Time Temperature Relations During Rubber Forming Continuous Fiber Reinforced Thermoplastics

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SUMMARY.

Preliminary tests have been carried out in order to determine the cooling speeds of CFRTP laminates during rubber forming. It appeared that cooling speeds are rather high, which might become a problem when a large reconsolidation process during the pressing phase is obligated. However, several heat-saving measurements are mentionable and can be taken to tune the cooling ratio in such a way that optimum material properties with respect to optimum production cycle times are obtained.

The test setup needs to be altered. It is strongly recommended that an automated data acquisition system is purchased in order to ease further experiments with respect to this highly interesting subject.
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1. INTRODUCTION

Rubber forming continuous fiber reinforced thermoplastic materials is a feasible technology with respect to commercially viable production methods of CFRTP parts. One of the major advantages of the rubber forming principle is the possibility of fast production cycles. The inherently high cooling rate put on the laminate is an important issue for investigation.

Some preliminary experiments have been conducted in order to get a first impression of the magnitude of these cooling rates. Tests have been carried out in which the cooling speeds of carbon/PEI and glass/PC laminates under different conditions have been determined.

The results of these experiments indicate that feasible cooling speeds can be obtained by tuning the time-temperature history of the laminate. This tuning can be realized by adjusting starting temperature, temperature of the metal mould and the use of co-heated, isolating layers (PI-foils), given the time span dictated by the used equipment.

The tests have been carried out without these isolating layers which means that the given results give the cooling speeds under the most stringent conditions.

In the second chapter some background information on the rubber forming process will be given. The tests (materials and set-up) are described in chapter 3 while in chapter 4 the results will be discussed. The last chapter is contributed to the conclusions and recommendations for further investigations.
2. BACKGROUND AND OBJECTIVES OF THE EXPERIMENTS.

2.1 Introduction.

The rubber forming process can be described using a time-temperature history of a product from blank to the actual form. This history belongs to a method of production as given in fig.2.1, which is: separate heating in a hot platen press, forming in a fast moving press, transport by hand. A number of different stages can be distinguished, see fig.2.2:

0-1: heating period in hot platen press
1-2: temperature stabilizing period, also buffering possibilities
2-3: transport from heating to forming press
3-4: forming phase
4-5: pressing phase
5-6: Opening of the mould, product temperature well below $T_s$
6-7: Further cooling down to handling conditions

The interesting part of this figure is the period between 2 and 6, which in fact is the actual production time of one product for one mould. The parts 0-2 and 5-7 will not be discussed in detail because their contribution to the success of the forming and pressing phase is nihil.

Keeping period 2-3 as short as possible will enhance the quality of the product without the necessity of heating up to higher buffer temperatures to cope with excessive cooling during handling. Of course, the importance of this period increases when large blanks have to be handled.

In the following emphasis will be put on the periods 3-4 and 4-5, the forming and the pressing period.

The most stringent condition with respect to the time-temperature relation is investigated which occurs when during the forming phase the laminate makes contact with both moulds.
In fact this is only valid for certain parts of a certain range of products. If the flanges of a product are parallel with the movement of the press immediate contact takes place. Other forms give the possibility of sustained high temperatures during the forming phase. Further investigations on this field will be carried out.

First the differences between the forming and the pressing phase will be defined.

2.2. The forming phase.

The forming phase in the rubber forming process is defined as this period in which the blank is formed from a flat plate into a 2- or 3-dimensionally formed product. Depending on a number of parameters (thickness of the laminate, the complexity of the form etc.) ply-slip and shearing of the fiber bundles will occur. Each mechanism has been utterly described in literature\textsuperscript{1,2,3}, hence no explanation of these phenomena will be given here.

It is emphasized that these mechanisms occur smoothly only when the temperature of the thermoplastic matrix material is well above the minimum autohesion-temperature. Depending on the complexity of the form the effects of the forming mechanisms on the homogeneity of the laminate do urge for a certain amount of reconsolidation in order to retain the desired mechanical properties of the composite material. It is clear that this reconsolidation proces, which has to be started well before the minimum autohesion temperature has been passed, puts restraints on the forming time.

2.3. The pressing phase.

The pressing phase starts when the large forming mechanisms have stopped. No ply-slip or bundle shearing will occur anymore and
all the pressure put on the product is used to reconsolidate the laminate. This reconsolidation process can only be achieved when the parameters pressure, time and temperature \((p,t,T)\) are within a feasible window.

In this report it is assumed that during the pressing phase no large matrix movements (for instance needed for impregnation or a smooth surface) will occur. In other words, only the retention of the interply cohesion after pressing is important. Restoring the broken, cohesive bonds between the slipped layers is called autohesion. The minimum autohesion temperature is that temperature below which no cohesion (under low pressure) can occur anymore. The autohesion time, the time needed to obtain a 100% cohesive area, depends strongly on the viscosity of the matrix material, hence on the temperature of the material. It is generally acknowledged that at a rather low viscosity (e.g., high temperature) the autohesion period of even high performance thermoplastics \((PEI,PES)\) can be very short.

However, all tests regarding this topic have been conducted at constant temperatures, while the principle of rubber forming is the short production cycle, hence a large temperature gradient is wanted. The maximum allowed gradient (from material point of view) is dictated by the time needed for a 100% restored autohesion \((t_{4.5})\) and the minimum autohesion temperature \((T_{5})\). Regarding the used equipment it is important to know how much time can be consumed for forming \((t_{3.4})\) before the pressing phase has to be started. This period in turn depends on the cooling speed of the laminate during forming.

Therefore a thorough knowledge of the time temperature relations during rubber forming is very important.

In order to get global information on these topics the cooling speeds of hot laminates during handling as well as during forming have been determined.
3. PRELIMINARY EXPERIMENTS.

3.1. Introduction.

The used material has been a carbon/PEI combination often used in our laboratory (CD282 produced by Ten Cate Glas), the number of layers has been varied from 2, 4, 6 to 8. Furthermore, some tests have been carried out on a glass/PC combination (thickness 1 mm). First, the cooling speed of a hot laminate (with varied thickness) has been determined in various media:

- water  (for comparative reasons)
- silent air  (transport period 2-3)
- turbulent air  (idem)
- cold aluminium and PU moulds  (forming and pressing)

From the results it appeared that the cooling speed in cold moulds is very high, hence the same tests have been conducted using a hot aluminium mould.

3.2 Test setup.

The test setup is shown in fig. 3.1. For the heating of the blank a hot platen press has been used. After heating up to the desired starting temperature ($T_s$) the specimen was taken out the platen press and put on the rubber mould in the fast closing press, which was then closed (within 0.5 s) for one minute. Using the same fast method of transport throughout the experiments the cooling of the laminates during transport was minimized and equalized for all tests. The heating of the aluminium mould has been realized by hot air guns. By adjusting the distance of these guns to the aluminium mould steady temperature conditions of the mould could be obtained.
K-type thermocouple wires were used to measure the temperatures in the blank and the moulds. Measurements have been performed at three different locations in the laminate to account for the heat capacity of the material. Fig. 3.2 shows that three different temperature gradients exist over the cross section of the laminate due to different heat absorption coefficients of the rubber and aluminium moulds.

The temperatures in the aluminium and rubber mould were measured simultaneously. The position of the thermocouple wires in the moulds has been 1 mm from the surface. This position will obviously result in a time delay of the measured temperature, which will be altered in further investigations.

In order to minimize the influence of the 'dead time' of the used equipment, the output voltages were sent to a X-t writer directly. The X-t writers were adjusted to a speed of 600 cm/min. After reading the curves the obtained data were adapted into figures using a spread sheet program.

3.3. Results.

3.3.1. Cooling in different media.

First test have been carried out cooling a hot Carbon/PEI laminate (T_s = 300°) in various media (water, AL/PU moulds, silent and turbulent air). By adding two layers (upper and lower side) subsequently the influence of increasing thickness has been determined. Note that the thermocouple was situated in the middle of the laminate for all tests, hence the heat capacity of the outer layers are incorporated in the results, (fig.3.3a to 3.3d). From fig.3.3 it can be deduced that the cooling behaviour of a heated laminate in air (turbulent or not) gives enough opportunity to replace the blank from the platen press to the forming press. The results also show that the rubber forming process using cold moulds result in a very fast cooling rate.
Excerpted from these results fig. 3.4 shows the cooling of 2, 4, 6 and 8 layers between cold aluminium and PU-moulds. Because of the position of the thermocouple (midplane) these results are rather optimistic, as will be shown later.

3.3.2. Cooling with a hot aluminium mould.

The results of the tests using a hot aluminium mould are shown in fig. 3.5a to 3.5f (C/PEI) and in fig. 3.6a to 3.6e (Glass/PC) for different temperatures of the aluminium mould. The temperature in the mid-plane of the laminate (8 layers) has not been recorded. The cooling and heating aspects of the various parts are evident. The time-temperature relations (average temperature) with different aluminium mould temperatures for the C/PEI laminate have been summarized in fig. 3.7. It can be seen that by using a hot aluminium mould an (expected) increase in available processing time can be realized. If, more or less arbitrary, the $T_s$ of PEI (217°C) is taken as a lower boundary, the period available for the pressing phase with a 200°C aluminium mould is more than two times as large as the period in which a cold aluminium mould (50°C) is used.
4. DISCUSSION OF THE RESULTS.

The major conclusion resulting from the experiments is that the available time for processing using cold moulds is quite short. However, from the autohesion model described by Jaarsma\(^4\) and others\(^5\) this short period of time should be enough for an autohesion process after forming, especially in relation to high pressures. Of course, the available time left for a reconsolidation in the pressing phase is depending on the time consumed during transport and the forming phase. Measures to be taken if the transport and/or the forming phase take too much time can vary from using isolating foils, heated metal moulds up to higher starting temperatures. Experiments in which actual (small) products have been rubber formed applying heat-saving measurements indicate that reconsolidation occurs.

In the above the physical behaviour of the matrix material is not considered yet. However, the viscosity of the matrix material at higher temperatures is a very important parameter regarding reconsolidation in short times. Other experiments indicate that rubber forming CFRTP with low-viscous matrix materials yields good results (with regard to reconsolidation) in short production cycle times.

Using low-viscous matrix materials it can even be considered to use unconsolidated prepregs (cowovens, woven comingled) as blank material. The events taking place during the total process as shown in fig.2.1 will then be: a halfway consolidation during the heating phase (0-2) while during the pressing phase a 100% consolidation is achieved. This topic is further investigated using matrix materials like PA, PET and PP.

One of main problems however will still be the determination of the degree of consolidation and the degree of reconsolidation. Some experience in this field has been obtained. For instance, to determine the (qualitative) degree of consolidation of a glass/TP laminate a visual inspection can be performed.
A good consolidated glass/PEI laminate will be transparent (yellow) before reheating. This is a result of a smooth, equalized refraction of the light rays through the laminate, the matrix material does enclose the fibers to such an extent that no false refractions occur. When taking out a hot (glass/PEI) blank from the hot platen press it is obviously observed that the transparency has gone, indicating a loss in homogeneity. After pressing (cooling under pressure) the degree of recovered consolidation can be 'measured' from the grade and homogeneity of the transparency.

This visual inspection is one way of determining the restoring influence of the pressing phase, other methods include testing of the mechanical properties (ILS-values, ±45 tensile test and so on).

In order to define the processing windows for different materials more accurately these topics need to be investigated yet.

With regard to the forming and pressing phase the most important issue is the minimum value of the temperature at point 5 (the end of the pressing phase) coupled to the minimum period needed for a 100% reconsolidation. This issue is now being investigated for glass and carbon fiber reinforced PEI materials.

Furthermore, a processing window with respect to the processing parameters pressure, time and temperature must be evaluated for different materials and different thicknesses of these materials.
5. CONCLUSIONS AND RECOMMENDATIONS.

5.1. Conclusions.

From the results of the preliminary tests a number of conclusions can be drawn:

- Measurements of cooling speeds during fast forming processes are possible.
- The cooling speed during transport is low.
- The influence of the laminate thickness on the cooling speed is large.
- The influence of hot moulds on the cooling speed is large.
- Cooling speeds during rubber forming CFRTP are high, but can be adapted to desired levels using various heat saving methods.

5.2. Recommendations.

It is recommended:

with regard to the test set-up:

It is strongly recommended that an automated data acquisition system is purchased. This will enhance the accuracy of the measurements and the tests can be conducted faster. Furthermore, using such a system opens the possibilities of simultaneously monitoring pressures and temperatures which will give several features:

- By conducting a number of simple tests the processing (ptT) window of new fiber/TP combinations can be determined.
- Existing processing windows (for known combinations) can be optimized and determined with regard to thickness, fibervolume contents and so on.
- With regard to the industry: "Live" quality control

Furthermore the location of the thermocouple wires in the specimen (delays due to heat capacity of layers between contact area and TC wire), and in particular the aluminium mould (galvanic conduction) needs to be chosen well. For instance, the hot weld of the TC wires could be dipped in a PI solution to prevent galvanic contact between the weld and the aluminium block.

With regard to the material:

 Tests have to be carried out in order to investigate the role and effects of:
- laminate thickness
- sort of laminate
  - fibers
  - matrix
- degree of preconsolidation or not
  - two stage consolidation depending on the viscosity of the matrix material.
  (From earlier investigations it appears that the pressing time needed for a 100% reconsolidation depends (among other parameters) on the grade of consolidation before forming, given the autohesion time of the matrix material. see also literature (8)).

In general:

- Investigation of the rather clear proposition:
  "surface quality 'ok' & autohesion 'ok'"
- A theoretical background of the time-temperature relations\(^7\) must be matched to the experimental results. For instance in order to create an expertsystem which can determine the ptT window for various combinations of CFRTP.

- Finding ways to determine the influence of the rubber forming process on the material properties. Preliminary tests have been carried using the ILSS method with plausible results, but no standard is known yet.

Further actions related to the rubber forming process will also include discussions of the industrial possibilities of this process. One of the main goals with respect to a commercially viable production method is to keep the production cycle short. Rubber forming offers cycle times per mould from 5 to 60 seconds, which is possible from material point of view. However, this objective can only be purchased if all used materials (moulds, isolating foils and so on) are capable of withstanding high temperature shock loads combined with high pressures.

For instance, because of the (known) rather low continuous use temperature of the material of the rubber mould (PU) the steady state temperature during production should be kept under a certain value to avoid excessive wearing and sustained deformation of the PU-mould. Forced cooling of the rubber mould might be necessary when forming at high speeds.

Investigations on all of these issues are ongoing.
LITERATURE.

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fig. 2.1: Method of transport (by hand)

Fig. 2.2: Typical $T-t$ relation for a rubber forming process cycle.
Fig. 3.1 Test set-up.
Cooling of an 8 layer C/PEI laminate during rubber forming cold molds, different locations

![Graph showing cooling over time](image)
cooling of a 2-layer C/PEI laminate in different media
thermocouple situated in midplane laminate
cooling of a 4-layer C/PEI laminate in different media

thermocouple situated in midplane laminate

TIME [h]

0 5 10 15 20 25 30

[Temperature]

0 50 100 150 200 250 300

water - - - - - - -
Al-FU molds - - - - - - -
silent air - - - - - - -
turbulent - - - - - - -

Fig. 3.3b.
cooling of a 6-layer C/PEI laminate in different media
thermocouple situated in midplane laminate

TEMP [°C]

TIME [s]

water
AI–PU molds
calient air
turbulair

Fig. 3.3c
cooling of a 8-layer C/PEI laminate in different media
thermocouple situated in midplane laminate

\[
\begin{align*}
\text{TEMP [°C]} & \\
300 & \quad 250 & \quad 200 & \quad 150 & \quad 100 & \quad 50 & \quad 0 \\
0 & \quad 5 & \quad 10 & \quad 15 & \quad 20 & \quad 25 & \quad 30
\end{align*}
\]

- water
- Al-PU molds
- silent air
- turbulent
Cooling of 2, 4, 6 and 8 layer C/PEI laminate during rubber forming using cold moulds
Cooling of an 8 layer C/PEI laminate during rubber forming

$T_A = 50^\circ C$

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**Diagram:**

- **Laminate**
- **Al side**
- **Laminate**
- **PU side**
- **Al mould**
- **PU mould**

**Graph:**

- **Y-axis:** Temp (°C)
- **X-axis:** Time [s]

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FIG. 3.5a.
Cooling of an 8 layer C/PEI laminate during rubber forming

\[ T_{AI} = 100^\circ\text{C} \]
Cooling of an 8 layer C/PEI laminate during rubber forming

\[ T_A = 125^\circ C \]
Cooling of an 8 layer C/PEI laminate during rubber forming

\[ T_{AI} = 150^\circ C \]
Cooling of an 8 layer C/PEI laminate during rubber forming

$T_A = 175^\circ C$
Cooling of an 8 layer C/PEI laminate during rubber forming

\[ T_{AI} = 200^\circ C \]
Cooling glass/pc (t=1 mm) during rubberforming

$T_A = 50^\circ C$
Cooling glass/pc (t=1 mm) during rubberforming

$T_{Al} = 100^\circ C$
Cooling glas/pc (t=1 mm) during rubberforming

$T_A = 125^\circ C$
Cooling glass/pc (t=1 mm) during rubberforming

$T_A = 150^\circ C$
Cooling glass/pc (t=1 mm) during rubberforming

$T_A = 175 \degree C$
cooling an 8-layer C/PEI laminate during rubber forming

different Al-mold temperatures

Tₚₑᵢ

TIME [s]

0 1 2 3 4

TEMP [°C]

50°C 100°C 125°C 150°C 175°C 200°C

Fig. 3.7.