Quantitative analysis of a new SMC generation during compression moulding

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Abstract
This study is dedicated to the characterization of the behaviour of new SMC generation from an instrumented specific device, in conditions representative to those encountered in compression moulding. More specifically, we focused on the couplings between heat transfer, SMC flow and crosslinking through heat flux, temperature and pressure measurements during the whole process. Important industrial parameters were considered: the mould temperature, the deformation rate during the SMC flow and the sample residence time before the beginning of the closure of the mould.
The coupling between the flow and the crosslinking was found to be negligible in the industrial conditions and the self-heating of SMC during the flow was very low. On the contrary, we recorded significant variations of the surface temperature of the mould, which also depended of a thermal contact resistance at the interface between the composite and the mold. The decrease of the SMC deformation rate favoured the coupling between the flow and the curing. A lower mold temperature increased the window of moldability but at the expense of the cycle time.

1. Introduction

Nowadays, the automotive industry works actively to reduce fuel consumption and carbon dioxide emissions. One of the main levers consists in reducing significantly weight of structural parts of vehicles. In this framework, composite materials seem to be an ideal choice in accordance with their potential to combine mechanical resistance and lightness.
Sheet Molding Compounds (SMC) materials, which are mainly constituted of thermoset resin, mineral fillers and reinforced with chopped glass fibers, are already extensively used to manufacture semi-structural parts. In order to design and produce structural parts, new formulations, *e.g.* more reactive and/or containing a higher fiber content, are being developed. SMC compression moulding process imposes strong thermo-mechanical loadings coupled with thermal and chemical phenomena, such as heating, flowing, crosslinking reaction and cooling.
To characterize and deeply understand these couplings during the whole compression process, it is necessary to get quantitative data from mechanical and thermal points of view, with an accurate control of the boundary conditions and process parameters. Several papers deal with the experimental approach of SMC compression molding with *in situ* measurements and especially thermal ones. Such results are then used to improve or to validate numerical models and the associated assumptions [1-3]. However, the characterization of the material behavior in representative conditions of industrial process required the development of specific and instrumented devices. They are designed for compression moulding and these moulds are generally smaller than real ones to facilitate their handling but also installation in laboratory press allowing the accurate control of the force and closing

rates imposed during the test. Their geometries are mainly simples such as rectangular or square shapes [Ref] but sometimes more complex with e.g. different thicknesses, steps [Ref]… There are also developed to enable a simple closing kinematics for in-plane strain compression. Sometimes the flow is avoided to focus only on the curing step [4].
From the instrumentation point of view, these specific moulds are equipped with different types of sensors: thermocouples, pressure, dielectric and ultrasonic sensors. Thermocouples are clearly the most used type of sensor, which are often placed at different locations of the SMC preform before the compression test [1-3,5]. Their instrusivity can lead to several problems: possible evolution of their positions, effects of mechanical stresses (tension, compression…) and size on temperature measurement reliability. As a consequence, many precautions have to be taken to get accurate measurements. Some authors instrument their laboratory mold with pressure sensors as a complementary information [1,3-4]. Judicious locations enable to detect the flow front but also possible pressure heterogeneities during the filling step and the curing reaction. The use of dielectric sensors is rare [4,6-7] and even more for ultrasonic ones [4]. However, it seems that ionic viscosity is a good parameter to qualitatively follow the different steps of the compression molding. It is thus well adapted to on-line production control [4]. Note that the thermal and chemical shrinkages are also important parameters in thermoset composite processing. They are not investigated in this paper but can be accurately determined in real time from an other laboratory device [8], which was used for these new SMC [9].
From the literature review, it appeared that there is almost no accurate and reliable measurements of heat transfer nor fine analysis of the couplings between flow, heat transfer and curing during SMC compression moulding. For this purpose, we proposed a new instrumented bench to quantify all these physics from the preform deposition to the end of moulding cycle, in representative process conditions. We considered two important industrial parameters: the variations of the mould temperature and the deformation rate during the SMC flow.

2. Material and methods

In this work, the considered thermoset prepreg was a new industrial SMC formulation consisting in a high-reactivity polyester-based paste (matrix) and reinforced with hollow glass beads (4,1wt%) to reduce the density and chopped glass fiber content (28 wt%).
The device we developed was a home-made instrumented compression mold (Figure 1) equipped with pressure and heat flux sensors. We also record the force and the displacement of the piston from sensors of the press (Instron ElectroPulse E10000) used for compression forming. Data are collected from the placement of SMC plies in the molding cavity up to the end of curing. This device is able to work under experimental conditions close to industrial ones in terms of temperature (up to 200°C), pressure (4.2 MPa on the initial preform and 1.8 MPa after the mold filling) and strain rate during the SMC flow (mold filling step, up to 0.3 s^{-1}). It can be thus considered as a characterization tool of the material behavior during its processing.
The molding cavity is a channel (140mm x 40mm), initially entirely filled in the width direction, that ensures a planar flow kinematics of the SMC flow. The mold is furthermore designed to have 1D heat transfer through the sample thickness. Sensors have been located in the piston and the bottom of the cavity such that we can record heat transfer (sensor developed in our laboratory) and pressure (Kistler sensor serie 6167A) as soon as the placement of SMC plies and then during compression (detection of the flow front) and curing. Heat flux sensors are denoted PO1, PO2 and PO4 whereas pressure sensors are denoted K-62 to K-64, as depicted in Figure 1.

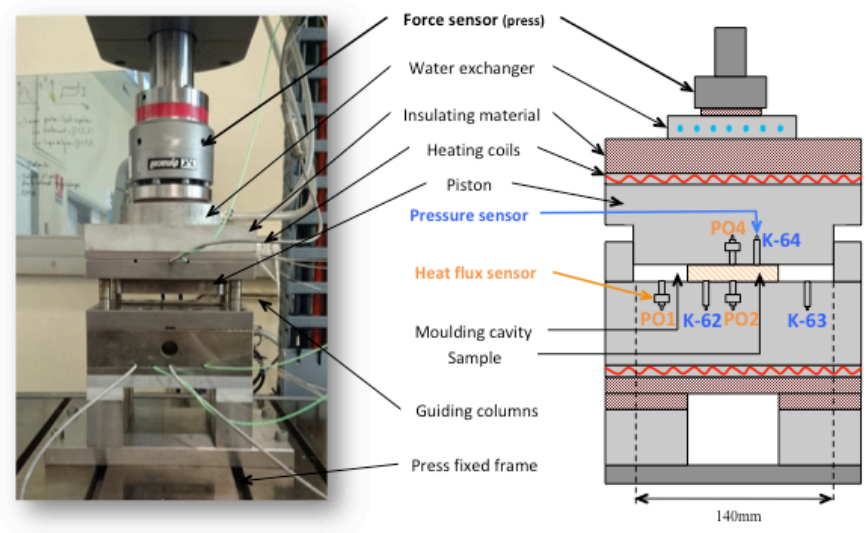

Figure 1: (a) Picture of the instrumented compression mould placed on an Instron ElectroPulse E10000 press. (b) Schematic view of the mould with sensors positions.

The thickness of the sample is initially equal to 6-7mm and finally reaches about 3mm after the complete filling. Heat flux sensors are thermally not intrusive and measure the heat flux exchanged between the mould and the SMC but also the surface temperature of the mould according to a sequential inverse method (see [8]). We thus detected the flow front and calculated the conversion degree of the crosslinking from the integration of the associated heat flux peak. Pressure sensors were used to detect the closure of the mould and the charge loss due to the SMC flow.
The thermal regulation was done with a PID controller connected to heating coils. The control of experimental conditions and data acquisition were managed from a dedicated software developed under LabVIEW® environment. A raw data file was finally exported and treated with a home-made Matlab® script.
Unfortunately the constant strain rate during the SMC flow could not be directly experimentally imposed by the software of the press. As a consequence, the exponential decreasing of the piston position was approximated by successive linear small portions with constant velocities.

3. Results

3.1. Analysis of moulding cycle for typical industrial conditions

Typical industrial parameters are set to quantify heat transfer and pressure variations during SMC compression moulding. The mould was thus regulated at 150°C, the closing rate was first equal to 10mm.s^{-1} until detecting the contact with the SMC. Afterwards, a strain rate of 0.3s^{-1} was applied, corresponding to around 4-5s as filling time. At the end of the flow, a constant force of 9000N was imposed till the end of the experiment (in-mould pressure : 1.64 MPa).
The data provided by heat flux sensors enabled to get the surface temperature of the mould and the parietal heat flux exchanged between the SMC and the mould during the whole forming cycle (Figures 2a and 2b). The temperature evolution showed that the deposition of the cold SMC on the lower part of the mould led to a first temperature drop (sensor PO2, zone 1). The mould started to close (zone 2) and when the piston came into contact with the upper surface of the preform, another strong temperature decrease (sensor PO4, zone 3) was observed. Assuming a perfect thermal contact, the contact temperature can be easily calculated [10] and was found to be equal to 135°C, which is close to the measured value. In the same time, sensor PO2 continued to record lower temperature values due

to a better thermal contact induced by the compression process. During the SMC flow, the material started to be heated and the flow front was detected by PO1 (smaller temperature drop) few seconds after PO4. At the end of the mould filling (zone 4), the surface temperatures smoothly increased until about 45s. This stage was immediately followed by the crosslinking reaction characterized by a temperature overshoot of 3-4°C. After the complete curing, the temperatues tended to come back to a constant value close to the initial one.

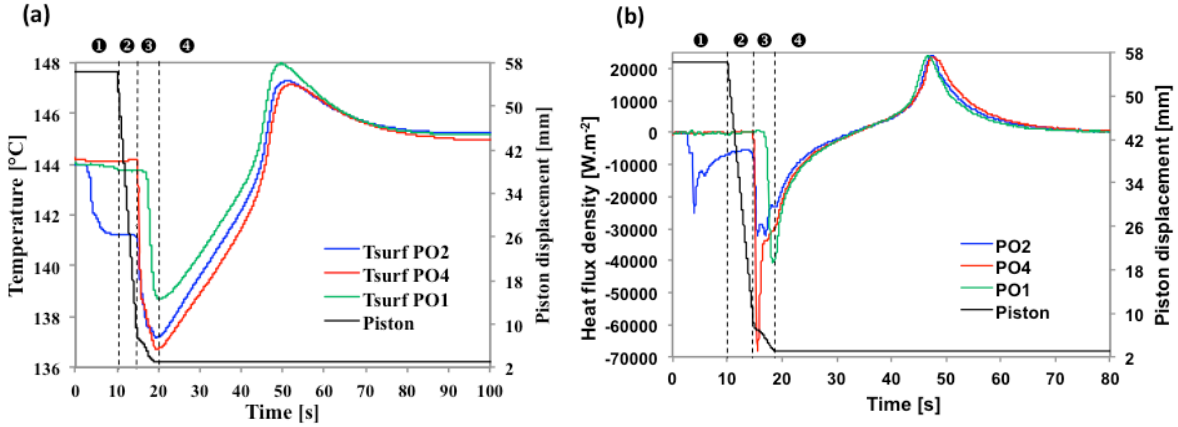

Figure 2: Evolutions of (a) the surface temperatures of the mould and (b) the heat flux densities exchanged between the mould and the sample during the compression test in industrial conditions. The displacement of the piston is also plotted.

Heat flux density curves provided the same kind of information for identifying each step of the process. For these sensors, a negative value indicated an endothermal phenomenon for the SMC, such as the contact between the composite and the mould and the flow front detection. During steps 1-3, heat flux density fluctuations were associated to the evolution of thermal contact resistance. After the filling of the mould, heat flux density tended to come back to zero but it became positive, which indicated an exothermal signal associated to the curing reaction. Note that the three recorded signals reached the same value at 35s, meaning that heat transfer were identical between the mould and SMC. The curves then were almost superimposed during the curing and up to the end of the experiment.

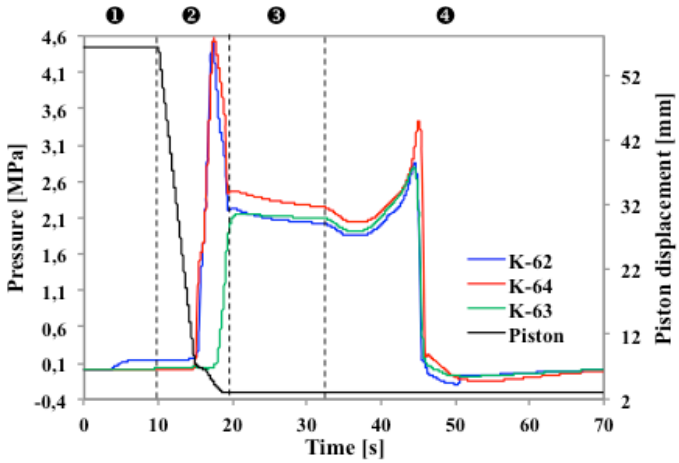

Figure 3: Evolution of the pressures in the moulding cavity during the SMC compression test in industrial conditions. Piston displacement is also displayed.

Such results demonstrated an homogeneity of the reaction through the whole sample: the relatively low filling time limited an heterogeneous distribution of temperature in the preform plane, which ensured a decoupling between the flow and the crosslinking in this case. The mass enthalpy was calculated by integration of the peak in the time range of positive values of the heat flux density; We found 137J.g^{-1}, which was in good agreement with values obtained in previous studies [9].
The pressure curves were also characteristic of the different steps of the compression test. The deposition of the preform on the lower part of the mould induced a small increase of pressure (sensor K-62, zone 1). When the contact between the SMC and the piston occurred, the pressure level for both sensors K-62 and K-64 sharply increased up to a maximum (zone 2). Such behavior was interpreted as the threshold value to enable the composite flow and it was thus followed by a pressure drop, due to the increase of the sample surface during squeezing. During the filling, the flow front was detected by a pressure increase by the sensor K-63. When the applied pressure was constant, the recorded pressures tended to slightly decrease linearly after the mould filling (zone 3), which was attributed to a progressive uniformisation of the stress state induced by the flow. The normal stress tends to p_0=1.8 MPa, which is simply the ratio of the constant applied force versus the total surface. In zone 4, corresponding to the curing phase, one could observe for the three sensors, a small decrease of the pressure level followed by a pressure peak and finally a dramatic drop down to zero.
A detailed analysis demonstrated that the slight pressure decrease matched with the beginning of the chemical shrinkage along the centerline of the sample. The pressure peak was induced by the thermal expansion since the reaction is exothermal. Shrinkage became finally the main phenomenon and led to the rapid pressure decrease. A zero pressure indicated a loss of contact (unsticking of the sample from the piston and matrix surfaces) at the end of curing. This observation was correlated to a higher thickness at the edges compared to the central zone of the sample and was explained by a heterogeneous crosslinking inherent to the thermal history of the SMC during the compression test. In the same way, this spatially heterogeneous curing also explains the information recorded by the pressure sensors that do not always match the average imposed pressure p_0.

3.2. Evaluation of the self-heating during SMC flow

Compaction and flow phases of the preform could potentially lead to the self-heating of the material due to frictions in its volume and at the surface of the mould. One question was thus to quantify this phenomenon to know if it could influence significantly the temperature field in the part.

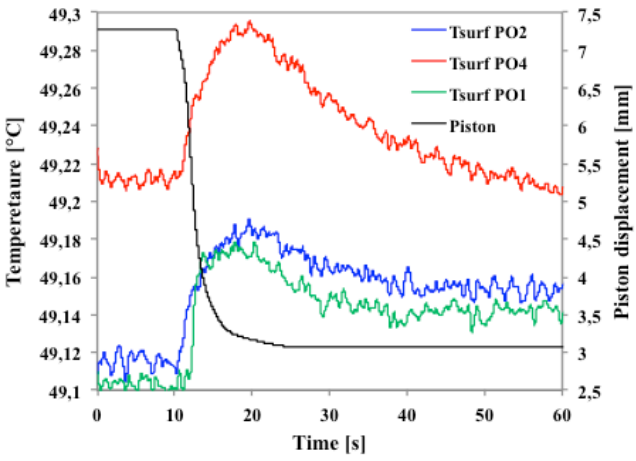

Figure 4: Evolution of the surface temperatures during an isothermal filling test at 50°C. The piston displacement is also displayed.

For this purpose, some tests were carried out in isothermal conditions (50°C and 80°C) for which curing reaction was prevented. In this specific cases, both SMC and mould had the same temperature. The expected strain rate imposed during the piston displacement was $0.3s^{-1}$.
The heating induced by the filling of the flow was recorded with the heat flux sensors, which could also provide the surface temperature. Figure 4 presents these temperatures for the three sensor locations and an isothermal condition of 50°C. Note that the actual strain rate was $0.2s^{-1}$ due to SMC difficulty to be deformed (high viscosity) with respect to the press capability. As expected, compression and flow of SMC generated a temperature increase at the mould surface. However, its value remained small and lower than 0.1°C. Such result was also similar for a moulding condition at 80°C and the self-heating was even lower (around 0.05°C) whereas the strain rate was the initial expected one. As a conclusion, the viscous dissipation term in the conductive heat transfer equation could be neglected.

3.3. Processability window: modification of the deformation rate and appearance of defects

The possible coupling between the flow phase and the curing reaction was driven by several experimental parameters such as the compression strain rate (which imposed the filling time) and the mould temperature. Thus, we aimed to investigate the conditions for which such coupling existed and to evaluate the processability window of this SMC formulation.
As a first step, the mould temperature was kept constant (T=150°C) for all experiments but the strain rate was decreased progressively up to obtain incomplete cured samples. The operating mode was the same as the one depicted in section 3.1. The evolution of heat flux densities recorded by sensors PO2 and PO4 were presented in Figure 5a.

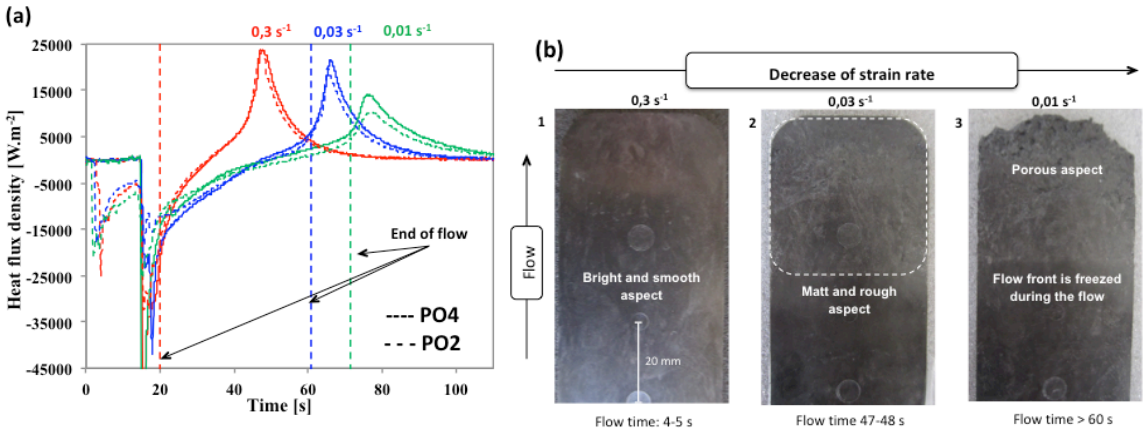

Figure 5: Evolution of the heat flux densities for different compression strain rates, T=150°C; the end of filling is indicated by a vertical dotted line (a); Pictures of SMC plates after compression test (b).

The decrease of the strain rate during SMC compression induced a change of the thermal history of the material. The time range to reach a good thermal contact between the lower part of mould increased and the sample thickness reduction was slowered, leading to an increase of the heating time. As a consequence the reaction was delayed, as shown by the shift of the heat flux peaks. Above $0.055s^{-1}$, the filling of the mould was achieved before the exothermal signal and the peaks recorded by both sensors were identical, suggesting that no coupling existed.
However, from $0.03s^{-1}$ and below, the reaction obviously started before the end of the flow and the amplitude of the exothermal peak recorded by PO2 sensor was smaller, suggesting that a part of the energy released by the crosslinking occurred during the sample heating during its flow. Such results

suggested an increase of the reaction heterogeneity in the part volume and a modification of the flow, which could have some consequence on the part quality and especially on the apparition of defects.
This aspect was confirmed from pictures presented in Figure 6. We could observe that for the highest strain rate, the mould filling was complete and the sample was characterized by smooth and bright surfaces. However, these surfaces became matt and rougher from a compression strain rate equal to $0.055s^{-1}$ and an example was given for $0.03s^{-1}$. Such an observation was generally linked to a lack of pressure during the compression [4] and the beginning of crosslinking during the flow. Decreasing more the strain rate finally led to partial filling and thus uncomplete samples with more holes and even lack of material. The observation of the sample lateral surfaces (i.e. through the thickness) clearly showed the progressive apparition of cracks oriented in the flow direction for low strain rates. We assumed that they were induced by internal stresses generated by the curing of the SMC during its flow.

3.4. Processability window: modification of the mould temperature

We also analyzed the effect of the mould temperature on the SMC behavior and more specifically on the coupling flow/curing. Figure 5b compared the evolution of heat flux density curves recorded by PO4 sensor during isothermal compression moulding at 130°C and 150°C. A first effect of the decrease of the mould temperature was the shift of the beginning of the curing reaction. Compared to experiments carried out at 150°C, the reaction at 130°C was delayed by 15s for a $0.3s^{-1}$ strain rate and by about 30s for a strain rate equal to $0.01s^{-1}$. Such result was explained by a lower rate of inhibitor consumption at a lower temperature. However, another important effect was the decrease of the crosslinking kinetics. As a consequence, a decrease of the moulding temperature enabled to decouple the material flow from the consolidation/curing step, which enhanced the window processability (lower strain rates were available) but to the expense of the cycle time. Moreover, bright and smooth surfaces were observed at T=130°C for compression strain rates down to $0.02s^{-1}$ (filling time of 50s). Defaults on the SMC samples appeared below this value. As an example, we obtained full plates but with material lacks and surface roughness at $0.01s^{-1}$.

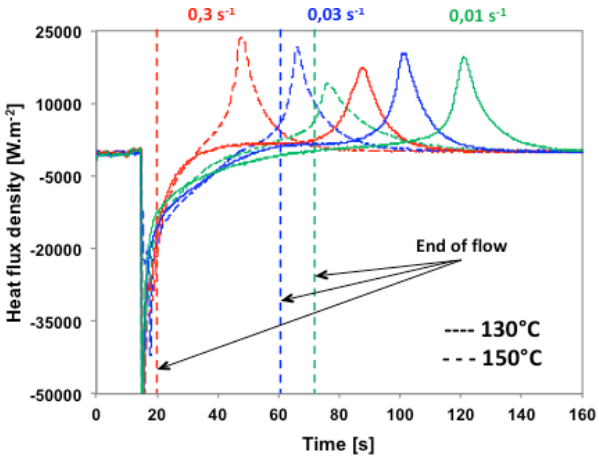

Figure 6: Evolution of the heat flux densities for different compression strain rates, for two different moulding temperatures, 130°C and 150°C. The end of filling is indicated by a vertical dotted line.

4. Conclusions

This work presented the development of a new instrumented device to study the compression moulding of SMC material with a focus on heat transfer and pressure evolution in relevant industrial

conditions. Experimental results demonstrated that the coupling between the curing and the material flow could be neglected under typical industrial conditions and the increase of temperature induced by viscous dissipation during the flow was negligible. Heat flux sensors were very useful to quantify the variations of surface temperature (the hypothesis of a constant value was not valid) and heat flux densities exchanged during all steps of the compression test. The evolution of the pressure curves could be also explained from each phase of the process and were influenced by the ability of SMC to flow, its thermal expansion and cure-induced shrinkage. Finally, the possible coupling between the flow and the crosslinking reaction was investigated by varying the compression strain rate applied during the flow and the temperature mould. Heat flux analysis demonstrated that the decrease of the strain rate induced a longer time for flow but also for sample heating (due to worse thermal contact), leading to a delay of the curing reaction. The lower the strain rate, the higher the coupling flow/curing. Qualitative observations of sample surfaces indicated a progressive apparition of defects with this coupling. The main effect of the decrease of mould temperature was the delay of the reaction starting. It thus favoured the decoupling between the flow and the curing and increased the processability window but with larger cycle time.

Acknowledgments

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