

Effect of Film Draw Temperature (FDT) on Physical Properties of Polyethylene

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ABSTRACT: Thin polymer films (0.06 mm thick) were prepared with LLDPE (coded as A) and LDPE (coded as D) at different film draw temperatures (FDTs) from 5 to 65°C. There was about a 42% enhancement of the tensile strength when the LLDPE film was drawn at 45°C and LDPE at 35°C and the ultimate elongation increased between 14 and 32%. When various additives were incorporated into these resins A and D, the tensile strength slightly decreased, but the ultimate elongation increased. Films which attained the

highest tensile properties showed the maximum resistance against degradation by natural outdoor weathering. Although irradiation of these films by a gamma source caused reduction of their tensile properties, there was a general tendency of resisting this reduction with increase of the FDT. © 2003 Wiley Periodicals, Inc. *J Appl Polym Sci* 88: 459–465, 2003

Key words: thin films; radiation; additives; HALS; thermal properties

INTRODUCTION

Polymer is a versatile material. Its diverse applications are increasing daily in different fields as technology advances. Thus, it is imperative to design new formulations to match the desired applications of the polymer. Various additives are used with their variable contents during the compounding process of the polymer. These additives thus influence the properties of the polymer. Some additives are used as antioxidants and to add heat resistant to protect the polymer against the effects of UV radiation and the heat of the sun during their outdoor applications. Based on their properties and manufacturing techniques, polyethylene, particularly commercial polyethylene, is classified as high-density polyethylene (HDPE), low-density polyethylene (LDPE), linear low-density polyethylene (LLDPE), and ultralow-density polyethylene (ULDPE).^{1,2} These polymers also have different specific applications because of their different physical properties induced by different manufacturing techniques.^{3–5} Polyethylene films are generally prepared by melting their pellets in an extruder and the melt is then drawn into a film through a set of stainless-steel drums cooled to a desired temperature by air or water circulation through the drums. Physical and mechanical properties of these films are expected to be largely

dependent on the rate and temperature of the film drawn as well as on the additives incorporated into the system during the compounding process.^{6–12}

The present article deals with the effect of the film draw temperature (FDT) and various additives, particularly antioxidants (both primary and secondary antioxidants), light stabilizers (HALS), and ultraviolet radiation absorbers (UVAs) of the sun on the physical properties of thin PE films of two different resins: LLDPE (coded as A) and LDPE (coded as D).

EXPERIMENTAL

Materials

Two types of basic resins, namely, LLDPE (A) and LDPE (D), were obtained from the local market. The antioxidants (AOs) Irganox 1010 and Irgafos 168, hindered amine light stabilizers (HALS) Tinuvin 494 and Chemiasorb 944 and UV absorber (UVA) Chemiasorb 81 were obtained from the Ciba Speciality Chemicals Co. (Basel, Switzerland), and the secondary antioxidant (SAO) Alkanox TNPP was procured from the Great Lakes Chemical Co. (Persan, France).

Methods

Films preparation

Thin polyethylene films (0.06 mm) were prepared with masterbatch materials which were prepared earlier by mixing appropriate additives at 5 phr with a basic resin LLDPE (A) or LDPE (D) according to formulations of Table I. The mixing was done in an

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TABLE I
Formulations for LLDPE (Code A) and LDPE (Code D) Using Various Additives

Code of films	Irganox 1010 (%) PAO	Irgafos 168 (%) SAO	Alkanox TNPP (%) SAO	Chimassorb 944 (%) HALS	Tinuvin 494 (%) HALS	Chimassorb 81 (%) UVA
A or D	—	—	—	—	—	—
A1 or D1	0.05	0.1	—	0.08	—	—
A2 or D2	0.05	—	0.1	0.08	—	—
A3 or D3	0.05	0.1	—	—	0.08	—
A4 or D4	0.05	—	0.1	—	0.08	—
A5 or D5	0.05	0.1	—	—	—	0.08
A6 or D6	0.05	—	0.1	—	—	0.08

PAO, primary antioxidant; SAO, secondary antioxidant; HALS, hindered amine light stabilizer; UVA, ultraviolet radiation absorber.

extruder (Model L/D 19/25) attached to a Plasti-Corder (Model PL 2200) from Brabender Co. (Germany). Compounding was carried out in the extruder at 200–230°C under a nitrogen gas flush (20 mL/min). Films were drawn with a speed of 2 m/min at a desired temperature from 5 to 65°C controlled by a thermostated water-cooling unit attached to a Univex take-off machine (Model D 47055) of the Brabender Co.

Natural outdoor weathering and irradiation

PE films were exposed up to 165 days in the sun at an angle of 45° toward south for natural outdoor weathering as per ASTM D1435. Films were also irradiated with a ⁶⁰Co gamma cell of Nordion MDS Int. (Model GC 220; Canada) under nitrogen gas at total doses of 20–200 kGy at a dose rate of 6.5 kGy/h.

Tensile properties measurement

Tensile strength (TS) and ultimate elongation (Eb) of all the PE films were determined as per ASTM D882 using a universal testing machine from Instron Co. (Model 4505; UK) using a gauge length of 2.5 cm with a speed of 50 cm/min.

RESULTS AND DISCUSSION

Effect of FDT

Thin films of LLDPE (A) were prepared at various FDTs from 5 to 65°C, while LDPE (D) films were prepared between 20 and 65°C, because it was not possible to draw PE film of LDPE (D) at a temperature lower than 20°C. The tensile properties (TS and Eb) of these films are plotted in Figure 1 against the FDT. It is clearly observed that the TS starts increasing from the FDT at 20°C, reaches a maximum at 35°C for LDPE and 45°C for LLDPE films, and then decreases to a minimum which is lower than the value obtained at 20°C in each case. There is not much change in the Eb

with the FDT. In fact, it shows slight enhancement of elongation with increase of the FDT. Higher TS values at 35 and 45°C for films of LDPE and LLDPE, respectively, may be attributed to the fact that the respective polymer chain is capable of proper orientation of its backbone chain with respect to its respective monomer units and side branches at that particular FDT, after which this orientation becomes more fragile at higher temperature due to softening of the polymer matrix. On the other hand, since the polymer backbone chains are straightened longitudinally at higher temperatures with proper arrangement of their different monomer and side units, Eb is not much affected by increase of the temperature.

Effect of additives on FDT

Six different types of PE films were prepared with each of LLDPE and LDPE according to the formulations in Table I. The tensile properties of these films

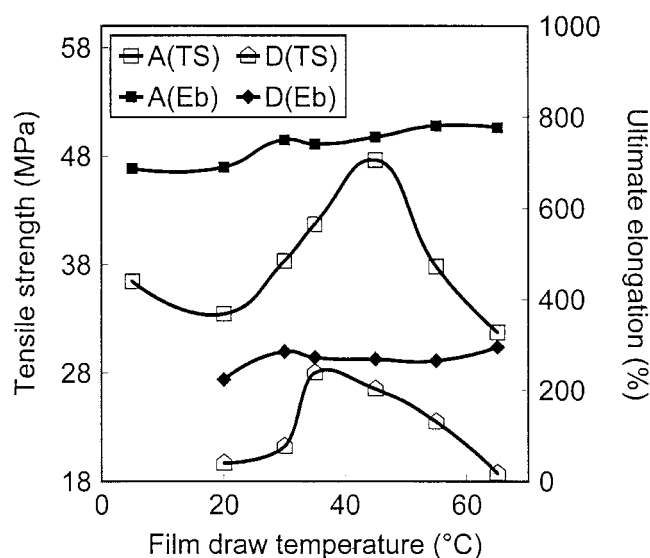


Figure 1 Tensile properties of PE films of LLDPE (A) and LDPE (D) as a function of the FDT.

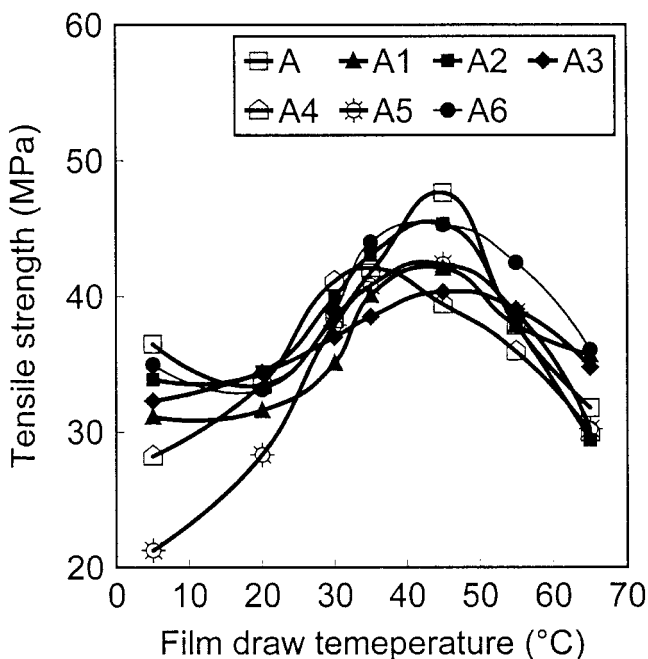


Figure 2 TS of various PE films of LLDPE (A) prepared at different FDTs.

are plotted in Figures 2–5. The TSs decreased as a result of the incorporation of additives in each case. These plots show a maximum TS at 35°C for LDPE (Fig. 3) and at 45°C for LLDPE (Fig. 2). The decrease in the TS is due mainly to the filling effect of these additives in the polymer matrices, because the additives are not monomers and, thus, cause hindrance during orientation of the polymer backbone chains at

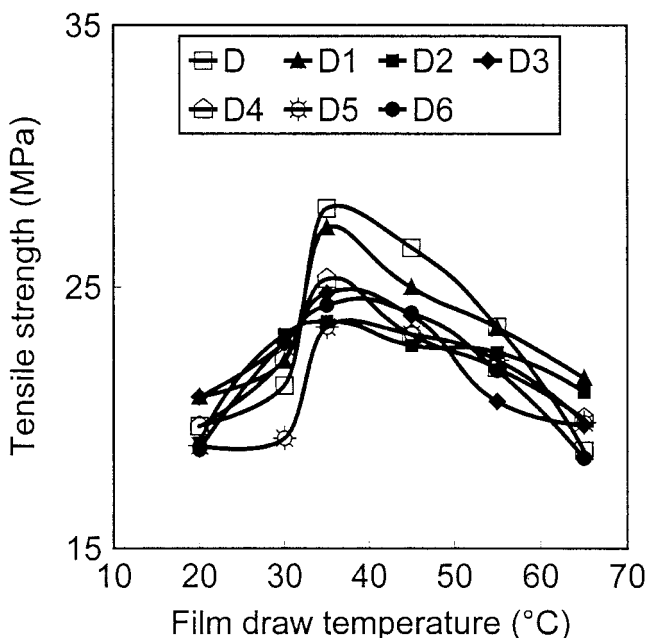


Figure 3 TS of various PE films of LDPE (D) prepared at different FDTs.

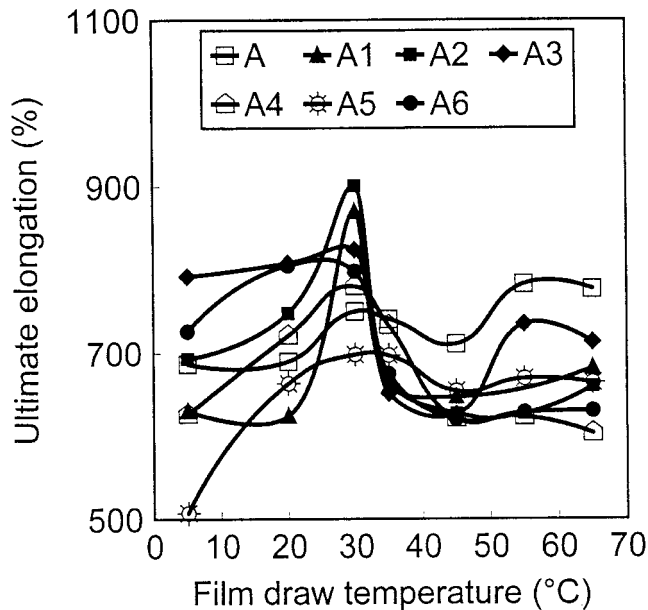


Figure 4 Eb of various PE films of LLDPE (A) prepared at different FDTs.

the time of extrusion and film draw. On the other hand, Eb increased in the presence of additives in each case except with films of A5 (Fig. 4). The PE is composed of several long chains in a polymer matrix; in these matrices, all the additives incorporated remain scattered in between the long chains. The polymer backbone chains thus remain longitudinally oriented and this helps to impart a higher elongation to the polymer matrix.

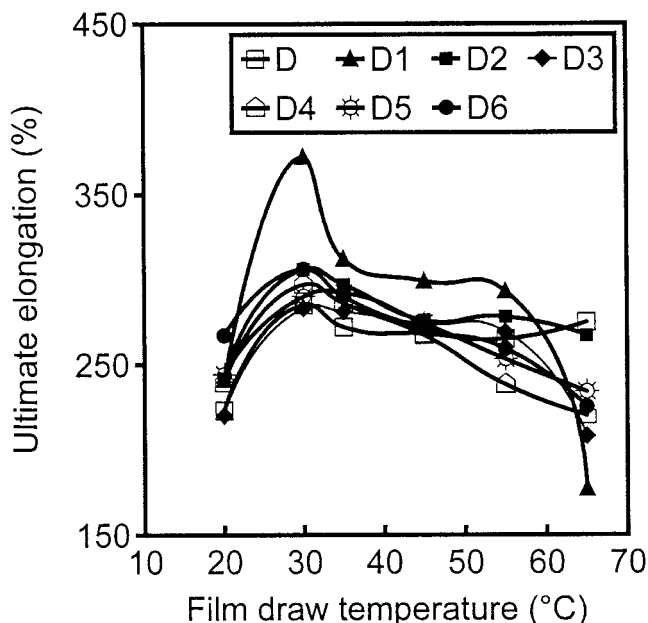


Figure 5 Eb of various PE films of LDPE (D) prepared at different FDTs.

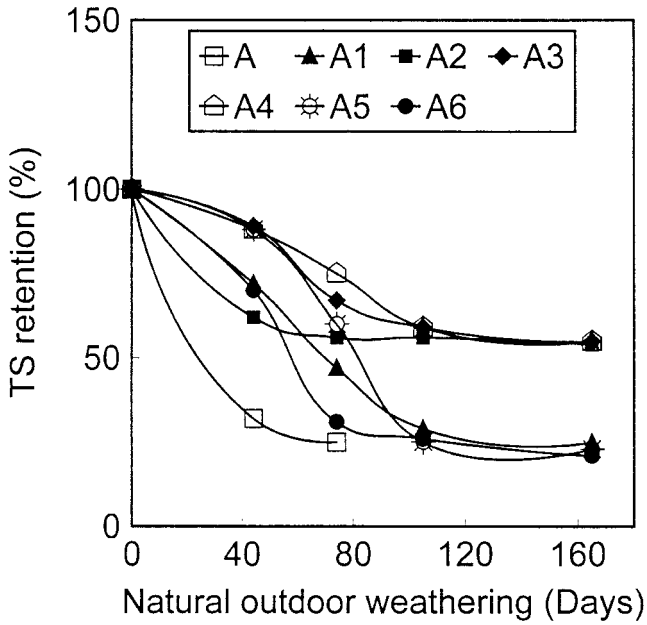


Figure 6 Retention of TS of LLDPE (A) films made at FDT of 45°C after natural outdoor weathering for various durations.

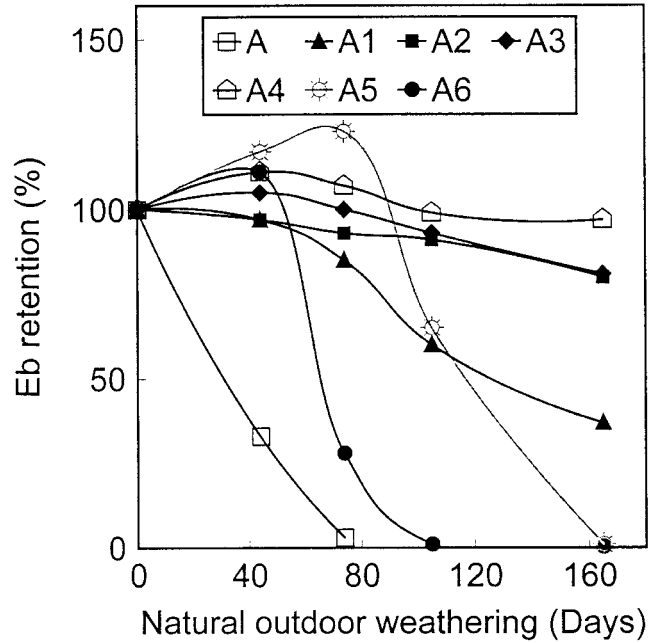


Figure 8 Retention of Eb of LLDPE (A) films made at FDT of 45°C after natural outdoor weathering for various durations.

Natural outdoor weathering

Various PE films of LLDPE (A) and LDPE (D) were prepared at an FDT of 45°C following the formulations in Table I and were subjected to natural outdoor weathering for different periods in the sun as per ASTM D1435. The retention of their TSs and Eb's after natural outdoor weathering is shown in Figures 6–9. Films of pure resins A and D were degraded quickly

during natural outdoor weathering. More than 50% of their tensile properties (TS and Eb) were destroyed in less than 40 days of their exposure in the sun, whereas films containing additives showed much resistance to such degradation. In fact, retention of the tensile properties of films A2, A3, and A4 remained higher than 50% (Figs. 6 and 7) even after 165 days of natural outdoor weathering which was carried out from June to November. But retention of the TS of the D2, D3,

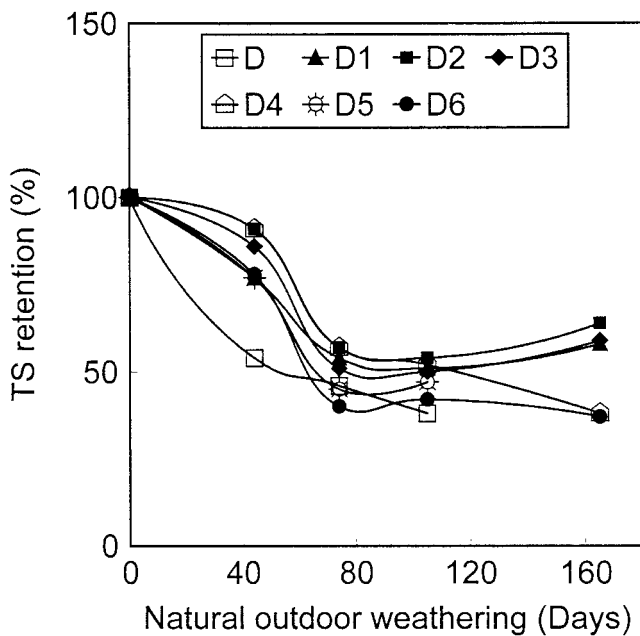


Figure 7 Retention of TS of LDPE (D) films made at FDT of 45°C after natural outdoor weathering for various durations.

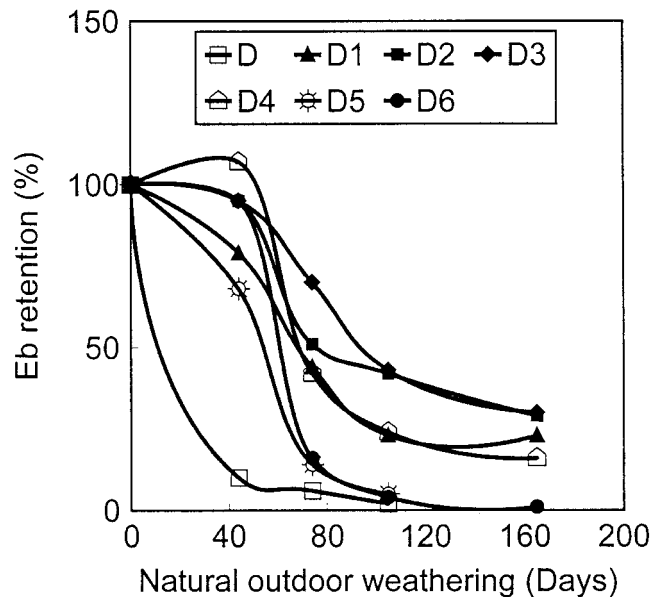


Figure 9 Retention of Eb of LDPE (D) films made at FDT of 45°C after natural outdoor weathering for various durations.

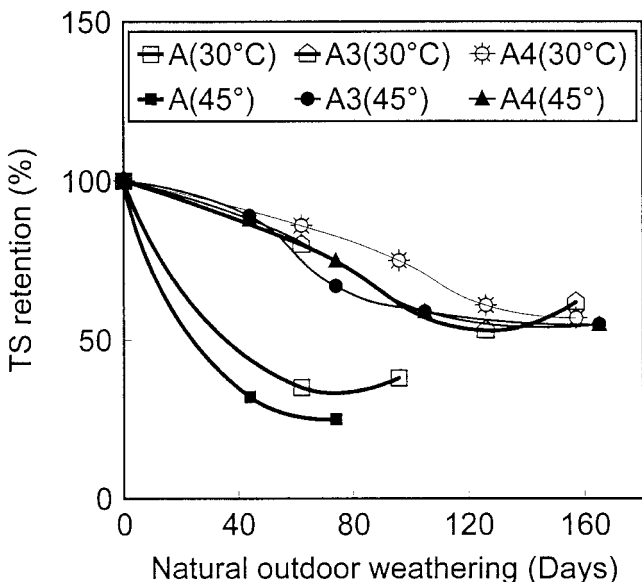


Figure 10 Comparison of retention of TS of LLDPE films made at FDTs of 45 and 30°C after natural outdoor weathering for various durations.

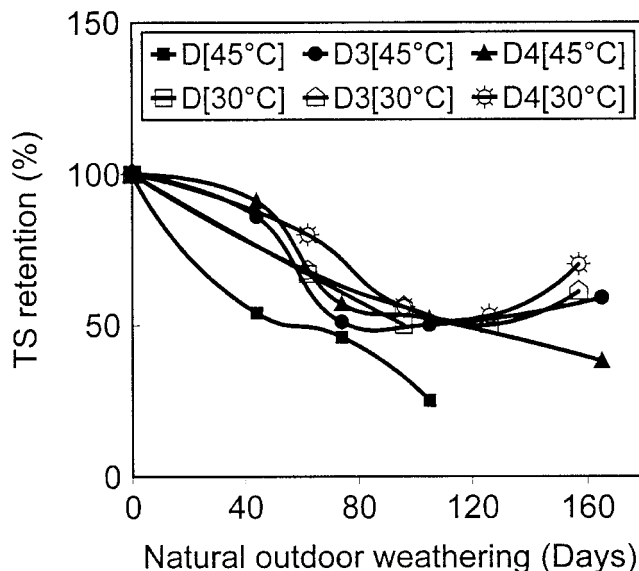


Figure 11 Comparison of retention of TS of LDPE (D) films made at FDTs of 45 and 30°C after natural outdoor weathering for various durations.

and D4 films was almost 50% within 100 days of the outdoor weathering test (Fig. 8) and that of Eb of these films was lower than 50% within 80 days of the outdoor weathering (Fig. 9). This difference among various films of resins A and D in resisting degradation due to outdoor weathering is related mainly to the intrinsic character of the pure resins LLDPE and LDPE. LDPE is highly branched compared to LLDPE and the latter contains short branches and greater tensile properties with higher environmental stress-cracking resistance.

Elongation of some of the films of resins A and D showed slight enhancement of more than 100% at the initial weathering period (Figs. 8 and 9). This enhancement may have been due to scorching sun rays observed during summer in Riyadh (Saudi Arabia). It is observed from Figures 8 and 9 that films of A2, A3, A4, D2, D3, and D4 exhibited maximum resistance against outdoor weathering degradation. It is known from Table I that formulations 3 and 4 contain the same additive (Tinuvin 494). This means that the films of A3, A4, D3, and D4 contain the same HALS (Tinuvin 494). Films of A3, A4, D3, and D4 were then prepared at an FDT of 30 and 45°C and were subjected to natural outdoor weathering. The results are plotted in Figures 10–13.

The TS retention is shown in Figures 10 and 11 for A and D, respectively, and Figures 12 and 13 represent the Eb retention of these films, respectively. The TS retention is observed to be slightly higher for both LLDPE (A) and LDPE (D) films when they are prepared at an FDT of 30°C instead of 45°C. Almost similar behavior of the TS retention is also observed when additives are incorporated into these resins (Figs. 10

and 11). On the contrary, the Eb retention of A3, A4, D3, and D4 films is much higher at an FDT of 45°C than at an FDT of 30°C (Figs. 12 and 13). This is clear evidence that these films prepared at an FDT of 45°C are more capable of resistance to outdoor weathering degradation than are films prepared at an FDT of 30°C, particularly, the A3 and A4 films show much a higher resistance in this condition (Fig. 12).

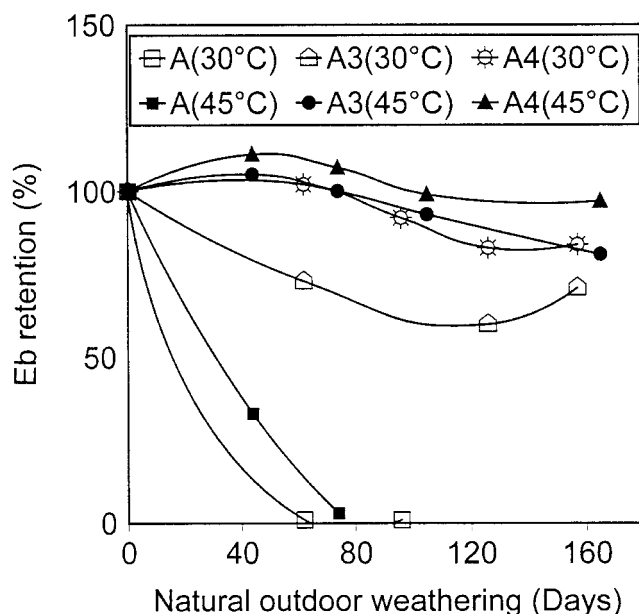


Figure 12 Comparison of retention of Eb of LLDPE films made at FDTs of 45 and 30°C after natural outdoor weathering for various durations.

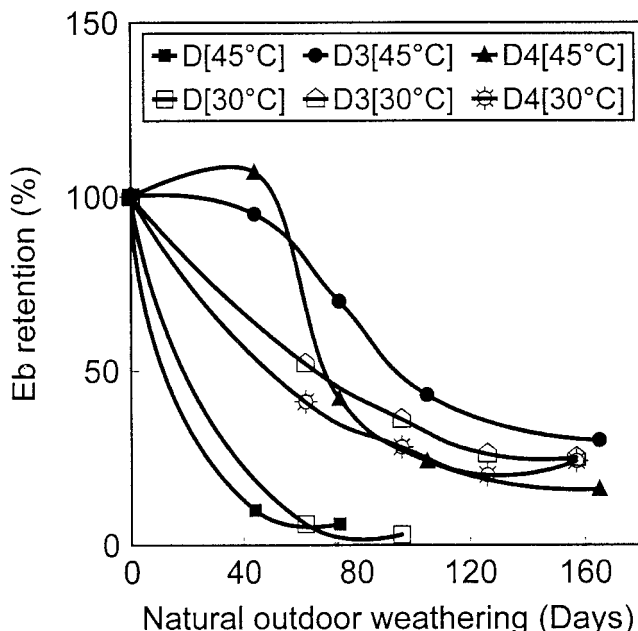


Figure 13 Comparison of retention of Eb of LDPE (D) films made at FDTs of 45 and 30°C after natural outdoor weathering for various durations.

Radiation-induced crosslinking

PE films of both LLDPE and LDPE prepared at various FDTs from 5 to 65°C were irradiated with a gamma source at different doses from 20 to 200 kGy. Tensile strengths are plotted against the FDT as a function of the irradiation and are shown in Figures 14 (LLDPE) and 15 (LDPE). LLDPE films of an FDT of 5 and 20°C showed a slight increase in the TS when irradiated

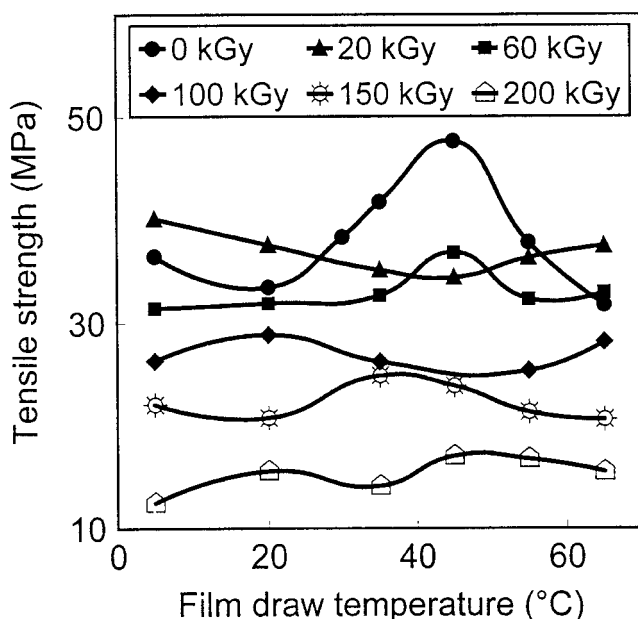


Figure 14 TS of LLDPE (A) films prepared at different FDTs as a function of irradiation dose.

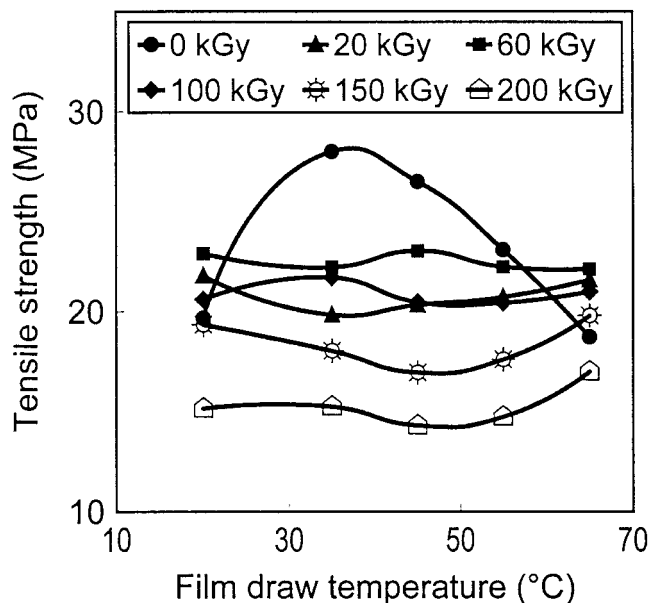


Figure 15 TS of LDPE (D) films prepared at different FDTs as a function of irradiation dose.

with 20 kGy and then the TS slowly decreased with increase of the irradiation dose. The decrease seems to remain similar for all films prepared at all FDTs. The additives (antioxidants, UV absorbers, and HALS) used may play a significant role in protecting the polymer against the effect of UV radiation, that is, these radicals might react with polymer radicals to result in reduction in the crosslinking and chain scission in the polymer matrix with increase of the irradiation dose. The enhanced crosslinking density may not necessarily increase the tensile properties of the polymer, because of the radiation-induced scission of the long polymer backbone chain as well as the brittleness character of the polymer induced at a higher crosslinking density. However, in some cases, there is some trend of enhancement of the TS with increase of the FDT (Fig. 14). The enhancement of the TS is more prominent with LDPE films from an FDT of 45°C upward at all irradiation doses. The gel content, an index of crosslinking density, was determined for various films and a typical plot is shown in Figure 16 for the LDPE system only at an FDT of 30°C. The results indicate that radiation-induced crosslinking took place in the films.

CONCLUSIONS

The following conclusions can be drawn:

1. There is an effect of the FDT on the tensile properties of PE films prepared at different FDTs. There is about a 42% enhancement of the TS with both LLDPE and LDPE films when the LLDPE film was prepared at an FDT of 45°C and LDPE

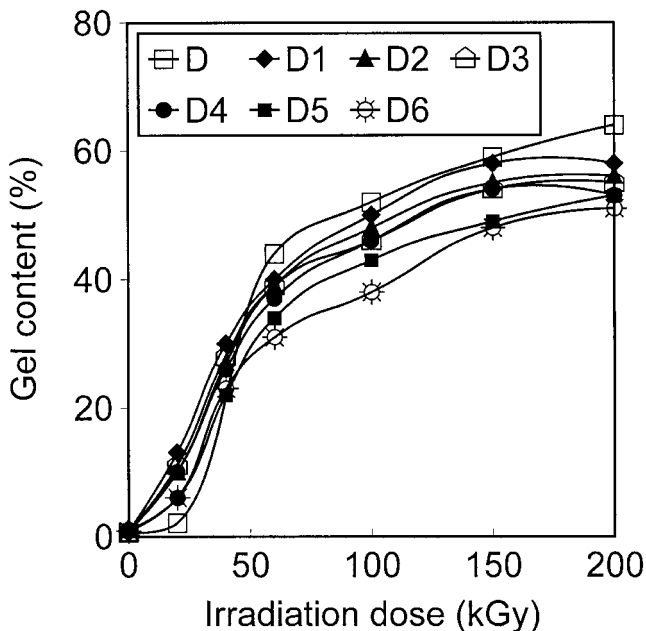


Figure 16 Gel content of radiation-crosslinked PE films of LDPE (D) at 30°C with different doses.

prepared at an FDT of 35°C. Enhancement of the Eb is 14% for LLDPE and 32% for LDPE (Fig. 1).

- When additives are incorporated into pure resins, the enhancement of the TS is slightly decreased (Figs. 2 and 3), but the Eb is generally increased (Figs. 4 and 5).
- There is a general tendency of showing more resistance against degradation of natural outdoor weathering by films prepared at an FDT of 45°C for LLDPE and 35°C for LDPE (Figs. 10–13).

- TS decreases for both LLDPE and LDPE films with increase of the irradiation dose, but there is also a general tendency of slightly enhancing the tensile property with increase of the FDT (Figs. 14 and 15).

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