

# Towards a Holistic Approach for an Efficient Determination of the Rheological Behavior of Sheet Molding Compounds for Process Simulation

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**Abstract.** Sheet Molding Compound (SMC) materials offer attractive specific strength and stiffness properties. With an addition of short cycle times, the possibility for complex geometries and a high recycling potential, this material is a promising solution for the manufacture of lightweight components of numerous industrial sectors. Accurate and reliable simulation can contribute to a fast and competitive development process and effectively reduce the time-to-market. Especially, the accuracy of the simulative filling process is crucial to be able to predict the mechanical properties and hence, pave the way to structural parts. An appropriate characterization of the rheological behavior is therefore, crucial for accurate simulation results. To this purpose bar flow tests are performed and the experimental results are compared to the numerical results. A sensitivity analysis is performed to investigate the influence of each parameter on the simulation outcome. Furthermore, guidelines to improve the simulation accuracy and the testing equipment are derived. In the testing method some improvement potential was identified and the necessity for a testing method closer to the compression process could be established.

## Introduction

For several decades SMC has been widely adopted for non-structural parts especially in the automotive industry for its advantages of high stiffness-to-weight ratio, very short cycle times and its recycling potential [1]. In the last years its use has been broadened to include semi- and structural parts, like rims or e-bike break handles. The material consists in about 2.5 mm thick sheets with typically 25 to 50 mm long fibers in evenly distributed bundles of 3k to 50k fibers. The sheets are impregnated with a thermoset resin system with a number of additives and fillers, mixed for a specific application. For the compression step the sheets are cut to a desired geometry, stacked and placed into a heated mold. The press applies a specific pressure until the part is completely filled and cured, normally in a range from 3 to 10 minutes before the part can be demolded [2]. Due to the flowability of the material, very complex parts can be produced, representing one of the main advantages of the material compared to many other fiber-reinforced parts. During the compression, the fibers move depending on the flow pattern, changing orientation, bending, spreading and also entangling. Defects, such as unfilled areas or weld lines can occur during the flow, reducing the strength and stiffness significantly [3,4]. These defects depend on the flow direction and velocity, the shape of the part, and different process configurations, like the orientation of the part in the tool and the charge placement. The resulting mechanical properties are directly dependent on the defects in the part and the final orientation of the fibers. Therefore, one of the major challenges is a relatively long trial and error procedure to obtain the optimal part geometry, the respective tooling and the necessary process

parameters for the ideal fiber orientation and to avoid flow-induced defects. The more complex a part is, the more it is crucial to predict the flow pattern and consequently resulting defects and the final fiber orientation. Especially for structural parts it is necessary to reach the required mechanical properties in every part, which means that the process has to be reproducible in order to avoid lower local load bearing characteristics. Process simulations offer a strong tool to reduce the time and cost of long trial and error approaches as they make the prediction of flow induced defects and the final fiber orientation possible. However, a simulation always represents a compromise between computation time and the level of detail. Especially when considering a material as complicated and variate as SMC, simplifications in the representation of the reality are compulsory and are found in all commercially available SMC process simulation tools. Consequently, numerical and experimental results of an identical setup will not be the same [5]. To reduce the difference between simulation and reality an appropriate characterization of the rheological behavior is necessary in order to identify the parameters for the viscosity model and the curing kinetics for each material.

### Rheological Description of SMC in Process Simulation

Several rheological descriptions for SMC process simulations have been created, aiming for a balance between the level of detail and the subsequent computation time. Many models were developed focusing on one specific factor, such as the influence of fiber knots, fiber length, voids or fillers [6]. However, the functional dependency of the viscosity of a thermosetting compound without considering the influence of the fibers is generally regarded as [7,8]:

$$\eta = f(T, \dot{\gamma}, \alpha) \quad (1)$$

with  $T$  being the temperature,  $\dot{\gamma}$  the shear rate and  $\alpha$  the degree of curing.

Several rheological models describe this dependency, but a very common industrial standard also implemented in the commercially available FE software 3DTimon by Toray Engineering Co., Ltd. [9] and Moldflow by Autodesk [10] is the combination of the Andrade, the Castro-Macosko and the Cross models [8]:

- temperature-dependent Andrade model

$$\eta_0 = a * e^{-b/T} * T \quad (2)$$

- curing reaction rate-dependent Castro-Macosko model [11,12]

$$\eta_1 = \eta_0 * \left( \frac{\alpha_{gel}}{\alpha_{gel} - \alpha} \right)^{D+E\alpha} \quad (3)$$

- shear rate-dependent Cross model

$$\eta = \frac{\eta_1}{1 + \left( \frac{\eta_1 \dot{\gamma}}{\tau^*} \right)^{1-n}} \quad (4)$$

where  $T$  is the temperature,  $\alpha$  is the degree of curing,  $\alpha_{gel}$  is the gelation point of the material,  $a, b, \tau^*, n, D$  and  $E$  are material specific constants. In the numerical description of the degree of curing, several factors can be and have been taken into account in the past years, like fillers or additives [13–15]. An industrial standard is the Kamal equation, which is applied in different commercially available software solutions, such as 3DTimon by Toray Engineering Co., Ltd. and Cadpress by The Madison Group [9,2]. The Kamal equation was developed specifically for epoxy

and polyester resin, not considering the above mentioned aspects, but showing a reasonable accuracy in the simulated data compared to the experimental data [16]:

$$\frac{d\alpha}{dt} = (A_1 e^{-E_1/T} + A_2 e^{-E_2/T} \alpha^m) * (1 - \alpha)^n \quad (5)$$

T is the temperature,  $\alpha$  is the degree of curing,  $A_1, A_2, E_1, E_2$  are material specific constants to describe the Arrhenius temperature dependency,  $m$  and  $n$  are kinetic exponents.

**Determination of parameters for process simulation.** To obtain the parameters necessary to describe the rheological behavior of a specific material, the equations have to be fitted to curves obtained with experimental data. In a previous study this procedure was performed for the curing kinetics, developing an approach allowing a fast and accurate determination of the Kamal parameters [17]. For the viscosity model the same procedure is the current state of the art. A flow test has to be performed and the experimental curve fitted to the model. However, as the SMC material is highly variable and the fibers have an influence on the rheological behavior, which is not included in the mathematical description in commercially available simulation tools, the obtained parameter sets for both the curing kinetics and the rheological behavior have to be calibrated. In a previous study squeeze flow tests were performed to obtain the first set of viscosity parameters [18]. A first calibration was also performed for the in plane flow by reproducing the tests in simulation together with a sensitivity analysis for the parameters. However, only the relatively simple in plane flow has been calibrated and more complex flow patterns may not be represented appropriately. Therefore, a complex flow test method has to be selected, performed and reproduced in simulation to evaluate the simulation outcome. A guidelines for future viscosity characterizations and related advice shall be derived within this study.

### Bar Flow Tests

For the second calibration step a complex flow test with out of plane flow is necessary. Previous research documented spiral flow tests or bar flow tests [19]. Also squeeze flow tests with a different sheet stacking method could be used, as well as any cavity geometry forcing the material to flow in a direction that is not the same as the sheet plane. Bar flow tests were selected for this study, as the cavity can be easily transferred to simulation and previous research were available.

**Setup.** For any SMC process, it is necessary to have a force and heat generation principle, as well as a tool for the material to fill. The general setup in CAD is shown in Fig. 1a [20]. A pneumatic actuator on the top of the setup moves a piston into the cavity for the charge placement. Below the charge holding opening is the tool, which consists in a channel with a length of 450 mm, a width of 30 mm and a thickness of 2,5 mm. For the heat generation an additional steel block is placed below the tool, which contains heating cartridges. To track and collect the necessary data seven temperature sensors are applied on the top mold along the channel and a linear potentiometer tracks the piston displacement during the compression. Fig. 1b shows the raw material SMC specimens before the testing. They consist in 55 mm wide long sheet strips. The material used was Astar CARBKID PGK5250-R63 [21]. The aim was to put as much material as possible into the charge holder, making a comparison with simulation easier. With a maximum diameter of the opening of 30 mm the length of several specimens had to be slightly adapted as the thickness of SMC sheets varies.

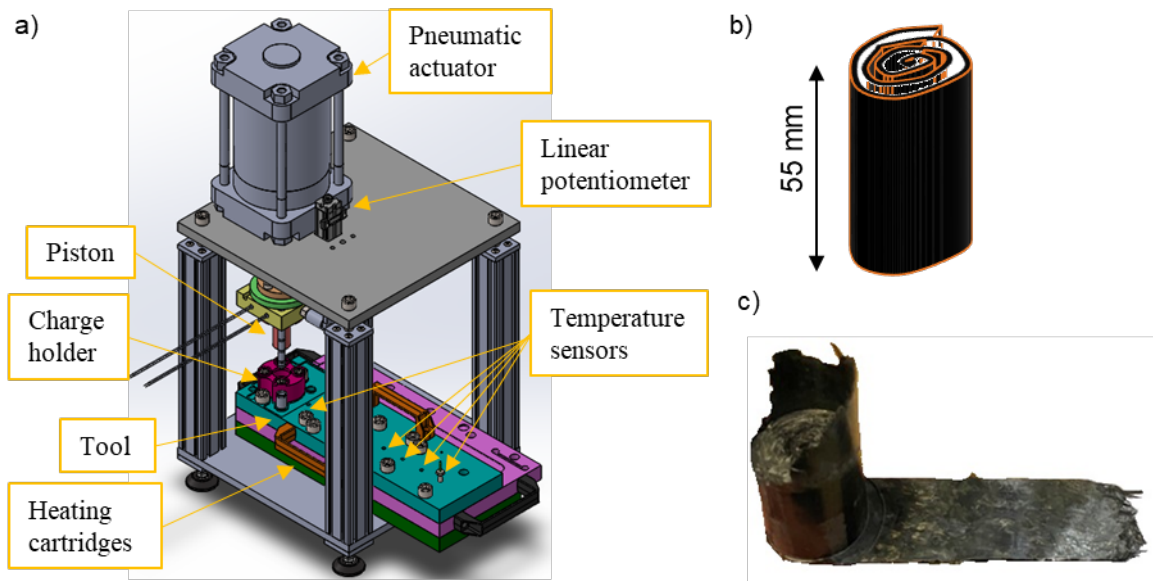


Figure 1: a) Bar flow test setup in CAD [20]; b) rolled up raw SMC material specimen as prepared for the testing; c) cured SMC specimen after test completion

In Fig. 1c a fully cured specimen at the end of the test can be seen. Usually the material cures before filling the whole channel, which allows conclusions about the flow speed at specific temperatures.

**Process parameters.** The applied testing parameters are summarized in Table 1. The pneumatic actuator was permanently set to the maximum compression pressure of 9 bar, which results in a generally low force. In a conventional SMC process the press closing speed is constant and around 1-3 mm/sec, with the force adapting. For the bar flow test bench the compression is force controlled, leading to variations in the closing speed. Based on the recommendations from [20], a test was performed when all the temperature sensors showed a relatively stable value within a 4 °C difference. The two tested temperature intervals were selected based on the material used, which cures at relatively low temperatures [21]. Therefore, the first tests were performed at 130-134 °C as suggested in [20] and the second interval was chosen to be 126-130 °C in order to give the material more time to cure and therefore, have a longer flow path.

Table 1: Process parameters applied for the bar flow tests

Process parameter	
Temperature intervals	126 – 130 [°C] 130 – 134 [°C]
Pressure	9 [bar]
Material batches	2
Repetitions	5

Two different batches of the same material were tested, both stored at -21°C, as suggested by the manufacturer, one being one year old, the second one being nine months old. The first material to be tested was the fresher material, which was out of the freezer four days before the actual testing. The older material batch was taken out of the freezer only one day before the testing. For every temperature interval five tests were performed, following previously set recommendations with regard to SMC due to the variability of the material resulting in a broad range of flow behaviors.

As the material cures very fast once the material is in the charge holder, the compression has to start immediately. Therefore, the data recording has to be started, then the specimen placed into the charge holder and finally the compression has to be started. To provide a basis for the comparison between the tests, the time between the start of the recording until the start of the compression was

set to be 10 seconds. From the start of the recording until drawing the piston back up five minutes passed, to ensure that the curing of the material had completed. Afterwards the top half of the mold was unscrewed and the specimen as shown in Fig. 1c removed before closing the mold again in order for it to heat up to the desired temperature range for the next test.

The raw data obtained during the tests include the temperature values recorded by the seven temperature sensors along the flow channel, as well as the tension recorded by the linear potentiometer. In order to compare the experimental results to the simulation, the piston position during the compression has to be derived with a linear dependency. Two points with known tension and position are necessary to obtain the constants  $m$  and  $c$ . In this study the values that can be used are the first contact of the piston with the material, which corresponds to the width of the SMC strips of 55 mm for the piston position and for the tension it can be identified in the resulting curve as the tension value abruptly stops its fast and linear decrease. The second point to be considered is the last data set of the experiment, which has a corresponding tension value and the thickness, representing the final piston position can be measured on the final specimen.

**Simulation.** The bar flow tests were implemented into the FE simulation software 3DTimon. The temperatures of the upper and lower mold were set to 128 °C and 132 °C, corresponding to the intermediate temperatures of the tested temperature ranges. The charge was placed in the cylinder with the average volume of the tested SMC of 35000 mm<sup>3</sup>. As the experiment, the simulation was performed with constant force, set at 636,173 N corresponding to the pressure of 9 bar applied on the surface of the specimen in the cylinder of 30 mm in diameter. For the meshing method Euler elements were selected, with an edge length of 4 mm and ratio 1. Given the reduced size of the specimen and the limitation to flow simulations not considering the fibers separately nor residual stresses the compilation time remains reasonable despite the high number of elements. The closing speed before the contact with the material was set to 11 mm/s, as the value measured during the experiments was 10,76 mm/s and 3DTimon only allows the use of math. integers. The temperature of the material at the beginning of the process was set at the respective mold temperature as an average of 22 seconds passes before the piston touches the material starting the actual compression and therefore the material has time to already heat up. The material parameters adopted as basis for the simulations were obtained from the previous studies [17,18].

## Results and Discussion

A first evaluation was pursued by selecting the material batch to be considered in the subsequent step. In Fig. 2 the relevant results for two tests in the interval of 130-134 °C, one for each batch are shown. The temperature sensors were numbered in the order of possible contact with the material, starting with 1 for the closest to the charge holder and ending with 7 for the furthest. For better overview only the relevant signals are shown in the diagrams in Fig. 2.

As can be seen in Fig. 2a the first material shows a visible temperature increase of 1 °C only for sensor 1 at around 85 s. Sensor 2 only displays a very small increase around the same time, sensor 3 did not record any disturbance. This means that the material did not reach sensor 3 in the flow and barely reached sensor 2. The increase in temperatures signifies a curing reaction happening, which is exothermal. However, compared to Fig. 2b the temperature increase is only half, signifying that the curing of the material, which steadily progresses at room temperature, had already progressed significantly. In Fig. 2b even sensor 2 shows a curing reaction at around 75 s. In this test, sensor 3 only shows a small increase in temperature, whereas sensor 4 was not reached. The signal of the linear potentiometer also confirmed the reduced flow and therefore, piston movement, by showing a smaller tension difference over the whole compression cycle. Therefore, the second material that was produced earlier, but defrosted only one day before the tests had a more significant curing reaction and flew further. Given the already far proceeded curing of the first batch, it was excluded from further evaluations, as the evaluation has to be performed for material with a comparable stage, as it has an impact on the resulting viscosity and curing kinetics. For future evaluations the time of the material outside the freezer has to be reduced as much as possible, while still ensuring full defrosting

and the production date of the SMC should be as recent as possible in order to ensure the best comparability.

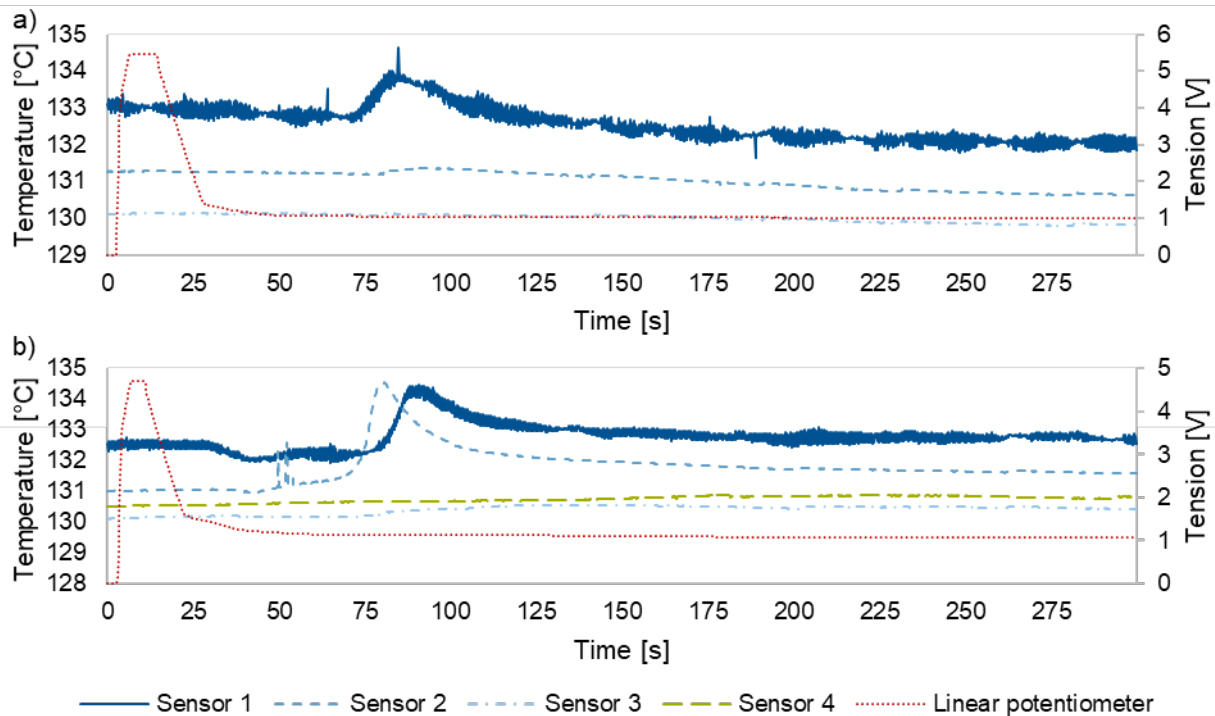


Figure 2 Bar flow test results for the interval 130-134 °C for a) the material from March 2020, out of the freezer for four days, b) the material from November 2019, out of the freezer for one day

The result of the piston displacement for each experiment was obtained from the tension data from the linear potentiometer based on the analysis presented above. The curves can be seen for the tests at the temperature range 126-130 °C in Fig. 3a and for the range 130-134 °C in Fig. 3b.

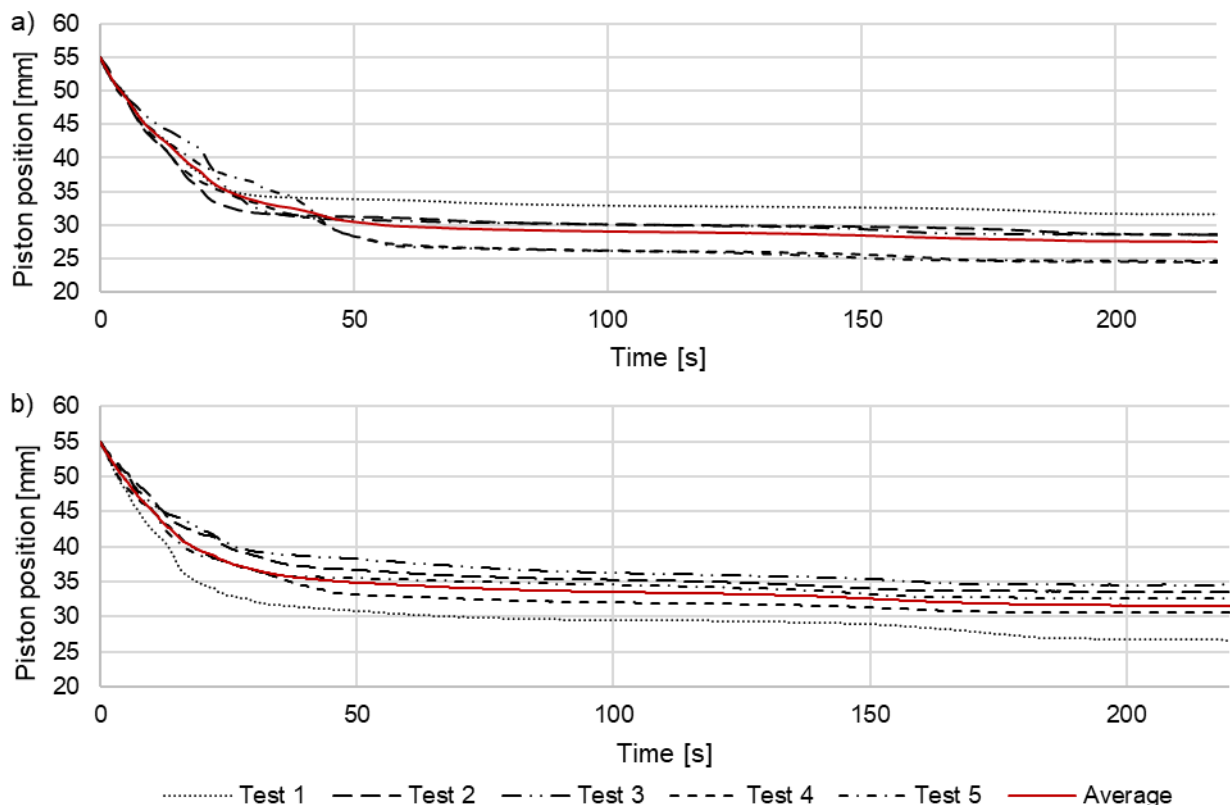


Figure 3 Piston displacement resulting from the data collected from the linear potentiometer for the tests performed in the temperature range a) 126-130 °C and b) 130-134 °C

According to a previously derived guideline for SMC in testing, given the extremely high variability at least five tests should be performed for each setting. The high variability is also visible here, as each curve displays a unique development. Typically the material heats up decreasing the viscosity, which allows the material to flow faster. Once the material is cured, the piston cannot move further. Therefore, most tests show a fast descent of the piston at the beginning, which then steadily slows down until plateauing around 50 seconds. However, in Fig. 3a tests 3 and 5 show two steps where the piston slows down. This could be due to compaction steps within partially cured material, to local defects in the raw material, but also to the setup itself. To develop a material model that is the best representative of all of these possible behaviors and influences it is necessary to build an average of the tests, represented as a line in Fig. 3a and Fig. 3b. It can be seen, that the average for 128 °C shows the piston stopping around 28 mm, while for 132 °C it stops around 32 mm. This is due to the material curing much faster for the higher temperature.

The comparison of the average for the two temperature ranges with the respective simulation with the material parameters obtained from previous studies can be seen in Fig. 4. For both Fig 4a for 128 °C and Fig. 4b for 132 °C a similar behavior in both the experiment and the simulation can be observed. In all data the piston movement is fast until around 50 s and then slows down significantly, which means the curing is completed around the same time for the simulations and the experiment at 128 °C. For 132 °C on the other hand the curing is complete earlier in the experiment than in the simulation. However, in both cases the piston movement in simulation is much slower than in the experiments, which is mainly due to the viscosity being lower in reality. Once most of the material is cured the lowering of the piston happens with comparable speed for both temperatures.

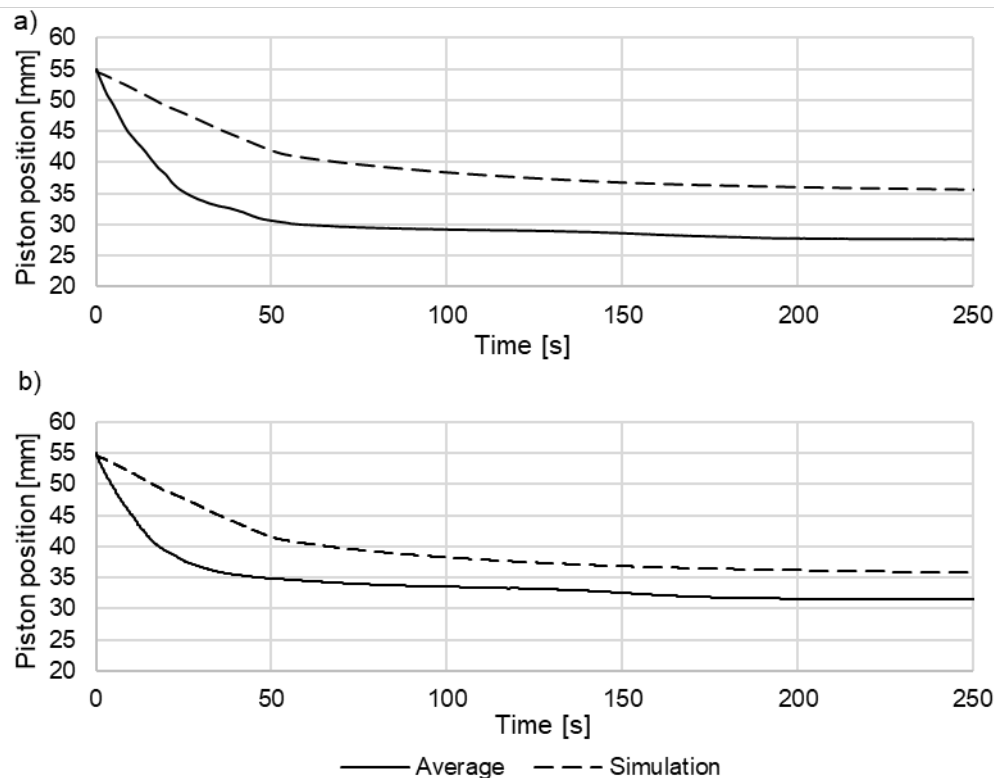


Figure 4 Comparison of simulation result with the average of the experimental results for a) 128 °C and b) 132 °C

The change in the piston speed is also more gradual in the experiments than in the simulation and for 132 °C, a difference to be addressed by changing the viscosity parameters. Furthermore, it can be seen that as in the experiments in simulation higher temperature corresponds to faster curing and therefore, less movement of the piston. However, the difference is extremely small in the numerical result. Despite several aspects to be comparable between reality and simulation, it has to be kept in mind that the temperature control, and the signal of the temperature sensors was not optimal. Therefore, it is also not exactly known what the temperature of the material was at the start of the

compression, nor what the exact temperature of the upper and lower mold were during the tests. Small adaptations, like the charge temperature or the mold temperature have an influence on the simulation outcome. It is therefore, crucial to have testing results that show these values together with the actual force applied on the piston. It also needs to be kept in mind, that the standard simulation runs are speed-controlled, whereas in this case, due to the test equipment the option of force-control was selected. Therefore, a speed-controlled testing equipment could improve the simulation outcome.

As mentioned, a few discrepancies between the above shown curves are related to the viscosity parameters. Therefore, a sensitivity analysis was performed to derive guidelines about each parameter to improve the simulation outcome. Each parameter of the curing kinetics and viscosity was set once to the upper and once to the lower limit in the material card while keeping all other parameters unchanged from the reference. The piston position for each simulation was compared to the reference simulation as shown in Fig. 5a for  $n$  curing and Fig. 5b for the kinetic exponent  $m$ .

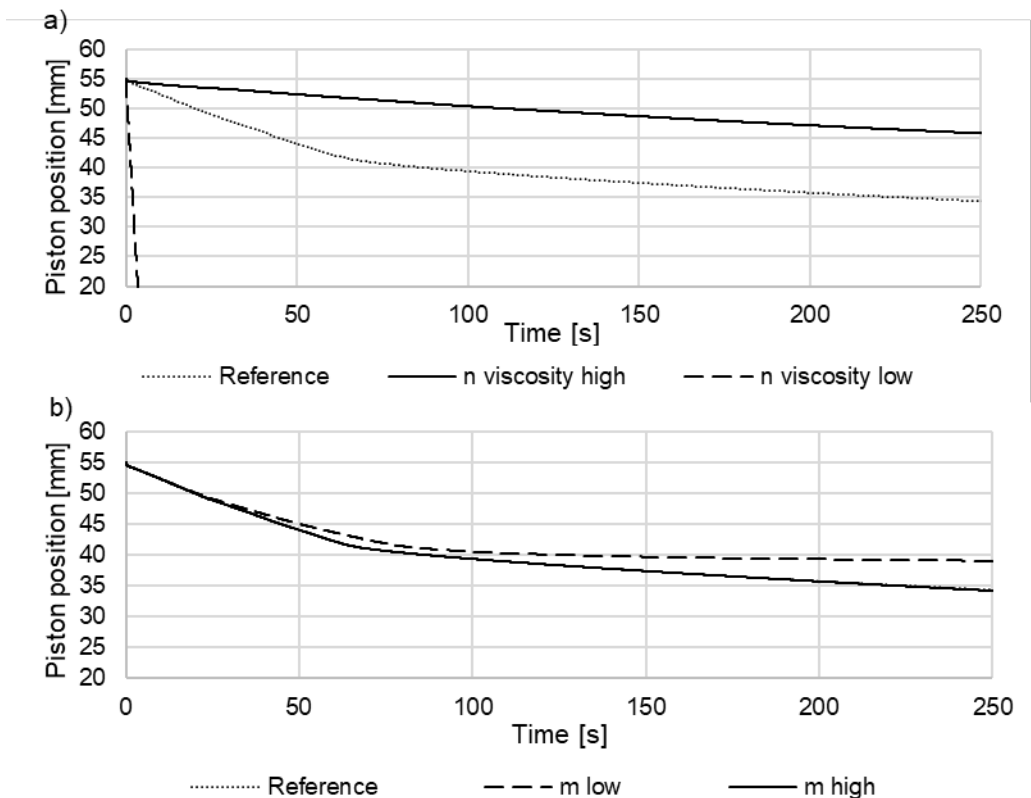


Figure 5 Results for upper and lower limit in the sensitivity analysis for a)  $n$  curing and b)  $m$

From the comparison of these curves to the reference the influence of each parameter could be identified and a guideline about an increase or decrease in the parameter can be derived. These are summarized in Table 2.



Table 2 Sensitivity analysis of the curing parameters for the compression speed in SMC flow tests

	Parameter	Compression speed with lower limit	Compression speed with upper limit
Curing kinetics	m	same beginning earlier, but more steady curing	unchanged
	n	unchanged	unchanged
	A1	unchanged	unchanged
	A2	unchanged	earlier and steadier curing
	E1	short shot	unchanged
	E2	same beginning much faster decrease	minimal decrease towards end
Viscosity	n	constant and slower	almost maximum
	tau	constant and slower	almost maximum
	a	constant and slower	almost maximum
	b	short shot	almost maximum
	D	constant and slower	maximum
	E	minimal increase towards end	unchanged

Based on these findings, for this bar flow case the accuracy of the simulation can be improved by decreasing  $\tau$ ,  $a$  or  $D$ . However, given the above mentioned limitations in the data collection and consequently, the definition of the simulation case, the uncertainties related to this very simulation case do induce rather significant changes in the simulation outcome. It is therefore, necessary, to improve the test equipment in order avoid all above mentioned issues and obtain data, that can be reliably transferred to process simulation. This means having a test equipment closer to the actual process, allowing for closing speed controlled compression and for a more uniform temperature distribution, avoiding the issues with the heating cartridges and their control system. Furthermore, for a precise comparison it is crucial to have a precise signal of the force applied in order to rebuild it appropriately in simulation.

## Summary

Within this study bar flow tests were performed and evaluated in order to calibrate the out of plane flow behavior of SMC materials in process simulation. The tests, the analysis method as well as the simulation approach are presented together with the comparison of the analyzed data from the tests with the simulation outcome. Furthermore, guidelines are derived based on a sensitivity analysis performed with process simulations for the calibration of the rheological parameters based on the outcome of the comparison and independent from the specific testing method. Several challenging aspects could be identified with regard to the used bar flow setup, among which the differences it has from the actual SMC process, leading to the necessity of developing a new flow test bench.

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