Journal of Metals, Materials and Minerals, Vol. 31, No. 1, pp. 11-16, 2021

Graphene enhanced linear low-density polyethylene nanocomposites by premixing and melt compounding

Wan Nazri WAN BUSU[1,2,3,*], Ruey Shan CHEN[2,3], Muhammad Jefri MOHD YUSOF[4], and Sahrim AHMAD[2,3]

[1] *Food Science & Technology Research Center, Malaysia Agricultural Research and Development Institute, PO Box 12101, 50774 Kuala Lumpur, Malaysia*

[2] *Centre for Frontier Sciences, Faculty of Science and Technology, Universiti Kebangsaan Malaysia, 43600 Bangi, Selangor, Malaysia*

[3] *Materials Science Program, Faculty of Science and Technology, Universiti Kebangsaan Malaysia, 43600 Bangi, Selangor, Malaysia*

[4] *Department of Chemistry, Faculty of Science and Technology, Universiti Kebangsaan Malaysia, 43600 Bangi, Selangor, Malaysia*

***Corresponding author e-mail:** wannazri@mardi.gov.my

Received date:
6 June 2020
Revised date
11 November 2020
Accepted date:
12 December 2020

Keywords:
Graphene nanoplatelets;
LLDPE;
Premix;
Masterbatch;
Indirect mixing technique

Abstract

In this study, linear low-density polyethylene/graphene nanoplatelets (LLDPE/GNPs) nanocomposites were prepared from conventional melt-mixing method and our new approach; two-step premix mixing technique. Indirect mixing technique (IDT) was employed to fabricate a premix of LLDPE/GNPs in the ratio of 80:20 wt%. The effects of GNPs loadings and the processing method of nanocomposites on the mechanical strength, decomposition temperature, crystallinity, electrical impedance and morphology were investigated. Irrespective of processing methods, the prepared nanocomposites exhibited crystalline structure due to the presence of GNPs whilst the degradation temperature was recorded to be increased with GNPs loadings that signified improved thermal stability. The inclusion of GNPs provided electrical impedance ability on LLDPE matrix as a result from the formation of conductive networks of GNPs. LLDPE/GNPs nanocomposites prepared from two-step premix mixing technique showed better mechanical properties than those of melt-mixing method. Apparently, two-step premix mixing of LLDPE/GNPs nanocomposites promoted better dispersion of GNPs in matrix based on SEM images. Our findings have proved that our new, profound technique of preparing premix before mixing could result in enhanced quality of nanocomposites that are potentially useful in packaging or electronic applications.

1. Introduction

Graphene-based composites have been extensively studied and prepared as the resulting products are useful in sensing applications, high conductivity and large specific surface area [1]. The incorporation of graphene or its derivatives with a series of polymer groups exhibits emergence of composites bearing improved mechanical, thermal and electrical properties with strong flexible properties [2,3]. Preparation of these composites requires delicate approach because the properties of the end products might vary and depend on the preparation manner. Particularly in preparing nanocomposites where the dispersion of nanofillers is the main concern, selective blending style should be adopted to ensure homogeneity of nanofillers in the polymer matrices [4]. Nasir and Choo [5] compared their mechanical and cure characteristics of carbon black filled styrene-butadiene rubber/epoxidized natural rubber blends that were prepared from masterbatch technique as opposed to conventional melt-mixing method. Better degree of dispersion was achieved via masterbatch preparation than melt-mixing technique that produced blends with more consistent cure characteristics and mechanical properties.

Process modifications are another scientific milestone pursued by research scientists in many fields to allow more production of products with upgraded physical features and performances [6]. During preparation of polymeric nanocomposites, the homogeneity of nanoparticulate clusters embedded in the matrix is important to yield products with enhanced properties [7]. Baek *et al.* [8] proposed a term "clustering density" to indicate the agglomeration index of nanoparticulate. They found that when the clustering density was increased, a notable reduction in the stiffness of the nanocomposites was recorded from 10 to 30% due to homogeneity decrement. Likewise, the notion of premix homogeny can be applied to explain the findings from a study conducted by Pera and Ambroise [9], stating that their pre-mixed fibre-reinforced magnesia-phosphate cement composites displayed significant mechanical strength.

In this study, we report on the comparison of physico-mechanical properties of linear low-density polyethylene (LLDPE) and graphene nanoplatelets (GNPs) nanocomposites prepared from different techniques (i.e two-step premix mixing versus conventional melt-mixing). This novel two-step premix mixing method acted as masterbatch-like material to prepare LLDPE/GNPs nanocomposites

DOI: 10.14456/jmmm.2021.2

at certain GNPS percentages to tally the GNPs compositions (1-5%) in nanocomposites prepared from conventional melt-mixing method [10]. GNPs in premix condition were expected to become a better nucleating agent in the later blending than one-pot melt-mixing method that subsequently could yield in better dispersion of GNPs in LLDPE matrix. This has been demonstrated by Morin [11] who developed enhanced polypropylene of stretching resistance, increased crystallinity and tenacity, thanks to the presence of nucleating agent used. Previously, similar concept of this study's hypothesis has been presented by past studies. For instance, in situ fusion of thermoplastic liquid resin is required before extrusion to achieve stronger biocomposites [12]. Extrusion of preimpregnated fibers [13] and in situ fusion of fibers with thermoplastics in molten state at the nozzle [14] will both produce composites with enhanced properties. Based on past studies, GNPs as nanofillers in the nanocomposites demand high degree of dispersion in order to execute products with enhanced properties.

This study aimed to explore the new LLDPE/GNPs nanocomposites preparation method by introducing two-step of melt-mixing and comparing the resulting properties with nanocomposites from conventional one-step melt-mixing method. In this new technique, the first step of melt-mixing is called premix that contained high amount of GNPs (20%) that would be subsequently diluted in the second step of melt-mixing. Improvement of GNPs dispersion in LLDPE matrix was expected to be achieved in this new, simple technique that will distinguish it from conventional melt-mixing method.

2. Experimental

2.1 Materials

LLDPE (density: 0.918 g·cm^{-3}, melt flow index: 1.0 g·min^{-1}, melting temperature: 120-160°C) was purchased from Exxon Mobile Chemical Corporation to be used as thermoplastic resin. GNPs (KNG-150) with thickness of 5-15 nm, 5 µm diameter, and density of 2.25 g·cm^{-3} were supplied by KNANO.

2.2 Preparation LLDPE/GNPs Nanocomposites

2.2.1 Melt-mixing method

LLDPE was dried overnight in a vacuum oven at 80°C to prevent hydrolytic degradation during melt processing in the mixer. About 55 g of LLDPE.GNPs nanocomposites were obtained using melt-mixing method LLDPE in internal mixer (Brabender W50EHT, Germany) at 140°C, 40 rpm for 13 min. LLDPE was added first and allowed to be melt-mixed for 5 min. Next, GNPSs were slowly added. The GNP loadings were varied at 1, 2, 3, 4 and 5 wt%.

2.2.2 Two-step premixing

Preparation of LLDPE/GNPs via this technique was similar with melt-mixing method except that there was an introduction of indirect mixing technique (IDT). IDT was used to make a premix of LLDPE/GNPs. The premix was prepared in the ratio of 80:20 wt% of LLDPE:GNPs in Brabender internal mixer. This nanocomposite with such ratio acted as masterbatch-like material to produce another sets of LLDPE/GNPs at different percentages of GNPs (1-5 wt%). The blend was discharged into the mixer for 3 min before LLDPE/GNPs pre-mixture. To obtain LLDPE/GNPs nanocomposites with certain percentage of GNPs, LLDPE was added to dilute the so-called masterbatch (Table 1). The next procedures followed the same routines as melt-mixing method.

2.3 Composite sheets preparation

Next, hot and cold press process (14 x 14 x 1 cm spacer mold with temperature 180°C for upper and lower platens, compression pressure of 1 kpsi) (LP50 LABTECH Engineering Company) was used to compress the blends into sheets.

2.4 Characterization

2.4.1 Mechanical properties

ASTM D 638-03 protocol (Testometric M350-10CT with 10 kN load cell) was used to measure the tensile stress at break (MPa) of the specimens. At least three specimens with each dimension of 165 mm length, 19 mm width and 3 mm thickness were used to get the average value. Impact test was conducted using Izod GOTECH GT7045 impact protocol according to ASTM D256. The impact energy (kJ·m^{-2}) was obtained for the average at least three specimens by calculating the difference in the potential energy of pendulum before and after it was latched.

2.4.2 Thermal stability

To study the thermal stability of the nanocomposites, thermogravimetric analysis (TGA) and X-ray diffraction (XRD) were conducted to observe the decomposition temperature and crystallinity/amorphous segments of the nanocomposites, respectively, to see the effects of GNPs dispersion in LLDPE matrix. Thermogravimetric Analyzer (Mettler Toledo SDTA 851e model) was used to analyze the decomposition temperature of the samples. The analysis was performed by STARe automated computer software. Meanwhile, Bruker AXS Germany D8 Advance model was used in XRD analysis with EVA as the running software.

Table 1. Dilution of LLDPE/GNPs nanocomposites from 80:20 wt% premix.

Loading of GNPs In LLDPE/GNPs (wt%)	Amount of LLDPE (g)	Amount of premix (g)
1	52.25	2.75
2	49.80	5.50
3	46.75	8.25
4	44.00	11.00
5	41.25	13.75

2.4.3 Electrical conductivity

The electrical resistance of LLDPE/GNPs was measured via complex impedance method using high frequency resonance analyzer (HFRA) solvent analyzer (Solartron Model 1255). The upper and lower surfaces of the plaques were coated with silver paint to ensure good electrical conductivity. Average resistance, sample thickness and electrodes area were recorded to calculate the electrical conductivity (S·cm^{-1}) using the formula:

$$\text{Electrical conductivity} = 1/\text{resistivity of specimen, } p \qquad (1)$$

$$\text{Resistivity of specimen, } p = RA/t \qquad (2)$$

where R is the resistance of specimen, A is the area of circle specimen and t is the thickness of specimen.

2.4.4 Morphology

The tensile-fractured surface of the specimens was used to study the morphology of the nanocomposites using scanning electron microscope (SEM Phillips XL30) at 15 kV voltage up to 5000x magnification. Samples were coated with thin layer of gold before being viewed by SEM.

3. Results and Discussion

3.1 Mechanical Properties

Figure 1 shows the comparison of mechanical strengths of LLDPE/GNPs nanocomposites prepared from two-step premix mixing technique and melt-mixing method. The tensile strength of the nanocomposites was in the range of 26.49-30.04 MPa and 29.62-31.88 MPa, with 2% and 4% GNPs were the maximum MPa for melt-mixing method and two-step premix mixing technique, respectively. In this new approach of two-step premix mixing, the increment of GNPs loading exhibited increase in mechanical strength. The impact energy of LLDPE/GNPs prepared from this new technique (39.57-42.91 kJ·m^{-2}) was always higher than those prepared from conventional melt-mixing method (30.86-36.07 kJ·m^{-2}). Similar findings were reported by Beretta *et al* [15] where the increment of carbon nanotubes (CNTs) percentages in their polyether ether ketone (PEEK) composite filaments yielded in better mechanical strength but dropped at CNTs > 5%. Molecular-level dispersion of GNPs aided in strong interfacial adhesion that can be attributed to the better tensile strength [16].

Especially for two-step mixing method, there was less agglomeration of GNPs from double extrusion time of LLDPE with GNPs, so they could be well dispersed resulting in higher mechanical strength. In method comparison study of preparing exfoliated graphite–polypropylene (xGnP-15/PP) nanocomposites by Kalaitzidou *et al.* [17], they discussed that melt-mixing alone has no enough shear to break down the xGnP-15 agglomerates and homogeneously disperse the graphite platelets when compared with in situ and premix techniques.

Meanwhile, for both methods, certain high amount of nanofillers (4-5% in this study), the interaction between the reinforcement (GNPs) and the polymer matrix during mixing would introduce air gap that led to detrimental mechanical properties.

3.2 Thermal Properties

TGA analysis was performed to investigate the decomposition temperature of LLDPE/GNPs nanocomposites. With the increasing amount of GNP loadings, it was found that the thermal stability of the nanocomposites was improved (Figure 2). LLDPE/GNPs nanocomposites prepared from two-step premix mixing displayed slightly higher and more consistent decomposition temperature than conventional melt-mixing method. Similar findings were reported by Maiti *et al.* [18] who performed TGA analysis on polyhydroxybutyrate/layered silicate nanocomposites. They explained that the intercalation of well dispersed fillers in polymer matrix would discriminate the decomposition temperature of same materials.

When the percentage of GNPs was lowered, isolated GNPs did not necessarily form a continuous network and conductivity pathway that was responsible for high thermal resistance [19]. Khanam *et al.* [20] described a phonon scattering phenomenon to explain poor interaction between GNPs and LLDPE at low amount of GNPs loading [21]. GNPs raised the decomposition temperature of LLDPE by acting as thermal barrier to hinder the degradation of LLDPE.

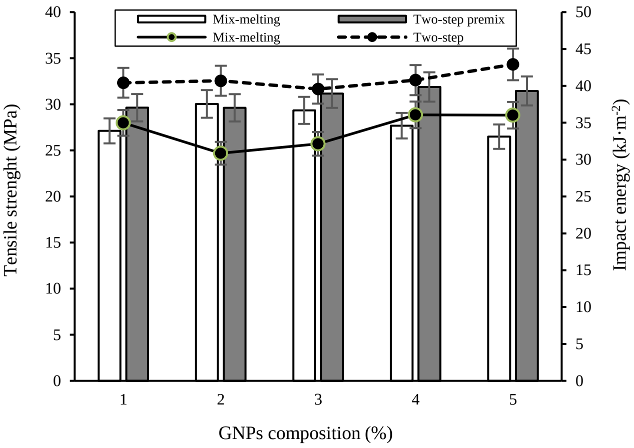

Figure 1. Mechanical properties of LLDPE/GNPs nanocomposites

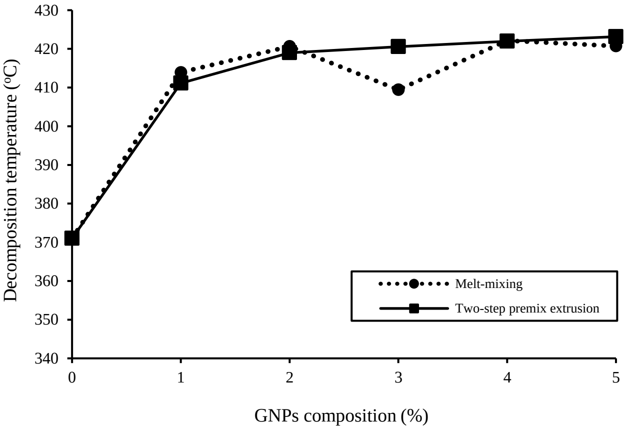

Figure 2. Decomposition temperature of LLDPE/GNPs nanocomposites.

3.3 XRD Analysis

XRD analysis is usually used to check on the amorphous and crystallinity segments of a material. Figure 3 shows the diffractogram pattern of LLDPE/GNPs nanocomposite. GNPs have been reported to increase the crystallinity of resulting products as it reduces the co-monomer incorporation [22]. High aspect ratio and GNPs distribution would cause stronger interphase bonding between the LLDPE matrix and GNPs, hence explaining the manifestation of the crystalline peaks [23]. The nucleating action of GNPs promotes in the increment of crystallinity [24].

All samples however exhibited identical pattern of diffractogram that produced several peaks indicating that crystalline structure was formed as a result of GNPs presence. It is noted that the same diffractogram pattern for all samples regardless the preparation method as they consisted of the same components of LLDPE and GNPs. Similarly, this observation has also been reported by Ngo *et al.* [25] who recorded identical peaks appearance to indicate crystallinity of their tested clay/epoxy nanocomposites, regardless the preparation manners.

3.4 Electrical Conductivity

The electrical impedance of LLDPE/GNPs nanocomposites is tabulated in Table 2. Generally, the addition of GNPs in LLDPE matrix directly proportionated the electrical conductivity of these nanocomposites. The formation of conductive network by graphene had converted the electric-insulating behavior of LLDPE into a significant electrical conductor in all samples [26,27]. At high percentages of GNPs, the low electrical conductivity could be attributed to the saturation level of GNPs content that displayed a slower rate in the conductivity channel of the nanocomposites [28].

Better dispersion of GNPs in two-step premix method was manifested when the electrical impedance values of the samples were higher than samples prepared from melt-mixing method. This is in agreement with a study by Potts *et al.* [29] that stated that the difference in electrical properties of their thermally-exfoliated graphite oxide/natural rubber nanocomposites was mainly the result of enhanced dispersion.

3.5 Morphology

Better dispersion of GNPs in polymer matrices always results in improved physical properties of the nanocomposites as it can be evident by SEM images [30]. Figure 4 displays the comparison of GNPs dispersion in melt-mixing method and two-step premix mixing technique for selected LLDPE/GNPs nanocomposites. This observation is convenient to explain the disparity properties exhibited by nanocomposites prepared from both procedures as two-step premix mixing yielded in significantly better physico-mechanical properties of nanocomposites than melt-mixing method [31].

It can be deduced that nanocomposites prepared from two-step mixing technique contained more dispersed GNPs that incorporated in the LLDPE matrix. The premix stage in this two-step premix mixing offered better homogeneity of GNPs in the LLDPE matrix. Premix condition is beneficial to ensure homogeneity as it prevents segregation of components during processing [32,33]

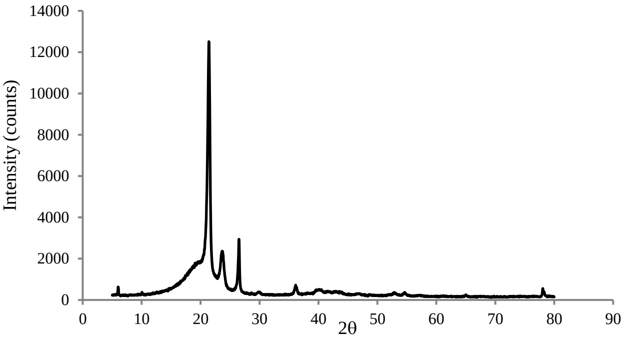

Figure 3. Diffractogram pattern of LLDPE/GNPs nanocomposites 1 µm.

Table 2. Electrical impedance of LLDPE/GNPs nanocomposites.

Samples	Resistivity (Ω)		Conductivity (S·cm^{-1})	
	Melt-Mixing	Two-step premix	Melt-mixing	Two-step premix
0 wt% GNP	2.44E+09	2.30E+08	4.09E-10	4.09E-10
1 wt% GNP	2.18E+07	7.56E+07	4.86E-09	1.32E-08
3 wt% GNP	1.96E+07	8.96E+07	5.43E-09	1.12E-08
4 wt% GNP	2.04E+07	9.81E+07	5.21E-09	1.02E-08
5 wt% GNP	2.33E+07	1.17E+08	4.55E-09	8.55E-09

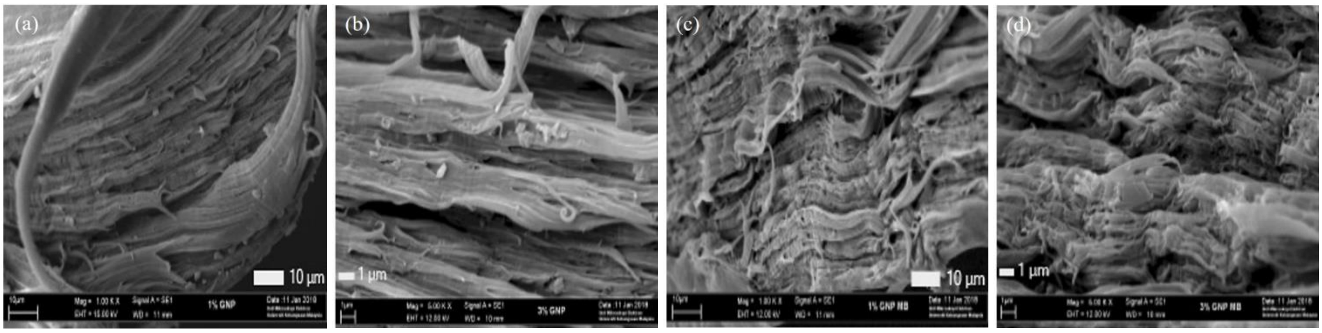

Figure 4. SEM images of LLDPE/GNPS nanocomposites prepared from melt-mixing (a) 1%, (b) 3% and two-step premix mixing (c) 1%, (d) 3%.

4. Conclusion

We have successfully demonstrated a new, simple approach of preparing LLDPE/GNPs nanocomposites by introducing premix of those materials before mixing. The physico-mechanical properties of the resulting nanocomposites were better than using conventional melt-mixing method, mostly in terms of their mechanical strength, thermal properties and morphology. The premix nature of our new technique helped in better dispersion of GNPS in the LLDPE matrix. The efficiency of this two-step premix mixing can be explored further for other nanocomposites preparation to achieve superior end products.

Acknowledgement

We would like to thank Universiti Kebangsaan Malaysia (UKM) for funding this experiment under FRGS grant (FRGS/1/2018/STG07/UKM/01/2) and Centre for Research and Instrumentation (CRIM) UKM for providing instrumentations and facilities.

References

[1] A. Popelka, P.N. Khanam, and M.A. AlMaadeed, "Surface modification of polyethylene/graphene composite using corona discharge," Journal of Physics D: Applied Physics, vol. 51(10), pp. 105302, 2018.

[2] H. Kim, S. Kobayashi, M.A. Abdurrahim, M.J. Zhang, A. Khusainova, M.A. Hillmyer, A.A. Abdala, and C.W. MacOsko, "Graphene/polyethylene nanocomposites: effect of polyethylene functionalization and blending methods," Polymer, vol. 52(8), pp. 1837-1846, 2011.

[3] T. Kuilla, S. Bhadra, D. Yao, N.H. Kim, S. Bose, and J.H. Lee, "Recent advances in graphene based polymer composites," Progress in Polymer Science, vol. 35(11), pp. 1350-1375, 2010.

[4] M.F.H.M. Ruf, S. Ahmad, R.S. Chen, D. Shahdan, and F. Zailan, "Liquid natural rubber toughened poly(Lactic acid) blend: Effects of compatibilizer types and loadings on thermo-mechanical properties," Malaysian Journal of Analytical Sciences, vol. 22, pp. 885-891, 2018.

[5] M. Nasir, and C.H. Choo, "Cure characteristics and mechanical properties of carbon black filled styrene-butadiene rubber and epoxidized natural rubber blends," European Polymer Journal, vol. 25(4), pp. 355-359, 1989.

[6] G.D. Goh, Y.L. Yap, S. Agarwala, and W.Y. Yeong, "Recent progress in additive manufacturing of fiber reinforced polymer composite," Advanced Materials Technologies, vol. 4(1), pp. 1800271, 2019.

[7] H. Shin, K. Baek, J.G Han, and M. Cho, "Homogenization analysis of polymeric nanocomposites containing nanoparticulate clusters," Composites Science and Technology," vol. 138, pp. 217-224, 2017.

[8] K. Baek, H. Shin, T. Yoo, and M. Cho, "Two-step multiscale homogenization for mechanical behaviour of polymeric nanocomposites with nanoparticulate agglomerations," Composites Science and Technology, vol. 179, pp. 97-105, 2019.

[9] J. Pera, and J. Ambroise, "Fiber-reinforced magnesia-phosphate cement composites for rapid repair," Cement and Concrete Composites, vol. 20(1), pp. 31-39, 1998.

[10] P.N. Khanam, M.A. AlMaadeed, M. Ouederni, E. Harkin-Jones, B. Mayoral, A. Hamilton, and D. Sun, "Melt processing and properties of linear low density polyethylene-graphene nanoplatelet composites," Vacuum, vol. 130, pp. 63-71, 2016.

[11] B.G. Morin, "Melt-spun multifilament polyolefin yarn formation processes and yarns formed therefrom," Google Patents, Nov. 04, 2008.

[12] Y. Nakagawa, K. Mori, and T. Maeno, "3D printing of carbon fibre-reinforced plastic parts," The International Journal of Advanced Manufacturing Technology, vol. 91(5-8), pp. 2811-2817, 2017.

[13] G.W. Melenka, B.K.O. Cheung, J.S. Schofield, M.R. Dawson, and J.P. Carey, "Evaluation and prediction of the tensile properties of continuous fiber-reinforced 3D printed structures," Composite Structures, vol. 153, pp. 866-875, 2016.

[14] K. Mori, T. Maeno, and Y. Nakagawa, "Dieless forming of carbon fibre reinforced plastic parts using 3D printer," Procedia Engineering, vol. 81, pp. 1595-1600, 2014.

[15] S. Berretta, R. Davies, Y.T. Shyng, Y. Wang, and O. Ghita, "Fused Deposition Modelling of high temperature polymers: Exploring CNT PEEK composites," Polymer Testing, vol. 63, pp. 251-262, 2017.

[16] T. Kuila, S. Bose, C.E. Hong, M.E. Uddin, P. Khanra, N.H. Kim, and J.H. Lee, "Preparation of functionalized graphene/linear low density polyethylene composites by a solution mixing method," Carbon, vol. 49(3), pp. 1033-1037, 2011.

[17] K. Kalaitzidou, H. Fukushima, and L.T. Drzal, "A new compounding method for exfoliated graphite–polypropylene nanocomposites with enhanced flexural properties and lower percolation threshold," Composites Science and Technology, vol. 67(10), pp. 2045-2051, 2007.

[18] P. Maiti, C.A. Batt, and E.P. Giannelis, "New biodegradable polyhydroxybutyrate/layered silicate nanocomposites," Biomacromolecules, vol. 8(11), pp. 3393-3400, 2007.

[19] M.E. Achaby, F. Arrakhiz, S. Vaudreuil, A.K. Qaiss, M. Bousmina, and O. Fassi-Fehri, "Mechanical, thermal, and rheological properties of graphene-based polypropylene nanocomposites prepared by melt mixing," Polymer Composites, vol. 33(5), pp. 733-744, 2012.

[20] P.N. Khanam, M.A. AlMaadeed, M. Ouederni, B. Mayoral, A. Hamilton, and D. Sun, "Effect of two types of graphene nanoplatelets on the physico–mechanical properties of linear low–density polyethylene composites," Advanced Manufacturing: Polymer & Composites Science, vol. 2(2), pp. 67-73, 2016.

[21] S.M. Ardekani, A. Dehghani, M.A. Al-Maadeed, M.U. Wahit, and A. Hassan, "Mechanical and thermal properties of recycled poly-(ethylene terephthalate) reinforced newspaper fiber composites," Fibers Polym., Fibers and Polymers, vol. 15(7), pp. 1531-1538, 2014.

[22] N. Nabizadeh, G.H. Zohuri, M. Khoshsefat, N. Ramezanian, and S. Ahmadjo, "Ethylene/1-Hexene Copolymerization and Synthesis of LLDPE/Nanocarbon Composite through In Situ Polymerization," Polymer Science, Series B, vol. 60(1), pp. 122-129, 2018.

[23] K. Zhou, W. Yang, G. Tang, B. Wang, S. Jiang, Y. Hu, and Z. Gui., "Comparative study on the thermal stability, flame retardancy and smoke suppression properties of polystyrene composites containing molybdenum disulfide and graphene," RSC Advances, vol. 3(47), pp. 25030-25040, 2013.

[24] N.E. Zakaria, I. Ahmad, W.N.W. Busu, H. Khalid, and A. Baharum, "Kesan Penambahan Kepingan Nanozarah Grafin terhadap Sifat Mekanik dan Terma Hibrid Komposit Serabut Sansevieria-Getah Asli-Polietilena Berketumpatan Tinggi," Sains Malaysiana, vol. 48(5), pp. 1121-1128, 2019.

[25] T.D Ngo, M.T. Ton-That, S.V. Hoa, and K.C. Cole, "Effect of temperature, duration and speed of pre-mixing on the dispersion of clay/epoxy nanocomposites," Composites Science and Technology, vol. 69(11-12), pp. 1831-1840, 2009.

[26] P.N. Khanam, M.A. AlMaadeed, S. AlMaadeed, S. Kunhoth, M. Ouederni, D. Sun, A. Hamilton, E.H. Jones, and B. Mayoral, "Optimization and prediction of mechanical and thermal properties of graphene/LLDPE nanocomposites by using artificial neural networks," International Journal of Polymer Science, vol. 2016, 2016.

[27] V. Sridhar, I. Lee, H.H. Chun, and H. Park, "Graphene reinforced biodegradable poly(3-hydroxybutyrate-co-4-hydroxybutyrate) nano-composites.," Express Polymer Letters, vol. 7(4), 2013.

[28] F.D. Zailan, R.S. Chen, D. Shahdan, and S. Ahmad, "Effect of conductive polyaniline in thermoplastic natural rubber blends on the mechanical, thermal stability, and electrical conductivity properties," Journal of Applied Polymer Science, vol. 136(20), pp. 47527, 2019.

[29] J.R. Potts, O. Shankar, S. Murali, L. Du, and R.S. Ruoff, "Latex and two-roll mill processing of thermally-exfoliated graphite oxide/natural rubber nanocomposites," Composites Science and Technology, vol. 74, pp. 166-172, 2013.

[30] H. Algul, M. Tokur, S. Ozcan, M. Uysal, T. Cetinkaya, and H. Akbulut, "The effect of graphene content and sliding speed on the wear mechanism of nickel–graphene nanocomposites," Applied Surface Science, vol. 359, pp. 340-348, 2015.

[31] F.D. Zailan, R.S. Chen, S. Ahmad, D. Shahdan, A.M. Ali, and M.F.H. Mohd Ruf, "Blends of linear low-density polyethylene, natural rubber and polyaniline: Tensile properties and thermal stability," Malaysian Journal of Analytical Sciences, vol. 22(6), pp. 999-1006, 2018.

[32] J.E. Japka. "Carbonyl iron power premix composition," U.S. Patent 5 401 292, Mar. 28, 1995.

[33] R. Quintana, O. Persenaire, Y. Lemmouchi, L. Bonnaud, and P. Dubois, "Compatibilization of co-plasticized cellulose acetate/water soluble polymers blends by reactive extrusion," Polymer Degradation and Stability, vol. 126, pp. 31-38, 2016.