Activation of Bifunctional Coupling Agents in Fiberglass/Polyethylene Composites by Electron Beam

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ABSTRACT: Electron beam (EB) radiation was investigated as a means to initiate coupling between the fiberglass and plastic phases in fiberglass/polyethylene plastic composites using two bifunctional compounds, 12-azido-1-diazo-2-dodecanone (**A**) and 1-diazo-17-octadecene-2-one (**B**). Chemical studies reveal that EB radiation has the potential to bind both of these compounds to fiberglass. Fiberglass coated with either **A** or **B** shows reduced values of percentage recovery upon exposure to EB, indicating a reaction between these compounds and the glass surface. However, even 400 kGy of radiation was not as effective as a heat treatment for 45 min at 150°C. To test the effectiveness of EB radiation to couple these compounds to polyethylene, fiberglass samples were heat-treated with compounds **A** and **B**, followed by extrusion mixing with polyethylene, and exposure of molded tensile and impact samples to EB radiation. Compound **B** showed the best overall ability to couple with the polyethylene matrix, but a 400-kGy dose was necessary to bring about substantial coupling. At 400 kGy, samples containing **B** showed a 23% improvement in tensile properties and a 30% change in Izod impact. © 2002 John Wiley & Sons, Inc. J Appl Polym Sci 83: 2579–2594, 2002

Key words: electron beam radiation; fiberglass/polyethylene composites; bifunctional compounds; coupling agents

INTRODUCTION

Novel bifunctional compounds have the capacity to improve the mechanical properties in fiberglass-filled polyethylene composites. The presence of these surface-active compounds allows the two chemically dissimilar materials, fiberglass and polyethylene, to be coupled together, so that the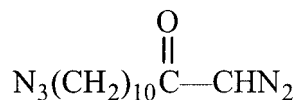 resulting composites display increased hardness and tensile modulus, and generally a range of properties that extend beyond those exhibited by unaltered polyethylene. Covalent bonding between high-density polyethylene (HDPE) and glass spheres with a silane coupling agent has been shown to have a significant influence on the stress/strain behavior[1–3] and creep properties of these composites at room temperature. As glass is added, the modulus drops, but increases again as the coupling agent is added. Creep behavior drops dramatically when a silane coupling agent is present.[4] In the first study of this series,[5] the decomposition of two compounds, 12-azido-1-diazo-2-dodecanone (compound **A**) and 1-diazo-
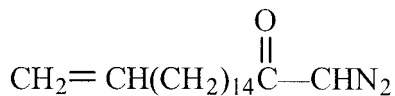
17-octadecene-2-one (compound **B**), under heat and UV light was studied. The effectiveness with which heat and UV light were able to bind these compounds to the surface of glass fibers was also investigated. Finally, several polyethylene/fiberglass composites were prepared by extrusion with **A** and **B** as coupling agents.

Compounds **A** and **B** are both part of a family of bifunctional compounds originally synthesized by McGarvey and Holden,[6,7] and intended to act as amphiphilic bifunctional compounds in composites containing glass and Al_2O_3. At one end of the molecule both **A** and **B** contain a heat- or light-sensitive diazoketone group, which can bind to the hydroxyl groups on a glass surface. At the other end of the molecule, **A** contains a heat-sensitive azide group and **B** an alkene group, which can bind to the polymer in the presence of the free radicals produced during processing. Thus, both of these compounds are excellent candidates to serve as coupling agents between fiberglass and polyethylene. The structures of compounds **A** and **B** are as follows:

$$N_3(CH_2)_{10}\overset{\overset{\displaystyle O}{\|}}{C}\text{---}CHN_2$$

Compound A

$$CH_2\text{==}CH(CH_2)_{14}\overset{\overset{\displaystyle O}{\|}}{C}\text{---}CHN_2$$

Compound B

Previous work to couple glass and polyolefin plastics has concentrated on the use of silane based coupling agents containing either a heat sensitive azide group or amino group at one end, and alkoxysilane groups at the other end of the molecule.[8–12] These coupling agents have also been shown to be useful in polyolefin composites containing mica, TiO_2, carbon black, flax, and hard clay.[13–15] Maurer and Welander[16] have shown that an increase in silane coupling agent in these composites reduces the free volume in the interphase region between glass and polyethylene, indicating that bridging takes place between the two phases. However, silane-based coupling agents are hydrolyzable, and the Si–O–glass bond can be broken to form an organosilane triol, resulting in poor chemical bonding between filler,

coupling agent, and the polymer.[17,18] The coupling agents that are the subject of this series of studies are not subject to disadvantageous hydrolysis, making them potentially more suitable as bridging compounds in fiberglass/polyethylene composites.

In the previous study of this series[5] treated fiberglass was prepared by applying the coupling agents to the surface of the fiberglass, followed by a heat or UV treatment. These were extrusion mixed with high-density and linear low-density polyethylene, and the mechanical properties of the composites were determined. Each one produced a composite that had improved properties over composites to which no coupling agent had been applied. A variety of extrusion conditions with variations in temperature and residence time, N_2 blanketing, and addition of benzoyl peroxide or AIBN to the extruder provided relatively little change in properties with variation in these conditions. The best composites were produced when compound **A** was applied by heat at 150°C, and extrusion undertaken at high temperatures and relatively long residence times. The increase in tensile properties in these samples was approximately 20%, and reduction in Izod impact approximately 35%. Composites made from both **A** and **B** showed better properties than those prepared with a commercial organosilane-based coupling agent. Izod impact appears to be the most sensitive indicator of coupling between the two phases.

In the present work compounds **A** and **B** were studied under electron beam (EB) to determine whether this method could be used to activate these compounds as coupling agents in fiberglass/polyethylene composites. The decomposition rates of the diazoketone, azide, and alkene groups on these molecules were observed by FTIR and NMR spectroscopy. Samples of compounds **A** and **B** were also coated onto fiberglass and then reacted under EB radiation. The extent to which these compounds could bind themselves to the fiberglass surface was determined by the ease with which they could be extracted from the fiberglass surface after exposure to EB.

Several samples of fiberglass, with compounds **A** and **B** bonded to the surface by heat, were extrusion-mixed with polyethylene at relatively low temperatures. Molded tensile and impact specimens of the composites were exposed to EB radiation at various doses, and the test results were used to determine the effectiveness of EB

radiation to bind treated fiberglass to the polyethylene matrix.

EXPERIMENTAL

To undertake decomposition experiments under electron beam, several samples of compounds **A** and **B** were deposited from $CHCl_3$ solution onto NaCl disks for FTIR analysis, and onto small watch glasses for NMR analysis. Each of these was exposed to varying doses of EB radiation. To determine whether decomposition of the diazoketone group in compounds **A** and **B** permits a bond to form with the surface of fiberglass, a 0.1% w/w sample was deposited onto the surface of 20-g samples of fiberglass, and treated with EB radiation in air and nitrogen atmosphere. A portion of the sample was slurried in hexane, and the amount of compound **A** or **B** that could be extracted from the surface of the fiberglass was measured by UV spectroscopy. The percentage recovered compound was plotted against the EB dose.

To undertake the preparation of treated fiberglass for extrusion blending, compounds **A** and **B** were deposited onto the surface of 200-g quantities of fiberglass (Fiberglass Canada 739DD, average length 0.8 mm, average filament diameter 15.8 μm) from solutions in $CHCl_3$. The standard application rate for both compounds was 0.1% w/w, approximately four times the calculated amount needed to form a monolayer, which allows for evaporation from the surface of the fiberglass during the heat treatment. The flasks containing the coated fiberglass were immersed into a heating bath and heated under nitrogen at 150°C for 45 min, with occasional shaking to facilitate heat transfer.

To undertake extrusion mixing, HDPE (Sclair 2710) was ground into a powder, and the treated fiberglass dry-mixed with the polyethylene at a 20% w/w level. This mixture was extruded and pelletized in a single-screw extruder with all zones adjusted to a temperature of 180°C. This temperature was chosen to be just high enough to process polyethylene, yet low enough to minimize the thermal reaction between treated fiberglass and polyethylene. Standard tensile and impact specimens were prepared in a reciprocating screw injection-molding machine, with all zones adjusted to 190°C. These were exposed to EB radiation of varying doses, and then tested for tensile

strength and elongation by ASTM D638M (type 1 specimen). Notched impact strength values were determined using a 2-lb. hammer according to ASTM D256 Type A with notch backward (reverse) for samples exposed to low EB doses [0–100 kiloGray (kGy)], and notch forward for samples exposed to high EB doses (100–400 kGy).

All samples for either laboratory analysis or mechanical testing were separated into trays corresponding to EB doses of 10, 50, 100, 200, and 400 kGy. A fast pass through the beam produced the 10-kGy dose. A slower speed produced the 50-kGy dose, and samples scheduled for 50, 100, 200, and 400 kGy were passed through the beam either 1, 2, 4, or 8 times to achieve their required dose. Between each 50-kGy dose the trays were allowed to cool before a subsequent pass through the electron beam. After the correct dose had been applied to each sample, the decomposition experiments were completed in the laboratory, and the molded specimens were tested for tensile and Izod impact properties.

RESULTS OF DECOMPOSITION STUDIES UNDER EB RADIATION

FTIR Results

Figures 1 and 2 show the IR spectra of compounds **A** and **B** after exposure to EB radiation, respectively, at doses of 10, 50, 100, 200, and 400 kGy. Decomposition of the diazoketone group in both cases is evidenced by a rapid reduction in the peaks at 3091 and 1645 cm^{-1}, corresponding to the C—H and C=O stretching frequencies of this group, respectively. (See peaks labeled X and Z in Fig. 1.) The new peaks that rise in the carbonyl region are the result of a new carbonyl species forming after breakdown of the diazoketone group. The decomposition of the azide group is observed at 2102 cm^{-1} (peak Y in Fig. 1). In compound **B** a peak at 2122 cm^{-1}, attributed to the N=N stretch in the diazoketone group, is reduced at the same time as the C—H and C=O stretches for this functional group. The residual absorption at 3080 cm^{-1} in compound **B** after exposure to 400 kGy is attributed to C—H stretching from the alkene group, which is not completely decomposed at this dose.

Figure 3 shows the absorbance values versus exposure for peaks X and Z in compound **A**. Cor-
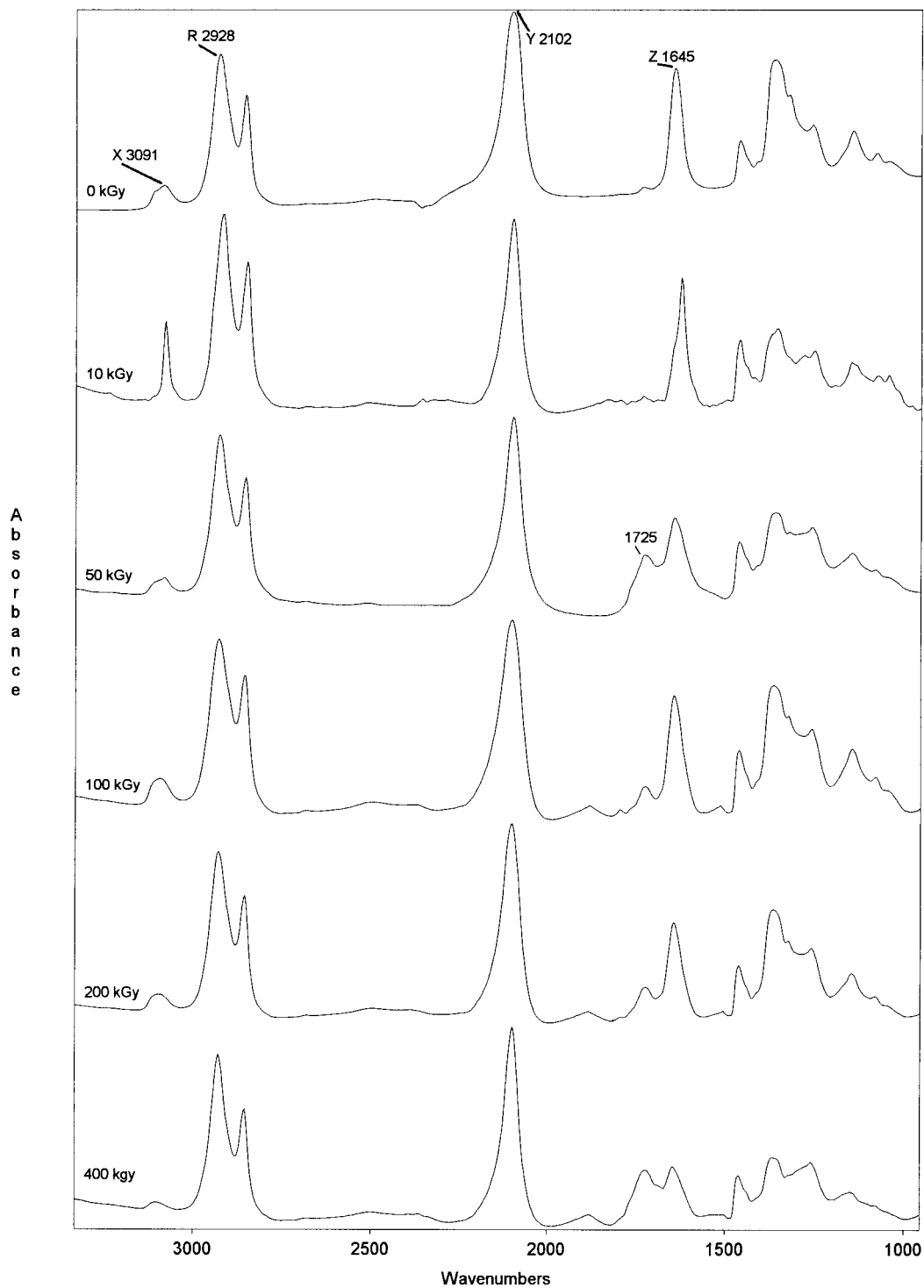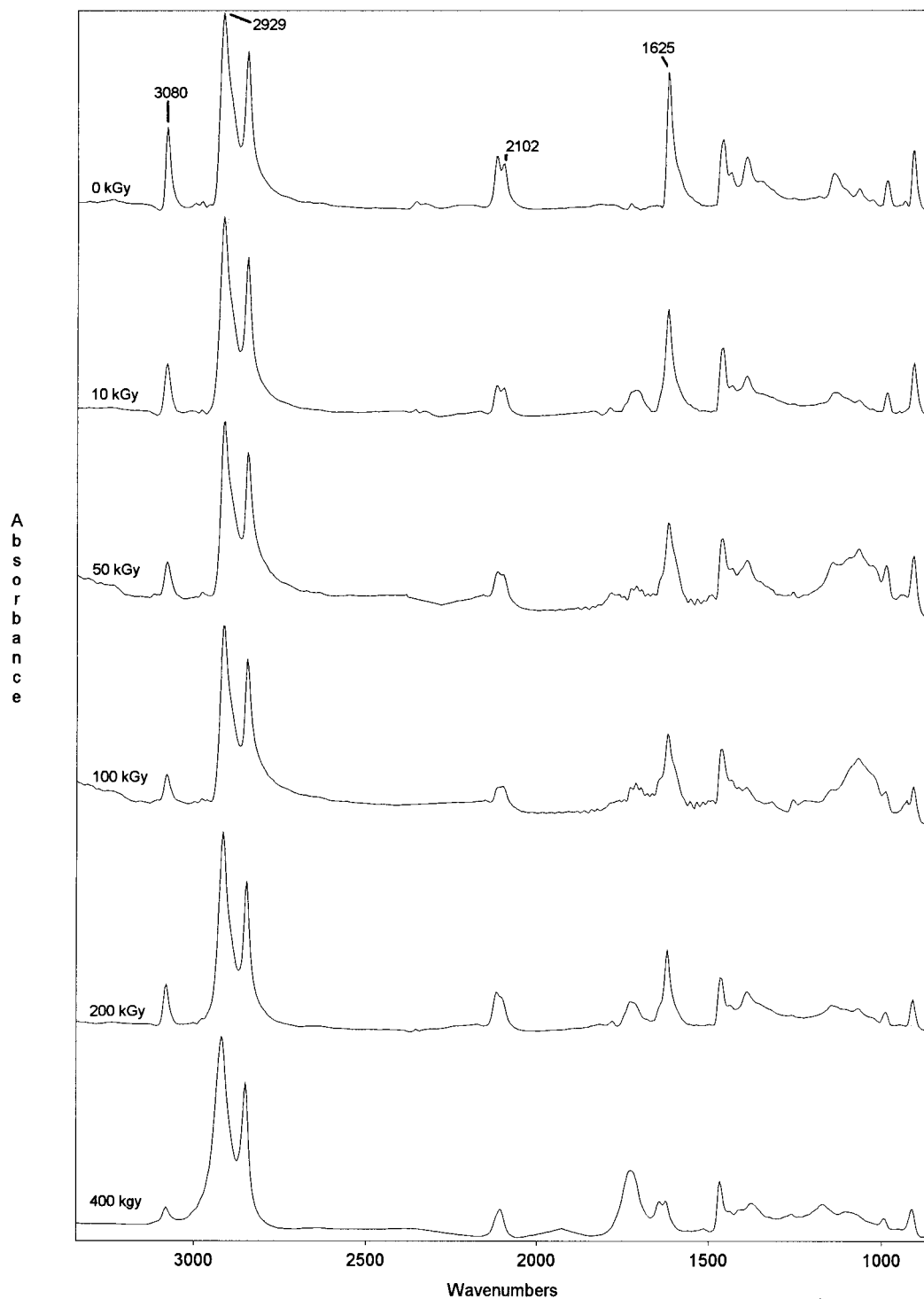
Figure 1   IR spectra of compound **A** after exposure to EB radiation from 0 to 400 kGy.

rection for differing amounts of sample on the IR disk was achieved by dividing each absorbance value by the methylene absorbance at 2928 cm^{-1} (labeled R), and then the curves were converted to the same scale for illustration purposes. The diazoketone group (3091 cm^{-1}) and the carbonyl group (1645 cm^{-1}) decompose during exposure to the electron beam, with steady

Figure 2   IR spectra of compound **B** after exposure to EB radiation from 0 to 400 kGy.

losses up to 400 kGy. However, decomposition is not complete at 400 kGy. Peak Y in **A**, consisting of an overlap of N=N absorbances from both the diazoketone and azide groups, does not appear to be affected by electron beam radiation. To determine whether oxygen or moisture has an effect on the decomposition of these groups under EB, these experiments were run
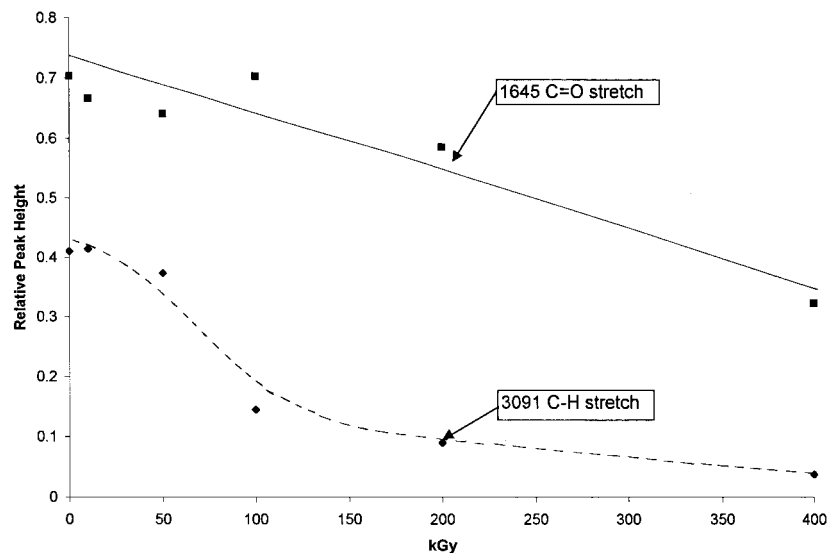
Figure 3 Absorbance values for diazoketone C—H (3091 cm^{-1}) and carbonyl (1645 cm^{-1}) functional groups in compound **A** after EB exposure from 0 to 400 kGy.

in both air and nitrogen. Very little difference was observed in the results.

NMR Results

Figures 4 and 5 show the NMR spectra for the decomposition of **A** and **B** under electron beam. For compound **A** (Fig. 4) the resonances at 3.25 and 5.20 ppm correspond to the hydrogens α to the azide group and the C—H hydrogen of the diazoketone, respectively. Loss of amplitude in these peaks is thus related to the decomposition of the azide and diazoketone groups. Normalization was undertaken by comparison to the hydrogens positioned β to the carbonyl group ($\delta = 1.60$), which are unaffected by chemical changes that take place in the diazoketone group during decomposition. For compound **B** (Fig. 5), the key resonances are at 5.20 ppm for the hydrogens α to the diazoketone group, and 4.95 and 5.80 ppm for the alkene hydrogens. Figure 6 indicates a plot of normalized peak areas versus time for the diazoketone, azide, and alkene groups taken from Figures 4 and 5. There is indication that the diazoketone group decomposes with EB radiation, but its decomposition is not complete with a 400-kGy dose. There appears to be essentially no decomposition of the azide group during exposure to electron beam. The alkene group decomposes with radiation, but more than half of the functional group is present after a 400-kGy dose. For

both compounds **A** and **B**, the results under air are very similar to those obtained under nitrogen.

Percentage Recovery from Fiberglass After EB Treatment

As samples of fiberglass coated with compounds **A** and **B** were exposed to electron beam, the recovery of unreacted compound in each case was reduced with increased time of exposure. Figure 7 summarizes the results of these experiments. The results show that the percentage recovery versus EB dose is relatively close for both compounds. Separate experiments in air and nitrogen indicate that the results are not affected significantly by the choice of atmosphere. In all cases, the percentage recovery is close to 50% after 200 kGy and 25% after 400 kGy. The results parallel the results of the decomposition studies on compounds **A** and **B**. Thus, it can be assumed that the decomposition of the diazoketone group in these compounds facilitates the attachment of the coupling agents to the fiberglass surface. Because percentage recovery values drop somewhat faster than the diazoketone peaks in the decomposition experiments, it also appears that spreading the compounds onto a glass surface facilitates decomposition under EB radiation. Nevertheless, extracted samples of **A** and **B** from EB-exposed fiberglass surfaces showed partial decomposition by IR and UV, indicating that the entire decom-
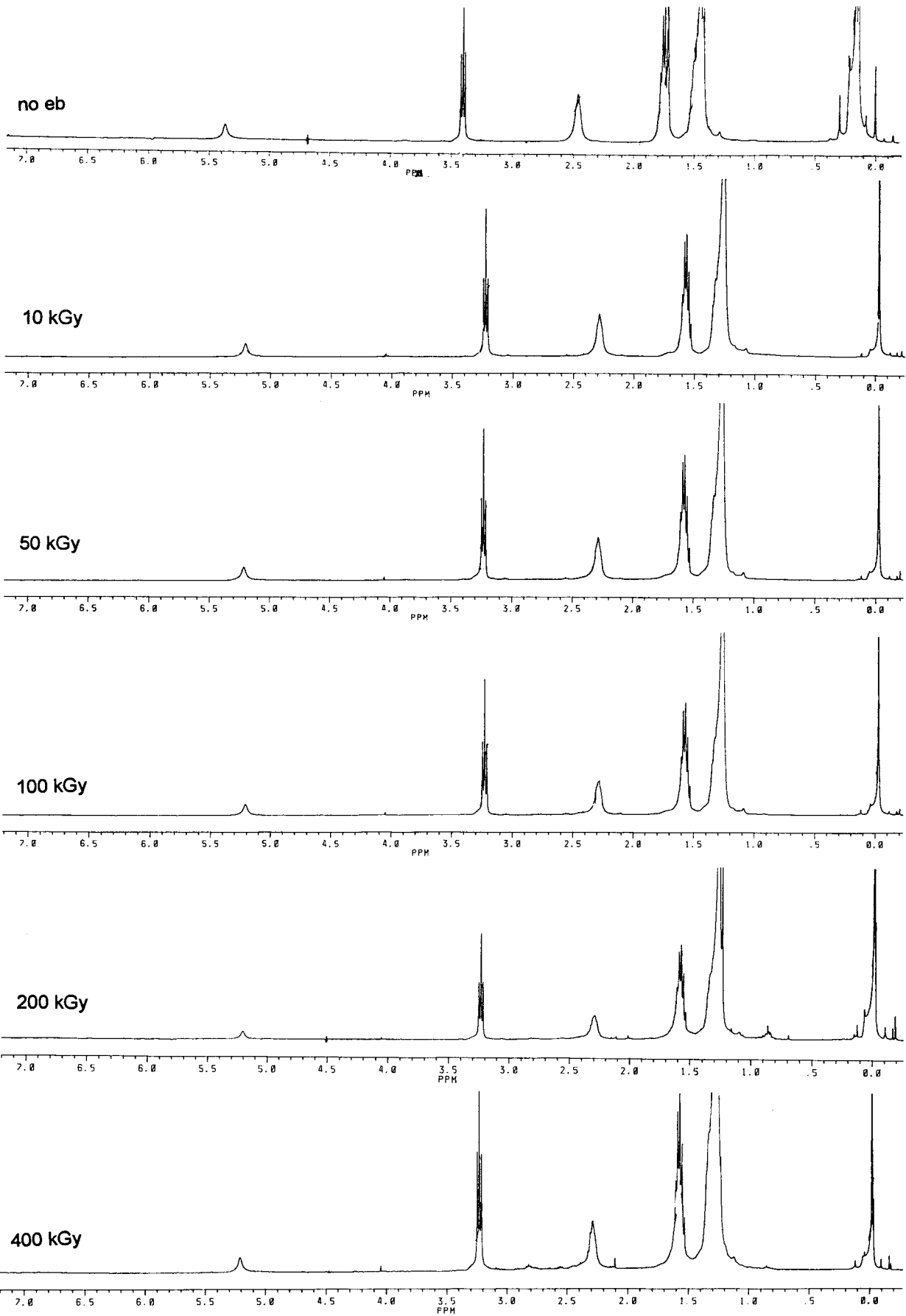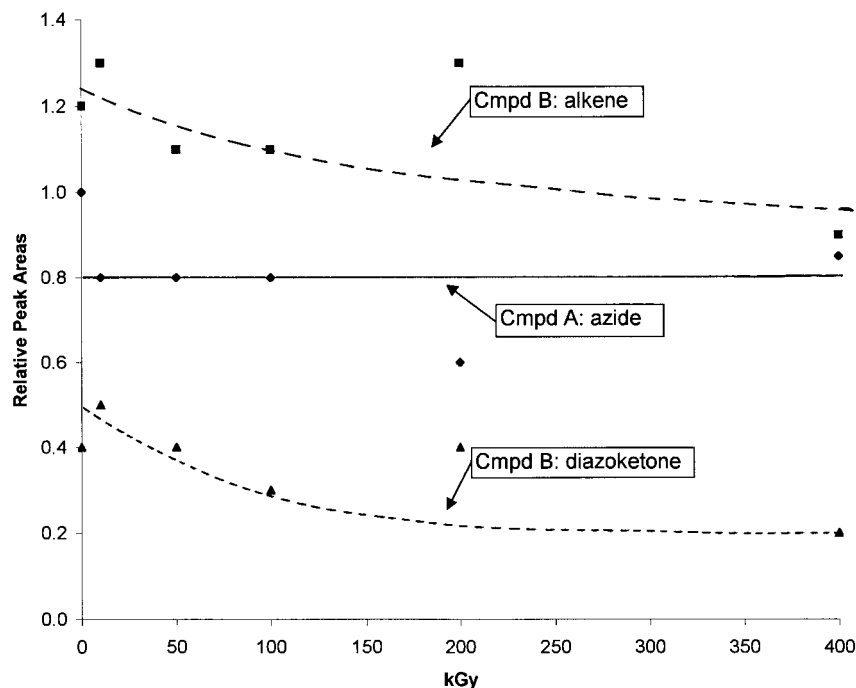
Figure 4 NMR spectra of compound **A** after exposure to EB radiation from 0 to 400 kGy.
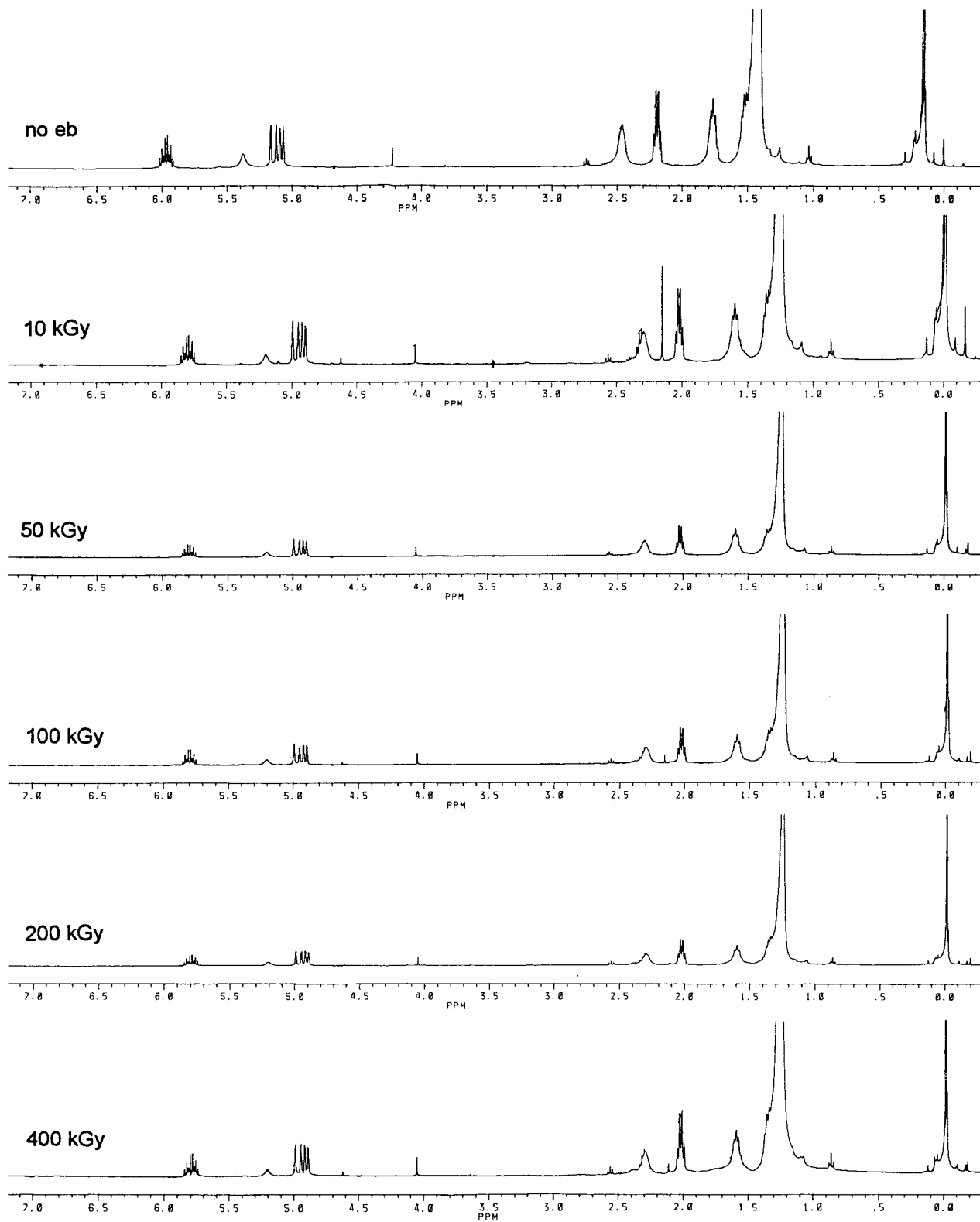
Figure 5   NMR spectra of compound **B** after exposure to EB radiation from 0 to 400 kGy.

Figure 6 Relative peak areas versus EB exposure, indicating the relative decomposition rates for diazoketone, alkene, and azide functional groups taken from NMR studies in Figures 4 and 5.

posed sample is not immediately bonded to the surface of the fiberglass.

It should be noted that EB treatment is both more difficult and less efficient than a heat treatment at bonding these coupling agents to fiberglass. In 10 min of heating at 150°C the decomposition reaction of the diazoketone group, and the subsequent reaction with the glass surface, is virtually complete.[5] Thus, heat is the preferred treatment to bind these coupling agents to a glass surface.

EFFECTS ON THE MECHANICAL PROPERTIES OF ADDING FIBERGLASS AND COUPLING AGENT TO POLYETHYLENE

As untreated fiberglass is added to polyethylene, one expects a rise in tensile modulus, a drop in elongation at yield, and a reduction in flexibility (measured by Izod impact) with the addition of this harder and chemically dissimilar material. Yield strength should also decrease, because the two phases do not adhere, making it impossible for the inherent strength of the fiberglass phase

to be imparted to the overall compound. If EB radiation is applied to a polyethylene/fiberglass composite that has no coupling between the phases, the samples should become more tough and rigid, reflecting the crosslinking that takes place in the polyethylene phase resulting from radiation. This will result in higher values of tensile modulus, yield strength, and Izod impact strength. If coupling takes place between the fiberglass and polyethylene phases in the absence of EB radiation, there should be an increase in the yield strength and tensile modulus, and a decrease in the impact strength, as the composite acquires more of the characteristics of the rigid glass phase.

Overall, a fiberglass/polyethylene composite with compound **A** or **B** reacted onto the surface of the fiberglass and exposed to EB radiation may yield a combination of all three observable effects noted above: the addition of fiberglass to the polyethylene polymer, crosslinking in the plastic matrix, and coupling at the plastic/fiberglass interface. These expected effects are summarized in Table I and are used to assess the effectiveness of the coupling agents in the study.
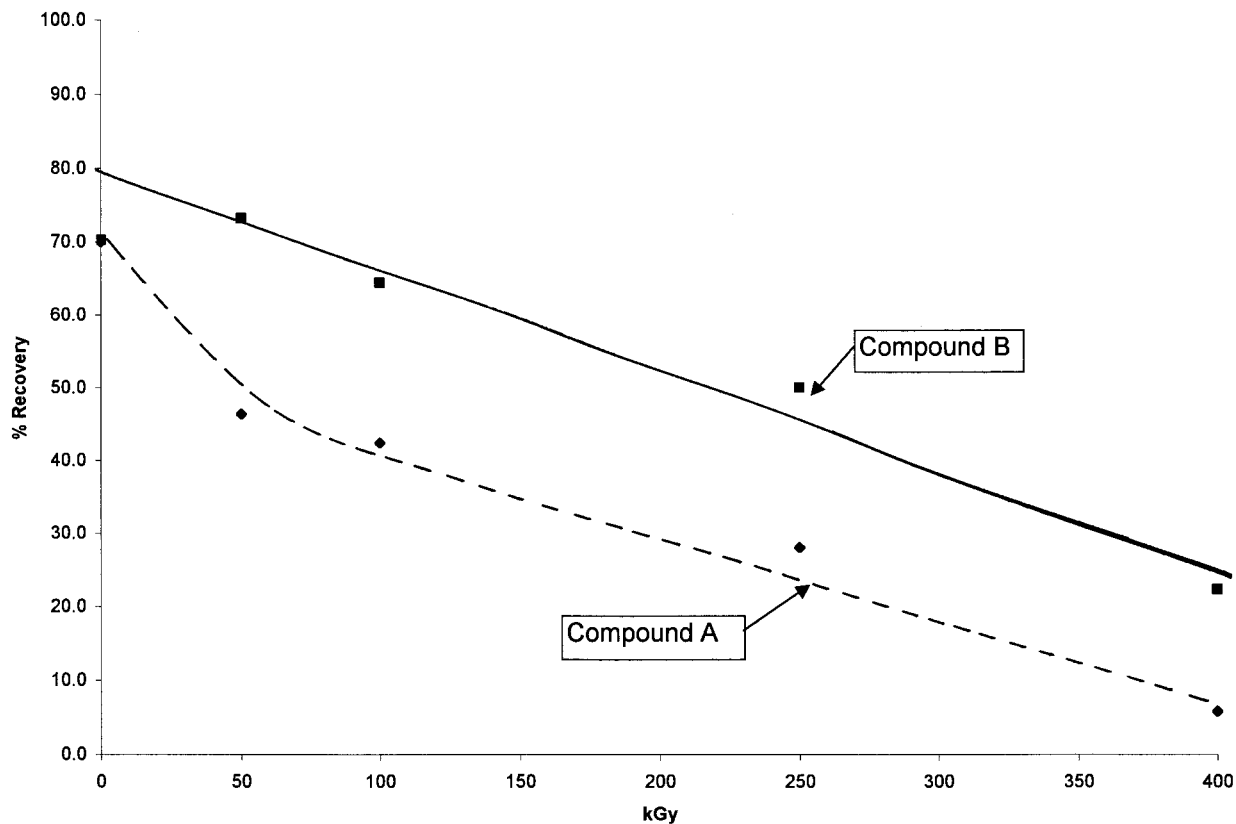
Figure 7   Percentage recovery of compounds **A** and **B** from coated samples of fiberglass after EB exposure.

RESULTS OF EXTRUSION STUDIES

To undertake the testing of polyethylene/fiberglass composites, compounds **A** or **B** were coated onto samples of dried fiberglass (0.1% w/w based on fiberglass) and treated by heat at 150°C. The glass samples were extrusion-mixed with pow-dered polyethylene, molded into test articles, and then passed through the electron beam. Table II lists the results of the tensile and impact testing for all the samples produced by this method. These include samples of HDPE with treated fiberglass made from either compound **A** or **B**, along with reference composites consisting of

Table I   Addition of Untreated and Treated Fiberglass to Polyethylene: Expected Effects on the Mechanical Properties with EB Radiation

Addition of Untreated Fiberglass to Polyethylene (No Coupling, No EB Radiation)	Addition of EB Radiation to HDPE and Uncoupled HDPE/ Fiberglass Composites (No Coupling Between Phases)	Addition of Coupling Agent to the Interface of the Two Phases in HDPE/Fiberglass Composites (No EB Radiation)
1. Yield strength decreased	1. Yield strength increased	1. Yield strength increased
2. Tensile modulus increased	2. Tensile modulus increased	2. Tensile modulus increased
	3. Elongation at yield relatively unchanged	
3. Elongation at yield decreased		3. Elongation at yield decreased
4. Notched impact strength decreased	4. Notched impact strength increased	4. Notched impact strength decreased

Table II   Mechanical Properties of Untreated and Treated Fiberglass in HDPE with EB Radiation[a,b]

Treatment	EB Dosage (kGy)	Yield Strength (MPa)[c]	Tensile Modulus (MPa)[d]	Impact Strength Reverse Notch (J/m)[e]	Impact Strength Forward Notch (J/m)[f]	Elongation at Yield (%)[g]	Elongation at Break (%)
Control							
(no fiberglass)	0	19.9	156	NB[h]		26.1	201
	10	20.4	169	NB		26.1	226
	50	20.6	181	NB		25.5	405
	100	20.7	164	NB	79	27.4	568
	200	21.9	191		145	26.4	509
	400	22.0	216		NB	27.1	217
Untreated							
fiberglass	0	18.5	235	397		21.0	77
	10	18.2	214	405		21.5	83
	50	18.3	234	462		20.9	59
	100	19.3	211	685	79	21.1	53
	200	19.5	204		101	22.6	62
	400	19.7	199		211	22.8	76
Treated with **A**	0	18.3	201	411		21.8	83
	10	18.4	226	376		22.5	76
	50	18.8	189	385		22.4	77
	100	19.6	196	500	76	21.6	62
	200	20.1	247		88	20.5	56
	400	20.7	218		184	20.8	60
Treated with **B**	0	17.9	171	428		24.6	98
	10	17.9	207	421		24.0	76
	50	18.4	170	511		24.0	83
	100	19.5	221	684	77	21.6	59
	200	20.0	251		82	20.5	60
	400	22.0	245		147	18.1	54

[a] Coupling agents **A** (12-azido-1-diazo-2-dodecanone) and **B** (1-diazo-17-octadecene-2-one) applied to Fiberglass Canada 739DD glass fibers. Standard application rate of 0.1% w/w based on fiberglass. Coated fiberglass was heat-treated at 150°C in an open vessel under N_2 blanket.

[b] Treated fiberglass dry-mixed with HDPE at 20% w/w level and extruded in a single-screw extruder with zones adjusted to 180, 180, 180, and 180°C. Injection-molded with zones adjusted to 190, 190, 190, and 50% (Nozzle).

[c] SE = ±0.1 MPa.

[d] SE = ±12 MPa.

[e] Impact strength for these samples measured with notch backward. SE = ±32 J/m.

[f] Impact strength for these samples measured with notch forward. SE = ±8 J/m.

[g] SE = ±0.8%.

[h] No break.

pure HDPE and HDPE with untreated fiberglass. The specimens containing the treated fiberglass will be affected both by crosslinking in the plastic matrix and by coupling at the plastic/fiberglass interface, if such takes place. Thus as the EB dose increases, the changes in yield strength, tensile modulus, elongation, and impact strength in these samples need to be seen in comparison to samples containing no coupling agent. For example, elevations in impact strength resulting from crosslinking should be reduced if coupling takes place between the rigid fiberglass and the more ductile polyethylene phase.

Based on the preceding comments and by reference to Table I, a number of observations can be made on the mechanical testing data shown in Table II.

1. EB generally causes an increase in yield strength. When fiberglass is present in the composite, the yield strength is reduced;
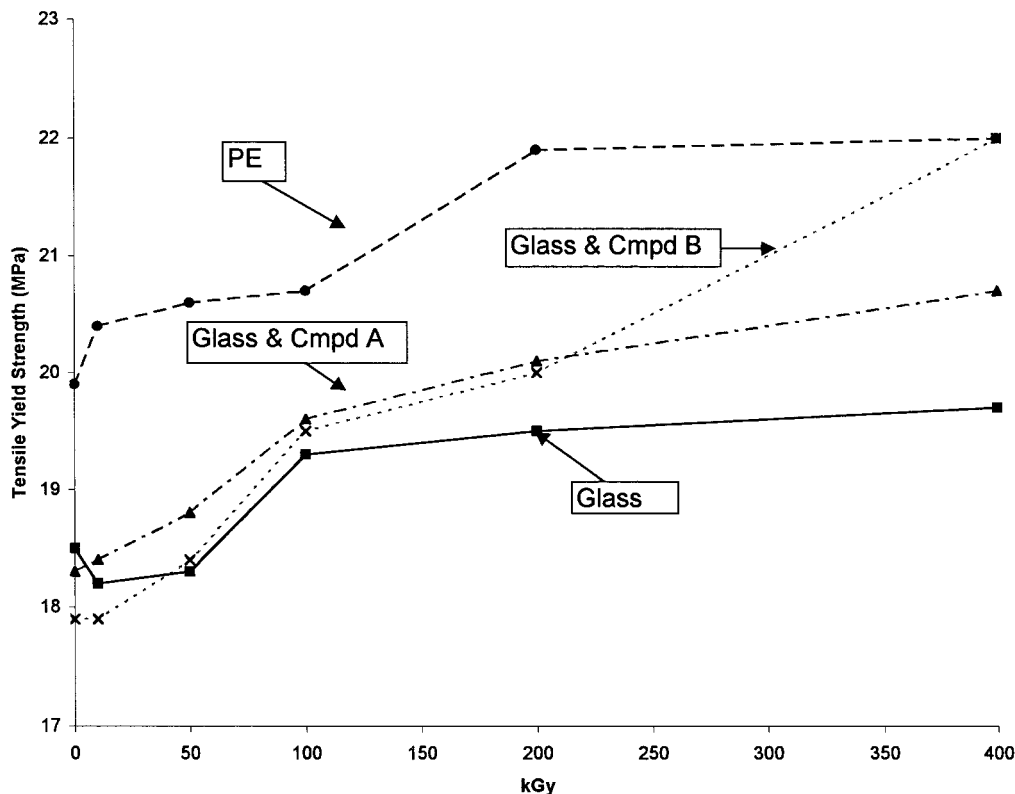
Figure 8   Effect of EB exposure on yield strength for polyethylene (PE), polyethylene/untreated fiberglass (glass), and polyethylene/treated fiberglass (glass and **A** or **B**).

however, when either compound **A** or **B** is present in the system, the yield strength values are higher than those for samples without coupling agent. At high EB dose levels the yield strength in composites containing compound **B** recover to similar levels seen for polyethylene alone (Fig. 8).

2. With the exception of untreated fiberglass/polyethylene specimens, there is a moderate, although somewhat irregular, increase in tensile modulus with EB dose (Fig. 9). Both **A** and **B** increase the value of tensile modulus and produce higher values than those for untreated fiberglass. Although compound **B** tends to produce higher values than compound **A**, both appear to act as coupling agents in these composites.

3. Elongation at yield values are generally decreased by the addition of fiberglass to polyethylene. Adding EB radiation to either polyethylene or a composite containing untreated glass has very little effect on this property. With either compound **A** or **B** present, there is a steady decrease in

values of this property with increasing EB radiation, suggesting that coupling is taking place at the interface of the two phases with EB radiation (Fig. 10). When experimental error is taken into account, both **A** and **B** have similar effects up to 200 kGy, but compound **B** produces the lowest values of this parameter at 400 kGy. A similar effect is seen with elongation at break values, although values of this parameter are generally less reliable than elongation at yield.

4. Izod impact strength values increase dramatically with EB dose, which is reflected in the values of impact strength for all samples. Impact values were measured with notch forward at high EB doses, whereas they were reversed for low-dose specimens to provide higher values and greater sensitivity. By observing the way in which either **A** or **B** is able to reduce the impact values compared to that of untreated fiberglass, it can be seen that compound **B** has no coupling effect in these
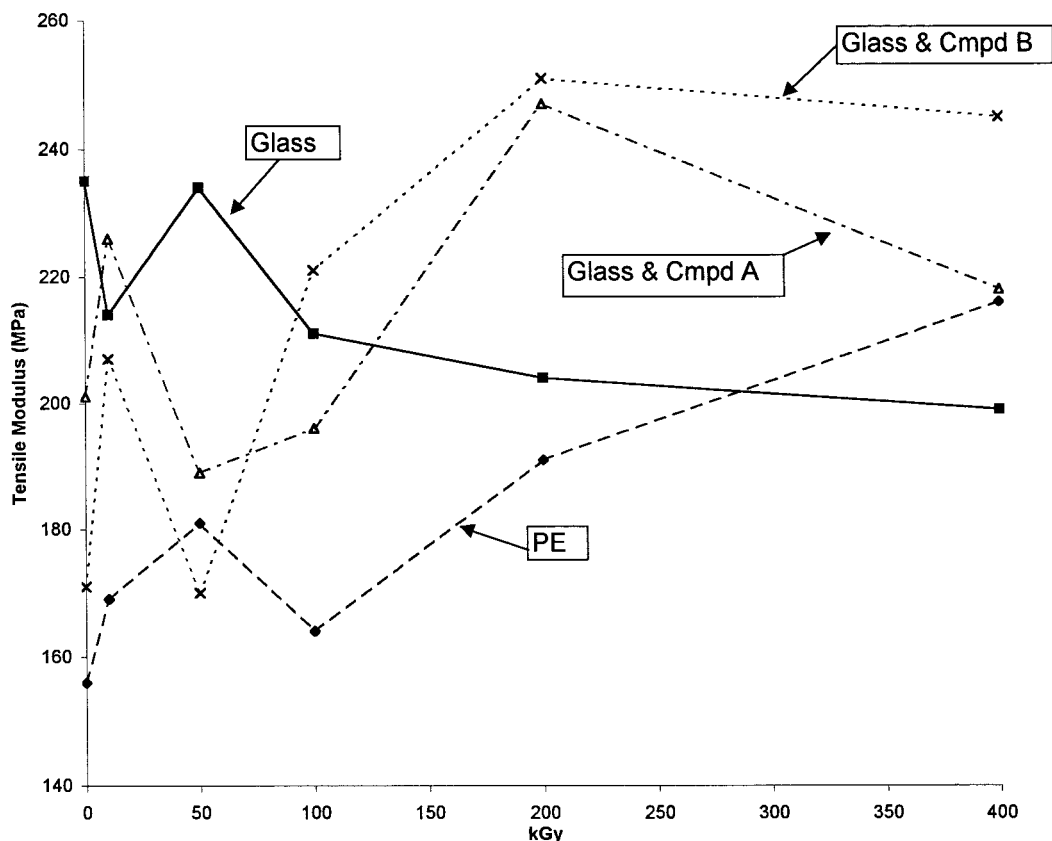
Figure 9 Effect of EB exposure on tensile modulus.

composites at low EB doses (≤100 kGy), whereas compound **A** has a moderate coupling effect (Fig. 11). At high EB doses (>100 kGy) both compounds act as coupling agents, but compound **B** is more effective than **A** (Fig. 12).

This study shows that both compounds **A** and **B** are able to act as coupling agents in fiberglass/polyethylene composites produced by the method used in this study, consisting of a heat treatment to bind **A** or **B** to the fiberglass phase, and EB radiation to bind the treated fiberglass with the plastic matrix. Free radicals produced in the polyethylene matrix by EB radiation are thus able to trigger a reaction with both compounds attached to the surface of fiberglass. The coupling effect is generally more pronounced for compound **B** than for **A**, especially at higher EB doses. This is not surprising, given that an alkene group is able to react with free radicals produced during the EB treatment. Higher doses (>100 kGy) are necessary to trigger the reaction with compound **B**,

whereas relatively low doses (<100 kGy) can trigger a coupling reaction in compound **A**.

Some comparisons to a heat treatment are in order at this point. When heat is used to bind compound **A** to fiberglass, and also used to trigger the reaction between the coupling agents and the plastic matrix, there is an increase in tensile properties of approximately 20% and a reduction in Izod impact of approximately 35%. In comparison, EB radiation on compound **B** is able to achieve a 23% increase in tensile modulus and a 20% reduction in impact at 200 kGy, whereas at 400 kGy there is a 23% increase in tensile modulus and a 30% reduction in Izod impact.

It should be noted that these coupling agents do not show any effect by EB radiation until the dose level reaches at least 50 kGy. This observation also confirms that little coupling to the plastic matrix of **A** or **B** has taken place during the low-temperature extrusion and molding steps to mix the treated fiberglass and polyethylene. At the higher doses used to initiate coupling between
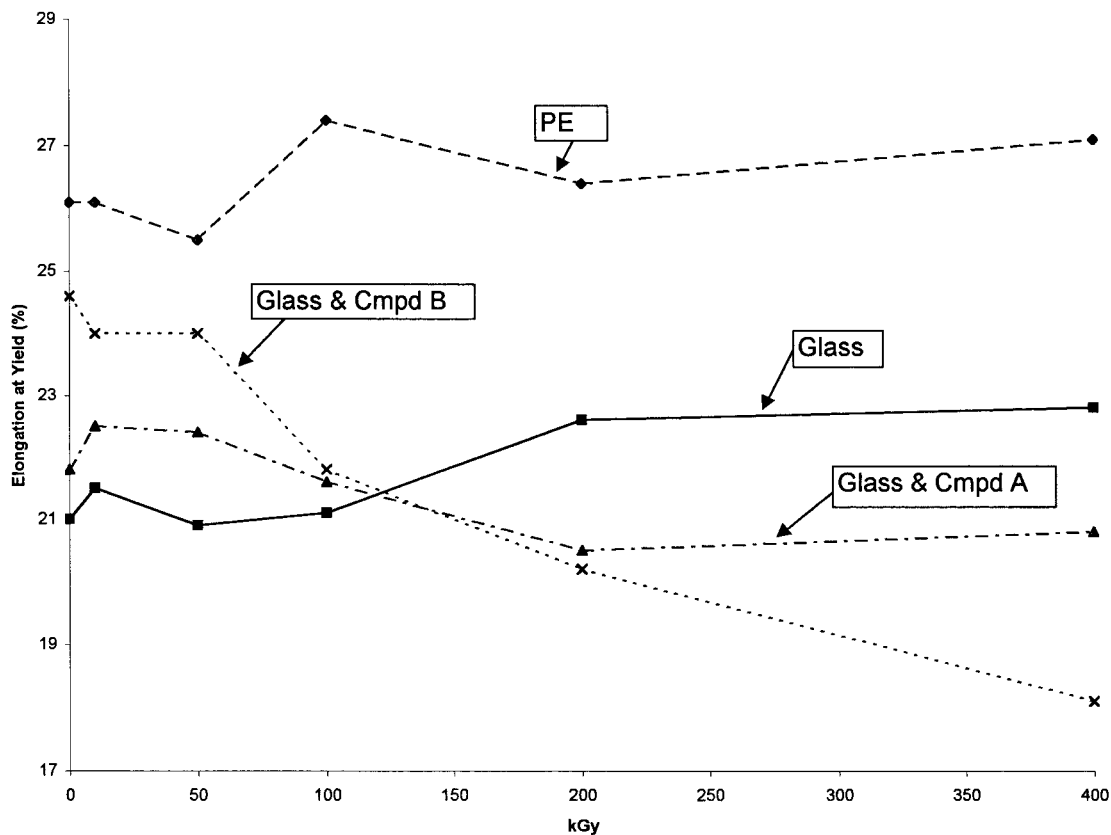
Figure 10   Effect of EB exposure on elongation at yield.

DISCUSSION

It has now been shown that electron beam radiation is not only able to decompose the diazoketone and alkene groups in compounds **A** and **B**, but also able to couple these molecules to fiberglass and to polyethylene. Compound **A** is more effective in coupling to polyethylene at low doses and compound **B** is more effective at high doses. At high doses the coupling of compound **B** to the plastic matrix is about equivalent to the best results obtained for compound **A** under thermal coupling. Although the preparation of these composites with EB radiation involves one more step than thermal coupling, and thus is an inherently more difficult and costly treatment, it may have

some advantages. For some applications involving polyethylene, an EB treatment may produce crosslinking along with coupling to a treated fiberglass, thus providing additional rigidity and toughness in the composite. This may be useful in certain situations.

Because a heat treatment is so efficient at coupling **A** and **B** to fiberglass, it is generally recommended to use this method to prepare treated samples of fiberglass.

CONCLUSIONS

Electron beam radiation is able to initiate coupling between fiberglass and two bifunctional compounds, 12-azido-1-diazo-2-dodecanone (**A**) and 1-diazo-17-octadecene-2-one (**B**). Fiberglass treated with either **A** or **B**, mixed by extrusion with polyethylene, and then exposed to EB radiation produces composites that exhibit coupling between the treated fiberglass phase and polyethylene. This allows for coupling between two un-
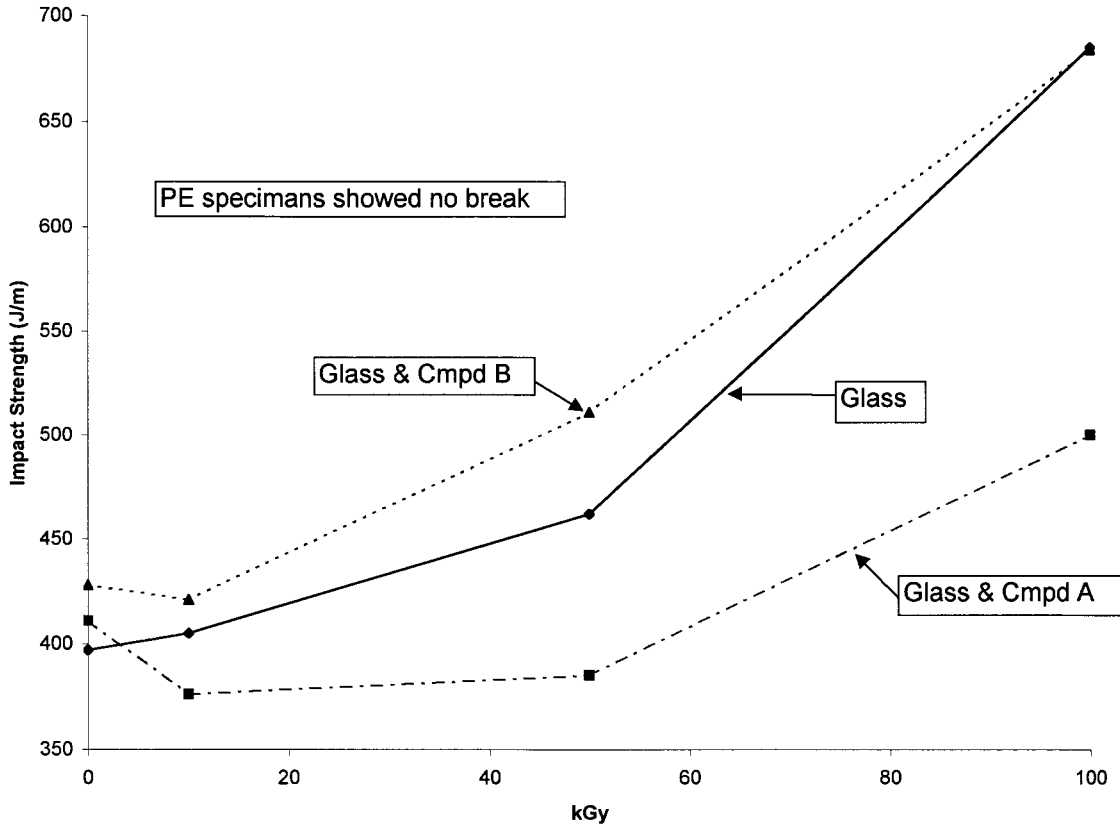
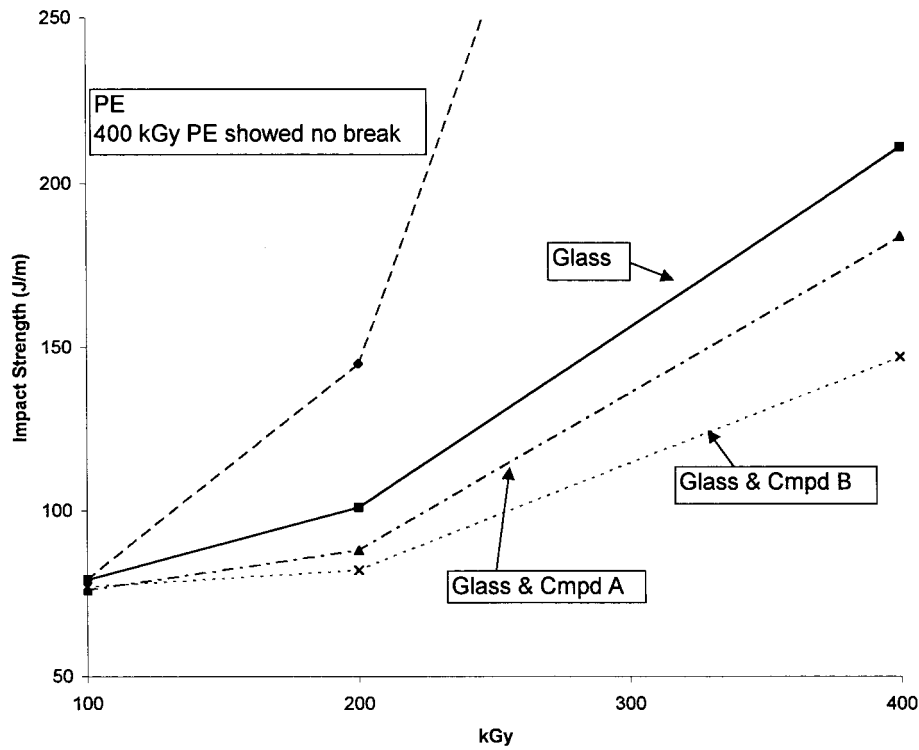**Figure 11**   Effect of EB exposure (0–100 kGy) on Izod impact strength (reverse notch).

Figure 12   Effect of EB exposure (100–400 kGy) on Izod impact strength (forward notch).

like phases. A number of fiberglass samples were heat-treated with compounds **A** and **B**, followed by extrusion mixing with polyethylene and exposure to EB radiation. Tensile and impact values in samples containing **B** showed the best overall coupling ability with the polyethylene matrix, providing a change of 23% in tensile properties and 30% in impact properties. However, these composites required a 400-kGy dose to bring about substantial coupling.

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