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DETERMINATION OF FIBER CONTENT IN LONG FIBER REINFORCED COMPOSITES BY SCANNING ELECTRON MICROSCOPY AND IMAGE ANALYSIS

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Abstract

Currently, there is a wide interest in further developing long fiber reinforced composites. Due to the dependence of a material's mechanical properties to its microstructure, further development of microanalytical techniques is needed to quantify the amount of fiber. In response, a standardless technique was developed to determine the volumetric fiber content and its variation in an oriented long fiber reinforced composite. The method utilizes scanning electron microscopy to acquire backscattered electron images of polished cross-sections. The images are then processed to determine the fiber area fraction which, in this particular case, is equal to the volume fraction. The results presented fall within 10% of the nominal bulk (fabricated) fiber content with the relative precision ≈2%. A large part of this difference can be due to local variations in fiber content.

KEY WORDS: Scanning Electron Microscopy, Image Analysis, Long Fiber Reinforced Composites % Fiber.

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Introduction

Novel materials such as advanced polymers, blends and composites are finding a wide range of applications in a diverse array of industries from aerospace to consumer products [1,2,12]. Advantages of using these materials include cost, ease of fabrication of complex shapes, corrosion and environmental resistance, and high strength to weight ratios. It is the interplay of these numerous factors that governs the selection of a particular material for a given application.

Of particular interest are long fiber reinforced polymer systems. They have found application where strength and load bearing capacity are the primary performance criteria. Long fiber reinforced polymers consist of oriented long fibers in a polymeric matrix. The fibers are usually aligned in a parallel manner and are typically composed of glass, carbon, nylon, or PET. The polymer matrix consists usually of a thermoset or thermoplastic resin. When a load or stress is applied the matrix transfers the force to the fibers which further distribute it to a larger volume of the material. This phenomenon accounts for the synergistic effect that allows development of materials that are stronger than their component parts.

The synergistic effect unfortunately is difficult to predict. It is however dependent on several variables: type of fiber and matrix, fiber content, interfacial strength between fiber and matrix, and fiber orientation within the matrix. To further understand the interaction of a composite's component parts and how they relate to mechanical properties, accurate analytical techniques are needed to determine these variables. A variety of such techniques are available; however, most are bulk methods.

Of particular interest is the determination of fiber content. Two direct methods are commonly used. These include wet digestion [14] and reflectance/ transmission measurements by optical microscopy [7,8]. Both methods suffer some drawbacks. In the case of wet digestion, the sample is destroyed, hazardous chemicals used in the digestion method must be disposed and the chemicals may attack or "etch" the fibers to some degree. Reflectance/transmission

experiments require preparation of standards characterized first by wet digestion and subsequent interpretation of standard curves. Optical methods also assume that the fiber content obtained by wet methods is directly comparable to that present in samples used in optical analysis. Experience has shown that composite samples are not uniform throughout. Additionally, there appear to be questions concerning the ASTM method for determining fiber loading [8].

Scanning electron microscopy has proven itself to be a very useful tool in the characterization of many materials. With the advent of coupled on-line image analysis, it has become possible to quantitatively determine various features (3,4,6,10,11) in a number of different matrices. This technique has been demonstrated in a number of different fields such as, biology (9), steel (13) and coal (5).

Scanning electron microscopy (SEM) with image analysis provide a standardless method for the determination of fiber content. The method consists of acquiring a digitized electron image from a previously polished specimen. A binary image is then constructed from the electron image which relates the feature of interest to the number of picture points (pixels) counted. Features such as fibers and voids can be highlighted to determine % fiber, % void or % matrix. The calculation is based on the number of pixels for the highlighted feature as compared to the total number of pixels in the frame. The percent area can then be calculated by the following equation:

$$\frac{\text{number of pixels of feature}}{\text{total number of pixels in frame}} \times 100 \qquad (1)$$

Using the above equation and assuming the composite is uniform with depth (z direction), the volume percent of the feature of interest is identical to the area coverage of feature of interest in the cross-section. Percent void content and matrix content can be calculated in the same manner. These three variables are interrelated using the following equation:

$$\text{total area analyzed-\% fiber-\% void=\% matrix} \qquad (2)$$

Experimental

An experimental long glass fiber composite was chosen to demonstrate the technique. SEM images were obtained with a JEOL 840 scanning electron microscope equipped with a LAB_6 source. The general operating conditions were: 12 kV accelerating voltage, 800 pA current (cup), working distance of 8 mm, and the compositional (backscattered) detector was used for imaging. Subsequent analyses of the images were performed using a Tracor Northern 5500/5600 system capable of obtaining and manipulating 512 by 512 pixel images with 256 gray level resolution. The Image Acquisition and Processing Program (IPP) was IPP 5B/80, also from Tracor Northern (Middleton, Wisconsin).

A typical analysis for fiber content consists of three parts; sample preparation, image acquisition, and image analysis of the features of interest. All three of these steps are extremely important as poor performance in any one will have detrimental effects on the final results. The three steps will now be considered in detail.

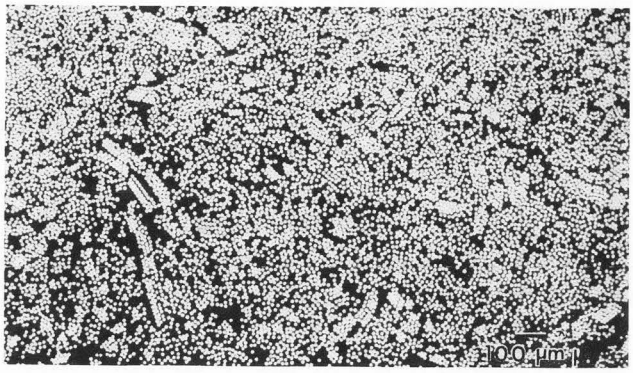
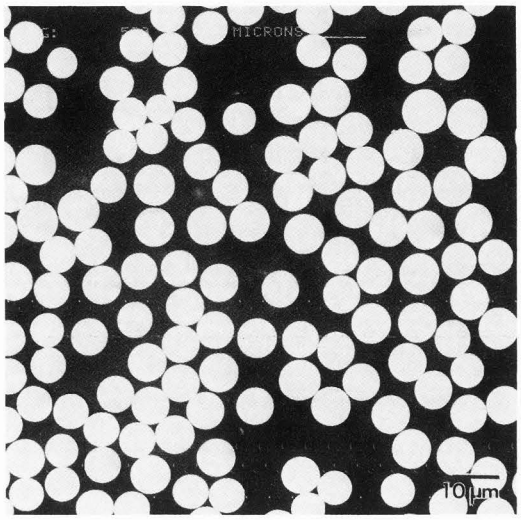


Fig. 1 Backscattered scanning electron micrograph of a typical polished cross-section of a glass filled composite showing non-uniform distribution of glass fiber. Bar = 100 µm.

Sample preparation usually consists of first isolating cross-sections with a metallurgical diamond saw. The typical sample is 1 mm by 10 mm which is embedded in epoxy such that the long axis of the fibers are perpendicular to the surface to be polished. Sample sizes up to 50 mm x 50 mm can be accommodated by the JEOL setup. Initial sample grinding consists of first abrading with successively finer grit silicon carbide paper to provide a flat surface for the final polish. Alumina particles of 9, 3, and 0.05 microns respectively are used to polish the sample. The surface is then buffed with microcloth to remove any finely adhered particles and coated with a thin layer of Au-Pd to provide a conductive surface for SEM analysis.

The electron microscopist has a wide range of instrument variables at his disposal to aid in acquisition of images which readily lend themselves to image analysis. These variables include accelerating voltage, current, type of detector(s) and magnification. As image analysis requires binary images, it is necessary to obtain high contrast images differentiating the feature of interest from the "background". With the Tracor Northern image analyzer it is preferable to have white features of interest (fibers) on a black background.

The binary images are created by first choosing a suitable threshold grey level within the grey level distribution of the digitized image. All pixels with grey levels above the threshold are assigned a "white" value whereas those below are "black". Therefore it becomes important to determine which grey level to choose for the threshold. Selection is done from the digitized electron image grey level distribution by plotting the frequency of pixels as a function of the grey level. From the grey



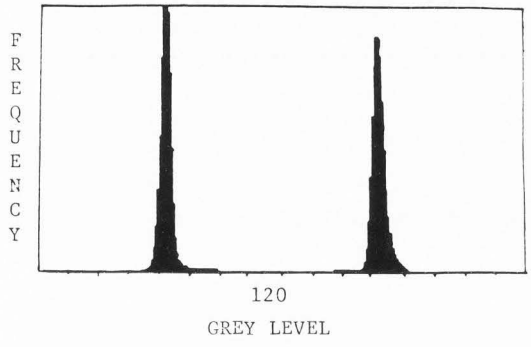
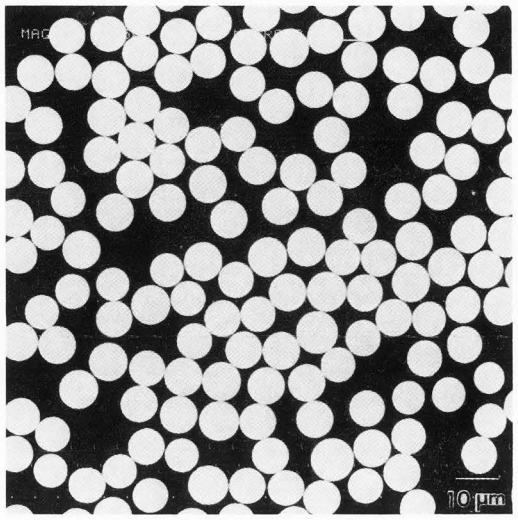


Fig. 2 a) Digitized binary image of polished cross-section (nominal fiber content = 58%). Bar = 10 μm.
b) Grey level frequency histogram of binary image shown in Fig. 2a.
c) Using grey level of 120 as a "threshold" the fiber content of Fig. 2a was found to be 49%.



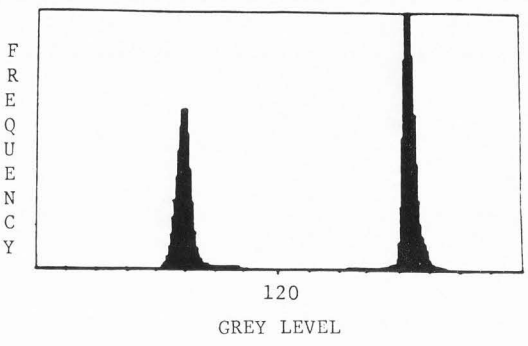


Fig. 3 a) Digitized binary image of polished cross-section (nominal fiber content = 58%). Bar = 10 μm.
b) Grey level frequency histogram of binary image shown in 3a.
c) Using grey level of 120 as a "threshold" the fiber content of Fig. 3a was found to be 56%.

level distribution (histogram) a high contrast image should have two peaks, the matrix (a low value of grey level or "dark") and the fibers (a high grey level, or "white"). Selection of one of the intermediate grey levels will then be used as the threshold level to create the binary image. Analysis of the image consists of counting the number of pixels that are white (as compared to the total number) from which percent cross-sectional fiber area can be determined. This value is numerically equal to the fiber content (volume percent) for the case of long parallel fibers.

Individual images can be analyzed to determine fiber content (volume %) of specific locations. Automation of the stage allows images to be collected from a number of positions. These images are subsequently analyzed and volume fraction of fibers for the total area scanned is calculated.

Results and Discussion

A typical long fiber reinforced polymer composite, when imaged using a backscattered electron detector in a scanning electron microscope, appears as shown in Figure 1. High contrast between the fiber (white) and the matrix (black) was obtained with backscattered electron images. Figure 1 shows many of the details found in a typical cross-section. Here fiber rich and poor areas are easily seen and, in general, the microstructure is not uniform. An initial survey of the cross-section is also performed at this point to determine whether a void analysis should be performed at a later time. Several areas should be examined to ensure small particulates did not adhere to the polished surface. If, however, small imperfections do exist, a cutoff is available in the IPP

program for areas smaller than a given value (usually the smallest diameter fiber used). This permits the rejection of any small defects found on the cross-section.

Analysis of the sample is usually performed at 500x with 512 by 512 images collected. Typical images, gray level histograms and the fiber content are shown in Figures 2 and 3. No imperfections arising from surface preparation are apparent. Once again the nonhomogeneous microstructure is evident as shown by clustered fibers and matrix rich regions.

Two composites, 46 and 58 percent fiber (as calculated during fabrication), were analyzed by obtaining cross-sections from different areas of the sheet. Fiber content was obtained by analyzing twenty frames of 31000 micrometer^2 area across the sample. The results obtained are reported in Table 1. Nominal fiber content was obtained by calculating the volume percent fiber from the weight of the fiber added to the resin. Calculations are based on the density of the starting materials and on the assumption that no voids are present. SEM results indicate close agreement in some cases to the calculated amounts. Variations of fiber content do occur in the cross-sections analyzed and are reflected in these values. To determine the variation in reproducibility a "58 percent by volume fiber" cross-section was run in duplicate (20 frames). The results of 61 and 63 volume percent indicate the reproducibility of the technique to approximately 2% relative precision.

Conclusion

Examination of numerous long fiber composites has shown a wide variation in fiber content within a given cross-section. SEM techniques, along with subsequent analysis of the acquired image for "% area of fiber", has been shown to be a useful technique for characterizing these composites. The results obtained indicate agreement to within 10% of the fabricated value and a relative precision of 2% for all cross sections analyzed. Some of this variation is undoubtedly due to variation within the microstructure. The ability to collect information on the variation of the microstructure, in a standardless manner, is unique among characterization methods. It is hoped that further work will reveal a relationship between micro-fiber content to the desired mechanical properties.

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Discussion with Reviewers

D.W. Strickler: You mention a number of fibers in the introduction, but do not discuss any of the problems involved in analyzing these fibers. Of course the glass fibers in a carbon matrix are ideal.

Author: Glass fibers in a carbon matrix are indeed ideal when considering the development of contrast. However, as you noted, there are problems associated with different reinforcement/matrix combinations. For example, organic fibers (nylon, polyethylene, etc.) for the most part polish better using diamond paste for the last polishing step. A carbon fiber in the carbon matrix gives problems when considering contrast. Here alumina works fine for polishing (as for most brittle materials), but one has to be very careful not to round the edges of the fiber by over-polishing. In today's ongoing research to develop more composites, a little trial and error is always necessary during preparation of the surface.

C. Jock: How are voids or void content determined by this SEM method?
Author: In general, any feature whose size is greater than a pixel can be counted. Voids can be caused by gas formation and non-wetting of fibers with matrix. Bubbles have been found to produce a higher grey level than the fiber, so it is relatively easy to isolate this grey level for counting. Voids between fibers, possibly caused by poor wetting, are typically "darker" than the matrix. Again, by isolating this grey level from the matrix, it is possible to "count" them.

C. Jock: How is perpendicularity of the fiber verified?
Author: If a cross-section of a fiber is taken perpendicular to its length, it should have an aspect ratio of 1. By checking several areas, one can determine whether the sample was prepared properly. However, there is no guarantee that all the fibers within a composite will be oriented identically.

J. Hefter: What are the effects of focus and astigmatism on the final imaging data?
Author: Incorrect focusing/astigmatism, will cause errors due to apparent increase of feature image size.

C. Jock: What effect does the polish have on the binary output image generated?
Author: Polishing is the most important step for accurate analysis. In particular, for brittle materials (carbon, glass), it is important not to chip them. Any damage done, or rounding of the edges, can effect the grey level distribution of the fiber making it difficult to distinguish from the matrix. This is a major problem with carbon fibers where the contrast between fiber and matrix is low.

J. Hefter: You have stated that the grey-level histogram generated from your digital image should have two peaks, one at the low end for the matrix (background), and one at the high end for the digitized features of interest. Since the images collected in this work are from the backscattered electron signal, why is it necessary?
Author: It is not always necessary to setup the image for two peaks. The important point is to isolate the feature of interest fiber, matrix, or voids from the other two. There are occasions when voids will appear brighter than the fiber, particularly in carbon fiber reinforced matrices requiring one to isolate the fibers between the matrix (darker) and voids (brighter).

D.W. Strickler: Your explanation of the analysis method does not make it clear that you are analyzing all of the pixels in every frame. Are you doing a lineal analysis or are you actually analyzing 512 by 512 pixels on each frame?
Author: To determine percent area, the grey level of interest is transformed into a binary image. The number of pixels in the binary are then counted and compared to the total.

Table 1: Nominal Versus Calculated Fiber Content in Volume Percent

	Composite 1	Composite 2
Calculated as Fabricated	46%	58%
Determined by SEM with image analysis		
Cross section 1	41%	58%
Cross section 2	48%	52%
Cross section 3		61,63%

D.W. Strickler: How many frames does a person have to collect to get a statistically valid volume percent? Certainly, the greater the variability among frames, the larger number of frames that would have to be analyzed.
Author: Currently, the analysis is conducted until the running average converges. Usually 20 frames is more than sufficient for this.

J. Hefter: Could you comment on the variability question insofar as the data taken for Composite 1 are substantially different from the "expected" value? How many more frames of data might need to be taken for adequate "average" values to be obtained?
Author: When composites are fabricated, the volume percent is usually calculated from the starting weight percent. Shrinkage, expansion (due to thermal mismatches), void formation and other fabrication concerns makes it impossible to know exactly what composition the finished material is. This is further complicated by the fact composites are not perfectly uniform on the microscale. So in answer to your question, certainly more areas should be analyzed to arrive at an "average" value. However, one of the advantages of this technique is to obtain loading information on "small" regions, thereby enabling one to study positional changes in loading in geometrically complex components. Hopefully, this work can then be extended so that one could correlate these data with micro failure mechanisms.

J. Hefter: From the digital images presented, it is apparent that some variation in fiber diameter exists. Might additional morphometric data readily available in IPP, such as average diameter and perimeter, be useful for further characterization of these materials?
Author: It is true that many other geometric parameters are very important in describing the fibers used in a composite. However, with our current Tracor system, it is necessary to manually separate fibers when they are touching, thereby eliminating the automation part of the image analysis. We are currently looking into modifications in the package to allow the determination of various geometric parameters when the fibers are touching. Average diameter, perimeter, and nearest neighbor distances and their variation are indeed important factors when characterizing these materials.

D.W. Strickler: Could you discuss the amount of time that is involved in an analysis of this type? It seems that this measurement could be done on the fly so that the pictures would not even have to be recorded and later processed. That should speed up the analysis.
Author: Several hours are required to analyze a set of samples. You are correct that the analysis could be sped up significantly by several methods. Unfortunately, this would require rewriting the software for the image analysis program presumably with some loss of versatility.

C. Jock: How long does this method take (sample preparation, monitoring, data collection and data reduction)? Does it compare favorably or unfavorably (with respect to time) with other methods?
Author: The two competing methods for determination of percent fiber are by wet digestion and optical techniques. Wet digestion takes about 1 day to digest, wash, dry, and calculate the results for several samples. Optical, as well as scanning techniques, requires a polished cross-section. These techniques require ~1/2 hour to isolate a cross-section and to embed it. I usually allow for drying overnight. Polishing takes about 1 hour followed by AuPd sputtering, 1-2 hours is required for data acquisition per cross-section (20 frames) and approximately 1 hour for data reduction. As with optical methods, it is possible to prepare and polish several cross-sections simultaneously. The difference lies in the analysis times. For optical techniques, this is a matter of minutes, whereas in scanning, it is a matter of hours. However, by automating the analysis, it is possible to run unattended overnight, greatly reducing the amount of operator time.