted "narrow mould");
  width of 53.25 mm (denoted "fitting mould");
  width of 54.25 mm (denoted "wide mould").

The hardness of the rubber stamp is chosen between 60, 78 and 90 Shore A.

- In **experiment p3**, test setup B from Fig. 4.25 is used for "rubber forming" of the fake laminate under tensile load. The tensile load on the laminate simulates blankholding forces as they are applied during a real rubber forming cycle. The imposed tension in the laminate has to be counteracted by the rubber stamp. In this case the hardness of 78 Shore A is used.
Fig. 4.25 Cross-sections of test setups for pressure experiments, mould geometries A, B, C.
4.3.3 Results of the pressure experiments.

The results presented in Figs. 4.26a,b,c give the influence of the width to height ratio of a rubber stamp (experiment p1). The general shape of the pressure profiles is similar in all graphs: the local pressure at the top and bottom corner of the side of the moulds is relatively low compared to the pressure at the middle of the side and the bottom. With increasing shape factor (increasing width), the measured local pressure at the mould sides and the bottom increases (at a specific applied pressure). Note that the relative pressure profiles are the same with increasing applied pressure: above a certain applied pressure, the stamp has completely filled the steel moulds.

In Figs. 4.27a,b,c, the results of experiment p2 are presented: the tolerance between the rubber stamp and the rigid mould is varied. It is noticed that when a rubber stamp is allowed more "space" in a mould, more pressure is absorbed by the barrelling effect of the rubber. The censored peak pressures (measured at the middle of the sides) are the lowest in the wide mould, especially for the hardest rubber (90 Shore A). The use of the 60 Shore A rubber results in relatively low corner pressures compared to the peak pressures, especially in the narrow mould. Apparently, friction between the mould sides and the barrelling rubber stamp lowers the pressure at the corner of the mould.

In Fig. 4.28, the influence of tension in the laminate on the pressure distribution in the mould is given (experiment p3). Because of the symmetry, only one half of the pressure profiles is presented in the figure. It can be recognized that the application of tension lowers the pressure in the corner of the mould (sensor 5). The other sensors do not measure any influence of the tension in the laminate.

(Fig. 4.26a)
Fig. 4.26 Results of the pressure measurements during the pressurization of moulds A, B, and C.
Optimisation of the rubber forming process parameters

(Fig. 4.27)
Fig. 4.27 Results of pressure measurements during the pressurization of a fake laminate in mould B, by a rubber stamp with hardness of 60 Shore A, 78 Shore A and 90 Shore A.

Fig. 4.28 Results of pressure measurements during the pressurization of a tensioned fake laminate in mould B by a rubber stamp.
4.3.4 Preliminary conclusions of the pressure experiments.

Some preliminary conclusions can be drawn from the pressure experiments to optimize the pressurization of a laminate during rubber forming:

- The use of rubber in a stamp enables the pressurization of a deforming laminate even at vertical sides of a mould.
- Once the rubber stamp has filled the mould completely, any further increase of the load on the stamp results in identical increases of the pressure on every spot of the laminate.
- If a rubber stamp is allowed to have much barreling deformation, the pressure, transferred to the laminate, will be relatively low. In general, a mould with a high shape factor $S$ can impose a higher overall pressure.
- A relatively soft rubber stamp can easily get stuck at the sides of the rigid mould, due to its high tendency for barreling deformation. This results in high friction on the mould sides and consequently in relatively low pressure on the corners of the rigid mould.
- Tension in a laminate, for instance due to blankholding forces, will deform the rubber stamp before it touches the rigid mould. This stamp deformation results in a lower pressure on the corners of the rigid mould when the mould is completely filled.

The influence of the rubber block parameters Young's modulus $E_g$ and shape factor $S$, can be implemented in a stamp by the choice of rubber hardness, the geometry or a combination of both. In 4.5, the results of the presented pressure experiments are discussed in relation to the general optimisation of rubber forming process parameters.

4.4 The application of external forming forces.

The application of external forming forces is a powerful tool for the control of laminate deformation and the inherent fibre movement. In Chapter 2, the effects of external forces on the behaviour of fabric reinforced thermoplastics are explained. To approach an ideal forming situation, external forming forces have to be imposed by technical means. The external forming forces are sometimes referred to as "blankholder forces".

In Section 4.4.1, the influence of additional forming forces to prevent wrinkling of fabric reinforced thermoplastics during interply slip is discussed. An illustration of the use of the Drape shearing simulation (version 2.0)\textsuperscript{30-31} is given in Section 4.4.2, to determine the geometrical effect of external forming forces on the shearing behaviour of fabrics.

In Section 4.4.3, the technical devices are presented with which it is proven to be possible to introduce external forming forces into the deforming composite material sheet. The use of friction and clamping frames to impose fabric deformation is illustrated with some examples.
4.4.1 Prevention of interply slip wrinkling.

The bending of a layered composite material in a thermoforming process is accompanied by the slipping of the reinforcement layers relatively to each other, as was already discussed in 2.3. The internal ply stresses for the interply slip deformation usually are compression at the inner side and tension at the other side, as illustrated in Fig. 4.29.

![Diagram of internal stresses in a laminate during thermoforming](image1)

*Fig. 4.29 Development of internal stresses in a laminate that is bent during thermoforming.*

During thermoforming the long and slender fibres are surrounded by soft matrix only. They therefore tend to buckle in compression. In Fig. 4.30 a picture is presented that shows compression induced interply slip wrinkling at the inner layer. At the sharp corners, the mould contact provides a certain stability of the compressed layer, preventing buckling. Aside of the places of mould contact, the compression forces in the layer can cause buckling of the ply.

![Image of laminate wrinkling](image2)

*Fig. 4.30 Laminate wrinkling caused by compressed layers due to insufficient interply slip.*
In Fig. 4.31, a typical, calculated stress distribution is presented for a pure interply slip deformation. In reality, however, interply slip results in higher tensile stresses in the outside layer and lower compression stresses in the inside layer of the curved laminate, compared to the presented theoretical results. This is due to the introduction of a general tension stress throughout the laminate during the forming process. By the application of additional forming forces, it is possible to increase this general stress level inside the laminate to a value where the inside layer is not in compression any more.

![Stress Evolution](image1)

**Fig. 4.31 Stress distribution through a bent, hot laminate. The graph is calculated by the Formsim software**.

The time in which a complete bending deformation of a hot laminate is completed is important for the generation of compressive stresses in the inner ply. The bending of a sharp 90° angle by a square rubber stamp imposes a high deformation rate (high $\alpha$) with a severe stress distribution through the laminate. In that case, wrinkling can only be avoided by imposing high blankholding forces.

![Curves A and B](image2)

**Fig. 4.32 Cross-section of a rubber formed product with a gentle curvature (A) and a sharp curvature (B).**
In Fig. 4.32 for instance, the bending of the laminate at A is less critical for wrinkles than the more rapid bending at B. Therefore, the deformation at B has to be considered to determine the blankholding force, necessary to avoid fibre buckling. However, the temperature distribution over the deforming laminate is also important: when the deformation at side A is performed at lower temperature than the B-curve, side A can become the most wrinkling sensitive.

It is important to minimize the necessary blankholding forces, because high laminate tension during rubber forming:
- disturbs the fibre distribution through the laminate,
- introduces residual internal stresses in the final product.

![Image](image_url)  
*Fig. 4.33 Imposing of laminate tension by the pressurization of the laminate during rubber forming.*

Tensile forces in the layers of the fibre reinforcement inside the laminate are counteracted by surface pressure on the composite sheet, as shown in Fig. 4.33. A pressure on the soft matrix interlayer will be created that can force the hot matrix through the underlying fabric reinforcement to the lower (pressure-free) surface. This "pressing out" of the matrix can be noticed best on sharp curves in the product, where a high resultant pressure is present. It will be revealed as small resin beads at the outer arc surface (Fig. 4.34). Although the surface quality can be restored during the consolidation, the fibre distribution at the curved section of the product is disturbed (Fig. 4.35).

The generation of compressive stresses in the inner ply of the curve is discussed in 2.5. A modification of the Formsim software program, introduced in that section, could be used as a tool to calculate the necessary blankholder force to avoid interply slip wrinkling. In Fig. 4.36, an example is given of the determination of the necessary blankholder force that a clamping point must generate in the illustrated rubber forming setup.
Fig. 4.34 Resin beads at the outside of a sharp curvature, imposed by a high resultant pressure at the inside of the curvature.

Fig. 4.35 Disturbance of the fibre distribution at a curvature due to tensile stresses in the fabric reinforcement.
4.4.2 Prevention of intraply shear wrinkling.

In 2.4, the intraply shearing is discussed as a deformation mode that can be used to shape fabric reinforced thermoplastic laminates into relatively complex composite parts. To realize the product fabrication however, the necessary shearing deformation has to be imposed and unwanted fabric wrinkling or tearing has to be avoided. In this section, it will be pointed out how and where external forming forces have to be applied in practice in order to control the intraply shearing behaviour of a CFRTP laminate during rubber forming.

Preliminary experiments on rubber forming of composite products where intraply shearing is involved showed the existence of two main wrinkling modes:

a) buckling of fibres at spots in the laminate that are compressed because of insufficient shearing elsewhere in the material,
b) wrinkling of fabric at spots in the laminate where the maximum shearing angle is exceeded.

In Fig. 4.37, the different locations of the two wrinkling modes are depicted. Mode a) wrinkles can be observed perpendicular to the buckled fibres. Mode b) wrinkling occurs at highly sheared laminate places. The mode b) wrinkles run parallel to the diagonal of the diamond patterns formed by the sheared fabric.

The application of external forming forces, provided by blankholding devices, can result in different ways of preventing material wrinkling:

○ Holding of the laminate edges during the forming process will avoid that more composite material is being drawn into the mould than is needed. In this way, the compression of possible excess CFRTP material in the mould is prevented. In combination with the enforcing of shearing, mode a) buckling is avoided.
Fig. 4.37 Photographs of a product with two kinds of intraply shear wrinkling: (top photo) buckling of fibres due to compression and (bottom photo) wrinkling of the fabric due to insufficient intraply shearing.
○ Tension stress caused by local clamping of the material will give the reinforcing fabric a higher degree of stability as is investigated in Section 2.4.3. Although the necessary shearing forces are also enlarged, a higher maximum shearing angle can be obtained. Mode b) wrinkling is postponed.
○ Blankholding devices can direct excess composite material away from mould cavities where a reduction of the original material sheet surface is necessary. This action can avoid the local compression of material and will lower the necessary shearing angle. Both mode a) and mode b) wrinkling are prevented.

To identify the positions of the clamping points that are needed on the laminate sheet surface, the already introduced Drape computer program can be used. Drape offers information about the shearing angles that have to be realized to cover a mould surface with a fabric. It can therefore help to identify product areas that are sensitive to fabric wrinkling. The shearing limits of the specific fabric reinforced thermoplastic material have to be known. Drape 2.0 can give an approximation of the geometrical and physical effect of locally applied external forces on the reinforcing fabric. It can indicate compressed areas of the product laminate which are sensitive for fibre buckling. The examples, as they are discussed in the following, give an indication of the possibility of the prevention of intraply shear wrinkling by the application of external forming forces. They demonstrate the use of Drape as a tool for problem shooting and solving.

![Fig. 4.38 Illustration of wrinkle sensitive areas in a fabric reinforced bicycle helmet.](image-url)
In Fig. 4.38, the mode a) and b) sensitive areas of a rubber formed bicycle helmet are pointed out. The application of fixed holding points at the corners of the composite material introduces additional tensile forces in the material. These forces cause the stretching of the material at mode a) critical positions and the increase of shearing forces at mode b) critical positions. From the applied grid which indicates the fibre directions, it can be noted that the clamping points at the corners of the sheet are positioned correctly to tension the fibres running along the edge. In this way, compression of those fibres is avoided there.

The possible occurrence of intraply shear wrinkles also depends on the kind and direction of the fibre reinforcement that is used in the product (Chapter 2). Influencing the shearing behaviour of a quasi-isotropic laminate, consisting of alternating 45º-45º and 0º/90º fabric layers, by means of external forming forces is technically difficult. The differently orientated layers require different blankholder forces which should not influence each other.

In Fig. 4.39, the shape of a composite car rim is illustrated⁹⁵. The rim is to be rubber formed out in two parts out of a quasi-isotropic laminate of approximately 6 mm thick G/PET material. To control the shearing deformation during rubber forming, two differently orientated fabric layers have to be loaded in tension. Because of the resemblance of the parts to a hemisphere, a preliminary investigation of a quasi-isotropic hemisphere is used to estimate the appropriate blankholder forces for the car rim manufacturing.

Fig. 4.39 Rubber formed car rim out of a quasi-isotropic G/PET laminate.
Fig. 4.40 DRAPE simulation of the local fibre orientations after shaping a hemispherical part out of a fabric.

In Fig. 4.40, the Drape simulation is presented of a hemispherical part that is clamped at the four corners of an originally rectangular (0°/90°) fabric reinforced laminate. Because the reinforcement in the quasi-isotropic hemisphere also contains 45°/-45° fabric layers, additional forming forces have to be introduced into these layers too. The laminate edges around the product area must be used to control the shearing deformation of the independent layers (Fig. 4.41).

However, the starting sheet for the car rim production is rectangular and does not offer separate free edges of the differently orientated layers. The blankholder forces on this laminate therefore have to be introduced in such a way that they give the same net shearing deformation at the critical spots of all the layers. To accomplish this, the positions of the blankholding points have to be chosen so that the resultant of the blankholder forces in the quasi-isotropic sheet approaches the resultants in the separate layers as much as possible (Fig. 4.42).

In Fig. 4.43, the result is given of a Drape simulation for a deformation with re-positioned clamping points. The re-positioning enables the use of the same blankholding positions for both orientations of the fabric layers. Additional experiments prove that the differently oriented layers hardly influence each others shearing deformation. Though the force distribution has changed somewhat theoretically, in practice wrinkling is avoided during the rubber forming of the product.
Fig. 4.41 Necessary introduction of tensile forces in a fabric reinforced laminate (0/90° and ±45° layers) to stimulate intraply shearing during the forming of a hemisphere.

Fig. 4.42 Alternative positions of the blankholding points in a fabric reinforced laminate (0/90° and ±45° layers) to enable the rubber forming of a car rim.
4.4.3 Technical realization of forming forces.

In general, it is technically impossible to apply the necessary tensile forces directly in the most suitable fibre bundles. Mostly, the only way to impose a certain directional forming force in the laminate is the clamping of the laminate sheet on a surface spot that has been chosen in advance. The clamping of the material together with the downstroke of the punch into the mould results in tensile stresses in certain directions.

In Fig. 4.44 is shown schematically how a tension is imposed from the mould edges into the deforming laminate. The tension will affect the whole laminate. In the figure, a fixed pin that pierces the material sheet is schematically presented. The generated tension in the laminate (F_lam) cannot be controlled (Fig. 4.45).

Another possibility is the use of friction between the laminate surface and the mould edge, as illustrated in Fig. 4.46. The necessary force F can be provided by the closing press or by a mould independent device that pressurizes the edge of the laminate before the stamp touches its surface. The friction on the laminate edge, however, can be varied only slightly with the level of the pressing force and the mould surface that contacts the laminate.

The use of friction surfaces or fixed pins, the so-called "passive blankholding", is not always suitable to generate additional forming forces. Neither can the direction and extent of the blankholding forces be accurately controlled by this passive blankholding.

To prevent wrinkling during the forming of a product that is deformed by interply slip only, an overall tension in the laminate, externally imposed by passive blankholding usually is sufficient. To prevent intraply shearing wrinkling during the shaping of more complex product forms, it is often necessary to direct the blankholder forces in an other direction.
Fig. 4.44 Introduction of tension loads by piercing pins in a deforming laminate during rubber forming.

Fig. 4.45 Tearing of the laminate due to the uncontrolled introduction of tensile loads during thermoforming.
Fig. 4.46 Introduction of tension loads by clamping of a laminate during rubber forming.

An example of the so-called "controlled blankholder concept" is presented in Fig. 4.47. It consists of a blankholder plate with clamping points or pins that are attached to an undercarriage that can slide through a slot in the plate. The sliding resistance of the undercarriage can be adjusted by mechanical (friction, spring) or hydraulical means. In a rubber forming cycle, the heated laminate sheet is placed over the mould and blankholder plate. Then, it is gripped by the clamping points. When the upper mould comes down, the laminate edges are forced to follow the movement of the undercarriages.

Fig. 4.47 Presentation of the controlled blankholder concept.
This method of applying external forming forces offers several advantages over the fixed blankholding setup:

- the level of external forming forces on the laminate can be adjusted by the sliding resistance of the undercarriage,
- during the shaping of the product, the direction of the external forming forces can be controlled by the track of the undercarriage,
- due to the absence of material tearing, the need for excess composite sheet material at the edges of the product is reduced, which will considerably lower the amount of scrap material.

Fig. 4.48 illustrates the results of active blankholding of a fabric reinforcement for the presented car rim.

Fig. 4.48 Results of controlled blankholding of a glass fabric during the forming of a car rim half.

4.5 General discussion.

The results from the temperature experiments from Section 4.2.3 and the pressure experiments from Section 4.3.3 are discussed in the context of the optimisation of the process parameters during a rubber forming cycle.
Optimisation of the temperature.

The infra-red heating of CFRTP materials is suitable for providing a sufficiently high cooling rate of a laminate. The developed software (Appendix B) can be used to calculate the setting of the heating equipment that is necessary to obtain a specific heating curve for a specific laminate. The resemblance between the measured heating curve and the calculated heating curve is best for the C/PEI laminate, heated at 100% IR power (Fig. 4.10). In general however, the calculated heating curve overestimates the real heating rate, especially in the case of relatively thick G/PET laminates (Figs. 4.13 and 4.14). Another discrepancy is that the calculated temperature distribution through the thickness of the laminate is more uniform than in reality, which means that the calculated heat flow through the thickness of the laminate is too high (Fig. 4.14). These discrepancies probably have multiple causes:

- the real IR heating power is lower than the power used in the calculations;
- the environment has an important influence on the heating rate of a laminate: possible air displacements over the laminate surface during the measurements and divergence of the IR radiation cause boundary conditions different from the ones used in the calculations;
- the models to calculate the thermal properties of the composite materials are not sufficiently realistic;
- because the laminate is not pressurized when heated, possible deconsolidation of the laminate can locally separate the layers which will obstruct internal heat conduction.

The cooling of the layers in a laminate in a mould can be predicted fairly well by the developed computer software. The influence of different boundary conditions during rubber forming are calculated with an acceptable degree of accuracy. This is for instance shown for the case of mould preheating (Figs. 4.17a,b) and for the case of locally different boundary conditions over the surface of the laminate (Fig. 4.20). The measured heat transfer through the thickness of a laminate is somewhat higher than the calculated one, as can be concluded from the smaller temperature differences between the rubber pressurized side and the aluminium pressurized side of the cooling laminates (Figs. 4.17a,b). The high heat transfer through the thickness is probably caused by the intimate contact of the pressurized layers in the laminate.

From the cooling measurements, presented in Figs. 4.15 and 4.16, the influence of the low conductivity of rubber compared to that of aluminium can be noticed. The preheating of the aluminium mould has to be intensive to lower the cooling rate of the laminate during the first few seconds. Mould preheating, however, will imply higher cycle times. Coating of the rigid mould by insulating materials like Polylimide (Fig. 4.18) is an economically more favourable solution.

Optimisation of the pressure.

The pressure measurements show that the use of a matching rubber stamp enables the homogeneous pressurization of a CFRTP product. Vertical sides of the mould can also be
pressurized in that way. If much deformation of a stamp is required to fill the mould (wide mould) and the stamp has a high deformation resistance (high hardness), the overall pressure on the product itself will be lower (Figs. 4.27). Because a non-matching rubber stamp "absorbs" pressure for its own deformation, the top loading of such a stamp by a press must be relatively high to pressurize a laminate in every spot of a product. Relatively massive press machinery is required, for example, for the so-called pressing technique, where a large flat rubber pad is used as a "stamp" (see 3.2). Consequently, when non-matching stamps are necessary to enable the forming of a difficult product shape, in general soft rubber has to be used to minimize pressure differences over the product area.

The required (local) deformation of the rubber stamp, to fill the rigid mould, determines the pressure distribution over the product. This pressure distribution, raised during the forming phase, will stay present during the consolidation phase. When the top loads on the stamp are increased and the mould is completely filled, the absolute pressure increase will be equivalent all over the product as is shown by Figs. 4.26.

The optimisation of the pressure distribution over the product in the mould necessitates a good stamp design. Up to now, a quantitative design method has not been found, although the use of shape factor calculations is promising (Appendix E). Obviously, the geometry and the rubber hardness of a rubber forming stamp are important design variables.

Because external tension-loading of a laminate can cause premature deformation of the rubber stamp (barrelling), additional forming forces must be minimized. Laminate tension will obstruct the pressurization of product details (Fig. 4.28). In 4.4, however, the need for external forming forces during thermoforming was emphasized to optimize the shaping of a CFRTP product. This need is strongly related to the temperature and pressure of a laminate during rubber forming, as demonstrated in Chapter 2.

4.6 Conclusions.

It has been demonstrated in this chapter, that the control of the rubber forming parameters temperature, pressure and external forming forces is technically possible. The rubber forming technique enables effective heating, forming and consolidation of CFRTP materials.

Infra-red heating power, radiation area, mould preheating and insulation are variables for the optimisation of heat flow during the rubber forming of laminate sheets. The influences of these variables on the temperature history of a laminate can be numerically calculated, using a one-dimensional energy equation. In general, the results of the calculations show relatively good resemblance to those of in-situ temperature measurements. The definition of the right boundary conditions for the calculations is rather difficult, however.
Geometry and hardness of the rubber stamp are the main variables for the optimisation of the pressurization in a rubber forming mould. A homogeneous pressurization of a CFRTP product in a mould is only possible if these variables are handled carefully. No directly applicable theory has been found suitable for the calculation of the shape and hardness of a rubber stamp to obtain an optimal pressure distribution and history. So-called "shape factor" considerations however, are believed to give useful, qualitative, information for rubber mould design.

The application of external forming forces on the laminate can be useful during thermoforming, to further optimize the shaping of a thermoplastic composite product. In some specific cases, the Drape simulation software can be helpful to determine the optimal external force introduction.
CHAPTER 5. Material response to rubber forming.

5.1 Introduction.

The rubber forming technique is set up as an efficient and fast technique to exploit the processing benefits of thermoplastic composites. The low cycle times of rubber forming are economical demands, however, and do not always offer optimal thermoforming conditions: The subprocesses heating, forming, consolidation and cooling are often performed in a way that the final product cannot achieve the highest possible (mechanical) properties. It is important, therefore, to gain more knowledge about the influence of specific rubber forming phenomena. The response of the CFRTP material to these phenomena must be known to be able to predict the quality of a product that is rubber formed at given conditions. In this way, it is possible to set the demanded product properties against the necessary processing conditions for every particular product application.

In 5.2, it is pointed out that the consolidation of a CFRTP part in the rubber forming process is not carried out in an isothermal environment. Consolidation theories, which have been developed for the use in isothermal and isobaric forming techniques, are reviewed therefore. Experimental work on the consolidation of fabric reinforced PEI laminates is discussed and a proposal for the prediction of the mechanical properties of rubber formed laminates is presented.

The high cooling rate at the laminate surface is another implication of rubber forming. Internal stresses can be raised, when the heated material is pressed against cold moulds, because of the sudden change of surface temperature of the laminate. The consequences of these stresses are discussed in 5.3: microcracks and geometrical distortions.

The forming forces applied in rubber forming can cause a significant change of the mechanical properties of composite materials like carbon- and glass fabric reinforced PEI laminates. The mechanical influence of forming forces is experimentally demonstrated in 5.4.
In 5.5, the main effects of rubber forming subprocesses on the composite material are summarized.

5.2 The influence of non-isothermal consolidation.

The processing of composite products that consist of layered material includes the consolidation of the material[25,26,96,97]; the fusion of the individual layers to one homogeneous laminate (Fig. 5.1).
Fig. 5.1 Schematic presentation of the consolidation process of laminate layers.

Sometimes only a stack of prepregs or commingled fabric\textsuperscript{24,98} is the starting material in the rubber forming process, but mostly a fully consolidated sheet is used. In both cases, however, the connection of the layers inside the laminate is absent during the forming process. (Re)consolidation must be carried out at the end of the forming phase, therefore. The point of time where the forming phase ends (no deformation possible any more) and the consolidation phase starts is called the "transition point".
In this section, consolidation experiments with 8-layered C/PEI laminates are used to describe the influence of non-isothermal consolidation.

5.2.1 Previously published work on consolidation.

Dara and Loos\textsuperscript{86} introduced a consolidation model in which the consolidation of the layers in a laminate is modelled by two phenomena, intimate contact and autohesion. First, the adjacent ply surfaces coalesce and have to realize "intimate contact", then the interfaces between the layers disappear by "autohesion", caused by diffusion of the polymer chains.

Lee and Springer\textsuperscript{25} further developed the consolidation model and represented the irregular ply surface by a surface consisting of a series of thermoplastic matrix rectangles (Fig. 5.2). The degree of intimate contact $D_i$ between two layers is defined as:

$$D_i = \frac{b}{w_0 + b_0}$$

\text{<5.2.1.1>}

where $b_0$ and $b$ are the initial ($t=0$) and instantaneous ($t=t$) widths of each rectangular element, respectively, and $w_0$ is the initial distance between two neighbouring rectangles.
During consolidation, flow is assumed laminar and by applying the law of conservation of mass, an expression is gained for the time required to complete intimate contact ($D_e = 1$):

$$t_{ic} = \frac{\mu_{mf}}{5p_{app}(1 + w_0/b_0)} \cdot \frac{b_0}{a_0} \cdot (1 + \frac{w_0}{b_0})^5 - 1$$

where $\mu_{mf}$ is the viscosity of the fibre-matrix mixture,

$a_0$ is the initial height of a rectangular element,

$w_0$ is the distance between two surface elements,

$p_{app}$ is the applied surface pressure.

As soon as two adjacent layers realize intimate contact, the autohesion phenomenon is started and the borderlines between the polymer surfaces are crossed by the diffusion of molecular chains (Fig. 5.3). Finally, this diffusion results in one homogeneous matrix interlayer.

**Fig. 5.3 Illustration of the autohesion process of polymer matrices.**
According to Dara and Loos, the degree of autohesion $D_{au}$ can be estimated by testing the bond strength between two plies. It is presented by the ratio of the realized bond strength $S$ and the ultimate bond strength $S_u$, $D_{au} = S/S_u$. The autohesion degree is approximated for amorphous polymers by:

$$D_{au} = C \cdot t_a^{0.25} \quad <5.2.1.3>$$

where $t_a$ is the time elapsed from the start of the autohesion process. $C$ is a constant that is related to the temperature by the Arrhenius relation:

$$C = C_0 \cdot e^{-\frac{E}{RT}} \quad <5.2.1.4>$$

where $C_0$ is a constant, $E$ is the so-called activation energy and $R$ is the universal gas constant.

The total consolidation time has been reached when intimate contact and autohesion are achieved all over the former matrix boundaries. A computer code is proposed to solve this calculation problem$^{25}$.

Muzzy, Norpoth and Varughese$^{26}$ suggest in their work that "bulk consolidation" is complete when the extent of spacial gaps between laminate plies is minimized by lateral squeeze flaw of the thermoplastic matrix. They refer to the earlier described intimate-contact submodel as "very difficult to quantify" in practice and suggest a simpler model in which the consolidation is directly related to the change $\dot{V}$ of the volume of the laminate. Since only the thickness $h$ of the stack of composite layers changes:

$$\dot{V} = A \cdot \frac{dh}{dt} \quad <5.2.1.5>$$

where $A$ is the laminate area normal to the applied force and $t$ is the time.

Muzzy et al. include the expelling of voids out of the individual plies. This model requires extensive viscosity and flow measurements which are considered out of the scope of the present work. Like Dara and Loos, Muzzy et al. point out that the controlling mechanism for achieving interlaminar adhesion is polymer diffusion across the ply surface. Consequently, the consolidation is governed by time and temperature as already presented by equation $<5.2.1.3>$. Resin flow may contribute to the interply adhesion.

5.2.2 Setup of the consolidation experiments.

The consolidation phenomenon implicates the disappearance of the matrix-matrix interphase between two adjacent layers. Consolidation quality has to be determined by measurements of mechanical properties that are likely to be influenced by the consistency of the matrix
layer. In the open literature, however, little discussion is presented about the influence of consolidation quality on material properties. In Fig. 5.4, the test setup is shown, used by Lee and Springer for the determination of the adhesion degree of consolidated APC-2™ carbon fibre reinforced PolyEther-EtherKetone. This test, however, is not easy to use for practical consolidation measurements and, moreover, has little affinity with strength determination of composite structures.

![Diagram](image)

*Fig. 5.4 Test setup for the measurement of the adhesion degree of CFRTP material.*

![Diagram](image)

*Fig. 5.5 Four-point bending test setup.*

In the present work, four-point bending tests (EN 2563 / ASTM D790-M) are used for the determination of the consolidation quality of laminates (Fig. 5.5). Bending of a specimen loads the fibre reinforcement (tension and compression), the matrix (shear) and their interaction. It is expected that a bending test can indicate the influence of process
parameters on the consolidation quality of a laminate. To minimize the local indentation of the material due to the test load introduction, the four-point bending setup is chosen. Relatively small specimen are required and the test is performed rather easily, which makes the bending test also practical for testing real products.

Tensile tests (ASTM D3039) are also performed, but tensile loading of a specimen in fibre direction mainly results in the stressing the fibre reinforcement. It is therefore not expected that a 0° tensile test will clearly discriminate differences of consolidation quality between the various specimens. Fig. 5.6 even shows a complete independence of tensile testing results of 8-layered G/PEI laminates and the consolidation parameter time.\(^9\) This independence, however, is very unlikely.

![consolidation time vs tensile strength](image_url)

**Fig. 5.6 Influence of consolidation time on tensile strength of 8-layered G/PEI laminates.**

The consolidation experiments, that are discussed in this section, are carried out on 8-layered C/PEI laminates. Symmetric stacks of eight (0,90°) prepreg layers are consolidated into composite plates, using 24 different combinations of the consolidation process parameters time, temperature and pressure. Much attention is paid to short consolidation, because of the short cycles that are aimed at in rubber forming. The plates are consolidated between the hot platens of a Fontijne TP500 press. The stack is pressurized (\(p_c\)) for a certain time (\(t_c\)) after reaching the estimated temperature (\(T_c\)). The temperature of the laminates is constantly monitored by two thermocouples between the middle layers of the laminate. After fast cooling, each of the composite laminates is cut into 8 tensile specimens and 8 bending specimens in the two principal directions of the sheet and mechanically tested.
5.2.3 Results and discussion.

In Figs. 5.7 and 5.8, the averaged results are presented of the tensile and four-point bending tests, respectively, on the specimens that are consolidated at a pressure of 0.1 MPa. The tensile results are presented as failure force per unit width of the specimen. The bending results are given as flexural strength, which is the maximal tensile stress in the specimen at failure during bending, according to beam theory.

![Graph showing influence of consolidation time and temperature on flexural strength of 8-layered C/PEI laminates (consolidation pressure 0.1 MPa).](image)

**Fig. 5.7 Influence of consolidation time and temperature on flexural strength of 8-layered C/PEI laminates (consolidation pressure 0.1 MPa).**

The bending specimens broke in tension at the lower side after so-called micro buckling occurred at the upper side (Fig. 5.9). This mixed failure mode supports the suggestion that the four-point bending test discriminates on a matrix-fibre influenced specimen property. The bending test results indicate a reproducible change of consolidation quality with the applied consolidation. As expected, the tensile test results do not show a significant influence of the consolidation time or consolidation temperature on the specimen quality.

The results of the bending tests on specimens that are consolidated at a pressure higher than 0.1 MPa are included in Fig. 5.10. The comparison of these results with those of Fig. 5.8 (specimens that are consolidated at 0.1 MPa), shows that the influence of pressure can be neglected during consolidation at 275°C or 300°C. Even during the consolidation at a lower temperature (250°C), a relatively large pressure increase results in a minor increase of flexural strength, only.
Fig. 5.8 Influence of consolidation time and temperature on tensile strength of 8-layered C/PEI laminates (consolidation pressure 0.1 MPa).

Fig. 5.9 Mixed failure mode of the tested four-point bending specimen: microbuckling at the compressed upper side and tensile failure at the lower side.
Fig. 5.10 Influence of consolidation pressure on the flexural strength of 8-layered C/PEI laminates.

Because consolidation of prepregs also implies the decrease of laminate thickness, the thickness of each of the specimen is carefully measured. It is found that the laminate thickness is related to the consolidation environment\(^*\) (Fig. 5.11). When the thickness-consolidation data is rearranged and the flexural strength of the specimens is set against their thickness, a relation is evident (Fig. 5.12). The flexural strength \(\sigma_{\text{max}}\) of a specimen, in the four-point bending test (the load span is one third of the support span) is calculated by\(^{108}\):

\[
\sigma_{\text{max}} = \frac{P \cdot L}{w \cdot t^2}
\]

where \(P\) is the failure load,
\(L\) is the support span,
\(w\) and \(t\) are width and thickness of the specimen, respectively.

\(^*\)The variation of thickness of differently consolidated specimen is the reason that tensile results are presented as (failure force)/(width of the specimen), which is an indication of the tensile failure stress of the fibres.
Fig. 5.11 Influence of consolidation time and temperature on the thickness of 8-layered C/PEI laminates (consolidation pressure 0.1 MPa).

Fig. 5.12 Relation between the flexural strength of an 8-layered C/PEI laminate and its thickness after consolidation.
Equation <5.2.2.1> already considers the geometrical effect of varying thickness. If the flexural strength of point A (Fig. 5.12) is taken as representative for optimally consolidated C/PEI material, the specimens with higher thickness should follow the trend as suggested by the dashed line in the figure, assuming fracture at the same load \( P \). In reality, the differently consolidated specimens do not achieve the suggested flexural strength. It is likely that the lower consolidation quality is caused by a worse removal of voids or a lower degree of molecular diffusion at the layer interfaces. A better consolidation inevitably results in a decrease of specimen thickness. The experimentally demonstrated relation between laminate thickness and flexural strength therefore can be used to qualify consolidation quality. A material thickness measurement is relatively easy and can be a useful non-destructive tool for the first determination of composite product quality.

Curve fitting of the influence of consolidation parameters on flexural strength.

A strong relation of intimate contact time with pressure is suggested by equation <5.2.1.2>. If the PEI matrix viscosity is sufficiently low, the relation predicts a short intimate contact time \( t_c \). The matrix surfaces are therefore smoothed almost instantly in that case, provided that the prepreg layers are at least slightly pressurized.

The autohesion of the thermoplastic matrix only depends on consolidation temperature and time. If the intimate contact time is neglected, the subsequent autohesion is the determining condition for realizing a certain consolidation quality in a laminate. In that case, the consolidation pressure does not influence the consolidation quality of CFRTP specimens. The experimental results, presented in Fig. 5.10, confirm this assumption. At 250°C on the contrary, it is expected that the viscosity of PEI is sufficiently high to influence the necessary contact time (see also Fig. 2.42) because the specimens consolidated at 250°C show some pressure influence.

The influence of consolidation time and temperature on specimen strength is evident. Fig. 5.13 shows the test results that are introduced earlier in Fig. 5.7. The relation between flexural strength and the consolidation time and temperature is curve-fitted by a relation, analogous to relation <5.2.1.3> for the degree of autohesion \((T>T_g)\):

\[
\sigma_{\text{max}} = \sigma_0 + C \cdot (T - T_g) \cdot t^n
\]  

<5.2.3.1>

where \( \sigma_0 \), \( C \) and \( n \) are fitting constants (\( \sigma_0 \) can be considered to be the flexural strength of a laminate consisting out of fully separated prepreg layers),

- \( T \) is the consolidation temperature,
- \( T_g \) is the glass transition temperature of the thermoplastic resin,
- \( t \) is the consolidation time.

It must be noted that the Dara and Loos relation <5.2.1.3> as well as <5.2.3.1> are not based on physical considerations and for instance the boundary condition \( \sigma_{\text{max}} \to \infty \) for \( t \to \infty \) is not realistic.
Fig. 5.13 Experimental relation between flexural strength and consolidation parameters, curve-fitted by eq. <5.2.3.1> \((\sigma_0 = 150 \text{ N/mm}^2, C = 1.6 \text{ s}^6 \text{KN/mm}^2, n = 0.17 \text{ and } T_g = 200^\circ \text{C})\).

The relatively simple relation <5.2.3.1> is suggested here to quantify the effect of the consolidation time and temperature on the consolidation quality within the framework of this investigation. As will be shown later, such a relation is necessary in order to estimate the effect of non-isothermal consolidation.

The factor \(t^n\) in the relation is included because of the autohesion dependence of the consolidation process (see equation <5.2.1.3>). In the experiments presented here, \(n = 0.17\) gives a rough, first approximation of the influence of time on \(\sigma_{\text{max}}\). The temperature dependence \((T - T_g)\) gives a better approximation of the experimental results than \(e^{-ERT}\) as it is present in the Dara and Loos relation <5.2.1.4>. It can be justified by assuming a total lack of molecular diffusion in the matrix layers below their glass transition temperature \(T_g\). A temperature of 200\(^\circ\text{C}\) has been chosen for the \(T_g\) of PolyEtherImide resin in Fig. 5.13. This temperature is lower than the neat resin \(T_g\) of about 215\(^\circ\text{C}\) as it is usually presented in data sheets. The presence of a few percents of the solvent N-MethylPyrrolidone (NMP) which is likely in the specimens, is known to lower substantially the glass transition temperature of the PEI matrix\(^84\). The constants \(\sigma_0\) and \(C\) are chosen 150 N/mm\(^2\) and 1.6 s\(^6\)KN/mm\(^2\), respectively, for the C/PEI laminates.

The curve-fitting results as presented in the figure obviously are not optimal. A better quantification is not estimated here, because of presumed test setup deficiencies. A precise control of consolidation time and temperature is difficult because:

- during heating and cooling the temperature through the laminate thickness is not completely uniform;
- it is not possible to create a uniform surface temperature over the hot platens of the press;
- in this stage of the investigation it is not clear what consolidation influence can be expected from the heating and cooling cycle of a laminate, which especially makes the combination of short consolidation times and high temperatures vulnerable for parameter determination errors in equation $<5.2.3.1>$.

A more precise quantification of the influence of consolidation parameters is being investigated by Bersee$^{39}$.

The results of the presented consolidation measurements can be summarized as follows:

- Bending tests are more discriminative than $0^\circ$ tensile tests for the determination of the consolidation quality of a laminate.
- The pressure on a laminate above a certain temperature during consolidation has a minor influence on consolidation quality, compared to the influence of temperature and time.
- The relative thickness of consolidated laminates can serve as a first indication of their mechanical properties.
- A preliminary relation to express the influence of process parameters on the consolidation quality can be estimated.

### 5.2.4 Superposition of non-isothermal consolidation increments.

A non-isothermal and non-isobaric consolidation is inherent to the rubber forming technique and its influence has to be investigated in order to control product quality. No literature is found, however, on the subject of non-isothermal or non-isobaric consolidation of CFRTP materials. The empirical approach, introduced in the preceding section, is further developed here to calculate the influence of non-isothermal consolidation on the CFRTP material.

Once a strength-consolidation parameter relation like $<5.2.3.1>$ is estimated, it is possible to consider the influence of non-isothermal consolidation. In the present work, the linear superposition of small isothermal consolidation increments is suggested to estimate the total effect of a non-isothermal consolidation cycle. After intimate contact has been established, the consolidation of a laminate is only determined by the diffusion of the polymer chains through the former matrix boundaries (autohesion). It is assumed that the effect of every increment in this diffusion can be linearly superposed to finally obtain the total consolidation quality.

Fig. 5.14a shows an arbitrary temperature-time relation. A possible subdivision of the temperature profile is given in Fig. 5.14b. This subdivision in discrete time steps $\Delta t$ is necessary to calculate the consolidation effect on the flexural strength from equation
<5.2.3.1>. The mean temperature $T_m$ during every time increment is considered to cause a specific amount of consolidation. The effects of each time step $\Delta t$ on consolidation are different because of the different temperature levels during each step. The cumulative effect of all the consolidation steps 1 to $n$ now can be calculated.

![Temperature Time Graph](image1)

**Fig. 5.14 Possible subdivision (b) of an arbitrary cooling curve (a) of a consolidating laminate.**

In Fig. 5.15, the strength relation <5.2.3.1> is visualized for various consolidation temperatures. Fig. 5.16 demonstrates the graphical determination of the cumulative consolidation effect of time step $t_i$ to $t_n$. During the first time step $\Delta t_1 = (t_{2(T_1)} - t_{1(T_1)})$ at consolidation temperature $T_1$, a mechanical strength $\sigma_1$ is realized:

$$\sigma_1 = \sigma_0 + \Delta \sigma_1 = \sigma_0 + C \cdot (T_1 - T_g) \cdot (\Delta t)^n$$  \[<5.2.3.2>\]

The next time step $\Delta t_2$ is carried out at a lower temperature $T_2$ and will realize an additional strength increase $\Delta \sigma_2$:  

![Temperature Time Graph](image2)
\[ \Delta \sigma_2 = C \cdot (T_2 - T_s) \cdot ((t_{2(T2)}^{n+1}) - (t_{2(T2)})^n) \]  

<5.2.3.3>

Since the physical laminate condition at the end of time step 1 is identical to the physical laminate condition at the beginning of time step 2, a 'new' time point \( t_{2(T2)} \) has to be calculated from the condition that:

\[ \sigma(t_2, T_2) = \sigma(t_2, T_1) \]  

<5.2.3.4>

Equations <5.2.3.1> and <5.2.3.4> then yield the equivalent starting time of step 2:

\[ t_{2(T2)} = \frac{n \cdot \sigma_1}{C \cdot (T_2 - T_s)} \]  

<5.2.3.5>

**Fig. 5.15** Flexural strength relations for various temperatures, according to eq. <5.2.3.1>.

**Fig. 5.16** Graphical determination of the cumulative effect of a non-isothermal consolidation cycle on the strength of a laminate.
The consolidation quality $\sigma_i$ at the end of an arbitrary step $i$ can be calculated from the consolidation quality $\sigma_{i-1}$ at the end of the preceding step (i-1) and time increment $\Delta t$ during which the material is consolidated at the new temperature $T_i$:

$$\sigma_i = \sigma_{i-1} + C \cdot (T_i - T_g) \cdot ((t_i + \Delta t)^n - t_i^n)$$  \hspace{1cm} <5.2.3.6>

where

$$t_i = \sqrt[n]{\frac{\sigma_{i-1}}{C \cdot (T_i - T_g)}}$$  \hspace{1cm} <5.2.3.7>

By summation of the increments 1 to n, the total consolidation quality $\sigma_n$ due to all the consolidation steps is estimated.

From Fig. 5.16, it can be concluded that during the last consolidation steps at lower temperatures the total consolidation quality will hardly be influenced. The first few moments at high temperatures in the consolidation phase of a cooling laminate are the most important for the final material quality. It is therefore strongly recommended to optimize the heat flow during rubber forming in such a way that the temperature of the laminate at the end of the forming phase is as high as possible to realize a satisfying consolidation quality.

The relation between consolidation quality and process parameters <5.2.3.1> has to be iteratively optimized by a large amount of consolidation data to take the heating and cooling of laminates into account during consolidation experiments. In this way, a calculation tool is created to predict the consolidation quality of non-isothermally thermoformed CFRTP products.

### 5.3 The influence of sudden cooling.

The rubber forming technique requires large changes in temperature of the CFRTP material in a relatively short time span, as is already demonstrated in Chapter 4. A hot laminate that is touched by a relatively cold mould will suddenly cool down. This sudden cooling can result in so-called microcracks in the matrix layer, which is discussed in Section 5.3.1.

Furthermore, the different materials of the rigid and the rubber mould in rubber forming impose different cooling rates of the two laminate surfaces. These cooling rates can cause geometrical distortions in the final product, due to residual internal stresses, as is shown in Section 5.3.2. The difference in thermal properties between matrix and fibre material will intensify the thermally induced deficiencies.
5.3.1 Microcracking of the matrix layers.

After rubber forming C/PEI, G/PEI and G/PET laminates, microcracks have been found frequently in the final products. Microcracks are (mostly microscopically) small cracks in a laminate that run through the matrix layer(s) or along the matrix-fibre interfaces. Examples of these cracks are shown in Fig. 5.17. The cracks are considered to be thermally induced, due to differences in thermal properties between matrix- and fibre material and the presence of an inhomogeneous temperature distribution through the laminate thickness\(^{63}\). It is illustrated in this section that the cooling rate of a thermoformed CFRTP product is very important, therefore.

Fig. 5.17 Microcracks in a suddenly cooled PEI-based laminate.

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Fig. 5.18 Schematic illustration of microcracks, perpendicular to the product surface.
In Fig. 5.18, an illustration is given of microcracks in the outer matrix layer, running perpendicular to the laminate surfaces. The microcracks are the result of the temperature "shock" that the hot laminate surfaces endure when they touch the cold(er) moulds. Local differences in shrinkage, imposed by the composite nature, or temperature gradients through the thickness cause internal stresses in the material. The cracking might proceed to the inner matrix layers, because a temperature gradient is present through the whole laminate thickness during cooling. How serious the microcracking will become strongly depends on the cooling rate and related internal stresses.

In experiments, microcracks are observed, both perpendicular and parallel to the fibres. Microscopic photographs of PEI-based laminates show microcracks in the matrix layers running from fibre layer to fibre layer. Fig. 5.19 shows a picture of a PET-based laminate where, obviously, the matrix-fibre interphase is the weakest spot. Microcracking occurs over the fibre surface inside the cooling laminate. While cooling a PET laminate in a rubber forming process, the cracking is even audible, especially at cooling rates where crystalline matrix parts are created. They cause a higher density and therefore more shrinkage during solidification\(^7\).

"The crystallizing tendency of semi-crystalline matrix based laminates will complicate the microcracking phenomenon\(^8\). This is not discussed here.
The effects of microcracking on the mechanical properties of a laminate are difficult to determine experimentally. Many factors that are considered to be important for this phenomenon can hardly be controlled or measured. Among them are the thermal properties of the composite material, the distribution of the fibres through a laminate and the temperature differences between matrix and fibre material parts. Microcracks, which run along the fibre surfaces or perpendicular to the surfaces, disturb the shear flow in a bent laminate and will consequently lower the bending stiffness and strength of a composite part.

In Figs. 5.20, the results are presented of a short beam three-point bending test (EN2377) on rubber formed 10-layered C/PEI specimens that have undergone different cooling histories in the mould after deformation. This is performed by using different insulation layers between the hot specimens and the cold mould.

Comparison of the 3-point bending shear stresses at failure shows that a higher stiffness and strength is gained by decreasing the cooling rate in the mould. Photographs of the specimens, however, do not clearly show a variation in the occurrence of microcracks. Therefore, (part of) the better mechanical properties of the series could be ascribed to a better overall consolidation quality. This was discussed in 5.2. The thicknesses of the specimens vary slightly.

![Figure 5.20a](image-url)  
*Fig. 5.20a Three-point bending deflection of differently insulated rubber formed laminates.*

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*In the results, an ILSS determined maximum shear stress is presented, although all the specimens failed in tension.*
Fig. 5.20b Three-point bending strength of differently insulated rubber formed laminates.

The presence of microcracks at the laminate surface under mechanical loading can be expected to ease the initiation of through cracks. Furthermore the presence of microcracks that reach the laminate surface can facilitate the penetration of the environment. The bearing strength and/or the product life can be seriously affected if this environment is aggressive for the particular composite material. Microcracks can also affect the optical surface quality so that even in non-structural applications, the presence of microcracks has to be minimized.

5.3.2 Thermally induced distortion of the product geometry.

The fact that the rubber forming method uses two different mould materials (i.e. rubber and metal) will cause different cooling histories at both sides of the composite sheet material, which will lead to residual stresses. An example is given in Fig. 5.21a. The higher initial temperature at A and B will still allow interply slip to relieve the induced compressive stresses. These stresses are raised by the (faster) shrinkage of layers D and C due to their higher cooling rate. When the material temperature of A and B finally drops below the minimum temperature at which interply slip is possible, no more stress relief is possible in the laminate. Layers A and B, however, still have a higher temperature than C and D and have to cool down further to room temperature. The higher residual shrinkage from this point will cause compression stresses in the layers C and D. Equilibrium at room temperature requires tensile stresses in A and B in the final product. A residual tensile stress at the rubber contact side and a compression stress at the metal contact side will cause a deformation when the product is taken out of the mould (Fig. 5.21b). The unintended curvature of the product will partly relieve the thermal residual stresses.
Fig. 5.21a Typical temperature distribution through the thickness of a rubber formed laminate in the consolidation phase.

Fig. 5.21b Deformation of a rubber formed product after removal from the mould due to residual internal stresses, which are thermally induced.

The so-called spring-forward effect in a product is another possible form of thermally induced distortion of the product. With "spring-forward" is meant that the final angle in a formed product is larger than the mould angle (Fig. 5.22). It is mostly noticed in thermoformed ledged products but it is believed to play a dominant role in more gently curved products also. The deviation is induced during the cooling of the composite material and several causes are depicted in the literature. The anisotropy in thermal expansion properties of the composite material is believed to be most important: The in-plane coefficient of thermal expansion of a fibre reinforced material can be more that 40 times higher than the through thickness coefficient.
A decrease of the cooling rate of a CFRTP laminate at the end of a thermoforming process is often restricted for technical and/or economical reasons. Some matrix materials however show to be susceptible to the non-uniform and sudden cooling and it depends on their final application whether temperature control has to be altered during rubber forming or not (higher mould temperature, insulation, etc.). More focused investigations have to be carried out with sufficiently sensitive temperature equipment to fully describe the presented thermal phenomena.

5.4 The influence of forming forces.

The fabric reinforced PEI prepgs, which are used in the investigations in this section, are manufactured by solvent impregnation of the matrix into the fabric reinforcement. The woven fabric is dipped in a bath of polymer solution and transported through a drying oven to remove the solvent (Fig. 5.23). Forces are imposed in the warp direction of the reinforcing fabric during this transport, which cause a straightening of the fabric yarns in this direction (Fig. 5.24). The yarns in the weft direction become more curved (Fig. 5.25). These differently curved prepreg yarns will cause a difference in mechanical properties in the warp and weft direction of the laminate, because the straightening of the fabric yarns is expected to influence the mechanical efficiency of the reinforcement\textsuperscript{103}.

The fabric reinforcement of a laminate is also loaded in tension during a rubber forming cycle: during the pre-consolidation of the prepreg sheets, during the forming phase of the laminate and during the (re)consolidation of the final part. During the forming phase, this loading is caused by (external) forming forces (Chapter 2). During the consolidation steps, the loading of the fabric is caused indirectly: by the matrix flow that occurs due to pressurization of the hot laminate. In this case too, curved yarns in a fabric will straighten in the force direction at the cost of the alignment of the interlaced yarns perpendicular to the force direction.
Fig. 5.23 Solution impregnation of PEI-based prepregs (the arrow denotes the warp direction of the reinforcing fabric).

Fig. 5.24 Curvature of warp yarns in a PEI-based prepreg.

Fig. 5.25 Curvature of weft yarns in a PEI-based prepreg.

In the present section, the mechanical influence is discussed of a tension in one of the principal directions of the fabric during rubber forming\textsuperscript{82,104-106}. Therefore, the changes in mechanical properties of composite material are estimated after each of the subprocesses in a rubber forming cycle. The mechanical effect of a distortion of the fibre directions, as it is imposed during intraply shearing, is not considered in the present work.
5.4.1 Setup of the forming force experiments.

Experiments are carried out on two kinds of CFRTP material: 10-layered G/PEI laminates and 8-layered C/PEI laminates (Appendix C1). Four laminates (500 mm by 500 mm) of each of the investigated materials are symmetrically stacked out of prepreg sheets. In a laminate the warp fibres of the prepreg sheets are all oriented in the same direction. This direction is called the warp direction of the laminate and is perpendicular to the so-called weft direction. The prepreg stacks are preconsolidated at different consolidation parameters in a Fontijne TP1000 hot platen press (Table 5.1).

<table>
<thead>
<tr>
<th>Table 5.1: Preconsolidation parameters for the forming force experiments.</th>
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<tr>
<td>consolidation code</td>
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<td>G/PEI</td>
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After preconsolidation, each laminate sheet is divided into three parts.

One part is used to obtain material data of laminates that are preconsolidated only. Specimens are prepared out of the laminates to perform tensile tests (ASTM D3039), compression tests (ASTM D3410 / EN2850) and four-point bending tests (ASTM D790M / EN2563), in warp and weft directions of the material.

The second part of the preconsolidated laminates is used in experiments to simulate the pressure process in a final consolidation setup: The laminate sheet is pressureless heated in the hot platen press (G/PEI specimens at 300°C, C/PEI specimens at 320°C), quickly transferred to a cold flat metal mould and pressurized by a flat rubber stamp at approximately 3 MPa. Tensile, compression and four-point bending specimen are cut from these sheets to obtain material data of laminates that are reconsolidated, but not directly stressed as in a forming phase.
The third part of the preconsolidated G/PEI and C/PEI plates is reheated and rubber formed into hat sections to investigate the influence of the stretching of the fabric during forming. The hat sections are rubber formed in a way that interply slip is imposed either in the warp direction or the weft direction of the laminate. Out of the sides and top of the obtained hat sections, specimens are cut which are tested in the four-point bending fixture.

In addition to the previously described investigations, the influences of forming speed, mould temperature and processing temperature on the mechanical properties of a rubber formed product have been investigated. 6-layered G/PEI laminates are preconsolidated at relatively high temperatures (variable: 280°C to 380°C) and relatively high pressure (3.75 MPa). These preconsolidated laminates are rubber formed into a hat shaped stiffener (Fig. 5.26) at various starting temperatures of the laminate and the mould (forming- and mould temperatures, respectively) and at various velocities of the press (forming speed). The bending angle of each of the final parts is monitored and four-point bending specimens, cut out of the rubber formed hat stiffeners are tested.

![Fig. 5.26 Shape of rubber formed G/PEI hat stiffeners.](image)

### 5.4.2 Test results.

In Figs. 5.27a,b, the test results are presented of the specimen out of the eight laminates that are preconsolidated only. The mean mechanical properties of the laminates are higher if a higher consolidation is applied by higher temperature and/or pressure. Moreover, the differences between warp and weft properties are decreased due to a higher consolidation.

In Figs. 5.28a,b, the test results of the flat rubber formed specimen are presented. The difference between warp and weft directional mechanical properties is somewhat smaller than before the forming cycle. The mean strength of flat rubber formed material is also lower, except for the worst pre-consolidated G/PEI and C/PEI laminates (GD and CA respectively).

The flexural strengths of the specimens cut from the hat sections are presented in Figs. 5.29 and 5.30. They are arranged according to the direction in the specimen in which interply slip is imposed during rubber forming.

The influence of a rubber forming cycle on laminate thickness can be noticed from Figs 5.31 and 5.32, which give the specimen thicknesses after each investigated subprocess for various specimen series.
The test results of the additionally performed investigation of the rubber forming of 6-layered G/PEI hat stiffeners are presented in Figs. 5.33a,b,c,d. They are arranged according to their different preconsolidation temperatures and the parameters forming temperature, mould temperature and forming speed. In Fig. 5.34, the final bending angles $\phi$ are presented in relation to the forming speed.

(Fig. 5.27a)
Fig. 5.27a Mechanical influence of preconsolidation on the warp and weft directional flexural, compression and tension strength of 10-layered G/PEI laminates.

(Fig. 5.27b)
Fig. 5.27b Mechanical influence of preconsolidation on the warp and weft directional flexural, compression and tension strength of 8-layered C/PEI laminates.
Material response to rubber forming

(Fig. 5.28a)
Fig. 5.28a Mechanical influence of additional flat rubber forming on the warp and weft directional flexural, compression and tension strength of 10-layered G/PEI laminates.
Fig. 5.28b Mechanical influence of additional flat rubber forming on the warp and weft directional flexural, compression and tension strength of 8-layered C/PEI laminates.
Fig. 5.29 Influence of the deformation direction on the flexural strength of 10-layered G/PEI laminates.

Fig. 5.30 Influence of the deformation direction on the flexural strength of 8-layered C/PEI laminates.
Fig. 5.31 Influence of a rubber forming cycle on the thickness of an 8-layered C/PEI laminate.

Fig. 5.32 Influence of a rubber forming cycle on the thickness of a 10-layered G/PEI laminate.
Fig. 5.33a Influence of the preconsolidation temperature on the flexural strength of rubber formed 6-layered G/PEI stiffeners.

Fig. 5.33b Influence of the rubber forming temperature on the flexural strength on rubber formed 6-layered G/PEI stiffeners.
Fig. 5.33c Influence of the mould temperature on the flexural strength of rubber formed 6-layered G/PEI stiffeners.

Fig. 5.33d Influence of the forming velocity on the flexural strength of rubber formed 6-layered G/PEI stiffeners.
Material response to rubber forming

Fig. 5.34 Influence of the forming velocity on the geometrical deviation of rubber formed 6-layered G/PEI stiffeners.

5.4.3 Discussion.

Preconsolidation results.

During the preconsolidation cycle of G/PEI and C/PEI laminates, the consolidation phenomenon occurs as it was discussed in Section 5.2.1. Moreover, the surface pressure, that forces the heated prepreg layers into intimate ply contact, causes a certain amount of matrix flow.

The flow of PEI resin, though very little, is directed from the middle of the laminate surface to the less pressurized edges of the laminate sheet and stretches the reinforcing fabric (Fig. 5.35). This results in a decreased difference between the curvatures of warp and weft bundles. Higher consolidation pressures and/or temperatures yield increasing matrix flows and therefore decreased warp-weft differences (Figs. 5.27a,b). The decrease of thickness with higher consolidation (Figs. 5.31 and 5.29) supports the assumption of the increase of the matrix flow.

matrix flow

Fig. 5.35 Schematic illustration of fibre stretching due to preconsolidation matrix flow.
Flat rubber forming results.

During the thermoforming of a CFRTP product out of a preconsolidated laminate, pressure is applied at elevated temperatures for the second time. The surface pressure on the laminate during forming and consolidating can induce further matrix flow. The difference between the mechanical properties in warp and weft direction, however, is only slightly lower after additional flat rubber forming (Figs. 5.28a,b). It is expected that little opportunity is given to the fabric to stretch any further due to matrix flow, because the cooling down of a laminate to the glass transition point only takes a few seconds.

The mean strengths of the specimens decrease due to deconsolidation of the layers during the flat rubber forming. This deconsolidation of the layers is caused by the lack of pressure during heating. Apparently, the (re)consolidation of the final specimens in the mould is insufficient. This explanation is supported by the higher specimen thickness after flat rubber forming (Figs. 5.31 and 5.32). It is shown however, that the influence of the different preceding preconsolidation cycles is still noticeable.

Hat section results.

The test results of the G/PEI specimens out of the rubber formed hat sections show an influence of the forming direction (Fig. 5.29). In Fig. 5.36, this influence of the subsequent subprocesses on the specimens of the GB series is visualized. It is shown for instance, that the mechanical properties in weft direction after interply slip in that direction have increased compared to the properties before rubber forming. They are also higher compared to the weft directional properties of a specimen that is deformed in the warp direction. It shows that for the G/PEI hat profiles the direction of the main deformation can be important for the final mechanical properties.

The bending test results on the C/PEI specimens do not show any consistent influence of the forming direction (Fig. 5.30). It is expected that a possible influence is not detected after rubber forming because a low overall reconsolidation quality of the C/PEI specimens after deformation dominates the bending test results. This explanation is supported by thickness measurements (Fig. 5.31). The measured thickness increase after rubber forming of the C/PEI specimens indicates a bad (re)consolidation of the deformed material. This insufficient reconsolidation also leads to a decreased average mechanical strength compared to the preconsolidated material (Fig. 5.27b).

The rubber forming of hat profiles does not affect the thickness of the G/PEI specimen (Fig. 5.32). The measured mechanical differences in the glass fabric reinforced material therefore mainly depend on the physical stretching of the fibre reinforcement.
Fig. 5.36 Influence of subsequent subprocesses on the flexural strength of 10-layered G/PEI laminates (GB preconsolidated).

The results of the additionally performed investigation of the rubber forming of 6-layered G/PEI hat sections (Fig. 5.33) show that a slight influence of the preconsolidation cycles is still present in the final products. A warp-weft difference cannot be detected. The difference apparently is lost in the scatter of the test results. This is attributed to the relatively severe preconsolidation. For the same reason the influence of the forming directions is assumed to be completely overshadowed by the preconsolidation subprocess.
Fig. 5.37 Internal stresses imposed by the bending of a laminate during thermoforming.

The test results in Figs. 5.33b to d also show that the influence of the parameters laminate forming temperature, mould temperature and forming speed cannot be estimated in a reliable way. They were expected to cause different internal forming forces.

The corner angles of the rubber formed hat profiles out of the 6-layered G/PEI laminates are smaller when a higher deformation velocity is applied in the rubber forming cycle (Fig. 5.34). A higher forming speed results in higher internal stresses in the internal stresses in the final hat section. The required stress equilibrium causes a "spring-back of the deformed laminate after removal of the product out of the mould (Fig. 5.37)."

The rubber forming of prototypes of G/PET suitcases is taken as an example to illustrate the effect of high forming stresses in the sides of a laminate. The bulging of the relatively large (originally flat) bottom area was noticed after rubber forming the first samples (Fig. 5.38).

A cross section, cut out of the bulging suitcase, is shown in the top of Fig. 5.39. To accomplish this shape by internal stresses, tensile stresses have to be introduced at the product out-side during rubber forming (like in the shaping of the hat shaped stiffener).

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"This effect is opposite to the spring-forward effect pointed out in Section 5.3.2, which is a thermally induced product distortion that is inherent to thermoforming CFRTP products in general."
Fig. 5.38 Bulging of the bottom area of a rubber formed CFRTP suitcase shell.

Fig. 5.39 Shape of cross-sections of rubber formed suitcase shells. The bottom one is rubber formed with reduced friction at the mould sides.
These tensile stresses can be caused during forming by laminate-mould friction only. Therefore, another G/PET suitcase is rubber formed with lubrication grease applied at the negative mould surface. A cross section of this suitcase is also shown in Fig. 5.39 and shows only little product-mould deviation. The lubrication obviously lowers the forming resistance of the laminate, which minimizes the introduction of tensile stresses at the product outside. This results in a higher mould resemblance of the final product.

A conclusion to prevent product mismatches might be forming at lower speed. This solution is however contradicted by the fact that a slower forming phase will result in forming and consolidating at a lower material temperature, which in itself can raise the internal stresses, as was discussed in 2.3.

5.4.4 Conclusions of the forming force experiments.

Some conclusions can be drawn from the forming force experiments, which are important if a specific product quality (shape, surface and/or mechanical properties) must be obtained:

- In a fast thermoforming process like rubber forming, good preconsolidation is necessary to enable the realisation of the highest material properties in a PEI-based fabric reinforced part.
- The fibre alignment of prepreg material in a laminate influences the mechanical properties of the final product, but can be (partly) compensated by forming forces during rubber forming. The last step in the rubber forming cycle is most important for the CFRTP part quality.
- Mechanical properties of thermoformed products can depend on the direction in which the laminate is deformed.
- The necessary forming forces during thermoforming have to be minimized to assure a high mould resemblance of the final product.

5.5 Concluding remarks.

The material response to rubber forming is difficult to quantify because the influences of the subprocesses, like heating, forming, consolidation and cooling, can be hardly isolated for investigation. In this chapter, several experiments are carried out on CFRTP materials that are exposed to rubber forming phenomena. From the results of these experiments some main conclusions can be given:

- The influence of non-isothermal consolidation on the mechanical properties of a laminate can probably be quantified. It is experimentally found that the pressure during consolidation has a minor effect on the final consolidation quality, if the consolidation
temperature is sufficiently high and time is sufficiently long. The superposition of the influence of isothermal temperature increments during non-isothermal consolidation is suggested to calculate the final properties of a rubber formed part. It is recommended however, to perform extended consolidation experiments to gain a more accurate relation between the applied consolidation parameters and the mechanical properties of a laminate.

- The sudden cooling of the laminate in the forming and consolidation phase of a rubber forming cycle can result in microcracks in the matrix layers and/or distortion of the final product shape. These phenomena are caused by the introduction of internal stresses, induced by large temperature differences through the thickness of the laminate. Microcracks and geometrical product mismatches due to residual internal stresses can be avoided, for instance, by mould insulation to lower the cooling rate of the CFRTP part.

- The fibre alignment in the reinforcement is important for the mechanical properties of the laminate. Tensile forces on the fabric can enhance the material properties in the force direction by straightening of the fabric yarns. These tensile forces can be imposed by (external) forming forces during forming or by matrix flow during consolidation. The properties of a final part are therefore influenced by the direction (warp or weft) in which the laminate is loaded in tension during forming. The forming forces have to be minimized to avoid residual internal stresses in the final part.

The application of the process parameters in the rubber forming technique is subject to technical, economical, and quality considerations. Only when the response of the composite material to the rubber forming process parameters is known, it is possible to determine the processing conditions that are necessary for the realisation of a cost effective structure.
CHAPTER 6. Quality of the rubber forming technique.

6.1 Introduction.

Structural composite parts can only be realized with rubber forming if this technique is sufficiently reproducible. Therefore, it must be demonstrated in what way a rubber forming cycle affects the mechanical properties of the CFRTP material.

In this chapter, the effect of the rubber forming technique on the mechanical behaviour of a composite part is investigated. Therefore a realistic structure, a 1.50m long part of an aircraft floor beam, is produced out of fabric reinforced thermoplastic material. The automated thermoforming facility (Fontijn NHR 134) as described in Chapter 3, is used for this purpose. The engineering bending theory is used to calculate material stiffness properties from the measured stiffness of a beam structure. It is emphasized, however, that this theory is used only as an example of a calculation tool. A different product geometry and/or laminate layup may require a more thorough stiffness analysis of the CFRTP product. This is considered outside the scope of the present work.

In 6.2, the manufacture of the floor beams is described. Coupon testing is performed on the starting material, the (pre)consolidated material and material taken from the final product. With these tests, the stiffness influences of the rubber forming process is determined. Full-scale stiffness measurements on the final product are performed as well. In 6.3, the results of the tests are discussed and used to demonstrate how the quality of rubber formed parts can be determined. To enable calculation of required starting material for a rubber formed part, a production-factor is introduced which depicts the overall rubber forming quality. In 6.4, it is shown that the process control during rubber forming determines the quality of the CFRTP parts.

6.2 Fabrication and testing of real-size CFRTP beams.

The influence of a rubber forming cycle on the mechanical properties of a carbon fabric reinforced PEI part is investigated with a 1.50m long U-shaped beam, (originally a part of an aircraft floor beam) rubber formed out of 8-layered laminates (Fig. 6.1).

The results of tensile tests and four-point bending tests on coupons of the preconsolidated starting material are compared to the test results of coupons out of the floor beam. Full-scale four-point bending tests and clamped-end tests are carried out on the beam to determine the stiffness of the beam as a whole. Simple engineering beam theory is used to investigate a possible relation between the measured modulus of elasticity of the material and the measured stiffness of the beam.
Fig. 6.1 Rubber formed C/PEI beams (1.50 m long).

6.2.1 Production of the U-shaped beams.

The starting material for the rubber forming cycle is a preconsolidated C/PEI sheet. Two types of layup are used for the stacking of the 8-layered laminates, which are denoted "crossply" and "combiply" in this investigation. The layup of the crossply consists of 8 layers of 5H satin-fabric reinforced prepreg sheets, positioned with the warp yarns in the same direction. This direction will be the length direction of the beam after forming (Fig. 6.2). The 8 prepreg layers for the combiply are stacked symmetrically with their warp direction alternating in 0°, +45°, -45°, 0°, 0°, -45°, +45° and 0° with respect to the later length direction of the beam. Because of the limited width of the available prepreg sheets, the ±45° prepreg layers are not continuous, as illustrated. The necessary seams in those layers, however, are distributed over the laminate as uniform as possible.

The stacked prepreg layers are (pre)consolidated by the supplier by use of a hot platen press. The consolidation temperature and pressure are 337°C and 2 MPa, respectively. They are applied for 20 minutes. The 1600 x 300 mm² laminate sheets are subsequently rubber formed into a U-shaped beam.

The percentage of the solvent NMP (N-Methyl-2-Pyrrolidinone) inside the solution impregnated prepreg layers is reduced beforehand to approximately 0.3% by a drying cycle of 3 hours at 300°C.
Crossply layup

prepreg warp direction:

0°

0°

0°

0°

0°

0°

0°

0°

0°

0°

3600

1000

Combiply layup

prepreg warp direction:

0°

0°

+45°

-45°

0°

0°

0°

-45°

+45°

0°

3600

1000

Fig. 6.2 Schematic presentation of the crossply and the combiply layup for the rubber forming of an 8-layered C/PEI laminate.
The rubber forming setup consists of a negative U-shaped steel mould and a rectangular rubber stamp in between which the hot laminate is formed (Fig. 6.3). The height of the flanges of the U-shaped beam is determined by the total thickness of the filler plates in the steel mould. To facilitate assembling, the moulds are divided by a seam as illustrated in the figure. The positive mould (stamp) consists of two pieces of casted PolyUrethane rubber with a hardness of 90 Shore A, which provide the pressurization during the rubber forming cycle.

Fig. 6.3 Rubber forming setup for the forming of the beam.

After insertion of the preconsolidated laminate in the preheated (330°C) transferring device, the C/PEI laminate is heated by the infrared radiation panels up to 320°C within 120 seconds. The hot laminate is quickly transferred to the press unit, released above the (cold) negative mould and pressurized during 30 seconds at 4 MPa to 6 MPa (Fig. 6.4).
6.2.2 Determination of material properties by coupon testing.

From the preconsolidated laminates, coupon specimens are cut, both from the crossplies and the comiplies. The rubber formed U-shaped beams are also cut into coupon specimens after the non-destructive full-size bending tests are performed. In Fig. 6.5 the original positions and numbering of the beam coupon-specimens are presented.

Fig. 6.4 Rubber forming of the C/PEI beam.

Fig. 6.5 Positions of the coupon specimens out of the rubber formed beam.
Using the coupon specimens, the stiffness of the material before and after rubber forming is determined in the length direction of the beam (denoted "warp" direction or 0° direction) and the direction perpendicular to the length direction (denoted "weft" direction or 90° direction). The stiffness determination is performed by four-point bending testing and tensile testing.

Four-point bending tests.

The specimens, used to determine the flexural strength and stiffness have the dimensions of approximately 10 mm by 40 mm. With these relatively small dimensions it is possible to measure the material properties very locally. The tests (ASTM D790-M / EN2563) are carried out in a 10 kN MTS testing facility (see also Fig. 5.5).

To determine the modulus of elasticity in bending $E_b$ of the specimens, a deflectometer is used to measure the centre deflection of the specimen during the test. From the estimated load-deflection curve the modulus is determined by:

$$ E_b = 0.21 \cdot \frac{m \cdot L^3}{w \cdot t^2} \quad \text{<6.2.2.1>} $$

where the factor 0.21 represents the geometry effect of the relative support span (the load span is one third of the support span), $m$ is the slope of the tangent to the initial straight section of the load-deflection curve, $L$ is the span of the support (30 mm), $w$ is the width of the specimen and $t$ is the thickness of the specimen.

The specimens fail by micro buckling at the upper side between the two central loading points, immediately followed by tension failure at the lower side. The flexural strength $\sigma_{\text{max}}$ is calculated with the simple expression:

$$ \sigma_{\text{max}} = \frac{P_{\text{max}} \cdot L}{w \cdot t^2} \quad \text{<6.2.2.2>} $$

where $P_{\text{max}}$ is the bending load at failure.

The four-point bending results of the initial crossply and combiply coupons are presented graphically in Figs. 6.6 and 6.7.

In Figs. 6.8a-d and 6.9a-d, the results are presented of the four-point bending tests on coupons of the laminates after forming. They are taken from the web and flange of the rubber formed U-beam. The results are grouped for different positions on the beam.

The results will be discussed in 6.3.
Fig. 6.6 Modulus of elasticity of preconsolidated C/PEI material determined by four-point bending.

Fig. 6.7 Flexural strength of preconsolidated C/PEI material determined by four-point bending.
Fig. 6.8a Four-point bending test results of material out of the crossply beams: modulus of elasticity of the web specimens.

Fig. 6.8b Four-point bending test results of material out of the crossply beams: flexural strength of the web specimens.
Fig. 6.8c Four-point bending test results of material out of the crossply beams: modulus of elasticity of the flange specimens.

Fig. 6.8d Four-point bending test results of material out of the crossply beams: flexural strength of the flange specimens.
Fig. 6.9a Four-point bending test results of material out of the combiply beams: modulus of elasticity of the web specimens.

Fig. 6.9b Four-point bending test results of material out of the combiply beams: flexural strength of the web specimens.
Fig. 6.9c Four-point bending test results of material out of the combipy beams: modulus of elasticity of the flange specimens.

Fig. 6.9d Four-point bending test results of material out of the combipy beams: flexural strength of the flange specimens.
Tensile tests.

The tensile tests are performed according to Fokker norms and carried out in a 100 kN MTS testing facility. During the tests, an extension meter is used to measure the elongation of the test specimens. The (tensile) modulus of elasticity $E$, can be obtained from the load-elongation curve. The estimated maximum tensile stress is denoted $\sigma_{\text{max}}$.

The tensile results of the crossply and combipy laminates before rubber forming are presented in Figs. 6.10 and 6.11.

In Figs. 6.12 and 6.13, the results are presented of coupons from the web of the rubber formed U-beam, grouped for different positions on the beam.

These results will be also included in the discussion in 6.3. (The relatively large dimensions of the tensile specimens of approximately 15 mm by 175 mm obstruct the cutting of tensile specimens from the flanges of the rubber formed beam.)

![Graph showing tensile test results](image)

Fig. 6.10 Test results of the tensile tests on preconsolidated material: modulus of elasticity.
Fig. 6.11 Test results of the tensile tests on preconsolidated material: tensile strength.

Fig. 6.12 Modulus of elasticity of material coupons out of the web of the crossply laminates.
6.2.3 Determination of material stiffness by full-scale testing.

To obtain the mechanical properties of the rubber formed beams as a whole, full-scale clamped edge and four-point bending tests are chosen, because the beams originally are applied in an airplane for their bending resistance. Moreover, a full-scale bending test setup is easier to realize than for instance a full-scale tensile test setup.

The bending loads during the full-scale tests are applied at the shear centre of the beams by testing pairs of identically processed specimens: for the four-point bending tests, two U-shaped beams are bonded backwards to each other (Fig. 6.14), using a 1% glass filled adhesive Scotch Weld® AW9323; for the clamped edge tests, the two U-shaped beams are (mechanically) fixed at the ends. The shear centre of the now obtained I-shaped beam is situated in the web of the beam.

The bending theory of thin-walled beams\textsuperscript{120} is used to transform the strain measurements in x-direction into stiffness data. Since the layup of the beams is chosen symmetrical and each layer is symmetrically reinforced, a bending load in z-direction on the bonded beams is considered to result in a bending deflection about the y-axis only. A coupling between a load in z-direction and twisting, due to typical coupling behaviour of the laminates, therefore is absent. If such a coupling would be present, the measured bending stiffness of
the system of the bonded beams would exceed the double bending stiffness of a single beam, because of the obstruction of twisting during the bending test. Furthermore, it is assumed that the shear deflection of the beam in bending can be neglected, because the shear modulus of the material is sufficiently high. The modulus of elasticity $E$, calculated by the bending theory, is actually the modulus of elasticity $E_x$ in $x$-direction.

![Diagram of a double U-shaped beam](attachment:image.png)

**Fig. 6.14 Doubling of the U-shaped beam for full-scale bending testing.**

**Full-scale four-point bending test.**

In Fig. 6.15 the four-point bending test setup is presented. The pair of bonded U-beams is loaded by an in-house developed 800 kN press facility. The bending load on the beam is increased by steps of 1000 N, intermitted by strain measurements of the 15 strain gages, positioned on the web and flanges as illustrated in Fig. 6.16. The relatively large number of straingages is applied to monitor the uniformity of the bending strain distribution.

![Schematic illustration of the full-scale four-point bending test](attachment:image.png)

**Fig. 6.15 Schematic illustration of the full-scale four-point bending test of the beam (distances in mm).**
Fig. 6.16 Positions of 15 attached straingages on the beam for deformation measurement during the full-scale bending test.

Between the two central loading points, the beam is subjected to a pure bending and a linear relation exists between the strain in x-direction ($\epsilon_x$) and the z-position of the strain gage (z):

$$\epsilon_x = \rho \cdot z$$  \hspace{1cm} \textit{<6.2.3.1>}

The curvature $\rho$ of the beam in the pure bending section is directly related to the bending stiffness $EI$ of the beam$^{110}$:

$$\rho = \frac{M}{EI}$$  \hspace{1cm} \textit{<6.2.3.2>}

where $M$ is the moment in the pure bending section,

$E$ is the modulus of elasticity in the x-direction of the beam,

$I$ is the moment of inertia with respect to the x-axis.

The modulus of elasticity $E$ of the material (in warp direction) can be determined from the $\epsilon_x$ strain gage measurements, using equations $<6.2.3.1>$ and $<6.2.3.2>$. The shear strain $\gamma_{xz}$ can be determined from the measurements of the "strain rosette" formed by the strain gages number 10, 5 and 11, to check the absence of shear strain in the pure bending section of the beam$^{107,110}$.

In Figs. 6.17 and 6.18, the strain $\epsilon_x$ is presented for the various z-positions as a function of increasing bending load (for both the crossply and the combiply beams). It can be noticed that the strain distribution in x-direction over the height of the beam is uniform during bending. In Figs. 6.19, it is demonstrated that the shear strain in the pure bending section of the beam can be neglected. The calculated E-moduli, abstracted from the mean values of the $\epsilon_x$ strain measurements are presented in Fig. 6.20.
Fig. 6.17 Measured strains in x-direction during the full-scale four-point bending test of the crossply beam.

Fig. 6.18 Measured strains in x-direction during the full-scale four-point bending test of the combiply beam.
Fig. 6.19a Measured shear strains during the full-scale four-point bending test in the pure bending section of the crossply beam.

Fig. 6.19b Measured shear strains during the full-scale four-point bending test in the pure bending section of the combiply beam.
Fig. 6.20 Moduli of elasticity of the crossply and combiply beam material, calculated from strain measurements during the full-scale four-point bending test.

The deviations of the moduli of elasticity at the start and at the end of the load ramp are probably caused by slack of the bending setup and lateral displacement of the beam, respectively. These deviations are neglected.

Clamped edge test.

In Fig. 6.21, the clamped edge test setup (cantilever beams) is presented. The ends of two U-shaped beams are fixed backwards onto both sides of a rigidly supported steel I-bar. The other, free ends of the beams are bolted on a tapered steel bar as illustrated. At these ends, the beams are loaded by a force controlled screw-jack. During the loading of the beams the free ends are guided against premature buckling.

Fig. 6.22 shows the placement of the strain gages, which are used to monitor the strains in length direction of the beams. The strain gages are also used in "strain rosettes" to measure the shear strain $\gamma_{xz}$. The strain measurements $e_x$ are presented in Fig. 6.23 and 6.24 for the crossply and combiply beams, respectively. Like in the four-point bending test, a uniform strain distribution over the height of the beam during bending is measured.

*Measurements of the bending deflection by linear voltage displacement transducers failed because of slight beam deflections perpendicular to the z-axis, discontinuities at the clamping and slack at the clamping.
Fig. 6.21 Schematic illustration of the (full-scale) clamped edge test setup.

Fig. 6.22 Positions of the attached straingages on the beam for the clamped edge bending test.
Fig. 6.23a Measured strains in x-direction during the clamped edge bending test of the crossply beam at position A.

Fig. 6.23b Measured strains in x-direction during the clamped edge bending test of the crossply beam at position B.
Fig. 6.24a Measured strains in x-direction during the clamped edge bending test of the combiply beam at position A.

Fig. 6.24b Measured strains in x-direction during the clamped edge bending test of the combiply beam at position B.
Equations \( <6.2.3.1> \) and \( <6.2.3.2> \) are used to calculate the modulus \( E_x \) from the strain measurements\(^{59}\). The calculated moduli of elasticity \( E_x \) (averaged over the strain gage positions) during the loading of the clamped edge beams are presented in Fig. 6.25 in both monitored cross-sections A and B. The calculated \( E_x \)-values from the measurements at position A approach those obtained in the full-scale four-point bending tests. The values at B show slight deviations, which can be caused by the fact that position B is close to (or situated in the disturbed tip area of the beam.

![Graph showing modulus of elasticity for crossply and combiply beam material calculated for different positions.](image)

**Fig. 6.25** Moduli of elasticity of the crossply and combiply beam material, calculated from strain measurements during the full-scale clamped edge bending test.

The shear modulus \( G_{\tau x} \) of the crossply and combiply material has to be calculated from the shear strain \( \gamma_{\tau x} \). They are related by:

\[
\tau_{\tau x} = G_{\tau x} \cdot \gamma_{\tau x}
\]

\(<6.2.3.3>\)

where \( \tau \) is the local shear stress. According to the beam theory\(^{107,116}\), the shear stress is varying over the height of the web. It can be calculated with:

---

\(^{59}\)This is valid because their curvatures \( \rho \) are measured only locally (although the cantilever beams are not in a state of pure bending during the tip loading).
\[
\tau_{xz} = \tau(v) = \frac{D_z \cdot S_{yy}}{I_y \cdot t_w} = \frac{D_z}{I_y \cdot t_w} (t_f \cdot b \cdot \sqrt{2}(h - t_f) + t_w \cdot v \cdot \sqrt{2}(h - 2t_f - v))
\]

where \( D_z \) is the tip load,

\( S_{yy} \) is the static moment with respect to the y-axis,

\( I_y \) is the moment of inertia with respect to the y-axis,

\( t_f \) and \( t_w \) are the thickness of the flange and the web of the beam, respectively,

\( h \) is the height of the beam,

\( v \) is the z-coordinate, according to Fig. 6.26.

Fig. 6.26 Geometry of the cantilever beams (\( L = 1500 \, mm \), \( h = 200 \, mm \), \( b = 25 \, mm \) and \( t_f = t_w = 2.5 \, mm \)).

The measured shear strain distribution over the height of the beams is presented in Figs. 6.27. The hyperbolic distribution, which is predicted by the isotropic beam theory, is more distinct in the results of the crossply beams than in those of the combiply beams. The visually monitored influence of lateral deflections at higher loads is clearly present. The calculated shear modulus for the combiply beams is ambiguous, because of this shear strain distribution. The shear modulus for the crossply beams is rather constant over the height of the beam and is approximately 3.9 GPa. Apparently, the results of the clamped edge tests must be considered with caution.

6.3 The influence of rubber forming on the stiffness properties.

Quantitative information about the expected final material properties is necessary, in order to design structural composite products to be manufactured with the rubber forming process.

In this section, the test results of the full-size beams are reviewed and compared to the results of tests on the coupons, from material before and after rubber forming. It is demonstrated that a "production factor" can be abstracted from test results. This factor can be used as a design tool to quantify the influence of the rubber forming process on CFRTP material.
Fig. 6.27a Measured shear strains at beam position A during the clamped edge test of the crossply beam.

Fig. 6.27b Measured shear strains at beam position A during the clamped edge test of the combiply beam.
6.3.1 Effect of a rubber forming cycle on the final product quality.

In Figs. 6.28 and 6.29, a comparison is presented of the various E-moduli as they were evaluated in 6.2. In Fig. 6.28a, the left bar represents the E-value in warp direction as it was supplied by the material manufacturer (Appendix C1). In Fig. 6.28b, the left bar gives the combibly E-modulus in "warp" direction\(^6\), calculated from the supplier's material data using the classical laminate theory\(^7\). The other bars in the figures indicate the averaged stiffness results of mechanical tests. The results are grouped according to the test method and specimen origin.

In Fig. 6.30, a comparison is given between the shear modulus of the combibly beam as given by the material manufacturer and the calculated values from the clamped edge test. Because of the unreliableness of the shear strain measurements (Section 6.2.3), the latter results are not discussed here.

![Graph showing average modulus of elasticity](image)

**Fig. 6.28a Average moduli of elasticity in warp direction of the crossply beams, determined by coupon and full-scale tests.**

\(^6\)As stated earlier, the warp direction of the material here is the direction in which most of the warp fibre bundles are oriented.

\(^7\)For the calculations, the computer program "Plamor" is used, which is developed at the Delft University to facilitate the stiffness calculations of laminated composites\(^{121}\).
Fig. 6.28b Average moduli of elasticity in warp direction of the combiply beams, determined by coupon and full-scale tests.

Fig. 6.29a Average moduli of elasticity in weft direction of the crossply beams, determined by coupon tests.
Fig. 6.29b Average moduli of elasticity in weft direction of the combiply beams, determined by coupon tests.

Fig. 6.30 Average shear moduli of the combiply beams, determined by the clamped edge test.
The stiffness data, obtained from the coupon four-point bending tests show lower E-values than those measured with the tensile tests, especially for the crossply material. It must be realized however, that the bending of a full-scale product always involves combined loading of the material. The material is neither fully loaded in tension nor fully in bending during the full-scale bending test of the beams. The comparison between the stiffness results out of coupon testing and the stiffness results out of full-scale testing must be considered with caution.

Conclusions from the coupon tests

The first conclusion drawn from the test results is that the mechanical properties in the weft directions are exaggerated in the manufacturer’s material specification. This specification is adequate only for the warp direction of both the crossply and the combibply material. The indicated exaggeration of material properties underlines the importance of verifying the quality of starting material.

The mechanical properties of the composite material slightly decrease during the rubber forming cycle. The fact that both the properties in warp direction and in weft direction diminish, demonstrates the influence of a non-optimal reconsolidation of the product. Probably because the high cooling rate of the beam by the use of the cold metal die does not provide an ideal consolidation environment.

The expected lower pressure at both ends of a beam during reconsolidation hardly influences the local material properties, as can be noticed from Figs. 6.8, 6.9 and 6.12, 6.13. Furthermore, the stiffness of the composite material in the flanges is the same as the stiffness of the material in the web (Fig. 6.31). This phenomenon supports the conclusion from Chapter 5 that the pressure on a laminate during consolidation only has a limited effect on its final mechanical properties, provided that the pressure exceeds a certain minimum level.

When the results for the stiffness properties in warp direction (Fig. 6.28) are compared to the stiffness properties in weft direction (Fig. 6.29), it can be noticed that the difference between the test results of the beam coupons and those of the flat laminate coupons are less for the properties in weft direction. This lower difference between material properties before and after rubber forming can be the result of stretching forces during the forming phase (Section 5.4.2), which are directed in weft direction in this case.

Both the effect of reconsolidation and the effect of stretching forces in the forming phase are process determined (Chapter 5) and appear to be the most influencing effects on the CFRTP material during the rubber forming of a U-shaped product.
Fig. 6.31a Comparison between calculated moduli of elasticity of material coupon tests out of the web and flanges of the crossply beam.

Fig. 6.31b Comparison between calculated moduli of elasticity of material coupon tests out of the web and flanges of the combiplly beam.
Conclusion from the full-scale tests.

An important conclusion is that the two different full-scale tests yield the same material stiffness in warp direction. They differ only slightly from the stiffness of the beam coupon tests (Fig. 6.28), which shows that the engineering beam theory (equations 6.2.3.1 and 6.2.3.2) can be used to calculate the moduli of elasticity in warp direction of both the full-scale four-point bending test and the clamped edge bending test from the measured strains. The clamped edge test also yields nearly equal stiffness of the beam in position A and position B (Fig. 6.25). This means that local material deficiencies like the prepreg seams in the ±45° layers or the rubber forming seam only have a limited effect on the stiffness of the rubber formed part.

6.3.2 Determination of the required starting material.

The mechanical requirements that have to be fulfilled in practice define the composition of the starting material necessary for a structural part. A composite part produced by a newly developed production technique, like the rubber forming technique, suffers from many still unknown process influences. This complicates the design of structures that are to be produced with such a technique. In this section the most important influences will be summarized, from the starting of the process to a final, rubber formed product. The rubber forming of the U-shaped beam is used to show how the necessary starting material can be determined in advance.

The stacking order of the prepregs plays an important role in the properties of a laminate. To avoid thermally and mechanically induced warping of a rubber formed product, the stacking must occur in a symmetrical way with respect to the mid-plane of the laminate. A fabric reinforced prepreg layer in itself is often not really symmetrically reinforced: depending on the kind of weave, the warp fibre bundles can dominate on one side of the laminate whereas the weft fibre bundles dominate on the other side (Fig. 6.32). In bending applications, a prepreg is most effective when most of the outer fibre bundles are directed in the load direction of the product. This is demonstrated by the results of bending tests on coupons out of two differently stacked laminates are presented in Figs. 6.33 and 6.34. Both 8-layered 0,90° layups consist of symmetrical satin weave C/PEI prepregs. In layup I, all the prepreg layers are stacked with the warp dominated surface outwards. In layup II however, the two outer prepreg layers have their weft dominated surface outwards. The remaining prepreg layers are warp dominated outwards.

The preconsolidation quality of the stacked prepreg material is still present in the final product. This phenomenon was already shown in 5.4 and has to be taken into account when calculating the properties of the final product. In the preceding sections it is demonstrated that it is not unusual that the real properties of the starting material differ from the properties presented by the material manufacturer. It is important to carry out material tests on representative coupon specimens to ensure a reliable knowledge of starting properties.
Fig. 6.32 Surface dominance of a specific fibre direction in a 5H satin fabric reinforced PEI prepreg.

Fig. 6.33 Mechanical influence of the stacking sequence of laminates, determined by the four-point bending test.
Fig. 6.34 Mechanical influence of the stacking sequence of laminates, determined by the tensile test.

When finally the CFRTP sheet is rubber formed, its mechanical properties are influenced again. Although the mechanical properties are not necessarily reduced (Chapter 5), the influence of the rubber forming cycle must be monitored carefully. As shown before, this influence strongly depends on the applied process parameters, the kind of material and the shape of the final product.

In Section 6.2.3, it was shown that the bending stiffness of the U-shaped beam is related to the stiffness properties of material coupons. So, if the required mechanical performance of a product is known, it is possible to calculate backwards the required properties of the material after rubber forming. Then the estimated influences of the rubber forming cycle and the stacking and preconsolidation subprocess are included. Finally (e.g. with the help of the classical laminate theory as it was mentioned in Section 6.2.1), the required mechanical properties of the prepreg material are determined and the necessary starting material can be estimated.

6.3.3 Introduction of the production-factor concept.

To quantify the influence of a rubber forming cycle on the mechanical properties of the composite material, a so-called production factor will be introduced. The production-factor $f_p$ can be defined as the fraction of the mechanical properties of the starting material that is realized after the material is rubber formed into a final product:
\[ f_p = \frac{\text{mechanical properties after rubber forming}}{\text{mechanical properties of starting material}} \]

The purpose of the introduction of a production-factor is to facilitate the quantification of the rubber forming influences under different processing conditions. By defining a specific processing condition, the designer of a CFRTP part is able to predict the final mechanical properties of a product more accurately. In Fig. 6.35, it is illustrated how a production factor for a specific forming environment closes the gap of uncertainty between the properties of the preconsolidated stacking material and the (local) material properties of the final product.

From Chapters 4 and 5, it can be concluded that the mechanical properties of rubber formed material and thus the production-factors are influenced by:

a) the composite material itself:
   - kind of material (reinforcement and resin)
   - thickness of the material
   - preconsolidation

b) the rubber forming environment:
   - starting temperature of the composite material
   - temperature of the moulds
   - mould material
   - pressure on the material
   - blankholder forces
   - forming speed
   - matching of the rubber and the rigid mould

c) the final product:
   - shape of the product (deformation sequence)
   - orientation of the reinforcement.

As an example, the calculated production-factors determined by the mechanical experiments on the relatively simple beam in the preceding sections are presented in Fig. 6.36. From the figure, the estimated production-factors are different for the different material directions warp and weft. They also depend on the layups of the material and the test method.
It is a difficult but necessary task to investigate the influences of the mentioned variables thoroughly. An example of necessary parameter understanding is described in Section 5.2.3 by equation <5.2.3.6> (covering the temperature dependence of mechanical properties). Although in Chapters 2, 4 and 5 the most important areas are covered, a vast amount of additional material investigations is necessary to be able to accurately design fabric reinforced thermoplastic structural parts. For the time being, a production-factor $f_p = 0.9$ is proposed for the use in the calculation of the bending stiffness of the rubber formed U-shaped beam, as described in the present chapter.

6.4 Quality control by process control.

In this work, it is demonstrated that it is mainly the processing environment that affects the quality of rubber formed parts of a specific product material and shape. The process parameters temperature, pressure and time are most important in the non-isothermal and non-isobaric, high-speed rubber forming technique. Especially in Chapters 3 and 4, it is explained how these process parameters can be influenced.
Fig. 6.36 Production-factors, calculated from mechanical tests on C/PEI material coupons.

The mechanical quality of the rubber formed parts can be assured, only if the rubber forming process is reproducible. The introduction of the production-factor for example is only useful if it is possible to reproduce processing conditions for each product, that fall inside a narrow process window. The technical development of the rubber forming technique, therefore, is necessary to guarantee a well controlled processing environment. The control of the rubber forming parameters can be improved by the full automation of the process, as described in 3.4. Apart from the increase of the manufacturing speed, the automation of the rubber forming technique facilitates the reproducibility of the applied process parameters.

Because of the achievement of a better understanding and control of thermoforming in general, the quality of the processing of continuous fibre reinforced thermoplastic products has been increased. The technical development of the rubber forming process, however, is being continued. This development, combined with research work on the response of the CFRTP material itself, is necessary to achieve an important goal in the manufacturing of composite parts: quality control by process control.
CHAPTER 7. Concluding remarks.

7.1 Conclusions.

A thermoforming technique for the production of composite parts out of fabric reinforced thermoplastic sheets is presented in this thesis. The technique is called rubber forming, because of the use of a flexible rubber mould to shape a hot composite sheet in a rigid (metal) mould. In order to successfully introduce rubber forming as a fast and efficient thermoforming technique, it is shown necessary to increase the existing knowledge and understanding of:

- the deformation of fabric reinforced thermoplastic sheets into three dimensional shapes during a thermoforming cycle (Chapter 2),
- the necessary process parameters for rubber forming (Chapters 3 and 4),
- the physical response of continuous fibre reinforced thermoplastic material on a rubber forming cycle (Chapters 5 and 6).

The rubber forming technique is proposed as an efficient thermoforming technique for the realisation of composite products in both the automotive and the aerospace industry. Therefore, practical solutions are pursued to solve manufacturing problems of thermoplastic composite products. In general, the solutions are supported by theoretical analysis and focused experiments.

From the analysis and experimental results of investigations into the deformation of fabric reinforced thermoplastics as they were discussed in Chapter 2, it can be concluded that:

☐ the two main deformation modes of the composite material during thermoforming, interply slipping and intraply shearing, are considerably facilitated by the application of high temperature, low surface pressure and low deformation velocity;

☐ the interply slip phenomenon can be modelled with a combination of powerlaw viscous behaviour of the polymer matrix interlayers and a mechanical sticking behaviour of the adjacent surfaces of the fabric reinforcement; the fabric layers inside a laminate have to be separated first before the actual interply slipping of the layers is started;

☐ the internal forces, necessary for intraply shearing of fabric reinforced thermoplastics can be qualitatively described by the Kawabata Finite-Deformation-Theory; the friction inside the reinforcing fabric determines the shearing behaviour of the composite laminate and the viscous thermoplastic matrix mainly lubricates the shearing fibre yarns.
the imposed deformation sequence of the composite sheet and thus the mould design are most critical aspects in the realization of sufficient material deformation in a thermoforming process.

The processing conditions during thermoforming can be optimized by technical means. To support the control of the process parameters, theoretical analysis of the heat flow and the pressurization of the composite material are proven to be valuable (Chapter 3 and 4):

- it is shown that replacing one of the two rigid moulds of a matched-die technique by a flexible rubber offers a better control of the pressurization of the laminate in both the forming phase and the consolidation phase of the thermoforming cycle; using a rubber mould, the forming sequence of a laminate sheet is less dependent on the product shape and possible changes in the thickness of the composite material (e.g. due to intraply shearing deformation) can be easily dealt with;

- rubber deformation theories like the theory of shape factors can be used for the design and qualitative description of the behaviour of rubber moulds in a rubber forming process;

- the heat flow during the strongly non-isothermal rubber forming cycles must be controlled to a high extent; heating and cooling histories of the thermoplastic laminates can be calculated using a one-dimensional energy equation; numerical computer programs are shown to be useful to determine the influence of technical process changes, provided that the thermal properties of the processed composite material are well known;

- wrinkling of the composite material during forming can be caused by deformation induced compression stresses in the plane of the laminate; it is technically possible to apply external tension forces on the deforming laminate to prevent this wrinkling;

- the full automation of the rubber forming technique is realized (it is, for instance, possible to rubber form a full-size car door every 55 seconds).

Investigations into the effect of rubber forming on the material properties of fabric reinforced thermoplastic composites (Chapter 5 and 6) have shown that:

- the influence of non-isothermal consolidation during rubber forming on the mechanical properties of the rubber formed material can be described and predicted from the temperature history of the material; the autohesion phenomenon determines the consolidation quality whereas the pressure during consolidation in a rubber forming cycle is less critical; the change of material thickness during thermoforming is shown to be inversely proportional to the change of mechanical properties of the material;
the fabric reinforcement can be slightly stretched during rubber forming due to the tension loading of the fibres; this increases the mechanical properties of the material in the stretch direction;

to avoid residual stresses and product shape distortions, it is important to minimize cooling rates during thermoforming and related internal laminate stresses;

the total influence of a rubber forming cycle, performed by the automated thermoforming facility, on the mechanical properties of CFRTP material is small and can be quantified by a so-called production-factor; the production-factor can be used to calculate the amount of starting material that is necessary to realize a specific mechanical performance of the rubber formed final product; the production-factor mainly depends on the physical condition of the composite starting material and the rubber forming process conditions.

7.2 Recommendations for future work.

The major recommendations for future work on rubber forming of continuous fibre reinforced thermoplastics can be summarized by two, quite general statements:
- fill in the gaps in the understanding of the thermoforming behaviour of thermoplastic composites;
- expand the flexibility in the application of thermoforming process parameters.

The technical and the material aspects have to be thoroughly combined to optimize the processing environment, this also includes the design of the (rubber) moulds. An important unknown factor is also the service-lif of rubber moulds which requires more research.

With respect to the prediction of the behaviour of thermoformed thermoplastic composites, a further coupling of intraply shear and interply slip phenomena of the deforming laminate as a function of the processing parameters is necessary. The interaction of internal forming forces and final material properties is to be investigated more precisely, too. It is shown that the use of high additional forming forces to facilitate the shaping of a CFRTP product can introduce important residual stresses in the product. The influence of these stresses on the mechanical properties and the shape of the product must be known to optimize the application of additional forming forces.

As was discussed in Chapter 5, the non-isothermal consolidation during a fast thermoforming process mainly determines the mechanical material properties. More research work must be carried out to relate those final properties to the application of the process parameters temperature, time and pressure during consolidation. Subsequently, a reliable consolidation model has to be implemented to calculate the mechanical properties of a final product out of a monitored (non-isothermal) rubber forming cycle to replace the preliminary
relation <5.2.3.1>.

In the present work only glass- and carbon fabric reinforced PolyEtherImide (CETEX*) and glass fabric reinforced PolyEthyleneTerephthalate (DESTEX*) have been used to show that rubber forming offers the potential to shape good quality three-dimensional CFRTP products. The investigations, that are performed to demonstrate the good control of pressure, temperature and forming force during the process, need to be extended to a large variety of product shapes and composite materials.

The availability of a broader knowledge of the material response on rubber forming is a precondition to present rubber forming as a reliable manufacturing technique of composite parts. A more thorough investigation of the change of material properties during a rubber forming cycle is necessary. Only then, the production-factor can be developed as a quantitative function of the applied processing conditions and the quality of each product can be assured by the control of the forming process.

The present work must be considered as a first step in the realisation of a fast and efficient thermoforming method of continuous fibre reinforced thermoplastics. The rubber forming technique is developed to show the potential of these materials for cost-effective, high quality composite parts. The present work provides constitutive equations, describing separate stages of the rubber forming process, which are verified for those separate stages. Complete integration of the equations in a numerical program, describing the complete process, and the resulting production factor for the part performance was beyond the scope of this thesis. However, the present results indicate that such a final integration may be quite well possible and it is strongly recommended for future work.
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**Acknowledgement.**

The author wishes to express his gratitude towards all who contributed to the research and helped to make the present work possible. Financial support has been gratefully received from DSM Research B.V., The Netherlands.
Summary.

Until now, continuous fibre reinforced thermoplastic composites are not applied at the large scale that was expected at their introduction. Because of the lack of suitable thermoforming techniques, the potential product manufacturing advantages could not be realized.

In the present work, a fast press technique is presented to shape flat thermoplastic laminate sheets into two- or three-dimensional composite products. In this so called rubber forming technique, a flexible, preshaped rubber die is used to form a softened laminate in/over a rigid (metal) mould.

For the change of flat, laminated sheets into products, two material deformation modes are pointed out; interply slipping of the layers in a laminate and intraply shearing (trellis effect) of the fabric reinforcement in each individual layer. Forming experiments show that the interply slipping in a laminate starts after two adjacent layers are pulled apart. The actual slipping of the layers is ruled by a powerlaw shear behaviour of the viscous thermoplastic resin between the layers. Intraply shearing is mainly determined by the surface friction of the interlacing fibres at the crossover points in a fabric layer. The required forming forces for both deformation modes are largely determined by the temperature, the surface pressure and the imposed deformation velocity of the material.

In order to develop the rubber forming technique into an efficient thermoforming process, technical improvements are carried out according to the results of the preceding analysis of the forming and consolidation behaviour of thermoplastic laminates. Calculations of the heating and cooling curves of laminated materials during the rubber forming process point out the necessity of heat flow optimization. The calculations show good agreement with experimental results. Pressure experiments and calculations demonstrate the strong influence of the geometry and hardness of the rubber mould on the pressure distribution over the surface of a rubber formed laminate. It is shown that additional (forming) forces can be applied to facilitate material deformation and avoid material wrinkling.

The mechanical material response to the rubber forming process is estimated to enable design calculations of structural rubber formed composite parts. The consolidation of the laminate is shown to be governed mainly by autohesion of the prepreg layers and is therefore most sensitive to the applied process temperature. The influence of pressure during consolidation on the mechanical properties of a final product is small. The mechanical properties of a composite material are not necessarily lowered by typical rubber forming subprocesses like the application of additional forming forces and the relatively fast, non-isothermal consolidation. The reduction of the mechanical properties during a complete rubber forming cycle can be quantified by a so-called production factor.
The viability of rubber forming is demonstrated by the realisation of a 4000 kN, fully automated thermoforming unit, which is proven to be capable of rubber forming, for instance, a composite car door every 55 seconds.
Samenvatting.

Continue-vezelversterkte thermoplasten worden tot op heden niet zo veelvuldig toegepast als werd verwacht ten tijde van hun introductie. Door het gebrek aan de juiste thermovormtechnieken konden de potentiële productievoordelen van deze materialen niet gerealiseerd worden.

In dit proefschrift wordt een snelle perstechnick gepresenteerd om vlakke thermoplastische laminaten te vervormen tot twee- of driedimensionale composiet-producten. In deze zogenaamde rubbervormtechniek wordt een flexibele, voorgevormde rubberen mal gebruikt om een verweekte laminaat in/over een stijve (metalen) mal te vormen.

Voor de verandering van vlakke laminaten in producten kunnen twee materiaal-vervormingen verantwoordelijk zijn; interlamineaire slip (interply slip) van de lagen in een laminaat en intralaminaire scharing (intraply shear) van de weefselsterkting van een individuele laag. Vervormingsexperimenten tonen aan dat interlamineaire slip in een laminaat pas kan plaatsvinden na het lostrekken van twee buurlagen. Het echte slippen van de lagen wordt gedomineerd door de viscoseuze matrix tussen de lagen. Dit slipgedrag voldoet aan een machtsregel. Het interlamineaire scharren van een weefsellag wordt voornamelijk gedomineerd door de knooppuntswijziging van de in elkaar gewoven vezelbundels. De voor de materiaalvervormingen benodigde vervormingskrachten worden grotendeels bepaald door de temperatuur, de oppervlaktezwarendruk en de opgelegde vervormingssnelheid van het materiaal. Deze invloeden kunnen beschreven worden met bestaande vervormingsmodellen.

Om de rubbervormtechniek te ontwikkelen tot een efficiënt thermovormproces zijn technische verbeteringen doorgevoerd, naar aanleiding van de resultaten van de voorgaande beschouwingen over het vervormings- en consolidatiegedrag van thermoplastische laminaten. De optimalisatie van warmte-overdracht gedurende het opwarmen en afkoelen van het gelamineerde materiaal blijkt noodzakelijk te zijn. Dit wordt geconcludeerd na experimentele verificatie van berekeningen van de temperatuurscurven van het materiaal. Met behulp van drukberekeningen en metingen wordt de grote invloed aangetoond van de geometrie en de hardheid van rubberen stempels op de drukverdeling over het oppervlak van een gerubbervormd laminaat. Ook wordt gedemonstreerd dat additionele vormkrachten kunnen worden toegepast om de materiaalvervormingen te stimuleren en plooivorming in het product tegen te gaan.

Het bestaansrecht van rubbervormen wordt aangetoond door de doorgevoerde automatisering van een thermovorminstallatie met een perscapaciteit van 4000 kN, waarmee daadwerkelijk bijvoorbeeld composieten autodeuren iedere 55 seconden worden gerubbervormd.
De invloed van het rubbervormproces op de mechanische eigenschappen van het composietmateriaal is onderzocht. Dit is nodig om ontwerpberekeningen mogelijk te maken voor structurele, gerubbervormde composietdelen. Het blijkt dat de consolidatie van het laminaat voornamelijk afhankelijk is van de zogenaamde autohesie tussen de prepreglagen. De consolidatie van een laminaat is daarmee het meest gevoelig voor de toegepaste processtemperatuur. De oppervlaktedruk op het consoliderende laminaat is minder belangrijk. De mechanische eigenschappen van het composietmateriaal worden niet noodzakelijkerwijs verlaagd tijdens de typische rubbervorm subprocessen zoals het gebruik van additionele vormkrachten en de relatief snelle, niet-isotherme consolidatie. Over een gehele rubbervormcyclus genomen, is de verandering van materiaaleigenschappen klein. Deze verandering kan kwantitatief weergegeven worden door een zogenaamde productiefactor.

About the author.

Lucien Robroek was born in Heerlen in the Netherlands on March 23, 1967. In 1985 he graduated from the Atheneum of the Coriovalium College in Heerlen. In the same year he started his study at the Faculty of Aerospace Engineering at the Delft University of Technology in Delft, the Netherlands. He joined the research group "Vormgeving, Fabricage en Materialen" (Structures and Materials) in 1988 and graduated under the supervision of Professor Dr.ir. Th. de Jong and Ir. A. Beukers in 1990. The author's master thesis was titled: The manufacturing process of continuous fibre reinforced thermoplastics in the TUBE MOULD. After his graduation he started his PhD-research on the processing of thermoplastic composites in the Structures and Materials Laboratory, under the supervision of Professor Dr.ir. Th. de Jong. The results of the research are described in the present work.
Appendix A: Calculation of the boundary conditions in case of radiation heating.

A surface in an environment will receive a certain amount of radiation from that environment. This will be partly reflected, partly absorbed and partly allowed to pass, depending on the kind and temperature of the surface. The total energy coming from a surface (reflection from other surfaces plus its emission) is called radiosity. The radiation emitted from a surface with no reflection at all (a so-called black surface) is related to the temperature of the surface as follows:

\[ \varphi_{bl} = \sigma \cdot T^4 \]  \hspace{1cm} \text{\textit{\textcopyright A.1}}

where \( \varphi_{bl} \) is the radiation that is emitted by a black surface [J/sm²],

\( T \) is the temperature of the material [K],

\( \sigma \) is a constant [J/sm²K⁴].

The infra-red (IR) panels that are used in the thermoforming facility (NHR134) are electrically heated to emit mainly IR radiation. It is physically impossible for the IR radiators to act as real black surfaces. Therefore, their radiation will be lower than that of a black surface at the same temperature:

\[ \varphi_e = \varepsilon \varphi_{bl} \]  \hspace{1cm} \text{\textit{\textcopyright A.2}}

where \( \varphi_e \) is the radiation emitted by the IR heaters (≈ radiosity of the heaters) [J/sm²],

\( \varphi_{bl} \) is the radiation emitted by a black surface [J/sm²],

\( \varepsilon \) is the emission coefficient.

The emission coefficient \( \varepsilon \) depends on the kind and temperature of the radiating surface. In practice, \( \varepsilon \) can be put equal to the absorption coefficient \( \alpha \) of the surface, which usually can be found in data books.

The fraction of the totally emitted amount of energy of a surface 1 that will reach another surface 2 can be described by the use of a configuration factor \( F_{1,2} \):

\[ F_{1,2} = \frac{1}{A_1} \int_{A_1} \int_{A_2} \frac{(\cos \phi_1 \cdot \cos \phi_2)}{r^2} \, dA_1 \, dA_2 \]  \hspace{1cm} \text{\textit{\textcopyright A.3}}

where \( A_1 \) and \( A_2 \) are the areas of the emitting and receiving surfaces, respectively. \( \phi_1, \phi_2 \) and \( r \) are geometrical variables as depicted in Fig. a.1.

It is possible to determine the boundary conditions needed for the solution of an energy equation from equations \textit{\textcopyright A.1} to \textit{\textcopyright A.3}, in the case of a laminate that is heated by IR panels.
Fig. a.1 Relative positions of a radiation emitting surface ($A_1$) and a radiation receiving surface ($A_2$).
Appendix B: Numerical calculation of the heat transfer through thin materials.

Two numerical computer programs are developed for the calculation of the temperature of a laminate as a function of time during a rubber forming cycle. One program calculates the infrared heating, the other calculates the conduction heat transfer in a laminate imposed to various boundaries. The software programs are developed in the QuickBasic programming environment of LabWindows® version 2.0.

Infrared heating model.

The calculation model for the infrared heating of a composite sheet between the radiation panels is based on a theory in which an environment is completely bounded by specified surfaces. These surfaces can be denoted to be insulated surfaces (total reflection), open surfaces (black surfaces at room temperature) or radiation surfaces. The model is developed to enable the calculation of the required IR heating time of a thermoplastic laminate in the Fontijne NHIR134 thermoforming setup in the laboratory.

![IR radiation panels and laminate](image)

Fig. b.1 Representation of ¼ of a laminate inside the infrared heating oven.

Because of the symmetry of heating oven and laminate sheet, only ¼ of the total system needs to be considered (Fig. b.1). In this figure, surfaces 1, 2, 3, 4 represent the infrared heating zones. In reality as well as in the model, they can be switch on separately. Surfaces 5, 6, 7, 8 are planes of horizontal symmetry of the heating "oven", surface 5 represents ¼
of the upper laminate surface. Surfaces 9, 10 are open surfaces around the panels and surfaces 11, 12 are planes of vertical symmetry of the heating oven, which are modelled as ideally insulated planes.

For each of these surfaces the configuration factor \( F_{ij} \) (Appendix A) can be calculated by using equation \(<A.3>\) and Fig. a.1. For the twelve surfaces this results in a 12 x 12 configuration matrix \( F \).

For every so-called grey\(^8\) surface (like a laminate), the radiosity \( \varphi_i \) equals the sum of its own emission and its reflection of the radiation received from the other surfaces:

\[
\varphi_i = \varepsilon_i \cdot \varphi_{i_{\text{em}}} + r_i \cdot (F_{i-1} \varphi_1 + F_{i-2} \varphi_2 + \ldots + F_{i-n} \varphi_n)
\]

\(<B.1>\)

where \( r_i \) is the reflection coefficient and from \(<A.1>: \varphi_{i_{\text{em}}} = \sigma \cdot T_i \). For solid materials\(^76,73\) \( r_i \approx 1 - \varepsilon_i \approx 1 - \varepsilon_i \). This system leaves 12 linear equations with 12 unknown parameters \( \varphi_i \) to be solved.

When the radiosity of a surface is calculated, it is possible to estimate the nett specific energy that is absorbed from the other surfaces:

\[
\varphi_{i_{\text{nett}}} = (F_{i-1} \varphi_1 + F_{i-2} \varphi_2 + \ldots + F_{i-n} \varphi_n) - \varphi_i
\]

\(<B.2>\)

The totally radiated nett energy of that surface then is:

\[
\Phi_{i_{\text{nett}}} = A_i \cdot \varphi_{i_{\text{nett}}}
\]

\(<B.3>\)

This energy \( \Phi_{\text{nett}} \) is added to the surface of the laminate and thus is the input heat flow into the material. It is used as the boundary condition for the calculation of the heat transfer through the thickness of the laminate by conduction\(^9\). During the radiation heating of the laminate, its surface temperature will rise, which causes an increase of the radiosity \( \varphi_i \) and thus a decrease of absorbed energy \( \Phi_{\text{nett}} \) with time. Therefore, the surface characteristics of the heating laminate have to be recalculated after each time increment \( dt \).

**Heat conduction model.**

The different areas in a rubber formed laminate are subject to different cooling environments, as is discussed in Section 4.2.1. The boundary conditions over the laminate

\(^8\)A grey surface is a surface that partly reflects and partly absorbs radiation from another surface.

\(^9\) For the numerical evaluation of every new temperature profile through the laminate, a forward difference approximation is presented in the next part of this appendix.
surfaces change gradually compared to the changes over the thickness of the laminate. Therefore, the assumption is made that neighbouring laminate cross sections are not influenced by each other\(^4\). The heat transfer problem of a cooling laminate is simplified to a one-dimensional problem through the thickness of the laminate.

The one-dimensional non-steady conduction equation <4.2.1.5> from Section 4.2.1 can be rewritten by a forward difference approximation\(^7\) (which is called "explicit"), using a Taylor expansion around \(x=x\) and \(t=t\) into:

\[
\frac{k}{dx^2} (T(x-dx,t) - 2T(x,t) + T(x+dx,t)) = \frac{\rho \cdot Cp}{dt} (T(x,t+dt) - T(x,t)) \tag{B.4}
\]

where \(k\) is the heat conduction, \(T(x,t)\) is the temperature on location \(x\) at time \(t\), \(\rho\) is the specific weight and \(C_p\) is the specific heat of the material.

For (one-dimensional) heat transfer in a bar, consisting of three neighbouring nodes \(b\), \(a\) and \(c\), respectively, <B.4> can be represented by:

\[
\frac{k}{dx^2} (T_b - 2T_a + T_c) = \frac{\rho \cdot Cp}{dt} (T'_a - T_a) \tag{B.5}
\]

where \(T_{a,b,c}\) are temperatures at \(t=t\) of node \(a\), \(b\), \(c\) respectively, and \(T'_a\) is the temperature of node \(a\) at \(t=t+dt\). This expression gives the future temperature \(T'_a\) at node \(a\), in terms of the present temperatures at node \(a\) and surrounding nodes \(b\) and \(c\).

If the specified nodes represent the centres of little lumps of material with volume \(V=A \cdot dx\) (Fig. b.2), the total heat flow is considered by multiplying both sides of equation <B.5> by the lump volume:

\[
\frac{T_b - T_a}{dx/k \cdot A} + \frac{T_c - T_a}{dx/k \cdot A} = \frac{V \cdot \rho \cdot Cp}{dt} (T'_a - T_a) \tag{B.6}
\]

The thermal capacity \(C_s\) of the lump volume surrounding the node \(a\) can be expressed by \(C_s=V \cdot \rho \cdot C_p\) and the heat resistance for conduction is defined as \(R=dx/k \cdot A\). With these expressions, the heat flow problem becomes equivalent to the equation for an electrical current in an electrical network, as it is represented in Fig. b.3:

\[
\frac{T_b - T_a}{R_{ab}} + \frac{T_c - T_a}{R_{ac}} = C_s \cdot \frac{T'_a - T_a}{dt} \tag{B.7}
\]

\(^*\) This also assumes the neglecting of temperature profile distortions by matrix flow phenomena as they occur during e.g. interply slip.
Fig. b.2 Lump presentation of a bar.

Fig. b.3 Nodal presentation of a one-dimensional heat flow.

From equation <B.7>, it can be concluded that if a geometry is divided into a nodal network, the following expression must be satisfied at each nodal point i:

$$\sum_j \frac{T_j - T_i}{R_{ij}} = \frac{C_i}{dt} (T_i - T_j) \Rightarrow T_i = T_i + dt (\sum_j \frac{T_j - T_i}{R_{ij} \cdot C_i})$$  \hspace{1cm} <B.8>

where $R_{ij} = \text{dx}/k \cdot A_{ij}$ for conduction boundaries, $R_{ij} = 1/h_{ij} \cdot A_{ij}$ for convection boundaries and $C_i = V_i \cdot \rho \cdot C_p$.

No iteration or matrix conversions are required for the calculation of the new temperature $T_i'$ of node i, when using the explicit expression <B.8>. A stability criterion is generally used in practical cases:

$$\sum_j \frac{dt}{R_{ij} \cdot C_i} \leq 1$$  \hspace{1cm} <B.9>

As long as a sufficiently small time increment $dt$ is chosen, the explicit expression will lead to satisfactory calculations.
Another approach for the calculation of a two-dimensional heat flow is possible by developing the *implicit* expression for equation <4.2.1.5> by Taylor expansions around $x=x$ and $t=t+\Delta t$, using the so-called backward finite difference. This leads to an expression for the future temperature at node $a$ in terms of its current value and the future values of its neighbour's temperatures. In that case however, a larger system of equations must be solved which leads to more programming work and longer calculation times.

The software program that is written to solve the heat conduction problem of a laminate during the whole rubber forming cycle successively calculates the new temperatures inside an 11-node laminate at each node, starting with a given initial temperature distribution. Time is then increased (satisfying the stability criterium <B.9>) and calculations are repeated. Several boundary conditions can be chosen in succession to simulate the subsequent cooling steps.

**Use of the heat transfer software.**

The heating program is active in the "LabWindows" environment and is interactive with its subprograms. To calculate the heating profile of a laminate, the following input is needed (Fig. b.4):

<table>
<thead>
<tr>
<th>Table B.1 Required input for the infrared heating software.</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>composite material data:</strong></td>
</tr>
<tr>
<td>specific weight $\rho$ [kg/m$^3$]</td>
</tr>
<tr>
<td>specific heat $C_p$ [J/kgK]</td>
</tr>
<tr>
<td>heat conductivity $k$ [W/mK]</td>
</tr>
<tr>
<td>emission coefficient $\epsilon$ [-]</td>
</tr>
</tbody>
</table>

The heat conduction program is also active in the LabWindows environment. Beside the material data of Table B.1, the characteristics of the environment like rigid mould, rubber mould, blankholder and insulation are needed as input (Fig. b.5). Then, the user can specify the subsequent cooling steps during the process. Finally, the local temperature profile through the laminate thickness is calculated as a function of time.
Fig. b.4 Input/output screen of the infrared heating software.

Fig. b.5 Input/output screen of the cooling software.
Appendix C1: Material data of the used CFRTP materials.

The three main fabric reinforced thermoplastic composites that are used during the investigations are:
- glass fabric reinforced PolyEtherImide (denoted G/PEI in the present work),
- carbon fabric reinforced PolyEtherImide (C/PEI in the present work),
- glass fabric reinforced PolyEthyleneTerephthalate (G/PET in the present work).

<table>
<thead>
<tr>
<th>Mechanical properties of G/PEI and C/PEI laminates*</th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>(CETEX manufactured by TenCate BV)</td>
<td></td>
</tr>
<tr>
<td>prepreg code</td>
<td>SS303 (G/PEI)</td>
</tr>
<tr>
<td>reinforcement</td>
<td>8H satin glass fabric</td>
</tr>
<tr>
<td>resin</td>
<td>PEI</td>
</tr>
<tr>
<td>density [kg/m$^3$]</td>
<td>1905</td>
</tr>
<tr>
<td>fibre volume [%]</td>
<td>50</td>
</tr>
<tr>
<td>tensile modulus [GPa]</td>
<td>20</td>
</tr>
<tr>
<td>tensile strength [MPa]</td>
<td>350</td>
</tr>
<tr>
<td>compr. strength [MPa]</td>
<td>230</td>
</tr>
<tr>
<td>shear modulus $G_{\text{TL}}$ [GPa]</td>
<td>5.0</td>
</tr>
<tr>
<td>shear modulus $G_{\text{I5}}$ [GPa]</td>
<td>7.9</td>
</tr>
<tr>
<td>consolidated thickness [mm]</td>
<td>0.23</td>
</tr>
</tbody>
</table>

*According to TenCate data sheets: Ten Cate Advanced Composites, P.O. Box 9, 7440 AA Nijverdal, The Netherlands.
### Mechanical properties of G/PET consolidated sheet**:
(DESTEX manufactured by DSM Resins BV)

<table>
<thead>
<tr>
<th>reinforcement</th>
<th>DESTEX TPC 3000 (G/PET)</th>
</tr>
</thead>
<tbody>
<tr>
<td>resin</td>
<td>glass fabric (50/50)</td>
</tr>
<tr>
<td>tensile modulus [GPa]</td>
<td>PET</td>
</tr>
<tr>
<td>tensile strength [MPa]</td>
<td>20</td>
</tr>
<tr>
<td>flexural modulus [GPa]</td>
<td>600</td>
</tr>
<tr>
<td>flexural strength [MPa]</td>
<td>23</td>
</tr>
<tr>
<td>ILLS [MPa]</td>
<td>550</td>
</tr>
<tr>
<td>fibre volume [%]</td>
<td>40</td>
</tr>
<tr>
<td>density [kg/m³]</td>
<td>50</td>
</tr>
<tr>
<td></td>
<td>1950</td>
</tr>
</tbody>
</table>

**According to DSM Resins data sheets: DSM Resins, P.O. Box 1151, 6160 BD Geleen, The Netherlands.
Appendix C2: Overview of rubber materials, used for rubber forming.

In the table below, the properties of Silicone rubber materials that can be used in rubber moulds are given. The properties are extracted from data sheets of the suppliers. The Silicone rubbers are composed from two or more fluid components that are blended in the appropriate ratio. The viscous blend is casted in the final mould form. PolyUrethane rubbers, obtained from Fokker Aircraft, are also used in the investigations. These rubbers are solid and have to be cut in order to realize a rubber mould. The PolyUrethane rubbers are available in several hardness degrees. No further information is quantified.

<table>
<thead>
<tr>
<th>type</th>
<th>hardness ShoreA</th>
<th>max. strain [%]</th>
<th>tensile strength [MPa]</th>
<th>tear strength [MPa]</th>
<th>max. temp. [°C]</th>
<th>casting viscosity [mPas]</th>
<th>avail. casting time [min]</th>
</tr>
</thead>
<tbody>
<tr>
<td>RTV265 + A60 1</td>
<td>2-3</td>
<td>350</td>
<td>1.15</td>
<td>1.0</td>
<td>5200</td>
<td>&gt; 300</td>
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</tr>
<tr>
<td>RTV265 + A61 1</td>
<td>5</td>
<td>450</td>
<td>2.0</td>
<td>4.0</td>
<td>9000</td>
<td>150</td>
<td></td>
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<tr>
<td>RTV260 + A28 1</td>
<td>18</td>
<td>680</td>
<td>4.5</td>
<td>13.5</td>
<td>38000</td>
<td>70</td>
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<tr>
<td>RTV260 + A20 1</td>
<td>56</td>
<td>220</td>
<td>4.5</td>
<td>4.5</td>
<td>240</td>
<td>100000</td>
<td>110</td>
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<tr>
<td>RTV4026+A98 1</td>
<td>67</td>
<td>71</td>
<td>6.1</td>
<td>2.4</td>
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<td>50</td>
<td></td>
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<tr>
<td>KE24 + Cat. 2</td>
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<td>50</td>
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<td>30</td>
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<tr>
<td>Silastic J 3</td>
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<td>250</td>
<td>5.5</td>
<td>14</td>
<td>100000</td>
<td>120</td>
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</table>

1 manufactured by Possehl Polymehrchemie GMBH, Germany
2 manufactured by Shin-Etsu, Japan
3 manufactured by Dow Corning GMBH, Germany
### Appendix C:

**Examples of thermoplastic composites components**

<table>
<thead>
<tr>
<th>polymer</th>
<th>temperature</th>
<th>tensile properties</th>
<th>composite material</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$T_m$ [°C]</td>
<td>strength [MPa]</td>
<td>modulus [MPa]</td>
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<tr>
<td>PP</td>
<td>10 165</td>
<td>31</td>
<td>1700</td>
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<tr>
<td>PA 6</td>
<td>180 215</td>
<td>45</td>
<td>1500</td>
</tr>
<tr>
<td>PA 12</td>
<td>160 190</td>
<td></td>
<td></td>
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<tr>
<td>PETP</td>
<td>85 260</td>
<td>DSM</td>
<td>Destex</td>
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<tr>
<td>PSU</td>
<td>190 -</td>
<td>76</td>
<td>2200</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>PES</td>
<td>220 -</td>
<td>83</td>
<td>2410</td>
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<tr>
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<td></td>
<td></td>
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<tr>
<td>PAS</td>
<td>220 -</td>
<td>82</td>
<td>2660</td>
</tr>
<tr>
<td>PEI</td>
<td>215 -</td>
<td>110</td>
<td>3300</td>
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<td></td>
<td></td>
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<tr>
<td>PPS</td>
<td>85 285</td>
<td>65</td>
<td>3800</td>
</tr>
<tr>
<td></td>
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<td></td>
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<tr>
<td>PEK</td>
<td>170 381</td>
<td>105</td>
<td>4000</td>
</tr>
<tr>
<td>PEEK</td>
<td>143 343</td>
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<td>3900</td>
</tr>
<tr>
<td></td>
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<td></td>
<td></td>
</tr>
</tbody>
</table>

*A prepreg is a composite sheet, in which a fibre reinforcement is impregnated by a*
thermoplastic matrix (by solution impregnation or film-stacking). The fit form means that the reinforcement yarns are encaptured by a small thermoplastic tube. A hybrid yarn is a reinforcement yarn either cowoven or commingled with a thermoplastic yarn respectively with thermoplastic filaments.

<table>
<thead>
<tr>
<th>Reinforcement fibres in composites.</th>
</tr>
</thead>
<tbody>
<tr>
<td>material</td>
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<tr>
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<td>glass</td>
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</tr>
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<td></td>
</tr>
<tr>
<td></td>
</tr>
<tr>
<td>carbon</td>
</tr>
<tr>
<td></td>
</tr>
<tr>
<td></td>
</tr>
<tr>
<td>aramid</td>
</tr>
<tr>
<td></td>
</tr>
</tbody>
</table>
Appendix D: Theoretical determination of the modulus of elasticity of rubber.

In the present appendix a demonstration is given of the determination of the modulus of elasticity by looking at the deformation of a piece of rubber chain due to an external force $F$ (Fig. d.1). The distortion of the rubbery netting (elongation of the rubber piece) because of this force is schematically shown. The force needed for the elongation of such a chain is:

$$ F = \frac{NkT}{l_x^0} \cdot \left( \frac{1}{\lambda_x} - \frac{1}{\lambda_x^2} \right) $$  \hspace{1cm} \text{<D.1>}

where $N$ is the number of chain parts in the piece of the rubber chain,
$k$ is the Boltzmann constant,
$T$ is the temperature,
$\lambda_x = 1/l_x^0$.

Relation <D.1> is obtained from thermodynamic laws: the necessary work to elongate the chain equals the change in free energy of the system.

![Fig. d.1 Schematic presentation of the elongation of a rubber chain.](image)

The volume of a piece of deforming rubber is considered to be nearly constant. So, with $V = \lambda_x \lambda_y \lambda_z \cdot V_0 = V_0$, it is found that $\lambda_x \lambda_y \lambda_z = 1$ and $\lambda_y^2 = \lambda_x^2 = 1/\lambda_x$ for $\lambda_y = \lambda_z = \lambda_x$. Equation <D.1> then yields for the stress:

$$ \sigma = \frac{F}{l_y l_z} = \frac{NkT}{l_x^0 (l_y^0)^2} \cdot \left( \frac{1}{\lambda_x} - \frac{1}{\lambda_x^2} \right) $$  \hspace{1cm} \text{or}  \hspace{1cm} \sigma = N^* kT \cdot \left( \frac{1}{\lambda_x} - \frac{1}{\lambda_x^2} \right) $$  \hspace{1cm} \text{<D.2>}

where $N^*$ is the number of chain parts per unit of volume.

For small strains is $\lambda_x = 1 + \varepsilon_x$, $\lambda_x^2 = 1 + 2\varepsilon_x$, and $1/\lambda_x = 1 - \varepsilon_x$, which gives:
\[ \sigma = 3N^*kT \cdot \varepsilon_x \]

and thus:

\[ E = \frac{\sigma}{\varepsilon_x} = 3N^*kT \]

Equation \(<D.2>\) yields for large strains:

\[ \sigma = N^*kT \cdot \lambda_x^2 \quad \text{or} \quad \sigma_{\text{nominal}} = \frac{F}{t^0_1 t^0_2} = N^*kT \cdot \lambda_x \]

These expressions for the "modulus of elasticity" of rubber are not sufficient for an accurate calculation of the forces that are imposed by the rubber when pressing a thermoplastic laminate into a certain shape. In the thermodynamic derivation the assumption is made that the volume of rubber is constant during deformation, which is not true.

For a pure homogeneous deformation, the statistical theory of rubber elasticity\textsuperscript{114} predicts:

\[ \sigma = C(\lambda - \frac{1}{\lambda^2}) \]

where \( \sigma \) is the nominal stress \( F/A_0 \) and \( C \) is a constant.

The Mooney-Rivlin theory\textsuperscript{115} gives another, often used, approximation:

\[ \sigma = 2(\lambda - \frac{1}{\lambda^2})(C_1 + \frac{C_2}{\lambda}) \quad \text{<D.3>\textsuperscript{}} \]

where \( \sigma \) is the nominal stress \( F/A_0 \) and \( C_{1,2} \) are constants.

The relatively good approximation of equation \(<D.3>\) for the rubber deformation behaviour of PolyUrethane rubber is shown by experiments of the rubber pressing of metal sheets at room temperature\textsuperscript{91,116}. Because PU rubber is also used in rubber forming moulds, this relation could be a first approximation for the determination of the modulus of elasticity of rubber in rubber forming moulds\textsuperscript{117}. 
Appendix E: Design of rubber stamps.

The design of a rubber stamp for a U-shaped beam is taken as an example in this appendix.

During the forming of a U-shaped beam, it is not tolerated for the rubber stamp to touch the sides, except at the ends of the mould where there is no laminate between the rubber and the rigid mould (the ends of the mould are the front and rear side of the rubber block in Fig. e.1). Because the ends of the rubber are enclosed by the rigid sides, the stamp can be modelled as an infinite strip of rubber where the strain in the direction of its length is negligible.

![Diagram of rubber forming setup](image)

*Fig. e.1 Schematic presentation of a rubber forming setup.*

![Diagram of rubber block dimensions](image)

*Fig. e.2 The dimensions of a rubber block before and after pressurization.*
Appendix E

Suppose that the force per unit length of the stamp that is necessary to push in the laminate equals F. The rubber stamp (width w and height h) will be compressed by dh, along with a bulging deformation of dw (Fig. e.2), as long as the rubber bulging is not restricted by the mould sides.

The bulging dw is directly related to the compression dh, because of the constant volume assumption. If it is assumed that the vertical sides of a bulging rubber block take a parabolic shape:\(^{88,111}\):

\[2 \cdot \frac{2}{3} (h - dh) \cdot dw = w \cdot dh \quad \text{or} \quad dw = \frac{3}{4} \cdot \frac{dh \cdot w}{h} \quad \text{<E.1>}

where dh is assumed to be small compared to h.

The deflection dh can be calculated from:

\[F = E_c \cdot w \cdot \frac{dh}{h} \quad \text{<E.2>}

where \(E_c\) is the apparent compression modulus.

From equation <4.3.2.2>, for the apparent compression modulus \(E_c\) for long strips, it can be calculated that:

\[F = \frac{4}{3} \cdot E_0(1 + kS^2) \cdot w \cdot \frac{dh}{h} \quad \text{<E.3>}

where the shape factor S for long strips:

\[S = \frac{w}{2h} \quad \text{<E.4>}

So, the compression displacement:

\[dh = \frac{3Fh}{4E_0w(1 + k \cdot \frac{w^2}{4h^2})} \quad \text{<E.5>}

In combination with equation <E.1>, this yields for the bulging maximum:
\[ dw = \frac{9F}{16E_0(1 + k \cdot \frac{w^2}{4h^2})} \quad \text{<E.6>} \]

Equation <E.6> shows the available parameters to minimize the bulging of a rubber stamp in the forming phase.

Lowering the necessary force F is possible by decreasing the external forming forces and the friction with which a laminate slips into the rigid mould. This friction in itself depends on the pressure on the sides of the moulds.
An increase of hardness of the rubber stamp lowers the barrelling effect by an increase of the Young's modulus \(E_0\), but the effect is somewhat lowered by the slight decrease of the numerical factor \(k\) with the hardness of the rubber.

**Inverse barrel shape of the stamp.**

When the possible change of the mentioned parameters (especially hardness) is not effective enough to obtain a homogeneous side pressurization, a practical solution is the change of the design of the U-beam rubber stamp into a stamp with "an inverse barrel shape", as illustrated in Fig. e.3.

![Fig. e.3 Schematic illustration of the deformation of a pressurized rubber shape with an initial "inverse barrel shape".](image)

To determine the appropriate shape of the rubber stamp so that it provides all-over surface contact at the sides at the transition point, equation <E.6> can serve as a practical approximation. The rubber stamp has to be \(dh\) (see equation <E.5>) higher than the inner height of the rigid negative mould in order to provide enough rubber to completely fill the rigid mould. With equation <E.6> it is also possible to give an approximation of the surface pressure \(F/w\) on the bottom of the U-shaped composite product, before the laminate is pressurized against the mould sides.
If the width to height ratio \( w/h \) of the composite product that has to be rubber formed is small, it is possible that the inverse barrel shaped stamp cannot be used. Then the width \( w \) of the stamp is too small for two parabolic notches of depth \( dw \).

**Subdivision of the stamp.**

Another approach of the increase of the bulging resistance of the rubber stamp is the subdivision of the stamp in smaller rectangular blocks that are bonded to each other with a stiff (metal) sheet in the bond line. These smaller blocks each have higher shape factors than the unreinforced rubber block and cause a higher apparent compression modulus of the stamp as a whole. In Fig. e.4, it is illustrated how the subdivision influences the sideways deformation of the rubber stamp.

![Diagram of subdivision of a rubber block](image)

**Fig. e.4 Restriction of the barreling of a rubber block by subdivision.**

The shape factor of a rubber block with height \( \frac{1}{2}h \) is \( S = w/h \). The compression can be calculated by substitution of \( h \) by \( \frac{1}{2}h \) in equation <E.5>, for the same applied force \( F \) per unit length. The total compression of the new stamp becomes:

\[
dh_{\text{div}} = 2dh = 2 \cdot \frac{3Fh}{8E_0 \cdot w(1 + k \cdot \frac{w^2}{h^2})} = \frac{3Fh}{4E_0 \cdot w(1 + k \cdot \frac{w^2}{h^2})} <E.7>
\]

which is smaller than \( dh \) for the undivided stamp (see equation <E.5>), so the subdivided stamp has a higher apparent stiffness during the forming phase. The maximum sideways deflection of the subdivided stamp can also be calculated by substitution of \( h \) by \( \frac{1}{2}h \) in equation <E.6>:
\[ dw_{\text{div.}} = \frac{9F}{16E_0(1+k\cdot\frac{w^2}{h^2})} \]  

<E.8>

A subdivision of a rubber stamp in more layers increases the bulging resistance even more, so in this way even a very shallow rubber stamp can be limited in its bulging tendency. The use of rubber reinforcements smaller than the width of the mould, provides a more smooth transition between the (now partly) separated rubber parts. In this way, the divided rubber stamp gives a more uniform pressure distribution at the sides.

The basic examples of mould design that are given here can be used alone or in combination, depending on the complexity of the composite product that has to be rubber formed. With the presented equations and the right boundary conditions, it must be possible to calculate the deformations behaviour of a composition of all kinds of small rubber blocks parallel or in series integrated in a rubber stamp. Further calculation is considered to be outside the scope of the present, surveying work. Some interesting examples of the design of non-matching rubber moulds are presented in Section 3.3.3 and Section 3.3.4.