



Ablation And Thermal Properties Of Ethylene Propylene Diene Rubber/Carbon Fiber Composites Cured By Ionizing Radiation For Heat Shielding Applications



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Abstract

In this study, new composites were prepared from ethylene propylene diene rubber (EPDM) that loaded with various concentrations of short carbon fibers (SCFs) ranged from 5 up to 20 part per hundred part of rubber (phr). Aluminum trihydroxide (ATH) was added by two concentrations, namely 10 and 30 phr to EPDM/SCFs composites to enhance its heat shielding. Foaming agent, azodicarbonamide (ADC) was added to increase ablation resistance. Composites were prepared on open rubber roll mills, and then the rubber composites were gamma ray irradiated at different doses of 25, 50, 75, 100 and 150 kGy. Fiber incorporation into rubber matrix has been proven as a key factor in enhancing tensile strength and flame retardant properties of fabricated composites. The prepared composites were characterized by using various analytical methods counting physico-mechanical properties, thermal gravimetric analysis (TGA), Ablation test (oxy acetylene test) and propagation flame test. The utmost mechanical performance was reported for the incorporated 5 phr of carbon fiber into EPDM at 100 kGy. Also, the flame retardancy and ablation properties for composites were enhanced by increasing addition of carbon fibers and ATH in presence of ADC.

Keywords: composite, Carbon fiber, Ethylene propylene diene rubber (EPDM), Gamma radiation. Ablation test

1. Introduction

The rubber compounds based on ethylene propylene diene rubber (EPDM) are attracted a lot of interest because of its outstanding heat, ozone and weather resistance. It exhibits good resistance to polar solvent substances and steam condition too. Further, it has excellent electrical insulating properties. The EPDM rubber can be filled with reinforcing fillers, resulting in reduction of cost price. For these reasons this rubber is widely applied in many applications. Generally, most rubbers must be reinforced by fillers before practical applications for their low modulus and strength [1]. Irradiation of rubbers with ionizing radiation presents some advantages over conventional processing because the crosslinking of EPDM is made at room temperature, thus avoiding polymer degradation and oxidative degeneration, as observed in classical vulcanization. The crosslinking via ionizing radiation is an alternative to classical vulcanization to obtain materials with high crosslinking degree; high tensile strength; good resistance to compression; extremely high resistance to oils, lubricants, and greases; short

processing times; and very low material wastes [2, 3]. Cross-linking of polymers induced by ionizing radiation provides increased stability and improves mechanical properties. Under the action of high energy radiation, EPDM, a saturated non-polar rubber, is able to form intermolecular links that exhibits several properties including balanced heat Stability, aging resistance, elasticity (especially at very low temperatures) and water resistance [4]. Carbon fibers (CFs) reinforced insulators have some advantages such as better ablation resistance and char yield, so that it play an important role in the improvement of thermal and ablative properties of insulators. On the other hand, thermal conductivity and breakage during compounding are two main disadvantages of the carbon fibers [5-7].

2. CFs are incorporated into EPDM by various content up to 6 wt%. The addition of CFs to EPDM rubber reduced ablation rate of composites. Thermal stability and endothermic capability were improved with increasing short fiber contents in the rubber matrix. Experimental thermal conductivity measurement results elucidate that thermal

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conductivity reduces 60% at 473 Kelvin with 6 wt% addition of the fibers.

A remarkable improvement was scrutinized in the tensile strength and rubber hardness with increasing fiber to matrix ratio. Scanning electron microscopy (SEM)/energy dispersive X-ray spectroscopy (EDS) analysis of the composite specimens revealed the uniform dispersion of CFs within the host matrix [8]. EPDM based insulation system is being globally used for case-bonded solid rocket motors. Prepared compositions based on EPDM, filled fibrous fillers as aramid, and carbon fibers have been studied for mechanical, rheological, thermal, and interface properties. Addition of fibrous filler (5 parts) improved the peel strength, and reduced the thermal conductivity and erosion rate[9]. To improve the ablative performance, fibrous fillers have been used with the other supplemental materials such as flame retardant or char former[10, 11].

3. Mineral flame retardant, such as aluminum trihydroxide (ATH) is used widely because of their low cost [12]. ATH acts through fuel replacement in the condensed phase and through fuel dilution and cooling in the gas phase, due to the release of water into the flame and the formation of a ceramic protective layer[13]. The thermal stability and the flame retardant properties were evaluated for composites based on EPDM with aluminum hydroxide (ATH), nanoclay. The results indicated that the synergistic flame retardancy of the nanoclay with ATH in EPDM matrix could imply that the formation of a reinforced char/nanoclay layer during combustion prevents the diffusion of the oxygen and the decomposed organic volatiles in the flame [14]. A new method for the development of asbestos free rubbers for use as rocket motor insulators is presented. Such insulation is based on chopped carbon fiber (CCF) and aramid fiber in pulp form as reinforcement for EPDM along with ammonium polyphosphate (AP) flame retardant agent. Six millimeters long CCFs and/or Kevlar pulp (KP) are dispersed in the EPDM polymeric matrix to obtain a homogenous master batch for curing. The new method involves the development of two types of prepregs and the lamination of these types of prepregs. The first one consists of CCF/EPDM/AP (50 phr CCF) and the second type of prepreg KP/EPDM/AP (30 phr KP). Laminates composed of six alternative layers of these prepregs have been shown to exhibit better thermal, mechanical, physical, and ablative properties than their non-laminated counterparts [15].

This work aimed to prepare composites form EPDM and SCFs in presence of flame retardant ATH and foaming agent to enhance ablative and flame resistance of these composites for different applications

Experimental Materials and experimental techniques

Materials

Ethylene propylene diene monomer (EPDM) rubber, with ethylene content 59% and 4.4% 5-ethylidene-2-norborene (ENB), as termonomer was supplied by Polimer Europa (Italy) under the trade name Dutral TER 4049: Mooney viscosity (ML_{1+4} , 125°C) 76. Carbon fiber with specification; yarn material: Carbon 3k, yarn number: 200 tex, supplied by Suter Kunststoffe AG, Aeßligenstrasse 3, Switzerland. Aluminum hydroxide as fire retardant, Molecular formula: Al (OH)₃, Molecular weight: 78, Supplier: Sigma-Aldrich, 3050 Spruce Street, Saint Louis, MO 63103, USA. The chemical blowing agent used in this work was azodicarbonamide (ADC) has gas yield 220 mL/g, and was supplied from Haihong Chemical, China. Zinc Oxide in the form of white powder was supplied by EL Nasr Chemical Co. (Egypt). A commercial grade stearic acid was supplied from China, m.p.: 54 °C. 1, 2-dihydro-2, 2, 4-trimethylquinoline (TMQ), supplied by Birla Tires Limited (India), was used as antioxidant.

Sample Sheet preparation

The warp carbon fibers cut into SCFs to length ~ 15 mm, then EPDM/ SCFs composites were mixed on open rubber roll mill (ASTM D3182 – 16) at different contents of SCFs and other ingredients like TMQ, stearic acid, ZnO, ATH and ADC, after mixing the composites were hot pressed at about 130°C under 10 MPa for 5 min into sheets of suitable thickness and size for examination.

Gamma irradiation

Irradiation of blends was performed at the National Centre for Radiation Research and Technology (NCRRT), Egyptian Atomic Energy Authority (EAEA), Cairo, Egypt. The blends were subjected to gamma radiation, gamma cell type 4000 A, India, at ambient air, humidity and room temperature with the radiation doses from 25 up to 150 kGy at a dose rate of 1.19 kGy/h.

Mechanical testing

Mechanical testing was carried out using a Zwick tensile testing machine (Model Z010;Germany)at room temperature and across head speed of 150 mm min⁻¹ using dumbbell-shaped tensile specimens according to ASTM D 412-16.

Determination of cross-link density of rubber

Measurement of stress-strain response gives an easy method for estimation of the cross-link density of polymer networks. According to the kinetic theory of rubber elasticity [16], the stress-strain cross-link density relationship is given by the following equation:

$$\sigma = \sigma_0 (\lambda) + G_e (\lambda^2 - 1/\lambda) \quad (1)$$

where, σ is the true stress, σ_0 is the stress, λ is the extension ratio, G_e is a parameter depends on the

degree of cross-linking density. The relation between $(\lambda^2 - \lambda^{-1})$ and true stress (σ) for rubber blends were plotted as straight lines, hence G_c is the slope of this straight lines and related to M_c , which is the average molecular weight between two cross-links, by the following equation:

$$G_c = A_{\phi} \rho RT / M_c \quad (2)$$

Cross-linking density is calculated depending on M_c and calculated as follows:

$$\text{Cross-link density } (\nu) = 1/2M_c \quad (3)$$

Hardness measurements

The preparation samples at smallest amount 1 mm in thickness with plane surface were cut for hardness examination. The measurement was carried out according to (ASTM D 2240 - 2000) by a durometer of model 306L type pacific transducer corp.,USA. The units of hardness are expressed in (Shore A).

Thermogravimetric analysis (TGA)

Thermal analysis (TGA) was performed with TA/TGA-55., instrument from Shimadzu (Japan) for testing the thermal behavior of samples. The heating was carried out at temperature range from room temperature to 600°C with a heating rate of 10°C/min under nitrogen gas atmosphere. The weight of samples ranged from 3 to 5 mg each.

Flame propagation test (FP)

Flame propagation test was performed according to ASTM C1166-06(2016) for cellular elastomers. A vertical burn test specimen that have evaluates that flame propagation and reports the burning behavior has 25cm length and 1cm diameter .the burning time for each samples was performed for 5 min. the flame propagation was calculated for average six samples as follow:

$$FP = R_o - R_f \quad (4)$$

Where, R_o =original length and R_f =final length.

Ablation test

The ablation properties prepared composites were measured by oxy acetylene torch (ASTM-E285-08(2015)) by means of equipment created in our laboratory. Samples of 10*10*0.5 cm³ were placed at 2 cm of the torch. The flame was controlled by changing oxygen and acetylene proportion. The ablation test was performed until the specimen was burnt through its entire thickness during a defined time. Linear and mass ablation rates of samples were measured for 30 seconds, and then the flame was extinguished allowing the samples to cool down naturally. The linear ablation rate(LAR)and mass ablation rate (MAR)and percentage char yield were calculating according to Eqs.5-7.

$$\text{Linear ablation rate (LAR) (mm/s)} = \frac{T_0 - T_1}{t} \quad (5)$$

$$\text{Mass ablation rate (MAR) (g/s)} = \frac{M_0 - M}{t} \quad (6)$$

$$\% \text{ Char yield} = \frac{[M_0 - M_1 / M_0] * 100}{\quad} \quad (7)$$

Where : T_0 , M_0 , T_1 and M_1 are the thickness and mass of the ablator specimen before and after ablation testing, respectively; t is the ablation time fixed at 30s.

Scanning electron microscopy (SEM)

An ISM-5400 scanning electron microscopy (SEM) , JEOL, Japan, was used for the morphological examination of fracture samples, The samples were coated with gold before testing.

Results and discussion

Physico-mechanical Properties

Yield Strength (YS)

Figure 1 Shows the variation of yield strength (YS) as a function of irradiation dose for EPDM rubber loaded with different concentrations of SCFs. It can be seen that the values of YS for EPDM as well as its composites with SCFs increase on applying the irradiation dose. The values of YS increases slowly with increasing irradiation dose up to 50 kGy .By increasing the irradiation dose beyond 50 kGy ,the values of YS increases sharply and reaching its maximum value at 150 kGy, it was observed that the EPDM composites with 20 phr of SCFs attained the highest values of YS over the whole range of irradiation dose. The YS reached the highest value at 20 phr SCFs, the increasing YS of composites by increasing SCFs loading due to increasing plasticity by included SCFs to EPDM rubbery matrix [17].

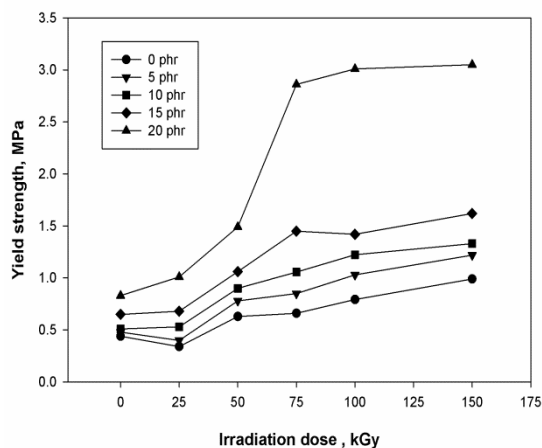


Fig. 1. Yield strength as a function of irradiation dose for EPDM loaded with carbon fibers

Tensile Strength (TS)

Figure 2 represents the TS as a function of irradiation dose for Ethylene propylene monomer rubber (EPDM) vulcanizates with different contents of SCFs, the results obtained for irradiated and unirradiated EPDM samples are also incorporated in the same figure as control for comparison. It can be seen that the values of TS for EPDM as well as its composite with SCFs increase on applying the irradiation dose. The values of TS increase gradually with increasing irradiation dose up to 100 kGy. The

increase in TS values may be attributed to increased crosslinking density that is occurring between macromolecules that found on both the EPDM and SCFs rubber. Beyond irradiation dose 100 kGy, the values of TS decreased on increasing the dose up to 150 kGy. This later reduces of TS values is affiliated increased rate of degradation with respect to crosslinking. Moreover, it may be practical that the EPDM composite with 5 phr of SCFs attained the highest values of TS over the whole range of radiation dose then it was decreased by increasing the content of SCFs up to 20 phr. When the fiber content was less than 20 phr, the ultimate TS of rubber composite was enhanced due to the amazing strength of the fiber and good fiber-matrix adhesion [8]. By increasing fiber loading the fiber not wetted by rubber matrix, as well as agglomeration of fibers takes place that totally lead to deterioration of TS of composites

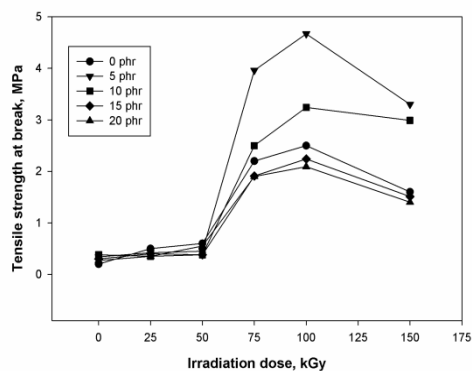


Fig. 2 Tensile strength at break as a function of irradiation dose for EPDM loaded with carbon fibers

Elongation at break (E_b %)

Figure 3 shows the variation in E_b of EPDM/SCFs rubber composites. As the radiation dose increases, the elongation for all composites increases up to first irradiation dose, namely 75 kGy, then it was decreased sharply up to 150 kGy. The increased elongation observed up to 75 kGy due to unattached macromolecules which are highly entangle but free reptate, i.e undergo reptile like motion through the three-dimensional structure[18]. Also at any irradiation dose it can be seen that the E_b decrease systematically with increasing SCFs percentage addition to EPDM. The decreasing in values of E_b by increasing SCFs loading due to higher modulus or rigidity of SCFs that causes its low elongation at break [8, 18] on the other hand, compatibility between component of composites by radiation lead to limited flow of macromolecules of rubber, then lead to decrease in elongation[19].

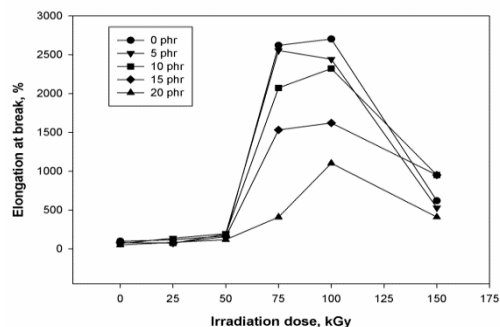


Fig. 3 Effect of irradiation dose on Elongation at break, % for EPDM composites loaded by carbon fibers

Tensile modulus at 25 % elongation (M_{25})

The stress at a definite elongation is inversely proportional to molecular weight between crosslinks (M_C)[20], which is related directly to the cross link density (number of network chains per unit volume) by inverse proportion. This observation would account for obtained M_{25} that are illustrated in Figure 4 as a function of irradiation dose, it was noticed that values of M_{25} for EPDM rubber and its composites with SCFs increases slowly with increasing irradiation dose up to 100 kGy, then leveling off at 150 kGy. This indicates the increase of cross link density of EPDM and its composites by radiation. Radiation induced crosslinking in rubber matrix that inhibits its flexibility, and hence increases its stiffness [21].

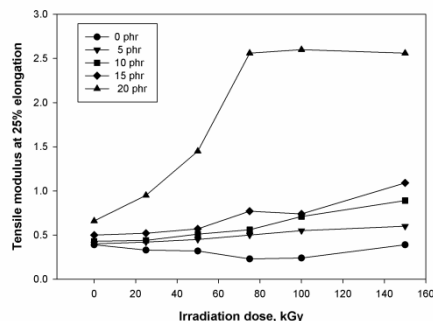


Fig. 4 Effect of irradiation dose on tensile modulus of EPDM loaded with carbon fibers

Crosslinking density

Figure 5 shows the effect of gamma rays on the crosslinking density of EPDM rubbers and its composites loaded with different amount of SCFs, the results of crosslinking density measurement of EPDM rubber and its composites showed that there is an increase in the crosslinking with increasing irradiation dose up to 150 kGy and by SCFs loading up to highest concentration namely, 20 phr. Results of crosslinking density are not similar to TS in behavior. Tensile strength, Tear strength, fatigue life, and toughness are related to the breaking energy. Values of these properties increase with small

amounts of crosslinking, but they are reduced by further crosslink formation [23].

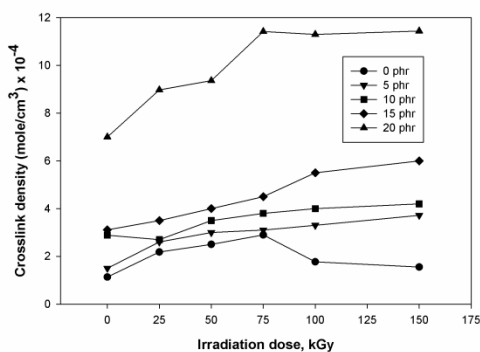


Fig. 5 Effect of irradiation dose on crosslink density for EPDM loaded with carbon fibers

Hardness

Figure 6 shows the data obtained for variation of hardness as a function of irradiation dose for unirradiated as well as irradiated EPDM samples that is either loaded or unloaded with SCFs. It may be observed that for all blank EPDM samples which not loaded with fibers, they have attained the lowest hardness values over the whole range of radiation, and this when compared with EPDM/SCFs composites. On the other hand, it may be seen that the increase in hardness was obtained by increase of irradiation doses for all composites of EPDM/SCFs. Meanwhile, at any irradiation dose, it can be observed that the values of hardness for composites increases with increasing SCFs content up to 20 phr. High rigidity of SCFs significantly decreased the chain mobility of the polymer matrix, and as a result, increased the hardness of final composites [22-24].

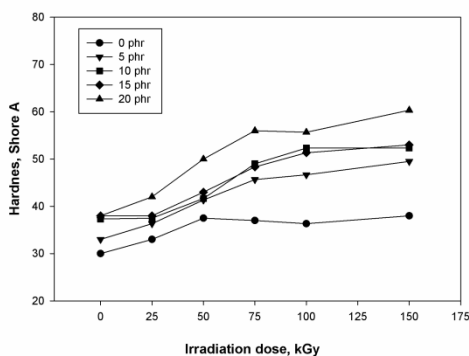


Fig. 6 Effect of irradiation dose on hardness of EPDM loaded with carbon fibers

Thermal gravimetric Analysis (TGA)

Figures 7 and 8 represent TGA and its rate of thermal decomposition (dw/dt) for EPDM/SCFs composites respectively. EPDM and EPDM/SCFs composites show one main step for thermal degradation that is recorded at temperatures of 400–500°C, due to polymer disintegration and molecular bond breakage of the polymer chains of the matrix. The data showed

that the EPDM has the maximum decomposition temperature (T_{max}) obtained at 467°C, while T_{max} for EPDM/5SCFs composites is obtained at 470°C, this due to adhesion between EPDM and SCFs at lower concentration. On the other hand, the T_{max} for EPDM/10SCFs composites is obtained at 469°C. The thermal stability for EPDM/20SCFs composites was decreased, and T_{max} was obtained at 464°C, the lower in thermal stability at high content of SCFs namely, 20 phr is due to agglomeration of fibers which lead to less adhesion between rubber matrix and fiber, the obtained results confirmed by mechanical properties that discussed before. The final residual weight of unloaded EPDM was obtained at 5.75%, while the final residue of composites at 20 SCFs increased to 19% after thermal exposure of the composite specimens up to 600°C. This is basically due to the presence of SCFs residue in the remaining char. According to Jiang et al. [25], the char structure of the EPDM composite was the prime factor that determined the ablation and insulation performance of EPDM. The enhanced char strength and thermal stability of the char layer played a decisive role in preventing heat transfer by hindering the spread of decomposition products and slowing down the rate of volatiles formation. Due to the intact and dense nature of the char layer, the physical barrier was augmented [26]. In addition to a dense char layer of EPDM matrix, SCFs content had a crucial effect on thermal degradation property. By increasing SCFs in the polymer matrix, weight loss of the composite slowed down and occurred at higher temperatures [27]

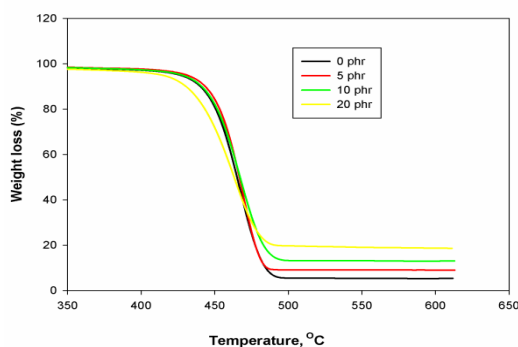


Fig. 7 Weight loss as a function of heating temperature for EPDM loaded with carbon fibers.

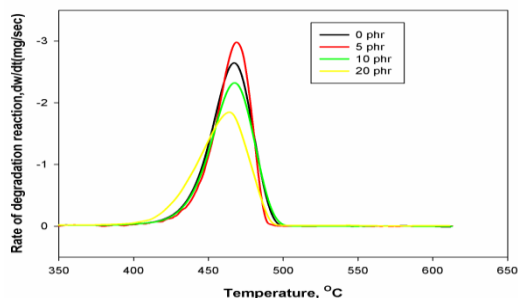


Fig. 8 DTG of EPDM loaded by carbon fibers

Flame propagation

The percentage burnet length form flame propagation test for unirradiated EPDM.SCFs/ATH at different of percent of ATH and concentrations of SCFs namely, 5 and 20 phr was presented in Figure 9(a) and (b) respectively. Generally, the results show that the Flame propagation values decreases, as the amount of ATH increases. When comparing the burnet length at different SCFs concentrations and at constancy of ATH and foaming agent, the burnet length was decreased at 20 phr more than 5 phr SCFs this due to increase of char formation. On the other hand adding foaming agent beside ATH more and more decrease burnet length due to at high temperature foaming agent decompose giving CO₂ and NO₂ gases which protect organic matter of EPDM rubber form degradation. The percentage burnet length for irradiated EPDM.SCFs/ATH composites at different of percent of ATH and concentrations of SCFs namely, 5 and 20 phr was presented in Figure 10(a) and (b) respectively. The irradiated composites take the same behavior of unirradiated ones except they have lower values for burnet length, this due to irradiation dose lead increase in flame retardancy[28].

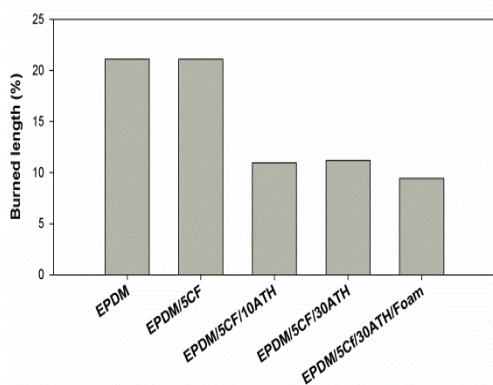


Fig. 9(a) Effect of ATH on flame propagation of unirradiated EPDM/SCF composites

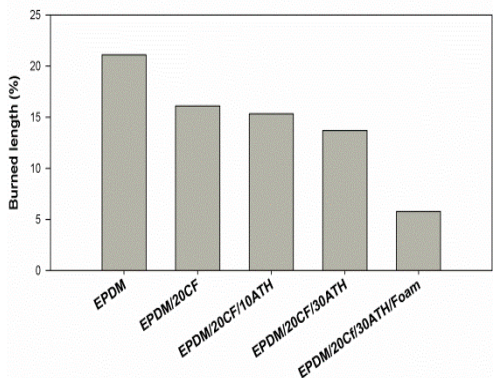


Fig. 9(b) Effect of ATH on flame propagation of unirradiated EPDM/20CF composites.

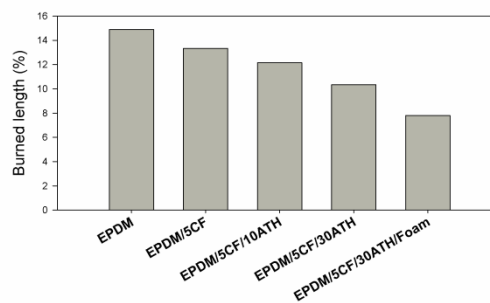


Fig. 10(a) Effect of ATH on flame propagation of EPDM/5CF composites irradiated at 100 kGy

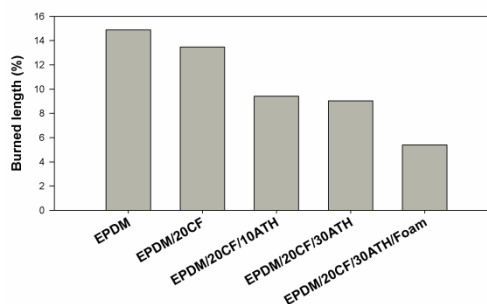


Fig. 10(b) Effect of ATH on Flame propagation of EPDM/20CF/ATH composites irradiated at 100 kGy.

Ablation testing

Tables 1 and 2 show the linear ablation rate, mass ablation rate and percent char yield for EPDM and its composites loaded with 5 and 20 phr SCFs respectively, and also all composites loaded with different concentrations of ATH. On the other hand all irradiated at 100 kGy. EPDM rubber is one of the best matrices to exhibit the lowest erosion rate that ultimately proves it the best insulation material and ablative material [23].

The values of linear, mass ablation rates and percent char yield of EPDM/SCFs composites are lower than EPDM rubber matrix, as addition of SCFs to EPDM enhances ablation performance of composites [8]. On the other hand, by increasing addition of ATH to EPDM/SCFs composites, the ablation resistance more increase, we believe that the ablation performance of EPDM/SCFs composites with different concentration of ATH is better than that of EPDM and EPDM/SCFs, also addition of foaming agent besides ATH can enhance the ablation resistance of the EPDM insulation material. The same behavior in is obtained for increasing ablation resistance by addition carbon nanotubes to EPDM in presence of fumed silica as flame retardant [29]. Nitrogen gas from the decomposing foaming agent produces expansion in depolymerized polymer from degrading insulation. The porous layer reduces the heat flow into the non-degraded insulation. Also nitrogen gas provide induction cooling in the char, it was moved from degraded layer to the outermost char surface [30].

Table 1. Linear ablation rates and percent char yield for EPDM/5SCFs composites after experimental ablation examination.

Sample	EPDM	EPDM/5CF	EPDM/5CF/10ATH	EPDM/5CF/30ATH	EPDM/5CF/30ATH+Foam
Linear mass ablation rates	0.1482	0.1333	0.1045	0.0869	0.0835
Linear ablation rates	0.2663	0.2306	0.0916	0.0833	0.0533
Linear percent char yield	14.550	12.600	8.8800	8.0400	5.9900

Table 2. Linear ablation rates and percent char yield for EPDM/20SCFs composites after experimental ablation examination

Sample	EPDM	EPDM/20CF	EPDM/20CF/10ATH	EPDM/20CF/30ATH	EPDM/20CF/30ATH+Foam
Linear mass ablation rates	0.1482	0.1348	0.0905	0.0883	0.0704
Linear ablation rates	0.2663	0.2180	0.0833	0.0733	0.0533
Linear percent char yield	14.5500	11.8600	7.3200	6.7700	5.3100

SEM studies of EPDM/ SCFs Composites

Figure 11 Shows SEM photomicrographs of tensile fracture surface for EPDM/SCFs composites. Figure 11(a) represents unirradiated EPDM rubber, It is not smooth and was shown as gum rubber, meanwhile, EPDM irradiated at 100kGy shows smooth surface due to curing by radiation as was shown in Figure 11(b). As can be seen from Figure 12(a) shows SEM for EPDM/SCFs composites loaded with 5 phr SCFs at 100kGy, the arrangement of the SCFs in the EPDM more orderly and uniformly distribute, besides the short length of fibers indication the adhesion of fiber to rubber matrix. Figure 12(b) shows SEM for EPDM/20SCFs at 100kGy, the fiber distribution is not uniform, and has obvious hole in the sample, as well as the long length of fibers. The EPDM/20SCFs are shown in Figure 12(c), with 30 ATH at same dose 100kGy, the fracture surface is not uniform in distribution. The EPDM/20SCFs/30ATH with foaming as shown in Figure 12(d) fibers appear agglomeration phenomenon in the EPDM, uneven distribution, and have many voids[31]

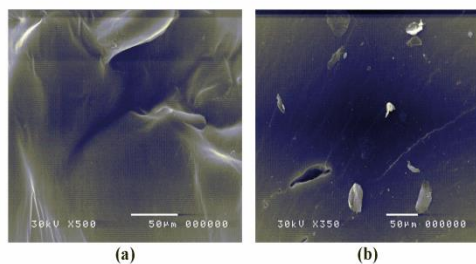


Fig. 11. SEM for EPDM, a- Unirradiated, b- Irradiated at 100 kGy

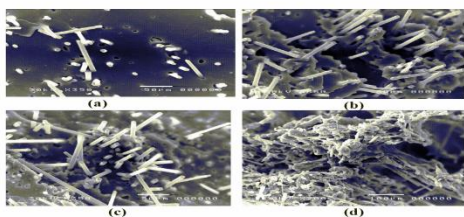


Fig. 12. SEM for EPDM/SCFs composites. (a), EPDM/5CF, (b), EPDM/20CF, (c), EPDM/20CF+30ATH, (d), EPDM/20CF+30ATH+foaming agent all composites irradiated at 100 kGy.

Conclusions

This article deals mostly with the use of the ionizing radiations for acting crosslinking in EPDM /SCFs composites. The characterization of prepared composites, namely, physical and mechanical was investigated. The obtained results are following:

The maximum TS for composites obtained at 5phr then decreased up to 20 phr. Meanwhile, the maximum YS, M_{25} , hardness and crosslink density obtained at 5 phr SCFs. All mechanical parameters increase by irradiation dose up to 100 kGy. The thermal stability and ablation characteristics were studied via TGA analysis, the char structure of the EPDM composite was determined the ablation and insulation performance of EPDM.

SCFs content had a crucial effect on thermal degradation property. By increasing SCFs in the polymer matrix, weight loss of the composite slowed down and occurred at higher temperatures. From Flame propagation test when used different amount of ATH with EPDM and SCFS, with increasing amount of ATH the value of flame propagation decreased. When comparing the burnet length at different SCFs concentrations and at constancy of ATH and foaming agent, the burnet length was decreased at 20 phr more than 5 phr SCFs this due to increase of char formation.

From ablation test the values of linear, mass ablation rates and percent char yield of EPDM/SCFs composites are lower than EPDM rubber matrix, as addition of SCFs to EPDM enhances ablation performance of composites. By increasing addition of ATH to EPDM/SCFs composites, the ablation resistance more increase. ATH and foaming agent exhibit good thermal conductivity; thus, from the perspective of heat transfer, the addition of ATH and foaming agent may adversely affect the ablation performance of EPDM.

References

- [1] R.M. Mohamed, K.F. EL-Nemr, H.A. Raslan, Properties of ethylene propylene diene rubber/white and black filler composites cured by gamma radiation in presence of sorbic acid, *Journal of Macromolecular Science, Part A* 56(5) (2019) 429-442.
- [2] M.-D. Stelescu, E. Manaila, G. Craciun, M. Dumitrascu, New green polymeric composites based on hemp and natural rubber processed by electron beam irradiation, *The Scientific World Journal* 2014 (2014).
- [3] M.D. Stelescu, A. Airinei, E. Manaila, N. Fifere, G. Craciun, C. Varganici, F. Doroftei, Exploring the effect of electron beam irradiation on the properties of some EPDM-flax fiber composites, *Polymer Composites* 40(1) (2019) 315-327.
- [4] S. Samaržija-Jovanović, V. Jovanović, G. Marković, S. Konstantinović, M. Marinović-Cincović, Nanocomposites based on silica-reinforced ethylene-propylene-diene-monomer/acrylonitrile-butadiene rubber blends, *Composites Part B: Engineering* 42(5) (2011) 1244-1250.
- [5] G. Gao, Z. Zhang, X. Li, Q. Meng, Y. Zheng, An excellent ablative composite based on PBO reinforced EPDM, *Polymer bulletin* 64(6) (2010) 607-622.
- [6] C.-j. Song, W. Zhang, J.-a. Mo, Effect of short fiber filler on the properties of NBR-matrix insulator, *JOURNAL OF SOLID ROCKET TECHNOLOGY* 27(1) (2004) 73-76.
- [7] T. Farajpour, Y. Bayat, M. Abdollahi, M.H. Keshavarz, Effect of borax on the thermal and mechanical properties of ethylene-propylene-diene terpolymer rubber-based heat insulator, *Journal of Applied Polymer Science* 132(19) (2015).
- [8] M. Asghar, N. Iqbal, S.S. Iqbal, M. Farooq, T. Jamil, Ablation and thermo-mechanical tailoring of EPDM rubber using carbon fibers, *Journal of Polymer Engineering* 36(7) (2016) 713-722.
- [9] C. Bhuvanewari, S. Kakade, V. Deuskar, A. Dange, M. Gupta, Filled Ethylene-propylene Diene Terpolymer Elastomer as Thermal Insulator for Case-bonded Solid Rocket Motors, *Defence science journal* 58(1) (2008) 94.
- [10] H. Lu, L. Song, Y. Hu, A review on flame retardant technology in China. Part II: flame retardant polymeric nanocomposites and coatings, *Polymers for Advanced Technologies* 22(4) (2011) 379-394.
- [11] C. Wei, M. Zeng, X. Xiong, F. Zhang, Thermal and frictional properties of modified sisal fibre/phenolic resin composites, *Plastics, Rubber and Composites* 39(2) (2010) 61-66.
- [12] D.J. Kind, T.R. Hull, A review of candidate fire retardants for polyisoprene, *Polymer Degradation and Stability* 97(3) (2012) 201-213.
- [13] B. Zirnstein, W. Tabaka, D. Frasca, D. Schulze, B. Schartel, Graphene/hydrogenated acrylonitrile-butadiene rubber nanocomposites: Dispersion, curing, mechanical reinforcement, multifunctional filler, *Polymer Testing* 66 (2018) 268-279.
- [14] Y.Y. Yen, H.T. Wang, W.J. Guo, Synergistic effect of aluminum hydroxide and nanoclay on flame retardancy and mechanical properties of EPDM composites, *Journal of Applied Polymer Science* 130(3) (2013) 2042-2048.
- [15] A.F. Ahmed, S.V. Hoa, Thermal insulation by heat resistant polymers for solid rocket motor insulation, *Journal of composite materials* 46(13) (2012) 1549-1559.
- [16] Y.-H. Zang, R. Muller, D. Froelich, Determination of crosslinking density of polymer networks by mechanical data in simple extension and by swelling degree at equilibrium, *Polymer* 30(11) (1989) 2060-2062.
- [17] K. Sau, T. Chaki, D. Khastgir, Carbon fibre filled conductive composites based on nitrile rubber (NBR), ethylene propylene diene rubber (EPDM) and their blend, *Polymer* 39(25) (1998) 6461-6471.
- [18] J. Augustine, S. Maiti, A. Gupta, Mechanical properties and crystallization behavior of toughened polyamide-6/carbon nanotube composites, *Journal of Applied Polymer Science* 125(S1) (2012) E478-E485.
- [19] M.M. Hassan, R.O. Aly, A. El-Ghandour, H.A. Abdelnaby, Effect of gamma irradiation on some properties of reclaimed rubber/nitrile-butadiene rubber blend and its swelling in motor and brake oils, *Journal of Elastomers & Plastics* 45(1) (2013) 77-94.
- [20] K.F. El-Nemr, M.A. Ali, M.M. Hassan, H.E. Hamed, Features of the Structure and Properties of Radiation Vulcanizates Based on Blends of Polybutadiene and Ethylene-Propylene Diene Rubber, *Journal of Vinyl and Additive Technology* 25(S1) (2019) E64-E72.
- [21] M.A. Ali, K.F. El-Nemr, M.M. Hassan, Waste newsprint fibers for reinforcement of radiation-cured styrene butadiene rubber-based composites-Part I: Mechanical and physical properties, *Journal of reinforced plastics and composites* 30(8) (2011) 721-737.
- [22] N. Iqbal, S. Sagar, M.B. Khan, H.M. Rafique, Elastomeric ablative nanocomposites used in hyperthermal environments, *Polymer Engineering & Science* 54(2) (2014) 255-263.

- [23] N. Iqbal, S. Sagar, M.B. Khan, M.I. Bassyouni, Z.M. Khan, Aluminum silicate fibers impregnated acrylonitrile butadiene rubber composites: Ablation, thermal transport/stability, and mechanical inspection, *Journal of Applied Polymer Science* 130(6) (2013) 4392-4400.
- [24] N. Iqbal, S. Sagar, M.B. Khan, Comprehensive ablation characteristics of ceramic fibers impregnated rubber composites, *International Journal of Engineering and Technology* 6(3) (2014) 162.
- [25] Y. Jiang, X. Zhang, J. He, L. Yu, R. Yang, Effect of polyphenylsilsesquioxane on the ablative and flame-retardation properties of ethylene propylene diene monomer (EPDM) composite, *Polymer degradation and stability* 96(5) (2011) 949-954.
- [26] T. Ogasawara, T. Ishikawa, T. Yamada, R. Yokota, M. Itoh, S. Nogi, Thermal response and ablation characteristics of carbon fiber reinforced composite with novel silicon containing polymer MSP, *Journal of composite materials* 36(2) (2002) 143-157.
- [27] G. Gao, Z. Zhang, X. Li, Q. Meng, Y. Zheng, Z. Jin, Effect of weight ratio of thermoplastic and thermosetting boron-containing phenolic resin on mechanical, bonding, and ablative properties of thermal insulating composites, *Journal of applied polymer science* 118(1) (2010) 266-274.
- [28] M.N. Alnajrani, E.M. Alosime, A.A. Basfar, Influence of irradiation crosslinking on the flame-retardant properties of polyolefin blends, *Journal of Applied Polymer Science* 137(18) (2020) 48649.
- [29] M. Guo, J. Li, K. Xi, Y. Liu, J. Ji, Effect of multi-walled carbon nanotubes on thermal stability and ablation properties of EPDM insulation materials for solid rocket motors, *Acta Astronautica* 159 (2019) 508-516.
- [30] J. Sommer, Trowelable rubber rocket insulation for use at 2800° C, *Rubber chemistry and technology* 57(4) (1984) 843-854.
- [31] W.L. Wu, J.K. Li, Study on carbon fiber reinforced chloroprene rubber composites, *Advanced Materials Research, Trans Tech Publ*, 2014, pp. 254-257.