### $\gamma$ -Irradiated Poly(tetrafluoroethylene) Particle-Filled Low-Density Polyethylene. II. UV Stability of LDPE in the Presence of 2°-PTFE Powder and Silane Coupling Agents

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**ABSTRACT:** Ultraviolet (UV) stability of  $\gamma$ -irradiated poly(tetrafluoroethylene), 2°-PTFE, powder-filled low-density polyethylene (LDPE) was studied in this work. The mechanical and thermal properties of 2°-PTFE powder-filled LDPE were discussed in our previous work (Akınay and Tinçer, 1999). It already has been shown that silane coupling agents (SCAs) result in improvements in mechanical properties. The UV stability of these samples was followed by a comparision between strain at break measured after UV irradiation and the initial strain at break, in terms of their ratio defined as residual elongation. The development of carbonyl index was also determined by infrared measurements. To get further UV stability, a hindered amine light stabilizer (HALS) was also used. Whereas addition of 2°-PTFE slightly enhanced the UV stability of LDPE, mercapto type silane (A-189) treatment appeared to increase the UV stability compared with the other types of silane treatments. HALS highly improved the UV stability of untreated and silane-treated 2°-PTFE-filled LDPE at given UV irradiation time. Although we observed some fluctuations in the experimental data of HALS and SCA treated 2°-PTFE-filled LDPE, the coaddition of HALS and SCAs showed synergetics effects in the UV stability compared with the separate additions. © 1999 John Wiley & Sons, Inc. J Appl Polym Sci 74: 877-888, 1999

**Key words:** UV stability; 2°-PTFE-filled LDPE; silane coupling agents; hindred amine light stabilizer

### INTRODUCTION

In the first part of our study,<sup>1</sup> we discussed the variation in mechanical and thermal properties of  $\gamma$ -irradiated poly(tetrafluoroethylene) (2°-PTFE)–filled low-density polyethylene (LDPE). As was mentioned, silane coupling agents (SCAs) were shown to promote the adhesion between 2°-PTFE

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and LDPE and the mechanical properties were also found to be quite acceptable in terms of some further applications. Fourier transform infrared spectroscopy (FTIR) study also revealed that the 2°-PTFE-filled LDPE possessed thermofilm property at even low concentrations. These films with and without SCA-treated 2°-PTFE could, therefore, be proposed to be used in greenhouse applications.

Ethylene-vinyl acetate copolymer (EVA) and LDPE are the most widely used plastics materials for greenhouse films. The former has excellent transparency and absorption bands in the long wave infrared (IR) corresponding to the spectral emission of soil (7–14  $\mu$ m) called thermofilms.<sup>2</sup> However, LDPE is highly susceptible to attack by

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ultraviolet (UV) light, heat, and oxygen, resulting in polymer brittleness, used preferably as greenhouse films because of its low price compared with EVA. Thus, LDPE should be stabilized against UV radiation to increase the useful life time as a greenhouse cover and also filled with special additives such as silicates and kaoline which are long wave IR absorbers. However, these additives led to decrease in the mechanical properties, especially in tensile strength and strain of the polymer.

In the early studies, Ni quenchers were used as photostabilizing agents for LDPE. Khraishi and Al-robaidi<sup>3</sup> found that bottom layers were least affected by UV radiation in Ni quencher stabilized multilayer greenhouse cover under natural weathering conditions. The effects of natural weathering on mechanical properties and chemical structure of Ni quenchers combined with UV absorber-stabilized LDPE were also studied by Akay et al.<sup>4</sup> No simple correlation was found to exist between the development of carbonyl groups and strain at break which was one of the most important parameters in following UV stability. It was also shown by Raab et al.<sup>5</sup> that the chemical changes might not greatly influence the bulk properties, such as complex modulus, but they significantly decreased strain at break because fracture followed the weakest path in the material.<sup>5</sup> Hence, in UV stability assessments, the change in elongation at break commonly is used rather than other mechanical properties. An initial increase in residual elongation (RE: the ratio of strain at break at the given UV exposure time to strain at break of unexposed sample) followed by steady decrease was described as chain stiffening effect.<sup>6</sup> This was also observed with a slight increase in RE, called a "recovery period," during the winter months in natural weathering.<sup>4</sup> Chain stiffening effect was defined by a partial increase in crystallinity due to the formation of smaller chains and also crosslinking reactions.<sup>7</sup>

Among these UV stabilizers, the development of hindered amine light stabilizers (HALS) was a big step forward in stabilization and this class of compounds has been increasingly used for PE stabilization. They are the scavengers of free radicals, especially for alkyl and alkylperoxy radicals. Photostabilization mechanism of HALS was proposed by Gugumus.<sup>8</sup> Durability of HALS-stabilized PE against UV irradiation in the model greenhouse and on aluminum racks was studied by Khan and Hamid.<sup>9</sup> It was reported that the film exposed in the model greenhouse showed a higher rate of photo-oxidative degradation in terms of higher crystallinity, carbonyl growth, and mechanical properties than the film exposed on aluminum racks. This was because of the conditions inside the greenhouse such as temperature, humidity, chemical fertilizers, etc. In another work, stability of HALS-stabilized PE was also examined in terms of oxygen uptake, formation of CO and  $CO_2$ , and the mechanical properties. It was noticed that HALS-stabilized PE showed superior properties against UV irradiation compared with unstabilized PE within the given time.<sup>10</sup>

As was discussed in our previous article,<sup>1</sup> 2°-PTFE powder showed thermofilm property to LDPE and improved processing aids of LDPE melt. Besides these, SCA treatments onto the surface of 2°-PTFE powder enhanced the interfacial adhesion between the filler and LDPE matrix, hence mechanical properties of the 2°-PTFE particle-filled LDPE. In addition to long wave IR absorption of PTFE, it is well known that PTFE also has high UV stability. Our objective in the second part of this study was to investigate the UV stability of LDPE filled with 2°-PTFE and the effect of SCAs and also the changes in the presence of HALS-type UV stabilizer under UV irradiation. The variations in the residual elongation and carbonyl index of the 2°-PTFE powder-filled LDPE were used to follow and compare the stability of all the sets of samples studied up to 1000 h UV irradiation. Because of the large number of samples and its variations, we had to follow UV irradiation up to 1000 h and the results were compared with each other for this final UV irradiation time.

### **EXPERIMENTAL**

LDPE was supplied from 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. Finally, degraded, brittle PTFE was grinded into powder PTFE (secondary, 2°-PTFE). The other details of the 2°-PTFE are discussed in our previous study.<sup>1</sup>

Various kinds of SCAs were used for the surface treatments of 2°-PTFE powder to increase the adhesion between 2°-PTFE powder and LDPE matrix, hence, UV stability of the samples. SCAs were provided from Union Carbide (now HULS 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°-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. The final mixture was then dried in an oven overnight at 60°C.

One of the HALS, Chimassorb 944-LD (944), was provided by CIBA-GEIGY (Switzerland) and utilized (0.5% by weight) in the study for further UV stabilization.

Values of 0.5, 1.5, 3, 5, and 10% (w/w) silane treated, untreated, and 944-stabilized 2°-PTFE powder-filled LDPE 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. On an adjustable speed conveyer belt, 2°-PTFE powder-filled LDPE films at 200  $\pm$  20  $\mu$ m thickness were obtained. Films were cut off using a standard dumbbell shaped die along the flow direction of the extruded films. These films were then exposed to UV irradiation by using four UV lamps (SYLVANA PAR 38 Mercury Lamp, USA) from both sides of samples at

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) <i>beta</i> -(3,4-epoxycyclohexyl)- ethyltrimethoxysilane	CH <sub>2</sub> CH <sub>2</sub> Si(OCH <sub>3</sub> ) <sub>3</sub>
A-187 (epoxy silane) gamma-glycidoxypropyltrimethoxysilane (DYNASYLAN GLYMO)	O CH <sub>2</sub> CHCH <sub>2</sub> O(CH <sub>2</sub> ) <sub>3</sub> Si(OCH <sub>3</sub> ) <sub>3</sub>
A-189 (mercapto silane) gamma-mercaptopropyltrimethoxysilane (DYNASYLAN MTMO)	$\mathbf{HSCH}_{2}\mathbf{CH}_{2}\mathbf{CH}_{2}\mathbf{Si}(\mathbf{OCH}_{3})_{3}$
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	$\begin{array}{c} \operatorname{CH}_3 \operatorname{O} \\ \mid & \parallel \\ \operatorname{CH}_2 = \operatorname{C-\!\!\!\!-C} \\ -\operatorname{O}(\operatorname{CH}_2)_3 \operatorname{Si}(\operatorname{OCH}_3)_3 \end{array}$

Table I Types and Chemical Formulas of Silane Coupling Agents

room temperature for 200, 400, 600, 800, and 1000 h. The radiation dose was  $30 \pm 10 \ \mu$ w/cm<sup>2</sup> during the study. UV stability 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.

RE, the ratio of strain at break at the given UV exposure time to the initial strain at break of the sample, was determined using 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 was reported.

The effect of UV irradiation on the samples was also examined by using a Shimadzu IR-470 Infrared Spectrometer. The development of carbonyl absorption (carbonyl index, COI) that appeared during UV exposure of the samples was calculated by using the following formula:

$$\text{COI} = A_{co}/A_{r}$$

where  $A_{co}$  is the amount of carbonyl absorbance at 1715 cm<sup>-1</sup> and  $A_r$  is the absorbance of CH<sub>2</sub> rocking at 722 cm<sup>-1</sup>, used as an internal reference.<sup>4</sup>

### **RESULTS AND DISCUSSION**

### Variations in the Residual Elongation Without UV Stabilizer

RE can be used as a measure of UV stability of the sample in terms of their mechanical property. This section covers the variation of the RE in LDPE, untreated, and different types of SCAtreated 2°-PTFE powder-filled samples.

The RE of pure LDPE decreased to a minimum value within 400 h of UV irradiation; then, after a slight increase in RE around 600 h irradiation, it was followed by a sharp decline until the sample becomes brittle, as given in Figure 1. This type of increase in RE, "recovery period," on exposure to sunlight in the degradation study of LDPE under natural weathering conditions was discussed previously.<sup>4</sup> This recovery period was also mentioned as the predominance of the crosslinking reactions over chain scission and it was usually referred to as "chain stiffening" in artificial weathering studies at the beginning of exposure.<sup>6,7</sup> However, in our work, we observed this chain stiffening after a



**Figure 1** Variation of RE of pure PTFE (P-PTFE) powder-filled LDPE with UV irradiation time.

certain time of irradiation in most of the samples. Therefore, in RE followed by a small increase in UV irradiation, time can be assessed with respect to chain scission and crosslinking reactions. Finally, pure LDPE showed approximately 30% RE at the end of 1000 h of UV irradiation. This simply meant that LDPE lost its flexibility and became brittle. The addition of pure PTFE powder into LDPE slightly affected the RE compared with that of LDPE, especially in 0.5, 1.5, and 3% compositions at the end of the 1000-h exposure, in which they retained 50% of their RE values (Fig. 1). The 2°-PTFE powder-filled LDPE showed even better RE at low concentrations with the exception at 10% PTFE composition, as shown in Figure 2. The 2°-PTFE containing carbonyl or similar chromophore groups, which can absorb UV radiation more readily than pure PTFE may also cause a faster degradation at high concentrations. Indeed, this was observed in 10% 2°-PTFE-filled LDPE. However, UV stability of 2°-PTFE powder filled appeared to be better than pure PTFE. This may be directly due to the fact that the UV energy is absorbed and dissipated by 2°-PTFE before



**Figure 2** Variation of RE of 2°-PTFE (IRR-PTFE) powder-filled LDPE with UV irradiation time.

LDPE absorbs UV light. A 1.5% addition of 2°-PTFE powder occasionally represents about 70% RE stability at 1000 h UV irradiation.

Silane-treated 2°-PTFE samples gave a variety of UV stability within this range of UV irradiation. Although the RE of A-1100 (1°-amine silane)-, A-186 (epoxycyclohexyl silane)-treated 2°-PTFE powder-filled samples decreased sharply just after 600 h of UV irradiation, these samples still represented better RE than that of LDPE (Figs. 3 and 4). Note that in 10% 2°-PTFE, the loss of RE at 1000 h  $\,$ of UV irradiation is worse than pure LDPE. An enhancement in RE was much clearer especially in the A-189 (mercapto silane)-treated samples which showed the best UV stability among all other silane-treated 2°-PTFE powder-filled samples (Fig. 5). The RE in all compositions of A-189-treated 2°-PTFE samples was found to be higher than 70% at the end of 1000 h of UV irradiation. It is known that sulfur (mercapto)-containing compounds can be used as photo-stabilizer agents.<sup>2</sup> Mercapto groups (SH) in this type of silane coupling agent therefore strongly enhanced the UV stability of the samples compared with the other types of silane coupling agents. Other types of silane coupling agents (as

given in Table I) resulted in lowering the UV stability of the samples, as can be followed from the Figure 6 drawn for A-1120 (2°-amine silane)-treated 2°-PTFE. These silanes, A-172 (vinyl silane), A-174 (methacrylo silane), and A-187 (epoxy silane) caused a fast decrease in RE, even lower than 80% RE. Because A-189 silane-treated samples gave the best UV stability, the mixed silane treatments of A-1100-189, A-187-189, and A-1120-189 were also studied. Yet, no further UV stability measured as RE was seen in these samples. The changes in RE of the samples at 1000 h UV irradiation are summarized in Figure 7. It is clear that pure PTFE for 0.5, 1.5, and 3% compositions in LDPE enhance the UV stability in terms of RE compared with LDPE. A further increase in RE is observed in 2°-PTFE-filled LDPE. Only A-1100 and A-186 SCA-treated 2°-PTFE powder-filled samples have shown good stabilization and A-189 silane represents the best RE among all types of silane treatments. The other types of silane treatments (A-1120, A-187, A-172, and A-174) depressed the RE, hence that is the UV stability of the samples. Photo-oxidative degradation may be said to be accelerated due to the chem-



**Figure 3** Variation of RE of A-186 silane-treated 2°-PTFE powder-filled LDPE with UV irradiation time.



**Figure 4** Variation of RE of A-1100 silane-treated 2°-PTFE powder-filled LDPE with UV irradiation time.

ical structure of these types of silane coupling agents.

# Variation in the Residual Elongation in the Presence of UV Stabilizer

The stress-strain curves of compositions in the presence of HALS, UV stabilized, and nonstabilized samples of A-189 with 2°-PTFE and pure LDPE are given in Figure 8. It is clear that the addition of 0.5% by weight HALS-944 UV stabilizer showed, to a certain extent, a synergetic effect on the ultimate mechanical property of LDPE. Similar kinds of improvements were also found in the other types of SCA-treated samples when they were combined with UV stabilizer compared with nonstabilized ones.

The variation of RE in the HALS-944-stabilized 2°-PTFE powder-filled LDPE with respect to time of UV irradiation is given in Figure 9. RE of 944-stabilized LDPE is highly improved. There is only about 5% loss in RE in pure LDPE at the end of 1000 h of UV irradiation. Note that 2°-PTFE powder-filled LDPE stabilized by 944 represented better RE than that of only HALS-stabilized LDPE. Almost higher than 100% RE was retained at the end of UV irradiation of these samples. This increase in RE cannot be simply discussed with respect to chain stiffening but also possible interactions between HALS and 2°-PTFE need to be considered. Therefore, it is very obvious that 2°-PTFE powder has shown a synergetic effect on UV stability with HALS stabilization.

Although A-172 and A-1120 silane-treated samples were shown to lose their RE faster (Figs. 6 and 7), the addition of 944 UV stabilizer in these samples appeared to improve RE, even better than HALS stabilization LDPE. The variation of the RE in bar graphics in HALS stabilized untreated, and SCA-treated 2°-PTFE powder-filled LDPE at 1000 h of UV irradiation is summarized in Figure 10. As mentioned, A-189 silane showed the best RE (Fig. 5) but when HALS and A-189 were combined it caused a sudden loss of RE after 750 h compared with other silane-HALS combinations (Fig. 11). This may be due to the inharmonious interactions between mercapto groups in A-189 silane coupling agent and hindered amine



**Figure 5** Variation of RE of A-189 silane-treated 2°-PTFE powder-filled LDPE with UV irradiation time.



**Figure 6** Variation of RE of A-1120 silane-treated 2°-PTFE powder-filled LDPE with UV irradiation time.

light stabilizer. Although RE of HALS stabilized untreated 2°-PTFE powder-filled samples reduced with increasing the filler content, they still



**Figure 8** Stress-strain curves of UV stabilized, 944, and nonstabilized samples of LDPE, 1.5% 2°-PTFE–filled LDPE and A-189 silane-treated 2°-PTFE–filled LDPE.

had higher RE in all compositions than only HALS-stabilized LDPE. Therefore, it is clear that 2°-PTFE powder shows a substantial synergetic



**Figure 7** Variation of RE of pure LDPE, silane-treated and untreated 2°-PTFE powder-filled samples at 1000 h of UV irradiation time.



**Figure 9** Variation of RE of HALS, 944, stabilized 2°-PTFE powder-filled LDPE with UV irradiation time.

**Figure 11** Variation of RE of HALS, 944, stabilized A-189 silane-treated 2°-PTFE powder-filled LDPE with UV irradiation time.



**Figure 10** Variation of RE of HALS, 944, stabilized pure LDPE, silane-treated and untreated 2°-PTFE powder-filled samples at 1000 h of UV irradiation time.



**Figure 12** Variation of COI of 2°-PTFE powder-filled LDPE with UV irradiation time.

effect when it is used with HALS. Whereas the presence of SCAs of the A-189 kind shows a slight adverse effect on the UV stability, an enhanced UV stability is observed by HALS stabilization in the presence of A-1120, A-187, A-172, and A-174 SCAs and this stabilization appears to be better than HALS alone at the end of the 1000 h of UV irradiation.

## Variation in the Carbonyl Index Without UV Stabilizer

The most important spectral change due to UV irradiation occurs in the carbonyl region of LDPE with maximum absorption around  $1715 \text{ cm}^{-1}$ . No simple correlation was found to exist between the development of carbonyl groups and RE.<sup>4</sup> Although there exist some fluctuations in RE (particularly in recovery periods), carbonyl index (COI) first develops gradually and then this slow increase is usually found to be autoaccelerated with a sudden jump. A sharp increase in COI mostly corresponds to an inevitable decrease in

the RE. The variation of COI in untreated 2°-PTFE powder-filled LDPE with respect to UV irradiation time is given in Figure 12. There was almost no difference in the COI variation of filled samples compared to that of LDPE, except 10% 2°-PTFE which showed higher COI than the others. This behavior was also observed in the pure PTFE powder-filled samples.

The extent of COI of A-189 silane-treated samples against the irradiation time is given in Figure 13. The development of COI in A-189 (mercapto silane)-treated samples were depressed. This decrease in COI appears to be in good agreement with the RE results (Fig. 5). A-1100 and A-186 silane treatments also depressed the oxidative degradation of the samples but not as much as the A-189 treated samples. The developments in COI with UV irradiation time for the other SCAs and their mixed compositions were found to be faster than pure LDPE. The type of SCA, with a particular active group, plays an important role in oxidative degradation. Those containing mercapto (A-189), 1°-amine (A-1100), epoxycyclohexyl



**Figure 13** Variation of COI of A-189 silane-treated 2°-PTFE powder-filled LDPE with UV irradiation time.



**Figure 14** Variation of COI of pure LDPE, silane treated and untreated 2°-PTFE powder-filled samples at 1000 h of UV irradiation time.

(A-186) with 2°-PTFE seem to provide a better protection against oxidative degradation whereas the rest cause a slight but noticeable increase in oxidative degradation. Actually, the variation of RE at 1000 h of UV irradiation showed a similar conclusion. The COI variations of the samples at the end of 1000 h of irradiation are summarized in Figure 14. Finally, one of the reasons why the untreated 2°-PTFE addition shows slightly higher COI than the pure PTFE-filled samples and LDPE is because PTFE already has carbonyl groups in its structure that is formed upon  $\gamma$ -irradiation of PTFE under oxygen atmosphere.

# Variation in the COI in the Presence of UV Stabilizer

When the COI scales between nonstabilized and HALS-stabilized samples were compared with each other, there was at least 10 times reduction in the later case as was expected. COI variation of HALS-stabilized untreated 2°-PTFE powderfilled LDPE is given in Figure 15. The COI of HALS-2°-PTFE samples were found to be greater than the HALS-stabilized pure LDPE, although their RE showed contrary results (Fig. 9). This may have arisen from enhancement in oxygen diffusivity, solubility in this sample due to particles of 2°-PTFE and the inherent carbonyl groups in the 2°-PTFE. The variation of COI in the HALS-stabilized samples at the end of 1000 h of UV irradiation is summarized in Figure 16. Similar results were also obtained in HALS-stabilized

A-172 and A-174 silane-treated 2°-PTFE powderfilled samples (compare Figs. 10 and 16). Although RE of HALS-stabilized A-189 silane-



**Figure 15** Variation of COI of HALS, 944, stabilized 2°-PTFE powder-filled LDPE with UV irradiation time.



**Figure 16** Variation of COI of HALS, 944, stabilized A-189 silane-treated 2°-PTFE powder-filled LDPE with UV irradiation time.

treated samples was lower than other samples (see Figs. 10 and 11), the development of COI of both nonstabilized (Fig. 14) and HALS-stabilized A-189-treated samples was found to be lower, especially in 1.5 and 3% compositions (Fig. 17, see also Fig. 16). However, there still exists no oneto-one correlation between RE and COI in this type of sample. This may be explained as mentioned before by a partial antagonism between mercapto silane and HALS. Similar results are also obtained for HALS-stabilized A-187, A-1100, and A-1120 silane-treated samples as shown in Figure 17. Whereas a depression exists in COI of A-1100, A-1120, A-187, and A-189 HALS-stabilized silane-treated samples, COI of HALS-stabilized A-172 and A-174 silane-treated samples show a faster development.

### **CONCLUSIONS**

The addition of 2°-PTFE, lower than 10% by weight, appeared to improve the UV stability of LDPE. SCA treatments resulted in further improvement, particularly the mercapto reactive silane-containing SCA (A-189). In all cases, we observed an increase in the RE after a certain time of UV irradiation. This could be attributed to the chain stiffening effect. HALS, as expected, added an extra stability in 2°-PTFE– filled LDPE and when HALS and SCAs coexisted, a better UV stability was observed with



**Figure 17** Variation of COI of HALS, 944, stabilized pure LDPE, silane treated and untreated 2°-PTFE powder-filled LDPE at 1000 h of UV irradiation time.

an exceptional mixture of HALS and A-189 SCA which possessed an antagonistic effect. Finally, the development of COI and the variation in the RE showed no direct correlation in assessing and following the UV stability of LDPE films.

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