FIBER LENGTH DISTRIBUTION MEASUREMENT FOR LONG GLASS AND CARBON FIBER REINFORCED INJECTION MOLDED THERMOPLASTICS

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Abstract

Procedures for fiber length distribution (FLD) measurement of long fiber reinforced injection molded thermoplastics were refined for glass and carbon fibers. Techniques for sample selection, fiber separation, digitization and length measurement for both fiber types are described in detail. Quantitative FLD results are provided for glass and carbon reinforced polypropylene samples molded with a nominal original fiber length of 12.7 mm (1/2 in.) using equipment optimized for molding short fiber reinforced thermoplastics.

Introduction

Long fiber injection molded thermoplastics (LFT) offer an attractive combination of desirable mechanical properties, low cost, and rapid processing make them suitable for structural or semi-structural automotive applications. Processing parameters and tool geometry used during injection strongly influence resulting fiber orientation, entanglement, fiber length attrition, matrix properties and residual stresses [1]. This work is part of a larger effort to develop a suite of predictive tools which would use process parameters, tool geometry and a limited number of experimentally obtained quantities as input and predict part's fiber architecture and mechanical properties [2,3]. In this paper, we focus on experimental techniques for fiber length measurement. Whether the measurements are used as inputs to models or for validations of models, a repeatable and consistent method is necessary if the quantitative results are to be used in the description of fiber architecture and calculations. An informal inquiry into fiber length distribution (FLD) measurement techniques used by the industry revealed that measurements on short fiber reinforced injection molded parts are usually performed by removing the matrix, selecting a sample of fibers, spreading them and measuring lengths from a set of digitized images. This general approach was adopted as a starting point for the development of the technique for LFT materials.

The initial fiber length in pellets is uniform and equal to the nominal length of the pellets. The fiber length subsequently degrades as the material passes through the screw and multiple flow restrictions. This process of fiber length attrition is statistical in nature and results in fiber length distribution in a given part at a particular point. It is therefore necessary to measure a rather large numbers of fibers to obtain meaningful data. The measured sample of fibers should represent fiber length distribution at a desired point in the part. This means that fibers cut during separation of a sample must not be measured and that their removal must not result in skewed distribution. This may not be a trivial issue with injection molded samples, since the injection process results in highly entangled and interlocked architecture. Furthermore, the selected fibers must truly represent our point of interest, not just a location conveniently accessible for measurement. Injection molded parts posses distinct core and shell structure [4]

(Figure 1) and our observations indicate that fiber length differs in these regions.

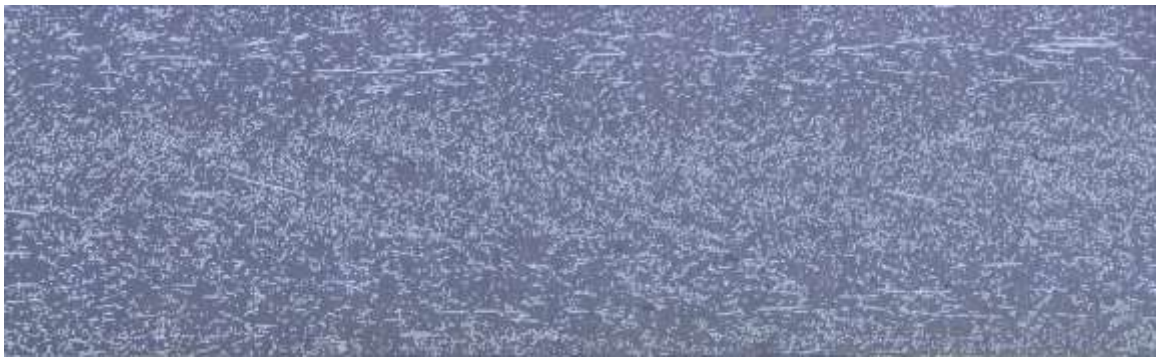

Figure 1. Micrograph of polypropylene/glass injection molded center-gated disk showing distinct core and shell structure.

Selecting a small amount of fibers with tweezers after removal of the matrix seems to be the standard technique for sample isolation in the industry. Aside from the obvious risk of fibers breaking and short fibers being dropped, this technique is likely to skew the results towards the fiber lengths seen in the shell. After isolating fibers from the region of interest, it is necessary to disperse individual filaments over a substrate well enough so that individual filaments and their ends are clearly distinguishable in a digitized image. Dispersion of long carbon fibers proved to be a challenging problem. The technique for dispersion of long carbon fibers will be discussed in detail.

The imaging technique depends on the desired resolution of digital images. Using a microscope provides high resolution, however long fibers may span several images. Stitching images accurately enough and avoiding creation of artificial fiber ends is time consuming and sometimes inaccurate. Scanning or imaging a larger area in one shot is far more expedient. Final measurement of fiber length is performed manually, because a suitable software for automated length measurement was not identified or developed.

Samples

Polypropylene pellets with glass or carbon fibers were procured from Montsinger Technologies and used to mold samples. The injection molding machine and molds were not optimized for process involving long fibers, which may explain rather significant fiber length attrition described later in the paper. The mold temperature was held at 78° C, while the inlet temperature of the melt was 240°C. Two test geometries were utilized for this study: an ISO plaque (Figure 2) and center-gated disk specimens (Figure 3).

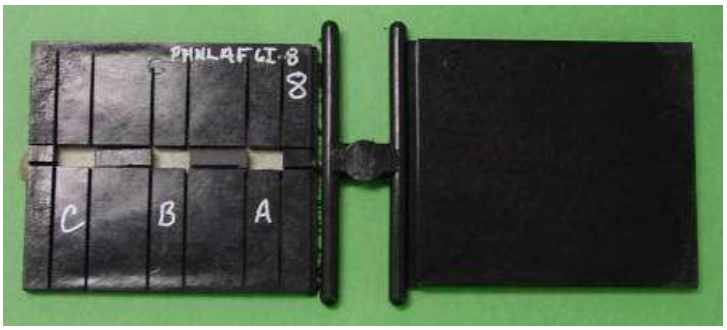

Figure 2. ISO plaque

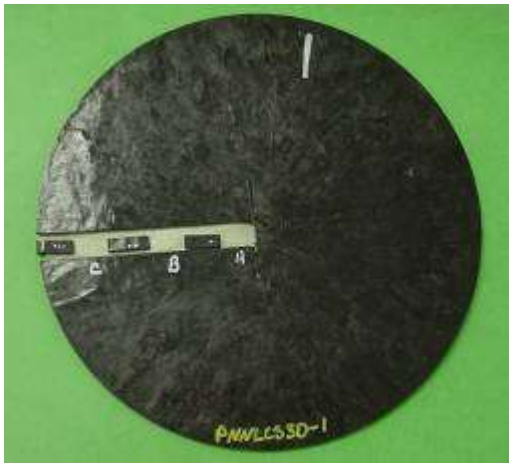

Figure 3. Center-Gated Disk

The ISO plaque was 90 mm long and 80 mm wide, while the disk specimen was 177.8 mm in diameter. All samples were 3 mm thick. Pellets containing either glass or carbon fibers were nominally 12. 7 mm (0.5 in.) long. The fiber content of pellets by weight was 40% for glass and 31% for carbon fiber, giving equal volumetric fiber content of 19.2% for both material types. In the subsequent sections, the specimens will be referred to by four letter designations. The first letter indicates the material type: A - polypropylene/glass fiber, B - polypropylene/ carbon fiber. The second letter indicates injection speed: S - slow 16.4 cc/sec (1 cu in./sec), F - fast 131.1 cc/sec (8 cu in./sec). The third letter indicates the specimen thickness in millimeters, while the last letter indicates the specimen geometry. Letters I and D designate ISO specimen and center-gated disk respectively.

Procedure

Preliminary burn-off tests revealed a wide range of fiber lengths present in molded samples. Standards and measurement techniques developed for short fiber reinforced plastics could not be used due to the wide range of fiber lengths and the tendency of the long fibers to bend. Procedures consisting of major steps identical to short fiber length measurement have been developed for fiber length measurement of glass and carbon filled LFTs. The execution of these steps varies due to different physical properties of glass and carbon fibers. The steps of fiber length measurement are:

1. composite coupon isolation
2. constrained removal of matrix material
3. fiber sample isolation
4. filament dispersion
5. imaging and individual filament length measurement

The objective of FLD measurement is to obtain distribution characterizing the fiber microstructure at a given point. Since the fibers are of finite size, this task is accomplished by measurement of FLD of fibers contained in a small volume and assuming that such measurement represents FLD at the point of interest. The size of a coupon cut out from a specimen must be large enough to prevent measurement of cut fibers. Materials examined in this project were molded with initial nominal fiber length of 12.7 mm (0.5 in.), therefore the distance of the center of the section - the location of interest - from any cut edge must be 12.7 mm or more. Cutting out and removing matrix from a larger coupon will not present any benefit, since all additional fibers will be discarded and only fibers passing through a small region in the center of this coupon will be counted.

The composite coupon is placed in an aluminum sheet metal form whose inside dimensions correspond to the shape and area dimensions of the coupon. The sheet metal form provides restraint at the edges of the coupon during the matrix removal step, which was performed via burn-off at 450°C. The form and its contents are sealed at the top and bottom with aluminum foil lids. The form height dimensions are greater than the coupon thickness to provide for some expansion of the fibers during matrix burn-off, which is caused by the elastic energy stored in the bent fibers. This expansion facilitates the separation and spreading of the filaments during the dispersal step that follows later in this procedure. Burn-off in an inert atmosphere as well as acid digestion was performed to validate that fibers are not damaged during atmospheric burn-off.

The fiber sample isolation step requires isolating the central fibers of the residual fiber mass for subsequent collection and characterization. The process involves inserting a needle attached to a hypodermic syringe loaded with a liquid epoxy through the center of both the top and bottom lids of the aluminum form containing the fiber. The needle is withdrawn from the form and through the fiber stack while dispensing the epoxy through the needle at a constant rate. The continuous stream of epoxy results in an approximately cylindrical column of resin that extends through the entire thickness of the fiber stack. The number of fibers collected from the specimen is proportional to the epoxy plug volume (diameter of the cylinder). Factors that control the volume include the resin's viscosity, the needle gage and the withdrawal rate of the needle through the fiber stack. Epon 828 resin mixed with 55 phr Versamid 125 supplied by Hexion along with an 18 gage hypodermic needle was used successfully for our measurements. The resin injection was conducted with the aid of an actuator for better control of needle withdrawal rate for the measurements provided below. After injection, the epoxy is allowed to gel at room temperature and the resin is subsequently cured. Filaments in the vicinity of the epoxy are thereby bonded in-situ to the epoxy cylinder. The bonded fibers are extracted by removing the fiber stack with epoxy plug from the aluminum form, manual shaking of the specimen and applying short blasts of low pressure air. The success of using low pressure air for removal of fibers not attached to the plug is largely dependent on the disentanglement of the fibers, which occurred during the expansion in the first burn-off. Figure 4 shows plots of the combined extracted fiber-and-epoxy plug weights as a function of resin needle withdrawal rate for carbon and glass composites evaluated using this procedure. The needle withdrawal rate was found to be the dominant factor in controlling the resin deposition rate compared to the

variations in pressure applied on the syringe plunger. The plot shows that the faster the resin deposition rate, the smaller weight plug specimen (epoxy and fiber) collected. Although the glass composites have nominal fiber weight contents that are higher (40 weight percent) than carbon composites (30 weight percent), the extracted glass fiber samples in this experiment have lower weights than their carbon fiber counterparts. It is speculated that this is because the carbon composites tended to have longer fiber lengths attached to the epoxy plug than the glass specimens, and that these longer fibers contribute to the higher overall specimen weight.

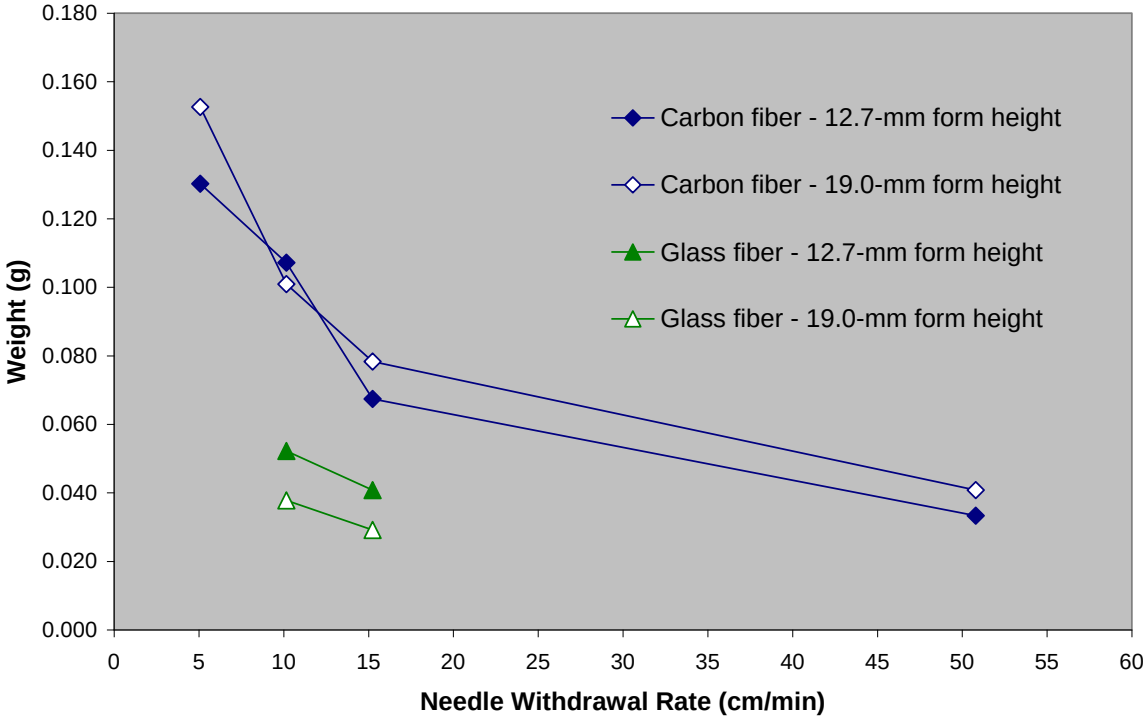

Figure 4. Plug weights as a function of resin deposition rate

Figure 5 is a photograph of an extracted carbon fiber-and-epoxy plug specimen. Long and shorter fibers are visibly attached to the plug specimen along its length. It is reasonable to assume that FLD of these fibers represents FLD at the center of the cylinder because of the small diameter of the cylinder.

Figure 5. Carbon fiber and epoxy plug.

Prior to dispersion of the fibers in the plug, burn-off of the epoxy resin is performed at the same temperature as the first burn-off. The dispersion techniques differ for glass and carbon fibers.

The ease of glass fiber dispersion depends on the proper amount of expansion during the first constrained burn-off. Complete constraint results in an entangled mass of fibers of approximately original sample thickness because glass fibers anneal at burn-off temperatures. Attempting to separate fibers from this entangled state is impractical. Constraint-free burn-off would result in dislocation of fibers, thereby preventing subsequent isolation of a representative sample. Dispersion of glass fibers from a properly processed sample can be performed readily by transferring mass of the fibers into a glass Petri dish and tapping the sides gently. Short fibers are the first to be attracted by the glass surface of the dish and separate from the fiber mass. Once most of the dish surface contains fibers, the remaining mass can be transferred to another dish. It is usually possible to disperse nearly all the fibers without using direct mechanical action, however wooden sticks can be carefully used to gently loosen any entanglement. It is prudent to perform any mechanical manipulation of fibers under low magnification microscope to make sure that the fibers are not being broken in the process. Because the use of plastic Petri dishes makes dispersion of glass fibers more difficult and the surface of the dish can be easily scratched, the use of glass Petri dishes is suggested for the dispersion. It is important to keep these dishes clean and free of scratches to make sure that these artifacts are not counted as fibers.

Long carbon fibers exhibit the tendency to form filament clumps, therefore preventing measurement of individual filament length. A number of unsuccessful trials involving liquids and various chemicals did not result in an acceptable technique. Mechanical methods usually caused breakage of the filaments.

The newly developed dispersal process relies on a corona field provided from a high frequency generator to create static charge in the vicinity of the carbon filaments and a substrate surface. In this case, the substrate is a ply of paper supported on a notebook pad of paper that is approximately 6-mm thick. The paper stack is elevated by supports so that the corona field from the tip of the high frequency generator can be applied from beneath. A portion

of the residual fiber clumps are placed on the paper and the high frequency generator is switched on to generate the corona field beneath the paper stack. As the paper and carbon filaments acquire static charge, two phenomena occur that aid in separation and dispersal of the carbon filaments. One is that the carbon filaments seem to repel each other and, when the residual fiber mass is sufficiently small, fly apart and disperse. The other is that the carbon fibers are attracted to the charged substrate and tend to adhere to the paper so long as the corona field is applied. The procedure for dispersal therefore involves gently moving the residual fiber clumps across the paper surface while generating the statically charged fields in their vicinity with the high frequency generator. The carbon filaments from the clumped fiber mass gradually are “shed” from the outside of the clump and onto the paper. This shedding process occurs with the smallest filaments first, leaving the longer filaments to be dispersed later in the process. Manipulation of the clumps is done manually. Wooden sticks from cotton swabs work well for this purpose as well as insulating the operator from electrical shocks. As-is, the carbon fiber dispersal technique is rudimentary, but effective. A little art and technique on the part of the operator is required to effectively disperse all of the fiber clumps and depending on circumstances (clump size, fiber length distribution, etc.) some strategies work better than others. Some care is required to avoid breaking the filaments during manipulation. Applying the corona field directly to the carbon fibers themselves will induce arcing that is definitely damaging to the fibers. The technique becomes less effective as the paper substrate becomes loaded with dispersed filaments, and so changing the paper at regular intervals is recommended.

Following dispersal, a layer of clear adhesive-backed laminating film is applied over the dispersed carbon filaments and the copier/printer paper substrate. The laminating film fixes the fibers in place to the paper so they cannot shift, re-combine, fly away, etc. These laminated sheets can then be transported, stored for future reference, etc. Digital images of the carbon filaments are scanned through the clear laminating film and into a computer file for later analysis.

Petri dishes with dispersed glass fibers or paper slides with dispersed carbon fiber can be scanned using a regular scanner. Traditionally, multiple images were collected from a microscope with an x-y stage and than joined. The use of a scanner considerably simplifies and shortens the measurement process. It is also possible to consider the use of an automated image analysis software for fiber length evaluation as problems with imperfectly joined images are eliminated.

Fiber length distribution is currently determined by manually measuring the fiber length of 2000 fibers using the Image J software [5].

Representation of results

Fiber length distribution may be represented via two rational approaches. In both approaches, a fiber is counted in a certain length range (bin), say 1-2 mm or 10-11 mm. The first approach is to simply report the number of fibers measured in a certain length range, while in the second approach the relative weight of the measured fiber is taken into account. A reasonable approach to the weighing is to report the fiber as a fraction of the original pellet length. If one starts with 12 mm pellet, for example, than 9.6 mm long fiber would add 9.6/12 = 0.8 to the 9-10 mm length range while 1.2 mm long fiber would add only 1.2/12 = 0.1 to the 1-2 mm range. It is clear that reporting number of fibers may accentuate the presence of very short fragments. These methods for reporting fiber length can also be thought of in the same terms as molecular weight by number and by weight (first and second moment) [6]. The histogram of experimentally measured FLD can be normalized and fitted with an assumed form of a

probability density function and used for thermo-mechanical LFT property calculations [7].

There are two corrections that one may want to make to the results obtained via the procedure described above to obtain an unbiased sample of the fibers in the material.

The first correction may be to account for very short fragments whose length to width ratio is small and they may have been discounted in the digitized image as noise. This correction may be performed by estimating the amount of fragments from digital images or by sieving and weighing these fragments before dispersion of the longer fibers.

The second correction can be applied to eliminate bias of the measurement method caused by preferential capture of long fibers. Figure 6 shows two long fibers, of length L, that are captured in the sampling region and two shorter fibers, of length L2 with similar centroid positions, one of which will not be part of the sample.

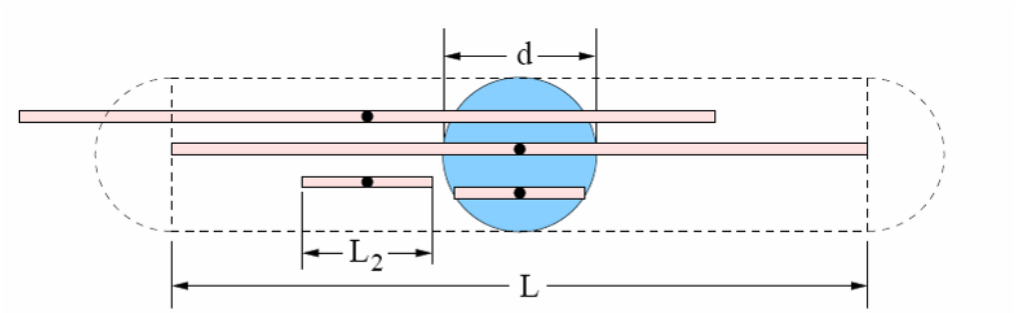

Figure 6. Sampling region of diameter d with three fibers captured for experimental measurement and only two centroids within the sampling region.

Assuming that the sampling region is a disk of diameter d and that all fibers lie in the same plane as the disk, then the actual number of centroids of fibers of length L within the sampling region N(L) can be obtained from the raw measurement of number of fibers of length L that pass through the sampling region Φ(L) as:

$$N(L) = \Phi(L)\left(1 + \frac{4L}{\pi d}\right)^{-1}$$

Pellet length

Fiber length measurement of fibers contained in pellets was performed to verify the majority of fibers were initially nominally 12.7 mm (0.5 in.) long. Burn-off was performed at 450°C for 60 minutes in a muffle furnace in air. Three pellets were randomly selected from each lot. All pellets are irregular in shape with cross-sections ranging from approximately circular, flat oval, to triangular. The original length of pellets was estimated by measuring distance of two randomly selected points on opposing surfaces using calipers. An exact measurement was not possible due to irregularity and lack of parallelism of opposing surfaces. Results from these measurements are presented in Table I.

Table I. Original pellet length

Length [mm]	Material A	Material B
pellet 1	12.92	13.13
pellet 2	13.02	12.43
pellet 3	12.41	12.61
average	12.78	12.72

The measurement of fiber length distribution was performed using the technique described above, however the number of measured fibers was smaller compared to measurement of FLD in molded samples. The vast majority of fibers in the pellet have the nominal pellet length, and nearly all of the fragments are very short. Therefore there was no need to measure a large number of fibers to establish the proper shape of the distribution.

Fiber Length Distribution in ISO plaques and Center-Gated Disk

Samples were selected from ISO plaque and center-gated disk from location B indicated in Figure 1 and Figure 2. The center of the samples was 45 mm from the gate of the ISO plaque and 34 mm from the center of the center-gated disk. The procedure described above was followed to obtain raw FLD measurements. These measurements were corrected to eliminate bias caused by preferential capture of long fibers. The sampling region diameter was estimated by weighing the extracted plugs before and after burn-off. Based on microscopic observations of the plugs it was assumed that the epoxy was deposited outside of the needle diameter, therefore the plug was a hollow cylinder. The fiber volume fraction in the expanded state was used to account for the presence of the fibers in the plug. Table II shows estimates of sampling region diameters for each sample measured, while Table III shows averages used for actual correction of the results presented below.

Table II. Estimates of sampling region diameter based on measured weights

Specimen	BF3D	BS3D	AF3D	AF3I	AS3D	AS3I
Expansion height [mm]	19.1	19.1	12.7	12.7	12.7	12.7
Selection diameter estimate [mm]	2.1	2.1	1.7	1.7	1.6	1.6

Table III. Sampling region diameter used for correction

	Carbon Specimens	Glass Specimens
Diameter for correction [mm]	2.1	1.6

Figure 7 shows results for center-gated disks with correction for preferential capture of long fibers and no correction for short fragment discounting. The relative weight contribution of the fibers is reported in the plots. It should be noted that samples with carbon fibers retain a higher fraction of long fibers when compared to samples with glass fibers and that fast mold fill processing results in greater fiber length attrition. Independent measurements of FLD on identical samples performed by Pacific North National Laboratory using a technique nearly identical to one described above have confirmed these measurements and by weighing very

short fragments prior to optical measurement have shown that the peak of the FLD is below 1 mm of fiber length when one accounts for short fiber discounting.

Figure 8 shows results for ISO plaques. Again, fast mold filling speeds resulted in greater attrition of fiber length.

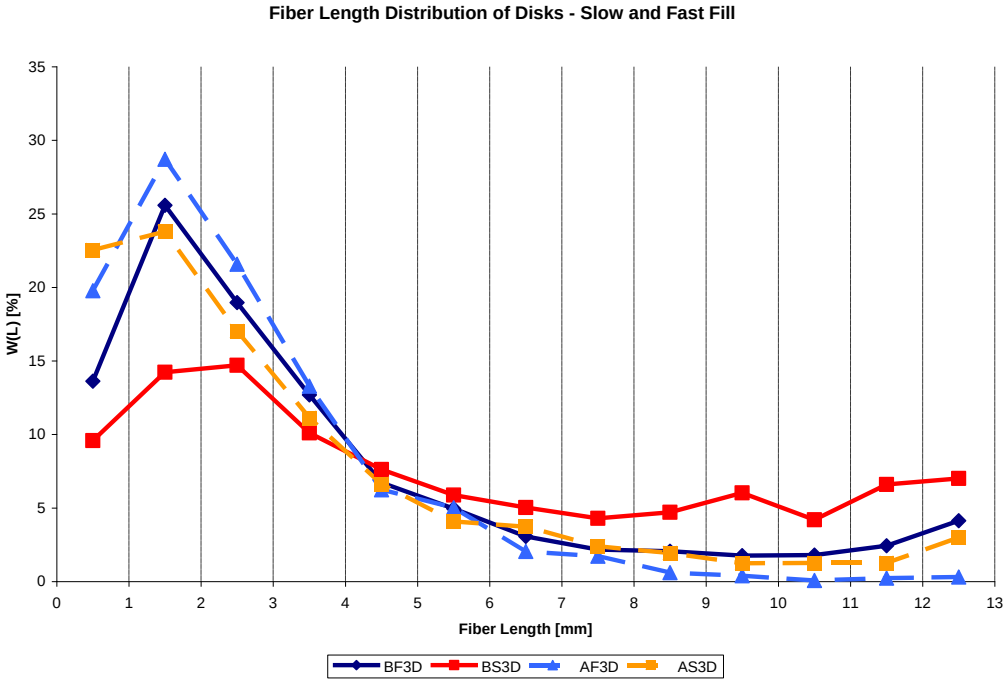

Figure 7. FLD for center-gated disks with correction for preferential capture of long fibers and no correction for short fragment discounting.

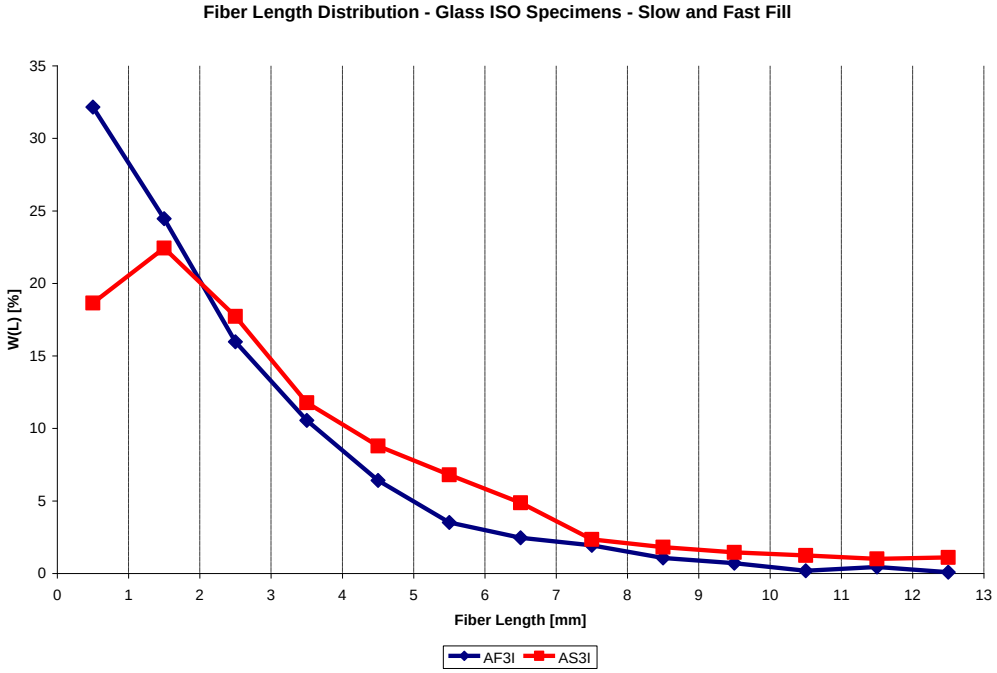

Figure 8. FLD for ISO plaques with correction for preferential capture of long fibers and no correction for short fragment discounting.

Conclusions

FLD measurement procedures for LFT materials containing glass and carbon fibers have been developed and validated. Measurements on samples molded using tools optimized for short fiber processing have revealed significant fiber length attrition. Samples containing carbon fibers retained larger portions of long fibers when compared to samples with glass fibers molded under the same conditions. Fast mold fill processing has resulted in greater fiber length attrition.

Acknowledgements

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