Process Simulation of LPM (Liquid Polymer Molding) in Special Consideration of Fluid Velocity and Viscosity Characteristics

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SUMMARY: Recently in RTM process technique in-situ polymerizing thermoplastics are a promising alternative for conventional thermoset matrix systems. The advantages of these systems are a low initial viscosity at the beginning of the process combined with the polymerization proceeding during injection. The low initial viscosity allows a fast mold filling even at a high fiber volume fraction which leads to a high fluid velocity especially at the beginning of the process. Additionally, the polymerization during injection leads to a change not only of the viscosity level but also of the viscosity characteristic to a shear rate dependent behavior. The high flow velocity as well as the shear dependent viscosity do not allow to model this kind of process by the generally accepted Darcy's Law which assumes low flow velocity and newtonian viscosity of the fluid. The intention of this paper is to investigate a high velocity fluid flow in a non-crimp fiber bed as well as the influence of visco-elastic fluid characteristic on the fiber bed microstructure.

KEYWORDS: Permeability, visco-elasticity, Newtonian, micro structure, Darcy's Law, high fluid velocity, viscosity characteristic, non-crimp

INTRODUCTION

RTM (Resin Transfer Molding) is a common process covered by the generic term LCM (Liquid Composite Molding) used to manufacture high performance parts. Recently an alternative for thermoset resins used in RTM is developing by in-situ polymerizing thermoplastics (ISPT). This kind of thermoplastics are polymerizing during mold filling which changes the flow behavior of the fluid from Newtonian to visco-elastic flow and causes a rise of viscosity by a factor of 100 starting from an initial viscosity of 17 mPas [1]. Due to these significant changes in flow characteristics it is necessary to adapt the process simulation software.

Most simulation tools are based on Darcy's law, which is one of the most common used empirical equation to describe flow through porous medias.

In its simplest formulation it takes the form:

$$v = \frac{K_f}{\eta_f} \cdot \frac{\Delta p}{\Delta x} \tag{1}$$

In this formulation v denotes the fluid velocity respectively the flow front speed, η_f represents the viscosity of the infiltrating fluid, K_f stands for the permeability of the porous media and $\Delta p/\Delta x$ represents the pressure gradient [2]. Darcy's Law is restricted to slow inertia free flow and a low, newtonian viscosity [3]. The key parameters in this equation are the permeability and in this particular case the viscosity. Usually the permeability is determined by one- or multidimensional flow experiments which are described elsewhere [3]. These experiments can for example be evaluated by the constant method, in which the permeability over the flow length is assumed to be constant. The following equation thus results directly by integration from Darcy's Law:

$$K_{const} = \frac{m \cdot \eta}{2 \cdot p_0} \tag{2}$$

In this formulation K_{const} denotes the permeability to be calculated, η represents the constant viscosity of the fluid, p_0 stands for the constant injection pressure and m represents the ascending slope of the straight line resulting from linearized experimental values in a diagram square of flow path over time. [4].

FLUID CHARACTERIZATION

Apart from the permeability the fluid viscosity plays a key role in the use of Darcy's Law. The change in viscosity over time and temperature as well as the dependency of the viscosity on the shear rate has to be included. Due to the difficult handling of ISPT systems a replacing fluid, a duromer system (Ly113 / Hy97) from the Huntsmann company was used for this study. For reference issues a standard vegetable oil with newtonian flow behavior was selected. The viscosity of the vegetable oil is ranging from 0,055 Pas at 20°C to 0,036 Pas at 27,5°C in which the all isothermal flow experiments have been executed.

To quantify the viscosity characteristics, measurements have been executed by using a plate-plate rheometer. The viscosity evolution of the epoxy resin at high shear rates and different temperatures is shown in fig. 1a. The initial viscosity is decreasing from 0,690 Pas at 20°C to 0,325 Pas at 27,5°C but at the same time the polymerization velocity is rising significantly. The shear rate dependency of the viscosity was measured in a range from 0,023 1/s to 225 1/s. The results are displayed in fig. 1b. These graphs illustrate that at low shear rates (> 10 1/s) the fluid viscosity is strongly shear rate dependent. Apparently the viscosity at shear rates lower than 0,1 1/s depends only on the shear rate but not on the degree of polymerization. This effect occurs because the macromolecular chains need only one energetic level to keep their degree of orientation independent from the chain length and therefore independent from the grade of polymerization. In respect of the shear rate dependency the epoxy is showing the same behavior as thermoplastic melts.

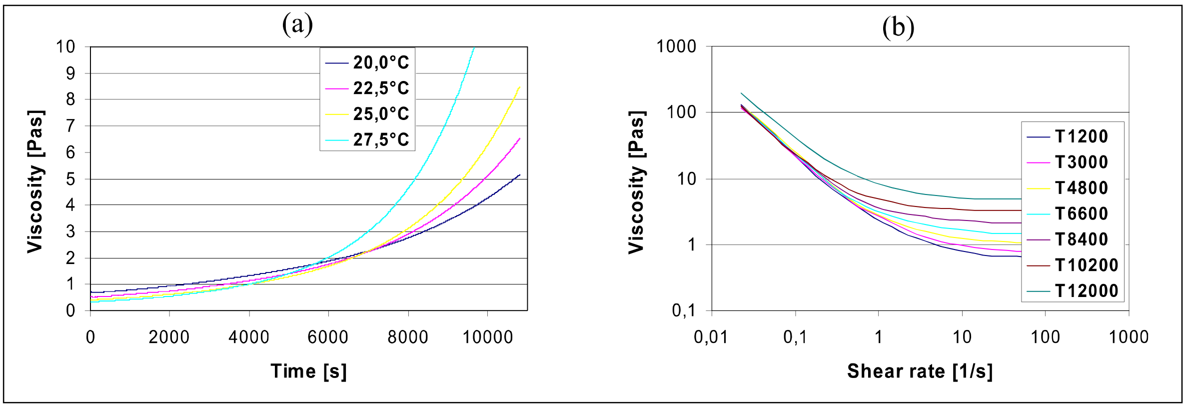

Fig. 1: a) Temperature dependent viscosity over time for the epoxy resin
b) Time dependent viscosity over shear rate for the epoxy resin

FLOW EXPERIMENTS

In order to get a reference, flow experiments were performed fulfilling the restrictions of Darcy's Law to sufficient low flow velocity and to shear rate independency of the fluid. Flow experiments have been carried out with vegetable oil using a fabric with a fiber volume content of 26% and an injection pressure of 0,2 bar. Fig. 2 (a) shows the square of flow path over time. It can be seen that the experiment displayed by graph 1 can be fitted quite well with a straight line through the origin of the diagram given by the Darcy's Law theory. This indicates that the experiment fulfills sufficiently the restrictions of Darcy's Law. Then, experiments with an injection pressure of 0,5 bar were executed. The results are shown by graph 2 in fig. 2 (a). It is obvious that the experimental values are differing from the straight line predicted by Darcy. Up to a flow length of approximately 0,17 m (0,03 m²) the flow is affected by inertia resulting in an overestimation of the flow front position by Darcy. From this point onward microstructural investigations are indicating flow channels which are apparently due to the higher pressure. This changes the permeability of the fiber bed resulting in a flow front velocity underestimated by theory.

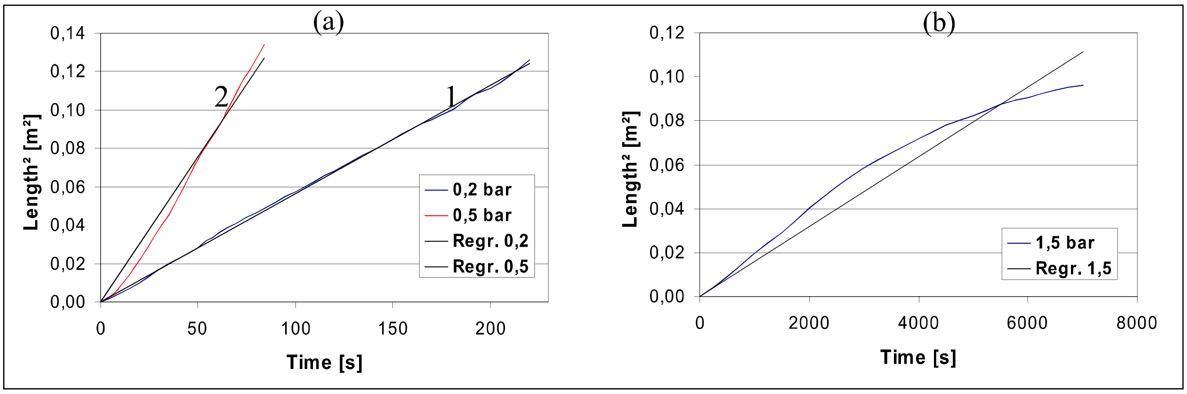

Fig. 2: Square of flow path over time at an injection pressure of 0,2 bar (a) and 0,5 bar (b)

In the next step the differences of Darcy- and visco elastic flow were quantified. Again, the experiment with vegetable oil and 0,2 bar injection pressure stands for reference. For this purpose a fiber volume fraction of 50%, an injection pressure of 1,5 bar and epoxy resin as fluid were used. Fig. 2 (b) shows the square of flow path over time of this experiment. Once more up to a flow length of approximately 0,17 m (0,03 m²) an inertia affected flow can be observed by a discontinuity in curve progression. A region of flow channel dominated the flow up to 0,24 m (0,06 m²) followed by the end section where the flow is decelerated because of a rising viscosity resulting from decreasing shear rates.

MICRO STRUCTURAL INVESTIGATION

The aim of the micro structural analysis was to investigate the influence of the fluid flow on the non-crimp fabric bed. The sample plates used for the investigation are produced with the same set of parameters mentioned in the paragraph above. In order to analyze the fluid influence polished cross-section cuts have been prepared from the plates impregnated by the epoxy resin. From this samples micrographs were taken to investigate the mesoscopic distribution of bundles, the developing flow channels inside the fiber bed and the distribution of the filament inside the bundles itself. The micrographs in fig. 4 are cut from the flow path length of 15 mm, 115 mm, 265 mm, 365 mm. It can be seen that the bundle distribution at the beginning of the flow path is nearly homogeneous. Picture 4 (b) and (c) at 115 mm and 256 mm flow length show, that with proceeding flow length a forming of flow channels takes place in the fiber bed influencing the flow front velocity during injection as already stated. This forming of flow channels can be observed reproducibly in all produced plates. In the boundary areas of the single fiber bundles a higher fiber volume fraction than in the mid area can be observed. A possible explanation for this effect is the transversal impregnation of the bundles [5]. Picture 4 (d) shows that the bundle distribution is again nearly homogeneous but the bundles are less compacted than at the beginning of flow. This change of the flow mechanism might be due to the lower flow speed and pressure gradient. It can be assumed, that the main flow switches from macroscopic to microscopic flow.

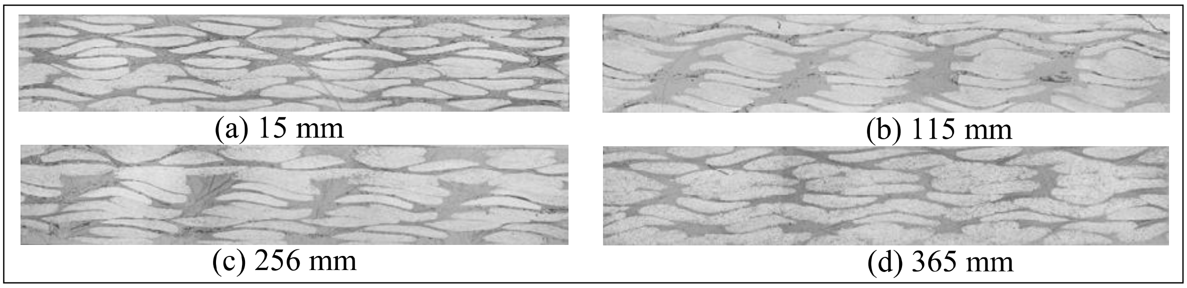
(a) 15 mm
(b) 115 mm
(c) 256 mm
(d) 365 mm

Fig. 4: micrograph of impregnated fiber bed at the (a) beginning, (b), (c)middle and (d) end of flow path

MICROSTRUCTURAL SIMULATION

In order to quantify the influence of the shear thinning characteristic of the fluid on the velocity distribution numerical simulations were performed. Fig. 5 shows the results of these simulations. The dimensionless flow speed v / v_{max} for a newtonian (center) and a shear-thinning fluid (left) in a highly resolved detail of a fiber bed, as well as for a newtonian (right top) and a shear-thinning (right bottom) fluid in the whole fiber bed at low resolution is displayed. All the results shown here are computed for periodic boundaries of the domain and a no-slip condition at the fiber boundaries. The results indicate that flow inside fiber bundles is negligible compared to flow between fiber bundles, and that for shear-thinning fluids the flow rate through the pores is higher than for comparable Newtonian fluids. This last result is in compliance to the experimental results which showed a higher flow front speed for the shear thinning fluid compared to the newtonian reference

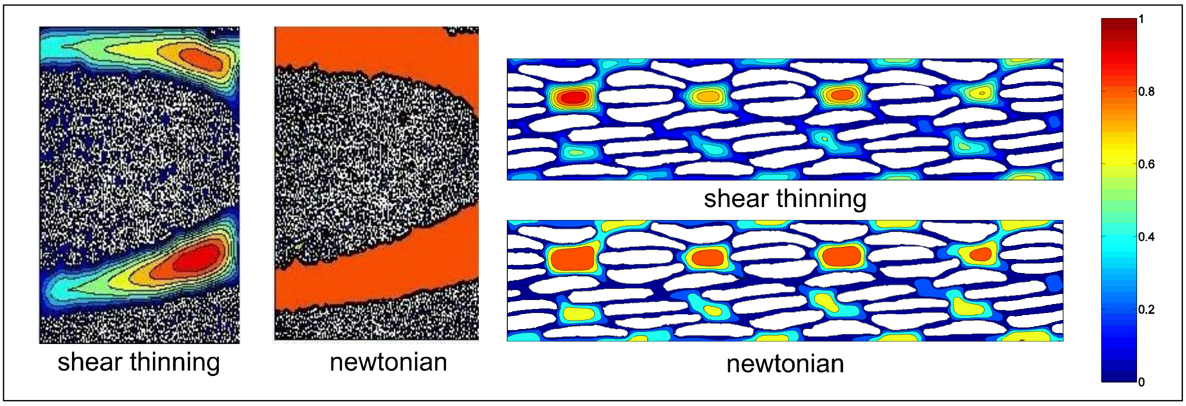

Fig 5: Dimensionless flow speed v / v_{max} for a newtonian and a shear-thinning fluid

CONCLUSION AND PERSPECTIVES

It can be stated that flow channel driven flow and therefore transversal impregnation of bundles is one of the most important mechanisms for transfer molding processes. From this arises the need to understand and to investigate the role of the microstructure during the process. One further aim is to detect the impregnation mechanisms on a micro structural level to achieve a more accurate flow simulation on a macroscopic scale.

REFERENCES

[1] R.H.J. Eder, Cyclics Thermoplastics - Properties and Processing, *IVW Schriftenreihe*, Volume 25, pp. 33-43, (2001)

[2] H.P. Darcy, Les Fontaines Publiques de la Ville Dijon, Victor Dalmont, Paris, 1856

[3] Huber, U., Zur methodischen Anwendung der Simulation der Harzinjektionsverfahren, *IVW Schriftenreihe*, Volume 26, Dissertation University Kaiserslautern, (2001)

[4] Flemming, M., Ziegmann, G. & Roth, S., Faserverbundbauweisen – Fertigungsverfahren mit duroplastischer Matrix, Springer-Verlag: Berlin and New York, 1999

[5] Huber, U., Maier, M.: Flow Behavior in saturated and unsaturated unidirectional fiber beds - a flow model, *Proceedings, FPCM-6 (2002)*, pp. 85-85, Auckland, New Zealand