An introduction to the rubber forming of thermoplastic composites

February 1992

L.M.J. Robroek

TU Delft
Faculty of Aerospace Engineering
Delft University of Technology
An introduction to the rubber forming of thermoplastic composites

L.M.J. Robroek
Abstract.

A rather new thermoforming technique known as rubber forming has been developed to process continuous fibre reinforced thermoplastics (CFRTP). In this introductory report a global summary of the rubber forming technique is presented.

To shape thermoplastic composite products, several subprocesses must take place during processing. Some of the thermoforming techniques with which this is currently done are pointed out. The main subprocesses of heating, pressurization and forming in the rubber forming method are dealt with separately. Also the shaping of a flat laminate during a rubber forming cycle by the necessary forming forces is discussed. The influence of the process parameters temperature and pressure on the deformation of a thermoplastic laminate is shown to be important.

Investigations indicate the large influence of several subprocesses in a rubber forming cycle on the mechanical properties of a CFRTP product. Heat transfer and pressurization are two important research items in the development of the rubber forming technique. They determine the composite product quality and have to be controlled to a high extent.

Further research and development in all of the mentioned process areas is necessary for better understanding of the rubber forming technique.
# Table of contents

<table>
<thead>
<tr>
<th>Section</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Abstract.</strong></td>
<td>5.</td>
</tr>
<tr>
<td><strong>Introduction.</strong></td>
<td>7.</td>
</tr>
<tr>
<td><strong>CHAPTER 1 The use of continuous fibre reinforced thermoplastics.</strong></td>
<td>9.</td>
</tr>
<tr>
<td>1.1 Potentials of thermoplastic composites.</td>
<td>13.</td>
</tr>
<tr>
<td>1.2 Basic thermoplastic prepreg forms.</td>
<td>17.</td>
</tr>
<tr>
<td>1.3 Processing of thermoplastic composites (terminology).</td>
<td>21.</td>
</tr>
<tr>
<td><strong>CHAPTER 2 Rubber forming as a thermoforming technique.</strong></td>
<td>27.</td>
</tr>
<tr>
<td>2.1 An overview of CFRTP processing techniques.</td>
<td>29.</td>
</tr>
<tr>
<td>2.2 The rubber forming technique.</td>
<td>31.</td>
</tr>
<tr>
<td>2.3 Subprocesses in rubber forming.</td>
<td>34.</td>
</tr>
<tr>
<td><strong>CHAPTER 3 The shaping of a thermoplastic composite product.</strong></td>
<td>37.</td>
</tr>
<tr>
<td>3.1 Deformation modes of fabric reinforced thermoplastics.</td>
<td>38.</td>
</tr>
<tr>
<td>3.2 Forming forces during rubber forming.</td>
<td>44.</td>
</tr>
<tr>
<td>3.3 Influence of process parameters on the forming cycle.</td>
<td>51.</td>
</tr>
<tr>
<td>3.4 Special fibre reinforcements.</td>
<td>52.</td>
</tr>
<tr>
<td><strong>CHAPTER 4 Temperature and pressure distributions during a rubber forming cycle.</strong></td>
<td>57.</td>
</tr>
<tr>
<td>4.1 Heat transfer in rubber forming.</td>
<td>59.</td>
</tr>
<tr>
<td>4.2 Pressurization during rubber forming.</td>
<td></td>
</tr>
<tr>
<td><strong>CHAPTER 5 The viability of rubber forming.</strong></td>
<td></td>
</tr>
<tr>
<td>5.1 Quality control by process control.</td>
<td></td>
</tr>
<tr>
<td>5.2 Experiments to determine the influence of rubber forming cycles.</td>
<td></td>
</tr>
<tr>
<td>5.3 Future investigations.</td>
<td></td>
</tr>
<tr>
<td><strong>References.</strong></td>
<td></td>
</tr>
</tbody>
</table>
Introduction.

As the composites market develops further, the number of realized applications of continuous fibre reinforced thermoplastics (CFRTP) is increasing. The advantages of the use of thermoplastic matrices in composites are widely known. One of the major advantages of CFRTP over competing thermosets is the potential ease and quickness of manufacturing of products.

To exploit the benefits of thermoplastic composites, these materials have to be transformed into high quality composite parts by a cost effective thermoforming process. In contrast to thermosets, however, the processing of thermoplastic composites is in its infancy. It is therefore not surprising that the number of investigations in CFRTP processing technologies is still expanding. The efforts to develop appropriate fabrication technology fall into two categories. In the first category, studies are made to adapt thermoset technology, such as diaphragm forming or filament winding. The second category deals with research on newly developed thermoforming technology to produce thermoplastic composite parts.

One of the newly developed fabrication technologies is rubber forming of continuous fibre reinforced thermoplastic composites. The development of this manufacturing process was started at the University of Technology in Delft several years ago now. The rubber forming technique is a modification of other rapid stamping processes that in fact are derived from forming processes in the metal processing industry. Most CFRTP-prepreg manufacturers supply the composite material in sheets, thereby providing the same starting point in the processing cycle as metal sheet suppliers do. When heated, thermoplastic composite sheets can be transformed into the desired shape by a rapid stamping process. One of the major differences with other formable materials however, is the continuity of the fibre reinforcement. The fact that CFRTP are materials composed of two different components that have to be fully impregnated and consolidated is a feature that requires care during thermoforming processes.

During the development of thermoforming techniques, the number of technical reports about processing of continuous fibre reinforced thermoplastics has risen almost exponentially. Often these reports cover one particular item to a high extent or they give a very global general overview. There is, however, still the need for an introductory report about the rubber forming process of thermoplastic composites. This need exists
not only for new students in the field of CFRTP materials, but also for more experienced composite workers who want to get a clear view of the rubber forming process.

Several topics that are dealt with in this report are often misunderstood or not considered at all. Sometimes, the solutions will be more difficult to find, but too often problems arise from a lack of basic understanding of the thermoforming technique that is used to create the projected composite products. In this report therefore only the fundamentals of the rubber forming technique will be discussed.

In the first chapter, an overview is given of the use of thermoplastic continuous-fibre reinforced composites. Some important basic subjects of processing of thermoplastic composites will be introduced and the diversification in available prepreg forms will be pinpointed.

The following chapter will describe some of the existing thermoforming techniques between which the rubber forming technique has claimed an important position. Each of these techniques is suitable for a specific manufacturing purpose. After a technical introduction, some fundamental subprocesses in the rubber forming cycle will be pointed out.

Chapter three describes the process of deformation which takes place during a thermoforming cycle. Important process variables that influence the deformation will be mentioned. Two basic experiments leading to a better understanding of the deformation variables of the deformation process will be described.

The next chapter deals with the two main process variables of thermoforming techniques, namely temperature and pressure. These parameters are regarded as the most essential elements in a rubber forming cycle. Several experimental results will prove the importance of heat transfer and pressurization.

The last chapter (chapter five) covers process and quality control. It is closely related to all of the items of the preceding chapters. The important issue of quality control of thermoplastic composite products will be looked at from the control of process parameters. Finally, some future investigations will be mentioned.
CHAPTER 1 The use of continuous fibre reinforced thermoplastics.

Thermoplastic matrix composites are of much interest for aerospace applications as well as for automotive structures. Generally, the components fabricated in these industries are of complex shapes. In aerospace structures, single or double curvature panels such as wing and fuselage skins, rudder and other control surfaces have to be manufactured. In the lightweight inner structure of an aircraft some reinforcing parts can be pinpointed: spars, stiffeners, ribs, but also floor beams that have various cross-sectional shapes (figure 1a). So far as the automotive industry is concerned, a much smaller variation in parts is necessary, but the shapes of these panels often are equivalent to those used in aerospace (figure 1b). Doubly curved panels cover an automobile and beams and stiffeners support them.

1.1 Potentials of thermoplastic composites.

Compared to thermosets, thermoplastic matrix composites are tougher, more impact resistant and can offer better resistance to chemicals and solvents\(^1\). They have unlimited shelf live and are more friendly to handle. These advantages of continuous fibre reinforced thermoplastics are often referred to. However, the larger part of the interest

![Figure 1a. Thermoformed CFRTP parts for aerospace applications.](image-url)
in CFRTP stems from their promise of processing ease. The capability of shaping and reshaping the material whenever it is heated makes it an attractive structural material for the manufacture of aerospace and automotive parts. It opens the possibility that manufacturing costs for relatively complicated thermoplastic components can be lower than for thermosetting or metallic parts.

Because composites have the possibility that their mechanical properties can be locally adapted to the need of the structure, designers of CFRTP products have an additional design liberty. This feature, however, demands extra skill. Another important point of attention is that, in contrast to thermosets where manufacturing and processing techniques have been known for many years, thermoplastic composites have to be processed by rather new thermoforming techniques. These techniques can give severe restrictions in part design but, on the other hand, often can create possibilities for complex shapes that hardly could be processed before. So the thermoplastic part designer must also be aware of the (still developing) thermoforming techniques that have to be used to realize the composite component.
1.2 Basic thermoplastic composite prepreg forms.

Many combinations of fibre reinforcement and thermoplastic polymers are commercially available. A few of the possible combinations will be pinpointed here and some basic CFRTP prepreg forms will be given.

Two main fibre types, carbon and glass fibres, are used in thermoplastic composite prepregs, though some aramide fibre reinforced composites are available also. Both carbon fibres and glass fibres can be subdivided in different grades. These grades of fibres each have different mechanical and/or chemical properties (see table 1) to serve in specific environments or fulfill specific mechanical demands.

<table>
<thead>
<tr>
<th></th>
<th>Specific gravity</th>
<th>Tensile strength MPa</th>
<th>Tensile modulus GPa</th>
</tr>
</thead>
<tbody>
<tr>
<td>E-glass</td>
<td>2.58</td>
<td>3450</td>
<td>73</td>
</tr>
<tr>
<td>A-glass</td>
<td>2.50</td>
<td>3040</td>
<td>69</td>
</tr>
<tr>
<td>ECR-glass</td>
<td>2.62</td>
<td>3625</td>
<td>73</td>
</tr>
<tr>
<td>S-glass</td>
<td>2.48</td>
<td>4590</td>
<td>86</td>
</tr>
<tr>
<td>T40-carbon (Toray)</td>
<td>1.74</td>
<td>4500</td>
<td>296</td>
</tr>
<tr>
<td>HMS4-carbon (Hercules)</td>
<td>1.78</td>
<td>3100</td>
<td>338</td>
</tr>
<tr>
<td>P100-carbon (Union Carbide)</td>
<td>2.15</td>
<td>2240</td>
<td>724</td>
</tr>
</tbody>
</table>

*Table 1. Mechanical properties of some fibres that are used in composite materials*.

To come to a composite material, the fibres have to be mingled with the thermoplastic polymer. In general this is done by the material supplier. So there is no need to apply any chemistry in composite part production by the end manufacturer.

To achieve the maximum properties of a composite material, it is necessary to obtain a good impregnation of the matrix in the fibre reinforcement. In general this is difficult with thermoplastic matrices because these resins have relatively high melt viscosities. Several impregnation techniques can be used by the prepreg supplier to obtain a good fibre-matrix interface. These techniques will determine or affect the handling as well as the processing and final properties of the CFRTP material.

The methods of impregnation can be divided into those that give a real pre-impregnated material and those where a final impregnation takes place during thermoforming of the material in the product mould (post-shaping impregnation)*.
Pre-impregnated materials are prepgs in which the reinforcing fibres are completely wetted and impregnated by the matrix (see 1.3). In post-shaping impregnation strategies only a physical mixing of the matrix and fibre is achieved by the prepreg manufacturer and thus a full wetting of the reinforcement does not occur.

A frequently used method of pre-impregnation is solution impregnation (figure 2). In this method the viscosity of the thermoplastic matrix is lowered by solvation in a suitable solvent. The acquired solution is used to thoroughly penetrate and wet the fibre reinforcement. Afterwards, the removal of the solvent is necessary. It can be difficult to remove the solvent totally, however, and often the part manufacturer has to carry out an additional drying (removal) cycle before thermoforming the material to avoid severe contamination of the composite by the solvent at high processing temperatures. Another disadvantage of the solvent-aided impregnation method is that polymers with good resistance to chemicals (e.g. most of the semi-crystalline polymers) cannot easily be dissolved. This method of impregnation therefore is the most common only for amorphous thermoplastics.

![Diagram of solution impregnation process](image)

**Figure 2. Schematic view of the solution impregnation process.**

Another method of reducing the viscosity of the matrix is heating it. However, melting the thermoplastic resin and wetting the fibre reinforcement with the melt is difficult. The melt viscosity of thermoplastics is often too high to realize good impregnation. In the *melt impregnation* technique fibre bundles can be pulled through a molten polymer or molten sheets of thermoplastic resin can be forced into the fibre bundles by pressure. This method, called film-stacking (figure 3), may be used to produce flat impregnated sheets that can be thermoformed in a subsequent stage. Sometimes, only a partial
impregnation is achieved only to add the polymer to the fibres before full impregnation is realized during the final thermoforming process. Therefore it sometimes is classified as a post-shaping impregnation method.

![Diagram of film-stacking process](image)

**Figure 3. Schematic view of the film-stacking process.**

Another technique of physically mixing fibres and matrix without (total) impregnation is *powder coating* (figure 4). Particles of the thermoplastic polymer are distributed over the fibre reinforcement. The fibre bundles have to be spread as much as possible to assure uniformly distributed particles. After the powder distribution, the polymer has to be prevented from segregation from the fibres. This can be realized by a tackifying agent on the fibres\(^3\), eventually followed by melting of the powder onto the fibres. Another approach is the addition of a small "tube" over the powder impregnated fibre bundle (figure 5).

![Diagram of powder coating process](image)

**Figure 4. Schematic view of the powder coating process.**
Cowearing and commingling as mixing techniques use the fact that some of the polymers can be spun and these polymer fibres can be mixed with reinforcing fibres on a fibre bundle level (coweaving) or on a fibre level (commingling), see figure 6a and 6b. The
real impregnation of the material finally takes place during the thermoforming process. In principle, it should be easier to achieve full impregnation using commingled material, rather than using cowoven material, because the mixing of fibre and matrix is more uniform and thorough.

1.3 Processing of thermoplastic composites (terminology).

In the terminology used in this report, the following subsequent processes (subprocesses) in thermoforming of CFRTP parts are distinguished:

- **Physical mixing** of fibre reinforcement and thermoplastic matrix is the combining of the two components only to a macroscopic level (wetting of fibre bundles, stacking, coweaving, commingling). One of the main features in physical mixing is that voids can still be present inside the reinforcing fibre bundles.

- **Impregnation** of the fibre reinforcement is the expulsion of the air or other "strange" particles out of the fibre bundles and the complete wetting of fibres inside those fibre bundles.
Consolidation is the subprocess where a voidless and homogenously fibre-reinforced material is realized by the complete fusion of fully impregnated layers (figure 7).

Forming of the composite laminate is the shaping of the material characterized by the deformation of the fibre reinforcement by intraply shear, interply slip or other motions of the fibre reinforcement. (During forming it is possible that already consolidated layers will debond due to the motion of the reinforcing fibre bundles, which is called deconsolidation. The final consolidation that is needed to undo deconsolidation is often called reconsolidation.)

A fully shaped and consolidated thermoplastic composite product has to cool down to room temperature. During this cooling down, the material will solidify.

Processing.

The quality of the final composite product can be influenced by the prepreg form that will be used for manufacturing of CFRTP parts. Not all of the mentioned impregnation or mixing techniques will be suitable for every thermoforming technique and/or material combination. In general it can be stated that the better the pre-impregnation of the composite, the less difficulties in the thermoforming process of a product.

Figure 7. Schematic illustration of the consolidation subprocess.
As mentioned before, the manufacturers of thermoplastic composite products usually will be supplied with prepregs in a sheet form when moulding techniques will be used to produce composite parts. The manufacturer can arrange the prepreg plies in a stacking sequence that is most suitable for the product to be made. The composite plies will be heated and formed by a forming device and after cooling down, the product can be depressurized and taken out of the mould.

In the ideal case, the final part should be free of voids with the plies well bonded to each other. This has to occur during the consolidation subprocess (figure 7). For good consolidation a high normal pressure on the plies is necessary at a temperature that is high enough to minimize the viscosity of the thermoplastic. Composite prepreg sheets have to be pressed well onto each other to assure intimate ply contact and interlaminar adhesion (autohesion). It is important that the consolidation subprocess takes place after deformation of the prepregs into the final shape of the product, because of the distortion of ply contact when the plies are in motion during thermoforming (deconsolidation).

It is difficult to determine the time, pressure and temperature, necessary for complete consolidation. From a manufacturing point of view it is favourable to avoid long consolidation time and high pressure or temperature. This functions not only to prevent the need for heavy duty forming and heating equipment, but also to keep the energy costs and cycle times low. It is therefore important to estimate optimal consolidation process parameters to realize high mechanical properties of the final product. In chapter five an attempt is made to establish the effects of consolidation time, temperature and pressure on mechanical quality for a specific thermoplastic composite. It will be clear that the degree of pre-impregnation of the prepreg used is important for the processing parameters that will be needed. When prepregs are used in which the matrix and the fibres are not more than physically mixed, the first part of the temperature-pressure-time cycle has to be used to fully impregnate the material.

After consolidation, the composite product has to cool down under pressure and the thermoplastic matrix will solidify. The solidification of (semi-)crystalline material will start when the temperature drops below the melting point \( T_m \) of the matrix (figure 8). Because of the influence of the crystallization degree on chemical and mechanical properties of (semi-)crystalline composites, it is important to control the cooling rate of these materials\(^4\), because the cooling rate determines the crystallization degree. With amorphous matrices, the solidification value is totally determined by the glass transition temperature \( T_g \).
Because of the absence of crystallization in amorphous matrix, temperature control during cooling will be less important with amorphous matrix composites than with semi-crystalline matrix composites. However, because of the different specific volume versus temperature of matrix and fibre material in all composites, residual stresses can develop during solidification\textsuperscript{5}, which can partially be relieved close to $T_g$. A good control of the thermal history all through the thermoforming cycle is therefore still needed. Finally, when the temperature of the newly formed composite part is far below its glass transition temperature, the pressure on the product can be relieved and it can be taken out of the mould.
CHAPTER 2 Rubber forming as a thermoforming technique.

To manufacture a two or three dimensional thermoplastic composite part out of a flat prepreg, it is necessary to choose an appropriate thermoforming technique. The thermoforming method that will be used to transform a flat laminate into the desired product shape will be determined by several technical and economical factors. Apart from the kind of prepreg, the choice can depend on the level of capital investment desired, estimated product series, cycle times and other economical characteristics. A discussion of the cost of thermoforming techniques is outside the scope of this report. In this chapter, the technical position of the rubber forming method among other thermoforming techniques is outlined. For this purpose, five other important thermoforming techniques are mentioned first. In the second section a technical description of the rubber forming method is given. Finally in this chapter, the most important subprocesses during rubber forming are pointed out.

2.1 An overview of CFRTP processing techniques.

The major processing methods to thermoform continuous fibre reinforced thermoplastics that are currently being used, are:

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

![Figure 9. Schematic view of diaphragm forming.](image)

17
In diaphragm forming, the laminate is held between two disposable plastically deformable diaphragms. The diaphragms are clamped onto the mould and a vacuum is applied between these foils. The whole setup is heated to the processing temperature. At this temperature, increased air pressure (in an autoclave) is used to force the diaphragms with the laminate to slide into the mould. With vacuum forming, this forming pressure is created only by a vacuum in the mould itself. Vacuum forming therefore can only deform with low forming forces on the diaphragms. During the forming process, the laminate can slide within the diaphragms. This sliding action creates tensile stresses in the sheet that reduce wrinkling and induce the deformation of the fibre reinforcement. Due to the slow deformation rate, the control of the deformation usually is good. Cycle times, however, are relatively high compared to other thermoforming techniques due to the extensive heating and cooling cycles. Another disadvantage is the deformation limitation of the diaphragms that are used and the diaphragm determined temperature range when deformation has to take place. Recent studies⁶ however, give good prospects for reducing these limitations.

Hydroforming⁷ is similar to diaphragm forming in that a fluid medium, usually a hydraulic fluid, behind a rubber diaphragm, is used to deform the laminate over or in a mould. The main differences are that the rubber diaphragms in hydroforming are a permanent part of the forming equipment, are usually much larger than the workpiece and only one is used. Extra disposable rubber sheets have to be placed over the hot material to prevent excessive wear or diaphragm rupture due to high temperature and sharp edges on the mould. This thermoforming technique stems directly from the metal sheet forming industry and usually implies the use of massive systems.

*Figure 10. Schematic view of hydroforming.*

18
Figure 11. Schematic view of rubber pressing.

ad c, figure 11).

Rubber pressing of thermoplastic composites is another forming technique that is 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 rubber pad is permanently attached to the press platen and is generally much larger than the tool. The process provides high forming forces but they are not uniformly distributed over the tool\(^8\). The local forming forces are determined by the shape of the die that counteracts the closing of the press. The complexity of parts is limited by the maximum local deformation of the rubber pad.

ad d, figure 12).

In pultrusion thermoplastic composite tapes are (semi-)continuously formed into a stringer or rod. In this technique it is possible to laminate, form and consolidate the material in one manufacturing street. Coming from rolls, the prepreg plies first have

Figure 12. Schematic illustration of thermoplastic pultrusion.
to be uniformly heated. This can be done by hot air, radiation or contact heat. When a sufficiently high temperature is reached, the plies must be compacted to a laminate. This hot laminate is formed directly or in various steps into the desired form by pultruding the material through one or a series of sequential dies. The necessary forming and consolidation pressure has to be induced by these dies while the material is still in motion. Mostly the cooling down of the pultrusion product takes place in the last die. The use of the pultrusion technique is limited to the forming of rod material with relatively simple cross-sections. Temperature and pressure control is often very difficult.

ad e, figure 13).

Matched-die forming is the most widely used thermoforming system. This is because forming presses are historically used in a broad range of other processes and are therefore available in a vast range of different sizes and designs. The use of matched metal dies in a forming technique ensures a proper surface quality and high detail on both sides of the thermoplastic composite part. The male and female dies usually can be internally heated which can increase the quality of the final product when a suitable temperature is used. High (local) pressures can be applied on the laminate during forming. A disadvantage of this thermoforming technique is that when there is even a small thickness mismatch between the formed part and the mould cavity, the pressure distribution on the material will be highly non-uniform during the final step of the moulding process and especially during the consolidation of the material. This will result in a non-uniform consolidation and/or varying fibre-volume fractions along the product. Furthermore, it is difficult to control the deformation of the fibre reinforcement, because the non-flexible forming dies introduce only highly localized forming forces on the surface of the deforming laminate.

![Figure 13. Schematic view of matched metal die forming.](image-url)
2.2 The rubber forming technique.

The rubber forming technique has been developed to overcome some of the main disadvantages of the matched metal-die forming method. The rubber forming process, however, has become an independent thermoforming technique. In this technique, one of the rigid metal dies in the matched metal-die technique has been replaced with a flexible rubber die (figure 14). The remaining rigid (metal) die determines the final shape of the thermoplastic composite product during forming and gives a good surface quality on that side of the product.

![Diagram of rubber forming](image)

*Figure 14. Schematic view of rubber forming.*

Using a matched rubber die in a rubber forming process results in a more homogeneous pressure distribution on the composite material. The flexibility of the die will account for the thickness mismatch or thickness variation that can be present during thermoforming of CFRTP laminates. However, the rubber die does not have to be totally matched with the rigid die-half. To obtain an optimal deformation of the fibre reinforcement in a thermoplastic laminate, it is often favourable to under- or overdimension the rubber die-half. The intentional mismatch avoids or creates certain local sheet pressures to facilitate the desired material deformation.

One of the important properties of the flexible rubber is that it will adapt during the forming process to completely fill the rigid die, as is the case with the rubber pressure medium in hydroforming and rubber pressing. Because of the limitation of the maximum local rubber deformation though, the rubber die usually must have a certain preshape that profiles the rigid die (figure 15). Rubber forming, therefore, also fills the gap between rubber pressing (using an unshaped rubber pad) and matched-die forming.
Another difference with the rubberpressing technique is that in rubber forming the rubber die half mostly is not larger than the rigid mould.

2.3 Subprocesses in rubber forming.

As in most of the mentioned thermoforming techniques, a rubber forming cycle consists of several stages (figure 16). Depending of the kind of prepreg that is used, the laminate that will be deformed often has to be pre-consolidated. Then the thermoplastic composite sheet has to be heated until the temperature of the material reaches the processing temperature. Finally, the laminate has to be quickly transferred to the forming device and the forming of the product can take place.
The pre-consolidation of a laminate, composed of several layers of prepreg, can be carried out in a hot platen press or an autoclave. At a temperature that is high enough to ensure a relatively low matrix viscosity, the layers are pressed together for a certain time to finally become one homogeneous flat sheet. For badly impregnated prepreg forms, this step is often necessary to ensure a good impregnation of the material. Rubber forming, in particular, is a rapid stamping process in which the thermoforming cycle only takes 1-3 minutes per part. Therefore the time that is available for the final consolidation will be very short (see chapter 4) and it is favourable to carry out a part of the consolidation or impregnation outside the real rubber forming cycle. Sometimes the prepreg material is supplied in the form of already (partially) consolidated sheets. It is clear that this has the cost-saving advantage of deleting the pre-consolidation stage out of the thermoforming cycle. A disadvantage is that the pre-consolidated sheets have to be supplied in the right configuration for the final products.

The preceding stage of the real rubber forming step is the heating of the thermoplastic composite laminate to bring it to a temperature that is high enough to reduce the matrix viscosity in a way that deformation of the fibre reinforcement will be allowed in the subsequent forming stage.

The laminate can be heated by contact heat (conduction heating) between two heated plates. When this is done at a high enough pressure, the above mentioned separate pre-consolidation step sometimes becomes unnecessary. A disadvantage of the conduction heating method is the fact that the laminate actually contacts the heating equipment. Good release agents must be used therefore to prevent the sticking of the material to the plates.

In dielectric and induction heating methods, a strong alternating magnetic field and electrical field is created in the composite material. These fields induce an electrical
current and agitation of the molecules in the material and only work with specific fibre/matrix combinations, because of the physical demands on the composites (conductivity and presence of dipoles).

*Convection heating* in an oven is also possible, but will usually take the longest to heat a laminate. The use of inert gas is preferable to prevent oxidation of the polymer at high temperatures.

Inert gas environments are also recommended when using *infrared heating methods*, though heating times usually are short. A disadvantage is that for thick sheets temperature gradients develop through the thickness and sometimes will restrain formability in the thermoforming technique. Radiation heating, however, can be a clean and quick heating method in general. It provides a flexible and a well-manageable heating device.

When the temperature of the thermoplastic composite laminate reaches the processing temperature, the material has to be quickly transferred to the forming device, or when the laminate is heated in the forming equipment, the heating device must be removed. Cooling of the composite sheet during this transfer has to be avoided as much as possible.

*Figure 17. Close-up of the (empty) blankholder on the metal die with clamping pins.*
As already mentioned, the forming device in rubber forming consists of one rigid (metal) die and a flexible rubber die. In general, the rubber die is made of polyurethane or a heat resistant castable silicone rubber. Apart from a possible intentional mismatch between the rubber and metal die, a "blankholder" can be used during rubber forming to control the laminate deformation. This blankholder serves to introduce local forming forces by clamping of the sheet on specific points (figure 17). In this way, certain deformations of the fibre reinforcement are imposed to facilitate the shaping of the composite product as a whole. The function of the blankholder can be integrated in the moulds or in a clamping ring in which the laminate is placed before the heating and forming cycles. The use of a clamping ring as blankholder improves the handling of the composite sheet, especially when transferring it from the heating to the forming device. A disadvantage is the required additional heating time because the clamping ring increases the heat capacity of the laminate system.

When the final shape of the composite part is realized and final consolidation is assured, the product must be cooled down under pressure (see chapter 1). When the temperature of the moulds is low enough, the part can be taken out and trimmed if necessary.

Rubber forming is a rapid stamping process; total production cycle times of 3-10 minutes for one product are realistic, largely depending on the heating method and material thickness. The actual forming subprocess can take place in less than five seconds. The quality of the final product largely depends on the control of (local) temperature and (local) pressure on the composite sheet during rubber forming, including the final consolidation subprocess. Therefore, in the next chapters the influence and control of temperature and pressure during a rubber forming cycle will be discussed more intensively.
CHAPTER 3 The shaping of a thermoplastic composite product.

When a reinforced thermoplastic laminate is forced in a specific form by a thermoforming technique, the fibre reinforcement has to adjust to that same form. The continuity of the fibres plays an essential role. In contrast to metals or unreinforced thermoplastics, continuous fibre reinforced thermoplastics cannot undergo a deformation with only local adjustment of the material (figure 18). Bending or curving on one spot of the hot material may influence the whole laminate. There are several ways, however, in which the adjustments of the fibre reinforcement can take place.

![Image](image_url)

Figure 18. Stretching of fibres along the edge of a lightening hole due to early clamping of the laminate.

3.1 Deformation modes of fabric reinforced thermoplastics.

Although composite products generally are formed of laminates of more than one layer, the deformation capacity of one individual layer plays a dominant role in the forming process (intraply deformation).
Some of the main intraply deformation modes will be given here:

- The Trellis effect of the fabric or intraply shearing. During thermoforming, tensile forces can be generated in a reinforcing fabric in directions other than the principal (main) directions of the fabric itself. They result in a rotation of the fibre bundles relative to each other (figure 19). The crossover angle of the fibre bundles will decrease during the forming process\textsuperscript{11}.

![Diagram of Trellis effect](image)

*Figure 19. Intraply shear (Trellis effect).*

- Fibre straightening and fibre elongation. The only two "local deformations" of a reinforcement that can be generated by tensile forces in the laminate are straightening and elongation of fibres. However, fibre straightening is the result of stretching a badly aligned laminate and should be considered as a corrective process. Fibre elongation mainly occurs when the fibre bundles are loaded too high and other deformations are obstructed. It will often lead to breakage or residual internal stresses in the final composite product.

Normally, a "laminate" consists of more than one layer. Such a laminate can schematically be presented as a stack of fibre rich layers alternated by thermoplastic resin rich layers. In a thermoforming process, the resin rich layers are soft and will have a certain viscosity. They will allow the relatively inextensible fibre rich layers to slip relative to each other when the laminate is bent by forming forces. This deformation mode is called interply slipping (figure 20). During this slipping the resin rich layer must act as a lubricant, otherwise the generated inplane compression stresses at the concave side of the product and tension stresses at the other side will rise too high during bending and can cause failure of the composite bend\textsuperscript{12}.

28
3.2 Forming forces during rubberforming.

To realize the deformation of a flat composite sheet and thus the deformation of the fibre reinforcement, forming forces have to act on the laminate. These forces have to be introduced by the moulds and/or the blankholder during the actual rubberforming (see chapter 2). Not only the complexity of the desired product shape, but also the initial fibre directions in the laminate are important in the deformation process. The continuous fibre will only transfer the applied deforming forces in their own direction (interply slipping or straightening), but by using a fabric reinforcement for instance it is possible to move fibres sideways or rotate them (intraply shear), because of the greater dimensional stability of a fabric.

Looking at figure 21a, areas in the originally flat laminate can be indicated where the reinforcing fabric has to shear. The necessary rotation of the fibre bundles has to be imposed by forming forces which are introduced in the fabric by a combination of pins in the blankholder and the downwards movement of the upper die half. The application of pins (figure 21b) restricts the sliding of the fabric at that point into the lower die half.

Figure 20. Interply slip.

Figure 21a. A product draped with a schematic fabric ("Drape" simulation by O.K. Bergsma).
This supplies the "clamped" fibre bundles with the necessary forces to rotate the fabric in the depicted area.

During intrapy shearing, the deformation can be obstructed by two kinds of friction. The first one is the regular friction of the rotating fibre bundles moving relatively to each other through a highly viscous medium. The other kind of friction occurs at the crossover points of the interlacing bundles where the rotation of the fibres is retarded.

In figure 21c a cross-section of the same product is given. It shows the difference in bending radii of the individual fabric layers after forming. When a laminate is bent during a thermoforming process, stresses are created in each layer by the forming forces until a certain "shear yield stress" is exceeded in the viscous resin rich layers between the fibre rich layers and interply slip is started. The (fibre rich) layers will move relatively to each other until the different bending radii of the individual fabric layers are realized. The necessary amount of slipping is a function of the bending angle. During this slipping a friction force must be overcome.

---

*Figure 21b. Application of clamping pins.*

*Figure 21c. Schematic cross-section of the formed product.*
3.3 Influence of process parameters on the forming cycle.

In thermoforming, the viscous matrix acts as a lubricant and decreases the internal friction during the motion of fibre bundles in a laminate. The forming process will be facilitated due to the presence of hot thermoplastic resin and the necessary forming forces on the laminate will be reduced.

![Graph showing the effect of matrix presence on shearing deformation](image)

*Figure 22. Influence of the presence of thermoplastic matrix on the maximum shearing deformation of a fabric.*

The influence of the presence of matrix in a glass fabric is illustrated by figure 22. In this figure is depicted that the maximum shearing angle increases when a fabric is impregnated with hot thermoplastic resin (here Polyetherimide at 300°C). The shearing angle is the angle with which the crossover angle between two fibre bundles reduces at a certain forming force\(^\text{14}\). Because of the role that the viscosity of the matrix plays, it is clear that a change in processing temperature, which changes matrix viscosity, has an important influence on the lubrication function. In figure 23 this is shown for a glass-fabric reinforced polyetherimide composite that with an increase of processing temperature a higher intraply shear deformation is possible. The applicable upper limit of processing temperature however is given by the degradation temperature of the matrix.
Figure 23. Influence of process temperature on the maximum shearing deformation of an impregnated fabric.

Figure 23 also gives the important influence of the normal pressure that is applied on the deforming laminate. An increase of normal pressure will increase the internal rotation friction as well as the sliding friction in the laminate. The normal pressure during a rubber forming cycle can be controlled by the shape of the rubber die-half. It is important to avoid or delay the presence of pressure on a specific part of the laminate that has not yet been deformed. This sometimes can be achieved by under-dimensioning of part of the rubber die. It is, of course, a highly product-dependent solution.

Experiments have shown that an increase of temperature also facilitates the interply slip deformation\textsuperscript{15}. When an individual layer is pulled out of a heated and pressurized laminate in one of its fibre directions, it is clear what important influence a lower matrix viscosity has on the deformation properties of that laminate (figure 24). Both the slip velocity and the possible final amount of slip deformation is facilitated with increasing temperature. In reference 14 is shown that the slip phenomena partially can be modelled out of the rheological behaviour of a polymer by a "power-law with yield value" as is depicted in figure 25. With the theoretical prediction the outcome of an interply slip experiment can be simulated by numerical integration. In order to use this model for designing a thermoforming process however, some of the factors in the equations have to be experimentally determined again for different circumstances such
as processing temperature. When the normal pressure on the deforming laminate increases, the interply slipping will proceed slightly slower. The slip velocity, however, will decrease more when the thickness of the resin rich interlayer is decreased due to a
higher pressure. In figure 26 is shown that the shear stress imposed on the viscous matrix layer to obtain a specific slip velocity does not have a clear relation with the normal force. The explanation for this observation probably has to come from rheology studies.

![Graph showing shear stress vs. slip velocity](image)

**Figure 26.** Influence of normal pressure on the interply slip deformation in a laminate.

The above considerations about the influence of the process parameters pressure and temperature underline the importance of investigations to determine and control these parameters. In this way it might be possible to find the optimum combinations of time, pressure and temperature in a rubber forming process of thermoplastic composite parts.

3.4 Special fibre reinforcements.

It is not always possible to realize a specific complex product shape in a rubber forming cycle. Sometimes however, the use of special kinds of composite material can be recommended. The use of Long Discontinuous Fibres (LDF)\(^{16}\) for instance, can be an alternative way of manufacturing a particular composite part if the use of LDF material is mechanically allowable in the application. Discontinuous fibres have the advantage that deformations of the composite material can be obtained very locally, depending on the length of the reinforcing fibres. The use of LDF can be a disadvantage in particular cases because the discontinuous fibres provide less stability in rubber forming or other
stamping processes. Besides that, the mechanical properties of discontinuous fibre reinforced thermoplastics are somewhat lower.

Another alternative for producing complex parts or products with very deep sections is the use of a cowoven or commingled fabric (see chapter 1). Such a "prepreg" material where the thermoplastic matrix is present as fibres in the reinforcing fabric, still possesses the drapeability of a real fabric. Therefore, the hybrid fabric can be placed or draped over or in one of the die-halves before heating (figure 27). The draped cowoven or commingled "laminate" can be heated in the mould and finally will be pressurized by the forming equipment. The actual forming cycle of a thermoforming technique is then reduced to a very small final deformation of the material.

It is also possible to process cowoven or commingled fabric the same as other prepregs, namely consolidation of a flat laminate and an additional thermoforming process. The use of these kinds of composite materials is especially justified in the manufacturing of product shapes that normally would be quite difficult or impossible to realize out of flat fabric reinforced (semi-)consolidated laminates. Another advantage is the possibility of the material to be knitted or woven in a custom prepreg form.

*Figure 27. Draping capability of a (room temperature) commingled glass/PET fabric.*
CHAPTER 4 Temperature and pressure distributions during a rubber forming cycle.

In the preceding two chapters, the importance of temperature and pressure in both the forming and the consolidation subprocesses was emphasized. In this chapter the distribution of the process parameters temperature and pressure will be dealt with more extensively. In the first section, the temperature history during rubber forming will be described; the second section of this chapter deals with the pressure parameter.

In this chapter, a typical temperature-pressure-time profile will be used from which several sections will be recognized, both in the temperature and the pressure lines. Such a general rubber forming process profile is given in figure 28. The most important points in the rubber forming cycle are distinguished and numbered.

![Diagram with temperature and pressure profiles and process stages]

Figure 28. General rubberforming process profile (temperature and pressure as a function of time).
Starting at point 0, the (preconsolidated) laminate is heated by a specific heating device until the processing temperature is reached at point 1. Here the laminate is quickly transferred to the forming equipment or the heating device is removed from the forming equipment. In this schematic presentation, the transportation step proceeds so smoothly that its influence on the temperature profile is neglected.

The following step is the forming of the laminate. At point 1 a rubber stamp is pressed against one side of the hot laminate and forces the laminate into the rigid die. At point 2, the other side of the laminate touches the rigid die. In the figure it is assumed that the mould is completely filled at point 2 and the composite material is pressurized to the desired consolidation pressure.

When the formed composite product is cooled below a certain temperature, the mould is opened (point 3) and the composite part is removed from the forming equipment. Then it will cool further to room temperature (point 4).

The above mentioned point numbers and indices from figure 28 will stay resident in this chapter and will be used to discuss the several subprocesses in the next sections.

4.1 Heat transfer in rubber forming.

Heat transfer is one of the most important issues in a thermoforming process\(^\text{17}\). Temperature is not only a main process parameter that is responsible for the quality of the formed product, but the rate of heat transfer is also the bottle-neck in the optimisation of thermoforming cycle times and indirectly in the processing costs. The heating cycle (point 0 to point 1) accounts for a large part of the processing time in the rubber forming cycle. Therefore it is important to make this step as short as possible.

In section 2.3 several heating devices were mentioned and some of their advantages and disadvantages were pointed out. In general it can be stated that heating of a laminate by IR radiation is favourable because of the ease and high heating rate of this system. In figure 29 the temperature profile is sketched of a 10-layered glass fabric reinforced PEI laminate that is heated by short wave infrared radiation. It shows not only the short heating time but the profiles also give an expression of the main disadvantage of IR-heating; the high temperature gradients through the thickness of such a quickly heated laminate.
Concerning cycle times, the importance of the heating rate will be less when the heating is taken care of outside the forming equipment in a way that does not occupy any other (costly) devices than the heating facility.

![Diagram of temperature distribution in a laminate.](image.png)

*Figure 29. Temperature profile of an infrared heated 10-layered glass/PEI laminate.*

At point 1 of the time scale of figure 28, the hot laminate is transported to the forming equipment. In reference 18 is demonstrated that this step takes a certain time during which the sheet will cool down. Because the transportation time can be reduced to obtain a minor temperature influence by more advanced mechanical transportation and/or insulation of the sheet, this transportation time and the involved cooling down will be neglected in this report. The dip in the temperature profile of the laminate at point 1 is caused by the sudden contact between the (cooler) rubber stamp and the laminate sheet.

As long as the laminate only contacts the rubber mould on one side, the physical boundary condition of that side has to imply a heat transfer due to the temperature difference between the rubber and the composite, probably with a certain thermal contact resistance. The heat transfer at the other side of the laminate will be relatively low because it is only dictated by free convection (radiation is neglected) due to the fact that the free lower side only contacts stagnant cool air (figure 30).
Figure 30. Cooling rates of an 8-layered carbon/PEI laminate in different media.

The boundary conditions however will change with time during rubber forming. The initial temperature difference between the rubber mould and the composite material will decrease and theoretically it will become zero when the system reaches a certain equilibrium temperature. The equilibrium temperature of the one-sided contact between the rubber mould and the laminate is called $T_{eq,\text{rubber}}$ in figure 28.

The forming time (point 1 to point 2) is too short for the system to actually reach the equilibrium temperature $T_{eq,\text{rubber}}$ because the laminate will be forced into the rigid metal die and then contacts it at point 2.

From point 2 the consolidation will start and the cooling rate of the laminate will increase because the convection of the former free lower side of the laminate will be replaced with the more rapid heat transfer to the metal die by conduction. Figure 31 taken from reference 18 gives an impression of the influence of the laminate-metal contact on the cooling rate. It is therefore no surprise that the cooling rate of the composite material will further increase from point 2 until the temperature of the system reaches a new equilibrium temperature $T_{eq,\text{total}}$. $T_{eq,\text{total}}$ is the equilibrium temperature of the system in which the laminate contacts the rubber stamp and the metal die.
The equilibrium temperature depends primarily on the relative heat capacity of material and moulds, whereas the time needed to reach this equilibrium mainly depends on the conductivity and the thermal contact resistance between the different materials in the system. When the temperature of the laminate drops below the opening temperature $T_{\text{open}}$ (determined by matrix solidification), the mould will be opened (point 3) and the composite part will cool down further outside the mould (point 3 to point 4).

![Cooling of an 8-layered carbon/PEI laminate during rubber forming cold molds, different locations](image)

*Figure 31. Cooling rates of an 8-layered carbon/PEI laminate, monitored through the laminate.*

In the preceding chapter, the importance of temperature control during forming is emphasized. The importance of temperature control in the consolidation phase will be explained somewhat further in this section.

To fully consolidate a thermoplastic laminate (see chapter 2), it is necessary that the material is pressurized during a certain time, at a sufficiently high temperature. Experiments prove that these three process parameters are partly interchangeable inside the processing window with regard to the mechanical properties of the final consolidated product. This means that to obtain good consolidation in CFRTP products in a thermoforming process that can only supply a limited amount of consolidation pressure,
it is necessary to optimize the temperature-time relation. For non-isothermal consolidation as is the case of rubber forming, it is important to use the short time span point 2 to point 2' of figure 28 where the temperature is high enough to consolidate.

Figure 28, however, only shows the mean-temperature profile during a rubber forming cycle. The temperature distribution over the laminate thickness as well as the temperature distribution along the laminate can be fairly different. In figure 31 is clearly depicted that the cooling rate of a laminate is higher at the side of the laminate that contacts the metal die.

During the rubber forming cycle, the heat transfer situation in the laminate that is introduced can be described by the general 2D energy equation\textsuperscript{19,20}:

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

where:

- \( k_x \) = thermal conductivity in the x-direction (W/m\(^\circ\)C)
- \( k_y \) = thermal conductivity in the y-direction (W/m\(^\circ\)C)
- \( \rho \) = density of the composite laminate (kg/m\(^3\))
- \( C \) = heat capacity of the composite laminate (J/kg\(^\circ\)C)
- \( g \) = heat generation due to chemical reactions or to crystallization (W/m\(^3\))
- \( T \) = temperature (\(^\circ\)C)
- \( t \) = time (sec)
In general, the different temperature distributions can be calculated from the energy equation by adding the right boundary conditions over time and place in the laminate. It will also be important to include the right material properties, concerning heat transfer.

The calculation of temperature distributions will imply the use of energy equations with time dependent boundary conditions. It will probably be necessary to add the third dimension to the heat equation in case of rubber forming of strongly asymmetric composite products.

The solution of the energy equation is beyond the scope of this introductory report, but it will be clear that the variations of the temperature through and along the laminate with time will be large during forming and consolidation.

In practice, full consolidation has to be obtained all over the laminate. The part with the highest cooling rate will determine the available consolidation time (point 2 to point 2'). Usually this bottle-neck in the laminate is the section just beneath the outer layer that has to be "consolidated" with the rest of the laminate which was the first spot to contact one of the dies. This section is the first part of the thermoplastic material that will start to cool down. Therefore it will be the critical part in the product and it is the appropriate spot to monitor the temperature to obtain the minimum consolidation degree.

As a last remark about the temperature history in this report, it must be noted that it is possible to create a more homogeneous temperature distribution during forming and the subsequent non-isothermal consolidation by:

- (partly) isolating the laminate from the cold moulds by using separation foils;
- using mould materials that have low conductivity and ensure low cooling rates;
- preheating of the dies to decrease temperature differences between mould and composite laminate (an example of lower cooling rates is given in figure 32).

In this way the temperature profile of the processed laminate is stabilized and more time will be available for both forming and (re)consolidation of the thermoplastic composite, without having to raise the temperature of a laminate too high at the start of a rubber forming cycle.
4.2 Pressurization during rubber forming.

The main characteristic of the rubber forming technique is the way in which the composite laminate is pressurized during the thermoforming cycle. To assure optimal deformation of the laminate and additional good consolidation, the right amount of pressure has to be applied at the right moment.

The influences of the pressure distributions on the forming and consolidation subprocesses have already been discussed. In this section the realisation and determination of the pressure profiles in a rubber forming cycle, using male rubber stamps will be dealt with.

Looking back at figure 28, the pressure in the forming phase (point 1 to point 2) is kept low, because high pressure has a negative influence on the formability of continuous fibre reinforced thermoplastic sheets. On the other hand, it is important to create local pressure on the laminate to introduce the necessary forming forces. The freedom to
optimize the form of the stamp for good deformation of the reinforcement is relatively high. This is because of the fact that the stamp is made of rubber that is flexible enough to be able to adapt to the rigid die together with the deforming laminate. The introduction of intentional "mismatches" between flexible rubber die and rigid metal die as a means of facilitating laminate deformation is a powerful tool in rubber forming but will not be discussed any further here. The full understanding of this feature requires modelling of the deformation of the rubber stamp.

To adequately control the forming and consolidation however, it is important to be able to determine the pressure profiles at the thermoplastic sheet during a rubber forming cycle. Up to now, pressure measurements have been successfully carried out only during the consolidation phase by measuring the imposed pressure of the laminate on the rigid die half. In this report only a two-dimensional product shape will be discussed, but more complicated mould shapes can often be divided in several simple two-dimensional sub-shapes.

In figure 33 from reference 21, a typical pressure profile is given of a U-shaped product in the consolidation phase. This profile is measured by pressure sensors in the rigid die half. Figure 33 shows the high influence of the mould shape on the pressure in the laminate. It is important to be aware of the consequences that a mould form has on the pressure distribution in the consolidating composite sheet, especially when the available consolidation time is short due to a high cooling rate in that particular mould.

![measured local pressure](image)

*Figure 33. Typical pressure distribution during the consolidation of a U-shaped beam.*
Two important mould shape parameters that influence the pressure distribution are:
- width to height ratio of the mould;
- tolerance between the rubber and the metal die.

Figure 34a gives the influence of the width to height ratio of the mould on the pressure profile in a laminate that is consolidating in a U-shaped mould. Figure 34b gives the influence of the tolerance of the dies. Friction on the laminate that is moving into the metal die (in the forming phase) is an important cause for these pressure phenomena in the consolidation phase, as will be explained later on.

![Figure 34a. Influence of the width to height ratio of a U-mould on the pressure profile.](image)

Another important mould parameter that determines the pressure profile during rubber forming is the hardness of the rubber die half. The hardness, that is mostly referred to in *shore A hardness* units, is directly related to the stiffness of the rubber. The capability of the rubber die to compensate a mismatch with the metal die is therefore determined by the hardness of the stamp. What hardness to use in the rubber forming process largely depends on the dimensional complexity of the shape of the final composite part.
In figure 35a the pressures in the corners of a consolidating U-shaped beam are depicted, measured in forming experiments with different rubber dies. It can be noticed that when a relatively hard rubber is used, the corner pressures will be relatively low. This occurs because a higher hardness rubber less easily fills up the corners in the rigid die in the end of the forming stage. This can also be translated to the fact that the use of a low hardness rubber will facilitate the forming of small details in a CFRTP product.

Figure 35b however, shows that when the width of the rigid metal die is decreased by 2% and the rubber die stays the same, the results will change. Though the used moulds in the experiments of figure 35b leave sufficient space for the laminate, there always will be some friction between the sliding laminate and the rigid die-half. This friction introduces tensile forces in the laminate that must be counteracted by the stamp. During forming the tensile forces in the laminate will deform the rubber stamp. A "soft" rubber stamp will deform most and a so-called barrelling effect will occur (figure 36). This barrelling will induce even more friction during forming and the rubber stamp can clamp itself in the female rigid die. In this way a kind of cavity is created in the corners, that will obstruct the pressurization of the laminate in the corners of the U-beam. The corner pressure will be lower using a 60 shore A rubber stamp, rather than using a 78 shore A rubber stamp when the sliding space between rubber and metal die is decreased, as depicted in figure 35b.
Figure 35a. Influence on the pressure profile of the hardness of the rubber die in a wide U-mould.

Figure 35b. Influence on the pressure profile of the hardness of the rubber die in a narrow U-mould.
Figure 36. Schematic view of the barreling effect of rubber dies during rubberforming.

It may be clear that there is a discrepancy in rubber mould design. To be able to realize highly detailed products, relatively low hardness rubbers have to be used but to ensure a high stability of the rubber die form, high hardness rubbers are recommended. Beside the choice of a high tolerance mould design, a possible solution to this discrepancy can be the use of a hybrid rubber stamp, composed of rubber sections with different hardnesses (figure 37). When these features really are controlled, it will be possible to optimize the pressure distributions in both the forming and consolidation cycle for a particular mould shape. Only then, the advantages of rubberforming can be exploited.

Figure 37. Hybrid rubber die composed of two kinds of rubber with different hardness.
CHAPTER 5 The viability of rubber forming.

As the rubber forming method is a rather new manufacturing technique developed for the processing of continuous fibre reinforced thermoplastics, a large amount of further research needs to be done. In this final chapter some additional points of attention of the development of rubber forming will be discussed.

First of all the importance of process control will be emphasized. Measurements and control of the parameters during a rubber forming cycle could perhaps ensure a certain predetermined product quality. In the second section, examples of research activities are given that attribute to the further understanding of rubber forming processes. Finally, an overview of expected future work is given.

5.1 Quality control by process control.

An important issue to investigate in the field of continuous fibre reinforced thermoplastics is the reliability of the thermoforming processes that is needed to accomplish the desired final composite parts. It is a fact that the full understanding of a thermoforming process and high product quality cannot be uncoupled. The foregoing sections of this report demonstrated the necessity of investigations of rubber forming subprocesses in order to obtain good thermoplastic composite products. In this section, however, a more far-reaching point of view in process control is discussed.

Often, material properties of thermoplastic composites are determined after consolidation and are used in strength and stiffness calculations of structures that are made from these materials. In section 5.2 however, will be depicted that the mechanical properties of consolidated material will be "deformed" together with the laminate in a thermoforming process, influenced by local fibre and matrix deformations and by the achievable reconsolidation degree. Making strength calculations using undeformed material properties therefore can locally result in serious mismatches with the real fabricated CFRTP structures.

The main question in this section is whether the understanding of the rubber forming technique could become complete enough to ensure a predicted quality of the composite part with a high degree of certainty. Is it possible to guarantee specific mechanical
properties of a rubberformed product by applying certain pre-determined rubber forming parameters?

In this way, process control will become even more important. If it is possible to prove that the mechanical properties of a composite part fully depend on the processing conditions and the quality of the "raw" material it is processed from, than it can be put forward that quality control of thermoplastic composite parts really is the process control of the thermoforming technique.

The mechanical destructive testing of shaped composite products is costly and often difficult to accomplish in a realistic way\textsuperscript{22}. Non-destructive testing is perhaps even more difficult and in critical spots like radii and thickness gradients even impossible. With quality control by severe (approved) process control, the need for controlling and/or testing each single product will decrease. The quality of CFRTP products can be determined by the measurement and control of the parameters in the rubber forming technique, preceded by actual mechanical tests on undeformed material (relatively easy to perform). The acceptance of such a quality control for rubberformed products by for instance airworthiness authorities will contribute to the potential of the rubber forming method of being a thermoforming process with low development and production cycle times.

Again it must be stated that to allow quality control of rubberformed CFRTP products by control of the rubber forming process, the forming technique with all of its features must be thoroughly investigated and understood.

5.2 Experiments to determine the influence of rubber forming cycles.

The lack of production process data for thermoplastic composite parts obstructs the application of these materials in primary load-bearing structures. Even more important perhaps, is the underestimated difference between the mechanical properties of composite materials that come directly from the material supplier and the mechanical properties of the same material after transformation into a final product. Therefore, beside the process investigations already mentioned in preceding chapters, research needs to be done more directly applied to the influence of rubber forming processes on the properties of CFRTP parts.
An example of research effort in the field of determination of the influence of a rubber forming cycle is given in this section. It is an illustration of the relations between material and process dominated phenomena in a rubber forming cycle.

To be able to get information about the influence of the (sub)processes that a laminate will undergo, the following (preliminary) experiments have been carried out.

Four laminates (500*500 mm²) of 10 layers TenCate glass fabric reinforced PolyEtherImide (glass/PEI) were symmetrically stacked. These laminates A, B, C and D were consolidated at different consolidation parameters. Each sheet was divided in three parts after consolidation. The first part was used to determine reference material data. The other parts were pressure-less heated again and respectively rubberformed into a flat panel (no deformation) and rubberformed into a hat section (interply slip deformation).

Specimens were prepared out of these parts to perform 0,90° tensile tests (ISO D3039), compression tests (EN2850) and four point bending tests (EN2563). By testing the specimen, the influence of the subprocesses were investigated:

- consolidation
- flat rubber forming
- 2D rubber forming

The results that are shown in the following sections are a representative selection of the numerous test data.

The four glass/PEI prepreg stacks were consolidated in a hot platen press at different parameters:

Specimen A: 20 minutes at 320°C at 1 N/mm² consolidation pressure
Specimen B: 20 minutes at 300°C at 1 N/mm² consolidation pressure
Specimen C: 20 minutes at 280°C at 1 N/mm² consolidation pressure
Specimen D: 20 minutes at 280°C at 0.5 N/mm² consolidation pressure
Because of the prepreg production process and transport, the woven fabric fibres in the prepreg are more curved in weft direction than they are in warp direction of the fabric (figure 38). This causes better mechanical properties in warp direction of the laminate (figure 39). Both stress at failure and stiffness of the material are higher in warp direction.

During consolidation of a fabric reinforced thermoplastic sheet, matrix flow is responsible for the fusion of the prepreg layers. It is, however, responsible for another phenomenon. Due to the matrix flow from the middle of the sheet (highest pressure) to the sides, the
reinforcing fabric will be stretched. The difference in curvature of warp and weft fibre bundles will be decreased. At higher consolidation pressures and/or temperatures, an increasing matrix flow will decrease the warp-weft difference more as is clearly shown in figure 39. The influence of the also occurring thickness variation in the laminates will not be discussed here.

Flat rubber forming

When after a separate consolidation cycle (as in the experiment) a thermoplastic composite product is thermoformed, an additional pressure process takes place at elevated temperature. During rubber forming, the cooling down of the laminate to the glass transition point only takes a few seconds and therefore gives little opportunity to the fabric to stretch any further due to matrix flow. However, mechanical tests on rubberformed flat laminates show that the difference between warp and weft directional mechanical properties further diminishes (figure 40). This phenomenon is clearest when a badly preconsolidated laminate (specimen D) is rubberformed or pressed (3 MPa at 300°C starting temperature). It shows that a subprocess can (partly) cover the influence of the preceding one.

Figure 40. Influence of several rubberforming subprocesses on four point bending results of glass/PEI laminates.
Rubber forming of a hat section

Parts of each preconsolidated glass/PEI sheet were heated up to 300°C and rubberformed into a hat section. Out of the sides and top of the hat section, specimen were cut which were tested in the four point bending fixture. The tests showed the influence of the direction in which interply slip has to take place during thermoforming. Because this slipping introduces tensile stresses in the layers of the laminate and therefore stretches the fibre bundles in slip direction, the stiffness of the material will increase in this direction and indirectly the strength will, too.

Although it actually needs a discussion of the failure mechanism, the differences in maximum stress at failure, shown in figure 40 show that the direction of the mean deformation has an influence on mechanical properties.

Discussion

The experiments were carried out on consolidated laminates from one particular prepreg lot. Each further process (flat and 2D rubber forming) therefore gives "transformed" material on which standard tests generate transformed material properties. The results of the four point bending test clearly indicate the influences of consolidation, additional heating and pressure cycles and deformation direction of the laminates.

Experimental conclusions

1. The quality of the fibre reinforcement of prepreg material influences the laminate’s properties.

2. The (pre)consolidation quality influences the final material properties of a glass/PEI product.

3. When more subprocesses are involved in a thermoforming cycle, the influence of one of the subprocesses on product quality decreases.

4. Product properties of thermoformed products can depend on deformation direction.

5. The experiments indicate the importance of the quality of the last step in the fabrication cycle of a thermoplastic composite product.
5.3 Future investigations.

The rubber forming technique is a fast and potentially adequate thermoforming technique that needs further development. In section 5.2 an example was given of a way of investigating the consequences of rubber forming on material properties. Only a simple product shape has been considered, but the need for this kind of research has been demonstrated. It can therefore be justified to carry on with investigations on the influence of the different kinds of subprocesses in rubber forming on the material properties of final CFRTP products.

First of all, the changes in the thermoplastic laminate during thermoforming have to be examined. The deformations as stated in chapter 3 will influence product properties other than those induced by a possible change in fibre directions. In section 5.2 for instance is shown how interply slip phenomena induce high mechanical properties in slip direction. It may be expected that other laminate deformations show similar influences.

The important process parameters time, temperature and pressure will most likely not only influence the deformation capabilities of thermoplastic laminates but the final mechanical properties of CFRTP parts as well. Because of the fact that the rubber forming technique is a highly non-isothermal and non-isobar thermoforming technique, the measurement and control of the process parameters during a rubber forming cycle is important. More research is desired on these subjects.

Because of the state of infancy of the rubber forming process, more research effort has to be made in optimisation of the technical side of the process. The application of pressure and temperature on laminates and the relation with time during a rubber forming cycle has to be guaranteed. Development of tools will play an important role. In order to prove rubber forming a viable thermoforming technique, the realisation of the "hardware" is needed.

The introduction and application of a new manufacturing technique is not merely a technical issue. The cost versus performance of the rubber forming process is an important consideration in a potential industrial market introduction. The existence of several kinds of CFRTP processing industries (e.g. aerospace and automotive industry) implies different approaches in manufacture of thermoplastic composite products. It is therefore necessary to investigate the economic features of the rubber forming technique. Material costs, mould costs and life times, energy consumption, cycle times, size of
product series, etc. have to be mapped.

However, to get a clear picture of the potential of rubber forming, there is still the need to know and understand the technical performance of this thermoforming process and it may be obvious that the limits of rubber forming are not yet clearly defined.
References.


9> Robroek, L.M.J., *Material Response of Advanced Thermoplastic Composites to the


