Distribution of fillers and reinforcements in injection moulded thermoplastic composites: Case study of glass bubbles

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Abstract. Apart from the complex nature of natural fibers, studying glass bubbles, as reinforcement phase, is considered a good start to understand the flow behaviour of fillers or reinforcements and hence their distribution in the final products after injection moulding. Glass bubbles are thermally stable within the processing temperatures of Polypropylene. They are also void from the effect of fibre aspect ratio. This work explores the uneven glass bubbles content along injection moulded spirals out of polypropylene (PP). Glass bubbles are experimented at 10 and 30% volume fraction Vf levels. The processing temperature during the injection moulding varies in the range of 180-200°C. Injection pressure is changed from 250 to 1000 bar. Flow speed is adjusted at two levels of 60 and 100 cm/s. The response measurement is the distribution of the glass bubbles across the spiral at different distances from the injection point. The experimental results show the significance of the nominal Vf, pressure, temperature and flow speed on the distribution homogeneity where the bubbles move further in case of 30% at high pressure of 1000 bar and higher temperature of 200°C that eases the flow. At 10% Vf, the fluctuations in the measured Vf is minimal. The reason of the fluctuation in case of 30% Vf at high temperature and pressure is the multi-phase flow where the investigation shows that bigger size of bubbles acquires greater momentum to flow. Scaling-up of this study to the synthetic fibres will cover fragmentation. More up-scaling to the case of the natural fibre thermoplastic composites NFTC can deal with other factors like thermal stability and fibre splicing or branching.

1. Introduction

NFTC have an increasing demand in European market. Natural fibre reinforced polypropylene composite (NF/PP) is significantly represented in the injection moulded NFTC of Europe with 92% of the 15000 ton total production volume of NFTC [1]. However, one concern of these composites is the strong fluctuation of the fibre content in comparison to the nominal fibre content [2]. As a direct result, a product with non-uniform mechanical and physical properties is produced. Accordingly, this will induce negative effect on both the production efficiency and the product cost.

The instability of the fibre content values in the injection moulded product is analysed. The reason is due to the separation flow phenomenon between the phases of fibres and the carrying polymer melt during injection into the mould [2]. This separation between phases is attributed to the difference in density between the fibre and the polymer. Also, the shape effect of the fibres whether it is straight or branched. Kubat et al [3] reported the same observations during the injection of flowability spiral mould using low density polyethylene (LDPE) reinforced with 25.7% glass fibres. Phase separation, as reported by [3] increases directly with increasing the injected spiral length as well as the polymer...
viscosity. The glass fibre content decreases obviously after the injection point. Then it jumps again along the length of the spiral. Helger \textit{et al} [4] studied the filler homogeneity in injected samples of dumb-bell form under different processing parameters. The significant factors affecting the homogeneity and the stable distribution of the glass bead fillers are namely: filler geometry, mould shape, temperature, injection time, filler content and polymer material. Helger [4] explained the phase separation in terms of the filler inertia along the flow direction. Consequently, fillers accumulate near to the axis with higher inertia leading to higher filler content at the far end of the injection moulded samples.

Peltola \textit{et al} [5] considered the phenomenon of changing fibre content, qualitatively, in case of natural fibres namely hemp and flax. Effects of processing melt viscosity and fibre type on the fibre length were investigated. Thermal processing, during injection, reduced the fibre length. This reduction in fibre length is attributed to the increasing shearing force in compounding. Hemp fibres keep higher aspect ratio due to the more fibrillation compared to flax. Higher aspect ratio results in higher reinforcing, Ramzy \textit{et al} [6] studied the flowability and fibre distribution of NFTC in case of hemp and sisal in spiral injection moulds. Increase of polymer temperature, mould temperature and injection pressure led to increased flowability (longer spiral length). The fibre content, similar to [3], drops after the injection point and then goes up again along the spiral length. The fluctuation in sisal was less than that measured in hemp because of the more agglomeration spots found in hemp case.

Table 1. Degradation modes of synthetic and natural fibres [6].

| Mode                | Description | Synthetic fibre | Natural fibre |
|---------------------|-------------|-----------------|---------------|
| Fragmentation       |             | X               | X             |
| Thermal degradation |             |                 | X             |
| Aspect ratio reduction |         | X               | X             |
| Stiffness           | X           | X               | X             |
| Branching           |             |                 | X             |
| Splicing            |             |                 | X             |

Studying the fluctuation of natural fibre is more complex than glass fibres and even more complex than glass bubbles as illustrated in table 1. Synthetic fibres undergo fragmentation during processing. Fragmentation, in turn, reduces the aspect ratio and the expected strength especially when the aspect
ratio fails to satisfy the critical ratio. Stiffness of the fibre [7] is a function of temperature. On the other hand, natural fibre suffers from all the above-mentioned drawbacks as well as other points. Low thermal stability leads to negative change in mass, size and diameter. Another characteristic of natural fibre is the branching and splicing.

Empirical modelling of NFTC during injection needs more understanding of the fibre degradation in shape and size on the flow behaviour [8]. Analytical modelling in literature addresses mostly the synthetic fibres and their flow behaviour without considering the problem of fibre uneven distribution across and along the injection moulding product [9]. Therefore, this work deals with the problem of filler content fluctuation by starting with glass bubbles and not the complex natural fibres. Glass bubbles are characterised with two advantages namely; thermal stability where no change in mass is accompanied, and unity aspect ratio where no effect of rotational moment is occurring.

The objective is to observe and understand the flow behaviour of glass bubbles in spiral flow-ability moulds during injection moulding. The work is limited in this study to the qualitative description. The distribution of the glass bubbles is measured within the cross section and along the spiral length. Effect of the processing parameters (Pressure, temperature, speed) in a well-designed plan is considered in this study which is not thoroughly done in previous work [3].

2. Experimental work
The carrying polymer matrix is a high flow PP of trade name Moplen EP 500 V developed by Lyondell Basell Industries. The glass bubbles are supplied by 3M GmbH with average size of 15 µm and bulk density of 460 kg/m³. Spirals are injected per a design of experiments (DoE) Plan using JMP software as listed in table 2.

| Temperature | Pressure | Flow speed | Fibre content |
|-------------|----------|------------|---------------|
| °C | bar | ccm/s | Vf% |
| 1 | 180 | 250 | 60 | 10 |
| 2 | 180 | 250 | 100 | 10 |
| 3 | 180 | 250 | 100 | 30 |
| 4 | 180 | 1000 | 100 | 30 |
| 5 | 180 | 1000 | 60 | 30 |
| 6 | 180 | 1000 | 100 | 10 |
| 7 | 200 | 250 | 60 | 10 |
| 8 | 200 | 250 | 60 | 30 |
| 9 | 200 | 250 | 100 | 30 |
| 10 | 200 | 1000 | 60 | 10 |
| 11 | 200 | 1000 | 100 | 10 |
| 12 | 200 | 1000 | 100 | 30 |
| 13 | 180 | 1000 | 60 | 30 |

Figure 1 presents the injected spiral and the positions of the segments to be cut and investigated. The spiral mould is an Archimedean spiral. The outer diameter is 250 mm and the step is 9.5 mm. The spiral flow cavity is 6 mm wide and the curvature denoted by the radius R=5 mm.

For each condition, almost 15 spirals are injected, and their lengths are measured for the sake of flow-ability. To measure the glass bubble content as well as its size, segments are cut at the injection point and then every 10 cm. Glass bubbles are extracted by dissolving the PP matrix (using dacaline solvent) at 160°C/ 2 hours. The extracted glass bubbles are then dried and preserved for weighing and further investigation of the glass bubble size development (length and diameter). The extraction process is carried out three times for each compound for three different samples of spirals. The average value of the glass bubble content and the standard deviations are then calculated.
Figure 1. Spiral test for injection moulding flow-ability and the places where the glass bubbles are extracted [Ram18].

3. Results and discussion
The results shown in Figure 2 for Vf=30% match the behaviour reported in [1] for NFTC. The scale of deviation in Vf is small [25.5-29%] relatively in comparison with that in natural fibre case (>10%) as in [1]. Effect of temperature is not observable because Vf fluctuation is reduced under mainly due to the pressure effect. On the other side, it is obvious that low pressure results in less flow-ability. Flow speed is significant where less contents of glass bubbles are measured indicating that portion of the bubbles is hindered before the injection. Also, it is obvious that high pressure increases the Vf fluctuation. This can be attributed to the accompanying increase in spiral length.

Figure 2. Glass bubbles distribution along spiral length at different processing parameters for 30% Vf of glass bubbles.
Reproducibility of the results is ensured by comparing the very close behaviours of samples 5 and 13 shown in Figure 2. Most important is that the measured Vf along the spiral is close to parabolic nature where the bubbles content is reduced just after the injection start and then increases again. The reason is either that more bubbles are flowing separately faster than the hosting polymer, or the bubbles of greater size have more momentum and got separated from bubbles with less diameter. This assumption will be investigated microscopically by samples taken from the spiral at different locations. Figure 3 shows the behaviour at 10% Vf, where the measured Vf show almost constant trend regardless the temperature and the flow speed. Error bars are removed for better illustration.

![Figure 3](image1.png)

Figure 3. shows the behaviour at 10% Vf, where the measured Vf show almost constant trend regardless the temperature and the flow speed.

Figure 3 Glass bubbles distribution along spiral length at 180-200°C injection temperature, 1000 bar with different flow speeds 60 and 100 ccm/s different processing parameters for 10% Vf of glass bubbles. Figure 4 shows the sites taken for microstructure investigation. Three places were cut from the spiral namely the injection start point, the middle of the spiral length and at the end tip. Then at each one of the three places, the glass bubbles were measured and counted at the upper surface, the cross section middle point and at the bottom curved region.

![Figure 4](image2.png)

Figure 4. Places of microscopic investigation for glass bubbles diameter along and across the spiral.

Table shows the significance of the processing parameters namely Vf and pressure on the measured diameters of the glass bubbles across the spiral. This result will be helpful in explaining the
effect of processing parameters with more complex shapes like synthetic fibres and finally branched flexible natural fibres. As seen in figure 5, the larger glass bubbles in diameter concentrate at the end along the flow. Also, relatively bigger bubbles accumulate at the curve region. This cannot be attributed to gravity because the mould is installed in a vertical position. However, the flow has less resistance at the curve than at the flat surface due to the geometrical effect on the computational fluid dynamics of the polymer/glass bubbles multi-phase flow.

| Source                              | Vf [%] | Pressure [bar] | Flow speed [ccm/s] | Temperature [° C] | Vf [%] | Pressure [bar] | Flow speed [ccm/s] | Temperature [° C] | Vf [%] |
|-------------------------------------|--------|----------------|--------------------|-------------------|--------|----------------|--------------------|-------------------|--------|
| Vf [%] (10,30)                      |        |                |                    |                   |        |                |                    |                   |        |
| Pressure [bar] * Vf [%]             |        |                |                    |                   |        |                |                    |                   |        |
| Pressure [bar] * Flow speed [ccm/s]|        |                |                    |                   |        |                |                    |                   |        |
| Flow speed [ccm/s] * Vf [%]         |        |                |                    |                   |        |                |                    |                   |        |
| Temperature [° C] * Flow speed [ccm/s] |        |                |                    |                   |        |                |                    |                   |        |
| Flow speed [ccm/s] (60,100)         |        |                |                    |                   |        |                |                    |                   |        |
| Pressure [bar] (250,1000)           |        |                |                    |                   |        |                |                    |                   |        |
| Temperature [° C] * Vf [%]          |        |                |                    |                   |        |                |                    |                   |        |
| Temperature [° C] (180,200)         |        |                |                    |                   |        |                |                    |                   |        |
| Temperature [° C] * Pressure [bar]  |        |                |                    |                   |        |                |                    |                   |        |
| Vf [%] * Vf [%]                     |        |                |                    |                   |        |                |                    |                   |        |

**Figure 5.** Glass bubble diameter along the spiral length in terms of its position in the cross section.

4. Conclusion and final remarks

The experimental results prove that the distribution of unity aspect ratio particles (glass bubbles) show phase separation. This separation is influenced by the nominal Vf, temperature, pressure and flow speed. The distribution of the glass bubbles behaves parabolically specially at the 30% Vf.

The increase of the measured Vf away from the injection point is interpreted in terms of the size of the bubbles where the glass bubbles of bigger diameter get separated and move further. Diameter of the bubbles across the spiral is influenced by the nominal Vf and pressure. This study can be used further in the analysis of NFTC to exclude the effect of the filler or the fibre size and focus on the other factors like thermal degradation and fibre aspect ratio.

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