SHEET EXTRUSION AND THERMOFORMING OF DISCRETE LONG GLASS FIBRE (LGF) REINFORCED POLYPROPYLENE

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Abstract

The present paper summarises the main aspects and developments of sheet extrusion and thermoforming of discrete LGF composites using the SAFIRE technology (Self Assembling Fibre Reinforcement). During extrusion the long glass fibres are organised into coherent fibre mats which persist into the solid state, and are able to withstand the deformation process that takes part during thermoforming. A process analysis has been performed for extrusion and thermoforming indicating the main individual processes and operations. Both processes have been studied with regard to their performance with the materials used in the studies, namely polypropylene homo- and copolymer, with and without LGF reinforcement. Significant improvements in mechanical properties relative to the unreinforced materials have been found for the extruded sheets and the thermoformed products. Major improvements in processability relative to unreinforced PP have been found for the LGF materials. These are discussed in terms of the coherent fibre mat concept.

1. INTRODUCTION

The work reported here results from a long term research programme on LGF composites under the generic name of Self Assembling Fibre Reinforcement (SAFIRE) in which fibre reinforcing mats are created in the melt during its passage through the process equipment. Previous reports from this laboratory have described results obtained in pipe and sheet extrusion, injection moulding (Bush et al. 1995), and blow moulding (Bush and Tonkin 1997), all of which have been developed at the industrial scale.

Thermoforming has become a major process with a growing range of applications in the automotive, aerospace, agricultural, building and packaging industries. Various types of composite materials have been used in thermoforming, presenting different types of reinforcements, e.g. continuous fibres, woven rovings, discrete aligned fibres (Cogswell, 1992, Schuster and Friedrich 1997). The main problem with continuous and textile reinforcements is associated with deep draw thermoforming because of their limited ability to stretch without disrupting the surface. This problem can be potentially alleviated by combining several laminae each with discrete fibres aligned in one direction into a single sheet so that viewed through the sheet the fibre orientation is tailored to the drawing requirements of a particular thermoforming. LGF composites produced by the SAFIRE technology can produce a nearly isotropic composite in one step using conventional processing techniques like extrusion or injection moulding.

The key to the ability of LGF extruded sheets to be thermoformed into 3D products, and therefore to stand the stretching process without failure, lies in the reinforcing fibre mat concept. During extrusion the fibres are organised in the flow field forming coherent fibre mats which persist into the solid state. These mats give the composite material improved mechanical and thermal properties (Bush and Tonkin, 1997) as well as improvements in processability in the thermoforming process as described below.

Another advantage of the SAFIRE LGF composites is that significant improvements in mechanical properties have been found at relative low concentrations (less than 5% in volume) compared with the other types of thermoformable composites.
The key stages in the SAFIRE technology applied to sheet extrusion and thermoforming are presented in Fig. 1. The first stage is the LGF granule manufacture in the GRANEX line, which is basically a modified thermoplastic pultrusion process, in which the fibre tows are wetted and encapsulated in a thermoplastic matrix and chopped into granules of given length in the range [3, 15mm]. The granules can be made from virgin or recycled materials, or a combination of both. The second stage is the extrusion of thermoformable sheets. In this stage novel fibre management devices are used to separate the fibre bundles in the melt and to form coherent fibre mats with nearly isotropic properties. The third stage is the thermoforming of the LGF composites, by any of the available thermoforming techniques. The final product is then ready for installation. Thermoformed and other scrap material can be recycled and added to the virgin material in the first or second stage of the process.

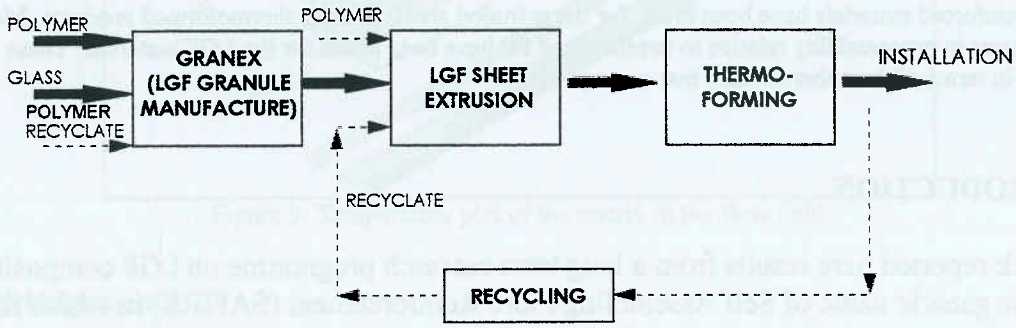


Fig. 1. SAFIRE technology applied to sheet extrusion and thermoforming

2. SHEET EXTRUSION

2.1 Process Analysis

Extrusion of LGF reinforced thermoplastics can be performed in conventional lines with only a few modifications. A layout of an extrusion line for the manufacture of thermoformable LGF reinforced sheet is given in Fig. 2. Significant characteristics of LGF composites with regard to processing are higher melt viscosity and thermal conductivity than the unreinforced polymers. Shear viscosities of more than double of that of the unreinforced polymer have been found for LGF reinforced polypropylene, with a fibre content of % by volume, and similar increases have been found for the extensional case (Bush et al. 1999). These properties have to be considered when determining the optimal flow and thermal conditions for the process.
Conventional extrusion is usually divided in three stages : solids conveying, melting or plastication, and melt conveying or metering. These are discussed next with regard to the processing of LGF composites.

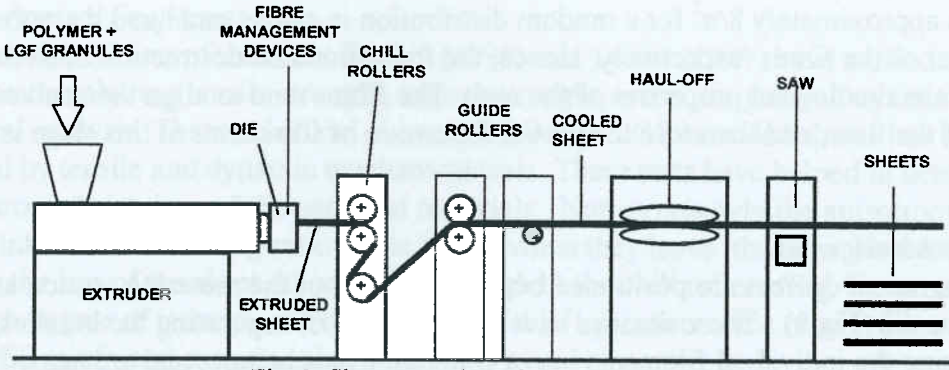


Fig. 2. Sheet extrusion of LGF composites

2.1.1 Solids Conveying

LGF granules are basically cylindrical and in these experiments had a length of 10mm. Since they present a different shape from standard unreinforced thermoplastic granules (approximately squashed cylinders of length c. 3mm), they potentially have a different solids conveying behaviour in the feeding zone of the barrel. However no significant oscillations in flowrate or pressure have been recorded. While still in the hopper, no bridging or interlocking effects have been encountered during processing.

2.1.2 Melting

The melting behaviour of LGF composites during extrusion has been studied by Wolf (1994) with regard to fibre attrition, and by Thielen (1995) with regard to extrusion blow moulding. Fibre attrition is closely linked to the plastication process in the screw channel, since it is believed that it is during that stage that most of the damage to the fibres occur. This is due to the way LGF granules melt when heated. Simple hot plate tests which we have done, show how the granule melts initially at the ends and leaves the fibres ends uncovered by the polymer. These ends are the first to be damaged. Fig. 3 shows diagrammatically the melting behaviour of an LGF granule.

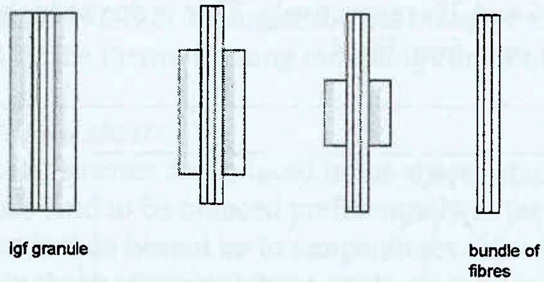


Fig. 3. Melting of a LGF granule on a hot plate

2.1.3. Metering

In the metering section of the extruder, the fibre-reinforced melt presents a considerably higher shear viscosity than an unreinforced polymer melt (Bush et al 1999). The viscosity of melt in this part of the flow path depends on the number of fibre touches N, which is given by (Bush 1993)

$$N = A.c.l/d \tag{1}$$

where, A is approximately $8/\pi^2$ for a random distribution in-plane, and l and d are the length and diameter of the fibres respectively. Hence, the formation and destruction of fibre touches determines the rheological properties of the melt. The fibres tend to align themselves in the direction of the flow, and therefore the partial formation of fibre mats at this stage is only temporary.

2.1.4 Fibre Management

Fibre Management devices are positioned between the exit of the metering section and the mouth of the die (Fig 2). These devices have the functions of separating the bundles of fibres and organising the individual filaments into a coherent mat structure, and are named after their main functions : Fibre Separating Device (FSD) which optimises the separation of the individual filaments in the melt, and the Fibre Lacing Device (FLD), which enhances the formation of permanent mat structures in the melt which will persist into the final solid state.

2.2 Study of the Extruded Sheets

2.2.1 Fibre Length Distributions

Fibre length is one of the factors that determine the formation of a coherent mat in the melt processing of LGF composites (see eq. (1)). The initial length of the filaments, while still embedded in the granule, is around 10mm. During extrusion, a large proportion of the fibres suffer a substantial reduction in their length due to attrition. Previous work on fibre attrition in LGF composites extrusion include those by Blackburn (1995) and by Wolf (1994). The fibre length distribution of a LGF composite can be estimated with the help of an image analyser (Torres and Bush, 1999).

Fig. 4. shows a typical fibre length distribution for an extruded sheet of Polypropylene (PP copolymer, Targor™ 2300L, MFI 6) with 3% of fibre content by volume. The samples were processed at 35 rpm and the temperature profile of the system varied from 170°C to 230°C. The weight average length l_w for LGF-PP at 6% v/v was 4.6mm and for LGF-PP at 3% 5.6mm. The corresponding number of touches N calculated from eq. (1), were 13 and 8 for a fibre volume fraction of 6% and 3% respectively. This is due to higher number of short fibres in LGF-PP 6%, as it can be seen from Fig.4.

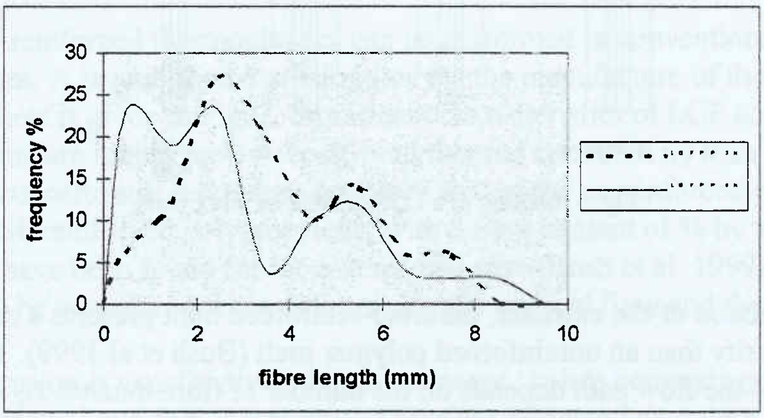


Fig. 4. Fibre length distribution for extruded LGF-PP at 3% and 6% of glass fibre by volume

2.2.2 Mechanical Properties

To characterise LGF reinforcement Tonkin (1998) and Torres (1999) have carried out a wide range of tests including tensile strength, impact strength, creep, fatigue and dynamic mechanical analysis. The mechanical characterisation of LGF extruded sheets has been performed by tensile and dynamic mechanical tests. These tests have helped in determining the anisotropic behaviour of the extruded materials. Not surprisingly the anisotropy is strongly linked to the drawing ratio of the sheets when they leave the die. However, providing the lips of the sheet die are close enough to the chill rollers (Fig. 2), structures with a very high degree of isotropy can be extruded. Table 1 presents some results from tensile tests of LGF reinforced extruded sheets.

Material	UTS (extrusion direction) MPa	UTS (transverse direction) MPa	Anisotropy ratio (UTS ext/ UTS trans)
PP 2300L (unreinforced)	27.8	23.9	0.86
LGF-PP 3% v/v	36.2	26.8	0.74
LGF-PP 6% v/v	54.7	39.7	0.73

Table 1. Ultimate Tensile Strengths (UTS) for extruded sheets (LGF and unreinforced)

In addition to determining the anisotropy in the sheets, Dynamic Mechanical Analysis (DMA) has shown that the softening behaviour in LGF sheets changes less rapidly with temperature than with unreinforced PP sheets, which represents a great advantage when it comes to thermoforming (Bush et al 1998).

2.2.3 Surface Finish

The surface finish of the extruded sheets depends mainly on the calibration and sizing equipment used. This may vary with application. The cooling rollers used in the processing of LGF composites must be combined with a robust transmission system since the cooled sheet is considerably stiffer than an unreinforced one. The sheet haul off speed and roller temperatures have to be controlled accurately and their set values have to be chosen considering that LGF composites have higher thermal conductivities than unreinforced polymers (Bush and Torres 1999). It is important to note however that the definitive surface finish will be determined by the thermoforming moulding conditions.

2.2.4 Shrinkage of the extruded sheets

During the extrusion process, stresses are induced in the sheet, which remain in the material after cooling. These stresses tend to be induced preferentially in the extrusion direction. When the extruded product is then heated up to temperatures close to its melting temperature (processing temperatures in thermoforming) these stresses are relieved from the material and it will suffer shrinkage and distortion, especially in the extrusion direction. A study was performed to determine the shrinkage of unreinforced and LGF-PP extruded sheets. LGF sheets have been extruded at different glass fibre concentrations, and a line speed of 0.3 m/min. Samples of 100mmx100mm were cut from the sheets and put in an oven at 180±1 °C for 30 minutes. After that the samples were left to cool and the dimensions measured after 24 hours. Table 2 shows the percentage shrinkage values found for PP copolymer with and without LGF reinforcement. As can be seen from Table 2., the LGF reinforced sheets present a much lower shrinkage than the unreinforced ones.

Fig. 5 shows an unreinforced and an LGF sample after the shrinkage test. Much lower levels of shrinkage and distortion with regard to the original sample shape can be observed for the LGF reinforced sample relative to the unreinforced one.

Direction	PP	LGFPP 3%	LGFPP 6%
Extrusion	34.5%	2.7%	1.2%
Transverse	(-)8.5%	(-)7.2%	(-)10.0%

Table 2. Shrinkage (%) of unreinforced and LGF-PP extruded sheets. (-) means expansion.

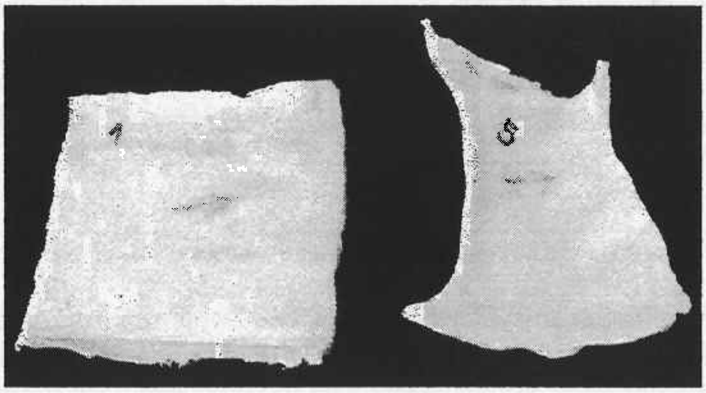


Fig. 5. Samples after shrinkage test.Oven Temp. : 180C. Time : 30min.Left: LGF-PP Right : Unreinforced PP.

3. THERMOFORMING

3.1 Process Analysis

As for extrusion, thermoforming can be divided in a series of operations. Fig. 6. presents a systems view of a basic thermoforming operation. Most thermoforming processes start with a flat sheet, which is heated and stretched. The stretching process may be assisted by vacuum or pressure. Then the deformed sheet cools and solidifies. The last operation is usually trimming and the application of any surface finishes required.

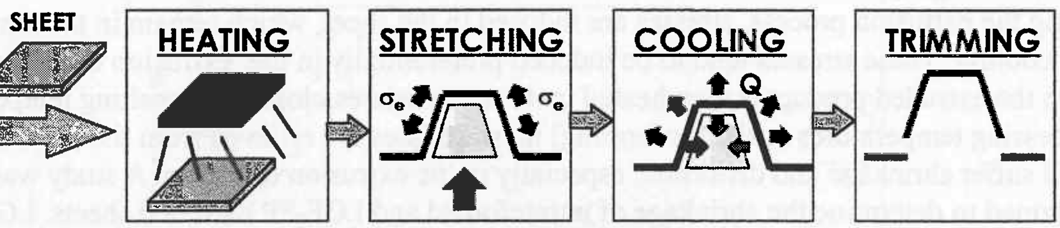


Fig. 6. Basic operations in thermoforming

3.2 Thermoformability

Thermoformability may be defined in various ways. In this paper, thermoformability is defined as the ability of a material to perform all the operations shown in Fig. 5. Therefore, the thermoformability of a sheet depends on (a) its heating behaviour, (b) its stretchability, (c) its cooling and (d) its trimming behaviour. The relative criticality of each of these four factors will vary from application to application. However, if a material cannot perform all the above operations successfully for a typical range of applications, it cannot be considered a thermoformable material. Within the SAFIRE programme the thermoformability of LGF sheets has been characterised by : resistance to local heating effects, breadth of the processing window of the materials, the softening behaviour of the sheets as determined by DMA, stretchability (simulated by hot tensile tests and by rheological tests), final thickness distribution and final mechanical properties of the thermoformed product. The results have been reported elsewhere (Bush et al 1998, Torres 1999, Bush and Torres 1999, Bush et al 1999).

Based on these results and being consistent with the definition of thermoformability given previously, it is possible to state that the thermoformability of polypropylene is considerably improved when it is reinforced with long glass fibres. The improved resistance to localised heating effects, the broader processing window and the progressive softening behaviour of LGF composites are a consequence of the modified thermal properties given to the composite by the coherent fibre mat structure. Stretchability is an important and unique feature of this type of composite which is due to the fact that fibres can move relatively to others without leading to the destruction of the fibre mat. The fibre mat deforms and adapts to a new shape rather like a textile structure with which it shares many features (section 3.3).

3.3 Thermoformed Products

The final properties of LGF thermoformed products have been studied as a method of quality control: Bush et al (1998) have measured the final tensile strength of LGFR thermoformed products, processed at different temperatures. Although a small decrease of strength was found as the temperature was raised within the broad processing window characteristic of LGF materials, important increases in after- moulding dimensional stability have been found for these compounds relative to the unreinforced polymers. Fig. 6(a) shows two thermoformed products, one moulded with unreinforced PP copolymer and the other one with LGF-PP at 6% glass fibres by volume. The unreinforced moulding shows severe after-moulding distortion whereas the LGF-PP presents no distortion. Fig. 6(b) is a picture obtained with the Scanning Electron Microscope (SEM) showing the reinforcing mat structure from the wall of a thermoformed part.

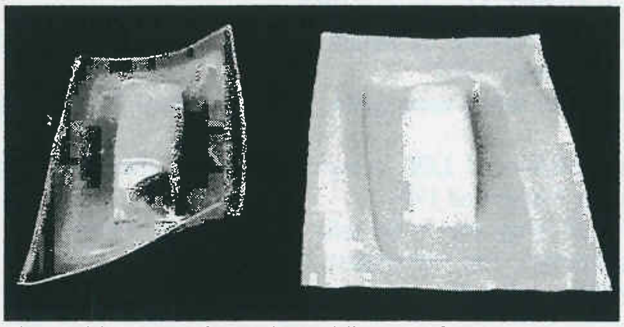
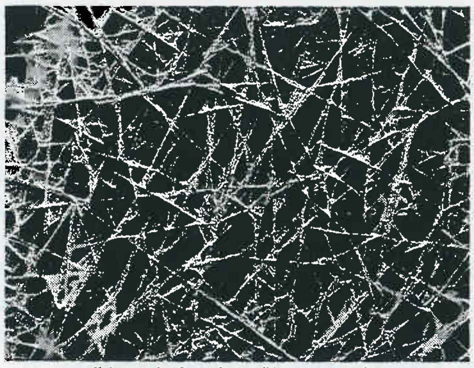


Fig. 6. (a) Thermoformed mouldings. Left : Unreinforced PP, right : LGF-PP with 6% glass fibres



(b) Reinforcing fibre mat from thermoformed part (SEM 15x)

Another important feature in thermoforming as in any other free surface moulding process, is that of surface finish. Surface finish of the free surfaces of thermoformed LGF composites has been studied using Scanning Electron Microscopy (SEM) and the results are reported elsewhere (Torres and Bush 1999). It has been found that when good fibre-matrix contact exists the long fibres do not stick out of the matrix, resulting in a relatively smooth surface of the composite. The level of smoothness depends on the viscosity (or MFI) of the matrix and of the fibre content of the composite. The surfaces in contact with the mould usually present a finish similar to those of the unreinforced polymers, and like them depend on the mould surface and on the cooling rate. It is typical in this case to find a fibre-free layer of polymer in contact with the mould.

4. CONCLUSIONS

A summary of the main aspects and developments in sheet extrusion and thermoforming of LGF composites using the SAFIRE technology has been presented. Sheet extrusion of LGF composites is feasible and has been performed successfully using conventional plastics processing equipment with minor modifications. A functional process analysis of the process has exposed the underlying principles which have to be used in scaling up the process to a fully industrial stage. It has been shown that LGF reinforcement provides the extruded sheets with improved mechanical and thermal properties. In addition, improved thermoformability, as defined in this paper, is a main characteristic of LGF-PP relative to unreinforced PP. Thermoformability has been defined as the ability of performing successfully each of the basic thermoforming operations.

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