

# Study of Influencing Factors on the Flowability of Jute Nonwoven Reinforced Sheet Molding Compound

Miriam I. Lautenschläger, Max H. Scheiwe, Kay A. Weidenmann, Frank Henning, Peter Elsner

**Abstract**—Due to increasing environmental awareness jute fibers are more often used in fiber reinforced composites. In the Sheet Molding Compound (SMC) process, the mold cavity is filled via material flow allowing more complex component design. But, the difficulty of using jute fibers in this process is the decreased capacity of fiber movement in the mold. A comparative flow study with jute nonwoven reinforced SMC was conducted examining the influence of the fiber volume content, the grammage of the jute nonwoven textile and a mechanical modification of the nonwoven textile on the flowability. The nonwoven textile reinforcement was selected to support homogeneous fiber distribution. Trials were performed using two SMC paste formulations differing only in filler type. Plate-shaped kaolin with a mean particle size of 0.8  $\mu\text{m}$  and ashlar calcium carbonate with a mean particle size of 2.7  $\mu\text{m}$  were selected as fillers. Ensuring comparability of the two SMC paste formulations the filler content was determined to reach equal initial viscosity for both systems. The calcium carbonate filled paste was set as reference. The flow study was conducted using a jute nonwoven textile with 300  $\text{g}/\text{m}^2$  as reference. The manufactured SMC sheets were stacked and centrally placed in a square mold. The mold coverage was varied between 25 and 90% keeping the weight of the stack for comparison constant. Comparing the influence of the two fillers kaolin yielded better results regarding a homogeneous fiber distribution. A mold coverage of about 68% was already sufficient to homogeneously fill the mold cavity whereas for calcium carbonate filled system about 79% mold coverage was necessary. The flow study revealed a strong influence of the fiber volume content on the flowability. A fiber volume content of 12 vol.-% and 25 vol.-% were compared for both SMC formulations. The lower fiber volume content strongly supported fiber transport whereas 25 vol.-% showed insignificant influence. The results indicate a limiting fiber volume content for the flowability. The influence of the nonwoven textile grammage was determined using nonwoven jute material with 500  $\text{g}/\text{m}^2$  and a fiber volume content of 20 vol.-%. The 500  $\text{g}/\text{m}^2$  reinforcement material showed inferior results with regard to fiber movement. A mold coverage of about 90 % was required to prevent the destruction of the nonwoven structure. Below this mold coverage the 500  $\text{g}/\text{m}^2$  nonwoven material was ripped and torn apart. Low mold coverages led to damage of the textile reinforcement. Due to the ripped nonwoven structure the textile was modified with cuts in order to facilitate fiber movement in the mold. Parallel cuts of about 20 mm length and 20 mm distance to each other were applied to the textile and stacked with varying orientations prior to molding. Stacks with unidirectional orientated cuts over stacks with cuts in various

directions e.g. (0°, 45°, 90°, -45°) were investigated. The mechanical modification supported tearing of the textile without achieving benefit for the flowability.

**Keywords**—Filler, flowability, jute fiber, nonwoven, sheet molding compound.

## I. INTRODUCTION

IN consequence of increasing environmental awareness natural fibers are more frequently used in fiber reinforced composites [1], [2]. In comparison to conventional reinforcing fibers, natural fibers offer many advantages such as sustainability, renewability or biodegradability [1], [3]. Jute fibers as one representative are used because of their low cost and their low density [4], [5]. With 1.45  $\text{g}/\text{cm}^3$ , the density of jute fibers is lower compared to glass fibers (2.56  $\text{g}/\text{cm}^3$ ) [5].

The SMC process allows a high degree of design freedom. Complex shapes such as ribs can be realized by the flow of material during compression molding. The challenge in the use of jute fibers is their reduced flowability in the mold cavity. Müssig et al. used the SMC process to produce an exterior component based on renewable resources [6]. The flow capacity of Cordenka and hemp fibers was investigated and it was found that natural fibers in the SMC process require higher SMC paste viscosities than synthetic fibers to allow flow. Compared to glass fibers natural fibers own a lower flexural stiffness which is the reason for fiber tangling. Tangled fibers decrease the fiber movement in the mold [6]. The observations of van Voorn et al. are comparable [7]. Furthermore, it was found that an even distribution of chopped flax fibers in the semi-finished product influences the homogeneity of the flow.

Besides chopped fibers, nonwoven textiles are an alternative reinforcing material in the SMC process. Due to their structure they offer nearly isotropic properties [2]. During processing barbed needles are punched through a fiber web and reorient some fibers in the vertical plane. The resulting structure affects the properties of the nonwoven textile [8]. Sengupta et al. studied the effect of area density, punch density, fiber orientation and depth of needle penetration of jute needle-punched nonwoven material [8]. Composites reinforced with nonwovens with different mass per unit area as well as jute woven reinforced fabric were manufactured using hand lay-up technique. Comparing the mechanical properties of jute nonwoven and woven reinforced composites, higher flexural and tensile strength as well as higher impact resistance for the nonwoven reinforced

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composites was found [8]. The use of nonwovens supports homogeneous fiber distribution due to its internal structure.

Fillers can be used to influence the viscosity of the SMC paste. Kiehl et al. investigated the influence of calcium carbonate in unsaturated polyester resin with regard to filler content and the shear stress on the viscosity [9]. Increasing filler content led to an increase of the viscosity. For a filler content beyond 53 wt.-% calcium carbonate, shear thickening behavior was observed [9]. Furthermore, Yuan and Murray studied the effect of particle morphology on the rheological behavior of concentrated kaolin suspensions [10]. Kaolin is subclassified in halloysite and kaolinite. Halloysite can be either tubular or spheroidal whereas kaolinite has a hexagonal platelet shape. Yuan and Murry found a relationship between particle shape of the kaolin and the viscosity independent of shear rates. Tubular halloysite filled suspensions showed the highest viscosity whereas spheroidal halloysite revealed the lowest viscosity. The viscosity of platy kaolinite filled suspension was in between [10]. Compared to kaolin, calcium carbonate has an ashlar shape. Fillers impact the viscosity of SMC paste, but no scientific work so far has focused on their influence on the fiber movement of jute nonwoven reinforced SMC.

The presented research work focuses on selected influencing factors on the flowability of jute nonwoven reinforced SMC. Material flow of jute nonwoven textiles is considered as expansion of the semi-finished product in the mold. Ashlar calcium carbonate and platy kaolin were selected to investigate their influence on fiber transportation during consolidation. Different flow studies with varying focus were performed. First the minimum mold coverage assuring homogeneous fiber distribution for each filler was determined. Moreover, the fiber volume content and the grammage of the used jute nonwoven textile were varied. In order to support even fiber distribution, the jute nonwoven material was modified by introducing cuts.

## II. MATERIALS

For the trials the unsaturated polyester resin Palapreg P17-02 supplied by Aliancys Deutschland GmbH was used. This resin is derived from standard glycols and orthophthalic acid diluted in styrene. As thickening agent Luvatol MK 35 from Lehmann & Voss & Co. was used. Additionally, styrene as solvent, peroxide as accelerator, BYK-W 9010 from BYK-Chemie GmbH as dispersant and Ceasit I from Baerlocher GmbH as release agent were added to the SMC paste. As fillers either calcium carbonate Omya Milicarb®-OG supplied by Omya GmbH or kaolin Chinafill KBE-1 supplied by Amberger Kaolinwerke akw were added to the SMC paste.

The calcium carbonate with an average particle size of 2.7  $\mu\text{m}$  is obtained from natural calcium carbonate. The kaolin filler with an aspect ratio of 20/1 has an average particle size of 0.8  $\mu\text{m}$ . This filler offers a high platiness.

As reinforcing textile needle-punched jute nonwoven material was selected with either 300 g/m<sup>2</sup> or 500 g/m<sup>2</sup>.

## III. METHODS

Assuring comparability between the calcium carbonate and the kaolin filled SMC paste the amount of the two fillers was determined to reach equal initial viscosity. Therefore, the initial viscosity of a calcium carbonate-polyester resin blend was measured using an Anton Paar Rheolab QC with a concentric cylinder. The result was set as reference for the kaolin-polyester blend. Kaolin filler was gradually added to the resin until the initial viscosity of the calcium carbonate-polyester blend was reached.

The SMC paste was produced mixing first all liquid components such as resin, peroxide, styrene and dispersant. Afterwards the solid components such as filler and release agent were introduced to the liquid. The paste was stirred with about 1500 rpm for 3 minutes with a dissolver (VMA Getzmann Dispermat® LC400) under vacuum to achieve a homogenized paste. The SMC material was produced applying a thin film on two carrier foils. After placing the dried jute nonwoven reinforcing textile on one paste film, the second carrier foil was applied on top forming a sandwich-like prepreg. This prepreg was consolidated and afterwards stored for 3 days at 30 °C. Until processing the produced SMC material was stored at 17 °C.

Jute nonwoven textile with 300 g/m<sup>2</sup> and 500 g/m<sup>2</sup> were used for the SMC production. SMC with both filler types was produced with 300 g/m<sup>2</sup> achieving a fiber volume content of about 12 vol.-%. Additionally, SMC material with 25 vol.-% was produced using the 300 g/m<sup>2</sup> jute nonwoven. The 500 g/m<sup>2</sup> nonwoven material was used to reinforce only kaolin filled SMC with a fiber volume content of 20 vol.-%. Moreover, the 300 g/m<sup>2</sup> nonwoven material was modified by applying cuts and used as reinforcing material in SMC with 25 vol.-% fiber volume content. A Zündt cutting table was used to slice the nonwoven textile. Cuts were either applied parallel or perpendicular to the direction of uncoiling the textile (Fig. 1). The direction of uncoiling equals the processing direction of the SMC material manufacturing process and is further on referred as processing direction. The cuts were of 20 mm length with a distance of 20 mm to each other. The jute nonwoven was not cut through. Instead considering process fluctuations about 1 mm of the 3 mm thick reinforcing material was left uncut.

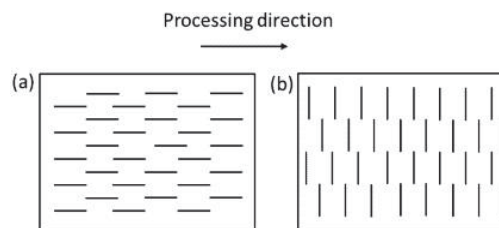


Fig. 1 Cutting pattern (a) parallel and (b) perpendicular to processing direction

SMC laminates were manufactured with varying initial mold coverage. For the flow study SMC material was stacked and centrally placed in the mold covering between 25 to 95%.

The SMC material with the modified nonwoven textile was first placed along one side of the cavity covering 25%. Afterwards the material was laid in one corner of the square-shaped mold. Fig. 2 shows exemplarily the material placement in the mold. The number of sheets forming the stack was varied between 2 to 9 layers. Because of the reduced flow capacity of the jute nonwoven only a distinct number of layers were stacked. To assure comparability the weight of the stack was maintained. Emerging these boundaries, the initial mold coverage was derived.

In the press, a pressure of 200 bar was applied to consolidate the material at 150 °C. The press parameters were maintained for all trials.

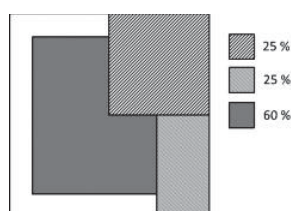


Fig. 2 Schematic drawing of the material placement in the mold

For qualitative evaluation of the produced laminates, transmitted light images were made using the light source of an overhead.

#### IV. RESULTS

In the following section the results of the influence of the filler type including the determination of the minimum required mold coverage, the effect of the variation of reinforcing material and fiber volume content as well as the impact of the modification of the jute nonwoven textile are described.

##### A. Influence of Filler Type

The flow study with calcium carbonate and kaolin filled SMC (300 g/m<sup>2</sup>, 12 vol.-%) revealed that the mold coverage has a strong influence on the homogeneity of the fiber distribution. A low mold coverage led to the destruction of the jute nonwoven textile structure. Ripped parts of the reinforcing material were transported to the extremities of the mold. Fig. 3 shows a calcium carbonate filled SMC plate that was produced with 27 % mold coverage. The placement of the square SMC stack was marked with a dashed line. The SMC filled the mold cavity, but the jute textile was ripped apart and the fibers were partially aligned. Furthermore, resin reservoirs formed in between.

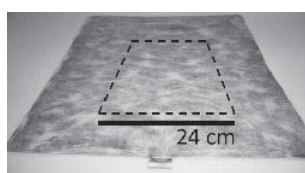


Fig. 3 Calcium carbonate filled SMC plate with 27 % mold coverage (the dashed line marks the stack position and size)

With high mold coverage none of these phenomena were observed. Thus, the minimum percentage mold coverage assuring homogeneous fiber distribution was determined by varying the size of the stack. Fig. 4 compares two calcium carbonate filled SMC plates with (a) 60.5% and (b) 79%. The scaled bar on the pictures is equal in length. The plate with 60.5% mold coverage shows a spotted inhomogeneous surface whereas fibers in the plate with 79% seem to be uniformly distributed. The minimum percentage mold coverage for the calcium carbonate filled SMC was determined to be at least 79%. The transmitted light images of these two plates are shown in Fig. 5. The image of the 60.5% mold coverage plates also shows locally concentrated fiber bulks and resin rich areas (a) whereas the image of the plate with 79% mold coverage exhibits even fiber arrangement. The transmitted light analysis supports the result.

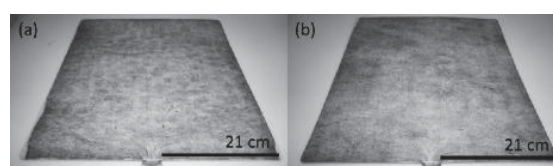


Fig. 4 Comparison of calcium carbonate filled SMC plates with (a) 60.5% mold coverage and (b) 79% mold coverage

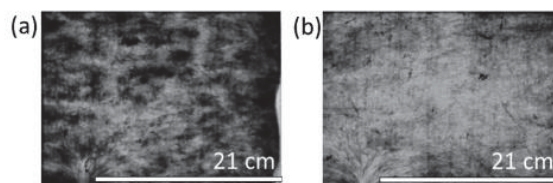


Fig. 5 Comparison of transmitted light images of calcium carbonate filled SMC plates with (a) 60.5% mold coverage and (b) 79% mold coverage

Fig. 6 shows kaolin filled SMC plates with (a) 60% mold coverage and (b) 68% mold coverage. The plate with the lower mold coverage reveals resin rich areas. With 68% mold coverage the plate shows homogeneous fiber distribution on the surface. Comparing the transmitted light images, the minimum percentage mold coverage for kaolin filled SMC was determined to be at least 68%.

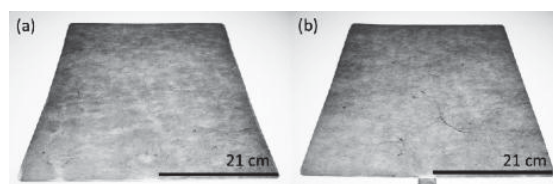


Fig. 6 Comparison of kaolin filled SMC plates with left 60 % mold coverage and right 68 % mold coverage

For the comparison the calcium carbonate and kaolin filled plates with stacks covering 53% of the mold consisting of 3 layers were manufactured. The stacks of the two SMC types



were of comparable weight. In Fig. 7, the different influence of the fillers can be observed. The calcium carbonate filled SMC plate shows fiber bulks and elongated shaped resin concentrations. Whereas the kaolin filled SMC plate reveals more uniformly distributed fibers. The beginning of the nonwoven textile destruction is already visible, but less progressed than for the calcium carbonate filled material. Fig. 7 also presents the transmitted light images of the calcium carbonate (c) and kaolin (d) filled SMC plates. The images support the results also throughout the plate not only the surface.

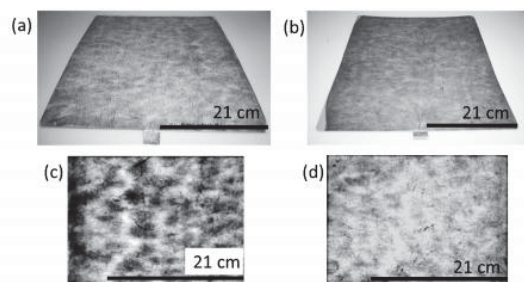


Fig. 7 Comparison of (a) calcium carbonate and (b) kaolin filled SMC plates and their corresponding transmitted light images (c) calcium carbonate filled SMC and (d) kaolin filled SMC with 53 % mold coverage and 3 layers stacked below

#### B. Variation of Reinforcing Material and Fiber Volume Content

A flow study with 12 and with 25 vol.-% fiber volume content was performed to investigate the influence of the fiber volume content on the flowability. For this flow study the SMC material was centrally placed in the mold covering 79 % of the mold surface. Fig. 8 compares the plates of kaolin filled SMC with (a) 12 vol.-% and (b) 25 vol.-%. The SMC material with the higher fiber volume content was not able to fill the mold cavity. Resin was squeezed out of the nonwoven reinforced SMC material instead. When comparing the molded plate with the size of the stack no difference was measured. The material with 25 vol.-% did not allow flow at all. In contrast, the SMC material with the lower fiber volume content completely filled the mold cavity with homogeneous fiber distribution. Similar results were achieved with calcium carbonate filled SMC.

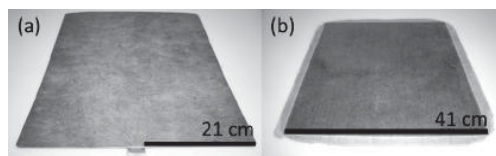


Fig. 8 Comparison of kaolin filled SMC plates with (a) 12 vol.-% and (b) 25 vol.-% fiber volume content

Next to the 300 g/m<sup>2</sup> jute nonwoven material and a fiber volume content of 12 vol.-% jute nonwoven textile with 500 g/m<sup>2</sup> was selected and a fiber volume content of 20 vol.-% was set. A flow study with kaolin filled SMC was performed

starting with 79% mold coverage and 2 layers of SMC. The stack was laid centrally in the mold. With keeping the weight of the SMC stack constant 3 layers of SMC material resulted in 53% mold coverage, 4 layers in 40.5% and 5 layers in 32%. Fig. 9 compares the result of the variation of the reinforcing material and the fiber volume content. The heavier nonwoven material shows already destruction with a mold coverage of 79%. With further decrease of the mold coverage diamond shaped pieces of nonwoven material form, separated by resin rich areas. In contrast, plates with the lower fiber volume content and 300 g/m<sup>2</sup> nonwoven show homogeneous fiber distribution for 79%. At 53% mold coverage initial approaches of destruction of the reinforcing material become visible. With further decrease of the mold coverage a diamond shaped structure also appears.

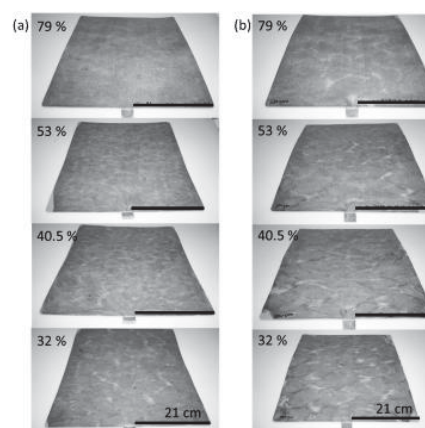


Fig. 9 Comparison of decreasing mold coverage from 79% to 32% of kaolin filled SMC plates with (a) 12 Vol.-% and 300 g/m<sup>2</sup> jute nonwoven and (b) 20 Vol.-% fiber content and 500 g/m<sup>2</sup> jute nonwoven

#### C. Modification of Jute Nonwoven Textile

Prior to the flow study with centrally placed SMC material the stack was laid along one side of the mold cavity allowing one-dimensional flow. Calcium carbonate filled SMC material with a fiber volume content of 25 vol.-% was used. Two basis observations were made during one-dimensional flow. It was found that partially resin accumulated in between the cuts and extended the cuts. Fig. 10 shows such a bloated cut which was expanded and connected to another cut. In this case the cuts were introduced to the nonwoven material parallel to the processing direction. The flow direction was perpendicular to the processing direction.

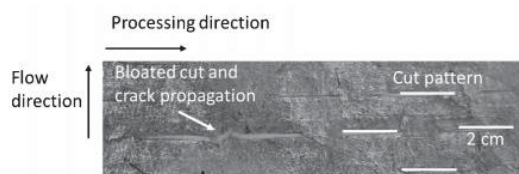


Fig. 10 Bloating of cuts and crack propagation of a calcium carbonate filled SMC plate with parallel to processing direction introduced cuts (white bars)

The second observed phenomenon is presented in Fig. 11. Instead of filling the cuts cracks form perpendicular to the introduced cuts. These cracks propagate along the tips of the cuts. In this case, the cuts were made perpendicular to the processing direction and the flow direction coincides with the processing direction.

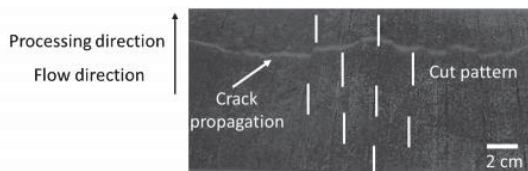


Fig. 11 Crack propagation of a calcium carbonate filled SMC plate with perpendicular to processing direction introduced cuts

In the next step, 4 layers of SMC material were stacked with the cut direction  $0^\circ/45^\circ/90^\circ/-45^\circ$ . Both parallel and perpendicular cuts to processing direction were investigated with 25% mold coverage under two-dimensional flow (stack placed in one corner). The results for both cut introduction directions showed comparable result. Fig. 12 shows a section of a photograph and transmitted light image of a calcium carbonate filled SMC plate with perpendicular to processing direction introduced cuts. The surface of the plate already reveals the rotation of the cut orientation in the nonwoven. Bloated cuts from the two layers in  $0^\circ$  and  $45^\circ$  close to the shown surface appear strongly on the surface. The transmitted light image indicates that not only cuts were bloated but cracks propagated in cascades.

Finally, a stack covering 80% of the mold surface was centrally placed in the mold. But, the results showed no support of the modifications.

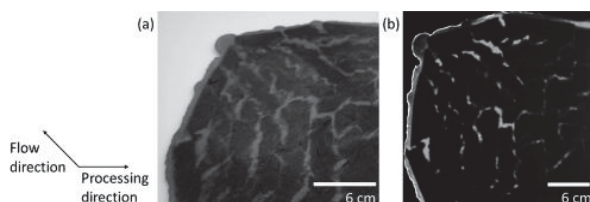


Fig. 12 Section of (a) photograph and (b) transmitted light image of calcium carbonate plate filled SMC plate with sheets stacked perpendicular to processing direction,  $45^\circ$ ,  $90^\circ$ , and  $-45^\circ$  rotated

## V. DISCUSSION

A flow study with calcium carbonate and kaolin filled SMC was performed to investigate the influence of the mold coverage on the homogeneity of the fiber distribution. The results for both filler systems indicated a correlation between the homogeneity of the fiber arrangement and the mold coverage. With increasing mold coverage, the fiber distribution became more uniform. The determination of the minimum mold coverage assuring even fiber arrangement revealed different behavior of the fillers calcium carbonate and kaolin in the SMC material. The minimum mold coverage

of kaolin filled SMC was lower (68%) compared to calcium carbonate filled SMC (79%). As the two SMC materials were manufactured with the same parameters besides the filler, the effect was ascribed to the filler type. Calcium carbonate and kaolin differ in their particle size and shape. Due to the ashlar shape, calcium carbonate is considered as isotropic particle [11]. These particles affect the compound equally in all directions. In contrast flaky kaolin shows anisotropic behavior. Anisotropic particles are assumed to align in the flow direction minimizing the resistance against flow [11]. Because of their two-dimensional structure kaolin particles seem to exert force on the jute nonwoven fabric and thus enable the material flow in the cavity. Furthermore the edges of kaolin platelets possess positive and negative charges building a three-dimensional structure comparable to a "house of cards" [12]. During maturation of the SMC material such a three-dimensional structure could develop. The construct would support the expansion of the nonwoven fabric and get destroyed by the material flow. With decreasing viscosity of the SMC material during consolidation, the kaolin particles may align in the material flow and minimize the destruction of the textile. In contrast, ashlar shaped calcium carbonate probably cannot reduce the exerted force on the jute nonwoven fabric and thus supports tearing, but it needs to be considered that with decreasing mold coverage the influence of the molding pressure increases which can also lead to the destruction of the textile.

Based on the results, it is assumed that a critical fiber volume content exists preventing fiber transport of jute nonwoven material when exceeded. SMC with 12 and 25 vol.-% was manufactured and compared. Following this assumption, the SMC with 25 vol.-% has already exceeded the critical fiber volume content as the consolidated plates show no fiber transport. Probably the lower filler-resin paste share in the SMC material was not able to exert enough force on the fabric. Only close to the edge of the SMC stack some paste was pushed out. Even though no direct comparison may be possible the result of the  $500 \text{ g/m}^2$  nonwoven SMC material with 20 vol.-% can be used as indication for a critical fiber volume content between 12 and 20 vol.-% jute fibers. For this SMC material, flow in the cavity was observed though inferior compared to the  $300 \text{ g/m}^2$  SMC material with lower fiber volume content. Next to the fiber volume content, the thicker textile seems to reduce the flowability. Sengupta et al. studied the effect of mass per unit area of jute nonwovens in the hand lay-up process [8]. It was found that increasing mass per unit area led to a stronger compaction of the fabric during needling. Simultaneously fiber entanglement increases [8]. Stronger fiber entanglement in the SMC process impedes fiber movement and thus fiber transportation. A  $300 \text{ g/m}^2$  jute nonwoven material is recommended to be chosen over a  $500 \text{ g/m}^2$  fabric.

Due to the destroyed nonwoven structure for low mold coverage the nonwoven fabric was modified by introducing cuts. The aim was to selectively weaken the fabric structure and at the same time reduce the initial fiber length. Müssig et al. have reported an optimal flow of hemp fibers with a length

of 20 mm [6]. The fiber length in nonwoven material exhibits a fiber length distribution between about 38 to 78 mm [13]. In fact, the introduced cuts did not support homogeneous fiber distribution nor facilitated fiber transportation.

## VI. CONCLUSION

The influencing factors filler type, fiber volume content, textile grammage and textile modification of jute nonwoven reinforced SMC were investigated. Ashlar calcium carbonate and platy kaolin were used as fillers in the SMC material. Both filler supported fiber transportation. Although kaolin revealed a better impact on the homogeneity of the fiber distribution than calcium carbonate at similar mold coverage. The fillers differ in the minimum mold coverage assuring even fiber arrangement. For kaolin filled SMC 68% mold coverage was determined whereas for calcium carbonate 79%. Differences were assumed to issue from particle morphology. Flow studies with 12 and 25 vol.-% fiber volume content indicated a critical fiber volume content preventing fiber transportation when exceeded. Due to more compact and entangled fiber structure 500 g/m<sup>2</sup> jute nonwoven fabric showed inferior flow properties compared to 300 g/m<sup>2</sup>.

The next steps of the work will be the determination of the critical fiber volume content and the mechanical characterization of the insertion area in the mold as well as the overflow area.

## ACKNOWLEDGMENT

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