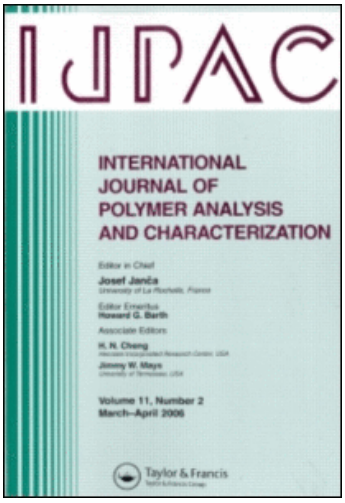

International Journal of Polymer Analysis and Characterization
Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713646643

Granulometric Characterization of Short Fiberglass in Reinforced Polypropylene. Relation to Processing Conditions and Mechanical Properties

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To cite this Article Avérous, L. , Quantin, J. C. , Lafon, D. and Crespy, A.(1995) 'Granulometric Characterization of Short Fiberglass in Reinforced Polypropylene. Relation to Processing Conditions and Mechanical Properties', International Journal of Polymer Analysis and Characterization, 1: 4, 339 — 347
To link to this Article: DOI: 10.1080/10236669508233886
URL: http://dx.doi.org/10.1080/10236669508233886

Int. J. Polymer Analysis & Characterization
1995, Vol. 1, pp. 339–347
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Granulometric Characterization of Short Fiberglass in Reinforced Polypropylene. Relation to Processing Conditions and Mechanical Properties

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(Received September 26, 1994; in final form January 30, 1995)

In most cases, for reinforced thermoplastics, a knowledge of average filler length is not enough information. The entire granulometric filler distribution needs to be known, if structure is to be correlated with properties. We have developed a technique of granulometric determination based on image analysis to analyse rapidly the evolution of the granulometry of short fiberglass. Once images in grey levels from polarized light microscopy have been acquired, each selected fiber on the frame is individualized and labelled before measurement and data treatment. For the fiber, the size factor chosen is the length, or the maximum Feret diameter. With this tool, we have been able to study more precisely the evolution of the fiber length distribution during processing with a polypropylene matrix. Finally, we elucidate the dependence of mechanical properties such as impact and tensile strengths on glass filler size.

KEY WORDS Fiber length, granulometry, image analysis, mechanical properties, reinforced polypropylene, short fiberglass.

I. INTRODUCTION

Reinforced thermoplastics are produced by incorporating reinforcing fillers such as fiberglass into polypropylene. They are being used increasingly in applications where a precise prediction of the material characteristics is essential. The properties and performances of such materials are conditioned by the microtextural characteristics brought by the fillers to the matrix. We can define microtextural characteristics by [1]: the filler parameters, such as granulometry (size) and granulomorphy (shape); and the filler-matrix parameters, such as spatial organization determined by the orientation and dispersion.

In this paper, the effects of processing on the mechanical properties of reinforced thermoplastics are reported as a function of a microtextural characteristic. In particular, we report the effect of varying filler granulometry (length distributions).

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Presented at the 7th International Symposium on Polymer Analysis and Characterization, Les Diablerets, Switzerland, May 24–26, 1994.

I.1. Granulometry of Fiberglass

During the two last decades, there has been considerable interest in granulometry of fibers. Different techniques, such as light scattering [2,3] or sieving [4], have been developed to determine fiberglass granulometry. However, most granulometry determinations (scattering, gravitational measurement, . . .)[5] are based on a spherical shape model, inadequate for investigating fiber-like particles. Therefore, the most usual technique is length measurement of fibers, using microscopy. For each selected fiber, the diameter of the circle circumscribed to the particle is determined. However, manual measurements are very tedious because the basic sampling is made up of hundreds of fibers.

To bypass methods like this, a semi-automatic image analyser process was developed. It is based on a digital table, where each extremity of the fiber representation is noted [6]. A computer program calculates the fiber length from the table coordinates of the endpoints. The length is expressed in the form of length distribution and calculated averages.

The last evolution is, with a minimization of human interventions and faster characterization, automatic images analyser [7–9]. The principal problem encountered particularly for long fiberglass, is the dispersion of fibers for image acquisition. Contacts between fibers must be minimized, in order to individualize each one easily, and to facilitate image treatment. To improve the dispersion, Sawyer [9] treated the fiber surface. The treatment is based on a silane-glass reaction with methyl trimethoxy silane catalysed by acetic acid and a cationic lubricant. However, there are a great diversity of other solutions [1,4,9] likely to improve, with different efficiencies, the dispersion of fibers.

For poorly dispersed fibers, it is also possible to treat images and to separate connected fibers with recently developed routines [7], although with current personal computers the treatment time is rather long.

I.2. Influence of the Processing on Fiberglass Granulometry

According to Turkowitch and Erwin [6], the principal causes of fiber damage are fiber-fiber interaction, fiber contact with processor and fiber interaction with polymer. These and other authors have shown that considerable fiber breakage occurs during processing. The breakage of fibers depends on their initial lengths [10] and is affected by processing conditions [4,6,10–14]. The most significant important cause of fiber breakage is shearing. For example, Vaxman et al [15]. observed extensive fiber damage at low shear. The conditions under which the fibers are mixed into the polymer [11,12,14,16] bring about significant reductions in fiber lengths.

I.3. Granulometry—Mechanical Properties Relationship

The relationship between fiberglass length and macroscopic mechanical properties is complex [17–20]. It is well known that the tensile behaviour is dependent on the fiber length. Indeed, elastic modulus can be described by Cox's rule of mixture [21], involving a fiber length factor.

Fracture mechanisms have been proposed by different authors [17]. They point out relationships between fracture energy and fiber length. In the same way, a critical length is introduced in the Kelly and Tyson's model [22] (load transfer matrix-fiber by sliding) which determines the kind of fracture mechanism.

II. EXPERIMENTAL PROCEDURES

II.1. Materials Used

Three different kinds of fiberglass filler (Vetrotex, Chambery-France) were used with different shape factors (f_s):

- Long fiber glass (**LFG**) with $f_s > 100$, obtained from 4.5 mm cut thread.
- Short fiber glass (**SFG1** and **SFG2**), $100 > f_s > 1$.
- Fiberglass powder (**FGP**); $1 > f_s$.

Shape factor is the ratio of length to diameter. Since filler populations have a constant diameter (13 micrometers), shape factor is characterized here by the length. The composites were prepared with a polypropylene matrix (Appryl 3030 MN1-Atochem). The filler weight content was constant at 30%.

II.2. Short Fiberglass and Fiberglass Powder Generation

Short fiberglass and fiberglass powder were obtained by fragmentation of the longest fibers. After fragmentation, short fiberglass and powder were separated by pneumatic selection (Alpine ATP50 Rotoplex, Augsburg-Germany). During another step, short fibers were divided by the pneumatic process into two populations (SFG1 and SFG2) characterized by two different average lengths, with two different granulometric distributions.

II.3. Sample Preparation

Processing conditions were identical for all specimens injected. A co-rotating twin screw extruder (Clextral BC45, Firminy-France) was used to prepare fiberglass-polypropylene pellets. The diameter of the screw was 50 mm. The extrusion temperature was 230°C and the screw speed was 100 rpm. An injection moulding machine (Billon, Bellignat-France) with a clamping force of 90 tons was used to mold standard dumbbells. The screw speed was 100 rpm and the injection temperature was 250°C. The holding pressure was 20 bars. The mold temperature was 20°C and the total cycle time was 1 minute. Dumbbell specimens produced had an active portion 10 mm wide and 4 mm thick (according to French standard NFT 51-034 1981). The Charpy samples were cut from the central part of the dumbbells to get samples without notches whose dimensions were 10*4*60 mm (according to French standard NFT 51-035 1983).

For granulometric analysis of the fibers in the molded samples and in the pellets (extruded product), the matrix was pyrolized in a muffle furnace for two hours at 500°C to separate the fibers.

II.4. Tensile Testing

An electro-mechanical tensile tester (Adamel Lhomargy DY26, Ivry sur Seine-France), coupled with a servo-controlled, optical, elongation-measuring system, was used to study the load-elongational behaviour of the specimens. A computer controls the testing machine and facilitates data processing. The testing conditions for elastic modulus deter-

mination was room temperature (20°C) and a speed of 1 mm/mn for the dumbbell calibrated length of 50 mm. For each determination, 10 specimens were tested.

II.5. Impact Strength Studies

A mechanical impact tester (Zwick S102, Ulm-Germany) with a 4 Joules pendulum was used for Charpy tests. The testing conditions for impact strength measurements were room temperature (20°C) and 10 specimens tested for each determination. The distance between supports was 40 mm. Impact strength is usually quoted as energy per unit of area.

II.6. Short Fiberglass Granulometry Determination by Image Analysis

The granulometric approach chosen is individual analysis, where each particle must be separated and individually measured [7]. The process was comprised of three principal steps [1]:

—Image acquisition: a system composed of a sensor (CCD camera) coupled with microscopy allowed images to be captured. Particles were dispersed between two glass microscope slides, in a solution of diethylene glycol (dispersing). The fiber concentration was very low to minimize fiber contacts. For a representative sampling, more than 800 fibers were analysed [24], requiring more than 25 different images. Observations were made with a polarizing microscope (Leitz, Wetzlar-Germany) in transmission mode.

—Image treatment: once the image had been filtered of noise, each selected fiber was individualized and labelled. Captured images were digitalized with a video card. The image treatments improved contrast and corrected light intensity variations. This was done using morphological tools [8]. Initial images (512*512 pixels) were in grey levels (256 levels). By thresholding, they were transformed into binary images (2 levels). Fibers were coded to 1 and the background to 0, each fiber thus being individualized. All the image manipulations were done with a computer and dedicated software: Visilog (Noesis, Vélizy-France).

—Quantitative analysis: for each labelled fiber, we determined the size factor. The data treatment allowed us to determine length distributions. The retained size factor for this cylindrical particle is the exodiameter or the maximum Feret diameter [23]. Each analysed particle had to be totally included in the frame of measurement. Feret diameter data were corrected for the probability of inclusion, longer fibers having a lower probability of inclusion. The Miles and Lantuejoul method [7] allowed us to correct this bias. Usually, granulometric determination offers two kinds of representation:

—Granulometry in number, where each particle is weighted by its own number. An average number length could be represented by:

$$L_n = \frac{\sum_i d_i \times n_i}{\sum_i n_i}$$

n_i is the number of fibers with a Feret diameter:d_i.

—Granulometry in weight, where each particle is weighted by its own length (fiber diameters and glass density are constant). A representative average weight length could be:

$$L_w = \frac{\sum_i d_i^2 \times n_i}{\sum_i d_i \times n_i}$$

Following Tancrez [17], we used a dispersity factor (D), representing the granulometric range:

$$D = \frac{L_w - L_n}{L_n}$$

and a variation coefficient (VC), the ratio between standard deviation and average length.

III. RESULTS AND DISCUSSION

III.1. Granulometric Distribution of SFG1 and SFG2

As with the majority of powders obtained by a breaking process, granulometric distributions have a log-normal shape [5] (see Figs. 1 and 2). The fitting of the granulometric distributions to a log-normal law was done with acceptable correlation coefficients (see Table II). Using the equation coefficients resulting from the fitting to a log-normal law, it was possible to determine other estimators of the average lengths (in number and weight) and dispersity coefficients (see Table II). Average lengths determined by this approach are lower, due to the length data display (histogram presentation).

In Table I, results of short fiberglass before processing show a sharp difference between both average lengths of SFG1 and SFG2. These quantitative results were confirmed by the granulometric distribution presentation in Figures 1 and 2. The graphs show clearly the positive result of the separation of two short fiberglass populations (SFG1 and SFG2),

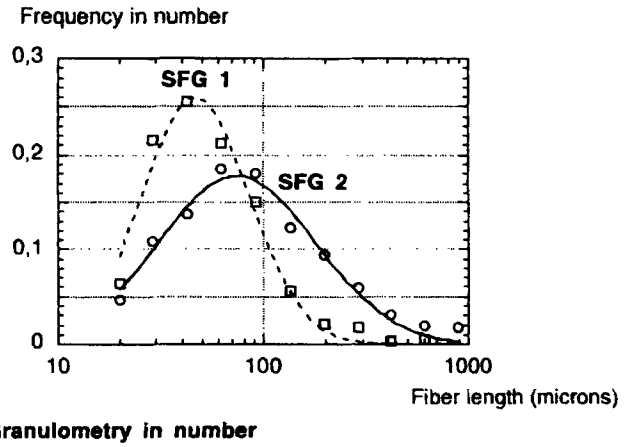

FIGURE 1 Fiber length distributions in number of SFG1 (initial) and SFG2 (initial) with log-normal fittings.

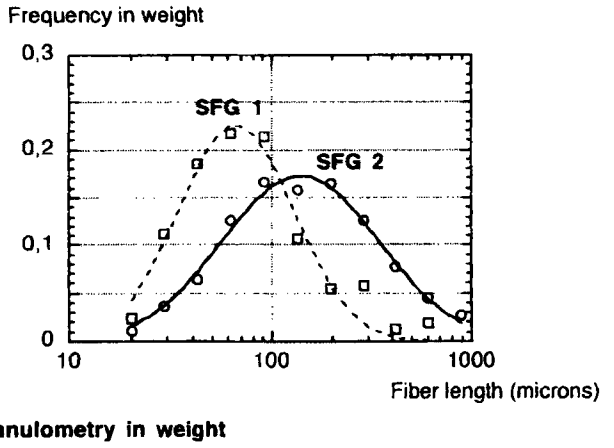

FIGURE 2 Fiber length distributions in weight of SFG1 (initial) and SFG2 (initial) with log-normal fittings.

TABLE I

Granulometric data in number and in weight

	SFG1 (initial)	*SFG2 (initial)*
Average length in number (L_n)	66 microns	127 microns
Average length in weight (L_w)	130 microns	297 microns
Variation coefficient (VC_n)	0,99	1,16

TABLE II

Results of fitting to log-normal curves

	SFG1 (initial)		SFG2 (initial)	
	In number	In weight	In number	In weight
Estimation of average length (microns)	56	82	108	206
Variation coefficient	0,65	0,73	1,06	1,11
Correlation coefficient (fitting)	0,987	0,984	0,985	0,991

made by an air selected process. In fact, we obtained two initial populations of short fiberglass different in size and shape ratio.

III.2. Evolution of the Granulometry of SFG1 and SFG2 with the Processing

The results of the evolution of the granulometry with extrusion and injection processes are presented in Table III. We can note a weak but progressive decrease in average length, for both granulometry by number and by weight, as the processing continues.

The variations in weight-average fiber lengths (L_w) are more important than those in number averages (L_n). In fact, the average L_w is more sensitive to fiber length. As confirmed by the representation in Figures 3 and 4, we show that it is mainly the longest fibers that deteriorate by breakage during the processing.

TABLE III

Granulometric data and processing

	Average length in number (*Ln* in microns)	Average length in weight (*Lw* in microns)	Variation coefficient (*VCn*)	Dispersity coefficient (*D*)
SFG1 (initial)	66	130	0,99	0,97
SFG1 (after extrusion)	62	124	1,00	1,00
SFG1 (after injection)	56	99	0,88	0,77
SFG2 (initial)	127	297	1,16	1,34
SFG2 (after extrusion)	119	234	0,98	0,97
SFG2 (after injection)	107	209	0,97	0,95

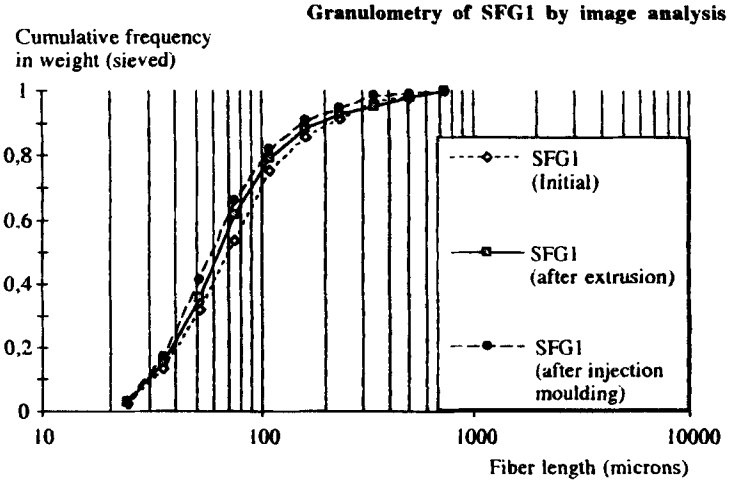

FIGURE 3 Evolution of SFG1 granulometry (in weight) with the processing.

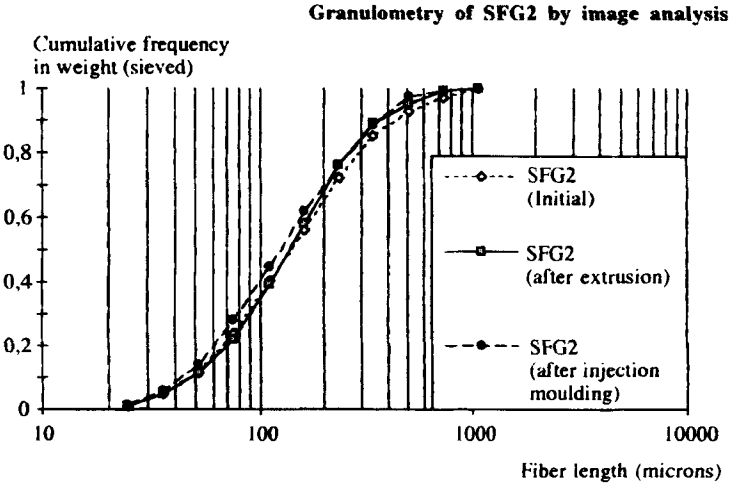

FIGURE 4 Evolution of SFG2 granulometry (in weight) with the processing.

The dispersity coefficient (D) and variation coefficient (VC) follow the same trend, and can be interpreted in the same way. For SFG2, dispersity decreases with the processing, due to the disappearance of some of the longest fibers by fracture and, consequently, a narrowing of the granulometric distribution. This effect is less important for SFG1, because its initial granulometric distribution is narrower.

However, with processing conditions chosen, the size of short fiberglass is little affected by processing.

III.3. Relationship Between Granulometry and Mechanical Properties

The results of impact (Charpy) and tensile tests are presented in Table IV. A statistic equality Student's test is carried out on the averages so that each result appears significantly different from the other with a confidence threshold greater than 99,9%. A synthetic representation is presented in Figure 5. Mechanical properties variations with filler length are important. The evolution of the impact strength as a function of the filler size is inversely proportional to the elastic modulus evolution. For such material, a compromise in filler size is necessary to optimize the mechanical properties.

IV. CONCLUSIONS

We have developed and described a granulometric tool based on automatic image analysis. With this tool, we can analyse more precisely the evolution of short fiberglass length distribution during processing. For our processing conditions, fiber mechanical degradation is low and affects mainly the longest fibers, with little effect on the number average length.

Different filler sizes were chosen in order to test the mechanical behaviour of reinforced thermoplastics. It is interesting to note different responses even for two such similar products as SFG1 and SFG2, which present principally a different granulometric distribution but are in the same granulometric range. The main trend is that the evolution of the resilience as a function of the filler size is inversely proportional to the elastic modulus evolution.

TABLE IV

Stiffness and impact strength data

	PP Matrix	LFG	FGP	SFG1	SFG2
Elastic modulus	1529	6847	2855	3088	4280
(MPa)	(2,5%)	(3,3%)	(2,6%)	(1,9%)	(2,6%)
E composite/E matrix	—	4,48	1,87	2,02	2,80
Impact strength	—	16,9	27,6	22,9	19,9
(kJ/m^2)		(6,0%)	(8,8%)	(3,1%)	(6,4%)

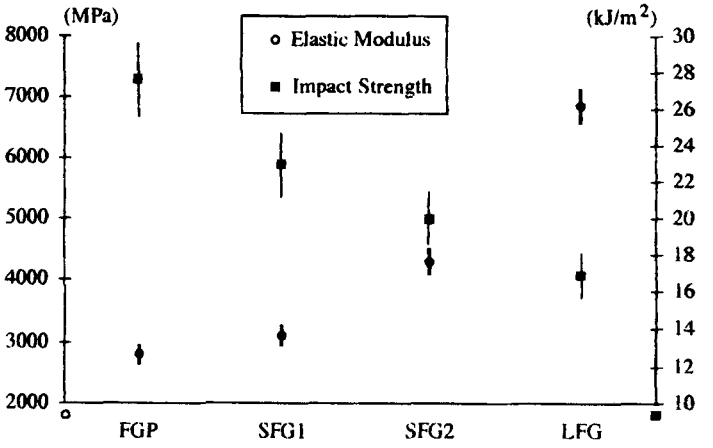

FIGURE 5 Results of impact and tensile test on powder, short and long fiberglass.

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