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Sample preparation and image acquisition using optical-reflective microscopy in the measurement of fiber orientation in thermoplastic composites

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Key words. fiber composites, fiber orientation, optical microscopy, sample preparation.

Summary

A complete sample preparation procedure used to determine three-dimensional fiber orientation from optical micrographs of glass fiber-reinforced thermoplastic composites is presented. Considerations for elimination of irregularities in the elliptical footprints, contrast enhancement between fibers and surrounding polymer matrix, controlled-etching that allows the identification of small shadows where fiber recedes into the matrix, and topographical reconstruction of the elliptical footprint are described in the procedure. This procedure has produced high-quality optical micrographs employed to obtain accurate fiber orientation data for thermoplastic composites using the method of ellipses. The optimal definition of the nonelliptical footprints' borders allows an accurate measurement of orientation in small sampling areas.

Introduction

Fiber-reinforced composites have enhanced physical properties relative to the polymer matrix. However, the physical properties, for example, thermal and mechanical, of these materials are dependent on the local orientation of fibers in the parts, which varies with position. Therefore, obtaining reliable measurements of orientation is important for estimating the properties of these materials.

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The method of ellipses (MoE) is the standard method used to determine the orientation in polymer composites, due to its low cost and simplicity. In the MoE, the orientation of fibers is obtained from the characteristic values of complete or incomplete elliptical footprints and rectangular footprints seen in micrographs (Zhu *et al.*, 1997; Vélez-García *et al.*, 2012). Characteristic values for elliptical footprints are the center position, the minor (m) and major (M) axes lengths, and the angle between the horizontal and the major axis. In the case of rectangles, the axis lengths are substituted by the footprints' height and length. Each footprint represents a cylindrical rod with fiber orientation expressed by the angles of orientation, that is, in-plane (ϕ_f) and out-of-plane angles (θ_f). Figure 1 illustrates the angles of orientation for a single fiber.

The problem of determining fiber orientation reduces to obtaining accurate measurement of the footprints' characteristic values. For this, it is essential to procure high-quality images showing footprints with well-defined borders as well as excellent contrast between the fibers and the surrounding polymer matrix. Moreover, the use of high-quality images is a crucial element for automatic detection of footprints based on image analysis tools (Mlekusch *et al.*, 1999). Corroboration of automatically detected footprint borders via manual measurement is time consuming and it is at odds with the purpose of image analysis tools. Therefore, the imprecisions detected in footprint borders automatically cause inaccurate measurements of fiber orientation which are consequently accepted as correct results.

Measurement of orientation in small sampling regions can be obtained by considering the contribution of fractional footprints as demonstrated by Vélez-García *et al.* (2012). This

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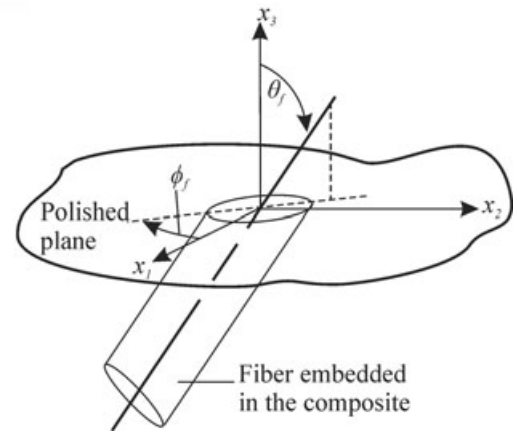

Fig. 1. In-plane angle (ϕ_f) and out-of-plane angle (θ_f) describing the orientation of a single fiber.

is particularly important for regions with rapidly evolving orientation or with low fiber concentration. Fractional footprints seen in the polished plane may arise when the polished plane intersect the tip of a fiber. However, inappropriate sample preparation can cause fiber breakage which also produces fractional objects. Therefore, a good experimental procedure is critical in order to avoid undesired footprints.

The ambiguity problem is one of the major limitations of the MoE and arises from the fact that every elliptical footprint has two possible values, ϕ_f or $\phi_f + \pi$ (Clarke & Eberhardt, 2002). Recently, Vélez-García *et al.* (2012) developed a method in which the tips of the fibers underneath the polished plane of a footprint are exposed by the plasma etching treatment. This results in a dark region in the optical reflection micrograph next to the major axis of footprints for inclined fibers and can be used to determine unambiguously the in-plane angle. The correct sign of the in-plane angle is determined from the position of the dark region (shadow). An effective visualization of the shadow depends on the controlled excavation of the polymer matrix during the etching step, the quality of the in-focus image, and the inclination of the fiber. The identification of this new parameter requires a distinctive sample etching procedure.

The sample preparation procedure has a direct impact on the production of high-quality images for obtaining reliable and complete fiber orientation in thermoplastic composites. A standardized method, however, is not available in the literature which can lead to the use of low-quality images for evaluating fiber orientation. These low-quality images show low contrast and poor footprint border definition, for example, irregular or diffuse borders, which prevent obtaining the footprints' characteristic values correctly. Imprecision in elliptical footprint dimensions may increase the systematic error significantly (Bay & Tucker III, 1992b).

There are three reasons for the absence of guidelines encompassing sample preparation of fiber composites for orientation analysis. First, the American Society for Testing and Materials (ASTM) standards, that is, ASTM E3 and ASTM E 2015, are only for metals and neat polymers. Second, very few publications encompass all steps of sample preparation for fiber composites. Typically, the only sample preparation step mentioned is sample polishing, referred to as metallographic polishing (Bay & Tucker III, 1992a; Lee *et al.*, 2003; Hine & Duckett, 2004; Vélez-García *et al.*, 2012). Third, literature considering fiber composites provides only general aspects involved in sample preparation for fiber thermoplastic composites, for example, Sawyer *et al.* (2008) and Hayes and Gammon (2010), but these references lack sufficient details.

The objective of this paper is to provide a sample preparation procedure that will serve as guidance for obtaining optimal definition of the footprints' border and appropriate contrast enhancement. This experimental procedure attempts to provide sufficient detail in all steps involved in the sample preparation of fiber composites. In addition, we consider modifications in the etching procedure that allow identifying an additional parameter in the image used to unambiguously determine the fibers orientation. As a result of the sample preparation, high-quality images suitable for image analysis in a small sampling area are obtained.

Sample preparation

Sample preparation steps include cutting, marking, mounting, grinding, polishing, etching, and sputtering. The procedure presented in this paper has been applied to several commercially available resins containing short and long glass fibers, for example, 30 wt.% short glass fiber filled polybutylene terephthalate (SF-PBT, Valox 420, SABIC, Houston, TX, USA), 30 wt.% short fiber polypropylene (SF-PP, RTP-105, RTP Company, Winona, MN, USA), 30 wt.% long fiber polypropylene (LF-PP, LNP Verton MV006S, SABIC, Houston, TX, USA) and 30 wt.% long fiber polyamide 6 (LF-PA6, LNP Verton PX00700, SABIC, Houston, TX, USA). A detailed description that can be readily adapted to other composites is presented below.

Sample cutting

Appropriate cuttings are critical to avoid both fiber breakage and changes in the fiber orientation. First, aggressive cutting causes fractures and surface irregularities of fibers. This results in distorted shapes of fiber footprints in the final polished plane which increases the imprecision in their characteristic values. Second, the deformation and rise in temperature related to the cutting operation can change the orientation of the fibers. Especially for circular and nearly circular footprints, errors in characteristic values of the order of several pixels significantly

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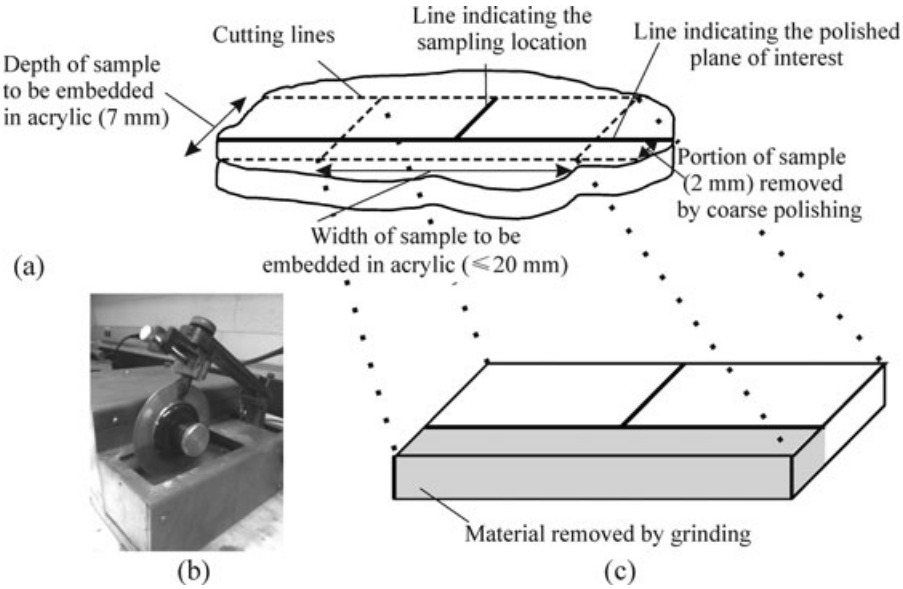

Fig. 2. Sample in molded part: (a) Lines indicating the location of polished plane, sampling location and cutting lines considering the distances away from the polished plane and width of the sample, (b) abrasive cut of sample, and (c) mounting sample with the gray area used to denote the material to be extracted by sample grinding.

increase the systematic error in orientation analysis (Vélez-García *et al.*, 2012). Setting a margin between the line denoting the measurement plane and the cutting lines, as well as performing the cut by abrasion using low speed, prevent effects of fiber breakage and alteration of orientation on the polished plane (Bates & Wang, 2003; Hayes & Gammon, 2010). The material in the margin region is subsequently removed during the grinding step to prevent effects related to the cutting operation. If cuts by abrasion are not possible, cuts using a band saw combined with larger margins are necessary. Cuts using a band saw have the advantage of reducing the cutting time considerably with respect to the low-speed abrasive cutter. However, the dimensions of the margins related to band saw cutting must be set judiciously. Small margins would suffer from the problems described above, while large margins as recommended by Sawyer *et al.* (2008), that is, 25 to 50 mm, are impractical and would increase the grinding time by as much as 12 times. The dimensions of the sample are important in order to avoid problems if compression molding is used for sample mounting. Large samples cause vertical inclination and deformation of the sample during compression molding.

The following procedure considers a careful cutting operation that has been proven to produce polished surfaces without irregular footprints caused by fiber breakage created during this step. Figure 2 summarizes the relevant aspects of the cuts made to obtain a mounting sample. Figure 2(a) illustrates the line indicating the plane of interest, the line indicating a sampling location, and the cutting lines. In addition, this figure depicts the dimensions, that is, 20 × 7 mm, of a rectangular mounting sample appropriate for a mounting mold of 25.4 mm. The 7 mm of final depth includes a margin of 2 mm of material removed during the grinding step. Increasing the margin from 2 to 5 mm showed no significant differences in footprints' shape. Also, our results in general indicate that a depth of 7 mm does not cause problems of sample tilting or deformation during the hot mounting. The mounting sample is cut using a low-speed diamond wheel saw (South Bay Technology, Inc.; San Clemente, CA, USA), as shown in Figure 2(b), with cutting oil (Precision Surfaces International, Inc., Houston, TX, USA) used as a cooling agent. At the end of the cutting operation, a mounting sample similar to the one shown in Figure 2(c) is obtained. In Figure 2(c), the gray color denotes the portion of material to be removed during the grinding step.

In the case of large samples, an initial sample cut around the region of interest is necessary and is illustrated in Figure 3. Figure 3(a) shows two samples of interest located in the interior of a large part and the cutting-lines' distance from the line of interest for cuts made with a band saw (e.g. Delta band saw model 28–150, Anderson, SC, USA; 14 TPI standard/deep cut portable band saw blade, Brookfield, WI, USA). Our results indicate that at least 10 mm margins between the line of interest and cutting lines do not cause fractures or promote changes in the polished samples. In Figure 3(b) an initial sample of ≈ 20 mm in depth with the important lines on it is shown. The dimensions of this sample are suitable for abrasive cutting using the low-speed diamond wheel saw.

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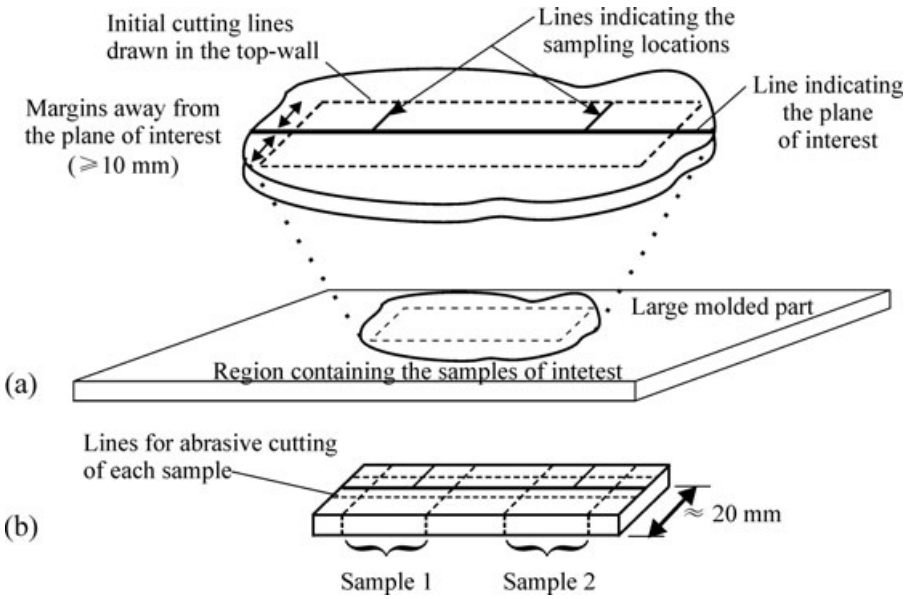

Fig. 3. Sampling cut in large samples: (a) Lines indicating precut of initial sample containing two sampling locations of interest (b) Lines for abrasive cuts in the initial sample for obtaining two mounting samples.

Sample marking

Identifying precise locations for measuring the fiber orientation is important in studies in which information of local orientation is relevant. However, sample marking on locations of interest is an aspect that has not yet been described in the literature. Tridimensional sample markers made of polymeric material are a feasible option for identifying specific locations during image acquisition. However, the "marker" polymer must be selected carefully to avoid problems in subsequent steps of sample preparation. For example, the polymer has to be softer than the composite material which is to be ground and polished, and it must resist deformation due to the heating operation if compression molding is used for sample mounting.

Tridimensional sample markers made of squared and colored polymeric pieces bonded on the mounting sample provide a means of identifying locations of interest during image acquisition. The idea is to use one side of the marker to stop the grinding step and to use the other side as a reference of the location of interest during the image acquisition as shown in Figure 4. Tridimensional markers based on red-colored polyethylene terephthalate (PET), and bonded with a commercially available cyanoacrylate adhesive (Krazy Glue, Columbus, OH, USA) are feasible options. This material resists the mounting temperature and is softer than the composites considered in this study.

Sample mounting

Appropriate sample mounting is essential because this step influences the sample preparation resources and time and keeps the sample in correct position during the mounting. Description of sample mounting of fiber-reinforced composites in the literature is limited to using epoxies (Sawyer *et al.*, 2008; Hayes & Gammon, 2010). This has the advantage that the sample is not exposed to thermal softening or mechanical distortion. However, when the thermal and structural characteristics of the fiber composite remain intact at the mounting temperature and pressure, compression molding is a faster and less expensive option for fiber composites. Notably, the compression pressure required to mount fiber thermoplastic composite samples is much lower, that is, ≈1 kPa, than that required for metals and ceramics (LECO Corporation, 2001). To reduce the time required for sample preparation and in order to save resources, multiple samples can be mounted in the same puck.

A consistent mounting operation with low values of compression pressure requires an appropriate control of the heating and cooling cycles. An effective heating cycle consists of compacting the sample with approximately 20 ml of

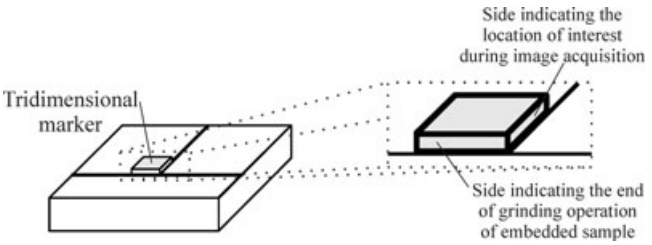

Fig. 4. Mounting sample with a tridimensional marker indicating the position where the location of interest and line indicating the polished plane meet.

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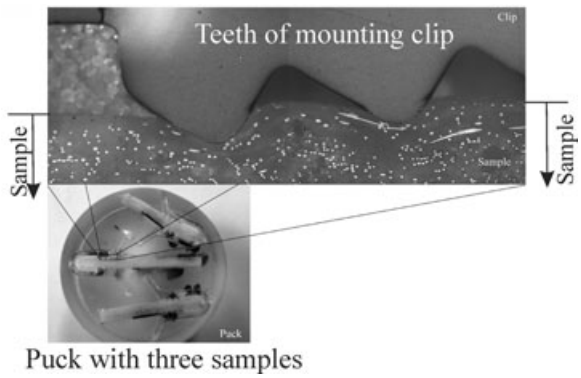

Fig. 5. Deformation of a sample of 30 wt.% long fiber polypropylene (LNP Verton MV006S) due to the indentation of mounting clips illustrates the sensitivity of fiber-reinforced composites to hot mounting conditions.

acrylic powder (thermoplastic powder; Precision Surfaces International, Inc., Houston, TX, USA) and preheating for 7 min at 458 K to melt the mounting material, slowly applying compression pressure until the material sustains 1 kPa, while maintaining the heat for 7 min. Subsequently, a transparent puck is obtained by allowing the sample to cool for 15 min while maintaining 1 kPa pressure. Our results indicate that the use of this cycle prevents the tilting of the sample during mounting and that the samples must be located away from the edges of the puck, for example, 1 or 2 mm, in order to avoid potential problems of uneven light intensity during image acquisition. We found that SF-PBT and LF-PA6 are able to resist the heating conditions with few problems, but materials like SF-PP and LF-PP can be negatively affected by the heating conditions. Figure 5 shows the deformation near the surface of LF-PP samples induced by the strong grip of support clips (UniClip; Buehler, Lake Bluff, IL, USA). This deformation can cause errors in the evaluation of orientation near the surface of the sample if the clips are near the sampling region. To avoid any distortion in the sample, soft clips (spring clips; Precision Surfaces International, Inc., Houston, TX, USA) with reduced heating time are recommended. Reduction in heating time can cause the "cotton ball" effect, that is, partially melted acrylic in the middle of the puck, but we observed no adverse impact of this cosmetic imperfection.

Sample grinding

Sample grinding is critical for removing material damaged by the cutting operation. The purpose of this section is to provide guidance for sample grinding specifically tailored for fiber composites that will be helpful in extrapolating the procedure for other composites. Factors inherent to the composite, that is, fiber type, fiber concentration, polymer type, glass transition temperature, and crystallinity, all make it challenging to derive a single distinctive procedure capable of meeting the above objective. Indeed, an adapted version of metallographic grinding suitable for fiber thermoplastic composites requires a set of experimental considerations and personal skills and judgment to tackle these factors. In contrast with recent literature that focused on general experimental considerations, that is, Sawyer *et al.* (2008) and Hayes and Gammon (2010), in this section we provide a specific procedure that can be used as a starting point for sample grinding. During the grinding operation, it is important to maintain wet conditions in order to avoid excessive fiber breakage caused by using dry grinding and simultaneously eliminating the debris. Periodic rotation of the sample is essential to keep the even height of the puck as well as to erase the grooves in the direction of the previous grinding step.

Effective sample grinding of fiber thermoplastic composites consists of a two-stage process which is summarized in Table 1. The objectives of these two stages are material removal from the margin region and gradual reduction of the grooves from previous grinding steps, respectively. The first stage consists of a semi-manual operation in which a wet-belt grinder (BG-32 dual belt grinder; LECO, St. Joseph, MI, USA) is used to remove material quickly, but its use requires careful operation due to safety and control issues. We have found that using 120—followed by 180—grit abrasive paper for the wet-belt grinder helps to provide control of material removal in the first stage. We also explored a fully manual execution for the first stage as well, using 120—grit abrasive paper, but it increases the grinding time for this stage from 4 min to at least 30 min. The second stage for the sample grinding involves manual operation using a wet hand grinder (HandiMet® 2 Roll Grinder; Buehler Co, Lake Bluff, IL, USA), while taking into account the considerations indicated in Table 1. It is critical to reach the plane of interest, defined by the border of the marker embedded in the puck, especially when multiple samples are embedded in a single puck. Uneven sample grinding affects the performance of the subsequent sample preparation steps as well as causes images to be acquired over a nondesired plane. To ensure even removal of material at each step of the sample grinding, the height of the puck is measured at a minimum of 4 locations after every complete puck rotation. Regions exhibiting greater height than others are targeted for additional grinding. A careful and systematic 90° rotation of the puck, as indicated in Table 1, showed no significant variation in the height of the puck, that is, less than 0.010 mm (dial test indicator, Mitutoyo Corporation, Kanagawa, Japan). The purpose of the values and conditions indicated in Table 1 is to provide a basic procedure for implementing the grinding operation. This information may be easily adapted and implemented to any other composite. For example, Figure 6 shows a linear change in the height of the puck after the different grinding steps for fiber composites with different polymeric matrices. The amount of material removed depends on the polymeric matrix and its morphological characteristics assuming that E-glass with the same dimensions and composition is used as reinforcing material.

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Table 1. Sample grinding procedure for fiber orientation studies in thermoplastic composites.

Stage (instrument)	CAMI† Grit designation (average sizes of the abrasive material in μm)	Time (min)	Interval for 90° rotation (s)	Objective
I (wet belt grinder)	120 (115)	1–2 (until blurry marker is perceived)	10	Removal of material until plane of interest is reached
	180 (82)	1–2 (until clear marker is seen)	10	
II (wet hand grinder)	240 (53)	2	30	Gradual removal of surface layers to eliminate grooves
	320 (36)	2	30	
	400 (23)	2	30	
	600 (16)	2	30	

†Coated Abrasives Manufacturer's Institute (CAMI).

Sample polishing

Sample polishing is critical for obtaining optimal footprint borders in the polished surface. At the end of the polishing step, the surface exposes the microstructure of the composite at the plane of interest. Factors inherent to the composite, that is, fiber type, fiber concentration, polymer type and glass transition temperature and crystallinity, all make it challenging to derive a distinctive procedure capable of meeting the sample polishing objective. The purpose of this section is to develop a procedure which can be extrapolated to other composites. During polishing, there are four experimental considerations in this step that promote obtaining optimal footprint borders in the polished surface (Hayes & Gammon, 2010; Sawyer *et al.*, 2008). First, automatic polishing provides precise control of the polishing conditions. Second, avoiding extreme pressure levels prevents undesired artifacts on the polished surface. Third, avoiding an excessive amount of suspension prevents the surface from slipping over the particles rather than polishing the surface. Fourth, removing the debris after each polishing step by putting the sample in an ultrasonic water bath for 30 sec. In addition we have found that repeating the polishing steps, based on visual inspection of the polished surface and checking the footprint border, are essential for obtaining a high-quality polished surface.

Adequate sample polishing that allows obtaining the polished surface with optimal footprint borders for the composites is presented. Table 2 summarizes the sample polishing based on the use of a semiautomatic single mount grinder/polisher (MiniMet® 1000 grinder/polisher; Buehler Co, Lake Bluff, IL, USA). The use of this type of instrument is crucial for providing precise control in polishing conditions and for obtaining the desired footprint borders indicated in

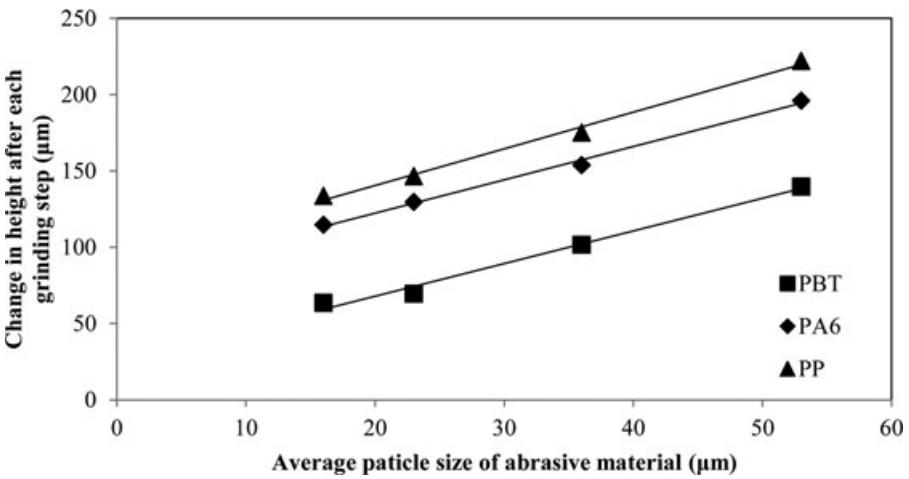

Fig. 6. Removal of material by abrasion during the second stage of sample grinding based on change in height after each grinding step. Sandpapers with average abrasive material sizes of 53, 36, 23, and 16 μm were used to remove the material. The material removed consisted of either polybutylene terephthalate (PBT), nylon 6 (PA6), or polypropylene (PP) composites embedded in acrylic. The concentration of glass fiber in the composites was 30 wt.%.

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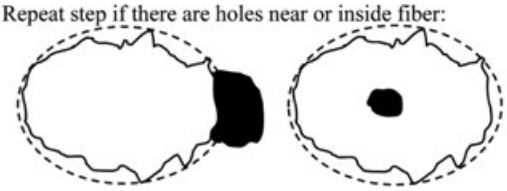
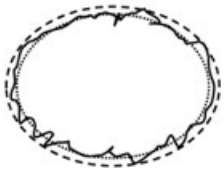
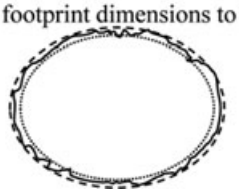
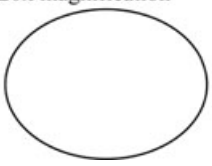
Table 2. Sample polishing procedure for fiber orientation studies in thermoplastic composites using a MiniMet® 1000 grinder-polisher.

Alumina particle size (μm)	Speed (rpm)	Vertical force (N)	Time (min)	Typical fiber border at the end of polishing step
5	40–50	22.2	60	Repeat step if there are holes near or inside fiber:  Acceptable: Irregularities at the edge of footprints allow ≈ 90% final footprint dimensions to be seen. 
1	40–50	22.2	30	Repeat step if the irregularities at the edge of the footprints are similar to previous step: Acceptable: Irregularities at the edge of footprints allow > 95% final footprint dimensions to be seen. 
0.05	40–50	22.2	30	Acceptable: No irregularities observed at the edge of footprints using 20x magnification 

the last column of Table 2. This column is included because we have found that tracking the elimination of holes and reduction in irregularities of footprint borders are better indicators than a fixed polishing time for the purpose of gauging the polishing progress. In contrast with a typical automatic polisher, the use of the MiniMet® 1000 requires a hole drilled perpendicular to the upper side of the puck to hold the sample in place and to avoid uneven polishing. We have found that the use of a single type of cloth, for example, synthetic nap cloth (Final finish cloth; Precision Surface International Inc., Houston, TX, USA), is capable of producing satisfactory results for the different polishing steps. However, changes in cloth every 15 to 30 min are necessary because of the moderate pressure levels dictated by the vertical force indicated in Table 2. To prevent excessive dryness in the polish cloth, three drops of the polishing suspension (Precision Surfaces International, Inc., Houston, TX, USA) must be added every 10 min. Removal of debris from the polishing step by putting the sample in an ultrasonic water bath for 30 sec keeps undesired artifacts from the polished surface. We have observed that sample polishing performance depends more significantly on the local fiber concentration than on other factors inherent to the composites. For example, a good polishing quality of a region having a large concentration of fibers is achieved much later than in regions of moderate concentration. Regions having a large number of fibers require careful visual inspection and may require some modifications to the polishing procedure indicated in Table 2.

Plasma etching and gold sputtering

When optical microscopy is used to image the sample, the etching of the polished surface with oxygen plasma is necessary to provide contrast enhancement between the fiber and polymer matrix. The contrast between the glass fibers

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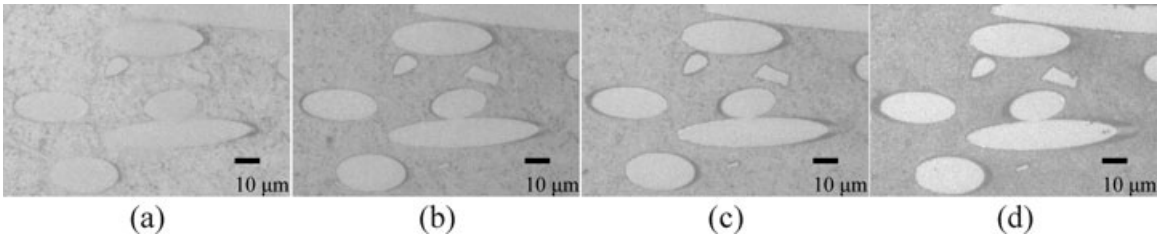

Fig. 7. Reflective-optical microscope images (objective 20×) showing the growth of shadows for footprints in the polished surface of 30 wt.% short glass fiber-PBT during plasma etching: (a) 0, (b) 20, (c) 40, and (d) 60 min of plasma etching.

and the polymer matrix in the polished surface is insufficient for producing quality images used to determine accurately the fiber orientation by image analysis. To overcome this limitation, oxygen plasma is used to excavate and to uniformly roughen the polymer matrix, causing the fibers to rise over the matrix producing a clear distinction between the fibers and polymer (Mlekusch *et al.*, 1999; Hine & Duckett, 2004). However, determining the etching time is important for reaching an appropriate depth of surface erosion that allows differentiating the direction of the fibers coming out of the polished plane. The rate of volatilization of the polymer roughening the surface depends on the polymer type and microstructure. Junkar *et al.* (2009) have found that the etching time increases with crystallinity because the rate of surface erosion is reduced due to the molecular arrangement in this morphological state. (The present authors also point out that crystal orientation may also be important). A detailed description of the mechanism and volatilization of resin surface due to the interaction of plasma and structural groups of different families of polymers is given by Egitto (1990). As a result of appropriate plasma treatment, the images of the surface allow for fast and reliable recognition of the elliptical footprints during image analysis and for identification of fiber direction.

Optimum etching times for different composites were determined from the analysis of surface erosion as a function of time. This analysis consists of tracking the sharpening of the footprints' borders, the increasing appearance of light-dark regions, and the lengthening of shadows from samples etched with oxygen plasma in 5 minute increments. Here the optimum etching time is defined as the minimum time required for both contrast enhancement and reliable identification of shadows. In Figure 7, the progression of contrast enhancement and increase in shadow size for SF-PBT at different etching times is shown. Also observed in Figure 7 is that the PBT acquires a mid-tone gray color but not black as suggested in the literature (Mlekusch *et al.*, 1999). On the other hand, of the three polymer types, PA6 is the only material that becomes practically black in about 4 min, but without showing appreciable surface erosion. These observations suggest that contrast enhancement is strongly dependent on the chemical reaction and not on the change in fiber height above the matrix background as suggested in Mlekusch *et al.* (1999). Figure 8 shows the impact of the material on the rate of the surface erosion. The difference in height between the polished surface of the glass fiber and the surface of the polymer matrix seen in Figure 8 was estimated from fibers having inclinations between 77 and 82°. Based on visual inspection, the optimum

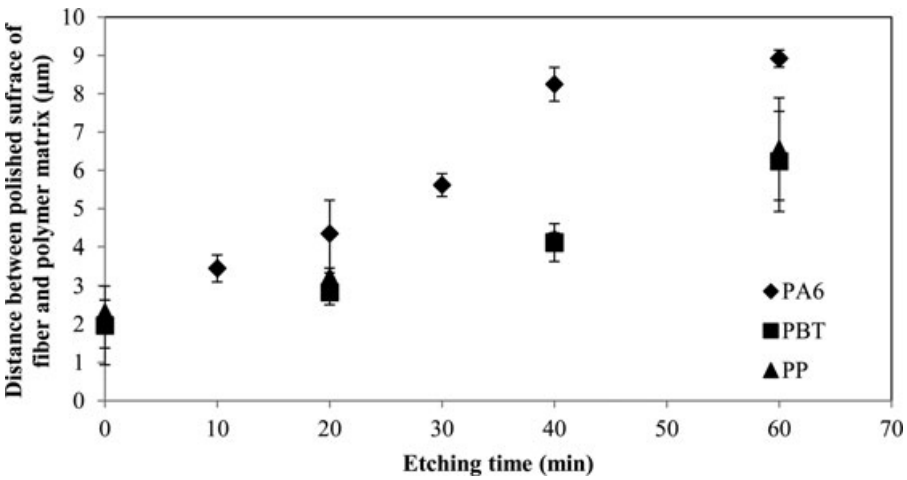

Fig. 8. Difference in height between the polymer matrix and the polished surface of fiber for three polymer composites as a result of varying exposure time to oxygen plasma. The polymer matrices consisted of polybutylene terephthalate (PBT), nylon 6 (PA6), and polypropylene (PP).

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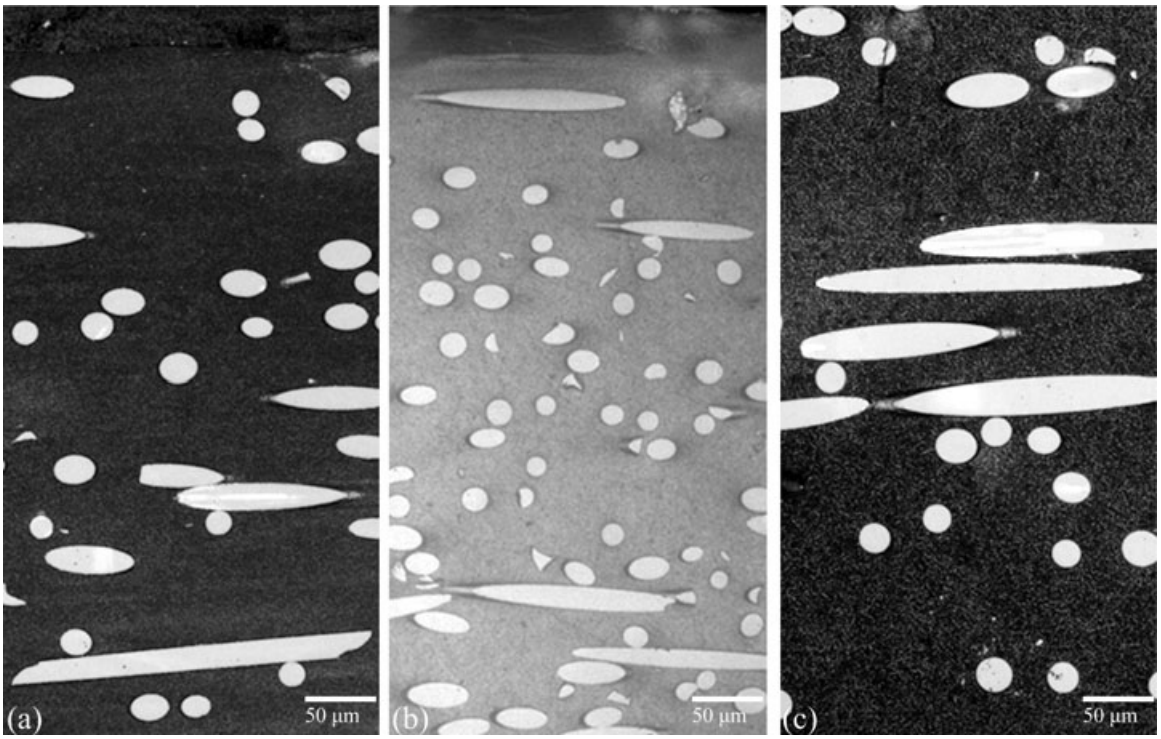

Fig. 9. Representative reflective-optical microscope single in-focus images (objective 20×) after polishing and plasma etching steps for glass fiber composites with various polymer matrices; (a) PP, (b) PBT, and (c) PA.

etching time for PBT and PP samples was determined to be 40 minutes while that for PA6 was found to be 60 minutes. The plasma etching was performed using a radio frequency (RF) plasma cleaner (Plasma Prep^{TM} II; SPI Supplies, West Chester, PA, USA) at a vacuum pressure of 26.66 kPa and a current of 75 mA. Our results demonstrate that this commonly available instrument is a feasible alternative to the more expensive, direct current (DC) plasma etcher used by Mlekusch *et al.* (1999) and Clarke and Eberhardt (2002). To obtain uniform etching, it was observed that only 2 or 3 pucks placed near the open end of the inner chamber of the Plasma Prep II should be used.

Gold sputtering after plasma etching may be used as a secondary means for contrast enhancement. A 30-sec gold sputtering step has been used as part of the sample preparation following standard polishing procedures. However, our results did not show the need for gold sputtering in any of the composites considered, due to the satisfactory contrast enhancement obtained with the plasma etching.

Image acquisition

Image acquisition requires setting appropriate magnification, uniform illumination and use of topographical correction to reduce systematic errors in fiber orientation analysis. Clarke and Eberhardt (2002) indicate that approximately 20× magnification provides sufficient resolution to reduce errors related to the pixilation of the digital image. A higher magnification can be used, but it increases the scanning time for and the noise in the images (Bay & Tucker III, 1992b; Davidson *et al.*, 1996). Use of uniform illumination avoids changes in appearance across the image. These changes alter the information during the image processing and cause imprecise measurement of orientation (Clarke & Eberhardt, 2002). Clarke and Eberhardt (2002) indicate that the use of autofocusing algorithms is essential to obtain a sharp contrast for large areas because the flatness of the polished surfaces cannot be guaranteed to be within the depth of field of the microscope objectives. The use of a motorized stage is also important for efficiently scanning the region of interest in the sample (Davidson *et al.*, 1996). Precise displacement of the stage is required in order to acquire multiple images with partial overlap. At the end of this step, a single image is reconstructed from multiple high quality images that are stitched together. Each reconstructed image is analyzed by a microprocessor and only characteristic values of footprints are stored. Advanced microscopes have software that performs the autofocusing and stitching automatically. However, these steps may be done manually by using commercially available software, as will be designated below. Storing of reconstructed images can be useful to perform additional image processing and analysis but it requires the preservation of the high-quality image. Therefore, if an image is stored, it must be saved with low amount of compression, which increases the image size.

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Microscopes with advanced features, for example, modules for scanning, stitching of images, and autofocusing correction, are able to produce the single global image automatically. Unlike recommendations provided by Clarke and Eberhardt (2002), our results indicate that the topographical corrections given by the autofocusing algorithm are important for obtaining high-quality images even for scans over small regions. The advanced features in the microscope indicated above are suitable for scanning the large areas required for determining orientation in long-fiber composites. If the microscope does not have these advanced features, image acquisition using a semi-manual procedure permits fiber orientation measurement with acceptable precision (Vélez-García *et al.*, 2012). An optical-reflection microscope (e.g. Axiovert 40 MAT; Carl Zeiss LLC, Thornwood, NY, USA) with a motorized stage allows for precise displacement between image frames. A scan over the region of interest must include images taken with 20× magnification, partial overlap, and uniform illumination. Each image was saved using the jpg compression format, the microscope's default setting. The dimension of each image was 700 μm × 525 μm (2310 × 1731 pixels). Additionally, each image has to be taken at different focus levels (depending on the irregularity of the topography) to capture the fiber detail from the matrix to the polished footprint with good resolution and to compensate for any variation in surface flatness. Subsequently, a single in-focus image is created from partially focused images using Helicon Focus (Helicon Soft Ltd., Kharkov, Ukraine) software. Examples of representative single in-focus images for PP, PBT, and PA after polishing and etching steps obtained with the procedure described in this paper are shown in Figure 9. Finally, the multiple in-focus local images are stitched together using Photoshop (Adobe, San Jose, CA, USA) to form a single stitched global image. The stitched global image was saved in jpg format using the highest resolution setting available (i.e. 12) in order to preserve most of the image quality. The size of each stitched global image was approximately 12 MB. Out-of-focus and single in-focus images can be discarded after this step to reduce storage requirements. Storing the stitched global image has the advantage that data incorrectly measured by automatic detection can be checked and corrected. Additionally, it can be used to analyze topographical corrections such as void spaces, clustering, and fiber bending.

Conclusions

We have presented a detailed sample preparation and image acquisition procedure for obtaining high-quality images from carefully prepared polished surfaces. The optimal footprint borders and contrast enhancement in those images allow accurate measurement of footprint characteristic values at locations of interest. The excellent definitions of the nonelliptical footprints allow us to include these shapes in the analysis of orientation, which reduces the sampling region. In addition, the described procedure of controlled etching enables the identification of small shadows where the fiber recedes into the matrix. This allows eliminating the ambiguity in the value of the in-plane angle of orientation. Subsequently, these values are used to compute reliable three-dimensional fiber orientation. This paper provides detailed yet simple guidelines specifically developed for the complete sample preparation procedure including cutting, marking, mounting, grinding, polishing, etching, and sputtering, as well as image acquisition used in the analysis of orientation for fiber composites. These guidelines take into consideration efficiency and time and cost reduction in every step. We indicated a number of aspects of sample preparation, not yet described in the literature, which should prove useful in further studies of fiber composites. The sample marking based on 3D markers presented here provides a simple but effective alternative for identifying specific regions of interest in the samples. Also, we demonstrated that compression mounting can be used to embed samples of thermoplastic composites as a less expensive and expedient alternative to cold mounting. The procedure outlined in this paper should be applicable to other composites, as well as to other means of image acquisition such as scanning electron or confocal laser scanning microscopy.

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References

Bates, P.J. & Wang, C.Y. (2003) The effect of sample preparation on the mechanical properties of nylon 66. *Polym. Eng. Sci.* **43**, 759–773.

Bay, R.S. & Tucker III, C.L. (1992a) Fiber orientation in simple injection moldings. 2. Experimental results. *Polym. Composite* **13**, 332–341.

Bay, R.S. & Tucker III, C.L. (1992b) Stereological measurement and error estimates for three-dimensional fiber orientation. *Polym. Eng. Sci.* **32**, 240–253.

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Clarke, A.R. & Eberhardt, C.N. (2002) *Microscopy techniques for materials science*. CRC, Boca Raton, FL, USA.

Davidson, N.C., Clarke, A. & Archenhold, G. (1996) Large-area, high-resolution image analysis of composite materials. *Proceedings of the Meeting of Microscopy of Composite Materials III*. Oxford, England.

Egitto, F.D. (1990) Plasma etching and modification of organic polymers. *Pure Appl. Chem.* **62**, 1699–1708.

Hayes, B.S. & Gammon, L.M. (2010) *Optical Microscopy of Fiber-Reinforced Composites*. ASM International, Materials Park, OH, USA.

Hine, P.J. & Duckett, R. (2004) Fiber orientation structures and mechanical properties of injection molded short glass fiber reinforced ribbed plates. *Polym. Composite* **25**, 237–254.

Junkar, I., Cvelbar, U., Vesel, A., Hauptman, N. & Mozetic, M. (2009) The role of crystallinity on polymer interaction with oxygen plasma. *Plasma Process. Polym.* **6**, 667–675.

LECO Corporation (2001) Metallography principles and procedures. *Form No: 200–860*. Leco Corporation, St. Joseph, MI, USA.

Lee, K.S., Lee, S., Chung, K., Kang, T. & Youn, J. (2003) Measurement and numerical simulation of three-dimensional fiber orientation states in injection-molded short-fiber-reinforced plastics. *J. Appl. Polym. Sci.* **88**, 500–509.

Mlekusch, B., Lehner, E. & Geymayer, W. (1999) Fibre orientation in short-fibre-reinforced thermoplastics. Part I. Contrast enhancement for image analysis. *Compos. Sci. Technol.* **59**, 543–545.

Sawyer, L.C., Grubb, D.T. & Meyers, G.F. (2008) *Polymer Microscopy*. Springer Science+Business Media, LLC, New York.

Vélez-García, G.M., Wapperom, P., Baird, D.G., Anning, A.O. & Kunc, V. (2012) Unambiguous orientation in short fiber composites over small sampling area in a center-gated disk. *Compos. Part A-Appl.* **43**, 104–113.

Zhu, Y.T., Blumenthal, W.R. & Lowe, T.C. (1997) Determination of non-symmetric 3-D fiber-orientation distribution and average fiber length in short-fiber composites. *J. Compos. Mater.* **31**, 1287–1301.

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