ELECTRICAL AND SHIELDING PROPERTIES OF ACRYLONITRILE BUTADIENE STYRENE FILLED WITH DIFFERENTLY TREATED RECYCLED CARBON FIBERS

A.M. Alkaseh^{1,2}, M.E.A Manaf¹ and N. Mohamad¹

¹Faculty of Manufacturing Engineering, Universiti Teknikal Malaysia Melaka, Hang Tuah Jaya, 76100 Durian Tunggal, Melaka, Malaysia.

²Department of Training and Cooperation, Polymer Research Centre, 83152 Tripoli, Libya.

Corresponding Author's Email: 1edee@utem.edu.my

Article History: Received 20 April 2021; Revised 30 March 2022; Accepted 3 May 2022

ABSTRACT: Polymeric composites have been studied for electromagnetic interference (EMI) shielding material due to their many advantages including low density, good flexural and impact strength, and ease of processing. Recycled carbon fiber has the potential as conductive filler in polymer composites, however the effects of different treatment such as irradiation on the properties is still lacking. A comparative study based on different fiber treatments and electron beam irradiation on recycled carbon fibers (rCF) filled acrylonitrile butadiene styrene (ABS) composites has been carried out and the results are presented in this paper. The main objective is to evaluate how these treatments impact properties such as conductivity and shielding effectiveness (SE) of the composites. Three types of filler, i.e., untreated rCF, chemically treated rCF, and chemically-mechanically treated rCF are used as to improve the electrical conductivity of ABS polymer. In order to enhance shielding effectiveness of the composites, electron beam (EB) irradiation is applied at 200 kGy to induce cross linked networks. The composite samples are prepared by using melt compounding method. In general, composites reinforced with treated rCF show better performance in both conductivity and shielding effectiveness. Moreover, for the same fiber amount, the electrical conductivity of the composites filled with irradiated-recycled carbon fibers is much higher than that of non-irradiated recycled carbon-fiber-filled composites. By using irradiated-recycled carbon fibers as filler, in order to achieve the same degree of conductivity as carbon fiber filled composites, the required volume fraction of fibers can be greatly reduced. From the results, the EB irradiated composite with 30 wt% chemically treated rCF shows the best shielding effectiveness and

electrical conductivity, where the highest values reached are 1.34×10^{-8} S/m and 46.13 dB, for electrical conductivity and shielding effectiveness, respectively. This finding is significant as to better understand the behavior of rCF filled ABS composites especially for EMI shielding application.

KEYWORDS: Recycled Carbon Fibers; Chemical Treatment; Electrical Conductivity; Shielding Effectiveness; Acrylonitrile Butadiene Styrene (ABS)

1.0 INTRODUCTION

With the widespread application of electronic devices and systems worldwide, especially in communications facilities and wireless networks, the problems associated with electromagnetic interference (EMI) has become a more important issue [1,2]. EMI can potentially lead to undesirable effects on highly sensitive devices, as well as on our health [3,4]. Therefore, the development of high performance EMI shielding material is crucial. EMI shielding materials find applications in various sectors including industrial, commercial, and military areas. For instance, it can be utilized for electromagnetic waves as castings for televisions and computers, in order to make them work more safely [5,6].

In the past few decades, due to their high electrical conductivity, metals have been utilized as the common materials for EMI shielding materials. However, their application for this purpose is restricted due to some weaknesses such as corrosion susceptibility, high density and difficulty in processing [1,2]. Conductive polymer based composites for EMI shielding have been studied greatly in recent years [7,8] due to their various advantages including low density, good mechanical properties, as well as significant reinforcing effect. These make them favorable for lightweight EMI shielding materials. Moreover, their ease of manufacturing by injection molding or extrusion, making them suitable to be manufactured in batches [9].

Carbon fibers reinforced polymers, having excellent mechanical characteristics and low density have been widely utilized in transport, aerospace, and sportive materials [10,11]. The excellent properties of CF including high specific tensile strength and good wear resistance have made it the material of choice to reinforce advanced composite materials. However, a significant shortcoming of the shielding material using CF reinforced polymer is that the electrical conductivity of CF is much lower compared to metals [12], thus requires a greater volume fraction of carbon fibers to achieve the same degree of shielding

effectiveness. Unfortunately, with an increase of CF amount, despite improvement in EMI shielding, the toughness of the composites, as well as the rheological properties of the fibers in extrusion or injection molding are negatively affected. Moreover, the increase in filler amount means an increase in production cost due to the high CF cost. The carbon fibers filled polymer therefore unable to meet the demands of shielding materials for highly demanding applications [13].

In order to reduce the fiber content and still preserve a satisfactory shielding effect, an electron beam (EB) irradiation technique can be used. The technique is usually applied to increase cross-linking of the composite matrices [14,15], which enhances mechanical properties of the irradiated material. Moreover, EB radiation also boosts excitation reactions on the fiber-matrix interfaces, which is useful in improving adhesion property. EB irradiation technique is commonly preferred owing to many advantages such as fast reaction time, low temperatures and low emission of volatile materials [16,17]. Moreover, it can be used to induce chemical and grafting reactions at any temperature in the gas, liquid, and solid phases without the use of catalysts [18,19]. Some studies have been made on the electrical and shielding properties of carbon fibers filled polymeric composites. However, to our knowledge, no study has focused on comparative performance regarding electrical conductivity and shielding effectiveness between differently treated recycled carbon fibers, i.e, untreated carbon fibers, chemically treated carbon fiber, and chemically-mechanically treated carbon fiber reinforced ABS composites. The objective of this study is to investigate how different treatments of recycled CF including EB irradiation affect the electrical conductivity and shielding effectiveness of recycled CF reinforced ABS composites.

2.0 MATERIALS AND METHODS

2.1 Materials

The matrix material used in this study was acrylonitrile butadiene styrene (ABS). Waste carbon fiber prepregs were used as the source of recycled carbon fibers (rCF) and supplied by a local composite company. The prepregs were made of carbon fiber reinforced epoxy resin. Their specific type is not reported because of a confidential agreement. They were used in three forms, i.e., as-received CF prepreg, chemically treated CF and chemically-mechanically treated CF. The chemicals used to treat the recycled carbon fibers were nitric acid (68%) and ethanol (95%).

2.2 Chemical and Mechanical Modification of rCF

The prepregs of recycled carbon fiber were first cut into smaller sizes of about 3 mm, before underwent the process of chemical modification by nitric acid treatment at 115 °C for 40 minutes to remove the resins on the surface. The fibers were then rinsed for a half hour with distilled water to achieve the same pH value as the distilled water (pH = 5.5-6). Then, the sample was soaked in an ultrasonic cleaner with ethanol for 1 hour. The obtained recycled carbon fibers (rCF) were dried in an oven at 60 °C for 24 h to remove the moisture.

The mechanical treatment was performed via pulverization technique. After chemical treatment and drying in the oven, the treated carbon fibers were pulverized to produce fine sizes of rCF. Sieve shaker was used to filter the fibers produced after the pulverization process. In this study, the fiber size obtained through the pulverizer was about 90 μ m. The characterisation of the chemically and mechanically treated recycled carbon fibers has been reported in our previous work [20].

2.3 Fabrication of rCF/ABS Composites

At this stage, rCF and ABS were melt-mixed using an internal mixer machine Haake Rheomix OS, at a screw speed of 25 rpm, followed with extrusion. Then, the mixtures were cooled and cut into composite master batches. Prior to the compounding process, crushing process by crusher machine TW-SC-400F was performed to obtain smaller size composite pallets. Then, hot pressing machine was used to form the samples of ABS/rCF composites.

2.4 Electron Beam Irradiation of ABS/rCF Composites

All samples of ABS/rCF composites were exposed to electron beam (EB) irradiation at 200 kGy dosage to assess the effects of EB radiation on the properties of the composites. EB irradiation (NHV EPS-3000) was used to develop EB irradiation induced rCF reinforced ABS composites as potential EMI shielding materials. This technique was applied as to enhance the electrical conductivity of the composites.

2.5 Characterization and Testing

Thermal analysis of the composites was performed by using thermogravimetric analysis (TGA Q50, TA Instruments). The analysis

was conducted from ambient temperature to 800 °C at a heating rate of 10 °C/min. The morphologies of the surface of non-irradiated and irradiated rCF/ABS composites were observed by using SEM (ZEISS EVO 50). The electrical conductivity was evaluated by measuring the surface resistivity (Ω /sq) of each sample by using four-probe meter. Shielding effectiveness was measured using vector network analyzer (VNA) instrument. The frequency range of scattering parameters was 8-12 GHz applied to the 2 mm thick samples using waveguide set up. The average value of three results was reported with percentage error of less than 5%.

3.0 RESULTS AND DISCUSSION

3.1 Morphological Analysis of rCF/ABS Composites

The surface morphologies of the samples of non-irradiated and irradiated rCF/ABS composites are shown in Figure 1. It is observed that non-irradiated rCF dispersed in polymer matrix exhibits significant amount of interfacial failures. This shows that deformation of the ABS resin matrix is restricted, and it can also be seen that the fibers are debonded. Furthermore, the edge of the void is clear, indicating weak adhesion interaction between the ABS and rCF. This result is very similar to that of the POM/CF [21]. Most of the fibers were pulled out from the matrix during deformation, therefore there is no significant adhesion between the rCF and ABS matrix. Upon EB irradiation, the rCF disperses in the ABS matrix shows changes in surface morphology. It was observed that there is a significant damage in the polymeric structure after irradiation, which contributes to the reduction of crystallinity in the material. The SEM images indicate that there is improvement in the interfacial adhesion between rCF and the polymer matrix in the irradiated samples.

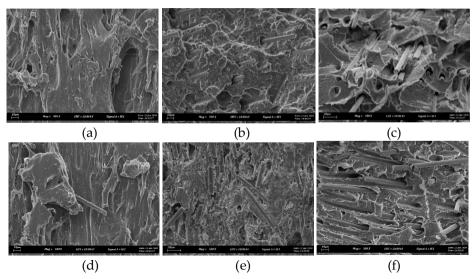


Figure 1: SEM Images for ABS/rCF (95/5) composites (500x) with Nonirradiated: (a) Untreated, (b) Chemically-mechanically treated, (c) Chemically treated rCF/ABS composite; and Irradiated: (d) Untreated, (e) Chemicallymechanically treated, (f) Chemically treated rCF/ABS composite

3.2 Electrical Properties of rCF/ABS Composites

Figure 2(a) displays the plots of electrical conductivity of nonirradiated differently treated rCF/ABS composites. It is observed that the electrical conductivity of non-irradiated samples increases with the increase of filler amount, and this type of behaviour is mainly due to the formation of conductive networks associated with the rCF. Composites with treated rCF consistently show higher conductivity than those filled with untreated rCF. For the chemically treated rCF/ABS composites, electrical conductivity shows a significant increase by four orders of magnitude (from 10⁻¹⁵ to 10⁻¹¹) at 25 wt% of rCF. However, there is no significant increase when the filler is further increased to 30 wt%. For the chemically-mechanical treated rCF/ABS composites, the electrical conductivity shows a significant increase by four orders of magnitude (from 10⁻¹³ to 10⁻¹⁰) at 20 wt% of rCF. However, the value is decreased slightly by one order of magnitude $(10^{-10} - 10^{-11})$ with increasing filler concentration (25 and 30 wt%). For the composites with untreated rCF, electrical conductivity shows the least increase with increasing fiber content.

Electrical and Shielding Properties of Acrylonitrile Butadiene Styrene Filled with Differently Treated Recycled Carbon Fibers

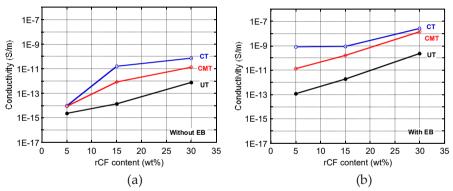


Figure 2: Electrical conductivity of differently treated ABS/rCF composites (a) without EB irradiation and (b) with EB irradiation

Figure 2(b) shows the plots of electrical conductivity of 200 kGy EB irradiated differently treated rCF/ABS composites. It is observed that regardless of the amount of rCF loading, the conductivity shows a respectable increase with EB irradiation, that is due to the increase in the continuous, interconnected conductive networks with EB dose in the ABS matrix, through which the charge particles able to move easily when external electric field is applied [22]. The EB irradiated chemically treated rCF/ABS composite show the highest electrical conductivity, achieving 2.68 × 10⁻⁸ (S/m) at 30 wt% rCF content. Meanwhile, the highest improvement due to EB irradiation is shown by the chemically-mechanically treated rCF/ABS composites, in which electrical conductivity increases from 1.36×10^{-11} to 1.34×10^{-8} (S/m) at 30 wt% rCF content. As for the irradiated untreated rCF/ABS composites, the electrical conductivity displays a humble increase to 2.32×10^{-10} (S/m) at 30 wt% rCF.

The lack of increase in electrical conductivity at 30 wt% rCF in the nonirradiated samples can be associated with the agglomeration of rCF in ABS, which hinders percolation, thus decreases the electrical conductivity of the composite. The dispersion level of rCF is essential in determining the final electrical properties of ABS composites. Upon EB irradiation, better dispersion level of rCF at 30 wt% in the composites is achieved, thus leads to formation of an effective network for electron path transmittance, which explains the higher electrical conductivity obtained in the irradiated ABS/rCF composites.

3.3 Electromagnetic Interference Shielding Effectiveness of Differently Treated rCF/ABS Composites

The results of shielding effectiveness (SE) for the non-irradiated and irradiated ABS composites as a function of frequency are shown in Figure 3. It is shown that increasing the amount of rCF causes SE value to increase. At higher frequency, the increase in SE can reach as high as 50% when the amount of rCF is increased from 10 to 30 wt%. The increase of SE with increasing frequency is anticipated and has been reported elsewhere [8]. The wavelength of the EM wave decreases as frequency increases and approaching the size o f the fiber. Thus, the higher frequency waves are more probably to encounter fibers distributed in the polymer matrix. Hence, the increase of the SE value as frequency increases.

Figure 4 shows the values of EMI SE for non-irradiated and irradiated rCF filled ABS composites. It is observed that for all the composites studied, irradiated composites samples exhibit better SE performance compared to the samples without EB irradiation. The non-irradiated ABS composite filled with 5 wt% of untreated rCF exhibits the lowest EMI SE and with increasing rCF loading, EMI SE value increases. Meanwhile, the irradiated ABS composite filled with chemically treated rCF at 30 wt% shows the highest EMI SE (46.13 dB) in the frequency range of 8 to 12 GHz. This is an increase of 41.5% compared to the same composite without EB irradiation. The results can be attributed to the formation of interconnected conductive networks by rCF in the composites due to the EB irradiation. The results of the EB irradiation effect on the SE of ABS/rCF is significant as it has never been examined previously, thus provides technical information necessary for further development of the material as an effective shielding material.

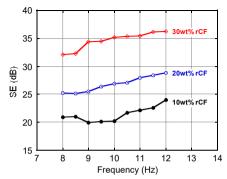


Figure 3: Frequency dependence of EMI SE for rCF/ABS composites containing various amount of rCF

Electrical and Shielding Properties of Acrylonitrile Butadiene Styrene Filled with Differently Treated Recycled Carbon Fibers

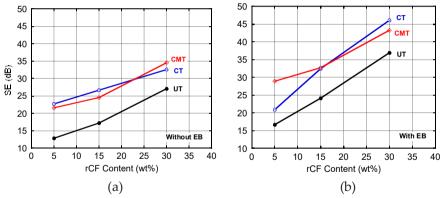


Figure 4: EMI SE for rCF/ABS composites (a) without EB irradiation and (b) with EB irradiation

The effect of EB irradiation dosage on the EMI shielding effectiveness of rCF/ABS blends has also been observed (not shown here). It has been found that EMI SE increases with the increase of EB irradiation dosage. Irradiation promotes the establishment of more interconnected conductive networks resulting from the formation and recombination of free radicals in the polymer-filler interfaces, which facilitates the movement of the mobile charge carriers in the rCF/ABS composites. The conductive fillers interact with the incoming electromagnetic radiation and facilitates the electron transport which is known as microwave absorption across the EMI shielding material by the conductive networks. This is effective in reducing the reflection and transmission of incident radiation. The irradiated ABS composites filled with treated rCF exhibit good electrical conductivity values, which is a determinant factor in a material to behave as an excellent shielding material, in order to be able to weaken the incident electromagnetic radiation and functions as an effective EMI shielding material.

4.0 CONCLUSION

The objective of the study, which is to investigate the effects of different treatments including chemical treatment of the rCF and electron beam irradiation on the ABS/rCF composites, on conductivity and SE effectiveness have been successfully achieved. It is observed that electrical conductivity of ABS/rCF composites increase with the increase in the rCF content. Treatment on the rCF also contributes positively to the improvement of the electrical conductivity and shielding effectiveness. The effects of EB irradiation on the electrical

conductivity and SE have also been investigated. Both electrical conductivity and EMI shielding effectiveness improve significantly with EB irradiation. This might be attributed to the breakage of chemical bonds and resulting in an increase of free radicals. An irradiated chemically treated rCF/ABS composite has shown the highest electrical conductivity reaching 1.34×10^{-8} (S/m) and EMI SE reaching 46.13 dB at 30 wt% of rCF content. It is shown in this study that recycled CF is a feasible material for the production of second-generation functional composites for EMI shielding application.

ACKNOWLEDGMENTS

The authors would like to acknowledge Universiti Teknikal Malaysia Melaka (UTeM) for the short-term grant, (PJP/2019/FKP(1A)/S01664) and for supporting this project through MOA on recycled carbon fiber between UTeM, USM and Malaysian Nuclear Agency.

REFERENCES

- [1] S. Ghosh, S. Ganguly, A. Maruthi, S. Jana, S. Remanan, P. Das, T.K. Das, S.K. Ghosh and N.C. Das, "Micro-computed tomography enhanced cross-linked carboxylated acrylonitrile butadiene rubber with the decoration of new generation conductive carbon black for high strain tolerant electromagnetic wave absorber", *Materials Today Communications*, vol. 24, pp. 1-13, 2020.
- [2] L. Vazhayal, P. Wilson and K. Prabhakaran, "Waste to wealth: Lightweight, mechanically strong and conductive carbon aerogels from waste tissue paper for electromagnetic shielding and CO2 adsorption", *Chemical Engineering Journal*, vol. 381, pp. 1-12, 2020.
- [3] J. Kang, D. Kim, Y. Kim, J.B. Choi, B.H. Hong and S.W. Kim, "Highperformance near-field electromagnetic wave attenuation in ultra-thin and transparent graphene films", *2D Materials*, vol. 4, no. 2, pp. 1-10, 2017.
- [4] S. Kashi, R.K. Gupta, T. Baum, N. Kao and S.N. Bhattacharya, "Dielectric properties and electromagnetic interference shielding effectiveness of graphene-based biodegradable nanocomposites", *Materials & Design*, vol. 109, pp. 68–78, 2016.
- [5] R. Ram, M. Rahaman and D. Khastgir, "Electromagnetic interference (EMI) shielding effectiveness (SE) of polymer-carbon composites", in *Carbon-Containing Polymer Composites*, S. Kalia. Springer Singapore, 2019, pp. 339–368.

- [6] Subhash B. Kondawar and Prema R. Modak, "Theory of EMI Shielding", in *Materials for Potential EMI Shielding Applications: Processing, Properties and Current Trends, J. Kuruvilla, R. Wilson and G.* Gejo, Elsevier, 2020, pp. 9-23.
- [7] D.D.L. Chung, "Electromagnetic interference shielding effectiveness of carbon materials", *Carbon*, vol. 39, no. 2, pp. 279–285, 2001.
- [8] L.C. Jia, L. Xu, F. Ren, P.G. Ren, D.X. Yan and Z.M. Li, "Stretchable and durable conductive fabric for ultrahigh performance electromagnetic interference shielding", *Carbon*, vol. 144, pp. 101–108, 2019.
- [9] H. Gaub, "Customization of mass-produced parts by combining injection molding and additive manufacturing with Industry 4.0 technologies", *Reinforced Plastics*, vol. 60, no. 6, pp. 401–404, 2016.
- [10] S.J. Park and M.K. Seo, "Carbon fiber-reinforced polymer composites: Preparation, Properties, and applications", *Polymer Composites*, vol. 1, no. 11, pp. 135–184, 2012.
- [11] H. Dhieb, J.G. Buijnsters, K. Elleuch and J. P. Celis, "Effect of relative humidity and full immersion in water on friction, wear and debonding of unidirectional carbon fiber reinforced epoxy under reciprocating sliding", *Composites Part B: Engineering*, vol. 88, pp. 240–252, 2016.
- [12] D.M. Bigg, "Mechanical, thermal, and electrical properties of metal fiber-filled polymer composites", *Polymer Engineering & Science*, vol. 19, no. 16, pp. 1188–1192, 1979.
- [13] G. Lu, X. Li and H. Jiang, "Electrical and shielding properties of ABS resin shielding with nickel-coated carbon fibers", *Composites Science and Technology*, vol. 56, no. 2, pp. 193–200, 1996.
- [14] R.L. Clough, "High-energy radiation and polymers: A review of commercial processes and emerging applications", *Nuclear Instruments* and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms, vol. 185, no. 1-4, pp. 8–33, 2001.
- [15] A.J. Berejka and C. Eberle, "Electron beam curing of composites in North America", *Radiation Physics and Chemistry*, vol. 63, no. 3-6, pp. 551–556, 2009.
- [16] B. Zsigmond, L. Halász and T. Czvikovszky, "EB processing of braided carbon fiber composite profiles", *Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms*, vol. 208, pp. 247–251, 2003.
- [17] J.G. Drobny, Radiation Technology for Polymers, CRC press, 2020.
- [18] F. Zhao and Y. Huang, "Uniform modification of carbon fibers in high density fabric by γ-ray irradiation grafting", *Materials Letters*, vol. 65, no. 23-24, pp. 3351–3353, 2011.

- [19] J. Li, Y. Huang, Z. Xu and Z. Wang, "High-energy radiation technique treat on the surface of carbon fiber", *Materials Chemistry and Physics*, vol. 94, no. 2-3, pp. 315–321, 2005.
- [20] A.M. Alkaseh, M.E.A. Manaf, Z. Shamsudin, J.A. Ghafar and E. Osman, "Characterization of Chemically and Mechanically Treated Recycled Carbon Fibers", *International Journal of Advanced Science and Technology*, vol. 29, no. 9s, pp. 1341–1347, 2020.
- [21] C.Y. Huang and T.W. Chiou, "The effect of reprocessing on the EMI shielding effectiveness of conductive fibre reinforced ABS composites", *European Polymer Journal*, vol. 34, no. 1, pp.37–43, 1998.
- [22] S. Bhadra, N.K. Singha and D. Khastgir, "Dielectric properties and EMI shielding efficiency of polyaniline and ethylene 1-octene based semiconducting composites", *Current Applied Physics*, vol. 9, no. 2, pp. 396– 403, 2009.