# $\gamma$ -Irradiated Poly(tetrafluoroethylene) Particle-Filled Low-Density Polyethylene. I. Effect of Silane Coupling Agents on Mechanical, Thermal, and Morphological Properties

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ABSTRACT: Poly(tetrafluoroethylene) (PTFE) scraps were recovered as a filler material for low-density polyethylene (LDPE) after they were degraded by Co-60  $\gamma$ -rays under atmospheric conditions to make small-size powder. The powder PTFE, which was called secondary PTFE (2°-PTFE), was melt mixed with LDPE and then extruded to obtain 200  $\mu$ m films. The mechanical and thermal properties and also the morphology of the fractured surface of these 2°-PTFE-filled LDPE were studied. It was found that the addition of 2°-PTFE resulted in thermofilm property of LDPE but it slightly decreased the thermal oxidative temperature of LDPE. The tensile strength and ultimate elongation of LDPE were found to decrease with the addition of 2°-PTFE. However, when it is compared to the addition of virgin PTFE into LDPE, 2°-PTFE shows better mechanical properties due to the presence of oxy groups which are capable of interacting with the main matrix. A further improvement in mechanical properties was achieved by silane coupling agent treatment of 2°-PTFE. Silane coupling agents were found to enhance the interfacial adhesion between 2°-PTFE and LDPE. The study on the fractured surfaces by scanning electron microscope revealed this adhesion between these two polymers. © 1999 John Wiley & Sons, Inc. J Appl Polym Sci 74: 866-876, 1999

**Key words:** 2°-PTFE particle-filled LDPE; silane coupling agents; mechanical properties; SEM characterization

## **INTRODUCTION**

Poly(tetrafluoroethylene) (PTFE) is well known for its good chemical and thermal inertness, its low coefficients of friction, and high electrical resistance. Besides these, it also has long wave infrared (IR) absorption, and those containing fluorine among the carbon-based polymers are distinguished by high stability to light. However, it is

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not possible to use these remarkable properties in any applications that require a good bonding ability to other materials, such as in polymer blends, due to the immiscibility of PTFE. Many methods have been proposed to modify the surface of PTFE; chemical<sup>1-3</sup> and physical treatments,<sup>4-8</sup> plasma modifications,<sup>9</sup> and radiation treatments<sup>10–16</sup> were performed for the adhesion between the PTFE and the other polymers in making blends or composites.

Among these chemical surface treatments, the most popular chemical surface treatment method of PTFE is known as the sodium etching method. Extremely reductive solutions of Na-naphthalene complexes, radical anion naphthalene complexes, or Na metal in liquid ammonia solutions were used to modify the surface of PTFE. However,

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PTFE in these modifications demonstrated extensive defluorination followed by oxygen incorporation and surface unsaturation resulted in a hydrophilic surface of PTFE.<sup>1–3</sup> Mechanical alloying of PTFE powder and transcrystallization is another example of physical surface treatments on PTFE. Mechanical alloying of PTFE powder with polyethylene (PE) powder showed that the compatibility of PTFE and PE began to occur after a period of ball milling.<sup>4</sup> Butadiene, ethylene, and methylmethacrylate monomers were block copolymerized by the initiation of mechanoradicals on PTFE powders with extremely low grafting ratio less than 0.1 to 3%.<sup>5,6</sup> The transcrystallization of polypropylene (PP) onto the surface of PTFE fibers was also found to improve the interfacial strength between two immiscible polymers. The mechanical properties were improved with a suitable transcrystalline thickness, particularly the adhesive fracture energy and the interfacial shear strength.<sup>7,8</sup> Plasma surface treatments were also used to change the adhesion properties of PTFE under inert or oxidizing gases such as air, nitrogen, argon, oxygen, etc.<sup>9</sup> Ionizing or high-energy radiation were particularly studied for radiation grafting of vinyl monomers into PTFE films. Acrylic acid,<sup>10–12</sup> methacrylic acid,<sup>13</sup> 4-vinylpyridine,<sup>10,11,14</sup> N-vinylpyrrolidone,<sup>10,11</sup> and vinylfluoride<sup>15,16</sup> are some examples of these monomers.

Although PTFE has outstanding thermal and chemical stability, it is very sensitive to highenergy radiation. The dominant effect of ionizing radiation on PTFE is molecular weight degradation caused by polymer chain scission. Even a small dose can make it lose most of its mechanical properties such as tensile strength, ultimate elongation, and embrittlement of the polymer. It has been shown that PTFE radicals reacted rapidly with oxygen to give the corresponding peroxy radicals when PTFE is exposed to ionizing radiation. Electron spin resonance spectroscopy showed that mid-chain and end-chain radicals were produced by high-energy radiation under vacuum and air irradiation. These radicals react with oxygen to yield corresponding peroxy radicals then followed by carboxylation of the molecules. Furthermore, the formation of vinyl end groups were identified under  $\gamma$ -radiation in the presence of oxygen atmosphere.<sup>17-22</sup> However, high-energy radiation under vacuum showed the characteristics of olefin formation in the polymer chain and was responsible for the possible branching or crosslinking reactions.<sup>23</sup> The changes in thermal

expansion coefficient,<sup>24</sup> thermal conductivity,<sup>25</sup> and dielectric properties<sup>26</sup> of PTFE were also studied when PTFE was exposed to  $\gamma$ -irradiation.

Some important PTFE blends and composites were reported in the literature with polyacetal (POM),<sup>27,28</sup> PE,<sup>29–33</sup> polyimide,<sup>34</sup> poly(ethylacrylate),<sup>35</sup> and poly(etheretherketone).<sup>36</sup> In all these studies, some properties were found to be improved such as mechanical, thermal, and wear resistance of the corresponding blend and composite application of PTFE. Although the miscibility between POM and PTFE was poor, the blend showed very good wear resistance, and strong bonding and homogeneous dispersion were provided between POM and PTFE through a chemical treatment of the surface of PTFE by an Na-etching method.<sup>1–3</sup>

Commercially available powders of high-density polyethylene (HDPE) and PTFE were used to make HDPE-PTFE samples with different compositions. Preparation of homogeneous mixtures by a dispersion mixing followed by sintering of the compacts at 200°C, above the melting point of PE but well below that of PTFE. IR, nuclear magnetic resonance, and electron spectroscopy for chemical analysis studies indicated that there might be a possible interaction between the two crystalline polymers on the molecular level. Although both PE and PTFE chains are nonpolar, C-F and C-H bonds are polarized and the high electronegativity of fluorine might be responsible for the interactions with hydrogen in PE. Hence, the interdiffusion of PE and PTFE chains that occurred at the interface resulted in the transitional layers.<sup>29,30</sup> As a processing aid, fluoropolymers blended with linear low density polyethylene (LLDPE) were found to improve the appearance of extrudates and reduce the melt viscosity. Fluoropolymers acted as an adhesion promoter between flowing melt and die wall, thus acting as a lubricant at the polymer stationary phase interface and promoting the slip of LLDPE melts.<sup>31</sup> Morphology and mechanical properties of ultrahigh molecular weight polyethylene (UHMWPE)-PTFE blends and composites prepared by kneading techniques to promote both the heat resistance and the wear property of PE were also studied.<sup>32,33</sup> Despite the incompatibility, with a composition 75 PTFE/25 UHMWPE, UHMWPE chains were found to diffuse within the PTFE fibrillar texture and could not be separated from the PTFE matrix. The addition of PTFE micropowder resulted in an increase in modulus but significantly lowered the strain at break values

while a slight improvement on wear resistance of UHMWPE was seen.

Our objective in this study can be described in two parts: 1. the recycling of PTFE scraps by grinding into microsize powders after  $\gamma$ -irradiation where these PTFE powders were described as 2°-PTFE, and 2. to introduce these 2°-PTFE into LDPE to improve processibility, UV-stability, and also to enhance the IR keeping capacity of LDPE films if they are used in greenhouses. The  $\gamma$ -irradiated PTFE containing oxy groups like carbonyl and alcohol could be used to enhance the interfacial interaction between these two polymers. Silane coupling agents (SCAs) were applied for further enhancement in the interaction. Varying amounts of 2°-PTFE powder (0.5 to 10 % by weight) was added into LDPE. SCAs were applied to the surface of freshly ground PTFE powders. In this part of work, the mechanical and thermal properties of these 2°-PTFE powder-filled LDPE were evaluated.

## **EXPERIMENTAL**

LDPE was supplied by Turkish Petrochemical Industry "PETKIM" coded as G 03-5. Melt flow index and density of the polymer were 0.3 g/10 min and 0.921 g/cm<sup>3</sup>, respectively.

PTFE powder used in the study was obtained by  $\gamma$ -irradiation of scraped PTFE in the forms of turnings and ribbons during the reshaping procedure of PTFE blocks. The oxidative and chain scission degradation of PTFE induced by Co-60  $\gamma$ -irradiation source (GAMMACELL 200, Atomic Energy of Canada Ltd.) continued to the 70 kGy absorbed dose as optimum dose. Dose rate was about 0.2 kGy/h during the study. Finally, degraded, brittle PTFE was ground into powder PTFE (secondary, 2°-PTFE) and sieved from a 270 mesh sieve by using an Octagon 200 test sieve shaker. Ninety percent of the 2°-PTFE powder obtained by this method had under 90  $\mu$ m particle size (Fig. 1) which was determined by using the particle size analyzer, Mastersizer S.

Various kinds of SCAs were used for the surface treatments of 2°-PTFE powder to enhance the adhesion between 2°-PTFE powder and LDPE matrix. SCAs were provided from Union Carbide (now HÜLS Group Company, Germany). Types, codes, and chemical formulations of SCAs, with new abbreviations in italic, are also given in Table I. In all cases, the amount of SCAs was about 2% by weight with respect to the amount of 2°-



**Figure 1** Particle size analysis of 2°-PTFE powder, as volume average distribution.

PTFE powder and totally 2%, 1 : 1 mixture of A-1100/189, A-186/189, and A-172/189 types of SCAs were also used for this purpose. Diethylether was used as solvent for SCAs and the surface treatment of the 2°-powder was performed in a slurry mixture of required compositions in 50-mL diethylether. The slurry was continuously mixed until most of the solvent was evaporated in 2 h at room temperature. Final mixture was then dried in an oven overnight at 60°C.

Silane-treated and untreated 2°-PTFE powderfilled LDPE, 0.5, 1.5, 3, 5, and 10% (w/w), were first mixed in a mixing head (W30H) of Brabender Plastic Corder Torque Rheometer, PLV-151. To provide a uniform dispersion of the filler (PTFE) in LDPE matrix was performed at 60 rpm of speed of rotation and 170°C for 5 min. Compression-molded sheets of around 4-mm thickness were prepared from these compositions between two aluminum plates at 190°C in a hot platen press, just after the complete mixing. These polymer sheets were cut into small pieces which were then extruded by using a single screw extruder (Model CS-194) attached to the Brabender in which the zone temperatures were adjusted to 175, 190, 200, and 210°C at a speed of screw 30 rpm. The changes in torque on the screw and pressure in the head of the extruder were also measured with respect to 2°-PTFE composition by using the torque rheometer of Brabender and Dynisco melt pressure transducer, respectively. As given in Figure 2, after an initial decrease in torque and pressure with 1.5% addition of 2°-PTFE, both variables were increased slightly, then remained almost unchanged. The processing aid property of 2°-PTFE appears to be very effective without any doubt. SCAs treatment, however, did not greatly

Types of Silane Coupling Agents	Chemical Formula
A-1100 (amino silane) gamma-aminopropyltriethoxysilane (DYNASYLAN AMEO)	$H_2NCH_2CH_2CH_2Si(OC_2H_5)_3$
A-1120 (amino silane) N- <i>beta</i> -(aminoethyl)- <i>gamma</i> - aminopropyltrimethoxysilane ( <i>DYNASYLAN DAMO</i> )	$H_2N(CH_2)_2NH(CH_2)_3Si(OCH_3)_3$
A-186 (epoxy silane) beta-(3,4-epoxycyclohexyl)- ethyltrimethoxysilane	$CH_2CH_2Si(OCH_3)_3$
A-187 (epoxy silane) gamma-glycidoxypropyltrimethoxysilane (DYNASYLAN GLYMO)	$\overset{O}{\swarrow}_{CH_2CHCH_2O(CH_2)_3Si(OCH_3)_3}$
A-189 (mercapto silane) gamma-mercaptopropyltrimethoxysilane (DYNASYLAN MTMO)	$HSCH_2CH_2CH_2Si(OCH_3)_3$
A-172 (vinyl silane) vinyl-tris-( <i>beta</i> -methoxyethoxy)silane ( <i>DYNASYLAN VTMOEO</i> )	$CH_2 = CHSi(OC_2H_4OCH_3)_3$
A-174 (methacrylo silane) gamma-methacryloxypropyltrimethoxysilane	$\begin{array}{c} \mathrm{CH}_3 & \mathrm{O} \\   &    \\ \mathrm{CH}_2 = \mathrm{C} - \mathrm{C} - \mathrm{O}(\mathrm{CH}_2)_3 \mathrm{Si}(\mathrm{OCH}_3)_3 \end{array}$

Table I Types and Chemical Formulas of Silane Coupling Agents

change these values. On an adjustable speed conveyer belt, 2°-PTFE powder-filled LDPE films at 200  $\pm$  20  $\mu$ m thickness were obtained. Mechanical properties of 2°-PTFE powder-filled LDPE samples were also compared with pure PTFE powder (Hostaflon TF-9205, Hoechst, Germany, 30- $\mu$ m average particle size) filled samples.

Tensile properties, stress and elongation at break, of modified and unmodified 2°-PTFE powder-filled films were determined by an Instron Tensile Testing Machine (Model TM 1102). Crosshead speed and gauge length in measurements were 5.0 cm/min and 3.5 cm, respectively. Tests were performed at room temperature and the average of at least five samples is reported.

SEM and X-ray fluorescence (XRF) analysis were held on tensile fractured surfaces of the modified and unmodified 2°-PTFE powder-filled LDPE film samples at various magnifications after gold plating by using a scanning electron microscope, JEOL, JSM-6400.

Thermal properties were followed as the thermal oxidation temperature (TOXT) of film samples by differential scanning calorimeter (DSC) (TA instrument of DSC 910 S). TOXT of film samples were investigated under  $O_2$  atmosphere. Heating rate and amount of samples were used in all of the measurements 10°C/min and 3–10 mg, respectively.

## **RESULTS AND DISCUSSION**

### Spectroscopic and Thermal Characterization

The difference Fourier transform infrared spectroscopy (FTIR) spectrum between 70 kGy irradiated PTFE and pure PTFE is given in Figure 3. The carboxylation of PTFE with irradiation is clearly visible. OH-stretching vibrations of the acid group above  $3000 \text{ cm}^{-1}$ , a band of the terminal carboxylic acid fluoride group at  $1880 \text{ cm}^{-1}$ , the carbonyl vibrations at  $1780 \text{ cm}^{-1}$ , and isolated carboxyl groups at  $1810 \text{ cm}^{-1}$  were observed with good agreement of Lunkwitz et al.<sup>17</sup>

The IR spectra of LDPE and 5% 2°-PTFE powder-filled LDPE films between the region 900– 1400 cm<sup>-1</sup> are shown in Figure 4. The long wave IR absorption peaks at 1145 cm<sup>-1</sup> and 1203 cm<sup>-1</sup> originated from 2°-PTFE powder. This region cor-



**Figure 2** Variations of the torque and the pressure in processing of the PTFE-LDPE mixtures vs. composition.

responds to the black body IR radiation of soil inside a greenhouse where LDPE has no strong absorption in this region. The 2°-PTFE introduces a thermofilm property to a greenhouse which can be kept warm for a definite but a longer time compared with the pure LDPE films.

TOXT vs. percent composition of various kinds of silane-treated and untreated PTFE-filled LDPE films are given in Figure 5. TOXT was determined as the onset point in DSC with 10°C/



**Figure 3** The difference absorption FTIR spectrum of 70 kGy irradiated PTFE from pure PTFE.



**Figure 4** IR spectra of LDPE and 5% 2°-PTFE powder-filled films in long IR region.

min heating rate where the exothermic slope change was observed in the DSC thermogram of the samples. A bold curve indicates the average value of TOXT of all corresponding compositions of the samples. A slight decrease of the TOXT of the samples compared with pure LDPE was observed as an average value. The broken lines



**Figure 5** The variation of TOXT with respect to composition of PTFE-filled LDPE. Symbols inside the figure show the silanes and PTFE. The bold line shows the average of all measurements and the broken lines are drawn for pure and 2°-PTFE powder-filled LDPE.

show both pure and 2°-PTFE-filled LDPE. The decrease in TOXT appeared to be clearer in both cases. For 1.5% composition, this decrease in TOXT was found to be surprisingly higher than others. Thermal conductivities of PTFE and LDPE are about 0.27 and 0.35 W/m.K, respectively. This value was shown to increase up to 0.5 W/m.K by  $\gamma$ -irradiation of PTFE, due to the enhancement in the crystallinity of PTFE.<sup>25</sup> So higher thermal conductivity of filler and the discontinuity created by filler through the matrix should be responsible for the decrease in the TOXT compared with pure PE. The average values of TOXT for the rest of the compositions were almost constant but slightly higher than the minimum observed at 1.5% composition. As average values, the similar behavior was seen in the silane-treated samples but there existed no regularity with respect to the type and mixture of SCAs.

#### **Mechanical Properties**

Values of 0.5, 1.5, 3, 5, and 10% by weight silanetreated, untreated 2°-PTFE powder, and pure PTFE powder-filled LDPE samples were tested mechanically. Stress-strain curves of 3 and 5% SCAs treated, untreated, and pure PTFE powderfilled samples are given in Figure 6(a,b). (To avoid overcrowding, only some selected stress-strain curves are given.) Elastic modulus of the samples appeared almost the same value within the experimental errors. The mechanical strength of the pure PTFE powder-filled samples was apparently found to be less than that of 2°-PTFE powderfilled samples and further enhancement in ultimate mechanical properties were obtained by the SCA treatments of the surface of 2°-PTFE powder.

The ultimate tensile strength and strain at break (%) of the filled samples are given in Figures 7 and 8, respectively. The addition of pure PTFE powder caused a sharp decrease in the strain and stress at break of the samples compared with LDPE, with increasing in the amount of pure PTFE powder resulting in a further decrease in these ultimate properties due to the incompatibility of PTFE. However, 2°-PTFE powder apparently yielded better results for these mechanical properties compared with pure PTFE. There was almost no change in the strain at break values of the samples with increasing amounts of 2°-PTFE powder (Fig. 8). The increase in the surface energy of the 2°-PTFE powder, due to  $\gamma$ -irradiation-induced oxidative chain degradation, revealed better incorporation with LDPE and hence better mechanical properties were achieved when it was compared with that of pure PTFE powder. Indeed, 2°-PTFE showed comparably higher val-



**Figure 6** Stress-strain curves for pure LDPE, 3 and 5 % PTFE powder-filled LDPE: (a) indicates 3% IRR-PTFE, A-1100, and a mixture of A-1100-189 silane-treated 2°-PTFE powder-filled samples, and (b) 5% pure, IRR-PTFE and A-189 silane-treated 2°-PTFE powder-filled samples.

ues for both mechanical properties at 5 and 10% PTFE.

Silane treatments of 2°-PTFE powder caused significant enhancement in ultimate properties. The behavior of SCAs, however, varied with their types when the interfacial adhesion was involved. In general, it is not difficult to say some particular reactive sites in SCAs acted well compared with others. On the other hand, some silanes having the same reactive site showed contrary effects. A-186 (epoxycyclohexyl) type silane, showed an antagonistic effect in the mechanical properties, yet another kind of epoxy silane, A-187, improved the ultimate properties, especially 1.5, 3, and 5 % compositions. Although A-189 (mercapto) silane treatment enhanced strain at break, no improvement was seen in its ultimate stress to that of nontreated powder-filled samples (Fig. 7). A-189 type silane may be said to increase the ultimate elongation with a possible plasticizing effect; on the other hand, it adversely affects the stress at break. There were also improvements in ultimate properties of 1.5 and 3 % compositions of A-1100 (a 1°-amine silane), 1.5, 3, and 5 % compositions of A-1120 (a 2°-amine silane), and all compositions in A-172 (a vinyl silane), and A-174 (a methacrylo silane) treated 2°-PTFE powder-filled samples compared with untreated one. Furthermore, these last two types of silane coupling agents represent more reproducible results within the experimental fluctuations in all compositions. Silane coupling agents, therefore, can be used as a material to enhance the interfacial interactions between the polymer and the organic filler where in this case the organic filler is also an extreme polymer with a very low adhesion property, if it is chosen properly.

A-1100-189, A-1120-189, and A-187-189 silane mixtures were also tested to obtain any synergetic effect in the mechanical properties. In these mixtures, A-189 type of silane was hold fixed because of its high UV stability, which will be discussed in the second part of our work. Only A-1100-189 type silane mixture showed a synergetic result on strain at break compared with the presence of individual silanes. The others present intermediate results at the 1.5 and 3% compositions.

The 2°-PTFE-filled films started to lose their transparency after 5%, and 10% addition of filler resulted in highly opaque films. Because transparency is one of the important parameters in greenhouse applications, these concentrations were not used for further analysis. For this rea-



**Figure 7** Bar graphics of tensile strength for LDPE and untreated pure PTFE and 2°-PTFE-filled LDPE, and silane-treated 2°-PTFE powder-filled LDPE. The type of silane treatment is given below each composition.

son, only some samples were studied in the presence of SCAs for 5 and 10% compositions, to check further enhancement.

#### **SEM Studies**

An SEM fractograph of the mechanically tested surfaces of an untreated 2°-PTFE powder-filled sample is given in Figure 9. Irregular-shaped powders were distributed homogeneously into an LDPE matrix without any visible adhesion between the filler and the matrix, but it should be noted that 2°-PTFE resulted in better mechanical properties. Surface treatment of 2°-PTFE powder with different types of silane coupling agents enhanced the adhesion of the filler with LDPE (Figs. 10–13). As shown in Figure



**Figure 8** Bar graphics of strain at break for LDPE and untreated pure PTFE and 2°-PTFE-filled LDPE, and silane-treated 2°-PTFE powder-filled LDPE. The type of silane treatment is given below each composition.

10, the filler surface was covered completely with matrix due to the A-1100, 1°-amine silane, treatment.

XRF analysis was performed on these particles. A sharp increase in F atom intensity proved that these particles were embedded 2°-PTFE particles into LDPE as given in Figure 14. The same kind of analysis was also done for the presence of 2°-PTFE particles in all these fractographs of the composites to be sure if they were 2°-PTFE. The low particle concentration, small thickness of the samples, and the ductile fracture of LDPE with a certain extent of fibrillation usually covered and hid the 2°-PTFE in the fractured surface. The possibility of obser-



Figure 9 SEM fractograph of the fractured surface of untreated 2°-PTFE powder-filled LDPE (1cm =  $6.25 \mu$ m).



**Figure 10** SEM fractograph of the fractured surface of A-1100 silane coupling agent-treated  $2^{\circ}$ -PTFE powder-filled LDPE (1cm = 10  $\mu$ m).

vation of any interference because of dust and foreign particles was therefore eliminated by XRF analyses. For these, A-1100, 1°-amine silane- (Figs. 10 and 11), A-1120, 2°-amine silane-(Fig. 12), and A-189, mercapto silane-treated (Fig. 13) particles showed the adhesion with the LDPE matrix. Besides these observations, in some certain silane treatments, like A-1120, a 2°-amine silane, it was seen that LDPE fibrils were also stuck on the surface of the filler upon fracture (Fig. 12).

Therefore, silane coupling agents can also be used to increase adhesion between the polymers. Possible bonding mechanism of 2°-PTFE powder to PE can be explained as the reaction sequence



**Figure 11** SEM fractograph of the fractured surface of A-1100 silane coupling agent-treated  $2^{\circ}$ -PTFE powder-filled LDPE (1cm = 10  $\mu$ m).



**Figure 12** SEM fractograph of the fractured surface of A-1120 silane coupling agent-treated  $2^{\circ}$ -PTFE powder-filled LDPE (1cm = 5  $\mu$ m).

shown. Carboxylic acid end groups, which arise from  $\gamma$ -irradiation induced oxidative chain scission of the PTFE, can be considered to be the most probable reaction center of the 2°-PTFE powder with silane coupling agents. R' in SCA represents the other reactive site that is capable of interacting with PE.





**Figure 13** SEM fractograph of the fractured surface of A-189 silane coupling agent-treated 2°-PTFE powder-filled LDPE (1cm = 10  $\mu$ m).





**Figure 14** XRF analysis to indicate embedded 2°-PTFE powder in the LDPE matrix.

## **CONCLUSION**

The 2°-PTFE powder, obtained by  $\gamma$ -irradiation– induced oxidative and chain scission degradation of PTFE scrap, can be used as a suitable filler that provides thermofilm property to LDPE in addition to the process aids of LDPE melt. The developed oxy groups like carbonyl, peroxy during the  $\gamma$ -irradiation are obviously considered to be the potential reactive sites ready for the interactions with the main matrix LDPE. When the mechanical properties of pure PTFE and 2°-PTFE-filled LDPE, with the size difference between them considered, 2°-PTFE showed observable and distinguishable results with increasing 2°-PTFE content compared with that of pure PTFE-filled samples. The lowest melt viscosity and TOXT of LDPE are obtained at 1.5% 2°-PTFE powder concentration. This also reveals that a better incorporation is achieved between the filler and LDPE matrix. Silane coupling agent treatment of 2°-PTFE particles appears to be very effective in inducing further interfacial adhesion between these two polymers. The effectiveness of SCAs seems to depend on the present reactive sites in SCAs. In general, with the exceptions of the mercapto (A-189) and one of the epoxy silanes (A-186), SCAs can be considered and proposed as

materials that can promote the interfacial adhesion between two polymers, not only for mineral fillers or glass but also for a case of PTFE.

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