

Preparation and Characterization of Poly(ethylene terephthalate) Powder-Filled High-Density Polyethylene in the Presence of Silane Coupling Agents

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ABSTRACT: Micron-size crystalline particles of Poly(ethylene terephthalate) (PET), obtained from PET bottles by crystallization and grinding, were used as a filler in high-density polyethylene (HDPE). The composite of PET particle-filled HDPE was prepared by melt mixing at 190°C, which was well below the melting temperature of PET. Silane coupling agents (SCAs) were used to enhance the interaction between PET and HDPE in the composite. A chain extender (CE) and maleic anhydride (MA) were also used to provide further interaction with SCAs between these two materials. The ultimate tensile strength, especially at highest content 40% PET-filled HDPE, and the impact strength of SCAs-treated PET-filled HDPE was found to be highly improved compared to untreated PET filling into HDPE. Dynamic mechanical analyses (DMA) demonstrated that T_g of the main matrix polyethylene was depressed from 3 to 10°C. Scanning electron microscopy (SEM) studies indicated a strong interaction between PET powder and HDPE when SCAs were present in the system. © 2000 John Wiley & Sons, Inc. *J Appl Polym Sci* 79: 827–835, 2001

Key words: poly(ethylene terephthalate) powder; PET powder-filled HDPE; silane coupling agents

INTRODUCTION

Concerns for the environment and the limited sources of the earth bring about the need for more efficient and conservative use of resources. Among the solid wastes, plastics occupy a special place because of their unique advantages such as easy collection, cleaning, handling, and high economic benefits as reviewed in many books and articles.^{1–4} Poly(ethylene terephthalate) (PET) resin has been used in textile industry, Dupont's

Dacron, and ICI's Terylene, since the late 1950s. After introduction of PET tire fabric by Goodyear in 1962, polyesters of the PET family were developed specifically for packaging, film, sheet, coatings during the late 60s, and soft drink bottles after 1978.^{1,2} PET recycling is economically viable because some significant price difference between virgin PET resin and postconsumer recycled PET in addition to high-quality availability of the PET bottle waste stream.^{1,3,4}

Recycling options for PET can be divided into three categories: chemical recycling (depolymerization), mechanical recycling, and multilayer forming. Recycling of PET based on depolymerization of the polymer was well known and fundamentally established in the last decade. Recycling through multilayer processing and also co-extrusion is, on the other hand, one of the recent

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technologies of recovering of PET. The process involves multilayer preforms obtained by injection molding of three to five layers with an inner layer of recycled PET.^{3,4} The mechanical processing in PET becomes important because it is less expensive than the chemical process and also adequate even for monolayered containers in direct food contact.^{3,4} Besides these recycling methods, the efforts to develop PET-based blends and composites have been recently intensified as a result of the availability of PET as a postconsumer waste.

PET–Polyolefin blends are the preferred systems because of lower cost and also the resistance of polyolefins to high processing temperatures needed for PET (230–270°C) under suitable conditions. However, one of the most important restrictions in these blends is that they are incompatible, and exhibit extremely poor mechanical properties. The studies on the PET–Polyolefin blends were, therefore, focused on the techniques to improve the compatibility between them. The first successful compatibilizers used in PET–Polyethylene blend were hydrogenated styrene–butadiene–styrene (SBS) block copolymer, applied by Traugott, Barlow, and Paul in 1983.⁵ Maleic anhydride grafted PE (Pe-g-MA),^{6–8} sodium ionomer of poly(ethylene-co-methacrylic acid),⁹ ethylene–glycidyl methacrylate copolymer (EGMA),^{10,11} poly(ethylene-co-acrylic acid) (PEAA),¹² styrene ethylene/propylene(S-PE) b-diblock copolymer,¹³ maleated PP,¹⁴ and acrylic acid grafted PP^{15,16} are examples of the compatibilizers used in the PET–Polyolefin blends. Besides the PET–Polyolefin blends, recycling and blending of plasma glow discharge modified PET with PVC in solvent,¹⁷ PET powder with PVC in melt compression,¹⁸ a tetra functional epoxy resin as a compatibilizer in PET/polyphenylene ether (PPE) blends¹⁹ have appeared in the literature. The results of these studies suggested that the enhancement in mechanical properties (especially in tensile and impact strength) accompanied by a better dispersion and the ease of processing were the benefits of the compatibilization. In one of the recent studies, a composite was suggested from commingled PET, HDPE, and newspaper fibers.²⁰ The incompatibility of each component and the poor adhesion between them did not produce a high-quality material.

In this study, amorphous PET, after crystallization at high temperature, was ground into a powder of almost homogeneous size 90–100 μm and introduced as filler in an HDPE matrix by melt

mixing. The weak interaction filler PET particles with HDPE was still present in the composition studied, and this occasionally caused weak mechanical properties. The application of silane coupling agents (SCAs) to the PET particles, despite their general utilization in glassy or mineral fillers to enhance the interaction between polymer and filler, was found to be effective in improving the mechanical properties of these PET-filled HDPE composites. Further possible improvement in some of the studied properties of the composites were observed when maleic anhydride and a chain extender were coupled with silane coupling agents.

EXPERIMENTAL

The granular HDPE (S0464) was supplied from Turkish Petrochemical Industry (PETKIM), and it had melt flow index of 0.35 g/10 min and density of 0.964 g/cm³ with a weight average molecular weight of 124,000. PET soft drink bottles produced by the SASA Co. (Sabanci Holding Company in Turkey) were collected in the campus area of the university. No further attempt was carried out for the characterization of these bottles. They were washed with tap water and dried in ambient atmosphere.

The dried PET bottles were cut into small flakes (approximately 1 \times 1 cm), and were put into an oven of 250°C for 10 min. Then, they were removed to room temperature for a complete crystallization. The total time of cooling took about 15 min. The products were white opaque PET flakes with high brittleness. These crystallized PET flakes (melting point of 249°C measured in DSC, TA 910S) were ground mechanically and sieved powders of about 90–100 μm size, which were used as filler for HDPE. The particle size distribution of PET powders obtained from particle size analyzer, Mastersizer S, is provided in Figure 1.

Silane coupling agents were the products of HÜLS (formerly Union Carbide) and their types (with old and new naming) and chemical names are given in Table I. Besides the hydrolyzable methoxy groups in SCAs, A-172 and A-174 contained vinyl polymerizable reactive sites while A-187 had an epoxy ring reactive site. The reactive groups, therefore, were also expected to play an important role in enhancing the interaction between PET powder and HDPE. The chain extender (CE) used in addition to SCAs was a linear alkane, the product of Amoco Chemicals Europe,

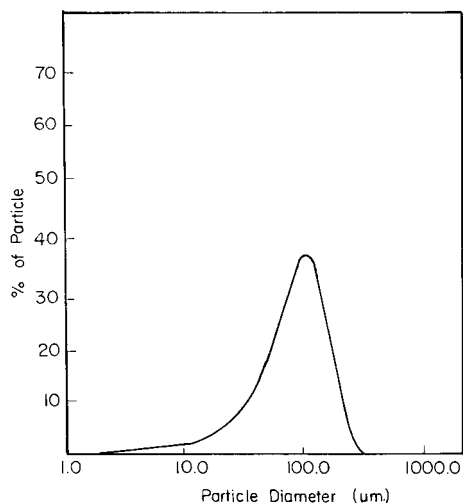


Figure 1 Particle size distribution of PET powder, as measured with respect to volume.

with a molecular weight of 1900 g/mol and contained two end —OH groups. Maleic anhydride (MA) was supplied from Merck Co.

SCAs of type A-187 and A-174 (as given in Table I) were applied to PET powder separately from their diethylether solutions. SCAs content in an ether solution was adjusted to have 2% by weight with respect to PET. A mixture of equal amounts of a vinyl silane (A-172) with A-174 was also applied onto the surface of PET powders with a final SCAs content to be 2%. The same procedure was followed in MA and CE treatments on the SCAs-treated PET powders where MA or CE content was kept 2% by weight. The ether was then removed completely in an oven at 45°C in 8 h.

Compositions of 5, 10, 15, and 40% by weight of surface-treated and untreated PET and HDPE composites were prepared in a Brabender mixing head (W30H, Brabender Plasticorder Torque Rheometer PLV-15) at 190°C and 60 rpm mixing speed for 15 min. Compression-molded sheets of 2 mm thick were prepared from these well-mixed composites between steel plates in hot platen press at 200°C. Molding continued under a pres-

sure of 20,000 psi for a minute, and the steel mold was quenched into cold water. A special steel mold with cavity dimensions of $0.75 \times 0.4 \times 6.0$ cm was used to prepare specimens for impact tests under the same conditions.

Pure HDPE was also passed through identical mixing and molding conditions to ensure an identical thermal history with the composites.

Tensile tests were carried out on standard dumbbell-shaped specimens cut from 2 mm-thick sheets by an Instron Tensile Testing Machine (TM 1102). Testing was conducted at 5.0 cm/min draw rate with a gauge length of 3.5 cm to evaluate the ultimate properties. Plastic Impact Machine of type H20 was used for the Charpy impact measurements. All mechanical tests were performed at ambient conditions, and the results reported were the average of five, at least, samples tested. The results of impact testing were given in terms kN/m (J/m^2) due to slight changes in thickness in the compression mold samples, where the surfaces of the samples were then polished before testing.

A TA Instruments 983 DMA was used to follow the thermal behavior of the composites from -140 to 110°C with a scan rate of $10^\circ\text{C}/\text{min}$ at the resonant mode (sweeping from 2 to 85 Hz) and with the oscillation amplitude of 0.2 mm. The sample size in these measurements were 10 mm wide, 50 mm long and 2 mm thick.

Fractured surface analysis of both tensile and impact test samples were made at various magnifications by using scanning electron microscopy (SEM), JEOL- JSM-6400A, after a protective coating was applied.

RESULTS AND DISCUSSION

Mechanical Properties and Dynamic Analyses

Fine powder of PET obtained by recrystallization followed by mechanical grinding was uniformly dispersed in HDPE matrix in melt mixing. Note that the processing temperature in mixing and

Table I Types of Silane Coupling Agents

Name of Silane Coupling Agent	Union Carbide	HÜLS Name
Gamma-glycidoxypropyltrimethoxysilane	A-187	Dynasylan-GYLMO
Vinyl-tris-(beta-methoxyethoxy)silane	A-172	Dynasylan-VTMOEO
Gamma-methacryloxypropyltrimethoxysilane	A-174	Dynasylan-MEMO

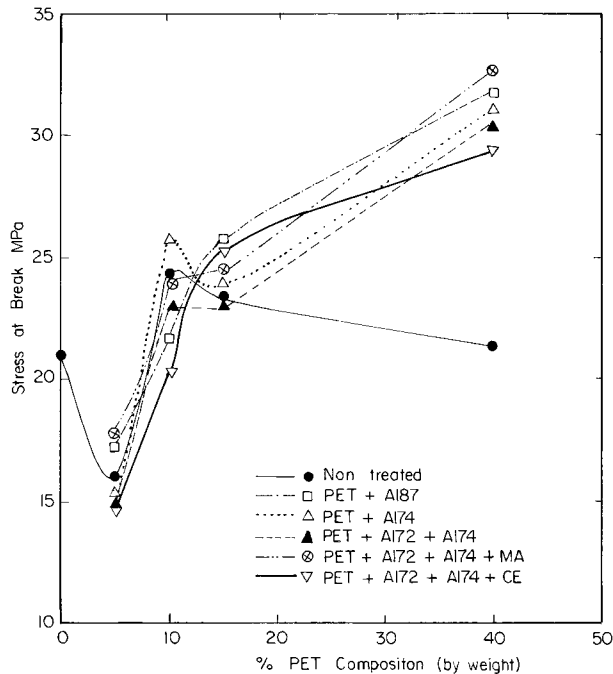


Figure 2 Variation of ultimate tensile strength with respect to PET filler concentration. (The standard deviation in tensile testing was found to be less than 10% of the mean values of tensile strength and elongation.)

compression molded was remained well below the melting point of PET particles in HDPE. As in all filler added polymers, the addition of powder PET improved tensile strength and elastic moduli while reducing the ultimate elongation and impact property of the final material. The results of tensile and impact testing are given in Figures 2–4. The variation of experimental standard deviation in these mechanical tests were about 10% of the symbols registered in these figures.

Tensile strengths of the composites were found to increase with PET, treated and untreated, as followed from Figure 2. The ultimate strength of composites with 5% treated and untreated PET were observed to be lower than that of the HDPE. The tensile strength of untreated PET followed an expected behavior of weak filler matrix adhesion. This decrease in the ultimate tensile strength compared to the virgin polymer could be attributed to the inhibition of orientation of HDPE (the absence of strain hardening) with the addition of PET by drawing after the yield point HDPE. Then, the recovery in the ultimate strength could be considered as the direct consequence of the crystalline PET filler in the matrix HDPE where it reached the maxima at 40% PET concentration. The low ultimate tensile strength

at 40% untreated PET-filled HDPE strongly indicates the weak adhesion between two polymers, whereas the SCAs-treated PET-filled composites showed substantial increases in the ultimate tensile strengths after 15% PET content. The maximum tensile strength was measured around 32 MPa in mixed SCAs treatment incorporated with MA. In addition to these, the presence of CE produced nearly 10% decrease in the stress at break at 40% PET-filled sample and also in mixed SCAs-treated PET composite come afterwards.

No clear yield point was observed after 15% untreated PET filling. These samples were broken usually just before or when they reached the yield point. Yet, SCAs-treated PET-filled samples were observed to fail just after an obvious point yield, which reached around 5 to 7% elongation. The decrease in elongation at break with PET content became inevitable after 10%. As mentioned above, the composites with 5% PET powder failed considerably at lower elongation than that of pure HDPE before the main matrix HDPE became oriented. A slight and noticeable enhancement in SCAs-treated PET, in particular mixed silanes and CE, is seen in the elongation at break compared to the untreated PET particles filled HDPE (Fig. 3).

Only unnotched impact strength of 15 and 40% PET-filled samples were studied because none of

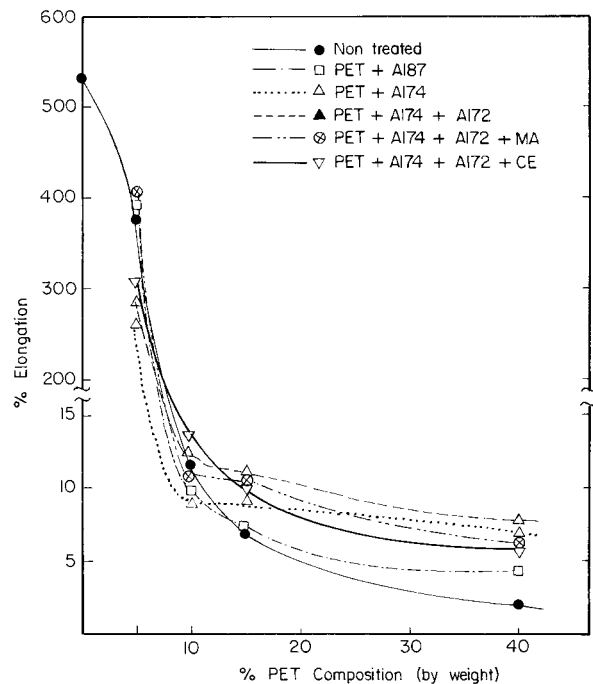


Figure 3 Variation of elongation at break with respect to PET filler concentration.

the samples of composites having 5 and 10% PET were broken and found to be highly resistant towards the impact force. PET surface treatment provided an improved impact resistance in all composites compared to nontreated PET-filled composites. The increase in the impact strength is almost four times of the nontreated PET (Fig. 4). It appeared that the improvement was the highest at lower content of PET (15%), MA, and CE in mixed silane treatment, and indeed, the MA mixed silane-treated sample was not broken in this composition. When PET content was increased to 40% the gap in the impact strength between SCAs-treated and untreated PET-filled composites was still found to remain where the presence of CE with mixed SCAs showed the best impact resistance.

Dynamic mechanical analysis (the average of two or three experiments) results are provided in Figure 5(A) and (B) as the variation of storage modulus with temperature, and $\tan \delta$ of the corresponding measurements in Figure 6(A) and (B) for 15 and 40% compositions. Elastic storage modulus increased with the addition of solid untreated PET powder into HDPE. Although the storage modulus of filled samples were comparably higher than that of pure HDPE, we found that

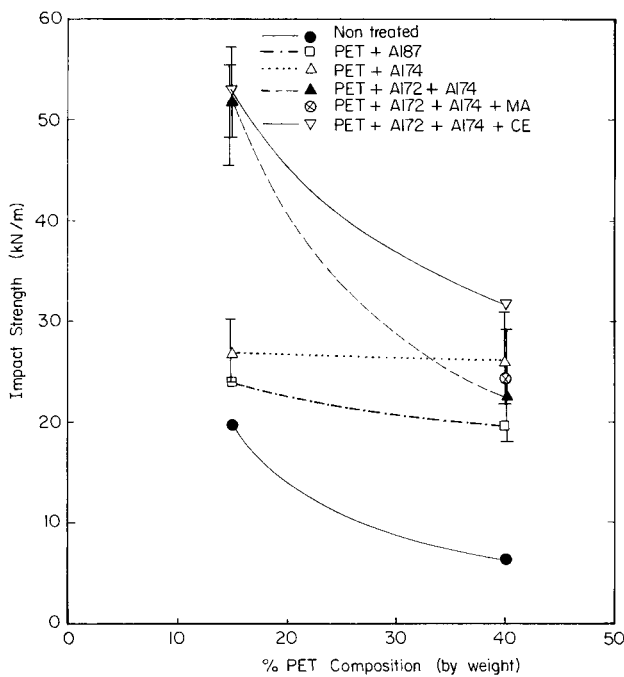


Figure 4 Variation of impact strength with respect to PET filler concentration. (Error bars were provided if the the standard deviation is more than 10% of the mean value.)

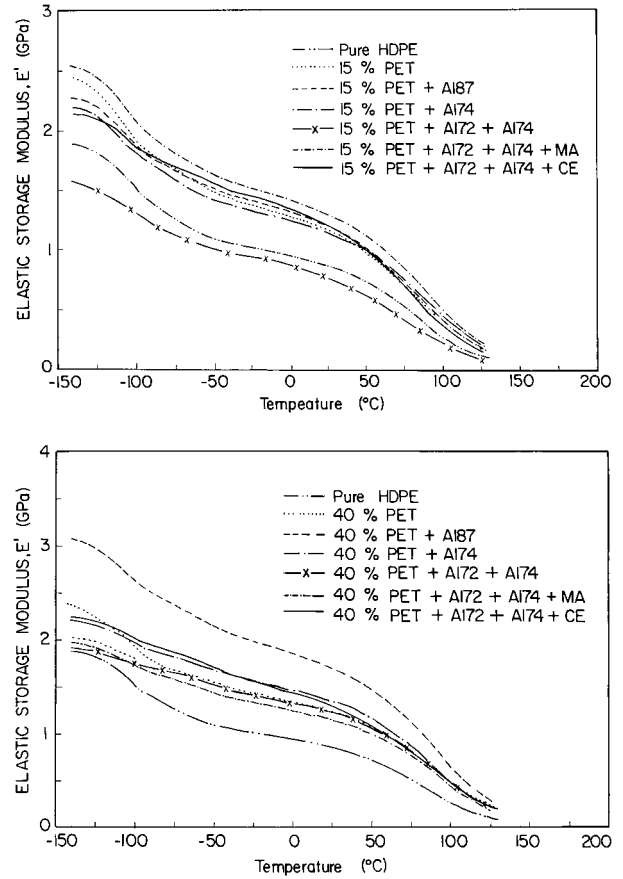


Figure 5 Elastic storage modulus of PET-filled HDPE composites measured in DMA: (A) 15% PET-filled HDPE with various types of silane treatment; (B) 40% PET-filled HDPE with various types of silane treatment.

there existed almost no difference between 15 and 40% PET-filled composites [Fig. 5(A) and (B)]. However, not shown here, 5 and 10% untreated PET-filled HDPE composites were found to be in between these lines but closer to the pure HDPE. The mechanical loss factor, $\tan \delta$, indicated that glass transition of HDPE shifted to lower values; nearly from -100 to -110°C , and became more diffused with increasing the PET filler content. The storage modulus variation for treated 15 and 40% PET filled composites as given in Figure 5(A) and (B) was found to be close to the untreated PET-filled samples without any distinct separation. There existed only two exceptions in the dynamic experimental data. The sample which contained A-187-treated 40% PET showed abnormally high modulus temperature change. This abrupt difference, however, was not completely reflected in static mechanical properties. Yet, this

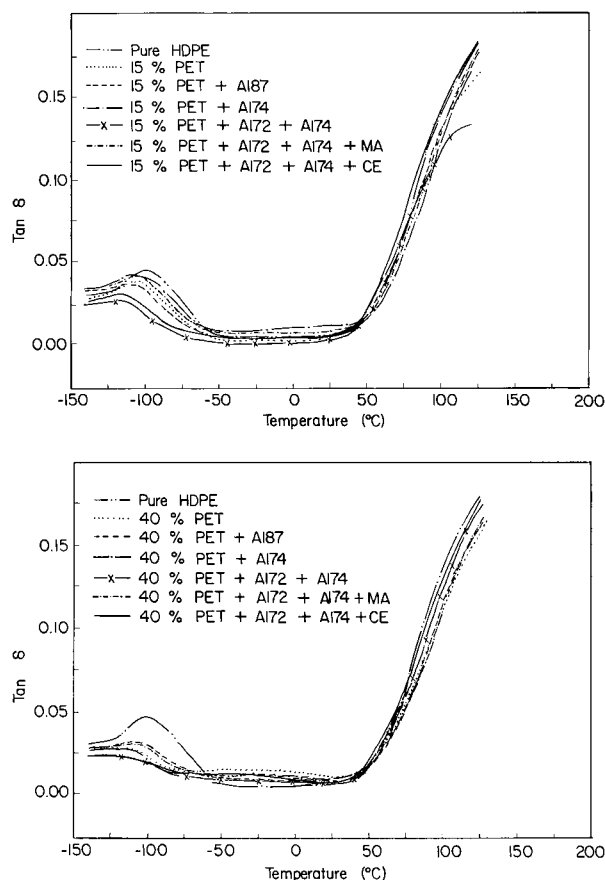


Figure 6 Tan delta ($\tan \delta$) of PET-filled HDPE composites as determined from DMA: (A) 15% PET-filled HDPE with various types of silane treatment; (B) 40% PET-filled HDPE with various types of silane treatment.

sample showed the next smallest elongation and the weakest impact strength after the untreated PET composites. The other experimental result that diverted from others was for the mixed silane-treated PET incorporated 15% with HDPE. This sample showed lower elastic storage modu-

lus than that of untreated PET-filled composite, and the same lowering was not observed in higher filling, 40% mixed silane-treated PET.

In general, addition of mineral fillers into polymers causes an increase in modulus and also glass transition temperature due to the polymer-filler interaction at the interface. This is the result of formation of partially immobilized layer between polymer and filler. In cases where the reduction of T_g is observed, the plasticization effect can be pronounced due to the surface treatment. In our case, the filler being a polymer appears to completely change the nature of interaction. Though the mechanical behavior is clearly followed by what is simply expected from a filled polymer, $\tan \delta$ variation obtained from DMA is just on the contrary of mineral fillers. All the glass transitions were found to be less than that of pure HDPE even in the untreated PET-filled samples, and the lowering in T_g was varied from 10 to 3°C. T_g values of PET-filled HDPE composites are given in Table II, based on Figure 6(A) and (B). Forty percent PET A-187 silane-treated composite had the closest T_g , around -104°C , to that of pure HDPE. It is very apparent that a possible plasticization effect of SCAs on the variation of T_g values HDPE can be suggested, and also the presence of CE with SCAs show a clean depression in T_g of HDPE while MA [see also Fig. 5(A), where MA and mixed silane containing 15% PET had higher modulus but not true for 40% PET-filled samples] results comparably in higher T_g in HDPE, the closest to pure HDPE.

In comparing the experimental results, it appears that the impact strength and ultimate mechanical properties are more effective in assessing and evaluating the behavior of the PET-filled composites with and without SCAs. Especially, the improvement in 40% PET composites, both

Table II Glass Transition Temperature of PET-Filled HDPE, Where T_g of Pure HDPE Was Measured as -100°C

Composition	15% PET-Filled HDPE T_g in $^\circ\text{C}$	40% PET-Filled HDPE T_g in $^\circ\text{C}$
Untreated PET	-103.8	-110.3
A-187 treated PET	-103.8	-105.2
A-174 treated PET	-106.7	-110.0
A-172 + A-174 treated PET	-113.3	-111.7
A-172 + A-174 and MA treated PET	-103.0	-103.8
A-172 + A-174 and CE treated PET	-106.7	-110.0

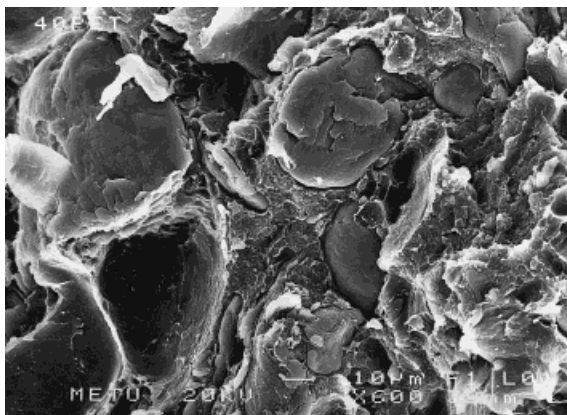


Figure 7 Impact fractured SEM micrograph of 40% untreated PET-filled HDPE.

impact and tensile strength, was worth mentioning.

SEM Studies

The improved mechanical properties, particularly in 40% treated PET inclusion, can be explained by interaction and adhesion between PET particles and HDPE. The basic idea in this work was to promote and apply SCAs as materials to make a chemical bond or to enhance interaction between two polymers that do not wet each other completely. Indeed, The fractograph (Fig. 7), shows that there is almost no interaction and adhesion between PET and HDPE. The surface of the particles was clean, and voids were observed around particles. The holes observed in SEM pictures of both tensile and impact fractured surfaces also indicated the weakness of the interaction between these two materials.

As it was explained in the works of Plueddemann,^{21,22} the coupling mechanism between polymers and mineral fillers of glass and also glass-like was achieved through the functional side of SCA for the former and the alkoxy side of SCA by hydrolysis and condensation processes for the latter. It has also been shown that γ -irradiated PTFE particles having oxygen bearing groups, like —OH and —COOH, could interact with SCAs and gave better mechanical properties when SCA-treated PTFE particles were introduced into LDPE.^{23,24} Both heat treatment and grinding processes in amorphous PET inevitably increased thermal decomposition and further oxidation. Zimmerman²⁵ showed that an extensive autocatalytic reaction by hydrolytic cleavage of polyester chains resulted in carboxyl groups under isother-

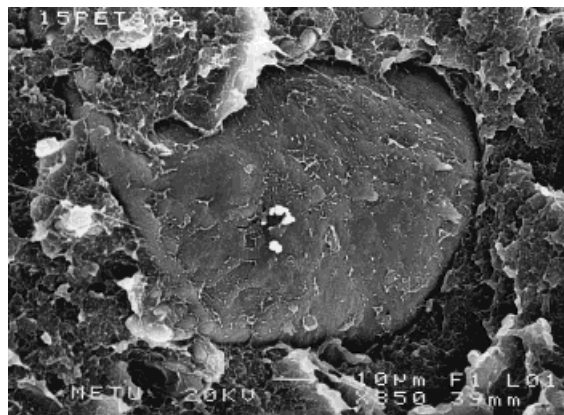


Figure 8 Impact fractured surface SEM micrograph of 15% A-187-treated PET-filled HDPE. (A-187 silane contains epoxy reactive groups.)

mal heating. Similar results were reported earlier by Buxbaum,²⁶ and the degradation mechanism was also reviewed by Jabarin.²⁷ Consequently, The silane treatment is expected to facilitate extensive hydrolysis and condensation, also possible alkoxy exchange reactions on the surface of the PET particles.

The following figures (Figs. 8–13) are SEM fractographs of tensile and impact testing for the treated samples. Impact and tensile fractured surfaces of A-187 and mixed silane-treated 15% PET filled HDPE are given in Figures 8 and 9, respectively. In both case the filler, PET, surface was covered and PET adhered to the matrix HDPE. A small gap all around the right side of the PET particle in the impact-fractured surface

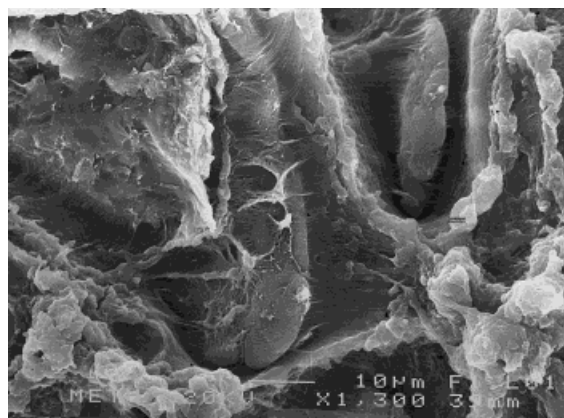


Figure 9 Tensile fractured surface of 15% mixed silane treated PET filled HDPE. Mixed silane was composed of A-172 and A174 in equal amounts as described in the text.

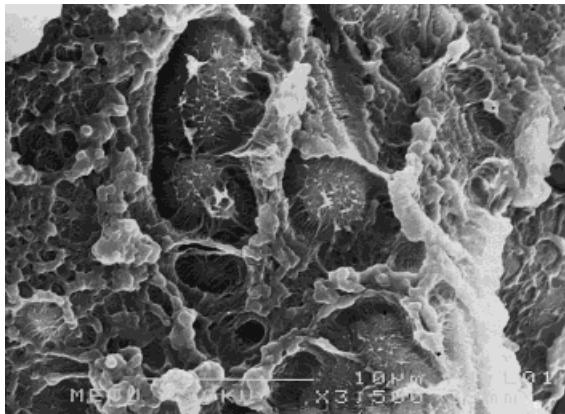


Figure 10 Tensile fractured surface of 15% A-174 silane-treated PET-filled HDPE.

might be directly due to the result of high impact energy in which the particle was forced to move. Yet, a strong interaction between PET particle and HDPE was still clearly seen. In tensile fractograph (Fig. 9), an orientation and fibrillation of HDPE around PET particles was observed due to slow drawing process and loading of energy on the sample compared to the impact energy. This orientation along the draw direction can also be observed in Figure 10, where A-174 silane-treated 40% PET-filled HDPE is the material.

Holes were observed in the tensile-fractured samples of mixed silane-treated samples when CE was present in the composite (Fig. 11). The number of holes increased if the sample was subjected to an impact force. However, no holes were seen in the impact-fractured surface of the mixed silane and MA-treated 40% PET-filled sample with a very strong interaction (Fig. 12). This dif-

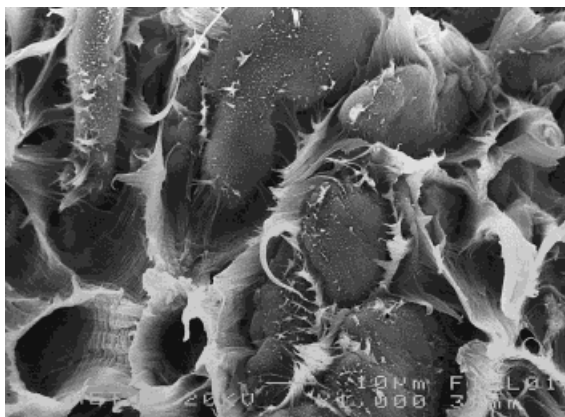


Figure 11 Tensile fractured surface view of mixed silane-treated 15% PET in cooperated CE.

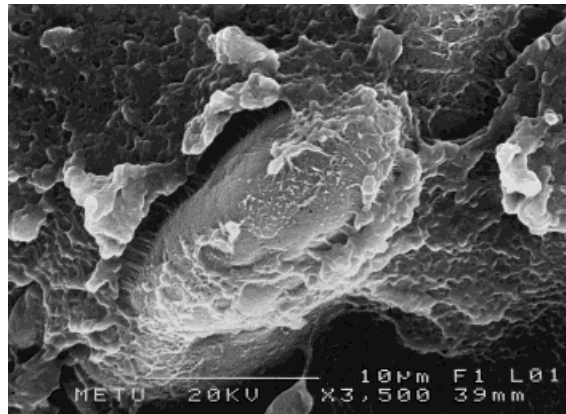


Figure 12 Impact fractured surface of 40% mixed silane-treated PET-filled HDPE where MA was present.

ference may be the result of the addition of CE and MA, in which CE possibly loosened the adhesion and creating some weak interaction areas while MA causes further interaction during processing. However, it should be noted that there exists no big difference in mechanical strengths of these two samples. Another observation was the peeling of the surface layer on PET particles only in mixed silane-treated PET-filled composite in impact testing, as given in Figure 13. The peeling in mixed silane-treated samples can be considered as a result of excessive self-hydrolysis and condensation process of SCAs on the PET particles. These comparatively weak interacted parts, expected to be oligomerized SCAs on PET, were peeled off from the surface of PET upon an impact force but not observed in tensile fractographs.

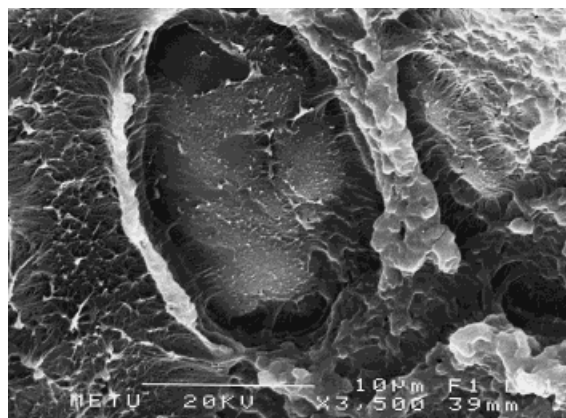


Figure 13 Tensile fractured surface of 40% mixed silane-treated PET-filled HDPE in the presence of CE. Note the peeling from the surface from PET particle becomes very important, particularly in impact force.

CONCLUSION

Silane coupling agents are used as the materials to promote the interfacial adhesion between fillers, especially glass and glass-like, and polymers. In this work, we, however, showed that SCAs could be also used to augment the interaction through a formation of an interfacial surface between two polymers where one of them was used as a solid filler, PET, in the polymer matrix of HDPE. The improvement in tensile and impact strength of the SCAs-treated PET-filled HDPE and the strong adhesion layer between PET and HDPE as followed from SEM fractographs compared to untreated PET samples were the strong indications of enhanced interaction between these two materials. The strength of the interaction can be evaluated from the impact and tensile strength results, especially in 40% PET-filled HDPE. Furthermore, the addition of PET filler was observed to decrease T_g of the main matrix on the contrary of mineral fillers added into the polymer matrices. It appears that SCAs act as plasticizers to a certain extent.

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