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Tensile, electrical properties and morphology of polyethylene/modified fly ash composites using ultraflow

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Abstract

This paper presents relative melt viscosity, tensile, electrical properties and morphology of high density polyethylene (HDPE)/organo-modified fly ash (MFA) and HDPE/MFA/ultraflow (UTF) composites which were prepared by melt mixing method. Relative melt viscosity of HDPE was decreased with adding MFA and UTF into HDPE. Tensile properties (tensile strength, elongation at break and Young's modulus) of HDPE/MFA/UTF composites were increased with rising UTF content to 5 wt.% and thereafter, they were dropped with the UTF content more than 5 wt.%. Electric properties (dielectric constant, dielectric loss and volume resistivity) of the HDPE/MFA and HDPE/MFA/UTF composites were investigated. The obtained parameters showed that the HDPE/MFA composites have electric insulation higher than HDPE/MFA/UTF composites. Morphology of the composite materials with and without using UTF was also studied by Field Emission Scanning Electron Microscopy (FESEM) images. The results indicated that the MFA was dispersed more regularly and less agglomerated in HDPE matrix with adding UTF into the HDPE/MFA composites.

Keywords. Tensile properties, electrical properties, Ultraflow, HDPE, modified fly ash.

1. INTRODUCTION

Fly ash (FA) is a waste of burning coal process from the thermal power plant. It is a mixture of oxides such as SiO_2, Fe_2O_3 and Al_2O_3, etc. FA has thermal stability, size stability and low cost. It is used very effectively in many fields to reduce the amount of waste from the thermal power plant. Especially, it is a useful additive in concrete and cement [1-4].

High density polyethylene (HDPE) is one of thermoplastic polymers widely used in the world. It has many advantages like good mechanical properties, relatively low cost, low permeability to moisture and non-toxic in the processing [5-8]. HDPE has been applied to fabricate wires, cables, packages, composite materials, etc. There are many kinds of fillers which are introduced into polymers to improve their properties such as tensile, thermal, electric and rheological properties [9-12].

In recent years, the HDPE/FA composite material has been interested in research [12-14]. C. Alkan et al. studied the tensile strength and chemical resistance of HDPE/FA composite materials [2]. Due to differences in the structure and chemical nature of polymers and fly ash, they are difficult compatible and phase separation. Therefore, using the compatibilizers or surface modification of FA to improved compatibility and miscible between FA and HDPE is very nessescary. Some our previous paper have been reported the surface modification of FA by some coupling agents or fatty acids [15-17] caused the positive effect on the mechanical, rheological, thermal properties and moisture absorption of modified FA (MFA)-filled polymer composites. However, the weak point of above published paper is that the modification process of FA occured in wet state. The solvent eliminated after this process needs to be re-treated.

One new method to modify FA has been reported in our literature [18]. Here, FA was modified with stearic acids in solid state. This is a friendly environmental and economical method. Up to now, the use of ultra flow (UTF) – a stearate zinc salt - an additive for preparing the composites based on HDPE and MFA has not been investigated fully. The relative melt viscosity, tensile properties, electrical properties and morphology of HDPE/MFA/UTF composites have been studied to prove important role of UTF in HDPE/MFA composites.

2. EXPERIMENTAL

2.1. Materials

High density polyethylene (HDPE), Honam Co. (Korea) with the density of 0.96 g/cm^3. Fly ash silo (FA) was provided by Pha Lai Thermoelectric Power Plant (Vietnam) after sink/flotation separation processes. The average particle size of selected FA is about 5 µm, total weight of SiO_2, Al_2O_3 and Fe_2O_3 more than 86 % and moisture content less than 0.3 %. FA was modified by stearic acids in solid state as process in [18]. Stearate zinc salt with commercial name of ultraflow (UTF), Korea.

2.2. Preparation of composites

The content of MFA was fixed in 10 wt.% while UTF weight is changed from 1 to 7 wt.% in comparison with HDPE weight in the composites. The composites preparation was carried out by melt mixing method in a Haake Rheomixer (Germany) at 180 ^oC and rotor speed of 50 rpm for 6 min. After that, the composites were molded by hydraulic press machine (Toyoseiky, Japan) at 180 ^oC for 3 min with pressing pressure of 12-15 MPa. Then the samples were cooled and stored at least 24 hours before determining properties and morphology. This process of composite preparation was conducted at Institute for Tropical Technology, Vietnam Academy of Science and Technology (VAST).

2.3. Characterizations

2.3.1. The relative melt viscosity

The relative melt viscosity or mixing torque in mixing process of HDPE/MFA composites using UTF was determined by Polylab 3.1 software connected to the Haake Rheomixer at Institute for Tropical Technology, VAST.

2.3.2. The tensile properties

The tensile properties (tensile strength, elongation at break and Young's modulus) of the HDPE/MFA/UTF composites were measured on Zwick Tensile 2.5 Machine (Germany) according to ASTM D638 at Institute for Tropical Technology, VAST.

2.3.3. The electrical properties

Dielectric constant (ε) and dielectric loss (tgδ) of the HDPE/MFA/UTF composites were measured on TR-10C Machine (Japan) according to ASTM D150 at frequency 1 kHz.

Volume resistivity (ρ_v) of the composites was measured on TR-8401C Machine (Takeda Ricken, Japan) by according to ASTM D257 with DC voltage 100V.

The above electrical properties were measured at Institute for Tropical Technology, VAST.

2.3.4. Field emission scanning electron microscopy (FESEM)

The morphology of the composites was examined by using Field Emission Scanning Electron Microscopy (FESEM) technique. The image of the samples was observed in an S-4800 FESEM instrument (Hitachi, Japan) at Institute of Material Science, VAST.

3. RESULTS AND DISCUSSION

3.1. Relative melt viscosity

Fig. 1 displays the torque curves expressing relative melt viscosity of HDPE, HDPE/MFA and HDPE/MFA/3 wt.% UTF composites. It is clear that the relative melt viscosity of HDPE was decreased with adding MFA and UTF into the HDPE matrix. Specially, the torque of composites using UTF from 2nd minute of melt mixing was decreased dramatically. This can be explained by the dispersion of UTF into HDPE contributed to increasing mobility of HDPE macromolecules, leading to reduction of internal friction in melt mixing process of HDPE and MFA. Therefore, melt mixing process of HDPE/MFA/UTF composite was carried out more easily than HDPE and HDPE/MFA composite as well as energy consumption of mixing HDPE/MFA/UTF composites was less than that with HDPE and HDPE/MFA composite [11].

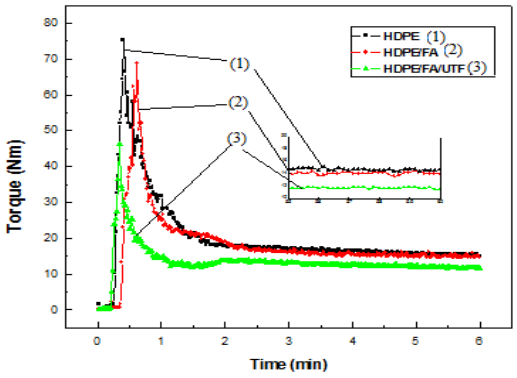

Figure 1: Torque curves of HDPE (1), HDPE/MFA (2) and HDPE/MFA/UTF (3) composites

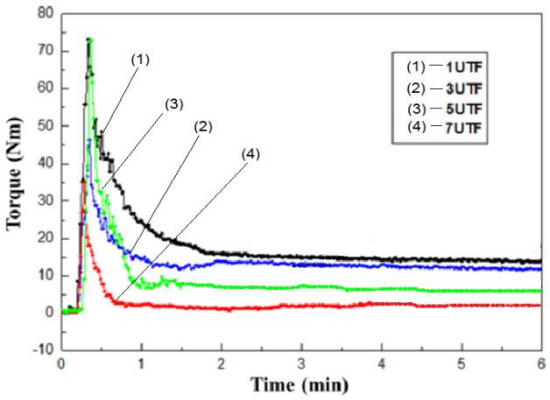

Figure 2: Torque curves of HDPE/MFA composites using various content of UTF

The torque curves of HDPE/MFA composites using various content of UTF are shown in Fig. 2. It can be seen the torque of HDPE/MFA/UTF composites was decreased with rising UTF content (from 1 to 7 wt.%). This can conclude that UTF plays the role as a processing aid agent and lubricant in mixing process of HDPE and MFA.

3.2. Tensile properties

The effect of UTF content on tensile properties of HDPE/MFA/UTF composites is demonstrated in Table 1. The tensile strength of HDPE/MFA/UTF composites was increased with rising UTF content up to 3 wt.%. For example, the tensile strength of HDPE/MFA/UTF composites is grown up from 29.26 MPa to 31.81 and 31.02 MPa at 1 and 3 wt.% of UTF and then dropped to 28.65 and 27.81 MPa at 5 and 7 wt.% of UTF. Similarly, Young's modulus of the composites had a tendency to increase up to 5 wt.% of UTF (1107.66 MPa). This can be attributed by UTF which was contributed in improvement of the dispersibility, adhering and mixing MFA and HDPE due to hydrogen bonds and dipole – dipole interactions between C=O, C−O−C groups of stearic acid grafted onto FA surface and C=O, C−O−C groups of UTF. Besides, the presence of stearate in UTF is easier to mix with ethylene unit chain in HDPE macromolecules. Here, UTF plays the role as not only a processing aid agent and a lubricant but also a compatibilizer in HDPE/MFA composites. In HDPE/MFA composites using UTF content more than 5 wt.%, excessed UTF weight may be to agglomerate in HDPE matrix and make weakening structure of the composites.

In contrast, from table 1, it can be seen that elongation at break of the HDPE/MFA composites was decreased as adding UTF. As our knowledge, the rule of change in mechanical properties of the composite materials is that the tensile strength increased as elongation at break reduced [4]. Thus, the going down in elongation at break of composites is reasonable. At the UTF content of 3 wt.%, elongation at break of the composites was increased. This result showed that UTF content of 3 wt.% is the most suitable for preparing HDPE/MFA/UTF composites.

Table 1: Tensile properties of HDPE/MFA/UTF composites at various contents of UTF

UTF content (wt.%)	Tensile strength (MPa)	Elongation at break (%)	Young's modulus (MPa)
0	29.26	854.00	917.22
1	31.81	509.31	1032.31
3	31.02	578.00	920.66
5	28.65	528.04	1107.66
7	27.81	192.23	826.36

3.3. Electrical properties

Table 2 presents the dielectric constant (ε), dielectric loss tangent (tan δ) and volume resistivity (ρ_v) measured at 1 kHz of HDPE/MFA composites using various content of UTF. It is clear that the dielectric constant, dielectric loss angle tangent (tan δ) and volume resistivity of HDPE/MFA/UTF composites were increased with rising of UTF content and higher than those of HDPE/MFA composite. The dielectric constant and tan δ of HDPE/MFA/UTF composites using UTF content (1 - 7 wt.%) were increased from 2.294 to 2.458 and 0.064 to 0.125, respectively. These results can be explained by the nature of an inorganic salt UTF. Therefore, the addition of UTF into composites can also make these materials become more polar and HDPE/MFA/UTF composites are more polar than HDPE/MFA composite.

Table 2: Dielectric constant (ε), dielectric loss tangent (tan δ) and volume resistivity (ρ_v) of the HDPE/MFA/UTF composites

UTF content (wt.%)	Dielectric constant (ε)	Dielectric loss angle tangent (tan δ)	Volume resistivity ρ_v (Ω.cm)
0	2.303	0.040	1.54×10^{12}
1	2.294	0.064	2.03×10^{12}
3	2.340	0.066	2.36×10^{12}
5	2.424	0.112	2.25×10^{12}
7	2.458	0.125	3.37×10^{12}

The volume resistivity of HDPE/MFA/UTF composites was raised from $2.03x10^{12}$ to $3.37x10^{12}$

(Ω.cm). This can be attributed to the presence of UTF as a compatibilizer in HDPE/MFA composites, the dispersion of MFA in HDPE matrix was improved significantly (see 3.4. Morphology). These electrical parameters of HDPE/MFA/UTF composites are to meet requirements for electric insulation materials such as electric wires and cables (according to TCVN 5935-1995).

3.1. Morphology

Morphology of HDPE/MFA and HDPE/MFA/UTF composites with and without 3 - 5 wt.% UTF is performed in FESEM images of their fracture surface (Fig 3). Observably, HDPE/MFA and HDPE/MFA/UTF composites have heterogeneous structure. The MFA dispersed more regularly and adhered better with HDPE in the presence of UTF (Fig. 3 b and c). This is explained by the hydrogen bonds and dipole – dipole interactions between UTF and MFA as well as easier mixing the moiety of stearate in UTF and ethylene unit chain in HDPE macromolecules. They are main reasons to improve dispersibility, adhering and mixing MFA and HDPE as above mentioned (section 3.1).

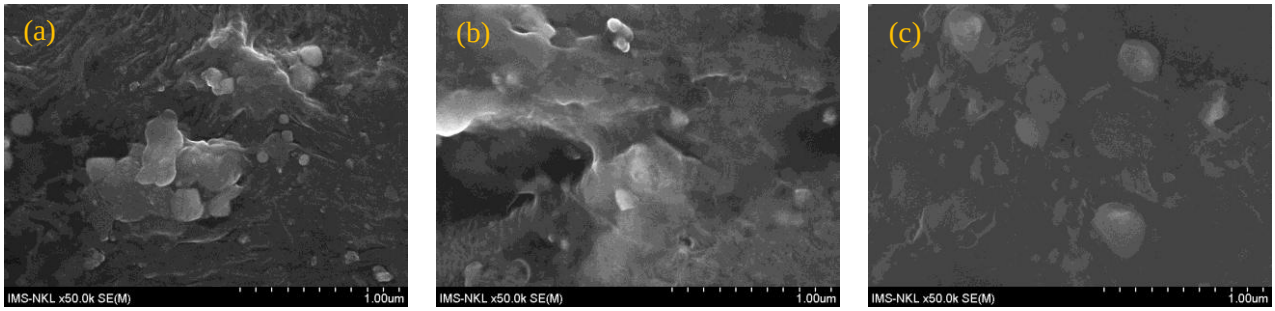

Figure 3: FESEM images of fracture surface of HDPE/MFA (a), HDPE/MFA/3 wt.% UTF (b) and HDPE/MFA/5 wt.% UTF (c) composites

4. CONCLUSION

The relative melt viscosity of HDPE/MFA composites was decreased as adding UTF into HDPE. The HDPE/MFA/UTF composites were prepared more easily than HDPE/MFA composite. According to tensile strength and elongation at break, the UTF content of 3 wt.% is the most suitable for preparing HDPE/MFA/UTF composites. The dielectric constant and dielectric loss angle tangent and volume resistivity of the HDPE/MFA/UTF composites are higher than those of HDPE/MFA composite. MFA particles were dispersed more regularly and adhered better with HDPE in the composites using UTF.

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REFERENCES

1. S. Tapkin. *Mechanical evaluation of asphalt-aggregate mixtures prepared with fly ash as a filler replacement*, Can. J. Civil Eng., **35(1)**, 27-40 (2008).
2. C. Alkan, M. Arslan, M. Cici, M. Kaya, M. Aksoy. *A study on the production of a new material from fly ash and polyethylene*, Resour. Conserv. Recycle, **139(3-4)**, 147-154 (1995).
3. S. Bonda, S. Mohanty, S. K. Nayak. *Viscoelastic, mechanical and thermal characterization of fly ash-filled ABS composites and comparison of fly ash surface treatments*, Polymer Composites, **33(1)**, 22-34 (2012).
4. S. G. Pardo, C. Bernal, A. Ares, M. J. Abad, J. Cano. *Rheological, thermal, and mechanical characterization of fly ash-thermoplastic composites with different coupling agents*, Polymer Composites, **31(10)**, 1722-1730 (2010).
5. K. Chrissafis, K. M. Paraskevopoulos, E. Pavlidou, D. Bikiaris. *Thermal degradation mechanism of HDPE nanocomposites containing fumed silica nanoparticles*, Thermochimica Acta, **485(1-2)**, 65-71 (2009).
6. D. Kulutas, I. H. Tavman. *A numerical and experimental study on thermal conductivity of particle filled polymer composites*, Journal of Thermoplastic Composite Materials, **19(4)**, 441-455 (2006)
7. Y. P. Mammya, V. V. Davydenko, P. Pissis, E. V. Lebedev. *Electrical and thermal conductivity of polymers filled with metal powders*, European Polymer Journal, **38(9)**, 1887-1897 (2002).
8. I. H. Tavman. *Thermal and mechanical properties of aluminum powder-filled high-density polyethylene*

composites, Journal of Applied Polymer Science, **62(12)**, 2161-2167 (1996).

9. U. Atikler, D. Basalp, F. Tihminlio Lu. *Mechanical and morphological properties of recycled high-density polyethylene, filled with calcium carbonate and fly ash*, J. Appl. Polym. Sci., **102(5)**, 4460-4467 (2006).
10. A. R. R. Menon, T. A. Sonia, J. D. Sudha. *Studies on fly-ash-filled natural rubber modified with cardanol derivatives: Processability, mechanical properties, fracture morphology, and thermal decomposition characteristics*, Journal of Applied Polymer Science, **102(5)**, 4801-4808 (2006).
11. S. A. R. Hashmi, P. Sharma, N. Chand. *Thermal and rheological behavior of ultrafine fly ash filled LDPE composites*, Journal of Applied Polymer Science, **107(4)**, 2196-2202 (2008).
12. T. Chaowasakoo, N. Sombatsompo. *Mechanical and morphological properties of fly ash/epoxy composites using conventional thermal and microwave curing methods*, Composites Science and Technology, **67(11-12)**, 2282-2291 (2007).
13. M. V. Deepthi, S. Madan, R. R. N. Sailaja. *Mechanical and thermal characteristics of high density polyethylene-fly ash Cenospheres composites*, Materials and Design of Nanomaterials and Nanostructures, **31(4)**, 2051-2060 (2010).
14. N. Chand, P. Sharma, F. Mahi. *Correlation of mechanical and tribological properties of organosilane modified cenosphere filled high density polyethylene*, Materials Science and Engineering: A, **527(21-22)**, 5873-5878 (2010).
15. Thai Hoang, Vu Minh Duc, Nguyen Vu Giang, Do Quang Tham, Nguyen Hong Quyen. *Relative melt viscosity, mechanical and electrical properties of polymer composites of HDPE/fly ash*, Vietnam Journal of Chemistry, **48(1)**, 85-88 (2010).
16. Thai Hoang, Nguyen Thuy Chinh, Nguyen Thi Thu Trang, Do Quang Tham, Tran Thi Thanh Van. *Mechanical properties, morphology and flammable retardancy of composites based on polyvinylchloride and fly ash modified with some silane coupling agents*, Vietnam Journal of Science and Technology, **51(5)**, 627-633 (2013).
17. Thai Hoang, Nguyen Thuy Chinh, Nguyen Thi Thu Trang, Do Quang Tham, Tran Thi Thanh Van, *Study on thermal and electrical properties of composites based on polyvinylchloride and fly ash modified with some organic acids*, Vietnam Journal of Chemistry, **51(2C)**, 882-887 (2013).
18. Do Quang Tham, Nguyen Thi Thu Trang, Nguyen Thuy Chinh, Tran Thi Mai, Nguyen Vu Giang, Nguyen Thi Kim Dung, Thai Hoang. *Fabrication and characterization of masterbatches made from poly(vinyl chloride) and modified fly ash*, Vietnam Journal of Chemistry, **54(2)**, 218-223 (2016).

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