

# A Positron Annihilation Spectroscopy Study of Carbon-Epoxy Composites\*

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## Synopsis

Many positron annihilation spectroscopy (PAS) studies have been carried out during the last several years for characterizing various polymeric materials. In this work, the PAS technique is used to investigate some positron annihilation characteristics of the combination of carbon fibers with epoxy polymers to form carbon-epoxy composites. These composites are receiving wide and increasing application in industrial markets where light-weight materials with high strength and excellent fatigue characteristics are required. We have found that PAS is capable of detecting different concentrations of carbon fiber in these composites and a relationship is shown to exist between fiber fractions found by PAS and apparent volume and weight fractions found by density measurements. Positron annihilation centers have also been found that we believe were created at the carbon-epoxy interface when the composite was prepared. Characterization of this interface is important for the development of composites with improved mechanical properties.

## INTRODUCTION

When a high-energy positron from a radioactive source, such as  $^{22}\text{Na}^1$  enters a solid molecular material, it will lose energy by colliding with electrons and thermalize in a few picoseconds.<sup>2,3</sup> After thermalization, the positron can remain in its free state, combine with molecular species in the solid, or extract an electron to form a bound state known as a positronium (Ps) atom. Positronium exists in two ground states, depending upon the relative spins of the positron and electron. *Para*-positronium (*p*-Ps) with spins antiparallel has a lifetime of about 125 ps, whereas *ortho*-positronium (*o*-Ps) with spins parallel has a lifetime of about 140 ns in vacuum.<sup>4</sup> Positrons and *p*-Ps annihilate with the formation of two photons, each having an energy of 511 KeV. In vacuum, annihilation of *o*-Ps produces three photons of varying energies which total to 1.022 MeV. In condensed matter, however, *o*-Ps during its lifetime undergoes many collisions with surrounding electrons which can react with the *o*-Ps atom to cause *para*-annihilation of the positron with the emission of only two photons.<sup>5</sup> This normal decay process known as pickoff

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reduces the lifetime of *o*-Ps to the order of a few nanoseconds. Much longer lifetimes of the order of 100 nanoseconds have been reported for finely divided materials with large surface areas.<sup>6,7</sup>

Following thermalization, positrons and positronium tend to become localized in regions of low electron density. These sites can be surfaces or defects such as vacancies, voids, and dislocations, or free volume in amorphous polymers. As the size of these regions increases the local electron density decreases. This causes an increase in the lifetime of the trapped *o*-Ps due to the lower probability of an interaction with an electron to undergo pickoff. The intensity of the *o*-Ps annihilations increases with an increase in population of the low electron density sites due to the higher *o*-Ps formation and trapping rate.

Experimental positron lifetime spectra of annihilation intensity versus lifetime are normally described as the sum of decaying exponentials.<sup>8-10</sup> This is shown in Eq. 1 where  $dn/dt$  is

$$-dn/dt = \sum_{i=1}^N I_i \lambda_i \exp(-t\lambda_i) \quad (1)$$

the count rate of the annihilating positrons at time  $t$  for a data channel width  $dt$ ,  $I_i$  represents the relative intensity, and  $\lambda_i$  is the positron annihilation decay rate. The model of the spectra usually contains two to four exponential terms which are convoluted with an instrumental resolution function. The number of terms is a function of the number of conditions under which the positrons annihilate in the material, that is, bulk, vacancy, region of free volume, etc. The magnitude or relative intensity of each term is a measure of the probability that the positron annihilated under the influence of a particular environment in the material and the decay rates represent the annihilation rates or the reciprocal of the positron mean lifetimes  $\tau_i$ .

Many positron annihilation spectroscopy (PAS) studies have been carried out during the last several years for characterizing various polymeric materials.<sup>11-18</sup> The positron lifetime spectra for these materials can usually be described with three exponential terms. The term with the shortest lifetime (0.1 to 0.3 ns) is usually associated with annihilation of *p*-Ps and Ps-molecular species, the intermediate lifetime (0.3 to 0.5 ns) with positrons and positron-molecular species, and the long lifetime (1 to 3 ns) with the decay of *o*-Ps by electron pickoff.<sup>19,20</sup> The annihilation of *o*-Ps in polymers is essentially controlled by the free volume characteristics of the polymer.<sup>18</sup> The size and concentration of the free volume "holes" are a function of the degree of cross linking in the polymer and depend upon the chemical formulation and the manner in which the polymer is prepared.

In this work the PAS technique is used to investigate some positron annihilation characteristics of carbon fibers, epoxy polymers, and carbon-epoxy composites. These composites are receiving wide application not only in the aircraft industry but also in recreational and other industrial markets. They are lightweight and are ideal in applications where strength and excellent fatigue characteristics are required.

## EXPERIMENTAL

### Composite Preparation

Six composites were used in this investigation. The epoxy (DER) for three of the composites was formed from diglycidyl ether of bisphenol A (Dow Chemical) and polyether triamine.<sup>21</sup> Two of these composites were prepared with carbon fibers<sup>22</sup> made from polyacrylonitrile (PAN); one with a low modulus fiber (PNL) and one with a high modulus (PNH). The third composite was prepared with a fiber made from pitch (PTC). The fibers were wound around the base of an aluminum mold which had been sprayed with a release agent. The epoxy resin was heated to 70°C, mixed with the amine in a ratio of 2.1:1 (resin:amine), and poured over the fibers to a depth of approximately 1 mm. The mold was placed in a vacuum chamber, degassed for 1 h at room temperature, and then vacuum cured at 80°C for 24 h. After compression molding for 16 h at 85°C the composite was allowed to cool to room temperature and then demolded. The other three composite samples were prepared by the Great Lakes Carbon Corporation. The epoxy (EPN) for these three composites was prepared from EPON 826 resin (Shell Chemical). This is a nondiluted bisphenol-A resin of low viscosity and an epoxide equivalent weight of 178–186. The EPON 826 was cured with metaphenylene diamine in a ratio of 7.1:1 (resin:amine). The curing cycle was 30 min at a set temperature of 100°C with pressure then being applied. The set temperature was adjusted to 150°C for the next 45 min, and the composites were then allowed to cool for demolding. These three samples differed in that one contained PAN fibers that had received a “high” surface treatment, one a “medium” surface treatment, and one a “low” surface treatment. The fibers are referred to as FHI, FME, and FLW, respectively. One point of the investigation was to determine if these proprietary surface treatments carried out by the Great Lakes Carbon Corporation were distinguishable with PAS.

### Density Measurements

Density measurements were made for the different carbon fibers, epoxies, and composites. The densities of the fibers were determined by the density gradient technique, and differential weights in air and alcohol were used for calculating the densities of the epoxies and composites. These density values were used for calculating the apparent volume and weight fractions of the carbon fibers in the composite materials.

### Positron Lifetime Measurements

The positron lifetime measurements were made using the standard fast-fast coincidence system.<sup>23,24</sup> The timing resolution of the system was determined by measuring coincident photons from <sup>60</sup>Co. The resolution function was determined to be the sum of two Gaussians with a combined FWHM of about 320 ps. The <sup>22</sup>Na source was prepared by evaporating about 40  $\mu$ Ci of a 200  $\mu$ Ci/mL solution of <sup>22</sup>NaCl (Amersham Corp.) onto an aluminum foil (8.5  $\mu$ m), which was then folded into a flat configuration. The 1.28 MeV photon from the decay of a <sup>22</sup>Na atom in the source was used as the start signal for

the birth of a positron, and detection of the coincident 0.51 MeV annihilation photon was used as the stop signal, the time interval between the two signals being equal to the lifetime for the particular positron. The positron annihilation spectra were recorded for the different samples by accumulating the annihilation events versus time until the prompt or "zero time" peak had accumulated 50,000 counts. The total number of annihilation events in each spectra from zero to 60 ns was about 600,000. About six spectra were taken for each sample. The zero time points of the spectra were adjusted to correct for instrument drift, and the counts were summed to improve the statistics. The computer program, Positronfit Extended,<sup>25</sup> was used for modeling the spectra, and a correction was applied for the positrons that annihilated in the source.

Spectra for the carbon fibers were taken on samples prepared by wrapping about one half of a fiber bundle around a small Teflon spool to the depth of several millimeters. The source was then inserted, and the rest of the bundle was wrapped. This assembly was placed in a chamber and evacuated to a pressure of about  $10^{-5}$  torr before taking the data. For the epoxy and composite samples, the source was inserted between two flat 1-mm thick sections of the materials, and the data were taken at atmospheric pressure in air.

## RESULTS AND DISCUSSION

Two approaches were taken for analyzing the positron annihilation information in the spectral curves. One approach involved the comparison of the entire PAS spectra for each carbon fiber, epoxy, and corresponding composite material. The other dealt with the lifetimes and intensities for these materials obtained with the computer modeling program.

### Positron Lifetime Spectra

The spectra of the PTC carbon fiber, the DER epoxy, and the PTC/DER composite are shown in Figure 1. A spectrum of pyrolytic graphite (PGR) is also shown for comparison. If the experimental system were capable of infinite resolution, each curve would appear as a delta function at time zero followed by the exponential decay. The convolution of a finite resolution function with the decay curve produces a peak near time zero with the accompanying tail shown at negative times.

Positrons annihilate very quickly in pyrolytic graphite due to its crystalline structure and low concentration of defects. The PGR curve decays very rapidly and is defined by two exponential terms as determined by the computer modeling program. Essentially all of the decay is related to a short lifetime component,  $\tau_1 = 0.2$  ns, with very little intermediate and no long lifetime component which agree with the reported values.<sup>26</sup> The PTC carbon fiber spectrum decays less rapidly and is defined by three exponentials with about 0.5% of the intensity due to a long lived component,  $\tau_3 = 3.0$  ns. The DER epoxy spectrum also requires three exponential terms for definition. However, the intensity of its longlived component,  $\tau_3 = 1.7$  ns, is about 27%, showing that an appreciable fraction of the positrons entering the sample result in the formation of *o*-Ps in the free volume "holes" of the epoxy. The

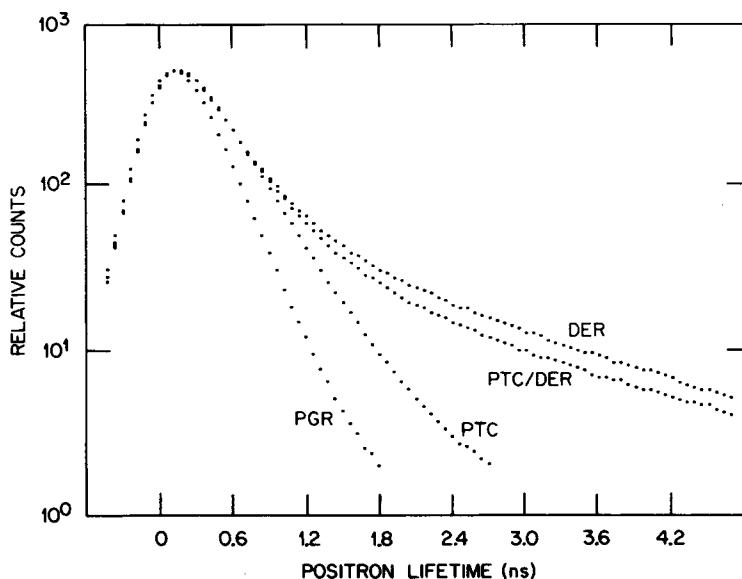


Fig. 1. Positron annihilation spectra for pyrolytic graphite (PGR), carbon fiber (PTC), composite (PTC/DER), and epoxy (DER).

PTC/DER composite curve decay rate is shown to be intermediate between the curves for the carbon fiber and the epoxy.

The PTC/DER composite was prepared to contain a relatively low concentration of carbon fibers, and as a result the decay curves for the epoxy and the composite are seen to be rather close at the longer lifetimes. As the concentration of fibers increases, these two curves become more widely separated, as shown in Figure 2, for the FME/EPN composite. This composite contains about 4.5 times as much carbon fiber by volume as the PTC/DER composite. The addition of fiber to the epoxy for composite preparation has the effect of diluting the epoxy, and the annihilation intensity becomes smaller as the relative amount of epoxy in the composite becomes smaller.

Because the composite is a physical and not a chemical mixture, it seemed reasonable that the addition of appropriate fractions of the carbon fiber spectrum and the epoxy spectrum should produce a fit of the experimental composite curve. For this to be true, it would be necessary that no appreciable effect be introduced by the junction created at the fiber-epoxy interface. The fractions of the carbon fiber and epoxy spectra necessary for the fit were calculated by minimizing the sum of the squares of the relative differences between the experimental data points of the composite spectrum and the fractional sums of the corresponding data points for the carbon fiber and the epoxy spectra (areas under the three curves were first normalized for equal number of total annihilation events). A representation of the degree of fit that was found is shown in Figures 3 and 4. Figure 3 is a semilog plot showing the comparison for the PTC/DER composite and the fractional sums of the corresponding PTC carbon fiber and DER epoxy spectra (pseudocomposite). Every other data point for the composite spectrum was plotted for clarity. A more revealing indication that the fit is quite good is shown in Figure 4, which

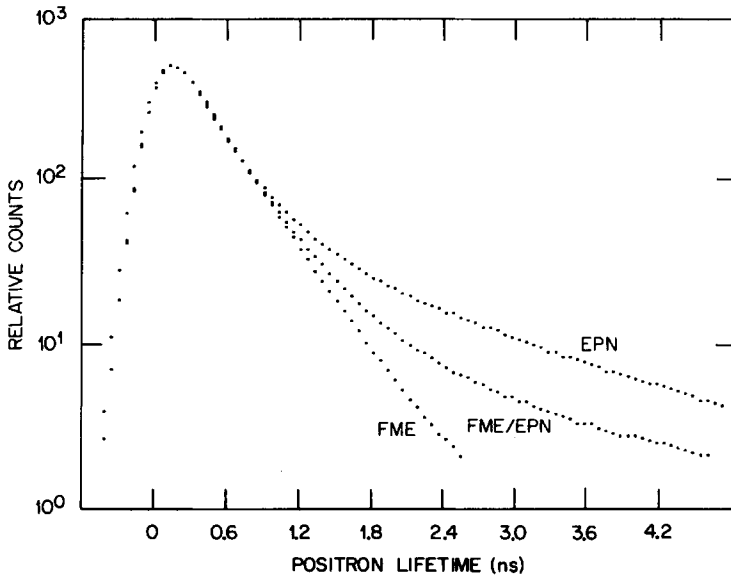


Fig. 2. Positron annihilation spectra for carbon fiber (FME), composite (FME/EPN), and epoxy (EPN).

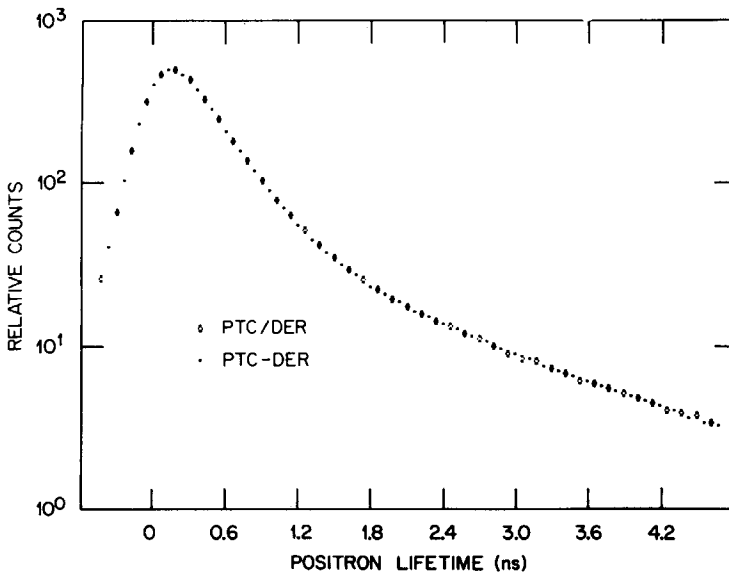


Fig. 3. Comparison of the fit of the composite spectrum (PTC/DER) with the fractional sum of the PTC carbon fiber and DER epoxy spectra (PTC-DER).

is a linear plot of the statistical variations in the relative differences between the corresponding points for the two curves. Similar relative difference curves were obtained for the PNL/DER and PNH/DER composites.

In some cases it is possible to distinguish between a fiber that is expected to be in a composite from one that is actually present. In cases where the spectra

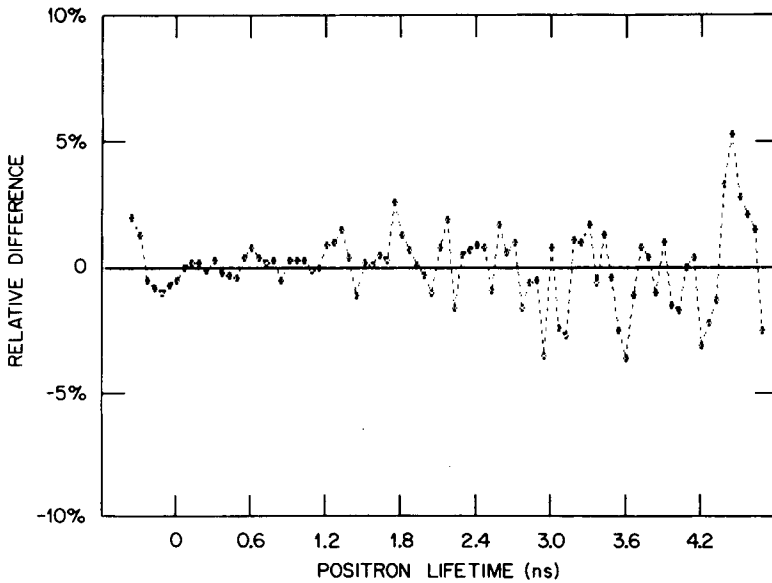


Fig. 4. Relative differences for the fit of the PTC/DER composite spectrum with the fractional sum of the PTC carbon fiber and DER epoxy spectra shown in Figure 3.

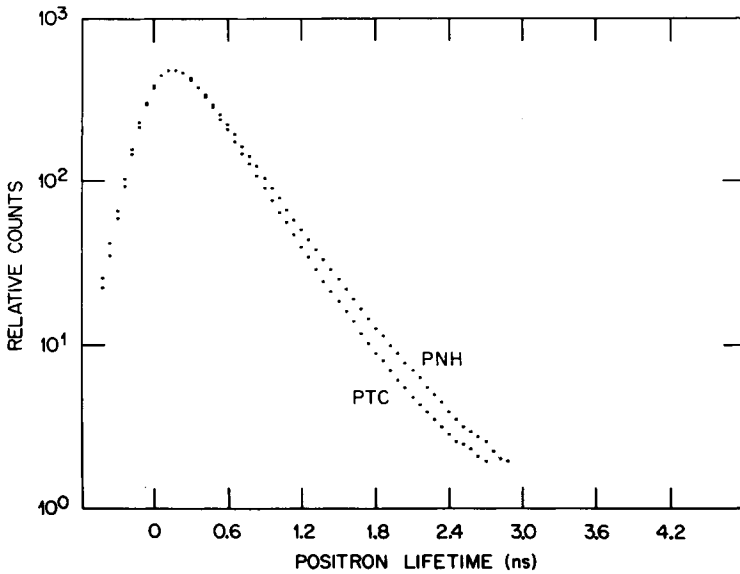


Fig. 5. Annihilation spectra for the PTC and PNH carbon fibers.

for the two fibers are sufficiently different, as shown in Figure 5 for the PNH and PTC fibers, attempts to make a least-squares fit of the spectrum of the composite using the spectrum of the fiber that was not present in the composite produced a poor fit. This is represented by the relative difference curve in Figure 6 where the spectrum for the high modulus fiber (PNH spectrum in Fig. 5) was used to fit the PTC/DER composite containing the

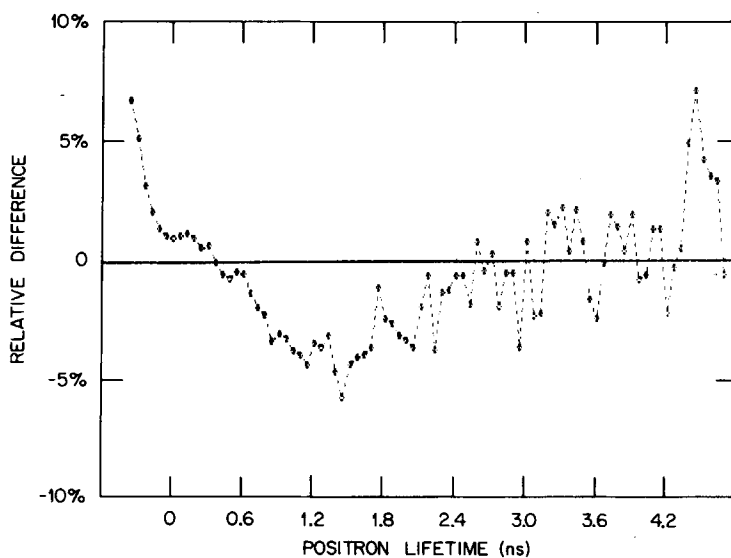


Fig. 6. Relative differences for the fit of the PTC/DER composite spectrum with the fractional sum of the PNH carbon fiber and DER epoxy spectra.

PTC fiber. The poor fit is obvious when compared to Figure 4 where the proper fiber spectrum was used.

It also appears that it is possible to detect the formation of new sites for positron annihilations in the composite that were not initially present in the fibers or the epoxy. This is represented in Figure 7, which is a relative difference plot for the FHI/EPN composite. The fit is good at the longer lifetimes where the intensity from the epoxy is predominant but consistently

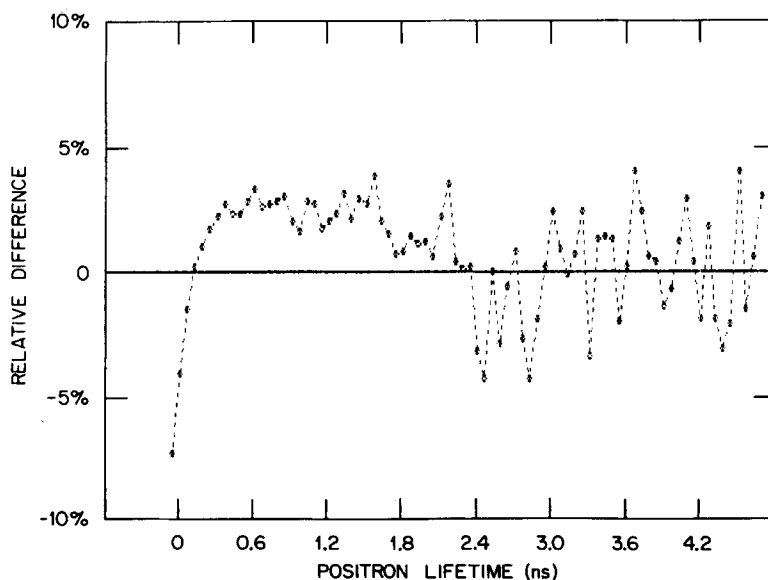


Fig. 7. Relative differences for the fit of the FHI/EPN composite spectra with the fractional sum of the FHI carbon fiber and DER epoxy spectra.



shows small positive deviations (composite > pseudocomposite) at lower lifetimes (< 2.0 ns) where the intensity from the fiber is large. This effect was also seen for the FME/EPN and FLW/EPN composites. As mentioned previously, the fibers in these composites received varying degrees of a proprietary surface treatment before fabrication. We conclude that this treatment has caused the formation of an interfacial component between the fiber and the epoxy that is detected as a higher intensity for the composite spectrum. This higher intensity is in the right direction for the presence of more trapping sites for the positrons in the prepared composite that would not be present in the pseudocomposite represented by the fractional sums of the fiber-epoxy spectra. Unless the surface treatment caused a change in the surface area of the fibers these sites would not necessarily show up in the fiber spectra alone because the fiber data were taken under vacuum. No degree of fiber surface treatment was evident for these three composites.

### Positron Lifetimes and Intensities

Each of the annihilation spectra for the carbon fibers, the epoxies, and the composites were modeled with the computer program Positronfit Extended. Best fits were obtained with the sum of three exponential terms for each spectrum. The values of the lifetimes and the intensities are listed in Table I.

The PNL/DER, PNH/DER, and PTC/DER composites were prepared to contain a relatively low concentration of carbon fibers (10 to 15% by volume), whereas the composites prepared with the EPN epoxy resin contained fiber concentrations within the range normally found in industrial materials (approx. 60%). These differences in fiber concentrations can be recognized in a

TABLE I  
Positron Lifetime and Intensity Results for Carbon Fibers, Epoxies, and Composites

	Lifetimes, ns			Intensities, %		
	$\tau_1$	$\tau_2$	$\tau_3$	$I_1$	$I_2$	$I_3$
<b>Fibers</b>						
PNL	0.20	0.40	3.00	21.3	78.3	0.4
PNH	0.25	0.43	2.80	26.1	73.2	0.7
PTC	0.29	0.40	2.88	49.0	49.7	1.3
FLW	0.14	0.39	1.55	12.3	87.2	0.5
FME	0.15	0.39	1.48	13.2	86.2	0.6
FHI	0.16	0.40	1.59	15.3	83.9	0.8
<b>Epoxies</b>						
DER	0.17	0.37	1.74	24.8	47.7	27.5
EPN	0.16	0.39	1.72	26.7	47.4	25.9
<b>Composites</b>						
PNL/DER	0.21	0.40	1.76	32.1	46.1	21.8
PNH/DER	0.24	0.45	1.78	41.9	38.8	19.6
PTC/DER	0.24	0.42	1.78	41.3	38.2	20.5
FLW/EPN	0.16	0.39	1.76	19.0	71.0	10.0
FME/EPN	0.16	0.39	1.74	18.7	71.1	10.2
FHI/EPN	0.17	0.41	1.74	21.1	69.1	9.8

qualitative manner from a cursory examination of the values for the  $I_3$  intensities listed in Table I. These are the annihilation intensities related to the long lifetime ( $\tau_3$ ) decay of *o*-Ps in each material. The  $I_3$  values for the two epoxies are in the 26 to 27% range, while those for all of the fibers are very low in comparison. Compared to the epoxies, the composites contain a smaller concentration of free volume "holes" per unit volume corresponding to how much of the carbon fiber was used in their preparation (dilution effect). This dilution effect is apparent from the lower values of the  $I_3$  intensities for the composites compared to those for the epoxies. The  $I_3$  values for the three DER composites with low fiber content are about 25% lower than that for the DER epoxy. The values for the three EPN composites with high fiber content are about 60% lower than the corresponding EPN epoxy.

### Carbon Fiber Fractions

The fraction of the carbon fibers in the composites, as determined by (1) the fiber-epoxy spectra summation technique and (2) the positron lifetime data, were correlated to the apparent volume and weight fractions determined from the density measurements of the fibers, epoxies, and composites. The basic equation used for estimating the fiber fraction (FF), both for the least-squares treatment for summing the fiber and epoxy spectra and from the lifetime data in Table I, is

$$FF \times f_i + (1 - FF) \times e_i = c_i \quad (2)$$

For the spectra summing method,  $f$ ,  $e$ , and  $c$  represent the corresponding data points on the normalized fiber, epoxy, and composite spectra, respectively. Approximately one hundred data sets of three points each were used for the parameters in Eq. 2 from time zero to about 5 ns. The expression used for determining the relative difference values for each data set for the least-squares calculation is

$$D_i = ((FF \times f_i + (1 - FF) \times e_i) - c_i)/c_i \quad (3)$$

The partial derivative of the sum of the squares of the relative differences with respect to FF was set to zero, and the resulting expression was solved for FF.

For the positron lifetime data, the parameters  $f$ ,  $e$ , and  $c$  in Eq. 2 represent evaluations of  $-dn/dt$  in Eq. 1 for the fiber, epoxy, and composite, respectively, using the intensity and lifetime values in Table I. Evaluation points on the time scale of the annihilation spectra were taken at times where most of the fiber spectrum had decayed and the major contribution to the intensity was from the longlived component, *o*-Ps. Values for the fiber fractions obtained from the lifetime data and from the spectra summing technique are compared in Table II to the volume fractions and weight fractions calculated from the density measurements. The values obtained from the lifetime data and the spectra-sum data agree very well.

A graphical comparison of the positron data and the volume fraction data is shown in Figure 8. It seems reasonable to assume that the zero coordinates lie on the correlation curve. Under this assumption, the relationship between the

TABLE II  
Composite Fiber Fraction Values from Positron Data and Density Measurements

Composites	Fiber Fraction			
	Positron data		Density data	
	LT <sup>a</sup>	SS <sup>b</sup>	VF <sup>c</sup>	WF <sup>d</sup>
PNL/DER	0.20	0.20	0.11	0.16
PNH/DER	0.27	0.26	0.14	0.20
PTC/DER	0.25	0.24	0.13	0.20
FLW/EPN	0.61	0.61	0.57	0.67
FME/EPN	0.62	0.61	0.59	0.68
FHI/EPN	0.64	0.64	0.58	0.67

<sup>a</sup>Lifetime measurements.

<sup>b</sup>Spectra-sum technique.

<sup>c</sup>Volume fraction.

<sup>d</sup>Weight fraction.

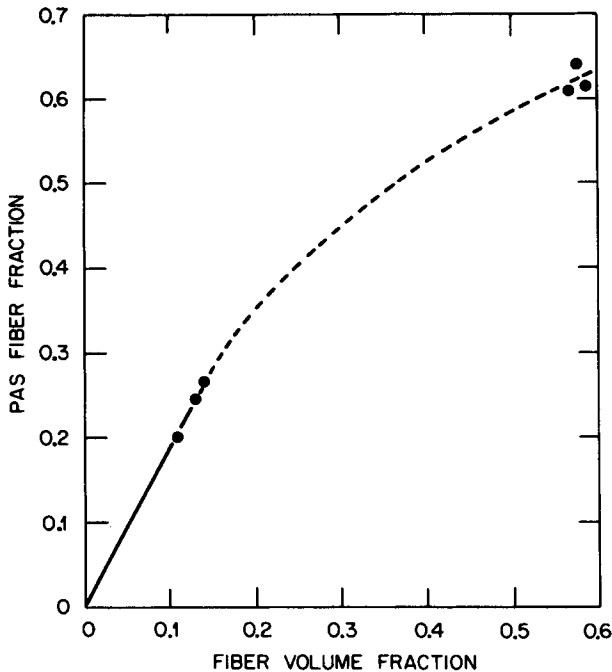


Fig. 8. Comparison of composite fiber fractions from positron data with apparent fiber volume fractions from density measurements (Table II).

PAS fiber fraction values and the fiber volume fraction values appears to be linear up to about 0.15 volume fraction. The shape of the curve above this point is presently uncertain. A similar correlation was found for the weight fraction data. For a more precise definition of the correlation, it will be necessary to analyze more composite systems. This is in our future plans where composites of lower and intermediate fiber concentrations will be investigated.

Although the spectra-summing technique did not reveal any relative differences for the degree of surface treatments for the FLW, FME, and FHI fibers in the EPN composites, it is interesting to note the monotonic changes in the intensity values ( $I_1$ ,  $I_2$ ,  $I_3$ ) for these fibers in Table I. More information would be necessary on the type and degree of surface treatment before any possibility for relevancy could be assigned to these intensity changes. Future work will include a more detailed study of the fiber-epoxy interface for fibers with well characterized surfaces.

## CONCLUSIONS

It has been shown that positron annihilation spectroscopy is a useful technique for the nondestructive characterization of carbon-epoxy composites. Annihilation parameters in this physical mixture are additive, and in some cases the presence of the correct fiber in the composite can be verified. It is also possible to detect the formation of positron annihilation centers created at the fiber-epoxy interface when the composite is prepared. It is important to be able to characterize this interface for the development of stronger and more elastic composites.

We feel that PAS can be developed for use as a quality assurance technique for composite fabrication, and the development of field test equipment for manufactured products is feasible. To this end, we are designing a source-detector combination which will incorporate  $^{68}\text{Ge}^{27}$  as a positron source instead of  $^{22}\text{Na}$ . The higher energy positrons from the  $^{68}\text{Ge}$  will have a greater depth of penetration and will therefore provide for the analysis of thicker composites.

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