METHODS TO IMPROVE MECHANICAL PERFORMANCE OF CARBON FIBER EPOXY SMC

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Abstract

There is an increasing uptake of composite materials in structural applications due to their high specific properties and the decreasing cost of carbon fiber. One of the most industrially-relevant manufacturing processes is sheet molding compound (SMC) due to its commonality and high-throughput. Epoxy carbon fiber SMC is a relatively new material system that has been developed to address issues with traditional SMC. Volatile organic compounds arising from the solvents present in traditional SMC are undesirable from both production and end-use perspectives; epoxy as the matrix can also best achieve the highest mechanical performance. Carbon fiber, however, poses a processing challenge since the small fiber diameter and high surface area are difficult to fully impregnate. Herein a systematic approach is taken to improve the mechanical performance of carbon fiber epoxy SMC by material selection and process-property relationships. Different fiber types, hybrids, roving sizes, processing parameters, the use of a fiber spreader, and the addition of continuous fiber prepreg are examined as methods to achieve the best mechanical performance.

Background

Sheet molding compound (SMC) has long been used with polyester and vinyl ester resin systems to achieve good overall performance and commercial acceptance in a range of applications [1]. Material systems to increase the mechanical performance, such as epoxy and polyurethane, have been recently developed with chemistries compatible to the SMC processing technique [2,3]. Epoxy, specifically, is suitable for numerous semi-structural and structural applications through low-viscosity formulation which can best wet the carbon fiber. Also, it is low in volatile organic compounds, and eliminates styrene monomer exposure [2].

Previous studies have examined some of the material format relationships of advanced carbon fiber composites, albeit with a vinyl ester matrix. Boylan found a relationship to the length of chopped fiber [4]. In her study, she found increasing fiber length did increase the mechanical properties nearly linearly, but caused an increase in cycle time as well as a reduction in the surface quality of moldings. Cabrera-Rios looked at hybrid sheet molding compound whereby the SMC laminates were fabricated with combinations of glass and carbon fiber layers after the individual material sheets had fully matured [5]. The result had quite a bit of scatter but generally suggested a linear trend in properties between the material formulations.

Studies with an epoxy matrix have focused on molding cut prepreg (rather than processing cut fiber into SMC sheet). Feraboli noted excellent properties with epoxy SMC and looked at the orientation effects in the prepreg chips [6]. Nicoletto discussed more of the damage evolution of the cut prepreg SMC and the relatively high variability in the observed performance [7]. While using prepreg eliminates the fiber wet-out issues, it adds a layer of cost and processing complexity not easily tenable in high-volume processing.

With the advent of new epoxy systems specifically formulated for the sheet molding compounding process [2], studies are required to understand the range of performance in carbon fiber composites. Carbon fiber, however, is notoriously difficult to process, often resulting in strengths much less than a similar glass based composite [1,2,4,5]. Thus, this report documents several of the process-property relationships of epoxy / carbon fiber SMC for semi-structural and structural applications.

Experimental Setup

SMC Compounding

Epoxy sheet molding compound (EP SMC) was manufactured on the Dieffenbacher equipment located at the Fraunhofer Project Center in London, Ontario. This industrial-scale line has a throughput up to 354 kg/hr and is suitable for both standard and direct SMC processing. In this instance, the in-line maturation features of the direct process were not used so as to replicate the standard conditions with which many SMC producers are already familiar. A schematic of this equipment and the line heating features are shown in Figure 1 with some of the heating capabilities highlighted. Though room temperature processing of epoxy carbon fiber is possible, it is common to heat the paste slightly to reduce its viscosity to better wet the chopped fiber.

Since the material was often made in small batches, a Bowers stand mixer with a low-shear dispersion blade was used instead of continuous dosing equipment such as a meter-mix-dispense system or an extruder. The low viscosity SMC epoxy from Hexion, EPIKOTE™ Resin TRAC 06605 and EPIKURE™ Curing Agent TRAC 06608, was provided with an internal mold release, HELOXY™ Additive 06805. The resin was mixed according to the data sheet with the exception that the optionally recommended BYK A-560 degassing agent was not used. The epoxy was always mixed for 5 minutes at moderate shear rate to keep the batches consistent.

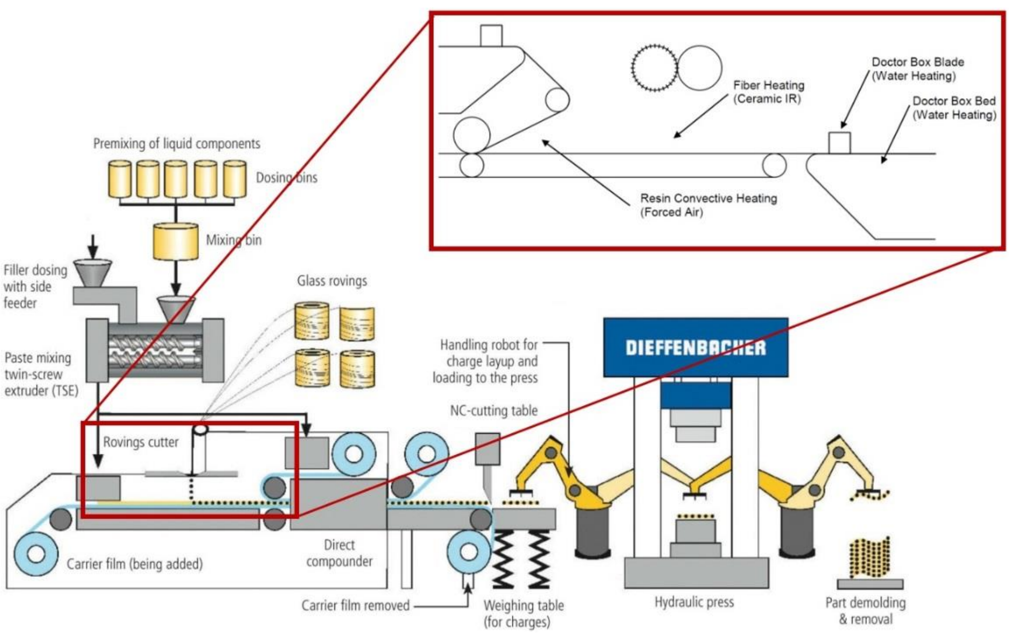

Figure 1 Schematic SMC Equipment at the Fraunhofer Project Center with heating callout

Unless otherwise stated, the carbon fiber was the Zoltek Panex 35 50K-T13. This standard modulus fiber has a low amount of epoxy compatible sizing such that the fiber spreads easily in the compounding and molding operations.

The quality of the SMC sheet is directly related to the selected processing parameters. While a full design of experiments was not conducted, from previous work, several key parameters were selected as the main optimization targets included the sheet basis weight, compaction pressure, and zone temperatures. The target fiber volume content was set to 45% for all the different materials created. The baseline parameters are documented in Table 1.

Table 1 Epoxy SMC compounding parameters

Parameter	Baseline setting	Anecdotes
Batch size	10 kg	
Fiber length	25 mm	
Sheet width	400 mm	
Resin film thickness	1.5 mm	Lower film thickness resulted in better, but less consistent sheet
Basis weight	3500 g/m^2	Lower basis weights resulted in better sheet material
Line speed	1.5 m/min	The line speed was capped at 1.5 m/min for operator safety
Fiber volume fraction	45%	
Line heating	Off	Line heat at 40°C resulted in improved fiber wet-out
Compaction 'pressure'	75 mm	Compaction set by roller offset with no simple way to measure the direct SMC sheet pressure (calculations estimate 0.25 bar)

Hybrid epoxy with glass and carbon fibers was also fabricated by layering glass and carbon SMC sheet prior to molding. Symmetric charges were composed of 0%, 33%, 66%, and 100% volume fraction glass SMC. The glass SMC was produced at the same fiber content level with the same epoxy from Hexion and with ME1510 glass fiber from Owens Corning.

Panels made from carbon fiber with different filament counts were also fabricated as previously described: 3K, 12K, 15K, and 50K. While the fibers were of different manufacturing origin, they were all epoxy sized at similar concentrations.

Tailored (also called co-cured) epoxy SMC with epoxy prepreg was created in a sandwich-panel style SMC charge such that both materials were simultaneously cured when molded together. Here, the carbon fiber was identical in both the SMC and prepreg materials and the respective resin contents similar. Though the prepreg did use a different epoxy resin system from Hexion, this system was selected with similar chemistry to ensure a miscible, uniform cure kinetic profile in the tailored panel. This prevents residual stress arising from differential curing in the mold which can cause micro cracking and delamination at the SMC/prepreg interface. The prepreg was added as unidirectional material aligned with the SMC machine direction. Several weight fractions of prepreg were selected to examine the trend of properties between pure SMC and pure prepreg materials. Charges were also fabricated with a variety of prepreg orientations and charge laminations including unidirectional, orthogonal, outer sandwich, inner sandwich, to name a few. This report discusses the agglomerate results, while future publications may describe the orientation effects of different co-cure arrangements.

A fiber spreading device was installed in the SMC line to better open the fiber bundles prior to compaction. The approach to spreading the fiber is proprietary to Dieffenbacher, but is designed specifically for high-tow carbon fiber. Panels were made using the same 50K carbon fiber as described, but with the added spreading processing equipment.

Finally, split-tow fiber is a fiber architecture technique to segment a large tow fiber into multiple smaller tows and is different to spreading; a technique to open the fiber bundles at the point of sheet compounding. This is not the same as an assembled roving whereby small tow creels are assembled and wound together to form a larger collective tow. Zoltek processed their Panex 50K in such a way as to effect a multi-ended roving; marketed under the moniker Kassen. Unfortunately, the results of this material were not available at the publication time, but the anticipated result based on trends with vinyl ester material is included for the purposes of expected directional trends and overall discussion.

SMC Maturation & Compression Molding

The epoxy SMC was matured according to the recommended guidelines at room temperature for at least a week prior to molding. Panels, 457.2 mm square, were compression molded for this study using a chromed tool and were always molded with a thickness in the range of 2-3 mm, depending on the sheet basis weight. For reference and context, this overall process is depicted in Figure 4: compounding > maturation > molding > testing.

Figure 2 Epoxy / Carbon SMC fabrication process: a) sheet compounding b) roll maturation c) compression molding d) panel testing

The SMC molding parameters were generally held constant; small changes were made trial-to-trial to improve the quality of the panels. The major settings are captured in Table 2 below. One of the critical parameters was found to be the mold filling speed (controlled by the press close speed), whereby a slower speed during the final few millimeters prevented several observed molding defects. Also, an external mold release, Henkel LOCTITE^{TM} FREKOTE 770NC, was applied at the start of each molding day.

Table 2 Epoxy SMC molding parameters

Parameter	Setting	Notes
Core Temperature	145 °C	
Cavity Temperature	135 °C	
Vacuum	On	Until resin gel point (~40s)
Load profile	75 bar	Constant
Speed profile (fast) Speed profile (slow)	80 mm/s 1 mm/s	Slow close only when tool contacts SMC charge
Mold coverage	50%	Center placed
Cure time	3 min	~60s/mm

SMC is known to have a 'machine direction' whereby some percentage of fibers align parallel to the compounding direction due to shear stress in the compaction zone of the line. This can help improve properties in this aligned direction, but reduce the transverse properties. To maintain this characteristic, all SMC charges were stacked with the individual cut blanks aligned; this was done to understand the processing effects and mechanical property variation.

Three exemplar panels are presented as-molded in Figure 3 from the baseline formulation, the spread tow, and one of the co-cure sandwich panels. These panels generally exhibited a clean, blister-free surface, but were not of class-A quality (a class-A surface was not the focus of this study series).

Figure 3 Sample SMC panels a) baseline b) spread tow c) co-cure sandwich

Composite Characterization

The molded panels were sectioned into testing specimens following European standards. The test coupon pattern is shown in Figure 4, with the tensile samples following ISO 527-4 and the flexure samples following ISO 14125. For reference, the disks and tensile gauges (post-test) were subjected to ignition loss testing following ISO 7822 to map out the panel fiber volume content and variability. Some dynamic mechanical analysis samples were also taken from each sample panel for separate study.

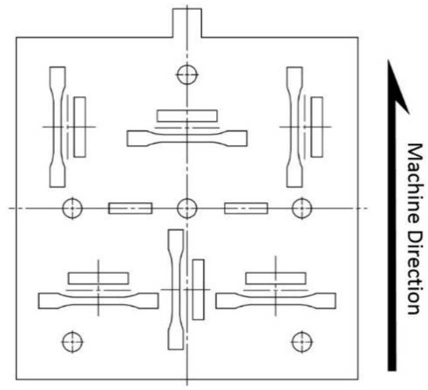

Figure 4 Flat panel geometry and test coupon pattern

Results and Discussion

The tensile results are presented for two selected carbon fiber and one glass fiber formulation in Table 3. These raw results are presented as averages for all the tested coupons in both the machine direction and counter machine direction.

Table 3 Selected epoxy SMC properties

	Baseline	Improved II	Glass/epoxy SMC
Density [g/cm^3]	1.44 ±0.04	1.47 ±0.04	1.94 ±0.05
Volume Fraction [%]	44.6 ±5.3	49.0 ±3.6	53.5 ±3.2
Tensile Modulus [GPa]	21.8 ±3.3	32.0 ±2.5	20.9 ±1.6
Tensile Strength [MPa]	87.0 ±17.9	142.6 ±15.7	304.8 ±18.3
Failure Strain [%]	0.44 ± 0.09	0.50 ±0.06	2.46 ±0.32

The glass epoxy SMC has excellent properties with high modulus and strength values for a chopped-fiber product, coming only with a penalty to the material density. The baseline carbon epoxy SMC represents a first effort with a new SMC matrix and exhibited sub-optimal fiber wetting and low properties. The primary issue is due to being able to fully wet all the fibers within a fiber bundle and the ability for that fiber bundle to open and separate during compaction and molding. The improved II epoxy/carbon SMC takes advantage of a lower basis weight, using line heating to lower the paste viscosity further, and slightly higher compaction pressure. These improvements helped to increase the carbon fiber wet out as well as reduce variability in the SMC sheet and subsequent molded panel.

To best compare all the results, the properties are normalized with respect to a given fiber volume fraction. The linear normalization is generally accepted as accurate for small volume fraction differences of <5% and follows Equation 1. The normalized property is the result of the ratio of the target fiber volume content to the measured fiber volume content multiplied by the measured property. This equation is also used to scale the measured standard deviation.

$$X^* = \frac{v_f^*}{v_f} X \quad (1)$$

The normalized results for the 50K carbon/epoxy SMC are shown together in Figure 5. Small improvements are observed by adjusting the compounding settings. Major improvements are seen by employing either spread- or split-tow material. The best properties were achieved by co-curing the epoxy SMC with 20%wt of epoxy prepreg in a sandwich type material.

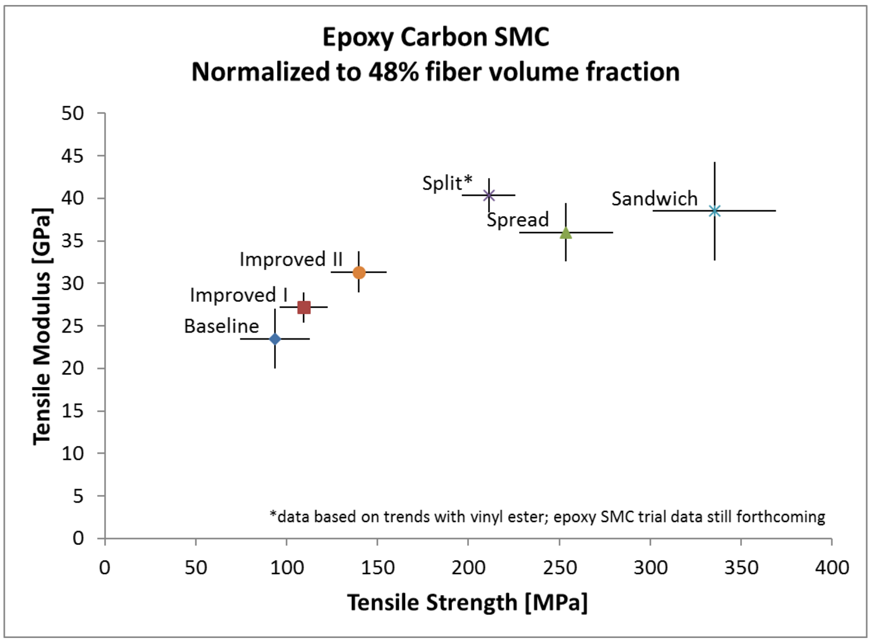

Figure 5 Tensile properties of various epoxy/carbon SMC products and production settings

Plotting the different co-cure materials (Figure 6) with respect to the baseline SMC material reveals a linear trend between the pure SMC and pure prepreg panel properties. This observation is especially important since it implies that epoxy SMC, a lower value product, can displace prepreg, a high value product, in some current designs without impacting weight. For example, a panel primarily in flexure could maintain its bending stiffness by using a sandwich panel with epoxy SMC as the core material rather than a pure prepreg alternative. Thus a simple relationship can be established between the desired material properties and the amount of continuous fiber reinforcement required in a given design.

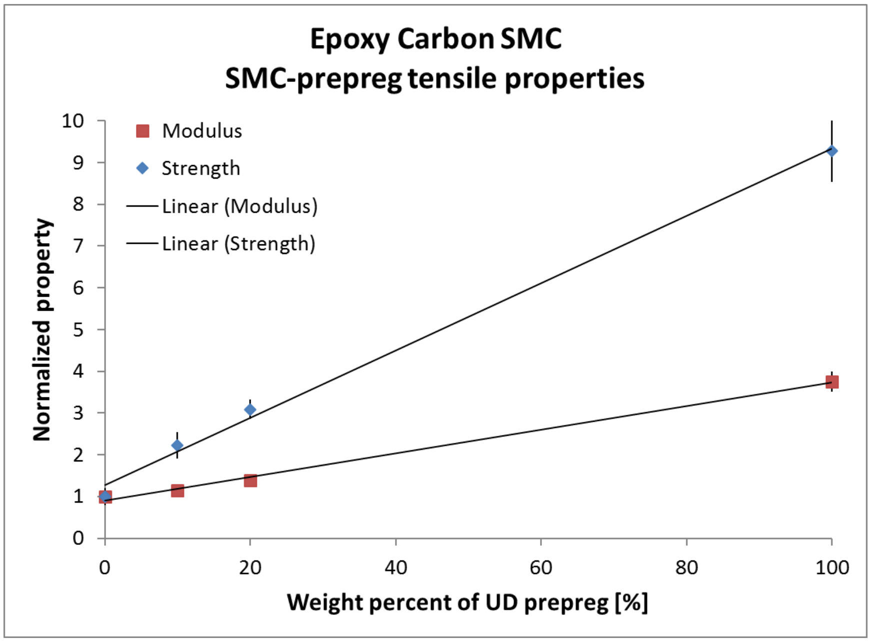

Figure 6 Tensile property trends of co-cured prepreg with epoxy/carbon SMC

The glass and carbon SMC hybrid materials resulted in similar linear property trends based on the component constituents, Figure 7, with respect to the carbon baseline material. Though the glass does offer excellent performance on its own, adding carbon is a viable method to reduce weight without the full cost implication of an all carbon fiber material. These property curves can act as a framework for a design engineer to best select the best material to balance the component cost, mass, and performance.

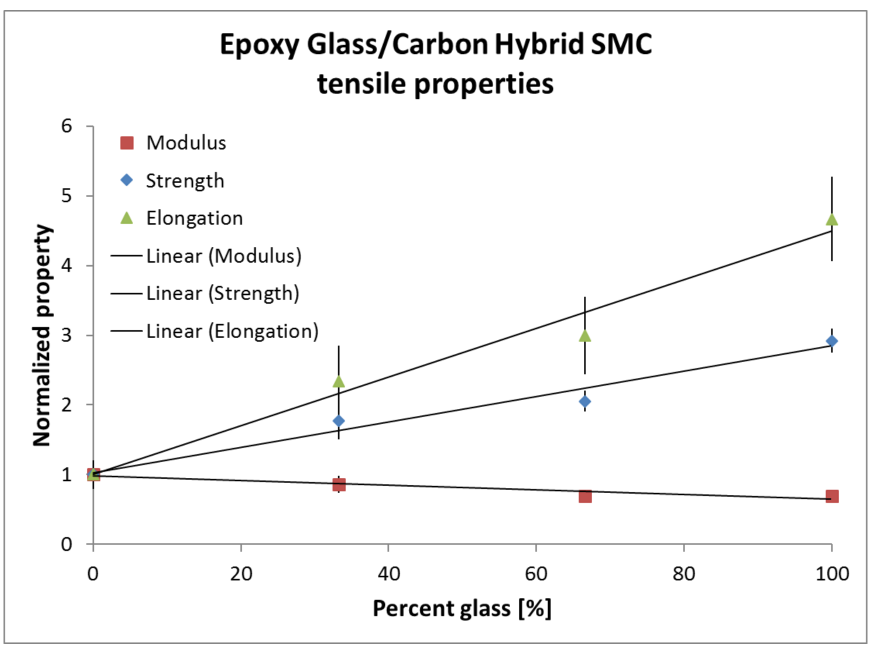

Figure 7 Glass/carbon hybrid epoxy SMC tensile performance trends

Finally, the different carbon fiber tow SMC materials are compared in Figure 8. As the tow size is decreased, both the stiffness and strength of the resulting epoxy SMC increase. At first glance it might have been expected that only the strength would increase since the volume fraction and fiber modulus are constant. However, smaller fiber bundles are more easily wetted during compounding and more easily dispersed during molding resulting in more complete load transfer between fibers and fiber bundles affecting the composite modulus and strength alike. The standard deviations are also observed to decrease with lower filament counts, resulting in molded SMC material with less variability. Thus a 50K spread-tow material performs at a similar level as a 12K standard-tow, but without any of the material cost premium.

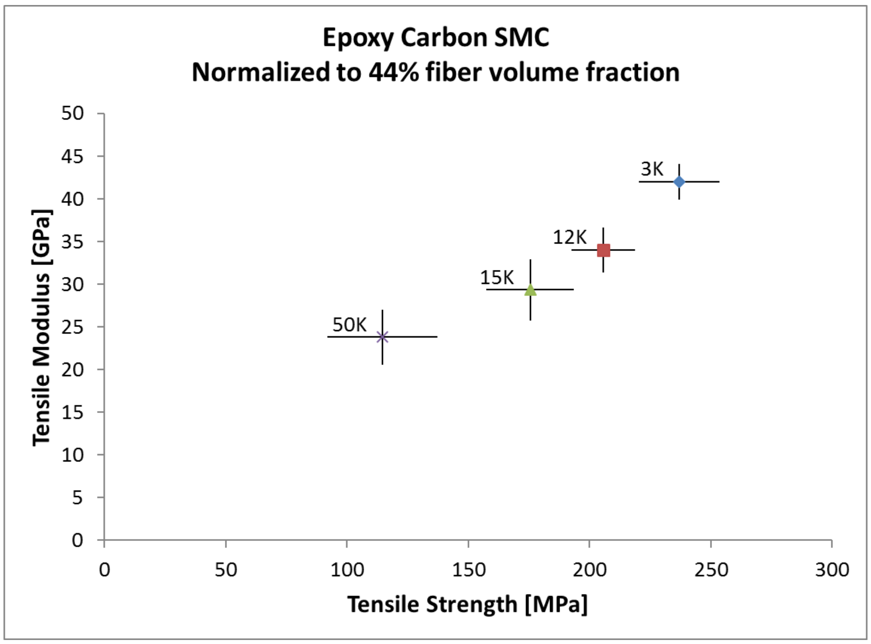

Figure 8 Epoxy/carbon SMC performance based on fiber tow size

Summary and Next Steps

A wide range of carbon fiber epoxy SMC panels were fabricated and tested in a series of industrial scale trials. These materials are suitable for high-volume production such as needed for the automotive industry. Several approaches were taken to improve the performance of epoxy/carbon SMC including process settings, carbon fiber type, and hybrid materials with glass or prepreg. Individually, each of the different approaches offer some level of possible improvement to the material properties. The combination of these approaches is expected to yield the best material solution. Thus future developments should focus on process optimization with fiber spreading technology and the combination of continuous fibers to produce high value, lightweight structural materials suitable for high-volume composite applications. On the chemistry side, future improvements are focused around reducing the system cure time and improving the system T_g without impact to any of the other processing characteristics.

While some optimization is possible with a given material combination and format, the extent of improvement is dwarfed by the performance gains made possible through a slightly different starting material reinforcing the importance of proper material selection in design. The next steps with this work is to analyze a full cost model for the different approaches and document a business case around these semi-structural and structural material systems.

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Bibliography

1. Kia, H.G. (1993) Sheet Molding Compounds: Science and Technology. Hanser/Gardner Publications Inc., Cincinnati, Ohio.
2. Swentek I. (2017) New epoxy systems enabling styrene-free high-performance SMC manufacturing. JEC world: presentation, Paris, France, 14-16 March 2017.
3. Park D. et al. (2016) Development of polyurethane sheet molding compound. Automotive Composites Conference and Exhibition, ACCE 2016; Novi, Michigan, USA, 6 – 8, September 2016.
4. Boylan S. and Castro J.M. (2003) Effect of reinforcement type and length on physical properties, surface quality, and cycle time for sheet molding compound (SMC) compression molded parts. Journal of Applied Polymer Science, vol. 90, 2557-2571.
5. Cabrera-Rios M. and Castro J.M. (2006) An economical way of using carbon fibers in sheet molding compound compression molding for automotive applications. Polymer Composites, vol. 27, 718-722.
6. Feraboli P. et al. (2009) Characterization of prepreg-based discontinuous carbon fiber/epoxy systems. Journal of Reinforced Plastics and Composites, vol. 28, no. 10, 1191-1214.
7. Nicoletto G. et al. (2016) Mechanical Characterization of advanced random discontinuous carbon/epoxy composites. Materials Today: Proceedings, 1079-1084.