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MULTISCALE MODELLING OF STOCHASTIC EFFECTS IN MOULD FILLING SIMULATIONS FOR THERMOPLASTIC COMPOSITES

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SUMMARY: Liquid moulding processes suffer from inherently present scatter in the textile reinforcement properties. This variability can lead to unwanted filling patterns within the mould resulting in bad parts. If thermoplastic resins are used with the in-situ polymerisation technique, an additional difficulty appears. The time window to inject the material is small if industrial processing parameters are used (<5 minutes). To model the stochastic nature of RTM, Darcy's description of the mould filling process has been used with the permeability distribution of the preform given as a random field. The random field of the permeability is constructed as a correlated field with an exponential correlation function. Optical microscopy and X-ray micro-CT have been used to study the stochastic parameters of the geometry for 2D and 3D woven textile preforms. The parameters describing the random permeability field (average, standard deviation and correlation length) are identified based on the stochastic parameters of the geometry for the preforms, analytical estimations and CFD modelling of the permeability. In order to implement the random field for the permeability and the variability for the resin viscosity, an add-on to the mould filling simulation software PAM-RTM™ has been developed. This analysis has been validated on case studies.

KEYWORDS: stochastic modeling, thermoplastic textile composite, X-ray µCT, viscosity measurements

INTRODUCTION

Resin transfer molding is a production process gaining a lot of interest to produce polymer composites. It allows producing parts of complex geometry. Those parts can be used in high performance applications. Another advantage of this production technique is the low emission levels during production. One disadvantage of the process is connected with the large variations that appear in the material properties governing the process. Those changes in values can be the cause of unwanted filling patterns during production. In the worst case, this can lead to rejection of the composite part. Up to now, there was no possibility to take into account stochastic factors to describe the material properties in a mould filling simulation software for RTM. It is only possible to perform a deterministic simulation resulting in a filling pattern, pressure field and possible air entrapment locations. This paper describes how the most important parameters for this process can be measured. Next, the implementation of the scatter for the different parameters in an existing mould simulation package is discussed. Finally, additional results from case studies are compared to a deterministic simulation.

PRODUCTION PROCESS

The different stages of the RTM process are shown in Fig 1:

- Textile reinforcement to be placed inside a mould (can be heated).
- Application of pressure to inject the resin (the maximum pressure is usually less than 1 MPa and the viscosity must remain below 1 Pa·s).
- Curing for a thermoset resin, polymerization for a thermoplastic resin.
- Cooling and subsequent demolding of the part.

The main parameters governing the process are the textile permeability and the resin viscosity. In combination with the dimensions of the mould and the applied pressure drop, these parameters are used in Darcy's law (Eqn. 1):

$$\langle v \rangle = -\frac{[K]\langle \nabla p \rangle}{\eta} \tag{1}$$

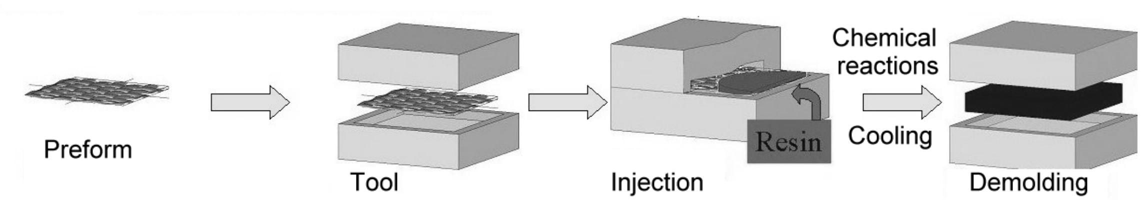

Fig. 1 Schematic showing the different stages of the RTM process.

TOPIC OF THE PAPER

Concerning Permeability

Trying to identify the permeability of textile reinforcement is already the topic for several years within the field of polymer composites [1]. Several experimental setups have been built to obtain a uniform permeability value for a given textile reinforcement. Each approach has its advantages and disadvantages. Performing permeability measurements always results in a high scatter for the permeability values. This is caused by several factors. The measurement technique and its corresponding disadvantages are of big influence together with the skills of the operator. Next, the arrangement of different layers of fabrics is of large influence together with the internal geometry variation of the textile layer. Hoes [3] tried to control all those influences by performing a lot of measurements for the same textile structure. This resulted in a coefficient of variation of 20% for specific textile reinforcement [2]. Hoes [3] thought that this is mainly due to the different nesting of the layers.

As performing a permeability measurement is time consuming and difficult, one tries to develop computer models to find permeability values based on a geometrical description for textile reinforcement. Generating a good geometrical description of textile reinforcement is another challenge. At the MTM department of KU Leuven, the WiseTex software is used to describe the textile geometry [4] and the FlowTex software to calculate the permeability of the textile [5]. Instead of trying to eliminate the variability of permeability while performing measurements, the idea of this paper is to take it into account in the flow modeling through a stochastic approach.

Concerning Viscosity

Next to the thermoset materials which have been used for quite some time with the RTM process interest in using thermoplastic materials is growing. Compared to thermoset materials, they have a higher impact resistance and allow higher deformations. Thermoplastic materials can't be used in their natural form together with the RTM technique, as their melt viscosity is much too high. A possibility to lower this viscosity is by using the "in-situ" polymerization process. The polymer is only formed if the basic material (pre-polymer or oligomer) is injected into the mould. Polymer materials that can be processed in this way are PA, PET and PBT. For those materials, the processing route followed is of paramount importance for the final mechanical properties of the produced parts. Parton [6] showed that processing the CBT® material together with the catalyst has to be done at 190°C to obtain the best mechanical properties. If this temperature value is used, there is only 5 minutes left to inject the material into the mould. After 5 minutes, the viscosity has passed the 1 Pa·s level.

If one wants to use the RTM process to inject thermoplastic material into a textile reinforcement, one must deal with a large scatter of textile properties together with a short time window to inject the material. This statement raises the topic of this paper, which aims to take into account the scatter of material properties in mould filling simulations [7].

DETERMINATION OF THE SCATTER FOR DIFFERENT MATERIALS

Variations in Textile Geometry

As indicated in the conclusions of Hoes [1], parts of the large scatter for the textile permeability can be explained by the local variations in the textile geometry. This is first part of this investigation. Different measurement techniques were compared to measure the internal geometry of 3D textile reinforcement. As a result, the X-ray µCT technique is the most useful [8]. With this technique, it is possible to measure the internal geometry without too much preparation. The result of this investigation showed that the spacing between two yarns can have a coefficient of variation up to 5% and the dimensions of the yarns have a coefficient of variation of 15%. If both variations are considered together, a variation for the gap can be found. For this particular 3D woven textile, the coefficient of variation for the gap dimensions is 81%.

Variations of Permeability

To identify this variation, several situations were considered. First of all, only the scatter for the spacing and yarn width were considered for a 3D woven textile. To obtain the permeability values, the Monte Carlo method was used based on 100 textile models built with WiseTex software. This resulted in a coefficient of variation of 28% for the permeability. Next, measurements were performed on a 2D woven textile for the gap width. This resulted in a coefficient of variation of 37%. As permeability is linked in a squared way to dimensions, the coefficient of variation would be 74% for the meso scale permeability.

Variations of Viscosity

If the in-situ polymerization process is applied with RTM, then there is only a small time window to inject the material into the textile reinforcement. This time window is defined between the time the catalyst is added to the oligomer and the time where the resin viscosity is larger than 1 Pa·s. To measure the viscosity behavior, this also raised some problems. Before the measurement could be started with the plate plate setup, 100 seconds were passed to stir the two components and to load the material in a proper way into the rheometer. To have an initial viscosity value, the oligomer viscosity was measured using a concentric rheometer. It is assumed that adding the catalyst doesn't have any influence on the viscosity during the first seconds it is added. The lacking information between 0 s and 100 s is approximated by a linear behavior of the viscosity between the measurement for the oligomer viscosity and the first measured value of the reacting material. Based on the experiments, a coefficient of variation up to 6% is found for the time to reach 1 Pa·s.

Correlated Permeability Distribution

Assigning a permeability value to a sub zone of the model can't be done in a random way without correlation. The resulting scatter is depending on the relative size of the sub zone to the whole model size. To avoid this problem, one needs to use a correlated permeability distribution along the three dimensions. To define this correlation, a simple exponential function has been used (Eqn. 2). The distance used is measured from the center of gravity of the two sub-zones for which the permeability has to be assigned. Next to the distance, there is the correlation length A. If A is large, a strong correlation exists for the permeability along a certain direction resulting in a low gradient.

$$R(\text{distance}) = \exp\left(-\frac{\text{distance}}{\text{A}}\right) \tag{2}$$

Measuring the Correlation Length for Textile Reinforcement Properties

Several strategies were tried to characterize this value. First of all, data for the channel width between two neighboring yarns was used for a 2D woven textile. From this measurement, a correlation length for the gap width of 10 mm was found. Using this information, new textile models were built with the WiseTex software taking into account a correlation length of 10mm for the gap width. Those models were solved with FlowTex to find the permeability values and its correlation length. An overview of the correlated gap width and the corresponding permeability values is given in Fig. 2. The correlation length found for the permeability is 5 mm. The correlation length for permeability is half the correlation length for the geometrical parameters.

Software Implementation

Once the variation is know for both the permeability values in the different directions together with the variation on the viscosity, all this information has to be used within a simulation tool. To obtain stochastic information with a simulation tool, the Monte Carlo principle was used. To do so, a pre and post-processing software has been developed around the commercially available PAM-RTM™ software. With this new development, it is possible to generate a correlated permeability distribution together with a randomly chosen viscosity behavior appropriate to the processing conditions. To generate a correlated permeability field, the covariance decomposition algorithm has been used. To perform stochastic simulations, one can setup the simulations based on a deterministic model. The mesh for such a model can be built with several commercially available finite element models. The universal file format was used as exchange format between the different software packages. The developed post-processing program allows generating additional results based on the standard output of the PAM-RTM™ program. For each element in the mesh, it is possible to calculate the average filling time, the standard deviation together with the coefficient of variation. Another interesting result is the possibility to have an air entrapment for a certain element. Using this extra information, it is possible to control a designed production process and see how sensitive it is for the material parameters.

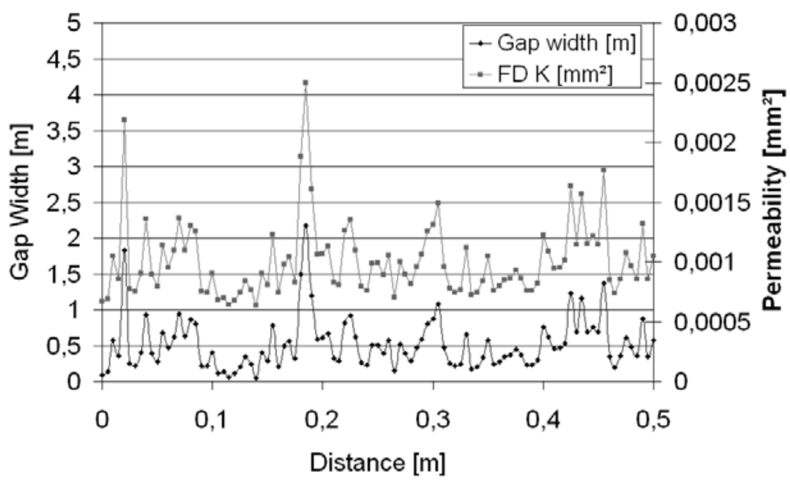

Fig. 2 Correlated gap width and corresponding permeability value.

EXAMPLES

Comparison with Experimental Values

Characterizing the permeability for textile reinforcement is most of the time done using experimental setups. A lot of practical problems arise to find identical values for an identical textile with similar boundary conditions. Due to this reason, next to the average value, also the standard deviation has to be defined. Measurements for permeability always are performed on macro scale. It is not possible to perform permeability measurements on unit cell level (mesoscopic scale). Hoes built a setup to measure permeability in a fast way based on sensor information. For a 2D woven textile R420 from Syncoglass, more than 80 measurements were performed which resulted in a coefficient of variation of 20%. Using the developed strategies, it is possible to investigate this experimental data using modeling. Starting from the measured variation in the textile geometry, permeability values can be assigned on mesoscopic scale level using an appropriate correlation. Based on the generated filling patterns using the Monte Carlo approach, it is possible to extract the permeability along a certain direction. This can be done using the sensor approach or by fitting of ellipses through the different flow fronts as function of time. Based on the data valid for a similar textile as used by Hoes, a coefficient of variation of 15% is found for the macro scale permeability.

Influence of the Most Important Parameters on Simulation Results

To identify the data for the correlated permeability distribution, there is up to now no exact technique available. For that reason, the influence of the value for the mesoscopic scale variation has been investigated. The influence of the correlation distance together with the influence of the coefficient of variation on meso scale for the permeability is displayed in Fig. 3. To end up with a macro scale coefficient of variation of 30%, one needs a coefficient of variation of 120% on meso scale in combination with a correlation distance of 5mm. If the correlation distance is 10 mm, still 80% is needed for the mesoscopic scale coefficient of variation. Using the developed program, one also has to pay attention that the number of sub-zones is large enough. The correlation length has to be larger than one third of the sub-zone dimensions. The mesh also has to contain enough elements, and each sub zone must contain at least 4 elements.

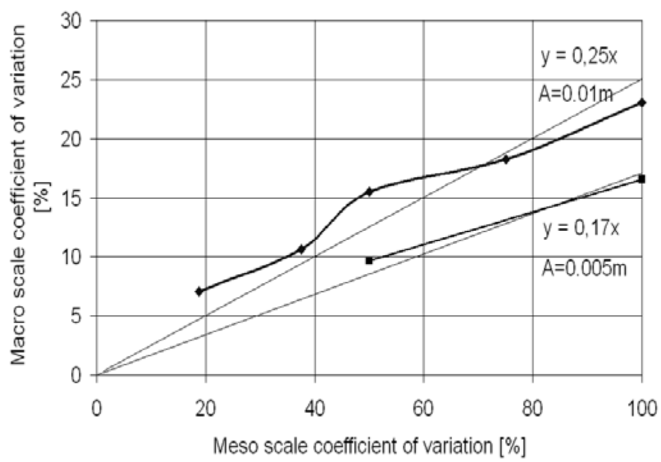

Fig. 3 Influence of the correlation distance and meso scale coefficient of variation.

Example with race tracking

With industrial applications of RTM, one often has to deal with race tracking along the boundaries of the mould due to low degree of fitting of the textile resulting in preferential flows. This can result in unwanted filling patterns possible causing air entrapments. If air entrapments are present, most of the times, the produced part has to be thrown away. Due to this investigation, it is also possible to implement a variation for the channel width and observe the influence of this variation on the complete mould filling. The geometry together with the boundary conditions is shown in Fig. 4.

If an average channel width of 0.5mm is considered, then no air entrapment would be found using a deterministic simulation. The filling pattern is shown in Fig. 5 left. If variation for the different parameters is taken into account, in 10% of the cases there would be air entrapments. In 2% of the cases, there even would be an air entrapment in the middle of the part (Fig. 5

right). This kind of simulation shows that implementing a stochastic approach to model the important parameters can result in relevant additional information for a particular process.

CONCLUSIONS

While mould filling simulation tools for RTM already exists for more than 10 years, it is still possible to improve the possibilities helping the people in real live production. Nowadays, a mould filling simulation package was used to get an idea about the filling pattern. One was aware about the large scatter which is present for the permeability values and the consequences for the RTM process.

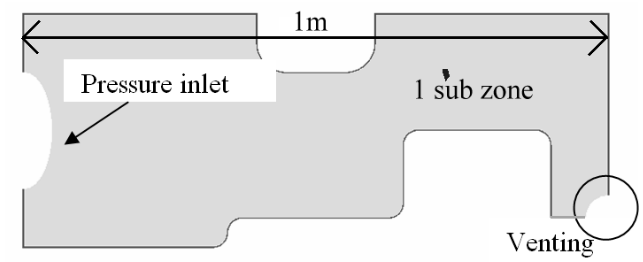

Fig. 4 Model with race tracking.

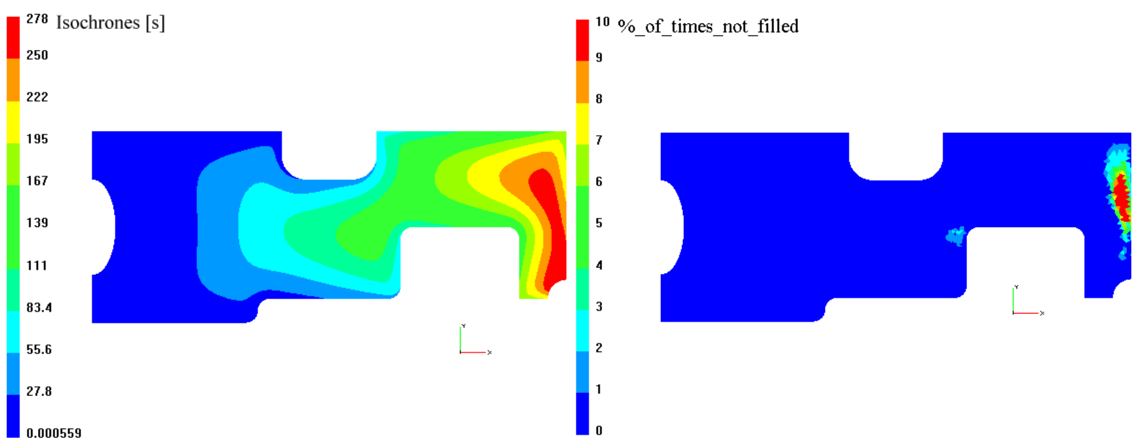

Fig. 5 Filling pattern in case of a deterministic study no air entrapments (left).
Possibility to have an air entrapment in case of a geometry sensitive to race tracking (right).

If one only performs a deterministic simulation, one has to accept that large differences between simulations and experiments can occur. It is only recently that one tries to take into account the scatter for the different parameters within the software's. This paper shows the different steps to realize this:

- Characterization of the internal geometry of textile reinforcement. From this investigation, it seems that the X-ray µCT is the most promising technique for 3D textiles. With this technique, a coefficient of variation of 5 percent was found for the spacing within a 3D textile in combination with a coefficient of variation of 15% for the yarn dimensions. This finally resulted in a coefficient of variation of 80% for the gap dimension.
- Trying to find the scatter for the permeability on mesoscopic scale. Several techniques have been tried together with the definition and use of the correlation distance. Depending

on the technique, a meso scale coefficient of variation of 75% was found in combination with a correlation distance of 5 mm

- Measuring the scatter for the viscosity for a thermoplastic resin. If the in-situ polymerization technique is applied, the process window is really small. For that reason, the smallest deviation on this time window can be of big importance. A technique was developed to measure the viscosity of these fast reacting systems. The coefficient of variation for the time to reach 1Pa·s is 6%.
- Implementation of the stochastic by means of a pre and post processing software around an existing mould filling simulation package. Applying the Monte Carlo technique allowed to generate the necessary and additional data.

With the current status of the investigation, it is already possible to perform stochastic simulations. Measuring strategies are developed to characterize the most important parameters. The results obtained for the scatter for the different parameters can be used in a fully automated way based on the Monte-Carlo principle with PAM-RTM^{TM}. This allows performing flow simulations that correspond better to reality.

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