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STUDIES ON PHYSICAL PROPERTIES AND FRACTOGRAPHY OF ELECTRON BEAM IRRADIATED ETHYLENE VINYL ACETATE/WASTE TYRE DUST BLEND IN THE PRESENCE OF polyethylene graft of maleic anhydride (PEgMAH)

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The effect of electron beam irradiation on the 80/20 EVA/WTD blend was studied in the presence of 0 to 5% of polyethylene graft of maleic anhydride (PEgMAH). The blend was irradiated by using a 3.0 MeV electron beam machine at doses ranging from 20 to 200 kGy in air and room temperature. The gel fraction, tensile strength(Ts), elongation at break(Eb) and Modulus at 100% elongation (M100) were measured. Electron beam irradiation of the EVA/WTD blend were found to cause crosslinking which in effect caused an enhancement in tensile strength, modulus and gel fraction together with a concomitant decline in elongation at break. maximum tensile strength of the blend was achieved at 150 kGy in the presence of 1% PEgMAH and it deteriorates at higher irradiation dosage and PEgMAH addition. The scanning electron micrographs of the tensile fracture surfaces of the irradiated blends show evidence to the enhancement irradiation -induce crosslinking consistent with the above contention.

Keywords: Ethylene Vinyl Acetate (EVA); Waste Tire dust (WTD); irradiation effect, crosslinking

INTRODUCTION

Rubber has become one of the most important material used widely daily in our lives, ranging from the simplest industries such as households to many other important industrial products. Rubber then had been vulcanized into a new product, which is called a synthetic rubber. Synthetic rubbers have been used widely in our days, especially in automotive industries. The production of tire is increasing every year due to the increasing sale of automotive industries, where tire is an integral part of automotive or vehicle. The generation and disposal of tire waste have indeed presented a serious problem to the human community.

The disposal of used automobile tires has caused many environmental and economical problems, due to the processed of being synthetic rubber. Tire disposal requires special solid waste management because of their particular properties. Natural rubber is often vulcanized by a process which the rubber is heated and sulfur, peroxide or bisphenol are added to improve resilience and elasticity, and to prevent it from perishing. Vulcanization greatly improved the durability and utility of rubber [1]. The durability and strength of tires make their disposal and reprocessing extremely difficult. Furthermore, tires are not very amenable to biodegradation [2].

Development of technologies for reducing polymeric waste, which are acceptable from the environmental aspects and which are cost-effective, has proven to be a difficult challenge due to complexities inherent in the reuse of polymers. Establishing optimal processes for the reuse/recycling of polymeric materials thus remain as a worldwide challenge [3].

Extensive studies have been carried out in the area of polymer blends. Blending also helps to improve properties such as processability, thermal, environment stability or the mechanical properties of final product [1]. Polymer blends and composites have been applied to conduct research for the development of new materials through blending and formation of composites using natural rubber, natural rubber latex and synthetic polymers, additives, fillers and compatibilizers at nano sizes for automotives or industrial applications. Radiation treatment of consumer waste polymers for the purpose of recycling is an immerging area due to environmental and economic importance. Thus radiation is often employed to enhance the mechanical properties and performance of recovered materials or materials blends.

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The potential to convert a conventional elastomer (thermoset) into thermoplastic elastomer through blending offers the potential for new market application for waste tire dust. Several studies have been reported on blending recycle tire with various types of thermoplastics and thermoset, such as polypropylene (PP) [4], high density polyethylene (HDPE) and low density polyethylene (LDPE) [5], recycle polyethylene (RPE) [6] and natural rubber [7]. In their work, they found that recycle tires possess relatively low interaction with the matrix when they are used as filler without any additives. High energy irradiation (gamma and electron beam) is an established technique for the modification of polymer.

Thus the study on a ethylene-vinyl acetate (EVA) and waste tire dust (WTD) blends with addition of compatibilizer polyethylene-graft-maleic anhydride (PEgMAH) at various percentage by weight have been carried out to obtain optimum processing conditions and properties. The focus of this research is to develop blend of ground scrap rubber with an irradiation application to develop material with useful range of properties for new markets.

MATERIALS AND METHODS

Material

Ethylene-vinyl acetate (Grade H2020) having 15% vinyl acetate content with MFI value of 1.5 g/10 min and density of 0.93 g/cm^3 was purchased from The Polyolefin Company, Singapore. Rubber dust from recycle tires (40 mesh) used in this study was obtain from Sin Rubtech Consultancy Sdn. Bhd. Polyethylene graft of Maleic anhydride (PEgMAH) functions as compatibilizer.

Sample Preparation

The blends of EVA/WTD with addition of of polyethylene graft of maleic anhydride (PEgMAH) by part per weight of EVA/WTD (0, 1, 2, 3, 4, 5)% were prepared by melt mixing the EVA and WTD in Haake Rheomix Polydrive R600/610 at 140°C and 50 rpm rotor speed for 10 minutes.

When the desire temperature was reached, EVA and PegMAH were charged into the mixing chamber and allowed to melt for 2 min. The WTD was then added to the molten EVA and PegMAH and the mixing was continued for further 8 minute. The blends from the Haake Rheomix were then compression molded into mm thick sheets under a pressure of 14.7 Mpa at 123°C for 5 minute. The sheets were immediately cooled between two plates of a cold press at 25 °C.

Irradiation

The molded sheets were irradiated using a 3 MeV electron beam accelerator NHV EPS-3000 at dose range of 0 to 200 kGy. The acceleration energy, beam current and dose rate were 2 MeV, 2mA and 50 kGy per pass, respectively.

Tensile properties

The tensile properties (tensile strength, Ts; Elongation at break, Eb; Modulus at 100% elongation; M100) were measured with Instron Universal Testing Machine 4301 H119 at 50mm/min crosshead speed. The molded samples of 1 mm thick were cut into dumb bell shape test pieces using BS6746 cutter. Five samples were used for tensile test and an average result was taken as the resultant value.

Gel Content

The gel content of the crosslinked samples were determined by the extraction of samples in boiling xylene for 24 h using Soxhlet apparatus. The extraction samples were dried in oven at 50°C till constant weight. The gel fraction was calculated as:

$$\% \text{ Gel content} = \frac{W_0 - W_1}{W_0} \times 100$$

where W_1 and W_0 are the weights of the dried samples after extraction and before extraction, respectively.

Morphology Study

Examination of tensile fractured surface using a scanning electron microscopic (SEM) model Fei Quanta 400. All samples were examined after sputter coating with gold to avoid electrostatic charging and poor image resolution.

RESULTS AND DISCUSSION

Gel Content Analysis

Gel content test is applied in order to determine the crosslinking built in the samples. Fig 1 shows the relationship between gel content with irradiation doses of 80/20 EVA/WTD blends containing different compositions of PEgMAH. As shown, the extent of gel formation increases with the increase radiation dose, indicating increases in crosslink density of the polymer. A similar increase in gel fraction on ENR and EVA blends was reported by Ratnam et al. [8]. However the percentage of gel content decrease with addition of compatibilizer. More PEgMAH content loaded meaning that less of formation radiation induced crosslinking network in the composite material. The Fig.1 revealed that the

gel content of EVA/WTD blends with 0% phr of PEgMAH shows the highest value than others almost at each irradiation dose while 5% phr shows the lowest value among others. It can conclude that addition of PEgMAH indicates that the additive accelerate the radiation-induced crosslinking in the blend. The higher dose increased formation of radiation-induced crosslinking network which is trigger the molecules to form more bridge also known as crosslink to link a chain to another.

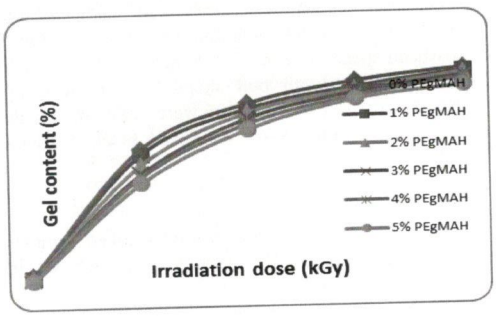

Fig. 1. The effect of PEgMAH content on gel content of EVA/WTD blend at different irradiation doses.

Tensile properties

The effects of the PEgMAH content on the mechanical properties at various doses irradiated of EVA/WTD blends are shown in Figs. 2-4. The PEgMAH content has little effect on the tensile strength or modulus at a definite elongation of the composites. The tensile strength reaches the maximum value when 1 phr PEgMAH added at 150 kGy irradiation. This is due to the enhancement of the crosslink density and with the addition of PEgMAH. In contrast the increment of the crosslink density leads to a reduction of the elongation at break as observed from Fig. 3.

It is evident from Figs. 2-4 that prior to irradiation, the tensile strength, elongation at break and M100 of the blends decrease with increasing PEgMAH content. This is due to lubrication effect of the compatiblizer, PEgMAH. The introduction of electron beam irradiation to EVA/WTD blends has enhanced the irradiation-induced crosslinking. It increased incrementally as the dose are higher. The same observation was also found by C. T. Ratnam et al. for Development novel applications for recycled rubber by using irradiation [8]. The incorporation of PEgMAH as compatibilizer leads to the improvement of mechanical properties. A decline in tensile strength of the EVA/WTD blend at 200 kGy irradiation dose in the presence of PEgMAH was observed. Such a decrease attributed to the embrittlement of the blend due to high extent of irradiation-induced crosslinking. PEgMAH was found to serve the optimum tensile properties upon 150 kGy EB irradiation of EVA/WTD blend.

Fig. 3 shows that elongation at break of all EVA/WTD blends decrease as irradiation dose increases. Such reduction is expected since this property is found to decrease invariably upon irradiation regardless of whether chain scission or crosslinking predominant [3]. This crosslinked network restricts the mobility of the polymer chains and causes the reduction in elongation at break.

Fig. 4 shows the effect of PEgMAH on the modulus at 100% strain (M100) of EVA/WTD blend. The trends reveal that M100 decrease at higher content of PEgMAH due to additivity effect of the PEgMAH. This result further supported previous observation in which PEgMAH provide lubricant effect to the blends. Fig 4 also indicates that the irradiated blends containing higher PEgMAH lead to less stiffer and more flexible blends.

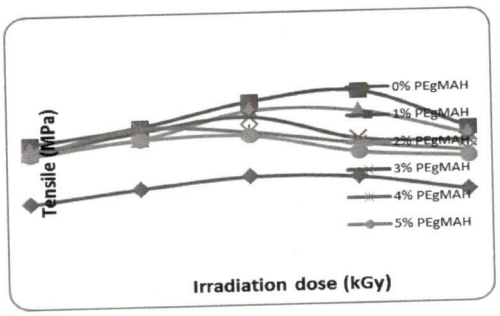

Fig. 2. The effect of PEgMAH content on tensile strength of EVA/WTD blend at different irradiation doses.

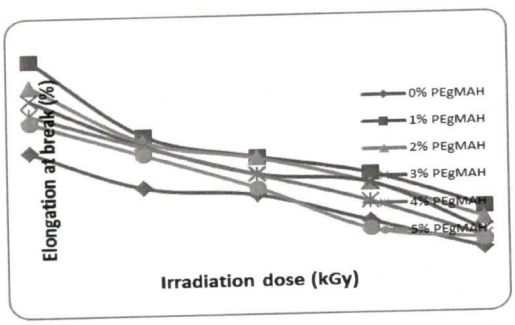

Fig. 3. The effect of PEgMAH content of elongation at break of EVA/WTD blend at different irradiation doses.

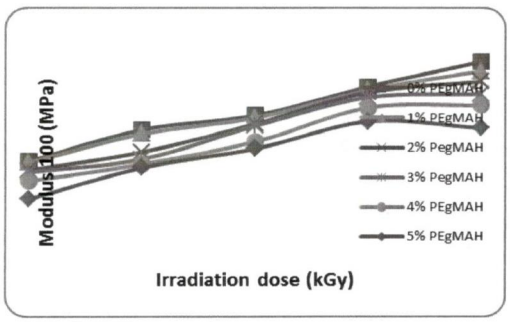

Fig. 4. The effect of PEgMAH content on M100 of EVA/WTD blend at different irradiation doses.

Morphological Analysis

Fig. 5 depicts the effect of PEgMAH on the morphological properties of EVA/WTD blends before and after irradiation. Figs $5_{a,b}$ show the morphological diagram of EVA/WTD blends before irradiation. For further investigate on tensile fractographs of distinc fibril-like morphology of various constructions are apparent for EVA/WTD blends with 1phr PEgMAH (Figs $5_{b,d,f}$) have been carried out. The interaction between the fiber and matrix is clear from SEM images of typical tensile fracture surfaces of composites as shown in Fig 5. Fig 5_a shows the presence of fibrils on the fractured surface indicate that it is a ductile type failure for un-irradiated EVA/WTD blend. The irradiated surface for 100 kGy of EVA/WTD blends (Figs $5_{c,d}$) showed continuous brittle cracks in the network formation. The formation of these cracks is attributed to the embrittlement of the material as a result of high extent of crosslinking [3, 11].

From Fig. $5_{b,d}$, it can be seen that the surface of WTD pulled out of composites with PEgMAH are relatively coarse, and plenty of WTD remains on the rough fiber surface. Good adhesion is clear from the matrix traces sticking to the surfaces of the fibers. It can be ascribed to the better transfer of frictional shear stress across the interface [9,10]. As shown Figs $6_{e,f}$, the fibre surfaces are smooth with little adhering matrix. This is due to the poor affinity between the fibre and matrix attributed to the embrittlement of the blend due to high extent of irradiation-induced crosslinking.

Comparing Figs $5_{a,c,e}$, the surface roughness of the blend become smoother attributed to the embrittlement of the blend due to high extent of irradiation-induced crosslinking at 200kGy irradiation and the low magnitude of inter molecular chain from elastomer without PEgMAH. In contrast with the 100kGy irradiated sample Figs $5_{c,d}$, it shows elastic type failure. The elastic natures that occur during failure are due to the crosslinked state of EVA/WTD phase. Fig 5_d showed that blend with addition of PEgMAH will cause the high magnitude of inter molecular chain that affected to produce fibre reinforced plastic. However sample irradiated at 200kGy (as shown in Figs $5_{e,f}$) will accelerate to degree of crosslink and make the natural stiffer. This indicates the improvement of hardness and modulus in blends due the increasing crosslink density [11]. Thus the SEM studies further testified to the improved interfacial adhesion with the addition of PEgMAH and the effect of electron beam irradiation crosslinking.

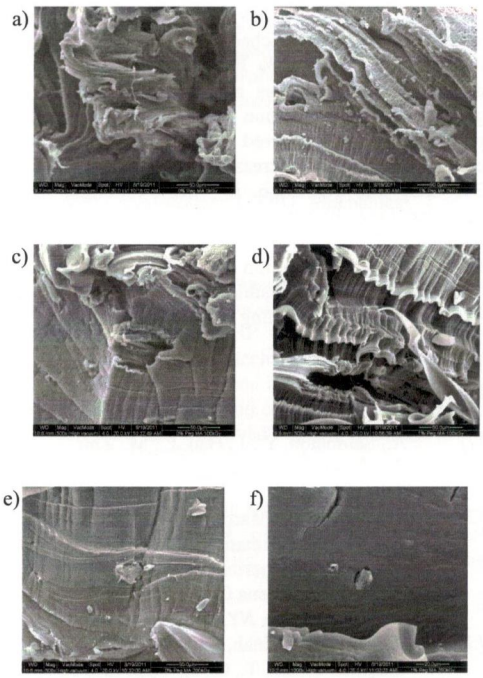

Fig. 5. SEM micrographs of the tensile fracture surface at (a) EVA/WTD 0 phr PEgMAH 0kGy, (b)EVA/WTD 1 phr PEgMAH 0kGy, (c) EVA/WTD 0 phr PEgMAH 100kGy, (d) EVA/WTD 1 phr PEgMAH 100kGy, (e) EVA/WTD 0 phr PEgMAH 200kGy and (f) EVA/WTD 1 phr PEgMAH 200kGy (magnification x1000).

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CONCLUSION

This research found that increase in irradiation doses increased the percentage of gel content in EVA/WTD blends. Gel content data shows that PEgMAH does not serve as crosslinking agent to accelerate irradiation-induce crosslinking of EVA/WTD blend. It was found that the tensile strength of the blend reached a maximum at 150 kGy in the presence of 1 % PEgMAH followed by a decrease at higher doses and higher PEgMAH. A gradual drop in elongation at break of blends upon irradiation is also observed. Irradiation enhances the crosslink density but increases the blend stiffness. At 200 kGy the reduction in tensile strength was observed due to high crosslink density. It was evident from morphological studies that poor interfacial bonding and agglomeration of EVA/WTD blends are the main factors responsible for the drop in tensile strength upon WTD loading

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