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Electron beam irradiation of carbon black filled linear low-density polyethylene

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Radiation as a technology has now been commercialized for over twenty five years. Radiations are used efficiently and economically for the production of new or modified polymers [1]. Radiation crosslinking can be applied to a great number of plastics, thermoplastics, elastomers and thermoplastic elastomers. Effects of irradiation on polyethylene have been studied extensively by several researchers [2–4]. Polyethylene undergoes considerable changes as a result of radiation induced cross-linkings. Fillers such as carbon black [5], glass bead [6], wax [7] and kaolin [8] filled polyethylene have been studied. The influence of radiation crosslinking on positive temperature coefficient (PTC) and negative temperature coefficient (NTC) on carbon black filled polyethylene also has been studied [7]. However, there is no study reported on electron beam irradiation of carbon black filled LLDPE. This article reports on the influence of radiation crosslinking and carbon black filler loading on the properties of LLDPE with respect to changes in mechanical properties and phase morphology.

Blends of LLDPE were prepared by melt blending in a Haake Rheomix at 140 °C, with a rotor speed of 50 rpm and 15 min mixing. LLDPE was charged into the mixing chamber and mixed for 2 min. Carbon black was then added, and the blending was continued for another 13 min. The blends were taken out and were compression molded into 1 mm thick sheets under a pressure of 150 MPa in an electrically heated hydraulic press at 140 °C for 3 min. The sheets obtained were then irradiated with electron beam (using 3 MeV electron-beam machine, manufactured by NHV, Japan) for 250 kGy.

Tensile test on the samples were done on dumbbell-shaped samples according BS 6746 standard using a Toyoseiki model Strograph-RI with a load cell of 1 kN at room temperature in accordance with ASTM D638 and a cross-head speed of 50 mm/min. The tests were carried out on seven sample pieces for each blend, and the readings were calculated from the best five samples. For morphological studies, the molded samples were fractured under liquid nitrogen. The fractured surfaces were coated with gold and examined under scanning electron microscope (SEM).

Variation of mechanical properties with the filler content is shown in Fig. 1 for both irradiated and unirradiated samples. Fig. 1 shows an increase in the tensile strength up to 10% carbon black loading for unirradiated samples. It indicates that the optimum carbon black loading in LLDPE is about 10 v%. The degree of reinforcement of LLDPE blends increases with filler loading and the extent of polymer–filler interaction. The reduction of tensile strength after 10 v% loading is due to less wetting of the black agglomerated particles in the matrix by the polymer chains [5]. However, the tensile strength decreases with increasing filler loading for irradiated samples. Electron beam irradiation induced chain scission and crosslinking in LLDPE phase [4] and at a large dose of 250 kGy, a high crosslink density is expected to form. The resulting cross-linked network reduces the migration of carbon black in the polymer matrix [5]. The influences of carbon black loading and electron beam crosslinking on the sample modulus at 100% are shown in the Fig. 2. An increase in modulus with filler loading is observed for both irradiated and unirradiated carbon black filled LLDPE, which indicates increased stiffness due to the rising filler content. A change of deformation behavior from ductile to more brittle occurred at higher contents of carbon black. At fixed filler loading, the modulus values for irradiated samples are higher than for

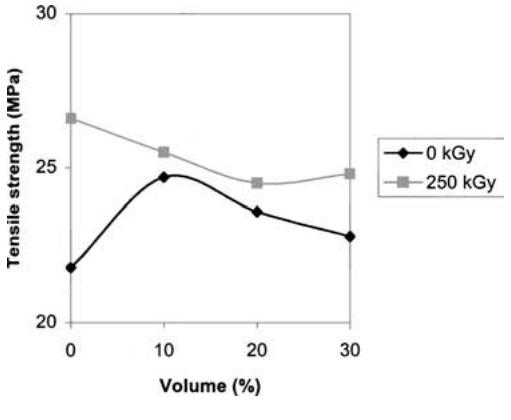

Figure 1 Tensile strength of carbon black filled LLDPE.

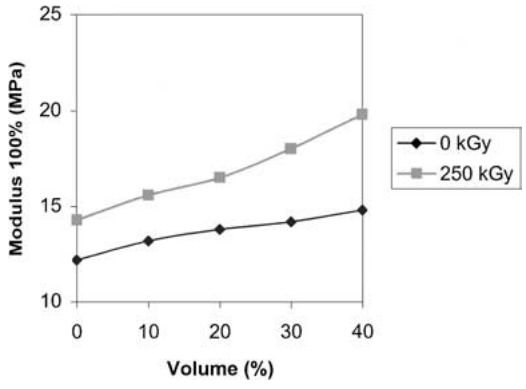

Figure 2 Modulus 100% of carbon black filled LLDPE.

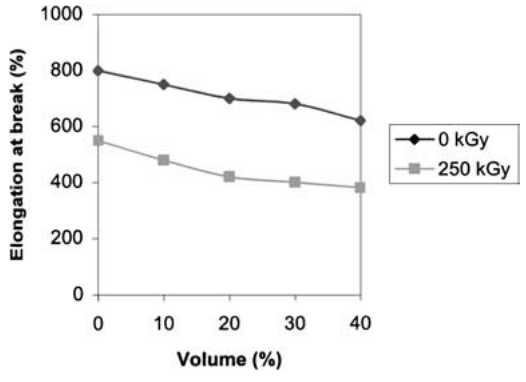

Figure 3 Elongation at break of carbon black filled LLDPE.

unirradiated samples, which is due to crosslinking in the polymer phase. Fig. 3 shows the decrease in elongation at break with increasing filler loading for both irradiated and unirradiated samples. The increased interaction between the polymers via the filler particles and enhanced by the radiation induced crosslinking causes the changes. Elongation at break for irradiated samples is lower than for unirradiated samples indicating a higher crosslinks density in the irradiated samples. Tensile strength and modulus at 100% strain are also higher for electron beam irradiated than the unirradiated samples.

Fig. 4a–f show the SEM photographs of the fractured morphology of the carbon black filled LLDPE. Fig. 4a and b shows the morphology of pure LLDPE and carbon black filled LLDPE, respectively. It is observed that at 10% loading, carbon black particles are well distributed in the polymer phase as shown in Fig. 4b. Therefore the tensile strength of LLDPE will be significantly improved. However, at higher filler loading which is shown in Fig. 4c, the carbon black particles agglomerated and dispersed inhomogeneously, which causes a reduction in the tensile strength. It is also observed that the network increases with 40 vol% of carbon black. It is believed that the carbon black particles were encapsulated by polymer matrix at low filler content. However at higher carbon black content, interaction between filler and filler is stronger than interaction

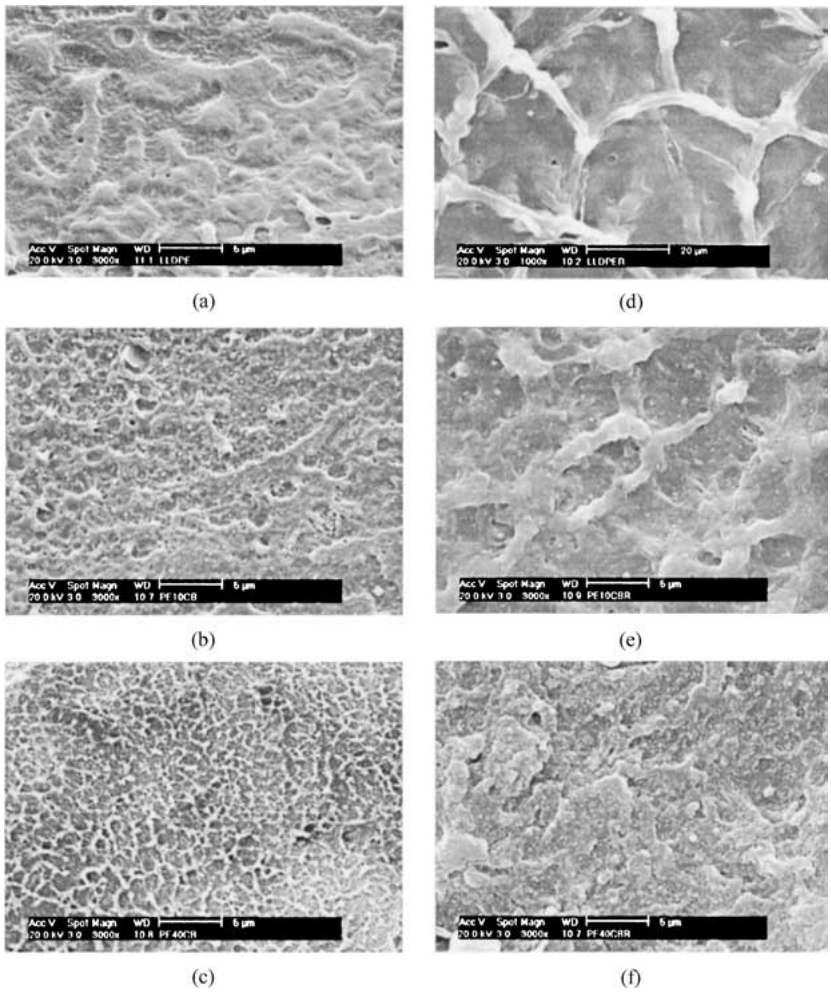

Figure 4 (a) pure LLDPE, (b) LLDPE (10%CB), (c) LLDPE (40%CB), (d) LLDPE (250 kGy), (e) LLDPE (10%CB, 250 kGy), and (f) LLDPE (40%CB, 250 kGy).

between polymer and filler and therefore filler networks are dominant in the polymer matrix.

In the Fig. 4d, the size of cavities of the irradiated LLDPE is smaller than those in pure LLDPE. It is suggested that crosslinking network formation occurs when LLDPE is exposed to electron beam irradiation. When the filled LLDPEs are irradiated at 250 kGy (Fig. 4e and f), the network takes place mainly in the amorphous phase. Consequently, the carbon aggregate or agglomerates are bound to the three dimensional networks of the irradiated samples. On the other hand, with radiation dose 250 kGy, many more carbon black particles were bound to the crosslinking networks, preventing the migration of the carbon black particles. It is due to formation of chemical interactions between filler surface and the polymer matrix. These interactions arise during the electron beam crosslinking of the matrix. It is assumed that radicals are formed by radiation and thermal mixing of the polymer and filler. Thus radicals are formed on the filler surface, which can combine with the polyethylene macroradicals. Grafting of the polyethylene macromolecules onto the filler surface results in the improvement of the wetting of the polymer surface and increase in the polymer-filler adhesion on the phase boundaries. The mechanism suggested above is responsible for the observed effect of electron beam irradiation on the mechanical properties of carbon black filled LLDPE. SEM micrograph also showed that the smooth structure is slowly transformed from ductile into less flexible and rigid structure (brittle) on increasing the carbon black filler loading and radiation dose [5]. In conclusion, the mechanical properties such as tensile strength, modulus at 100% and elongation at break of LLDPE are dependent on the polymer-filler interaction and radiation crosslinking. Irradiated carbon black filled LLDPE has higher mechanical properties than the unirradiated LLDPE. The morphology shows a smooth structure and is slowly transformed from ductile into less flexible and rigid structure (brittle) on increasing carbon black filler loading and radiation dose.

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