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Experiments and Simulation

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Abstract

Due to excellent properties and relatively low material and manufacturing costs, the use of fibre reinforced polymer composites have increased during the last decades. One method that is suitable for large scale productions of e.g. lightweight vehicle components is compression moulding of sheet moulding compound (SMC). Although the technique has been considerably improved since it first was introduced, some further improvements need to be done. The main reason why it has not come in wider use in the vehicle industry is unsatisfactory conditions of the surface finish of parts manufactured due to voids. In this work, experiments and numerical simulations has been performed in order to increase the knowledge of the flow behaviour during the compression moulding process and how the flow affect the quality of the finished product. A process parameter experiment of the compression moulding phase, carried out with a design of experiment approach, was performed in order to investigate the effect of vacuum assistance, mould temperature and ram velocity on the void transport and flow behaviour for SMC. The relative amount of voids has been quantified with a high voltage insulation test and the flow behaviour has been quantified with image analysis of samples moulded with coloured SMC. In conclusion, the setting of high vacuum, low ram velocity and low mould temperature creates a homogeneous flow and minimises the amount of voids. In order to further increase the understanding of void removal during compression moulding, a model experiment was performed where a non-Newtonian fluid (grease) with added bubbles was compressed between two plates whereas the motion of the bubbles were tracked and evaluated using Particle Image Velocimetry. The bubble motion was furthermore analytically modelled and coupled to the experimental results. The experiments reveal an increase in bubble speed compared to the surrounding grease during the compression of the plates. During the latter stage of the compression, the particles change form from initially being approximately spherical, to have the characteristic form of a falling raindrop. The change in form coincides with the increase in speed of the bubble. The developed analytical model supports the shown development in the experiments. A full general solution comprising an arbitrary value of the Power Law exponent, for the velocity fraction coefficient representing the relative bubble speed, is however not covered at the present stage. Finally, the commercial software Ansys CFX were used to perform computational fluid dynamics (CFD) modelling of the flow during compression moulding with a two different multiphase models. The first model treats the flow of SMC as purely extensional and dependent on temperature, fibre volume fraction and strain rate. While the other one sees the flow as mainly extensional but also with thin shear layers near the surfaces of the moulding tool. Where the viscosity, in addition to temperature, fibre volume fraction and strain rate, also is dependant on shear strain rate. Of the two models, the latter seems to be more robust in modelling the pressure during moulding.
PREFACE

This thesis was produced at the division of fluid mechanics at Luleå University of Technology, Sweden, as a part of the FYS-project which was financially supported by VINNOVA and is therefore gratefully acknowledged. Compression moulding experiments have been performed at Swerea SICOMP AB, Piteå, Sweden.

I have many to thank who have supported me in different ways during this project for which I am grateful, to mention everyone would be a too long list. However, there are some people that I especially would like to thank.

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The printing firm of the University for their charitable approach of my late submission...

I would like to thank my beloved Emma and our son Melvin for your encouraging help, especially the last few weeks!
PUBLICATIONS


Paper A
Design of experiment study of compression moulding of SMC

N. E. J. Olsson*1, T. S. Lundström1 and K. Olofsson2

The effect of vacuum assistance, mould temperature and ram velocity on the void transport and flow behaviour for sheet moulding compound (SMC) have been investigated with a design of experiment approach of the compression moulding phase. The relative amount of voids has been quantified with a high voltage insulation test and the flow behaviour has been quantified with image analysis of samples moulded with coloured SMC. In conclusion, the setting of high vacuum, low ram velocity and low mould temperature creates a homogeneous flow and minimises the amount of voids.

Keywords: SMC, Compression moulding, Vacuum, Flow, Void transport

Introduction

Owing to excellent properties and relatively low material and manufacturing costs, the use of fibre reinforced polymer composites have increased during the latest decades. One of the cost effective manufacturing methods suitable for large scale productions, in e.g. the vehicle industry, is compression moulding of sheet moulding compound (SMC). This manufacturing method can be briefly explained as a process with two major steps, manufacturing and compression moulding of SMC prepreg sheets. In the preparation step, a high concentration (15–35 wt-%) of chopped fibre strands is randomly spread in the plane between two layers of a paste that generally consists of three mixed ingredients: thermosetting resins, filler and additives. For enhanced impregnation, the sheets are then compounded. This is followed by a maturation phase, where the sheets are stored for some time whose length depends on composition of the paste. During the storage, the prepreg is chemically thickened, making the sheets more workable, and the dramatic increase in viscosity that results also aids transport of fibres into extremities of the geometry during compression. In the compression stage, the sheets are cut into pieces and placed inside a heated mould tool at which compression moulding is applied. During compression moulding, SMC undergo a very complex process with a first lowering of the viscoelastic properties initiated by heat transfer from the moulding tool. This is followed by solidification which occurs when the molecules start to grow in size by a cross-linking process, and a viscoelastic solid is finally formed through polymerisation.

Although inventions such as low profile additives and vacuum assisted mouldings have improved the quality for SMC products considerably since the technique was introduced in the 1950s, some further improvements need to be carried out. One issue that still concerns the industry is voids in the material and at the surface. Voids located at the surface may cause defects such as pinholes and blowouts which require rework or rejection for a large number of parts which inhibit the success for SMC in the vehicle industry where it is of great importance to have a surface appearance of high quality. Moreover, internal voids are also undesirable, especially in the high voltage industry since they decrease the electrical insulation properties.

During manufacturing, air may be entrapped in the prepreg manufacturing phase as voids in the paste and further by poor fibre impregnation. Moreover, voids may also originate from the compression moulding phase, due to air between the sheets in stacks, air entrapments due to complex flow and also boiling of styrene. Different methods have been developed to reduce the amount of voids in the prepreg sheets, in order to oblige reduce the amount of voids in manufactured products. However, results by Yamada et al. show that the void content in prepregs has to be very high to give an increased number of surface voids. This behaviour has also been confirmed by an initial experiment, carried out by Nilsson at Swerea SICOMP AB, where the effect of a vacuum treatment method for prepregs (Fig. 1) was investigated. The experiment revealed that the amount of voids in the prepregs had no significant effect on the amount of voids after compression moulding even though this method decreased the initial void content in the prepreg to approximately one-third of the void content for the regular industrially manufactured prepreg. Hence, it is important to gain knowledge of the flow behaviour during compression moulding and thereby diminish the amount of voids in SMC products and also to maintain flow control in order to achieve in-plane isotropic mechanical properties since the fibres align in the flow direction.

Owing to the anisotropic non-Newtonian behaviour of SMC, it is difficult to establish a fully accurate
model for the complex flow.\textsuperscript{11} Two practical methods that can be utilised to study the flow during compression moulding are capturing the flow front with a video and to utilising prepreg layers of various colours in order to study the final flow pattern. Odenberger \textit{et al.}\textsuperscript{5} preformed experimental flow front visualisations of the compression moulding process and observed three phases during mould flow:

(i) squish: a squish appears closest to the tool surfaces since a lowering of the temperature dependant viscosity is initiated there by the temperature gradient from the warm tool surface. Hence, the bulk material has a higher viscosity and has a higher resistance to flow

(ii) flow: a stable plug flow is formed

(iii) boiling: bubbles were observed at the low pressure region at the flow front. Based on chemical analysis, the gas was assumed to originate from boiling styrene.

Costigan \textit{et al.}\textsuperscript{14} also observed the squish effect which further became less distinct at higher ram velocities where the flow became more Newtonian and the charge layers became more disrupted as a consequence of increased shear extensional flow while using both of the abovementioned methods. It has also been observed that a higher ram velocity increases the flow of inner layers into deep ribs.\textsuperscript{15} The complexity of the flow is further increased by curing, which, for example, have been shown by Rabinovich \textit{et al.},\textsuperscript{18} where a spiral flow tool, allowing long flows, were used to characterise SMC. The test showed that higher mould temperatures decreased the flow length for SMC. This is because the chemical cross-linking is accelerated by the elevated temperature and thereby solidifies earlier. Le \textit{et al.}\textsuperscript{17} used X-ray phase contrast microtomography to analyse the fibre structure and void content in SMC pre- and post-moulding. A core zone was observed for the moulded plates, sandwiched between upper and lower skins. The skins contained less fibre bundles and porosity than the core zone, and moreover, the fibre bundles were severely broken up in the skins due to an effect of high shearing. Furthermore, the porosity was also found to be a decreasing function of the average moulding pressure.

Other than a high pressure, Yamada \textit{et al.}\textsuperscript{6} observed that vacuum assisted mouldings significantly reduce the amount of surface voids and that it is better to use a low mould temperature. In this paper, it will be studied how the flow behaviour during compression is affected by different settings of the vacuum level, ram velocity and mould temperature. Furthermore, the void contents will also be indirectly measured with an electrical insulation test in order to study how the process parameters and the flow behaviour affect the final void content after compression. All experiments are carried out with a design of experiment methodology.

**Experimental methods and equipment**

Two experimental series have been performed, one with different coloured SMC sheets to visualise the flow and one with sheets of uniform colour where the manufactured plates were analysed as to electrical insulation and thus indirectly void content. In the first series, 16 plates were manufactured and in the second series, 32 were manufactured. All compression moulding experiments have been carried out with a circular mould of diameter 0\,\texttimes\,32\,m with vacuum assistance capability, mounted in a modified Fjellman 310 ton press with parallel closure control. The SMC prepregs used were professionally manufactured by Reichhold AS and Polytec Composites Sweden AB where the coloured SMC were manufactured by adding different pigments to a mutual recipe. Hence, green, white and black prepregs were obtained with equal rheological properties, similar to those of the non-coloured SMC5412. Circular prepreg sheets were made by hand with a circular cutting tool with a diameter of 0\,\texttimes\,15\,m, which was placed in air sealed containers until moulding in order to prevent dry-out. The charges were prepared by randomly picking sheets from the containers. By exchanging sheets, the charges could obtain approximately equal weight. For the mouldings with coloured SMC, charges containing two green, two white and three black randomly picked layers were centrally placed by hand in the mould, as shown in Fig. 2, where aiming marks were used to aid fast and accurate displacement in the centre of the moulding tool. This was carried out in order to minimise possible pregelation effects and obtain equal flow conditions since the placement of the charge can affect the flow.\textsuperscript{7,18} The uniformed coloured charges were stacked in a similar fashion. For vacuum assisted mouldings, a switch was installed to turn on the vacuum so that it had reached full capacity just before the upper mould surface touched the charge.

![Illustration of vacuum treatment](image1)

**Fig. 1 Illustration of vacuum treatment**

2 stacks of coloured SMC with 32 different trials were manufactured. All compression moulding experiments were carried out with a circular mould of diameter 0\,\texttimes\,32\,m with vacuum assistance capability, mounted in a modified Fjellman 310 ton press with parallel closure control. The SMC prepregs used were professionally manufactured by Reichhold AS and Polytec Composites Sweden AB where the coloured SMC were manufactured by adding different pigments to a mutual recipe. Hence, green, white and black prepregs were obtained with equal rheological properties, similar to those of the non-coloured SMC5412. Circular prepreg sheets were made by hand with a circular cutting tool with a diameter of 0\,\texttimes\,15\,m, which was placed in air sealed containers until moulding in order to prevent dry-out. The charges were prepared by randomly picking sheets from the containers. By exchanging sheets, the charges could obtain approximately equal weight. For the mouldings with coloured SMC, charges containing two green, two white and three black randomly picked layers were centrally placed by hand in the mould, as shown in Fig. 2, where aiming marks were used to aid fast and accurate displacement in the centre of the moulding tool. This was carried out in order to minimise possible pregelation effects and obtain equal flow conditions since the placement of the charge can affect the flow.\textsuperscript{7,18} The uniformed coloured charges were stacked in a similar fashion. For vacuum assisted mouldings, a switch was installed to turn on the vacuum so that it had reached full capacity just before the upper mould surface touched the charge.

![Stacking order for coloured SMC](image2)

**Fig. 2 a stacking order for coloured SMC and b charge pattern for coloured stacks**

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![Stacking order for coloured SMC](image2)

**Fig. 2 a stacking order for coloured SMC and b charge pattern for coloured stacks**
The software Minitab was used to facilitate the analysis of the experiments carried out with a design of experiment methodology. Since the mould temperature is a hard to change factor, split plot designs have been used. Ninety-five per cent confidence intervals were utilised to determine important effects with one exception as mentioned later in the paper. The models were set up by first including all possible interaction up to the fourth order and then by removing the effect of highest order with the smallest significance one at a time. Moreover, when non-significant effects were removed from the model, the hierarchy principle was used, which means that non-significant main effects are remained in the model if they are involved in significant higher order interactions.

In order to perform statistical analysis, the sample needs to be quantified. Two types of measurements have been carried out, image analysis to quantify the flow and a high voltage insulation test to quantify the relative amount of voids. The image analysis was carried out with the software ImageJ. Images of the surfaces of the coloured plates were recorded with a regular digital system camera, while cross-sections were scanned with a regular desktop scanner. For the coloured plates created with centred circular charge, the area fractions of green and white were measured on the top (Fig. 3), as well as the width of the green and white inner layers. In Fig. 4, examples of cross-sections of the outermost 0.05 m can be seen.

Since the amount of voids highly affects the electrical insulation properties, a high voltage insulation test (IEC 60243-1) was performed at ABB AB Plast, Piteå, Sweden, in order to give an indication of the local void content. Because the amount of voids at the surface is roughly proportional to the void content inside a moulded plate, this test can also be interpreted as a quantification of surface quality. The test is carried out in an oil bath where the samples are squeezed between two electrodes, over which a voltage is applied. The voltage is increased by ~2.2 kV each second until a disruptive discharge. The resulting voltage gradient is then computed by dividing the discharge voltage with the local plate thickness. This measurement was performed at multiple locations over the diagonal of the plates, and the results were thereafter submitted into either one inner group, containing all results from discharges within the diameter of the original charge (0–15 m), or an outer group containing all results from outside the original charge diameter.

Experiments and results
The altered parameters for the experiments were the vacuum level, the ram velocity and the mould (top mould half) temperature (Table 1). For the flow visualisation experiments with coloured SMC, two replicates were made for each setting giving a total of 16 samples, while the mouldings for void content measurements were made with four replicates of each process setting giving a total of 32 samples. Moreover, since the void contents results were divided into two groups, an extra factor called section was added to the analysis where 1 represented the inner section with diameter 0–15 m, and 1 represented the surrounding area that the SMC is forced into during compression.

Flow visualisation with coloured SMC
A first observation from the compressions with coloured SMC was that SMC from the bottom green layer had moved to the top side, while the SMC in the middle green layer was sealed within the other layers and did not penetrate to the top. A high amount of green on the top side of the moulded plate can therefore be interpreted as a larger squish effect. Hence, to start

Table 1 Values of design parameters: temperature indicated is that at top surface; temperature at bottom surface was 3–4 °C higher to prevent tool damage

<table>
<thead>
<tr>
<th>Vacuum, %</th>
<th>Velocity, Mm s⁻¹</th>
<th>Temperature, °C</th>
<th>Pressure, kN</th>
</tr>
</thead>
<tbody>
<tr>
<td>-1</td>
<td>1</td>
<td>-1</td>
<td>1</td>
</tr>
<tr>
<td>75</td>
<td>0</td>
<td>2.5</td>
<td>144</td>
</tr>
</tbody>
</table>

![Image of samples moulded with coloured SMC](image-url)
with, the area fraction of green SMC was measured on the topside of the plates. The analysis of the response reveals that applying vacuum always reduces the effect of the squish but especially for the low ram velocity and for the low temperatures (Figs. 5 and 6). In addition, without vacuum assistance, the squish can be reduced by raising the ram velocity or the temperature.

Analysis of the area fraction of white for the top exposed a significant interaction between vacuum and ram velocity (Fig. 7). Here, a higher area fraction can be interpreted as a higher ability to flow. This area fraction increases with a higher ram velocity for mouldings without vacuum assistance. For vacuum assisted mouldings, the highest area fraction of white is obtained with slow ram velocity and is only slightly lowered with high ram velocity.

According to the analysis of variance, a complex three-way interaction between all factors exists for the width of the fifth green layer (Fig. 8). A larger width means a higher ability to flow and can be interpreted as a more homogeneous flow with less squish effect. By inspection, vacuum level seems to have the largest effect in the interaction, since the width clearly increases when vacuum is applied. The result is also dependent on the ram velocity and mould temperature. For vacuum assisted mouldings at high ram velocities, the width seems to be facilitated by a higher temperature. However, with vacuum assisted moulding at low ram velocities, the width decreases with higher temperatures. Without vacuum assistance, the smallest width is achieved when both the temperature and ram velocity are low. The largest width lengthening without vacuum assistance is at high ram velocity and a low mould temperature.

For the third layer, white, no effect was found using 95% confidence intervals. However, using 93% confidence intervals, it can be shown that vacuum assistance increases the width at low ram velocities. Without vacuum assistance, a clear increase of width can be carried out by increasing the ram velocity (Fig. 9) which
Void transport during compression moulding

The high voltage insulation test clearly showed that the outer zone which consists of the SMC that has flown the longest distance contained less voids than the inner zone. After removing three outliers from the 64 results, a main effect from the result was better insulating properties in the outer zone (section 1) than in the inner zone (section −1) as indicated with higher values in Fig. 10. The values in this figure are mean results of the discharge gradient (kV mm⁻¹) for each setting. Then by scrutinising the results in the inner and outer zones individually, it was found that interaction between temperature and vacuum as well as temperature and velocity behaved differently in respective zone. For the inner zone (section −1), it was found that the set-ups with high vacuum and high temperature dramatically decreased the electrical insulating properties (Fig. 10). In the outer zone (section 1), applied vacuum increased the insulation properties for both temperature levels (Fig. 10). High temperature combined with low velocity was found to decrease the insulation properties, while the combination of high temperature and low velocity increased the insulation properties in the inner zone (section −1). In contrary, the effect of high temperature and low velocity increases the insulation properties in the outer zone (section 1). The overall interaction effect between vacuum, velocity and temperature over the whole plates (sections −1 and 1) shows two process parameter settings that dramatically increase the insulation properties. For low temperatures, the insulation properties can be dramatically increased either by raising the vacuum to the higher level while keeping the velocity low, or by raising the velocity to the higher level while keeping the vacuum level low (Fig. 10). The set-up that seems to be the superior in order to increase the electrical insulation properties and hence decrease the void contents in both sections at the same time is the combination of high vacuum, low ram velocity and low mould temperature.

Discussion

The analysis presented indicates that an applied vacuum influences the global flow. The mechanism behind this is not fully understood, but one idea is that a pressure build-up by confined air inside the closed moulding tool is prevented. By assuming that higher electrical insulation properties can be correlated to a lower amount of internal voids and combining the results of the void transport test and the results from coloured SMC moulding, some conclusions can be made. First, flow of the SMC facilitates void removal during compression. Second, usage of vacuum assistance (75% vacuum) results in a less amount of voids in the outer zone. According to the test with coloured SMC, these settings cause a more uniform flow. Hence, a uniform flow should be used in order to decrease the amount of internal voids for freely flowing SMC.

According to the high voltage insulation test, the amount of voids seems to increase in the inner zone with the combination of high vacuum and high temperature. The reason for this may be due to boiling of styrene since the boiling point is lowered by the vacuum. Given that the curing is initiated at the outer border where the material has had an overall higher temperature due to the flow, the material may have started to cure before the boiling starts, explaining why this parameter combination does not increase the amount of voids in the outer zone to any large extent.

Combining the results from the coloured SMC experiment and the results from the large void transport experiment, it seems like the setting of high vacuum, low ram velocity and low mould temperature creates a homogeneous flow and minimises the amount of voids in the whole plate.

Finally, SMC have previously been described to have a strong shear thinning behaviour. This may be the reason why compression moulding without vacuum assistance seems to be more homogeneous with a high ram velocity (Figs. 5 and 7–9), and also significantly increases the electrical insulation properties in the outer zone (section 1) when low levels of vacuum and temperature is applied (Fig. 10).

Conclusions

The design of experiment approach to compression moulding of SMC reveals that the three tested factors, vacuum, ram velocity and mould temperature, all affect the global mould flow behaviour. The compression
moulding experiments with coloured SMC shows that high ram velocity tends to create a more uniform flow. Moreover, they also show that utilising vacuum assistance during moulding, a more uniform flow can be obtained also at low ram velocities. By increasing the mould temperatures, the inner layers are allowed to stretch further, but this also increase the squish effect. The high voltage insulation test indicates that the amount of voids is decreased by the flow. A minimisation of the amount of voids in both zones seems to be achieved with the setting of high vacuum (75%), low ram velocity (2.5 mm s\(^{-1}\)) and low mould temperature (144°C). From the experiment with coloured SMC, these settings seem to homogenise the flow and minimise the squish effect. Hence, a homogeneous flow is preferred for creating SMC products with a small amount of voids.

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Paper B
TRANSPORT OF BUBBLES DURING COMPRESSION IN A NON-NEWTONIAN FLUID

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ABSTRACT

Bubble transport is of importance in many applications for non-Newtonian fluids, such as in composite materials manufacturing where residual bubbles may impair electrical properties, surface appearance and the mechanical properties of the manufactured products. In this study, a model experiment is performed where a non-Newtonian fluid (grease) with bubbles is compressed between two plates whereas the motion of the bubbles is tracked and evaluated using Particle Image Velocimetry. The bubble motion is furthermore analytically modeled and coupled to the experimental results.

NOMENCLATURE

- \( \tau \) viscous stress
- \( \tau_0 \) yield stress
- \( u \) velocity in axial/main flow direction
- \( z \) z-coordinate
- \( r \) radial coordinate
- \( K \) grease consistency
- \( n \) Power-Law exponent
- \( h \) distance between plates
- \( U \) relative velocity between plates
- \( m \) friction factor
- \( \omega \) average coefficient of wall friction
- \( R \) plate radius
- \( U_b \) bubble speed
- \( W \) fractional velocity correction
- \( k_b \) bubble mean curvature
- \( V \) volume flux
- \( r_c \) sphere radius in Bretherton model
- \( r_s \) sphere radius in present squeeze flow geometry

1. INTRODUCTION

In many industrial applications such as classical lubrication of engineering bearings, compression moulding of composite materials and powder granulation, squeeze flow of a non-Newtonian fluid is highly relevant. Throughout the years there has been done a lot of research on squeeze flow, both for non-Newtonian fluids as well as for different geometries such as between plates and rigid spheres (see e.g. Sherwood [1]).

In this paper the objective is to investigate the bubble motion in a non-Newtonian (grease) squeeze flow. See Figure 1 for an overview of the experimental setup used in this study. The key objective is to examine whether (and if so, why) the bubbles move faster than the surrounding fluid – a question which, in terms of flow in capillary tubes, was first addressed by Fairbrother and Stubbs [3]. They showed that due to the bubble form - not behaving as a closely fitting piston, the bubble move somewhat faster than the surrounding fluid. In general, studies of droplet formation, bubble motion and two-phase flow in capillary tubes have been performed by numerous authors, see e.g. [4] and references therein. Bubble motion in squeeze flow has not been studied to the same extent. In the paper by Sherwood [8], bubble formation in dry foam is treated. The coupling of bubble motion in a non-Newtonian fluid with squeeze flow is however, to the authors’ knowledge much an untrodden path in this area of science.

A cornerstone work in the field of bubble motion is the analysis by Bretherton [9], where he derives an analytical expression for the thickness of the thin transition layer...
between the bubble and the wall in a pressure driven pipe flow. In this work, the Bretherton solving methodology is applied to a Herschel-Bulkley fluid in squeeze flow geometry. The analytical results covering the bubble motion are coupled to results from experiments for different fluids. The bubble motion is experimentally analyzed using Particle Image Velocimetry (PIV).

2. EXPERIMENTAL

Bubbles were observed by visual inspection in initial squeeze flow experiments to move significantly faster than the surrounding non-Newtonian medium. Therefore, the experimental setup was further evolved to render possible flow field measurements with Particle Image Velocimetry (PIV) methodology. During experimenting, two significant behaviours were observed during squeeze flow which will be presented in the results. Firstly, larger bubbles tend to translate faster through the medium than small bubbles. Secondly, bubbles with a higher velocity than the surrounding medium stretches from a circular-like shape into the shape of a falling rain drop, which seems to increase the velocity further.

2.1 SETUP AND PROCEDURE

Squeeze flow experiments have been performed with an experimental press which render possible image acquisitions of the flow and associated bubble transport, see Figure 1.

The particle doped grease was placed on the lower plate of the mould to a height of 6.2 mm with aid of a cylinder, with a diameter of 44 mm. The bubbles were then manually added with a syringe and the lower plate was forced to move towards the upper one with the aid of a mechanical screwing device generating a squeezing flow. As recording device a Dantec NanoSense MkIII camera was used along with a NIKON 60mm f/2.8D lens, where the image acquisition was controlled by the Motion Studio 2.06.05 software from IDT to 1280x1024 pixel, 8bit and 100 frames per second. The post-processing was performed with the software Dantec Dynamic Studio 2.3 which offered an adaptive cross correlation method, giving more reliable flow fields due to refinement steps through larger sub-images and high accuracy subpixel refinement. In the present case, the final interrogation area size was 8x8 pixel, numerically compiled through 4 refinements steps with 50% overlap from an initial area size of 128x128 pixel.

2.2 RESULTS

Experimental results from one designated squeeze flow experiment can be seen in Figure 3-Figure 6, where four different stages are presented in order to demonstrate how the encapsulated bubbles have been affected by the squeeze flow as a function of time. In each figure, three images are presented: the uppermost shows the original recorded image, the middle shows the same image with velocity vectors from the PIV measurements, and in the bottom one a close-up view of the PIV result is presented. Length scales in [mm] can be
seen to the left for respectively magnification and to the right, a colour scale for the vector velocities in [m/s] is placed.

Figure 3: Original, PIV result, and close-up view for stage 1.
Figure 4: Original, PIV result, and close-up view for stage 2.

Figure 5: Original, PIV result, and close-up view for stage 3.
A visual inspection of the original images in Figure 3-Figure 6 reveals that a large portion of the encapsulated bubbles in the outer regions are removed during pressing, especially the large bubbles. It can also be seen by inspection of the PIV results, that there is a correlation between areas of increased velocity and position of the bubbles. Comparisons between the original and the PIV image reveal that bubbles having a high velocity relative to the grease, tend to elongate and form a tail which narrows and gives the bubble the shape of a falling raindrop, which further increase the velocity. As a consequence, the grease starts to move inwards towards the bubble tail as seen in the close-up views representing the event history for three bubbles. In the first stage (see Figure 3), the largest bubble, which is also closest to the flow front, has the highest velocity. This bubble seems to affect the bubble above, since the velocity vectors points towards the larger bubble instead of in the radial direction. In the second stage (see Figure 4), the large bubble is elongated and has approached the grease flow front. In the third stage (see Figure 5), the large bubble from the previous stages has escaped the grease which, however, is still affected by its earlier presence since the grease flows towards the area previously occupied by the bubble. Moreover, the other two bubbles have started to elongate, in particular the uppermost which has gained velocity relative to the grease and made a radical approach towards the flow front. As a consequence, grease flows inwards towards the tail as it narrows. In the fourth stage (see Figure 6), this bubble has almost been removed from the grease and obtained an increased velocity. Again it can be seen how the grease move inwards towards the tail of the bubble as it narrows.

3. ANALYTICAL MODELING

In this section an analytical model for the bubble motion in the grease is sought.

3.1 Grease flow

Grease is due to its composition a complex fluid. It consists of a thickener system (usually a metal-soap), base oil and additives. The flow characteristics are dominated by its yield stress behavior, where a critical stress has to be applied in order for the fluid to flow. In a recent study treating grease flow in a rectangular channel, Westerberg et al. [11] showed that the Herschel-Bulkley rheology model can describe the flow well for the greases used in this study. This model is written as

$$\tau = \tau_0 + K \left( \frac{\dot{e} \alpha}{\xi} \right)^n,$$  \hspace{1cm} (1)
where $\tau_0$ is the yield stress, $K$ the consistency, and $n$ the Power Law exponent. $z$ is the coordinate perpendicular to the flow direction. Data characteristics for the NLGI2-grease used in this study are presented in Table 1. The NLGI grade indicates the thickness of the grease. As comparison in Table 1, a grease with NLGI grade 1, i.e. a less thick grease, is also presented. The characteristic shear thinning effect – typical for grease, is represented by an $n$ value < 1.

Table 1: Values for the parameters in the Herschel-Bulkley rheology model (Eq. 1). The NLGI2-grease is used in the present study. Values obtained from plate-plate rheometer. From Westerberg et al. [11].

<table>
<thead>
<tr>
<th>Grease</th>
<th>$\tau_0$ [Pa]</th>
<th>$K$ [Pa s]$^{1/n}$</th>
<th>$n$</th>
</tr>
</thead>
<tbody>
<tr>
<td>NLGI1</td>
<td>189</td>
<td>4.1</td>
<td>0.797</td>
</tr>
<tr>
<td>NLGI2</td>
<td>650</td>
<td>20.6</td>
<td>0.605</td>
</tr>
</tbody>
</table>

In Westerberg et al. [11] it is shown that there is a very thin shear/slip layer close to the boundaries, where the velocity gradient is very high. Instead of continuously approaching zero velocity at the boundaries, there is a discontinuous behaviour for the velocity in the slip layer. Here we investigate and discuss the influence of this layer on the bubble motion and the original theory by Fairbrother and Stubbs [3] that claims the shape of the bubble as a reason for its different speed; it does not behave like a closely fitting piston in the tube. In contradiction to Newtonian fluids having a parabolic velocity profile, grease take up a plug flow profile where the velocity is constant through a majority of the channel (see Figure 7). The effect of this property is also investigated for its impact on the bubble motion.

Figure 7: Velocity profile for the actual greases across a rectangular channel of width 1.5 mm. Dimensionless velocity $u$ scaled with maximum velocity in the channel. +:NLGI1, #:NLGI2. The third grease (dotted marker) having a Newtonian profile, is a NLGI00 grease. From Westerberg et al. [11].

3.2 Bubble motion analysis

3.2.1 The Bretherton model

Bubble motion in capillary tubes has been studied since the 1930’s when Fairbrother and Stubbs [3] discovered that the velocity of bubbles is slightly higher than the velocity of the surrounding fluid. In his benchmark work, Bretherton [9] showed that the bubble speed $U_b$ exceeds the average speed of the fluid in the tube by an amount $U_b W$, where

$$W = 1.29 \left( \frac{\mu U_b}{\sigma} \right)^{1/3} = 1.29 \left( \frac{C_b}{C_a} \right)^{1/3}, \quad C_a = \frac{\mu U}{\sigma} \rightarrow 0. \quad (2)$$

This result showed to underpredict the thickness of the film formed between the bubble and the tube wall, with a discrepancy being greatest at lowest speeds (Schwartz, Princen and Kiss [12]). However, though the Bretherton theory does not fit experiments in capillary tubes for low speeds, it is a good basis for investigating the bubble motion in the present study.

Figure 8 show an overview of a bubble being located between the plates (a) and the situation close to the boundary of one of the plates (b). Using the analogous analysis and notation as Bretherton [9], the bubble geometry and transition layer structure can be directly transferred to the plate-plate geometry following the experiments presented in the previous section. In traditional squeeze flow the two plates move towards each other with identical speed - meaning the origin is fixed in space and located in the middle of the gap between the plates. In the present approach however where the upper plate is fixed in space, the origin is not fixed in space, but moves with the same speed as the lower plate in order to have a fixed coordinate system with respect to the plates. Hence, in terms of this new coordinate system the analysis situation is analogous to traditional squeeze flow. This is most significant when considering the velocity profile in the whole gap between the plates (see Appendix A). For the analysis of the transition layer between the bubble and the plate (see §3.2.2) however, the introduced (local) coordinates are with respect to the plate only. The reason for choosing the upper plate to be fixed in space, is to better and more easily keep focus on the flow when recording it with the camera.
3.2.2 Transition layer analysis

In this section the fractional velocity correction $W$ (Eq. 2) for a Herschel-Bulkley fluid is derived. The analysis follows the methodology by Bretherton [9], where the thin transition layer with thickness $b$ (see Figure 8b) is the subject of investigation. For the transition layer analysis, Bretherton uses the Cartesian coordinates $x, y$. Here the geometry is cylindrical, and a coordinate transformation is necessary. It is a straightforward approach since no scale factors are introduced when going from $(x,y)$ to $(r,z)$, where $r$ is the radial co-ordinate and $z$ is normal to the plane.

Considering the bubble/fluid interface (see Figure 8b), zero tangential stress results in

$$\frac{\partial u_z}{\partial z} = 0, \quad z = z_1. \tag{3}$$

Normal stress condition at the interface is reduced to

$$p_1 + \sigma \cdot \kappa_z = p_1 + \sigma \frac{d^2 z_1}{dr^2} = 0, \tag{4}$$

applying the lubrication approximation where $\kappa_z$ is the mean curvature of the bubble (where the curvature of the plate-plate configuration is zero). The Navier-Stokes equation in cylindrical coordinates for a Herschel-Bulkley fluid, and for small Reynolds number, is written as

$$\frac{dp_1}{dr} = \frac{\partial}{\partial z} \left[ \tau_0 + K \left( \frac{\partial u_z}{\partial z} \right)^n \right]. \tag{5}$$

The boundary conditions are determined by the no slip condition at the wall

$$u_z = 0, \quad z = 0 \tag{6}$$

together with zero tangential stress at the interface (Eq. 3). The volume flux of the fluid is

$$V = \int_{0}^{z} u_z dz, \tag{7}$$

where the velocity $u_z$ is obtained by integrating Eq. 5, resulting in

$$K^{1/n} u_z(r,z) = n \frac{1}{n+1} \int \left[ \frac{dp_1}{dr} (z-z_i) \right]^{1/(n+1)} + C_2 \tag{8}$$

using the zero tangential stress B.C. (Eq. 3). Applying the no slip B.C. (Eq. 6) may be written as

$$K^{1/n} u_z(r,z) = \frac{n}{n+1} \left[ \frac{dp_1}{dr} (z-z_i) \right]^{1/(n+1)} - \left[ \frac{dp_1}{dr} z_i \right]^{1/(n+1)} \tag{9}$$

The resulting volume flux becomes

$$V = \int_{0}^{z} u_z dz =$$

$$= \frac{1}{K^{1/n}} \frac{1}{n+1} \left[ \frac{dp_1}{dr} \right]^{1/(n+1)} - \left[ \frac{dp_1}{dr} z_i \right]^{1/(n+1)} \tag{10}$$

Differentiating the normal stress condition at the interface (Eq. 4) yields the following expression

$$\left( \frac{dp_1}{dr} \right)^{1/n} = -\sigma \frac{d^3 z_i}{dr^3}^{1/n} \tag{11}$$

which together with continuity...
\[ V = U_j(z_i - c) \]  

(12)

results in

\[ \frac{d^2 z_i}{dr^2} = -U_j^* (z_i - c)^{n+1} \left( \frac{n+1}{n} \right) K z_i^{-n}. \]  

(13)

Here \( c \) and \( \zeta \) are constants where \( c \) is to be determined below, and \( \zeta \) equals

\[ \zeta_i = z_i^{1+n+2} \left\{ -\frac{n}{2n+1} (-1)^{1+n+2} - (-1)^{1+n+1} \right\}. \]  

(14)

\( c \) is determined from the condition

\[ \frac{d^2 z_i}{dr^2} = 0, \quad z_i = b, \]  

(15)

which leads to

\[ c = b. \]  

(16)

Eq. 13 can then be written as

\[ \frac{d^2 z_i}{dr^2} = -U_j^* (z_i - b)^{n+1} \left( \frac{n+1}{n} \right) K z_i^{-2n-2}, \]  

(17)

where

\[ \xi_i = -\frac{n}{2n+1} (-1)^{1+n+2} - (-1)^{1+n+1}. \]  

(18)

Introducing

\[ 3\mu^* = \left( \frac{n+1}{n} \right)^{n} K \xi_i^{-n}, \]  

(19)

Eq. 17 then writes as

\[ \frac{d^2 z_i}{dr^2} = 3\mu^* U_b (z_i - b)^{n} \left( \frac{3\mu^* U_b}{\sigma} \right)^{1/3} \left( \frac{3\mu^* U_b}{\sigma} \right)^{-1/3} \]  

(20)

This equation can be written on universal form by the following variable substitutions:

\[ z_i = b \eta, \quad r = b \left( \frac{3\mu^* U_b}{\sigma} \right)^{1/3} \xi \]  

(21)

leading to

\[ \frac{d^3 \eta}{d\xi^3} = \frac{(\eta - 1)^2}{\eta^{1/3}}. \]  

(22)

For sufficiently small values of \((3\mu^* U_b/b)^{1/3}\) there are regions in which

\[ \eta = \frac{z_i}{b} \gg 1, \]  

(23)

resulting in (Eq. 22)

\[ \frac{d^3 \eta}{d\xi^3} = 0, \]  

(24)

with the solution

\[ \eta = \frac{1}{2} A_1 \xi^2 + A_2 \xi + A_3. \]  

(25)

Returning to the original coordinates (Eq. 20) this solution is written as

\[ z_i = \frac{1}{2} A_1 \xi^2 + A_2 \xi + A_3 + A_4 b. \]  

(26)

In this region the mean curvature becomes

\[ K_1 = \frac{d^3 z_i}{d\xi^3} = \left( \frac{3\mu^* U_b}{\sigma} \right)^{2/3} \frac{A_1}{b^{1/3}} = \frac{1}{r}, \]  

(27)

where \( 1/r \) is the mean curvature of a cylinder with radius \( r \). The surface has constant mean curvature across the plates and can to the first approximation be expressed as a cylinder with radius \( r \). Eq. 26 and Eq. 27 then yields

\[ \frac{1}{r} = \left( \frac{3\mu^* U_b}{\sigma} \right)^{2/3} A_4. \]  

(28)

When \( n \) is different from zero it is not straightforward to proceed from this step of the analysis. In region CD (see Figure 8a), \( z_i \) approximately equals the transition layer.
thickness $b$, leading to a $\eta$-value close to unity. This yields (Eq. 22)

$$\frac{d^3\eta}{dz^3} \approx (\eta - 1)^n,$$

(29)

which is not possible to solve analytically for an arbitrary $n$-values. In this paper the greases have an $n$-value of approximately 0.6 and 0.8 respectively (c.f. Table 1). So, in order to solve for $A_1$ in Eq. 28, a numerical approach is necessary. Actual values of remaining parameters are however not known at the present stage. To proceed the current analysis, the case $n=1$ is considered henceforth. In terms of the Herschel-Bulkley rheology model (Eq. 1), this special (Newtonian) case yields the Bingham rheology model which also accounts for the yields stress behavior, but not for the shear thinning/thickening property. How the shear thinning property for the actual greases influences the physics in this study is discussed in §4.

### 3.2.3 Fractional velocity correction for the plate-plate configuration: Newtonian case ($n=1$)

When $n=1$, Eq. 28 is written as (c.f. Eq. 19)

$$\frac{b}{r_c} = \left(\frac{3KU_c}{\sigma}\right)^{2/3} A_1,$$

(30)

where $A_1=0.643$ following from the solution to

$$\frac{d^3\eta}{dz^3} \approx \eta - 1,$$

(31)

as shown in the analysis by Bretherton [9]. The Bretherton analysis is based on pipe flow geometry (see Figure 9a) with transition layer of thickness $b$ symmetrically near the pipe wall. The fractional velocity correction $W$ as measured by Fairbrother and Stubbs, is the proportion of the cross-sectional area of the pipe occupied by the transition layer [1], i.e.

$$W = \frac{\pi r_c^2 (r_c-b)^2}{\pi r_c^2} = \frac{2h}{r_c} - 1 \approx \frac{2h}{r_c} - 1 = 1.29 \left(\frac{3KU_c}{\sigma}\right)^{2/3}.$$

(32)

Considering a length $l$ of the plate-plate geometry (Figure 9b), the corresponding proportion is

$$W = \frac{bh}{hl} = \frac{2h}{h} \approx \frac{b}{r_c} = 0.643 \left(\frac{3KU_c}{\sigma}\right)^{2/3},$$

(33)

recalling Eq. 30 and that the radius $r_c$ approximately is half the distance between the plates $h$.

From Eq. 18 and 19 it is seen that when $n$ equals unity, the introduced apparent viscosity $\mu'$ equals $K$ which correspond the viscosity $\mu$ of a Newtonian fluid. Hence the result corresponding to $n=1$ is identical to the result by Bretherton [9], except for a factor two resulting from the change of geometry. Worth noticing is that the yield stress does not affect the solution.

![Figure 9: Cross-sectional view of the transition layer thickness for (a) the pipe flow used by Bretherton [9], and (b) the plate-plate configuration used in this study.](image)

### 4. DISCUSSION AND COUPLING TO EXPERIMENTS

Of certain interest and of vital importance, is to estimate the quality of the performed simplification corresponding to $n=1$. As measured in the rheometer, the greases used in this paper are shear thinning ($n<1$). This means that the shear stress decays as the rate of shear is increased - i.e. there is a non-linear relation between the two quantities. For a Newtonian fluid the relation is linear with a shear stress proportional to the shear rate. Furthermore, as shown by Westerberg et al. [11], that for grease it is likely to have a thin shear layer close to the boundaries where the velocity gradient is significantly higher than in the rest of the fluid. Key questions are whether the shear layer and the shear thinning property affect the bubble speed.
From the experimental results presented in §2, it is evident that the bubble speed is higher than for the surrounding grease. The analytical solution yields a velocity fractional correction proportional to the transition layer thickness divided by the mean radius of the bubble. The effective thickness of the layer is found knowing the Capillary number and the curvature radius of the bubble (i.e. approximately the distance between the plates for the squeeze flow case, and the pipe curvature for the pipe flow case). In Bretherton’s experiments [9] a tube with diameter 2 mm was used. In the present study, the distance between the plates at the latter part of the experiment (c.f. Figure 6) where the bubble speed is distinctively higher than the surrounding grease, is of the order of 2 mm. This should be compared to the distance of the order of 4 mm at the initial time of measurement presented in Figure 3, where the bubble speed not differ significantly from the grease speed.

As the bubble is highly compressible it is reasonable to assume that the transition layer thickness \( b \) between the bubble and the plate wall, remains approximately constant in comparison to the curvature of the bubble which becomes significantly smaller when the plate distance is reduced. According to the relation for the velocity fraction coefficient \( W \) in Eq. 33, the value of \( W \) then increases. Explicit values of the surface tension and the bubble curvature are not known at present stage. Hence explicit values of the Capillary number are also not known. However, the general behavior for the bubble speed presented in the analysis is to a large extent verified by the experiments.

From Eq. 18 and 19 it follows that the introduced apparent viscosity differs from the Newtonian value with a factor directly dependent of the \( n \)-value. This factor also affect the solution for \( z \) and \( b \) according to Eq. 21 and 26. For a factor larger than unity, it is not unreasonable to have an increase in the relation \( b/r \), and hence in the bubble speed compared to the surrounding grease.

There are several ways to go in order to develop the present study. To study the influence of the non-Newtonian properties, it is of interest to perform the experiments with grease having both thinner and thicker consistency. Furthermore, in order to enhance the coupling to the analytical model, it is of interest to measure the compression (plate) speed, surface tension, and bubble curvature. The natural step of improvement for the analytical model is to make it include a general value of the Power-Law coefficient \( n \). Also, the change of bubble form and how to include this in the model is a subject of great importance.

5. SUMMARY

In this paper bubble motion during squeeze flow of a non-Newtonian fluid (grease), is investigated using particle Image Velocimetry (PIV) and an analytical model. The experiments reveal an increase in bubble speed compared to the surrounding grease during the compression (squeeze) of the plates. During the latter stage of the compression, the particles change form from initially being approximately spherical, to have the characteristic form of a falling raindrop. The change in form coincides with the increase in speed of the bubble.

The analytical model is developed from the classical Bretherton analysis [9] for bubbles in pipes. It supports the shown development in the experiments. A full general solution comprising an arbitrary value of the Power Law exponent, for the velocity fraction coefficient representing the relative bubble speed, is however not covered at the present stage.

REFERENCES

APPENDIX A

Here solutions for the velocity profile of a Herschel-Bulkley fluid are presented. The velocity development between the plates is not of direct interest in the present approach as it can be treated separately from the flow in the transition layer. However, it is of great interest in general from a fluid mechanical aspect, and the subject for future studies using the μPIV method. In this Appendix a solution for the velocity profile is sought using a methodology analogous to the work by Laun, Rady and Hassager [13]. The governing equations are

\[
\frac{1}{r} \frac{du}{dr} + \frac{du}{dz} = 0 \tag{34}
\]

\[
-\frac{dp}{dr} + \frac{\tau_r}{\varepsilon_z} = 0, \tag{35}
\]

where

\[\tau_r = \tau_0 + K \left( \frac{\partial u}{\partial z} \right)^n. \tag{36}\]

Integrating (Eq. 34) with respect to \(r\) gives

\[u_r = rg(z), \tag{37}\]

where \(g(z)\) is an arbitrary function depending only on the \(z\)-coordinate. (\(u_r\) in Eq. 35 gives

\[2g(z) + \frac{du}{dz} = 0. \tag{38}\]

Integrating Eq. 36 results in

\[z \frac{dp}{dr} = \tau_0 + K \left( \frac{\partial u}{\partial z} \right)^n = \tau_0 + Kr^n g'(z)^n, \tag{39}\]

using the property

\[\frac{\partial u}{\partial z} = 0, z = 0. \tag{40}\]

Integrating Eq. 39 with respect to \(r\) then yields

\[z \cdot p(r) = \tau_0 r + \frac{K}{n+1} r^{n+1} g'(z)^n. \tag{41}\]

As the pressure is not dependent on \(z\), it can be written as an expansion in orders of \(r\) such that

\[p = A + \tau_0 r + Br^{n+1} + ... \tag{42}\]

Where \(A\) and \(B\) are constants. Applying Eq. 42 into Eq. 39 gives after integration with respect to \(z\)

\[\tau_0 (z-1) + (n+1)Br^z z = Kr^n g'(z)^n, \tag{43}\]

which after integration with respect to \(z\) gives

\[K^{\frac{1}{n}} g(z) = \frac{n}{n+1} \left[ \frac{1}{\tau_0 \frac{r^z}{n+1} + (n)B} \right]. \tag{44}\]

\[\left( \frac{\tau_0}{\varepsilon_z} (z-1) + (n+1)Bz \right)^{\frac{n+1}{n}} + C_2, \tag{44}\]

Using the partial slip boundary condition

\[u_r(r, h) = rg(h) = \frac{r}{R} u_s, \tag{45}\]

where \(u_s\) is the slip velocity at the boundary, \(R\) the radius, and \(h\) half the gap distance between the plates, the constant \(C_2\) is determined such that

\[C_2 = K^{\frac{1}{n}} \frac{u_s}{R} - \frac{n}{n+1} \left[ \frac{1}{\tau_0 \frac{r^z}{n+1} + (n+1)B} \right] \]

\[\left( \frac{\tau_0}{\varepsilon_z} (h-1) + (n+1)Bh \right)^{\frac{n+1}{n}}. \tag{46}\]

which completes Eq. 44. Considering the case \(n=1\) and neglecting the yield stress, \(g(z)\) is reduced to Eq. 43 (Power Law fluid) and Eq. 14 (Newtonian fluid) in Laun, Rady and Hassager [13] respectively. Eq. 44 and 46 in Eq. 38 gives after integration

\[11\] Paper No: ITP-09-55
\begin{align*}
u_z &= \frac{2n}{n+1} \left\{ \frac{1}{r^n + (n+1)B} \right\} \cdot \left[ \frac{\tau_0}{r} (R - 1) + (n+1)B \right]^{\frac{1}{n}} \cdot \left( 1 + \frac{\tau_0}{r^n + (n+1)B} \right)^{\frac{2-n}{2}} \cdot \frac{1}{\left( 2 + \frac{1}{n} \right) \left( r^n + (n+1)Bz \right)} z = \\
dh = \frac{2n}{n+1} \left\{ \frac{1}{r^n + (n+1)B} \right\} \cdot \left[ \frac{\tau_0}{r} (R - 1) + (n+1)Bh \right]^{\frac{1}{n}} \cdot \left( 1 + \frac{\tau_0}{r^n + (n+1)B} \right)^{\frac{2-n}{2}} \cdot \frac{1}{\left( 2 + \frac{1}{n} \right) \left( r^n + (n+1)Bh \right)} h = \\
-2K^2 \frac{u_z}{R} z + C_3, \\
\end{align*} 

(47)

where \( C_3 \) is determined from the boundary condition \( u_z(z=0)=0 \), giving

\begin{align*}
C_3 &= \frac{2n}{n+1} \left\{ \frac{1}{r^n + (n+1)B} \right\} \cdot \left[ \frac{\tau_0}{r^n + (n+1)B} \right]^{\frac{2-n}{2}} \cdot \frac{1}{\left( 2 + \frac{1}{n} \right) \left( r^n + (n+1)B \right)} \cdot \left( 1 + \frac{\tau_0}{r^n + (n+1)B} \right)^{\frac{2-n}{2}} \\
\end{align*} 

(48)

In order to solve for \( B \), the boundary condition \( u_z(z=h)=dh/dt \) is utilized, i.e.

To solve for \( B \) is not possible analytically. However, having explicit numbers on the parameters, a solution for \( B \) can be found numerically.
Paper C
Compression moulding of SMC: Modelling with CFD and validation

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ABSTRACT: Compression moulding experiments of Sheet Moulding Compound (SMC), visual observations of a vacuum test with prepregs, and numerical models for Computational Fluid Dynamics (CFD) for simulations of the present mould filling phase are here presented. The first model sees the flow as only being extensional while the other model sees the flow as mainly extensional but also shearing near the surface of the moulding tool. Pressure comparison with the experiments show that preheating effects can almost be neglected and that the pressure seems to be more accurately predicted if shear layers are present in the viscosity model of SMC. In the experiments it was confirmed that the pressure is predominantly affected by the mould closing velocity during mould filling, wherein three phases, separated by two pressure tops, also were observed. Regardless of here applied process settings, the first pressure top always appeared approximately at the logarithmic strain 0.25. The second top was associated with a slow down of the press and a subsequent decrease of the pressure until the applied pressure was reached when the mould was filled. The location at where this occurred was affected by the velocity and the vacuum level. Hence, it is logical to assume that vacuum assistance prevent a build up of back pressure. Furthermore, simple vacuum tests with prepregs indicated that prepreg also desiccate by vacuum, an effect that become more significant for heated prepregs which also swells immediately as vacuum assistance is applied if a critical temperature is reached.

KEYWORDS: SMC, Modelling, CFD

INTRODUCTION

Compression moulding of Sheet Moulding Compound (SMC) is a manufacturing method for composite products. The whole process can briefly be divided into two main steps, preparation and moulding of SMC prepreg, respectively. In the preparation step, prepreg sheets are manufactured which consists of a paste and fibres where the fibres are randomly spread and impregnated in the plane of the
sheets. Material formulations for these prepregs can be modified in uncountable combinations, for example to give them special rheological characteristics during manufacturing or to give the finalized product fire retardant properties, but in general they mainly consist of a thermosetting polyester resin, calcium carbonate fillers, and glass fibre reinforcements. In the moulding process, charges of stacked prepreg sheets are placed inside a heated moulding tool and as the mould closes, the charge is forced to flow and fill the cavity of the moulding tool. The mould is then kept closed until the part has cured. With this method, quite advanced shapes can be manufactured and due to the usage of prepregs, it is rather fast and therefore suitable for large scale productions of e.g. lightweight composite products in the vehicle industry where it is of great importance to achieve a high quality of the surface appearance. This is an issue for SMC manufacturers since voids located at the surface level sometimes cause so called pinholes or blowouts [1]. Voids inside the material is also of concern for the high voltage industry since voids decreases the electrical insulation properties [2]. Air entrapments during paste mixing and poor bundle impregnation have been observed as two sources to voids, but also that these voids are largely removed during compression moulding [3]. One possible cause of this is that the entrapped air can dissolve into the resin [4]. It is furthermore also possible that air entrapments can form due to complex flow as described in [5], where a squish effect was observed when the flow front was experimentally visualised during compression moulding. At the end of the flowing stage, boiling of styrene was observed as well [5], which also can result in voids in the final part. Several processing parameters have been found to reduce the amount of voids during compression moulding. For example, increased mould closing pressure, preheating of the SMC charge [6], vacuum assisted moulding [2, 7], and increased mould closing velocity [2]. A mutual effect for the just mentioned factors, except mould pressure, is that they seem to cause a more homogenous flow [2, 5, 8]. In [2], multi coloured SMC were used to study the effects from velocity, temperature, and vacuum assistance on the final flow pattern of moulded SMC plates. It was observed that at high mould closing velocity (10 [mm s⁻¹]), the flow become homogenous without the assistance of vacuum, but by applying vacuum assistance and using the lower mould temperature (420 [K] vs. 430 [K]), it also became homogenous at the low closing velocity (2.5 [mm s⁻¹]). Hence, it seems like a natural step towards total elimination of voids in finished SMC parts would be to develop models that accurately can predict the flow of SMC and from those models develop moulding tools, charge patterns, and prepreg materials in order to avoid flows that are more likely to cause voids. In addition to higher quality for manufactured products, a great deal of time and money could be saved since development of moulding tools and charge lay-ups for new products is often inhibited by some trial and error testing before the manufacturing comes to full capacity. The complexity of the prepreg material aggravates rheological measurements that are in need to develop models for computational simulation. Many have though made prominent work in the area through the years.

Rheology of SMC

Most SMC’s are thermosets, which itself can be seen as a Newtonian fluid, except at very high shear rates. However, by adding particles, the mix becomes non-Newtonian. This apply for SMC prepregs where the paste itself, without added fibres, behave as a non-Newtonian fluid with shear thinning properties [9, 10]. The viscosity of the paste as a function of temperature have been described as [10]
Eq. (1) is often combined with an expression stating that the shear viscosity is a power law function of the shear rate. But it has also been observed that the viscosity of the paste can be described by a Carreau viscosity model [9]. If the viscosity is assumed to be very small at infinitive shear, the Carreau viscosity model can be written as

$$\eta = \eta_s \left(1 + \left(\frac{\lambda \gamma}{\eta_s}\right)^{\frac{1}{n}}\right)^{\frac{n-1}{n}}$$

where $\eta_s$ is the zero shear rate viscosity, $\lambda$ the time constant, $\dot{\gamma}$ the shear strain rate and $n$ is the power law constant. This means that the paste behaves as a Newtonian fluid at low shear rates, and as the temperature increases, the Newtonian transition area moves to higher shear rates. It is widely accepted that unreinforced polymers and melts usually have a shear-dominated mould flow and that their flow behaviour can be modelled without considering the extensional viscosity dissipation [11]. However, when modelling materials such as SMC with planar fibre suspensions, it is popular to assume a gapwise velocity profile that is close to a plug flow and that the main viscosity dissipation being in-plane extension since many experiments have shown that SMC deform in a biaxial extensional flow mode with very little mixing between the prepreg layers and extensive slip along the mould walls [12]. It has also been shown that shearing effects can be significant even though the biaxial extension indeed dominates in most cases of fibre suspension squeeze flows [11]. When fibres are added into the plane of sheets, the compression viscosity, or elongation viscosity, is much more affected than the shear viscosity. The compression viscosity of SMC with fibres has been found to be a function of the paste viscosity, $\eta_{paste}$ and the volume fraction of fibres, $f'$ added to the paste according to the expression below [10].

$$\eta_{Compression} = 3 \cdot \eta_{paste}(T) \left(\frac{\dot{\varepsilon}}{\dot{\varepsilon}_0}\right)^{n-1} \cdot \left(1 + \beta \frac{f'}{1 - f'}\right)$$

Where $\beta$ is a constant that was found to be equal to 360, independently of temperature, and $\dot{\varepsilon}$ is the strain rate ($\dot{h}/h$). Later, in a subsequent study [13], Eq. (3) was revised into

$$\eta_{Compression} = 3 \cdot \eta_{paste}(T) \left(\frac{\dot{\varepsilon}}{\dot{\varepsilon}_0}\right)^{n-1} \cdot \left[1 + 100 \cdot f' + 1000 \cdot f' f''\right]$$

which gave a better fit to experiments, especially at low strain rates. It is in the context worth mentioning that the effect of fibres on the shear viscosity is not that dramatic, it have for example been observed that by adding 11.2 volume percent of fibres, the shear viscosity is only increased by a factor of 2.93 [10].
The fibrous microstructure of SMC have been investigated with X-ray phase contrast microtomography and it was observed that the plies before moulding consisted of a core zone with a high amount of fibres between two skins, upper and lower, that consisted of a small amount of fibres [6]. When the charges were moulded, the skins had disappeared for the plies placed inside the charge, creating a core zone with an even and high amount of fibres. However, the skins remained at the upper and lower plies which were in contact with the mould wall. The thickness of the skins remained approximately constant, 0.26 mm for the bottom layer, and 0.22 mm for the upper layer, independently of process settings, placement of charge, and preheat temperature of the charge. This imply that the SMC is highly sheared at the mould walls due to a low viscosity of a compound more or less without fibres and that plug flow models probably can be applied to describe the flow of the core zone [6].

**EXPERIMENTAL COMPRESSION MOULDING**

**Method and Materials**

Pressure data from compression mouldings were obtained from a process parameter experiments where the altered parameters were vacuum, mould closing speed, and the temperature of the moulding tool. The experiments were performed as a 2-level factorial design experiment with split plot design methodology since it was very time consuming to alter the temperature of the moulding tool, see Table 1 for process settings. Each setting was replicated 4 times, giving a total of 32 samples. The commercial software MiniTab 16 was used to facilitate planning and analysis of the experiments.

<table>
<thead>
<tr>
<th>Level</th>
<th>Low (-1)</th>
<th>High (1)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Vacuum</td>
<td>0 %</td>
<td>75 %</td>
</tr>
<tr>
<td>Velocity</td>
<td>2.5 [mm s⁻¹]</td>
<td>10 [mm s⁻¹]</td>
</tr>
<tr>
<td>Temperature</td>
<td>420 [K]</td>
<td>430 [K]</td>
</tr>
<tr>
<td>Pressure</td>
<td>450 [kN]</td>
<td></td>
</tr>
</tbody>
</table>

Table 1. Settings for the compression moulding experiments. Temperature is on the surface of the lower mould half. The upper surface was 3 °K lower.

An experimental moulding tool of circular geometry with two pressure sensors and with vacuum assistance capability was used for the experiments, see Figure 1. A standard industrial low-profile prepreg was used as experimental material, see Table 2 for a summary of the material composition. From this prepreg, circular sheets of diameter 0.16 [m] was prepared. They were thereafter stored in air sealed vessels in order to avoid significant styrene loss before they were randomly picked for the experiments where they were stacked in 7 layers. This resulted in an average height of 14.7 mm. The weight of the stack was controlled before it was placed in the centre of the moulding tool. To increase repeatability and shorten the time that the charge had to lay inside the hot moulding tool before it was moulded, markings were made inside the tool. This facilitated a fast and accurate manual loading of the charge.
Figure 1. Schematic illustration of the experimental setup. The radii of the SMC charge and the mould cavity is 80 mm and 159 mm, respectively. As the upper mould half goes down it encapsulates the cavity which enable the use of vacuum assistance.

Table 2. Composition of the paste. In addition fibres were added to an amount that took up a weight of 26-27 % of the total weight of the paste and fibres.

<table>
<thead>
<tr>
<th>Component</th>
<th>Mass %</th>
</tr>
</thead>
<tbody>
<tr>
<td>Resin</td>
<td>19.3</td>
</tr>
<tr>
<td>Low-profile additives</td>
<td>8.9</td>
</tr>
<tr>
<td>Fillers</td>
<td>66.8</td>
</tr>
<tr>
<td>Inhibitor</td>
<td>2</td>
</tr>
<tr>
<td>MgO</td>
<td>0.77</td>
</tr>
<tr>
<td>Internal release agents</td>
<td>1.5</td>
</tr>
<tr>
<td>Wetting agent</td>
<td>0.46</td>
</tr>
</tbody>
</table>

Experimental results
A summary of the experimental pressure response are presented in Figure 2 where each curve is the average pressure response from 4 separate mouldings. As seen in the figure, the pressure rapidly increases until a clear peak forms for the centre pressure sensor approximately at the height of 11.4 mm for both closing velocities. This peak is not as distinct for the outer sensor, instead the increase in pressure is flattened out. The pressure then drops a little at the centre sensor before it then starts to build up again. Interestingly the pressure curves from the outer sensor then border on the ones from the inner sensor indicating that there is a marginal flow and/or very low viscosity flow. During the period visualised in Figure 2, the press was capable of deforming the sample at constant velocity. However, as the press approached the lowest position, it suddenly decelerated and the pressure quickly decreased and finally reached the applied pressure when the press had reached the lowest position and the SMC had completely filled the cavity.
Statistical analysis of the experimental results shows several significant results that were found by using a confidence level of 95%. The results are here divided into two phases during the closing of the moulding tool.

**Phase 1 – Start to the first pressure top**

The height at which the pressure started to increase, ergo the height where the upper mould half reached the SMC charge, was found to be mainly affected by the vacuum. On average, the pressure for the samples with applied vacuum started to increase 0.64 mm before it started to increase for the samples that were moulded without vacuum assistance. The distance, 0.64 mm is not long, but it was a significant result. Therefore, an additional simple experiment was carried out where prepreg at different temperatures, room temperature, 50 °C, and 80 °C, were placed in a transparent vessel wherein vacuum was applied. For the prepregs at room temperature and 50 °C, nothing extraordinary occurred except that they seemed to dry out after a while. The sample that was preheated to 80 °C, however, immediately and rapidly swelled up when the vacuum was applied. A boiling liquid could also be seen at the surface of the transparent vacuum vessel. It is likely that the observed boiling liquid was styrene that had reached its boiling point due to the applied vacuum [5, 14]. Another significant effect was that the pressure at the first pressure top was mainly affected by the mould closing velocity. No significant effect was found to affect the difference in pressure when the first pressure peak occurred between the two sensors.

**Phase 2 – After the first pressure peak**

The distance from the first pressure top to the position at where the minimum pressure occurred was found to be significantly affected by the mould closing velocity. At average, the mould had enclosed 1.9 mm further for the slow than for the fast velocity. The distance between the first pressure top and the height at where the pressure reached the same pressure again was mainly affected by the velocity. At
average, the press had moved 2.1 [mm] further for the low velocity than for the high before it occurred. The closing velocity of the press was set to go at constant velocity the whole way. However, the press did not manage to do this as discussed previously. According to the statistical analysis there where two main effects, velocity and vacuum, and one interaction effect between velocity and temperature, that affected the height at where the deceleration was initiated. The largest difference was caused by the velocity setting. At the high velocity, the deceleration occurred earlier than at low velocity. The velocity was also involved in an interaction effect with the temperature level. At slow closing velocity, the press was capable to compress the SMC with constant velocity at the same distance for both the high and the low temperature. With the higher closing velocity, the press managed to keep constant velocity further with the higher temperature. Vacuum assistance seemed to aid the press at the end of the moulding phase since the press on average managed to compress the SMC further with constant velocity with applied vacuum assistance. This indicate that vacuum assistance prevent a pressure build up of encapsulated air inside of the moulding tool.

**Compression viscosity**

In [10], a compression viscosity was calculated as

\[ \eta_{\text{Compression}} = \frac{\sigma_{\text{Threshold}}}{\dot{\varepsilon}}. \]  

\[ (5) \]

It was found that the result followed a power law. Performing the same operation here with the pressure at the pressure peak registered by the centre sensor for the low and high mould closing velocity and with the average result for each process setting gives the average strain rate sensitivity \( n = 0.33 \), ranging from 0.28 to 0.36, see Figure 3.

![Figure 3. The compression viscosity at the first pressure peak, derived from average values from each setting. The average \( n \) is 0.33, ranging from 0.28 – 0.36.](image)

This result is however not fully trustworthy since a temperature gradient from the moulding tool was present in this work which probably lowers the compression viscosity more for the low closing velocity than for the high velocity due to longer time in contact with the mould before the pressure top was attained. It is therefore
reasonable to assume that if the moulding experiment should have been isothermal, the pressure at the low velocity should have been higher in comparison with the pressure at the high closing velocity than now. As a consequence, \( n \) would have approached the expected value 0.3, found from linearization of data in [15]. In [10] a constant strain rate and a homogeneous temperature distribution were applied. Here, the velocity was set to constant which gives an increasing strain rate as the mould closes and the gap between the mould halves decreases. From these results, curves could be plotted, see Figure 4 for one example. In the figure it can be seen that the compression viscosity decreases faster in the beginning, this is probably due to preheating from the moulding tool and the squish effect [5]. At the end, the curve flattens out and \( n \) approached the expected value 0.3.

**Figure 4.** Compression viscosity for a sample from start until the second pressure top seen in Figure 2. The highest value occurs at the logarithmic strain 0.25.

**NUMERICAL SIMULATION**

The commercial software Ansys CFX 12.1 has been used for the simulations. The starting point for this paper was to improve a previously developed viscosity model for CFD simulations of compression moulding of SMC [16].

\[
\mu(T) = A e^{\beta(T-T_a)}
\]

With a multiobjective surrogate-based inverse modelling technique Marjavaara et al. managed to obtain simulations that captured the flow behaviour of SMC, and the pressure could successfully be predicted but unfortunately not at two locations simultaneously. Hence, the main goal for his study was to improve the model so that the pressure could be predicted adequately at two locations at the same time. In addition it turned out early that Eq. (6), with only temperature dependence, is not suitable when the mould closing velocity is altered in the experiments as done in the present study. This was not observed in [16] since only one mould closing velocity was used (2 mm/s).
Model Setup
Several models were evaluated, and all of them had some geometrical and material properties in common. In the experiments, we have previously shown that there was a pressure top approximately at the same height regardless of processing parameters used. The same behaviour has previously been observed in [10], where it was concluded that the pressure seemed to reach a maximum at the logarithmic strain, \( \varepsilon = 0.25 \ln \left( \frac{h_0}{h} \right) \). By observing the SMC charge, it was concluded that the flow of it was initiated after the pressure top [10]. This phenomenon was addressed to air removal. Hence, it is likely that the SMC charge is only compressed until the first pressure top. Therefore, the model is setup to start at 11.4 [mm] which was the average height at where the pressure top occurred. This is actually when \( \varepsilon = 0.25 \). The cylindrical geometry of the experimental moulding tool is also applied in the numerical model. Since the geometry is axisymmetric, it can be modelled as a slice with symmetry boundary conditions. The slice is, in accordance with the Ansys guidelines, made up of one layer that is extruded 1 degree.

Thermal properties of the material are of great importance, especially when modelling the flow of SMC since the viscosity is directly dependent on the temperature. Thermal conductivity experiments performed on a similar prepreg yielded a value of 0.78 [Wm\(^{-1}\)K\(^{-1}\)] during heating up to 80 °C [17]. Later, during cooling from 120 °C to room temperature, the value of 0.98 [Wm\(^{-1}\)K\(^{-1}\)] was found. The increase was traced to decreased levels of air inside the stack after curing. Therefore, the value 0.78 [Wm\(^{-1}\)K\(^{-1}\)] will here be used to reflect the initial conditions in the stack while later during compression the value 0.98 [Wm\(^{-1}\)K\(^{-1}\)] is applied when much of the air is removed. When calculating these values, the density of the prepreg was measured to 1.816 [g cm\(^{-3}\)] and the value 1300 [J kg\(^{-1}\) C\(^{-1}\)] was used as specific heat capacity. These data will be applied here as well but, since the stack is assumed to be compressed before the flow is initiated, the density is adjusted to 2.33 [g cm\(^{-3}\)].

Heat will be transferred from the tools to the charge before the SMC starts to flow. During the experiment, the charges were placed by hand on the lower mould half before the press were manually started, a process approximated to 5 [s]. In addition we need to account for the time to move the upper mould half down to the height at where the flow is assumed to start, that is \( \varepsilon = 0.25 \). In the experimental compression mouldings, the press was programmed to go with very high velocity down to the height 40 [mm], then it slowed down and finally closed at desired constant velocity, 2.5 or 10 [mm s\(^{-1}\)]. As a consequence, the preheating time for samples before the actual mould flow was initiated varied between the two mould closing velocities. Initially, this whole time was accounted for before the actual flow actually was initiated in order to obtain accurate initial conditions, see Table 3 for details. However, during the simulations, it could be seen that the overall heat transfer probably was smaller in reality than in the simulation since very large differences in pressure compared to the experiments were predicted for the two velocities when different time was used to simulate the initial temperature in the stack. Instead, when the same initial temperature was used for the two velocities, the temperature became in better agreement with the experiments. It could furthermore also be seen in the simulations that as the time of preheating decreased, the difference in pressure also decreased between the two measuring points, approaching the experimental values. Therefore, the preheating time for the upper and lower side of the SMC were arbitrary set to 0.35 [s] and 1.3 [s], respectively in the final simulations.
Table 3. Initial elapsed time for the heat transfer simulations that were used as initial conditions in the moulding simulations. The time for the lower half is: approximated time from placing the stack inside the mould to start the press + time for the press to reach the upper surface + time to compress the stack to $H = 0.25$.

<table>
<thead>
<tr>
<th>Velocity [mm s$^{-1}$]</th>
<th>Lower [s]</th>
<th>Upper [s]</th>
</tr>
</thead>
<tbody>
<tr>
<td>2.5</td>
<td>5 + 12.7 + 1.3</td>
<td>1.3</td>
</tr>
<tr>
<td>10</td>
<td>5 + 3.6 + 0.35</td>
<td>0.35</td>
</tr>
</tbody>
</table>

Two different models for the viscosity have been evaluated. The paste viscosity is assumed to obey the model in equation (1), since it made a nicer fit with the data presented in [9] than equation (6). Fitting equation (1) to the data in [9] gives $b$ the value 11708, significantly higher than reported in [10] and [18] were 4900 and 4500 was found, respectively. The compression viscosity in both models are assumed to follow equation (4) with $\eta_0$ as 55000 [Pa s] similarly as [10]. The value 0.3 is used for the power law constant $n$. Hence, the only constant in the models that has been changed during simulation is $b$. First, a suitable value of $b$ was found for the low velocity and the low temperature for the used pre-heating time. Then the same $b$ was used for the remaining combinations of high and low velocity and temperature at that particular pre-heating time.

Model 1

Due to the common opinion that SMC mainly deforms in extensional flow mode during compression, this model is assumed to only depend on extensional viscosity during flow. The viscosity is based on findings in [10] and [13], where the compression viscosity was described as a function of paste viscosity, strain rate and fibre volume fraction. From [6], the $f/F$ value 0.188 were taken which was the average fibre volume content in the prepreg.

$$\eta_{\text{Compression}} = 3 \cdot \eta_{\text{Paste}}(T) \cdot \left( \frac{\dot{\varepsilon}}{\dot{\varepsilon}_0} \right)^{n-1} \cdot \left( 1 + 100 \cdot f/F + 1000 \cdot f/F^2 \right)$$

(7)

Model 2

Based on the observation in [6], that skins were formed during compression that were highly sheared and an inner core zone with a high volume percent of fibres which seemed to obey extensional flow, a thin layer of shear dominated flow was added to the model, representing the skins. It was observed in [6] that the core had a fibre fraction of 0.2, and that the skins had a ditto of about 0.12. In [10], the shear viscosity was observed to increase by a factor of 2.93 for the fibre fraction 0.112, therefore we simply multiplies the shear viscosity of the paste by 3 in the layers. In order to get rid of problems with shear rates under 1, the Carreau model (Eq. 2) was employed with the static value of 1 for the time constant, giving the curve a power-law-like appearance. Furthermore, the effect of a temperature dependent time constant was tested as well. Here, the data in [9] was used to fit a curve for the time constant. In the core zone, model 1 is still assumed to work. The average thickness of the skins in [6] was in average 0.22 [mm] for the upper one and 0.26 [mm] for the lower one. These thicknesses were unaffected by moulding conditions, the initial prepreg temperature, sample location, and if the prepreg stack were in contact with the mould for 10 [s] before the flow was initiated. The height of the skins in the
The present study are for simplicity set to 0.25 [mm]. The viscosity in the core zone obey model 1 (Eq. 7), and the viscosity in the skins is set to follow the following Equation

\[
\eta_{SW} = 3\eta_0 e^{-b \left( \frac{T}{T_b} \right)} \left( 1 + \left( \frac{T_b}{T} \right) \right)^{\frac{e-1}{2}}
\]  

\( (8) \)

**Results**

The modelled pressure for various pressing velocities, preheating times and values on the material property \( b \) is here presented. In Figure 5-12, solid lines show the pressure predicted in the centre and dotted lines show the pressure predicted at the radii 0.07 m, representing the location of the outer sensor. The curves named Centre and Outer are the experimental means presented in Figure 2. The preheating time is varied since it was noticed that the initial temperature of the modelled SMC charge highly affected the result while the material property \( b \) is matched to get a reasonable enough initial pressure while the material properties \( \eta_0 \) and \( n \) were held constant in order to reduce the number of modelling cases.

**Model 1**

For the initial estimated pre-heating times in Table 3 the pressure increases much too rapidly as the mould closes for both closing velocities, see Figure 5 and Figure 6. It can also be seen that the difference in pressure is large between the two points of measurements in comparison with experimental data. However, when the modelled initial temperature for the slow velocity is used, the initial pressure becomes much closer to the experimentally obtained pressure. Hence, it seems like the modelled initial temperature is overestimated even though a lower value of thermal conductivity was used during the period of pure heat transfer. Further modelling reveal that by decreasing the time for the initial temperature modelling, ergo assume that the heat transfer is less than what has been set, the predicted pressure approaches the experimental for both locations since the value of the constant \( b \) is allowed to increase as the initial temperature decreases. See Figure 5 and Figure 6 where the pressure curves also can be seen when \( b \) is adjusted to 7800 for the preheat time 8.6 [s] (0.35 [s] at the upper surface).

To go one step further it is simulated that the lower surface was heated for 1.3 [s] and the upper surface for 0.35 [s] at the two temperatures 420 and 430 [K] respectively. The result is that for the lower temperature, the simulated pressure follows the experimental results for the low temperature and the low velocity, see Figure 7. But, when the higher temperature was used, the pressure decreased too much as a consequence of the high value of the constant \( b \). For the higher velocity (Figure 8), the pressure increased too rapidly for both temperatures and there was a rather large difference in pressure between the two temperatures. In Figure 7, it can also be seen that the model is sensitive to changes in the value of \( \eta_0 \). By only increasing it to 60 000 [Pa s], the constant \( b \) can be increased to 9100, which gives a result that is closer to the experimental values.

**Model 2**

The modelled pressure for each setting and experimental means can be seen in Figure 9 - Figure 12. The value of the constant \( b \) could in this case be dramatically decreased by assuming that the outer layers behave as shear layers. Also for this
model, the predicted pressure came closer to the experimentally obtained pressure if the preheat time was decreased, therefore the preheat time 1.3 [s] for the lower surface and 0.35 [s] for the upper surface was utilized. By assuming that the shear layers have a time constant value of 1, which gives the viscosity curve a power law-like appearance, the constant $b$ could be adjusted to rather low values, see Figure 9 and Figure 10. As seen the fit between the predicted pressures and those obtained from the experiments is rather good for both velocities and best for the higher velocity. The low value of $b$ makes the viscosity less dependent on temperature which can be seen for both velocities. In Figure 10 and Figure 11, the predicted pressure can be seen when the time constant is chosen to be a function of the temperature. As a consequence, the shear thinning effect in the layers can almost be neglected due to the high temperature of the SMC in the layers. Therefore, the pressure increase a little too fast for the low velocity (Figure 11), which it also do for the high velocity, but there the initial pressure is too high as well, see Figure 12. The lower value $b$, compared to those in model 1, makes the viscosity less temperature dependent and resulting smaller differences in pressure for the two temperatures.

Figure 5. Model 1 and experimental pressure with velocity 2.5 [mm s$^{-1}$].
Figure 6. Model 1 and experimental pressure with velocity $10 \, [mm \, s^{-1}]$.

Figure 7. Model 1 and experimental pressure with velocity $2.5 \, [mm \, s^{-1}]$. 
Figure 8. Model 1 and experimental pressure with velocity 10 [mm s$^{-1}$].

Figure 9. Model 2 and experimental pressure with velocity 2.5 [mm s$^{-1}$].
Figure 10. Model 2 and experimental pressure with velocity 10 [mm s\(^{-1}\)].

Figure 11. Model 2 and experimental pressure with velocity 2.5 [mm s\(^{-1}\)].
Figure 12. Model 2 and experimental pressure with velocity 10 [mm s⁻¹].

DISCUSSION

The value of $\eta_0$ is of course a great source of vagueness, a proper rheological investigation of the paste would have been desirable. The same thing applies for the thickness of the shearing layer. However, both of those material [6, 10] were similar to the one used in these experiments.

The modelling of the initial conditions of the temperature in the stack seemed to overestimate the heat transfer even though a lower number of the thermal conductivity was utilised during this period. When the same initial conditions were used for the two velocities, the pressure approached the experimentally obtained values. Perhaps a more accurate initial condition of the temperature would be to fully neglect preheating effects or in some manner restrict the heat transfer to some limit. Noticeable from the simulations is that as the preheating time is increased, the difference between the pressure readings from the centre and outer pressure increase as well. A more accurate initial condition for the temperature could be found by adjusting the modelled preheating time until the pressure difference in the modelling become equal as it is in the experiments.

In model number 1 [10, 13], shear thinning effects are neglected. However, the high value of $b$, compared to 4900 and 4500 in [10] and [18], respectively, might cause similar effects as shearing would do. The high values of $b$ applied are not unlikely
and in the same order as the value predicted when fitting Eq. (2) to the data presented in [9] ($b=11708$).

Model 2 with a time constant equal to 1 showed very good agreement at the high velocity but not as good at the low velocity. It is possible that the time constant should be affected by the temperature, but not as much as in [9] due to the addition of fibres. An effect of this would have been that the shear thinning effect at the low velocity would have decreased due to the increased temperature. This could have made the pressure curve approach the experimental curve.

From this study, it was evident that the viscosity of SMC could not be described by only the temperature as in [16]. However, the approach, multiobjective surrogate-based inverse modelling, may very well be useful if a more advanced viscosity model is to be determined from e.g. model 2, since it turns out that initial conditions and material parameters can be adjusted to get very good fit between simulations and experimental results at different locations in the mould and at different compression speeds.

**CONCLUSIONS**

From the experiments it is clear that mould closing velocity is the main effect for the pressure. Some kind of yield point that was independent of altered process parameters was found approximately at the logarithmic strain 0.25. The compression viscosity behaves as a power law fluid, but due to the thermal gradients it is not possible to determine the exact value of the power law exponent. Vacuum assistance seems to prevent a pressure build up inside the moulding tool. Heated prepreg (80 °C) momentarily swells as vacuum is applied, hence the full function of vacuum assistance during compression moulding of SMC may be more complex than just removal of air.

The model that seemed most robust in predicting the pressure of the tested models was model number 2, where the SMC obey two deformation modes, extensional flow in the centre (Eq. 7), and shear flow near the surface of the moulding tool (Eq. 8).

**ACKNOWLEDGEMENTS**

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**REFERENCES**