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"Quantitative Non-Destructive Evaluation of Composite Materials Based on Ultrasonic Wave Propagation"

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I. INTRODUCTION

This report provides a summary of our continuing investigation of the application of ultrasound for the non-destructive evaluation of advanced composite materials. Our research has focused on deriving quantitative indices from fundamental acoustic parameters of the inhomogeneous materials being investigated. In the current grant period we are investigating several specific lines of investigation. Continuing work is being carried out on the examination of the acoustic Kramers-Kronig relationship between attenuation and phase velocity, examination of the problem of estimating attenuation from backscattered ultrasound, and the general wave propagation characteristics of anisotropic media such as fiber reinforced composites. Results of these on-going investigations will appear in subsequent progress reports.

In this progress report we shall focus on the results of one specific project. Much of the results contained in this report were reported at the 1985 Review of Progress in Quantitative Nondestructive Evaluation, held in Williamsburg Virginia.[1] Two innovative methods, one nondestructive and the other destructive, were used to characterize impact damage in a graphite-epoxy laminate. A nondestructive technique, polar backscatter, was employed at Washington University to detect and assess area, configuration and approximate interlaminar location of impact induced delaminations. In this technique the insonifying beam is incident on the sample at a non-zero polar angle, so that the specular echo from the water-composite interface does not dominate the backscattered signal. A destructive technique was employed at the Lockheed-Georgia Company[†] on the same impact sites to determine area and configurations of the delaminations at each interlaminar location. In this technique, deply, the internal matrix damage is marked with a gold solution, the laminate subjected to a partial pyrolysis and unstacked, and the damage area(s) quantified with an image analyzer. The successful correlation of the area and orientation of impact damage as measured by the polar backscatter technique with a lamina by lamina examination of the actual damage using the deply technique is reported below.

† The portion of this research carried out at Lockheed-Georgia Company was supported by Lockheed-Georgia Company's Independent Research Fund.

II. CORRELATION OF ULTRASONIC POLAR BACKSCATTER WITH THE DEPLY TECHNIQUE FOR ASSESSMENT OF IMPACT DAMAGE IN COMPOSITE LAMINATES

In this report we evaluate one technique for quantitative nondestructive evaluation of graphite fiber reinforced composites by correlation with a sensitive destructive evaluation technique. Although graphite fiber reinforced composites offer attractive strength to weight advantages for many applications, it is well known that one of the potential disadvantages of these materials is susceptibility to impact damage. For the case of low velocity impact, substantial structural damage can occur without significant exterior evidence.

One approach to the nondestructive evaluation of inherently inhomogeneous materials makes use of quantitative images based on ultrasonic backscatter.[2] A typical pulse-echo measurement is performed with the insonifying beam incident perpendicular to the specimen surface (a polar angle of zero degrees). Perpendicular insonification in an immersion measurement system results in a large specular reflection due to the acoustic impedance mismatch at the fluid/composite interface. This specular reflection may dominate the ultrasound backscattered from features of interest within the specimen. We note that effects of the large specular reflection on the backscattered signal can be significantly reduced by insonifying at non-perpendicular incidence (i.e., at a non-zero polar angle). An early application of this technique was used by Brown[3] in an investigation of the effects of fatigue in carbon fiber reinforced plastics. Brown's "dark-field" technique of insonifying at a non-zero polar angle was motivated by some observations on scattering by Bhatia.[4] A significant extension of the "dark-field" technique for anisotropic or quasi-isotropic materials such as fiber reinforced composites was independently introduced by Bar-Cohen and Crane.[5] This "polar backscatter" technique uses the fact that signals from cylindrical structures such as fibers are maximum when the insonifying beam is perpendicular to the long axis of the fiber, and falls substantially as the angle of insonification changes from perpendicular. Thus, the backscatter at a fixed polar angle exhibits a distinct, systematic azimuthal variation, with sharp peaks in backscatter that occur where the insonifying beam is perpendicular to any of the principal fiber orientations in the composite.

In a previous paper from the Washington University group,[6] we used quantitative images of polar backscatter to investigate impact and fatigue damage in graphite fiber reinforced laminates. In that study, images of polar backscatter were obtained with the azimuthal angle of insonification perpendicular to each of the four fiber directions present, so that each image was selectively sensitive to scatterers (fibers and damage) oriented along the specific fiber directions. The results suggest that low velocity impact results in more damage in laminae furthest from the side impacted, with damage in a specific lamina oriented along the fibers in that lamina. This research was supported during earlier funding periods of this grant, and portions of the research are described in several previous progress reports (March 1982 to September 1982, September 1982 to March 1983, and March 1983 to September 1983).

The destructive evaluation technique known as deply, developed by S.M. Freeman,[7] permits the characterization of impact damage at every interlaminar interface. In this technique, which is described in detail in Section IIc, the impact zone is saturated with a solution of AuCl, which penetrates into the regions of matrix cracking and delamination formed by the impact. The composite is then heated to partially pyrolyze the resin matrix and thus allow the lamina by lamina separation of the laminate. The damage at each interface is visible as a gold "fingerprint", allowing the characterization of the area, orientation, and shape of the damage as a function of depth.

In the work reported here, two sites of impact damage in each of two graphite-epoxy laminate panels were investigated ultrasonically and subsequently subjected to deply analysis. The orientation and shape of the damage determined by the two techniques were compared. Further, the area of damage estimated by the polar backscatter technique was correlated with that obtained from the deply technique.

IIa. SAMPLE PREPARATION

Test Panels for Impact

Two test panels, 6.0 x 10.0 inches, were removed from a 16-ply graphite-epoxy laminate fabricated from Hercules AS4/3502 prepreg tape. The stacking sequence for this laminate was $[0°/+45°/-45°/90_2°/-45°/+45°/0°]_s$ and consisted of 13 laminae with a possibility of 12 locations for interlaminar delamination and 13 locations for fiber-bundle fracture. The panels were ultrasonically "C" scanned to verify the absence of damage or defects before impacting.

Impacting of Test Panels

The panels were mounted in a special test fixture that provided vertical boundary supports spaced 3.0 inches apart. Two sites were impacted on each panel using a 0.5 inch diameter aluminum ball fired from a compressed air gun at a velocity of 150 feet per second. The end of the gun barrel was positioned 5 inches from the panel surface. Ball velocity was measured by two sensors spaced 6 inches apart on the gun barrel. For each panel both impact sites were between the same vertical boundary supports. Following impacting, the impact sites were subjected to TBE (tetrabromoethane) enhanced x-ray radiography to verify that impact induced damage was present. The panels were then baked at 150° F for 2 hours, cooled and one site on each panel infused with gold chloride. (See Section IIc. - Application of Marker Solution). After nondestructive evaluation (Section IIb.) the other impact sites were infused with gold chloride.

IIb. NON-DESTRUCTIVE CHARACTERIZATION METHOD: POLAR BACK-SCATTER

Each of the four impact sites was investigated using the polar backscatter technique. The experimental procedure was similar to that used in a previous study.[6] Four scans were obtained of each impact site. The scans corresponded to selective interrogation perpendicular to each of the four fiber orientations present in these samples. The panel being investigated was mounted on a motorized platform immersed in a water bath, with the back surface (defined as the surface opposite the impacted surface) facing the interrogating transducer. The (polar) angle of incidence was chosen to be 30° for all scans in this work. This polar angle is greater than the critical angle for quasi-longitudinal wave transmission into an anisotropic half-space for any azimuthal angle of incidence as determined from slowness surfaces based on the elastic constants for graphite-epoxy.[8,9,10] Consequently, interrogation of regions of damage interior to the specimen is expected to occur with quasi-shear waves. The azimuthal angle of the interrogating transducer was adjusted to be perpendicular to the selected fiber orientation of each scan.

A 0.5 inch diameter, 4 inch focal length broadband transducer, nominally centered at 5 MHz, was used in pulse-echo mode as the interrogating transducer. Approximately eight microseconds of backscatter from the sample were gated into an analog spectrum analyzer. The received spectrum at 288 frequencies over the range 2 to 8 MHz was averaged and normalized to the frequency average of the spectrum reflected from a flat stainless steel plate. Thus the numerical values of the polar backscatter reported here represent a quantitative measure of the broadband response of the interrogated material. The broadband frequency average of the backscatter reduces errors due to interference effects in the ultrasonic field and phase cancellation at the piezoelectric element of the transducer.[2, 11]

Each scan was 6.1 cm by 6.1 cm corresponding to 61 by 61 measurement locations with a spacing of 1 mm. The impact site was approximately centered in the region to be scanned.

Data Reduction

The ultrasonic information contained in the polar backscatter scans was cast into gray scale image format. Examination of gray scale images provides qualitative information regarding the shape and orientation of damage structures. An example of a gray scale image based on quantitative polar backscatter is presented in Figure 1. The image in Figure 1 is based on a polar backscatter scan of one impact site with the interrogating beam perpendicular to the +45° fiber orientation. (We note that the scan was performed from the back, resulting in an apparent reversal of the +45 and -45 directions.) The discrimination levels for the gray scale were chosen so that there are 16 equally spaced gray levels. The lightest level corresponds to backscatter less than -42.0 dB below that from a near perfect reflector, the darkest level to backscatter greater than -30.0 dB below that from a nearly perfect reflector. Although the damage is evident, the exact boundary of the damage zone is blurred by the beam width of the interrogating beam, which is several pixels wide at low frequencies.

Because the choice of discriminant levels can affect the qualitative aspects of a gray scale image, quantitative estimation of the area of damage based on a visual impression obtained from a gray scale image is often inaccurate. We have chosen a method which provides an unbiased estimate of the area of damage, based on the distributions of the measured polar backscatter values for undamaged and damaged zones. The damaged area shown in Figure 1 is characterized by stronger scattering than the nominally undamaged regions. Figure 2 represents a histogram of the distribution of the polar backscatter displayed in the image of Figure 1. The higher scattering values corresponding to the zones of damage can be seen as the "tail" extending from approximately -37 dB. The area of damage is a small fraction of the total image area, so that most of the histogram represents the distribution of backscatter from essentially undamaged

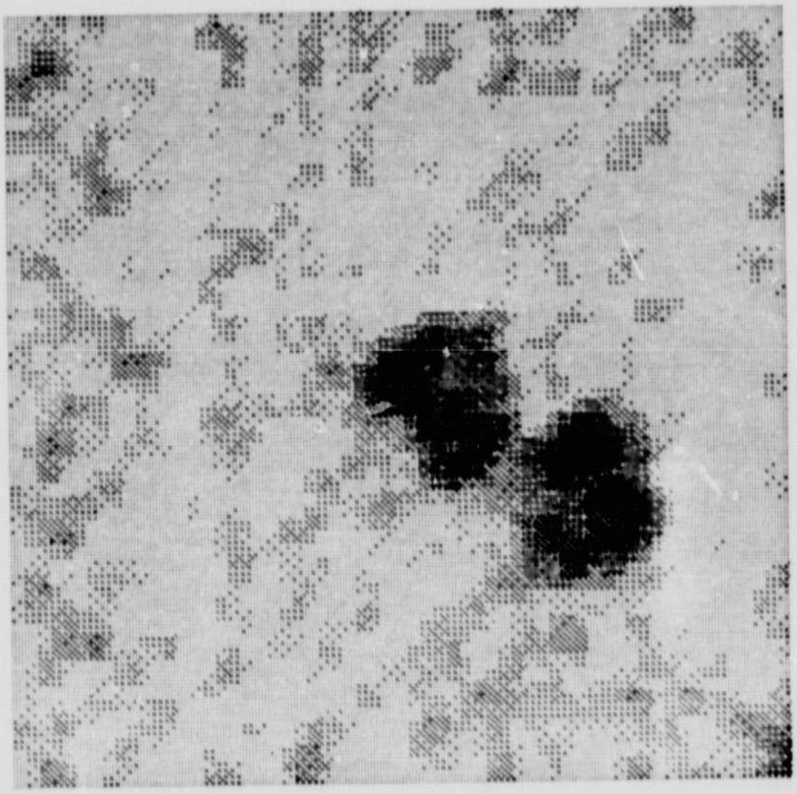


Figure 1 - Example of a gray scale image of a full 61 mm by 61 mm polar backscatter scan, interrogating perpendicular to the +45° orientation. The damage structure is orientated along the +45° direction.

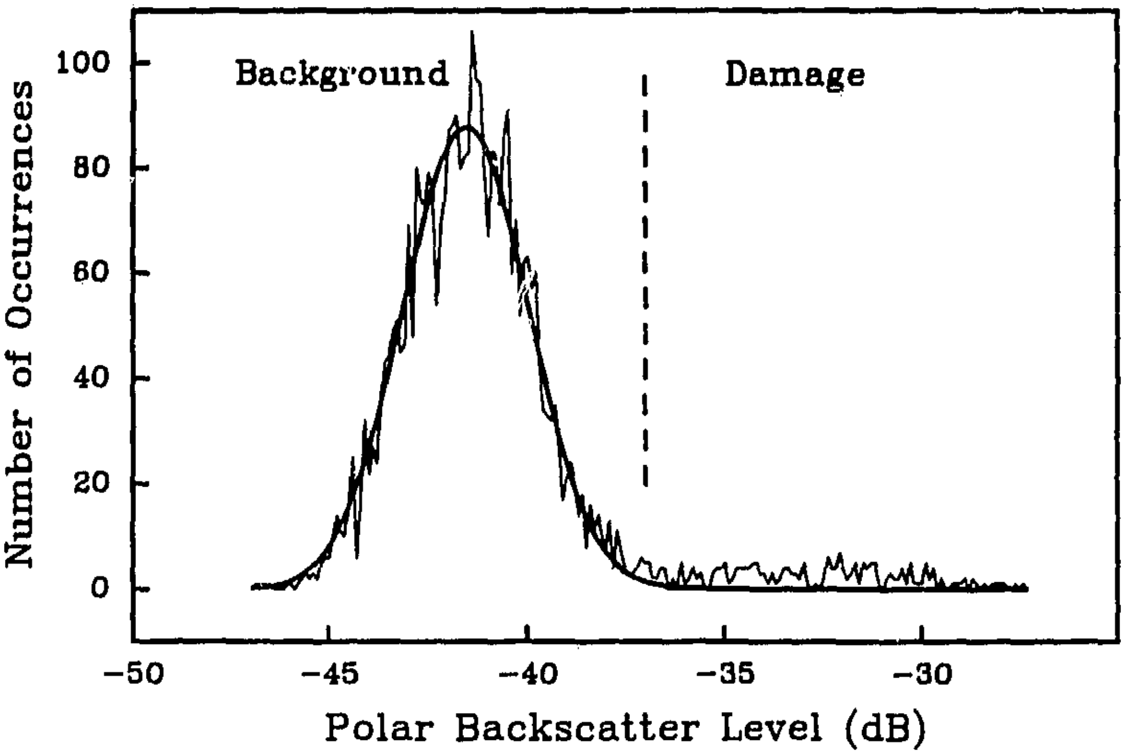


Figure 2 - Histogram of the quantitative backscatter values from the polar backscatter scan presented in Figure 1. The bin size was 0.1 dB. The heavy smooth line represents a normal distribution fit to the background.

regions. We chose to approximate this background by a normal distribution, which was determined by least-squares techniques. The smooth dark line in Figure 2 was generated from the least-squares parameters of the background distribution. An estimate of the area of damage was obtained by integrating the high scattering "tail" of the histogram and subtracting off the integral of the background distribution calculated over the same range of backscatter.

The apparent area of damage was determined by this histogram subtraction technique for all 16 images. The histogram information was also used to select appropriate threshold levels for the generation of the bi-stable gray level images shown in Section IId. The choice of a bi-stable display was made to simplify comparison with the photographs of damage obtained by the deply technique. In each image, the darker level represents higher scatter, starting at the (approximate) lowest scatter from damage, as determined from the histogram information.

IIc. DESTRUCTIVE CHARACTERIZATION METHOD: DEPLY

The deply inspection procedure consists of the application of a matrix damage marker solution to the graphite-epoxy panel followed by a partial pyrolysis of the resin matrix, unstacking the laminae, examination of the laminae, and damage or defect quantification.

Application of Marker Solution

A solution of gold chloride in diethyl ether (9.0% by weight gold) was applied to the composite face opposite the point of impact. There must be a pathway, even microscopic, that connects the damage area to the surface or edge of the composite to allow penetration of the marker solution. A dam of vacuum bag putty, with a mylar cover, was used to keep the solution in contact with the composite for about 60 minutes. Following the soak interval, the excess gold solution was removed and saved for recycling. The panels were heated to approximately 150° F, to remove the solvent before proceeding with the pyrolysis.

Pyrolysis

Segments of the graphite-epoxy composite containing the impact damage were placed on a stainless steel wire mesh holder and inserted into a zone of a tube furnace maintained at 785° F for 70 to 100 minutes. Following completion of the pyrolysis period the segments were removed from the furnace and allowed to cool. All segments were sufficiently pyrolyzed after 90 minutes to be suitable for unstacking.

Unstacking

The segments were carefully removed from the holder and placed on a work table. Each lamina of a segment was reinforced with transparent tape, lifted from the segment and stored in a small container. Normally the laminae are mounted on a worksheet with a piece of double-coated tape; however, these laminae were left unmounted to facilitate damage quantification. When unstacked in this manner the laminae were "flipped" so that the surface for observation was that of the "bottom surface" of the lamina just removed, or expressed in another way, the view was that of the "top half" of the interlaminar location. If one compares the gold marked area on the "bottom surface" of the lamina just removed with the gold marked area on the "top surface" of the remaining segment one observes that one area is a mirror image of the other. Care was exercised at this point to avoid touching the exposed surface of the lamina excessively with the finger, as this will sometimes blur the very small matrix crack indications that can be just a fraction of a millimeter in width and not readily apparent to the unaided eye. Figure 3 shows a typical view of gold marked impact damage.

Examination of Laminae

The most important requirement for observing the surface of a lamina for fiber-bundle fracture, matrix cracking indications and delaminations is proper illumination. Of course, some of the gross damage indications can be seen with makeshift lighting, but not the finer details. Fiber fracture is best observed with fluorescent light impinging at 90 degrees to the fiber direction. The optimum illumination for gold-chloride-marked matrix damage is a high-intensity light impinging on the lamina surface parallel to the fiber direction. For observing fiber-bundle fracture, small areas of delamination, and matrix cracking indications, a binocular microscope with a magnification range of approximately 7X to 50X is ideal.

Damage Quantification

The area of delamination for each interlaminar location at each impact site was determined with a Cambridge Q900 image analyzer using a macroviewer lens. A summary of these measurements is presented in Table 1, where the area of damage for each interlaminar location was obtained by averaging the

Figure 3 - Example of a photograph of the gold deposited at matrix damage.

Table 1 - Orientation of Damage and Average Area of Damage as a Function of Depth, from the Deply Technique

Interlaminar Location	Orientation of Damage	Orientation of Fibers Below Damage	Average Area of Damage (mm^2)
1-2	+45°	+45°	2.4
2-3	-45°	-45°	17.1
3-4	90°	90°	28.0
4-5	-45°	-45°	65.1
5-6	+45°	+45°	122.6
6-7	0°	0°	143.2
7-8	+45°	+45°	67.2
8-9	-45°	-45°	159.1
9-10	90°	90°	170.8
10-11	-45°	-45°	56.3
11-12	+45°	+45°	196.0
12-13	0°	0°	113.6

measurements from the four impact sites. The orientation of the damage with respect to adjacent fiber orientation will be discussed in the next section. The distribution of the delamination sizes through the thickness of the laminate can be readily visualized when the deply determined area for a single impact site versus interlaminar location is presented in a histogram format in Figure 4. Interlaminar location 1-2 on the histogram is adjacent to the impacted (front) side of the panel.

The trend of increased damage opposite the impacted face has been previously reported.[7,6,12] In thin, flexible plates such as the specimens in this study, this trend has been interpreted in terms of plate bending stresses.[13] Thicker, less flexible composite laminates may exhibit local subsurface damage more proximal to the impacted face.[13]

IId. CORRELATION OF RESULTS

The polar backscatter measurements represent a superposition of scattering from damage in several similarly oriented layers, with attenuation from intervening layers reducing the contributions from deeper layers. The deply technique provides information on damage at each interlaminar interface. We chose a subset of the deply technique information appropriate for correlation with the polar backscatter technique. Because we are testing the hypothesis that the polar backscatter technique is selectively sensitive to damage structures which are oriented perpendicular to the interrogating beam, the orientation of damage is the primary selection criterion for correlation. As an example, the polar backscatter scan of Figure 1 clearly indicates damage oriented in the +45° direction, as expected from the angle of interrogation. This polar backscatter scan should therefore be correlated with interlaminar locations which the deply technique indicates has damaged zones oriented in the +45° direction. Inspection of Table 1 reveals four interlaminar locations which exhibit this damage orientation: locations 1-2, 5-6, 7-8, and 11-12. Although one might initially envision the superposition of the damage zones in these four interlaminar locations as the damage zone which could be correlated with the polar backscatter technique, further consideration suggests that superposition may not be appropriate. The attenuation of quasi-shear waves in graphite-epoxy laminates can be substantial. Previous work from this laboratory[6] has shown that the polar backscatter technique in similar composite laminates is primarily sensitive to structures nearer the insonified surface. Thus, signals from damage farthest from the insonified

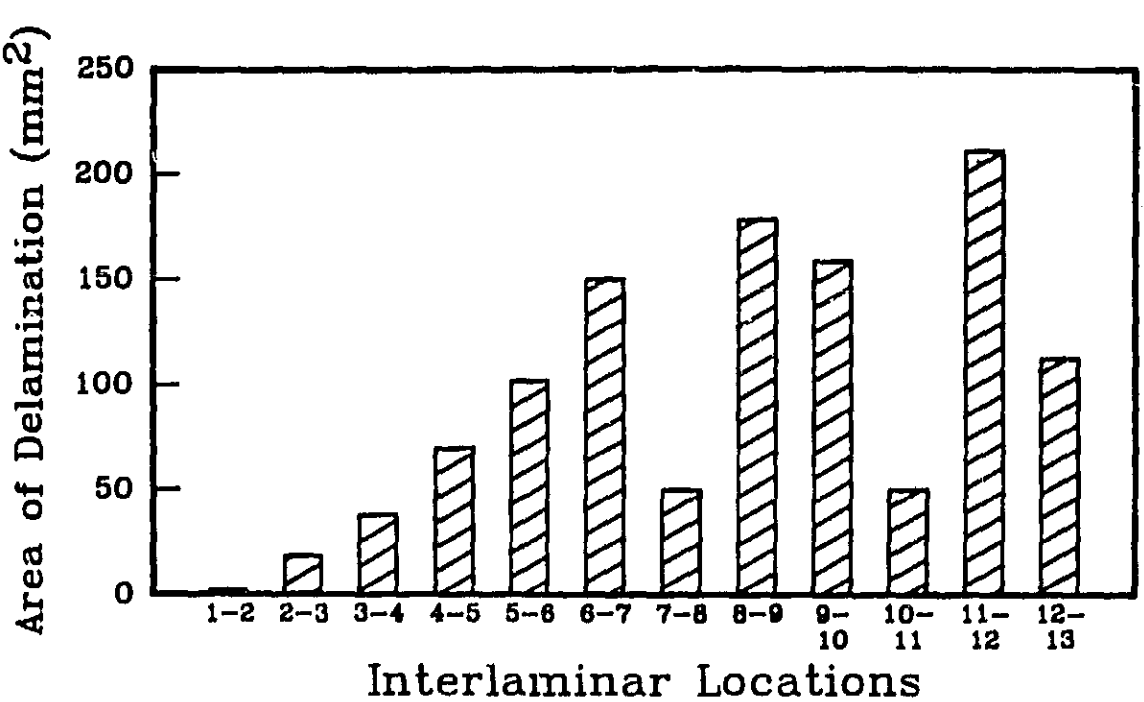


Figure 4 - Area of damage versus depth for one impact site. The impacted surface is adjacent to interlaminar location 1-2.

surface such as that in interlaminar location 1-2 will be significantly attenuated. We also note that qualitative superposition of similarly oriented damage zones suggests that the zones overlap to a significant extent, so that the largest damage zone provides a good estimate of the superposition. Combining these two considerations with the fact that we interrogated from the back surface where damage is more extensive led us to the following simple correlation criterion. The polar backscatter scan of a given orientation was correlated with the deply information from the interlaminar location exhibiting the largest area of similarly oriented damage, as indicated in Table 1. Specifically, we chose to correlate the deply information from interlaminar location 11-12 with the +45° polar backscatter image, interlaminar location 8-9 with the -45° polar backscatter, interlaminar location 9-10 with the 90° polar backscatter, and interlaminar location 12-13 with the 0° polar backscatter.

The approximate size, shape and orientation of damage is qualitatively correlated in Figures 5 and 6. The area represented by each image in these figures is approximately 2.6 cm by 3.7 cm. The scaling allows direct comparison between polar backscatter images and deply photographs.

The left panel of Figure 5 presents the polar backscatter image obtained with the interrogating ultrasound perpendicular to the +45° fiber orientation. The right panel presents the corresponding deply photograph from interlaminar location 11-12 of that impact site. The damage visualized by both techniques is clearly oriented along the +45° direction, as defined from the top of the sample. (Both of these evaluation techniques are examining the specimen from the bottom, so that the +45° and -45° orientations appear to be exchanged.) The shape and extent of damage in each panel are in good qualitative agreement.

Figure 6 presents the three remaining orientations for this impact site. The top panels present the results of interrogating perpendicular to the -45° orientation and the corresponding deply photograph from interlaminar location 8-9. The middle panels present the 90° orientation and interlaminar location 9-10, the lower panels 0° and interlaminar location 12-13. There is good qualitative agreement between polar backscatter and deply for the orientation, size, and general shape of the damaged regions. A quantitative correlation of these techniques can be obtained from the estimates of area. Figure 7 presents a correlation plot of the damage area as determined by polar backscatter versus the damage area determined by the deply technique. The linear correlation coefficient, calculated by including the error estimates shown in Figure 7 is $r = 0.88$.

Polar Backscatter Image

Deply Photograph of Interlaminar Location 11-12

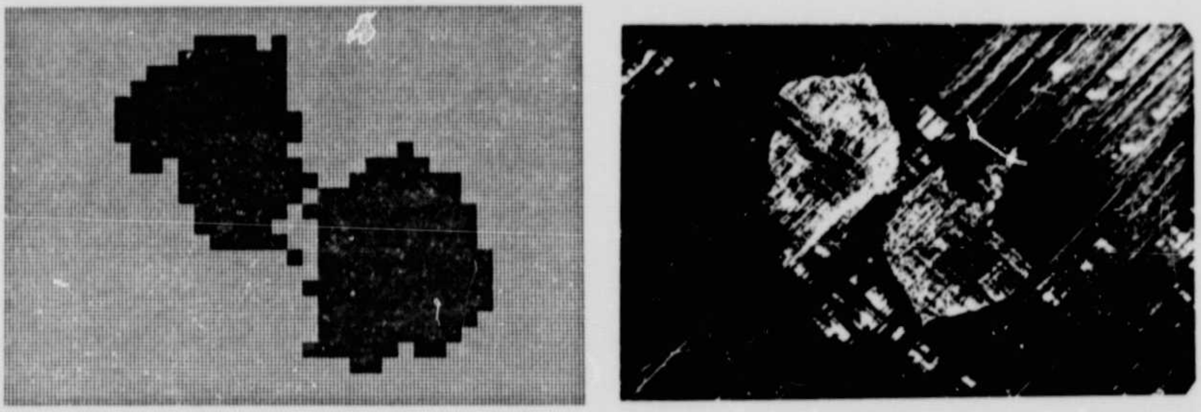
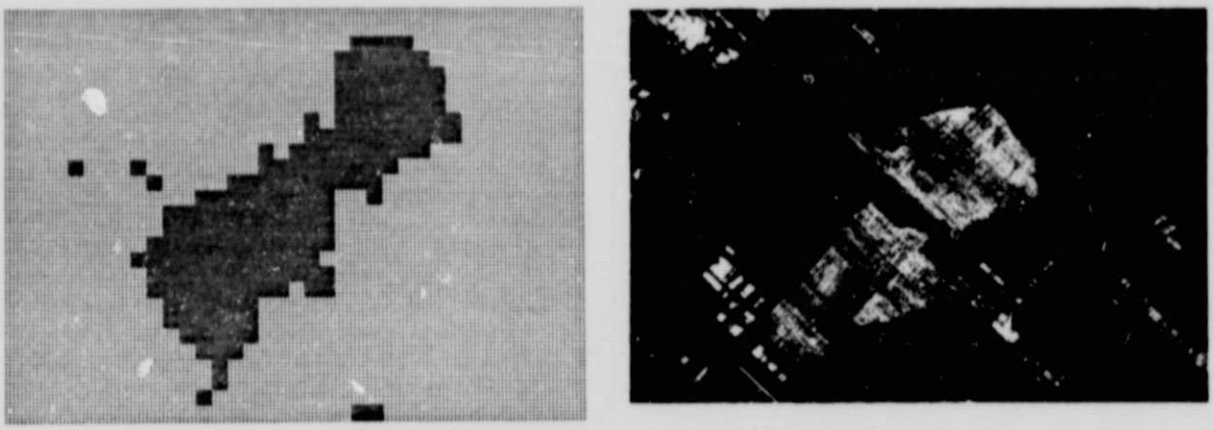
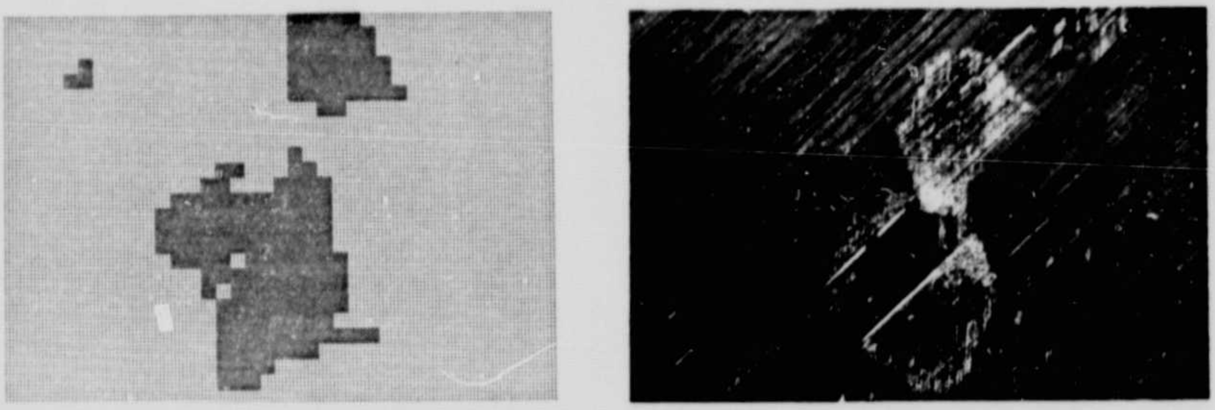


Figure 5 - Comparision of apparent zones of damage imaged by the polar backscatter technique (left panel) with photograph of damage indication from the deply technique. Orientation of the damage is along the +45° direction, as defined from the front (impacted) side.

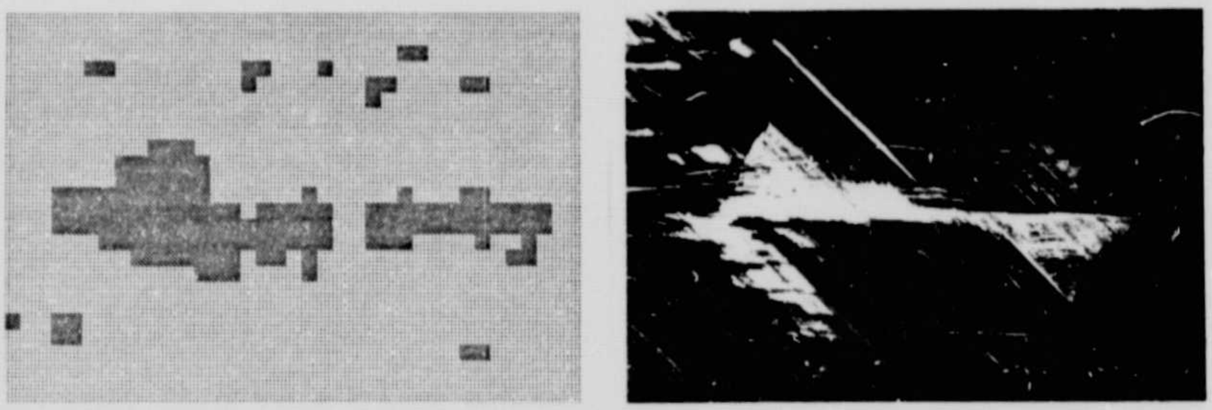
Polar Backscatter Image | **Deply Photographs**



a) -45° orientation : Interlaminar Location 8-9



b) 90° orientation : Interlaminar Location 9-10



c) 0° orientation : Interlaminar Location 12-13

Figure 6 - Comparision of apparent zones of damage as imaged by the polar backscatter technique (left panels) with photographs of damage indication from the deply technique (right panels).

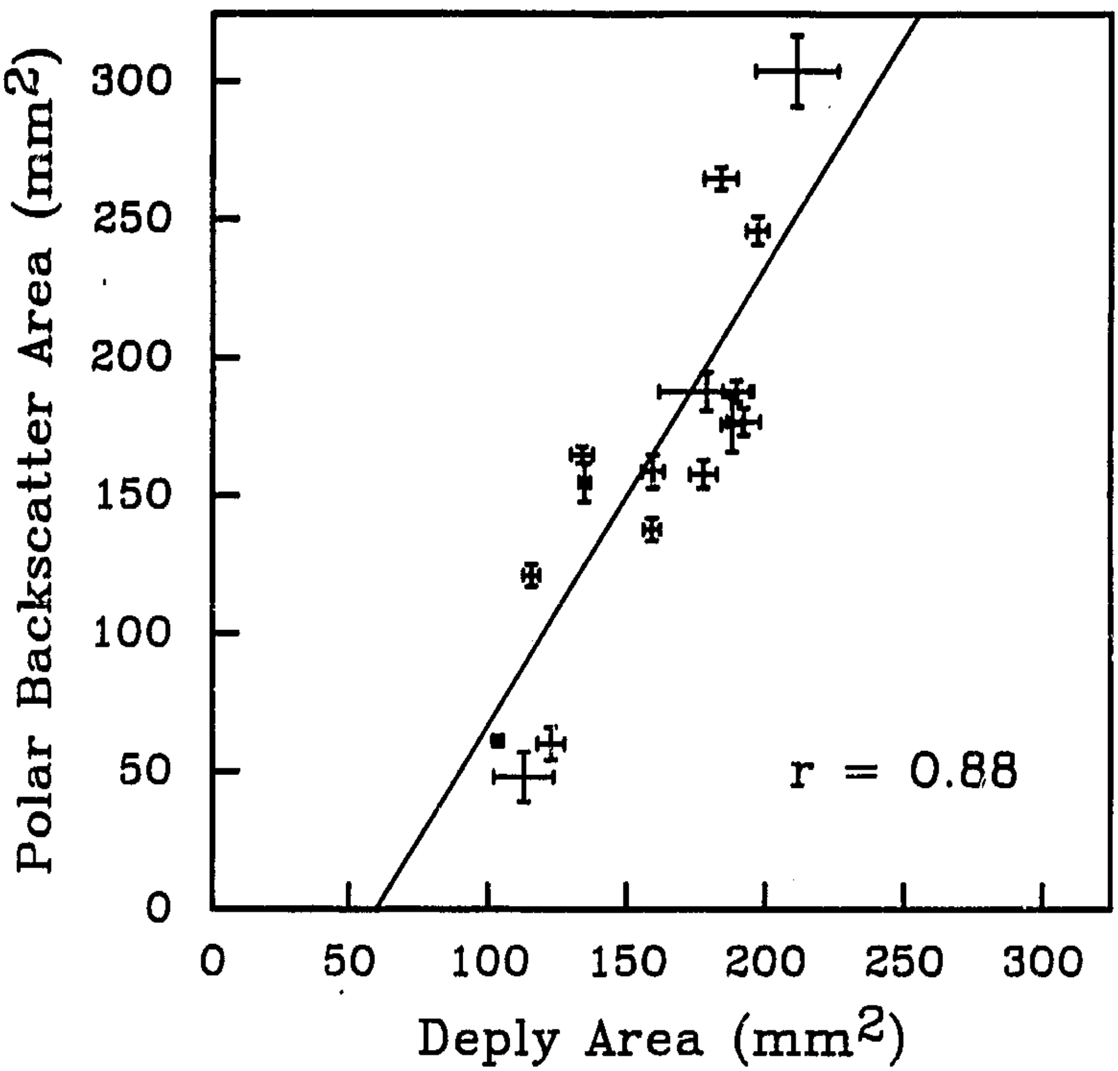


Figure 7 - Correlation of damage areas estimated from polar backscatter with corresponding damage areas derived from the deply technique.

III. CONCLUSIONS

The size, shape, and orientation of damage correlates well between the polar backscatter technique and the deply technique. Further, there is good quantitative correlation between the areas of damage indicated by the two techniques. These results suggest that the polar backscatter technique is sensitive to specific orientations of damage. The polar backscatter technique provides a good qualitative image of the size and shape of the largest zone of damage in each of the principal orientations. A quantitative estimate of the extent of these largest damage zones can be obtained from the polar backscatter technique. The selective sensitivity of polar backscatter may thus provide a useful tool for further studies of the mechanisms of impact damage in graphite-fiber reinforced composite laminates.

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