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Preparation and mechanical properties of graphite filled HDPE nanocomposites

M. Sarikanat ^a, K. Sever ^b, E. Erbay ^c, F. Güner ^c,

I. Tavman ^{b,*}, A. Turgut ^b, Y. Seki ^b, I. Özdemir ^d

^a Ege University, 35100 Bornova Izmir, Turkey

- ^b Dokuz Eylul University, 35100 Bornova Izmir, Turkey
- ^c Petkim Petrokimya Holding A.Ş., 35801 Aliaga-Izmir, Turkey
- ^d Bartın University, Bartın, Turkey
- * Corresponding author: E-mail address: ismail.tavman@deu.edu.tr

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ABSTRACT

Purpose: The design and manufacture of lightweight polymer composites with high electrical and thermal conductivity have been a research focus in recent years. In this study, tensile strength and modulus of elasticity of nanocomposites formed by high density polyethylene (HDPE) matrix and graphite powder filler material were determined.

Design/methodology/approach: In this study the conductive filler was graphite with an average particle size of 400 nm and purity of 99.9%, the matrix material was high density polyethylene (HDPE) with a density of 0.968 g/cm³ and a melt index of 5.8 g/10 min, supplied by Petkim A.Ş.- Izmir. Nanocomposites containing up to 30 weight percent of graphite powder filler material were prepared by mixing them in a Brabender Plasticorder at 180°C for 15 minutes. Tensile strength and modulus of elasticity of nanocomposites formed were determined as functions of graphite powder content.

Findings: An increase in tensile strength and modulus of elasticity was observed with increasing graphite powder content from 0 to 6%. However, for further increasing the graphite content, tensile strength decreases while modulus of elasticity continued to increase in the composite.

Practical implications: Since natural graphite (NG) has a high electrical conductivity at room temperature, it is considered an ideal candidate for manufacturing conductive polymer composites. The recent advancement of nano-scale compounding technique enables the preparation of highly electrically conductive polymeric nanocomposites with low loading of conductive fillers. Nanocomposites may offer enhanced physical features such as increased stiffness, strength, barrier properties and heat resistance, without loss of impact strength in a very broad range of common synthetic or natural polymers.

Originality/value: To see the effect of conducting fillers on mechanical properties of HDPE based nanocomposites, graphite particle 400 nm in size were used.

Keywords: HDPE; Conductive nanocomposites; Graphite; Tensile strength; Modulus of elasticity

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TECHNICAL PAPER

1. Introduction

In recent years, conducting polymer nanocomposites have attracted considerable attention due to their potential application in many areas, such as antistatic coatings, electromagnetic shielding materials, semiconductors and batteries [1-9]. The conductive fillers used mostly were metallic powders [1-3], carbon black [10-11] and graphite [12-16] usually incorporated into insulating polymers to produce electrical conducting composites [13-16]. Electrical conductivity values (S/cm) are typically 10^{-14} to 10^{-17} for polymers, 10^2 for carbon black, 10^5 for high-purity synthetic graphite, and 10^6 for metals such as aluminium and copper [17].

The presence of graphite particles has also great influence on the mechanical behaviour of polymeric composites [18-24]. Graphite/polyethylene composites have been studied intensively in recent years owing to the expected advantages of improving the electrical properties of composites. However, little effort is devoted to investigating the mechanical properties [24]. In the field of conducting polymeric composites, one main objective is to minimize the filler concentration because high-concentration of the conductive filler could lead to deterioration of mechanical properties of the composite [25]. For better process ability by conventional techniques such as injection moulding or compression moulding, a thermoplastic compound with as low a concentration of the conducting graphite additive as possible is preferred. This depends on the percolation threshold for a particular combination of graphite and the polymer matrix [26-27].

In this study, nanocomposites containing up to 30 weight % of graphite powder filler material were prepared by mixing them in a Brabender Plasticorder at 180°C for 15 minutes. Tensile strength and modulus of elasticity were determined as functions of content of graphite powder.

2. Materials and methods

2.1. Materials

The matrix used for this project was high density polyethylene (HDPE- I-668) with a density of 0.968 g/cm³ and a melt index of 5.8 g/10 min, supplied by Petkim A.Ş.- Izmir. The conductive filler used in this study was graphite with an average particle size of 400 nm and purity of 99.9%. The graphite powder was purchased from Nanostructured and Amorphous Materials, Inc.-Houston, USA.

Figure 1 shows the size of the graphite particles used in fabricating HDPE-Graphite nanocomposites. As can be seen from Figure 1, graphite particles have a different shapes and sizes.

2.2. Preparation of composite materials

HDPE-graphite mixtures in different weight percentage (wt.%) were prepared in a Brabender Plasticorder PLE 331 internal mixer at 180°C for a total mixing time of 15 min, the mixing chamber capacity being 30 ml. The rotors turned at

35 rpm in a counter-rotating fashion with a speed ratio of 1.1. After 10 minutes, the mixing chamber of the Brabender apparatus is opened and the resulting mixture is taken out, then after passing through the rollers the mixture is solidified. The HDPE-graphite mixture then put in a compression molding die and compressed in a compression molding press at 120°C, under 40 kPa pressure for one minute to obtain samples in the form of sheets of 1mm in thickness.





Fig. 1. The size of graphite particles

Morphological analysis of the graphite powder and nanocomposite sample was conducted with a scanning electron microscopy (JEOL JSM 6060). In order to reduce the extent of sample arcing, the samples were coated with a thin layer of metallic gold in an automatic sputter coater (Polaran SC7620) prior to examination by SEM. Graphite particles were dispersed in HDPE phase, as presented in Figure 2. When the concentration of graphite in HDPE phase is 4%, it is difficult to distinguish continuous phase (HDPE) and dispersed phase (graphite). However, when the concentration is increased, graphite phase can be seen clearly. Especially for the concentration of 30%, graphite particles are placed very closely to each other. When the SEM micrograph of graphite (Figure 2a) was compared with other SEM micrographs such as Figures 2d and 2e, it can be said that the accumulation of graphite particles is also possible due to higher graphite concentrations.



Fig. 2. a) Graphite powder b) HDPE-Graphite (4%), c) HDPE-Graphite (10%), d) HDPE-Graphite (20%), e) HDPE-Graphite (30%)

2.3. Tensile testing

The tensile specimens with a working area $35 \text{ mm} \times 3.6 \text{ mm} \times 1 \text{ mm}$ were punched out from the sheets. The mechanical properties were measured at room temperature using a

Shimadzu Autograph AG-IS Series universal testing machine equipped with a video extensioneter system at a cross head speed of 1 mm/min. The average values of three tests for tensile strength, tensile modulus and elongation at break are reported for each sample.

3. Results and discussion

Figure 3 shows the tensile strength with increasing graphite content in composite sheets. As can be noticed from Figure 3. filler content affects the tensile strength of nano composites. The tensile strength of composites increases from 23.31 to 35.72 MPa with increasing graphite content from 0 to 6%. However, increasing of filler content from 6 to 8%, tensile strength decreases to 34.82 MPa. Upon further increasing the graphite content, strength decreases in composite sheets. The tensile strength values of nanocomposite, HDPE-Graphite (10%), HDPE-Graphite (12%), HDPE-Graphite (15%), HDPE-Graphite (20%) and, HDPE-Graphite (30%) were obtained to be 32.74, 31.42, 30.43, 28.46 and 20.52 MPa, respectively. the detrimental mechanical properties are described on the basis of aggregation theory [28]. Hence, nanocomposites possess more matrix aggregation at low content of graphite. At higher content of graphite, due to its highly porous structure and high surface area, low content of polymeric matrix is insufficient to infiltrate in graphite and, as a result, aggregation of graphite in composites. However, at a lower content of larger graphite particles, the properties of composites are dominated by the matrix contents. At a lower content of graphite above the percolation threshold, the matrix causes the creation of voids in the composites [29]. Therefore, the stress concentration effect is due to the voids and also increases the degree of bonding between graphite particles and matrix. Thus, the importance of stress concentration increases which changes the mode of fracture behaviour of composite sheets.



Fig. 3. The variation of tensile strength with increasing graphite content in composites

At a higher content of graphite, graphite particles have a inclination to collect. Therefore, the matrix does not penetrate into the galleries between or inside graphite and graphite could not fully wet. This means that some graphite particles can overlap with each other during composite plate development, resulting in voids or defects in the interior of the composite structure where the strain is concentrated. This is because of a lack of percolation due to the lower content of matrix, which leads to composite plate that is fragile and can easily be fractured with the application of small stress [17].

The effect of graphite content on tensile modulus of composites was given in Figure 4. As can be seen in Figure 4, the modulus of HDPE increases with increasing graphite content. The modulus exhibits increasing trend with the amount of graphite, showing the dominant contribution of the filler particles to the modulus of the composite.



Fig. 4. The variation of tensile modulus with increasing graphite content in composites

4. Conclusions

HDPE-graphite mixtures in different weight percentage (wt.%) were prepared in a Brabender Plasticorder PLE 331 internal mixer and the resulting nanocomposite properties were compared with those of graphite in different weight percentage (wt.%) filled HDPE composite. With a loading of 6 wt.% expanded graphite in HDPE, the maximum tensile strength and the modulus improved over the neat HDPE film by about 53 and 46% respectively. The modulus of HDPE increases with increasing graphite content from 8% to 30%. But at a higher loading of 8 wt.%, the tensile strength exhibited significant decrement.

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References

- G. Pinto, A. Jimenez-Martin, Conducting aluminium-filled nylon 6 composites, Polymer Composites 22 (2001) 65-70.
- [2] L. Flandin, G. Bidan, Y. Brechet, J.Y. Cavaille, New nanocomposite materials made of an insulating matrix and conducting fillers, Processing and properties, Polymer Composites 21 (2000)165-174.
- [3] J. Stabik, A. Dybowska, J. Pluszyński, M. Szczepanik, Ł. Suchoń, Magnetic induction of polymer composites filled with ferrite powders, Archives of Materials Science and Engineering 41/1 (2010) 13-20.

- [4] P. Gramatyka, R. Nowosielski, P. Sakiewicz, Magnetic properties of polymer bonded nanocrystalline powder, Journal of Achievements in Materials and Manufacturing Engineering 20 (2007) 115-118.
- [5] B. Ziębowicz, M. Drak, L.A. Dobrzański, Corrosion resistance of the composite materials: nanocrystalline powder -polymer type in acid environment, Journal of Achievements in Materials and Manufacturing Engineering 36/2 (2009) 126-133.
- [6] S.S. Ray, M. Biswas, Water-dispersible conducting nanocomposites of polyaniline and poly(N-vinylcarbazole) with nanodimensional zirconium dioxide, Synthetic Metals 108 (2000) 231-236.
- [7] L.A. Dobrzański, A. Tomiczek, B. Tomiczek, A. Ślawska-Waniewska, O. Iesenchuk, Polymer matrix composite materials reinforced by Tb0.3Dy0.7Fe1.9 magnetostrictive particles, Journal of Achievements in Materials and Manufacturing Engineering 37/1 (2009) 16-23.
- [8] J. Stabik, A. Dybowska, Methods of preparing polymeric gradient composites, Journal of Achievements in Materials and Manufacturing Engineering 25/1 (2007) 67-70.
- [9] A. Gnatowski, J. Koszkul, Investigations of the influence of filler on the properties of chosen polymer blends with compatibilizer addition, Proceedings of the 13th Scientific International Conference "Achievements in Mechanical and Materials Engineering", AMME'2005, Gliwice-Wisla, 2005, 247-250.
- [10] Y. Ishigure, S. Iijima, H. Ito, T. Ota, H. Unuma, M. Takahashi, Y. Hikichi, H. Suzuki, Electrical and elastic properties of conductor-polymer composites, Journal of Materials Science 34 (1999) 2979-2985.
- [11] J. Stabik, Ł. Suchoń, M. Rojek, M. Szczepanik, Investigation of processing properties of polyamide filled with hard coal, Journal of Achievements in Materials and Manufacturing Engineering 33/2 (2009) 142-149.
- [12] D.S. Saunders, S.C. Galea, G.K. Deirmendjian, The development of fatigue damage around fastener holes in thick graphite-epoxy composite laminates, Composites 24 (1993) 309-321.
- [13] Y.H. She, G.H. Chen, D.J. Wu, Fabrication of polyethylene /graphite nanocomposite from modified expanded graphite, Polymer International 56 (2007) 679-685.
- [14] M. Szczepanik, J. Stabik, M. Łazarczyk, A. Dybowska, Influence of graphite on electrical properties of polymeric composites, Archives of Materials Science and Engineering 37/1 (2009) 37-44.
- [15] I. Tavman, V. Cecen, I. Ozdemir, A. Turgut, I. Krupa, M. Omastova, I. Novak, Preparation and characterization of highly electrically and thermally conductive polymeric nanocomposites, Archives of Materials Science and Engineering 29 (2009) 84-88.
- [16] J. Stabik, A. Dybowska, Electrical and tribological properties of gradient epoxy-graphite composites, Journal of

Achievements in Materials and Manufacturing Engineering 27/1 (2007) 39-42.

- [17] J.M. Keith, J.A. King, R.L. Barton, Electrical conductivity modelling of carbon-filled liquid-crystalline polymer composites, Journal of Applied Polymer Science 102 (2006) 3293-3300.
- [18] S. Kim, J. Seo, L.T. Drzal, Improvement of electric conductivity of LDPE based nanocomposite by paraffin coating on exfoliated graphite nanoplatelets, Composites Part A - Applied Science and Manufacturing 41 (2010) 581-587.
- [19] G.H. Chen, C.L. Wu, W.G. Weng, D.J. Wuand, W.L. Yan, Preparation of polystyrene/graphite nanosheet composite, Polymer 44 (2003) 1781-1784.
- [20] R.K. Goyal, A.N. Tiwari, U.P. Mulik, Y.S. Negi, Dynamic mechanical properties of Al₂O₃/poly(ether ether ketone) composites, Journal of Applied Polymer Science 104 (2007) 568-575.
- [21] A. Akinci, Mechanical and structural properties of polypropylene composites filled with graphite flakes, Archives of Materials Science and Engineering 35/2 (2009) 91-94.
- [22] Y.X. Pan, Z.Z. Yu, Y.C. Ou, G.H. Hu, A new process of fabricating electrically conducting nylon 6/graphite nanocomposites via intercalation polymerization, Journal of Polymer Science Part B - Polymer Physics 38 (2000) 1626-1633.
- [23] H. Kim, C.W. Macosko, Processing-property relationships of polycarbonate/graphene composites, Polymer 50 (2009) 3797-3809.
- [24] L.W. Wang, G.H. Chen, Dramatic improvement in mechanical properties of GNs-reinforced HDPE nanocomposites, Journal of Applied Polymer Science 116 (2010) 2029-2034.
- [25] J.R. Lu, X.F. Chen, W.Lu, G.H. Chen, The piezoresistive behaviors of polyethylene/foliated graphite nanocomposites, European Polymer Journal 42 (2006) 1015-1021.
- [26] T. Arai, Y. Tominaga, S. Asai, M. Sumita, Study on correlation between physical properties and interfacial characteristics in highly loaded graphite-polymer composites, Journal of Polymer Science Part B - Polymer Physics 43 (2005) 2568-2577.
- [27] S. Radhakrishnan, B.T.S. Ramanujam, A. Adhikari, S. Sivaram, High-temperature, polymer-graphite hybrid composites for bipolar plates: Effect of processing conditions on electrical properties, Journal of Power Sources 163 (2007) 702-707.
- [28] S.R. Dhakate, R.B. Mathur, S. Sharma, M. Borah, T.L. Dhami, Influence of expanded graphite particle size on the properties of composite bipolar plates for fuel cell application, Energy and Fuels 23 (2009) 934-941.
- [29] H.C. Kuan, C.C.M. Ma, K.H. Chen, S.M. Chen, Preparation, electrical, mechanical and thermal properties of composite bipolar plate for a fuel cell, Journal of Power Sources 134 (2004) 7-17.