EFFECT OF FLAKE DISTRIBUTION IN MOLD ON THE FLOW DURING COMPRESSION MOLDING OF UNIDIRECTIONAL LONG FIBER THERMOPLASTIC FLAKES

A Bhakta[1, 2], W Ali[1, 2], H.J.M. Geijselaers[1], U Sachs[2], M I A Rasheed[1], A.H.van den Boogaard[1]

1 University of Twente,7500 AE, Enschede, The Netherlands, mail: a.bhakta@utwente.nl
2 ThermoPlastic Composites Research Centre (TPRC), 7521 PN, Enschede, The Netherlands

Keywords: Composite flakes, Long fiber thermoplastics (LFTs), Compression molding, Composites recycling, Discontinuous fiber thermoplastics.

ABSTRACT

The growth of the use of composites in the industry generates a significant amount of composites waste. The thermoplastic wastes in the form of laminates and pre-pegs accounts for a significant percentage of the total material processed by composites manufacturing industries. These wastes could be shredded into smaller chips or flakes and compression molded into reusable products. Previous experiments have been carried out at the University of Twente in the Netherlands to produce an industrial product from recycled chopped woven flakes [1]. This paper discusses the compression molding process of chopped unidirectional (UD) flakes. The effect of consolidation pressure, dwell time and filling pattern of the mold on the laminates is studied. The paper shows the mechanisms of flake deformation generally observed during the molding process. Surface microscopy and density measurements show the extent of flake deformation and polymer flow. This qualitative analysis will later be used to model the process of flake deformation in melt to predict the mold filling behavior during compression molding of long (6-20 mm) thermoplastic flakes.

1 INTRODUCTION

Scraps generated in the composites industry are generally in the form of excess pre-pegs or laminates. Laminates cannot be directly compression molded. The scraps generally undergo a size reduction process through chopping and sieving before they are placed into the mold to be compression molded into a flake reinforced composite. Figure 1 shows the composite recycling for compression molded flake reinforced composite products.

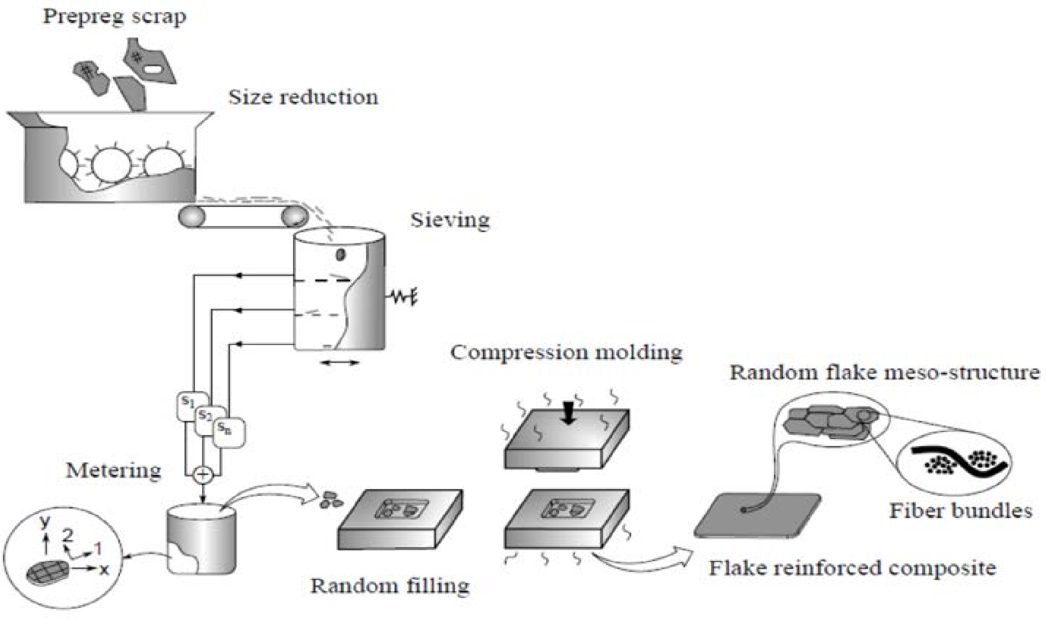


Figure 1: Conceptualized cycle for recycled compression molded thermoplastics [1]

For this paper the fiber length equals the flake length which in most cases is greater than or equal to 10 mm, which is considered as long fiber composites [3] . Discontinuous Sheet Molding Compounds (SMCs) are being used in the industry for quite long. For instance recently, Boeing used discontinuous SMCs to make window frames of its Dreamliner [2] aircraft. Thermoplastics too are increasingly being used in aviation industry for parts like clips, brackets, stiffeners and many other secondary products. Thermoplastics with long fibers could be a superior alternative to thermosets in near future. As both the aerospace and automotive industry shifts to use more of thermoplastic composites from thermosets, further work is needed to be done in the modelling of thermoplastic composites before they are fit to be used in aviation industry. Long fiber discontinuous thermoplastics are easier to process than continuous fiber thermoplastics and do not compromise much on the performance [4, 5] . This versatility brings the need to come with simulation tools able to predict their processing behavior.

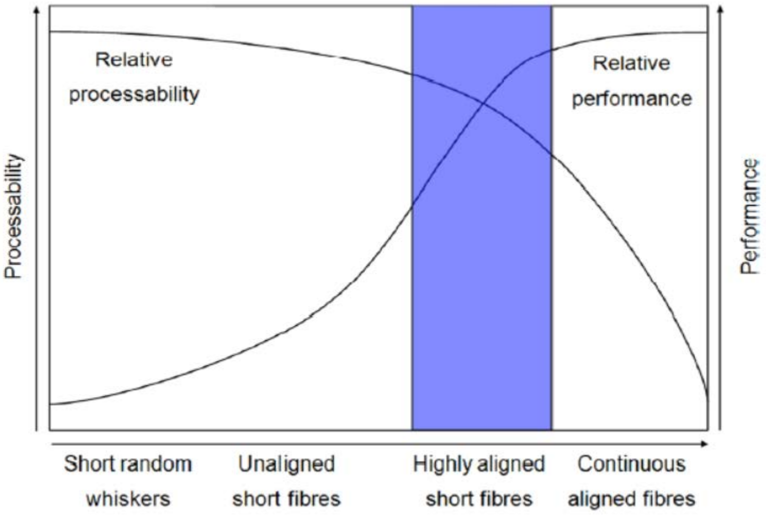


Figure 2: The processability performance schematic graph for different fiber lengths [4]

Long fiber composites also generally show properties on the higher side making them potent for aerospace usage.

Before concentrating on modelling of compression molding of recycled thermoplastic scraps, more efforts need to be taken in modelling of discontinuous long fiber thermoplastics. Till now accurate modelling tools to simulate filling behavior of long discontinuous fiber thermoplastics in melt do not exist. Aerospace grade parts demand thermoplastic matrix with long polymer chain and composites with fiber volume fractions of 50-60% for adequate performance. The long hydrocarbon chains give the polymers high melt viscosities. This highly viscous filling behavior with the densely packed fibers affects the fiber orientation. The fiber orientation again affects the filling behavior. Both mechanisms are coupled.

The initial distribution of material in the mold could have significant effect on the final properties thus affecting material properties. Ferraboli et al. [8] attempted to make laminates from randomly oriented discontinuous carbon-epoxy thermosets and estimated the stiffness from the fiber orientation of the laminates using a modified version of classical laminate theory. This work is preceded by Halpin and Pagano’s [6-7] work in which they estimated the stiffness of randomly oriented fiber composites from the properties of a quasi-isotropic laminate. Ferraboli followed a reverse approach to estimate the properties of the laminate. Their work is schematically denoted by figure 3:

Figure 3: Stiffness approximation for randomly oriented discontinuous plate [8]

Their work shows that for a compression molded composite if the local volume fraction and fiber orientation distribution are known, the variability in stiffness and thereby the strengths of composite laminates can be estimated.

Discontinuous long fiber reinforced thermoplastics generally come as at fixed length ranging from 3 mm to 48 mm or are chopped as per process' needs and manually filled into a mold of known geometry and size before being taken through the compression molding consolidation cycle. The nature of this manual filling pattern remains an unknown input to the physics of the compression molding process [9] . This paper begins with the initial distribution of material in the mold. This could have significant effect on the final properties thus affecting material properties. *One of the big needs the industries see is how to distribute the chips to avoid lumps and maintain consistent flatness* [9] .

2 NUMERICAL INVESTIGATION

Based on previous literature [10] a random flake distribution is numerically constructed. In the industry generally the distribution of flakes is done randomly without accounting much for the statistics of the flakes. In order to construct a demonstrator an unquantified distribution of material is made to make a discontinuous thermoplastic product out of woven or UD flakes.

Not much attention has been paid before to the initial statistics of the flakes: their position, flake count, flake overlaps and flake neighborhood. So the experiment is numerically replicated in a 250 mm x 250 mm flat mold.

A random distribution function in MATLAB generates the flakes' position in the mold. A code then checks the overlap of the flakes at 25 x 25 grid points. About 10000 flakes are randomly distributed and the number of flakes lying on top of each other is found as a function of position.

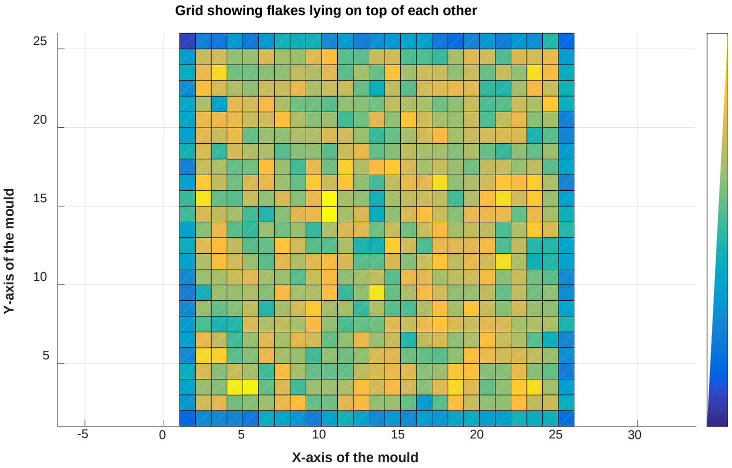


Figure 4: Numerical Distribution (generated in MATLAB)
X-axis and Y-axis show the mold's coordinates and legend shows number of flake overlaps

Figure 4 shows the flake count or overlap of flakes in different areas of the mold. The pictorial is a depiction of number of flakes which are lying on top of each other. The number of flakes lying on top of each other is arranged in a descending order in figure 5.

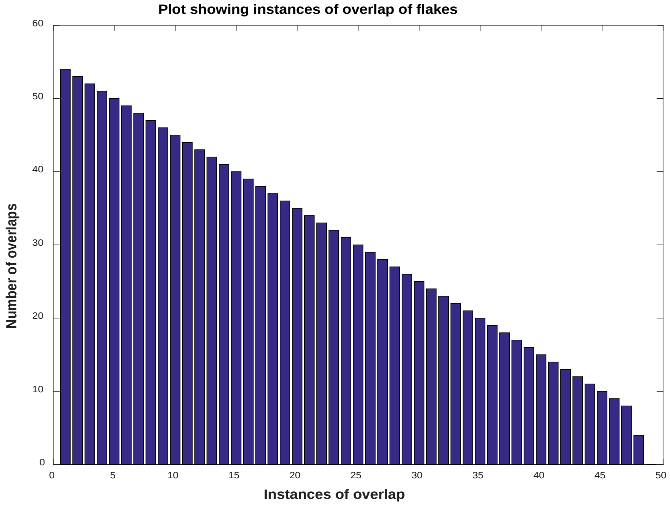


Figure 5: Descending Overlap Order of distributions
X-axis-number of variations and Y-axis-number of overlaps of flakes

These stacking columns of different heights could be randomly distributed at any position in the mold. This curve has a qualitative relation to the process. The mold comes in contact with the highest peak of flakes first then with the 2nd highest one and so on. There is an increase in force to be needed to squeeze the additional peak during the pressing [10] . The cumulative sum of this curve is in figure 6:

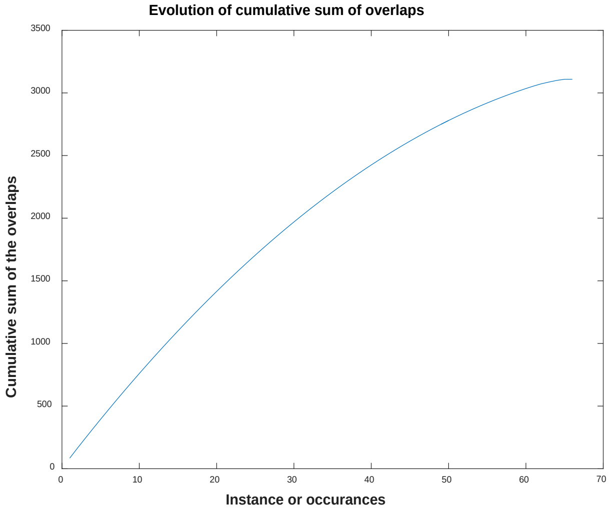
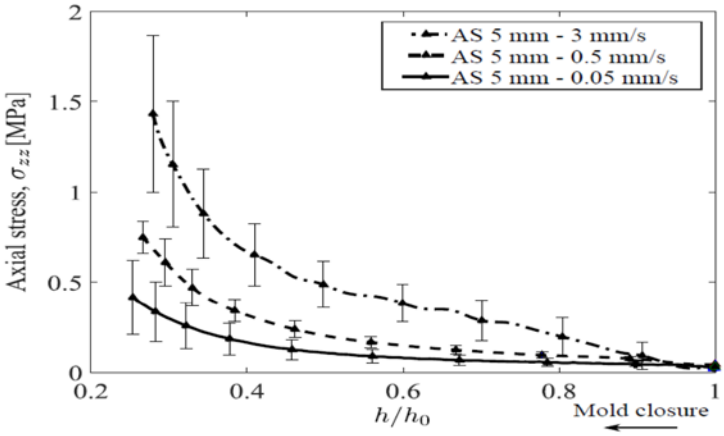


Figure 6: Cumulative Sum of overlaps
Y-axis: cumulative sum of overlaps in figure 5

Levy et al. [10] also came up with a model for the compression molding of randomly oriented carbon fiber PEEK pre-pegs strands. A qualitative relation between the flake overlap in the mold and squeeze force evolution during the compression molding process is also found in their work.

This cumulative force is related to the squeeze force evolution during the compression molding process. Figure 7 shows the squeezed force evolution with the closure of mold- following an increasing trend.



Experimentally observed axial stress vs. instantaneous mold gap for specimens with 5 mm flake size for different closing rates, under AS condition. Limited numbers of error bars are shown for clarity.

Figure 7: Squeeze force evolution for discontinuous woven thermoplastic flakes [1]

This qualitative relation arising from the arrangement of the flakes further requires that the initial material distribution needs to be studied in order to predict the flow behavior during the process.

3 EXPERIMENTAL PROCEDURE

3.1 Material Properties

In this paper the flakes are TenCate TC1100 Carbon reinforced Polyphenylene Sulphide (Carbon Fiber/PPS). Carbon Fiber/PPS are high temperature resistant thermoplastic pre-pegs which find use in the aerospace industry. Linear polymer chain of PPS is a semi-crystalline thermoplastic which is flame resistant, resists chemicals and oils. Carbon fiber/PPS was also used on press-formed ribs and spars for the Fokker 50 undercarriage door, as well as aileron structures on the Airbus A340 [11] . TC1100 tapes come industrially as 6 mm width tapes. The tapes are chopped in to 10 mm x 6 mm flakes. The flakes are chopped using a paper cutter which leaves a margin of error. The flakes used for compression molding show the material properties in table 1:

Material Properties of composite	
Volume fraction (carbon fiber)	60%
Density of composite	1.55 g/cm^3
Matrix Properties (PPS)	
Density of matrix	1.35 g/cm^3
Melt Viscosity at 300°C [20]	200 Pa·s
Processing Temperature	300 °C
Melting Temperature	275-290 °C
Glass Transition Temperature	85-100 °C
Young's Modulus	3700 MPa

Table 1: Properties of PPS flakes [12]

3.2 Compression Molding of Carbon fiber PPS flakes

The filling pattern of the mold is changed to have different count of flakes in the two halves of the mold. The idea is to have a different number of flake overlap distribution so as to understand the effect of variations during the compression molding flow. The flake overlaps have been previously studied in literature [10] as stacking columns. The flakes are used to make a lamina of weight 150 g. They are randomly distributed in the mold unequally with 50 g in one half and 100 g in the other half of the mold shown in figure 8:

Figure 8: Unequal Mold (25 cm x 25 cm) filling

In order to capture or freeze the flow during pressing modifications to the usual pressure cycle are done. The temperature cycle follows the usual one used in the industry. PPS being processible above 300 °C, the pressing was carried out at 320 °C. The laminate is then allowed to cool at room temperature. The experiments basically correspond to squeeze flow experiments [19] with constant loading without completing the consolidation cycle. Figure 9 shows the normal and modified pressure cycle so as to capture the state of laminate during the flow instead of continuing the full consolidation.

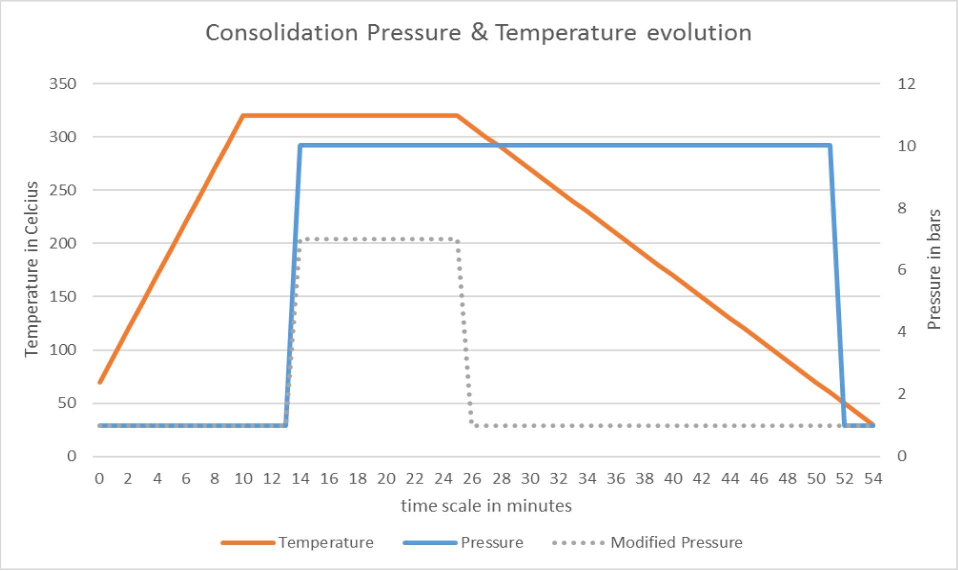


Figure 9: Usual and Modified Pressure Cycles

4 RESULTS AND DISCUSSIONS

Compression molding experiments were carried out for 3 sets of data points for the modified pressure cycle.

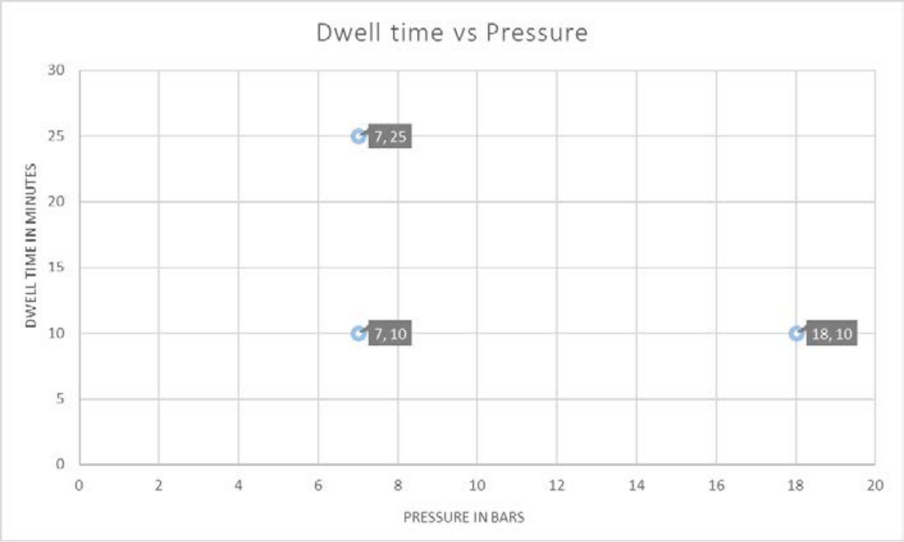


Figure 10: Settings used for compression molding

The laminates obtained for settings 7 bar, 25 minutes and 18 bar, 10 minutes show considerable amount of flow. However the one with 7 bar, 10 minutes does not show much flow on both the sides.

The laminates are then taken for surface microscopy to account for the flow on the heavy and low side flake distributions. Densities shown on the top of laminates are in gram per cubic centimeter measured using buoyancy method with pure ethanol as the immersing fluid. All regions of the composite have densities close to initial composite flake density of 1.55 g/cm^3.

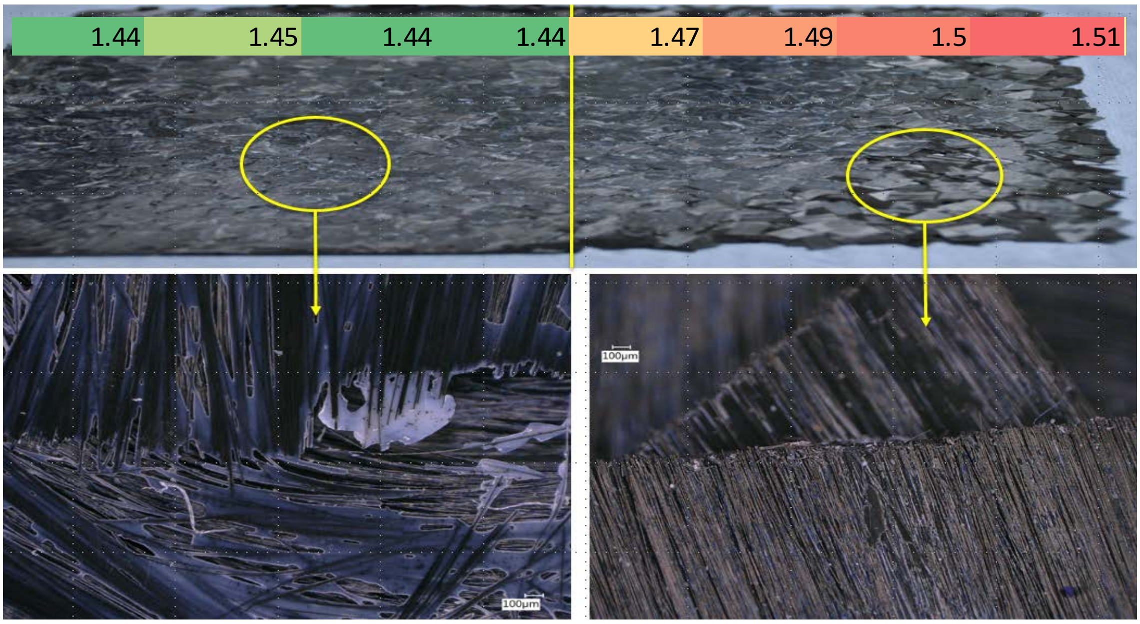


Figure 11-Case I: Pressure 7 bar Dwell time 10 minutes with regional densities on top (Left to Right: High to low material distribution)

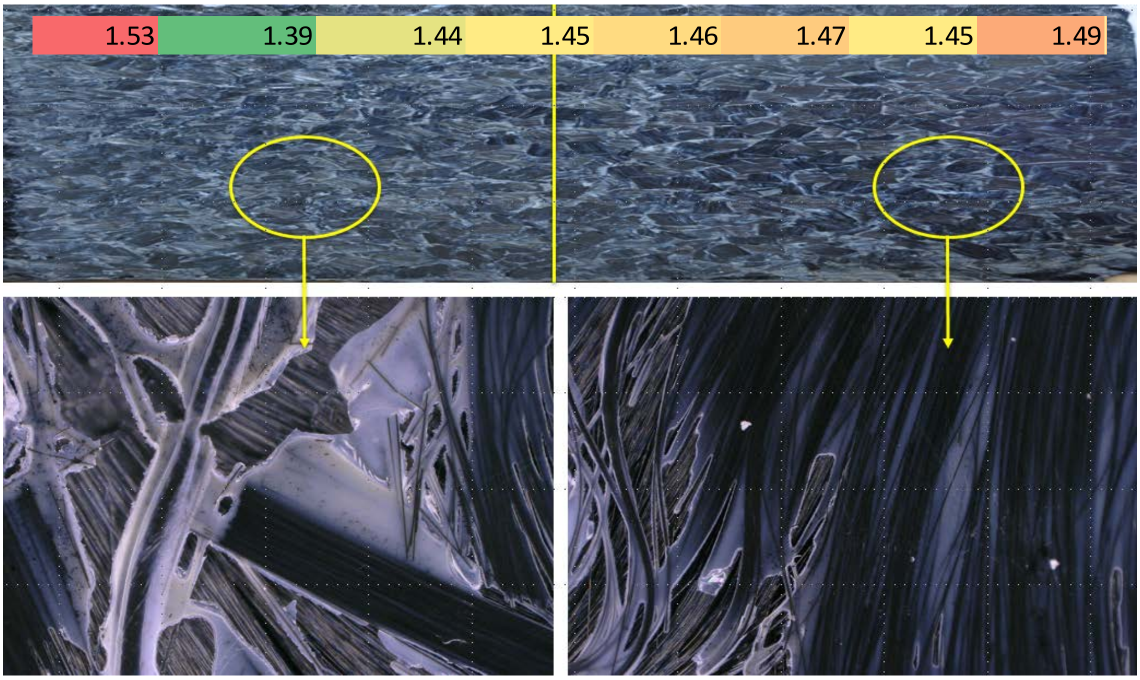


Figure 12-Case II: Pressure 18bar Dwell time 10 minutes with regional densities on top (Left to Right: High to low material distribution)

The flakes undergo considerable deformation in figure 12 and figure 13. There is significant redistribution of the material. They have deformed beyond recognition from their initial state. The images in figures 11, 12 and 13 are obtained from surface microscopy of the consolidated laminates.

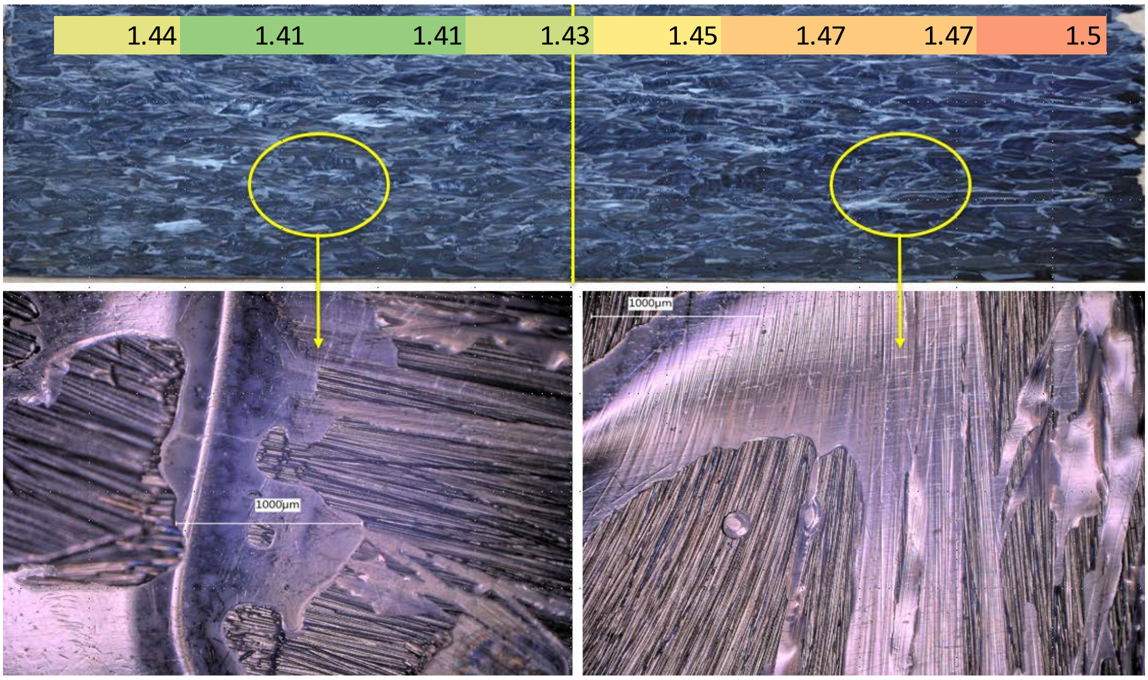


Figure 13-Case III: Pressure 7 bar Dwell time 25 minutes with regional densities on top (Left to Right: High to low material distribution)

Also some flakes are marked with ink whitener in order to capture their state before and after the compression molding flow in the laminate in figure 14.

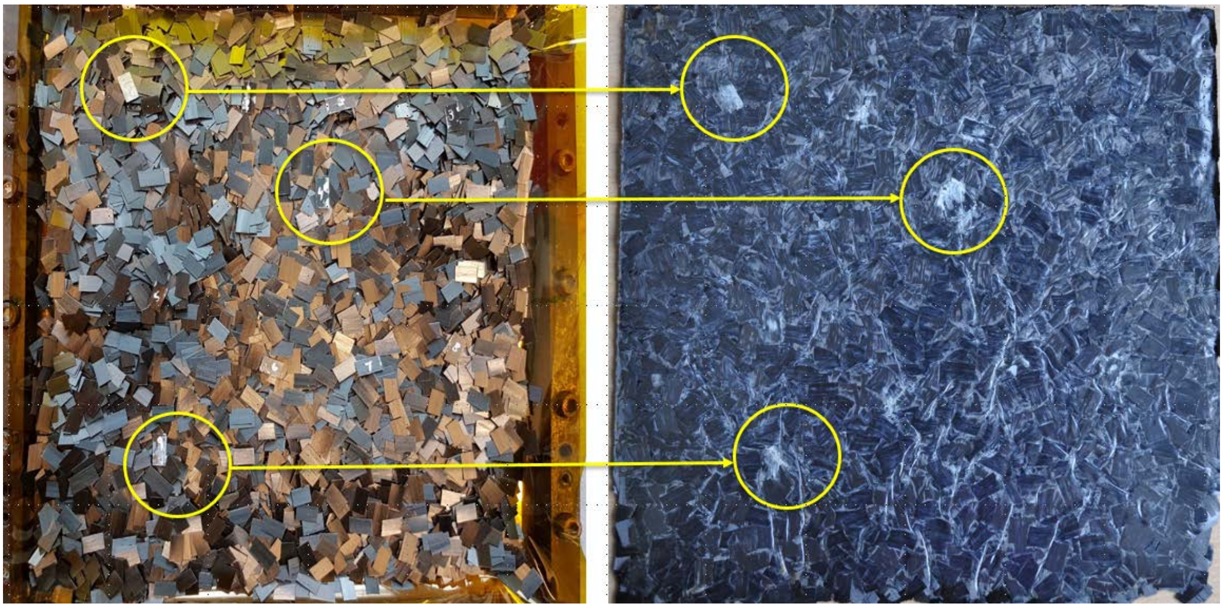


Figure 14: Marking of the flakes

The laminates obtained after pressing is followed by surface microscopy of the laminates which show that flow during compression molding is very much local. This was expected from the flow behavior of the thermoplastics. The movement of the matrix significantly relies on the filling pattern, neighborhood of the Long fiber thermoplastics and pressure cycle. In some cases the matrix moves almost 10% the length of the flakes which is quite significant at the meso-scale level. This can again significantly affect the local fiber volume fraction and also give rise to local fiber bending which is also evident from the microscopy results in figures 15-17. The deformation mechanisms observed in the process are shown in figure 15:

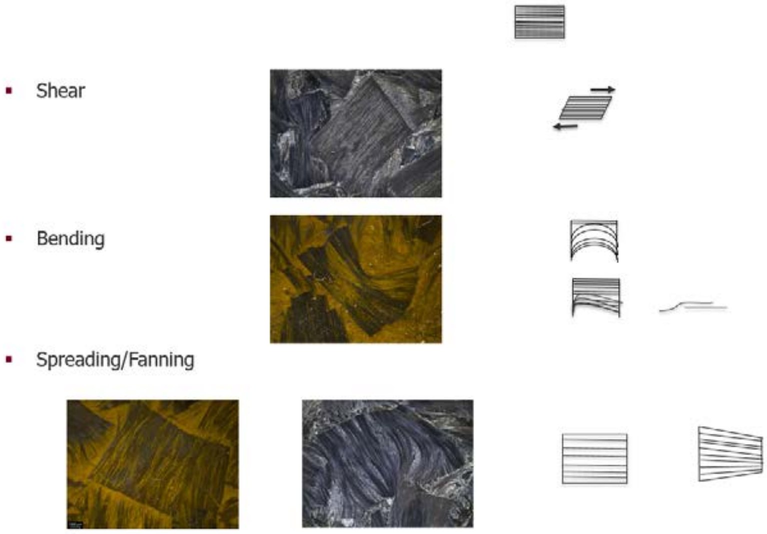


Figure 15: Deformation mechanisms in flakes with respect to original flake on top right

The deformation mechanisms observed in figure 15 can affect the properties and microstructure of composite products [13, 14] .

5 CONCLUSION

Compression molding experiments for different sets of pressure and time were conducted for unequal distributions of UD Carbon Fiber/PPS composites flakes. The material redistributes during the molding process to create laminates of nearly uniform thickness and material density. The higher is the pressure and time used better is the redistribution of the material. The negligible movement or reorientation of flakes in contact with mold surface suggests that no-slip condition is respected at the boundaries. The deformation mechanisms of flakes observed by surface microscopy of laminates were extension, shear, fanning and bending. This is suggestive of a local variation of fiber volume fractions starting with an initial distribution of material in the mold. However, more works need to be done to validate it.

6 FUTURE WORK

The local fiber volume fraction of the laminates need to be checked out using material decomposition experiments like polymer pyrolysis. Density correlations need to be corrected taking into account the presence of voids. The results for all cases needs to be replicated and compared with a normally filled lamina made in both modified and normal pressure cycles. The main aim of the work would be to find the cause of the various deformations observed in this paper and relate it with existing models [15-17] for prediction of fiber orientation and fiber volume fractions. Since the existing models cannot account sufficiently for fiber-fiber interactions and coupling between flow and fiber orientation. They cannot predict mechanisms like affine and percolation flows, fiber jamming and matrix rich regions observed in Rasheed’s work [1] .
The mold filling behavior for long discontinuous fiber thermoplastics corresponds to the concentrated region [18] of polymer flow models due to the high viscosity of polymer and high fiber volume fractions. An accurate simulation tool to predict the variability of long fiber reinforced thermoplastic composites would be main goal of the authors in the future publications.

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