

Nanocomposites Based on Magnesium-oxide/ Aluminum-Nitride/Polypropylene for HVDC Cable insulation

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Abstract—Polypropylene (PP) with high thermal stability and good electrical properties, has attracted much attention for its potential to take the place of cross-link polyethylene (XLPE) as HVDC insulation because PP is more easily recycled than XLPE due to its thermoplasticity. Due to the adverse effect of electric field reversal under HVDC application, there is a need to find the new polymer insulation material with higher thermal conductivity and good electrical performance. This paper investigates the effect of introducing aluminum nitride (AlN) and magnesium oxide (MgO) into PP on the electrical properties of the resulting the new nanocomposites. In the sample preparation, AlN and MgO were surface-modified by KH570 (γ -methacryloxypropyltrimethoxy silane) and then introduced into PP by the solution method to manufacture the nanocomposite materials. The measurements made were the voltage breakdown characteristics and the DC conductivity. The results obtained show that the combination of AlN and MgO can slightly decrease the DC conductivity of PP/AlN/MgO nanocomposites compared with pure PP. The breakdown strength was slightly decreased, which shows that the adverse effect of AlN on the electrical performance of PP can be compensated by introducing MgO nanoparticles. Hence, the new polymer with high thermal conductivity and good electrical properties could be manufactured by combining two kinds of nanoparticles.

Keywords — nanocomposites, magnesium-oxide, aluminum-nitride, polypropylene, electrical performance.

I. INTRODUCTION

Polymers filled with nanoparticles have attracted a large amount of attention in recent years since the first concept was proposed by Lewis in 1994 [1]. Due to the special characteristics of nanoparticles, the electrical, chemical, mechanical and thermal properties of polymers can be greatly improved by embedding nanoparticles [2-4]. For example, the space charge accumulation can be suppressed in polypropylene (PP) by introducing nano-MgO nanoparticles. The interfacial region between nanoparticles and polymer determines the characteristics of nanocomposites [6-7].

Polypropylene (PP) is one of the commercial thermoplastic polymers which has larger potential to be the next generation of new polymer insulation material.

Compared with LDPE and XLPE, PP has much higher operational temperature, excellent electrical properties including breakdown strength, space charge characteristics, low DC conductivity and low dielectric loss [8]. The nano-MgO can increase the breakdown strength and decrease the space charge accumulation but the thermal conductivity it has is only $42 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$, while nano-AlN has the thermal conductivity of $275 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ which is much higher than nano-MgO so that it can increase the thermal conductivity of polymer insulation materials then the electric field distribution would be more uniform and the effect of electric field reversal can be reduced. [9]. Also, both of nano-MgO and nano-AlN can reduce the dielectric loss of polymer insulation materials.

In this paper, PP was chosen to be the matrix material. Nano-MgO and nano-AlN were introduced into PP with different contents by using the solution method. The objective is to investigate whether it is possible to produce a nanocomposite that possesses the advantages of both additives. Finally, the properties of these nanocomposites were evaluated by DC conductivity and breakdown strength and the results showed that the synergistic effect of nano-AlN and nano-MgO on PP was good.

II. EXPERIMENTS

A. Materials and Preparation

The matrix material, polypropylene (PP) pellets with density of $0.92 \text{ g}/\text{cm}^3$ and nanoparticles MgO and AlN with a nominal diameter of 50 nm were provided by Aladdin Industrial Inc., China. The melt flow index of PP was 0.6g/min. The xylene solution with a purity of $\geq 98\%$ was provided by Sinopharm Chemical Reagent. Co. Ltd (China). The nanoparticles of MgO and AlN were surface modified by γ -methacryloxypropyltrimethoxysilane (MPS) in the xylene to improve the adhesion between the nanoparticles and polymer matrix [5]. Normally, it is widely believed that the xylene was used to dissolve the PP pellets at 120°C . The different contents of nanoparticles shown in Tab.I. were

dispersed in the mixture of PP and xylene and then the mixed solution was stirred by the blender as shown in Figure 1. The rotor speed was 500 rpm, the mixing time was 12 h and the operational temperature was 120 °C. Next, the mixed solution was dried in the turbo blower at 120 °C for 12 h. The film samples with different thickness were manufactured by using compression

TABLE I. SAMPLE NOTATION AND COMPOSITION

	Contents of nano-MgO, nano-AIN and PP		
	MgO (wt.%)	AIN (wt.%)	PP (wt.%)
No.1	0	0	100
No.2	1.0	2.0	97.0
No.3	2.0	1.0	97.0
No.4	3.0	0	97.0
No.5	0	3.0	97.0

molding at the temperature of 200 °C and the duration was 12 min under a pressure of 15 MPa. After that, the films were cooled to room temperature under a pressure of 10 Mpa for 15 min. The thickness of the samples was about 100 um for the breakdown test. Finally, the sample of pure PP and nanocomposites with different content of MgO/AIN were put in a vacuum oven (0.01 Pa) at 80 °C for 24 h and cooled down to room temperature at 0.1 K/min.

B. Detection Of Nanoparticles Dispersion

The nanoparticle dispersion was detected by the field emission scanning electron microscope (FE-SEM, Hitachi SU8010, Japan). The sample of nanocomposites were broken into several parts in liquid nitrogen, then covered with gold on the fractured surface.

C. DC Breakdown Tests

The DC breakdown strength was carried out using a sphere-sphere electrode configuration. The nanocomposite samples with the thickness of 100 um were put into silicone oil so that the nanocomposite samples were isolated from the air and flashover can be avoided during the breakdown test. There are 19 measuring points being marked uniformly on each film and placed in the middle of the sphere electrodes to make the breakdown points happen at the point which has been measured during DC breakdown test. The increasing rate of DC voltage was 1 kV/s until breakdown occurred.

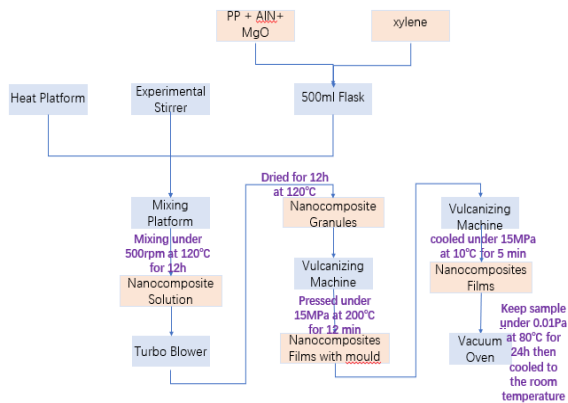


Fig.1. The procedure of sample preparation

D. DC Conductivity

The DC volume conductivity was carried out by a digital high-resolution electrometer (6517B, Keithley) equipped with standard three-electrodes system. The PP/MgO/AIN nanocomposites samples with the thickness of 100 um were measured under the DC electric field from 10 kV/mm to 80kV/mm with the step of 10 kV/mm. The duration of each step was 600 s. Before the sample measurement, each sample should be short-circuited for 10 min. Finally, the conductivity results will show the effect of nano-MgO and nano-AIN on the DC volume resistivity of PP.

III. RESULTS AND DISCUSSION

A. Scanning Electron Microscope Result

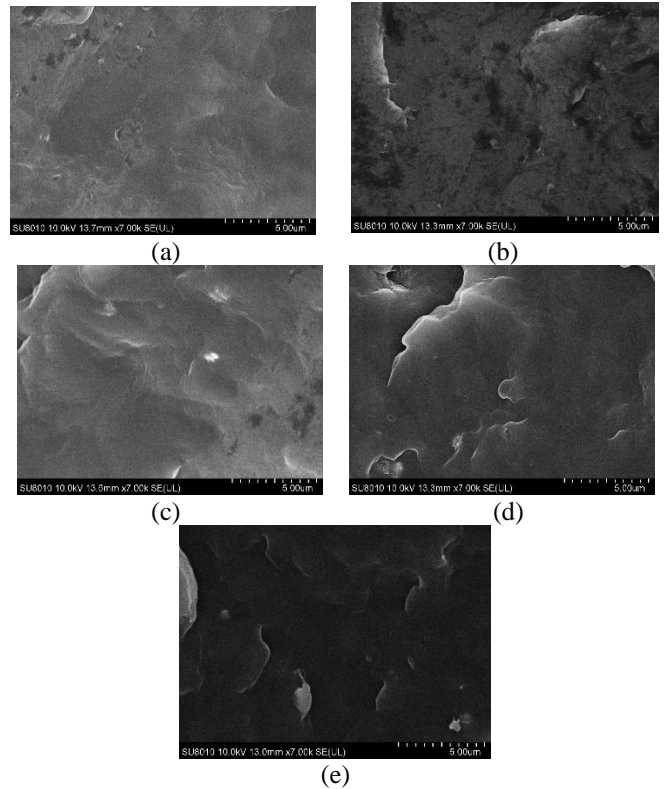


Fig. 2 SEM images of MgO/AIN/PP nanocomposites with different content (a). Pure PP (b). PP/1.0 phr MgO + 2phr AIN (c). PP/1.0 phr MgO + 2.0 phr AIN (d). PP/3.0 phr MgO (e). PP/3.0 phr AIN

Fig.2 has shown the distribution of nano-MgO and nano-AIN particles in the bulk of the matrix material, PP. Normally the dispersion of nanoparticles has great impact on the electrical properties of nanocomposites. According to the SEM images Fig.2(a)-(e), the surface modification by γ -methacryloxypropyltrimethoxysilane (MPS) has ensured most of nano-MgO and nano-AIN particles homogenously dispersed in the matrix material which reveals that the interface between the surface of nano-MgO and nano-AIN is compatible and the agglomeration would not cause severe degradation of electrical properties of MgO/AIN/PP nanocomposites.

B. DC Breakdown Tests Result

The experimental DC breakdown strength of nanocomposites are changeable, hence, the probability

distribution used to describe the variability was the 2-parameter cumulative Weibull distribution function (1) [10]:

$$P(E) = 1 - \exp \left[- \left(\frac{E}{E_0} \right)^\beta \right] \quad (1)$$

Where:

E : the measured electric field for each measurement point;

E_0 : the critical parameter of the breakdown for a probability of 63.2%;

β : the shape parameter of the Weibull distribution of breakdown strength data;

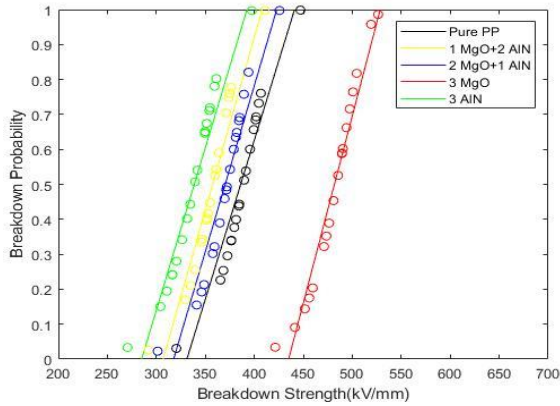


Fig. 3. Weibull description of DC breakdown strength of PP/MgO/AlN nanocomposites

Fig.3 and Tab. II shows the DC breakdown strength of PP/MgO/AlN nanocomposites with different contents of nano-MgO and nano-AlN. The surface of nano-AlN has been modified by MPS silane coupling agent to achieve good adhesion between nanoparticle and the matrix material PP, but some disadvantage of decreasing the electrical performance would be introduced into the polymer [11]. The nanocomposites with 3 wt% MgO had the highest breakdown strength of 492.2kV, while the breakdown strength of nanocomposites with 3 wt% AlN was reduced to 348.4kV by 12.3% compared with PP. The nanocomposites with 2wt% AlN/1wt% MgO and 1wt% AlN/2wt% MgO had only 7.8% and 4.17% lower than the breakdown strength of PP respectively. Thus, the adverse effect of nano-AlN on breakdown strength had been significantly compensated by nano-MgO. It is obvious that the DC breakdown strength of PP/MgO/AlN nanocomposites were reduced due to a higher AlN content. The reduction of DC breakdown strength may be caused by the defects in the interphase between AlN and PP [9].

TABLE II. THE CRITICAL VALUES OF THE BREAKDOWN STRENGTH FOR THE PROBABILITY OF 63.2%

	Order of Samples				
	1	2	3	4	5
E_0 (kV/mm)	397.7	366.6	381.1	492.2	348.4

C. DC Conductive Current

Through the applied DC electric field from 10 kV/mm to 80 kV/mm with the step of 10 kV/mm, the conductive current-

voltage (J-E) characteristics of all nanocomposites are shown in Fig.4. The DC conductive current increases with the increase of nano-AlN filler content. The nano-MgO could significantly suppress the conductive current. Hence, the increased DC conductivity should be attributed to the higher conductivity of AlN nanoparticles compared with the DC conductivity of PP and MgO. Otherwise, the low conductivity of MgO nanoparticles increases the resistivity of sample. Also, Fig.4. shows that the transition field decrease from 50 kV/mm to 30 kV/mm with the increase of nano-AlN. From [12], nano-MgO can significantly increase the depth of electron trap in PP, which can suppress the electron mobility in the nanocomposites. Hence, the adverse effect of nano-AlN on the conductive current can be compensated by introducing nano-MgO. This phenomenon is presumed to be related to the conductivity of AlN. This phenomenon is presumed to be related to the lower conductivity of nano-AlN than nano-MgO.

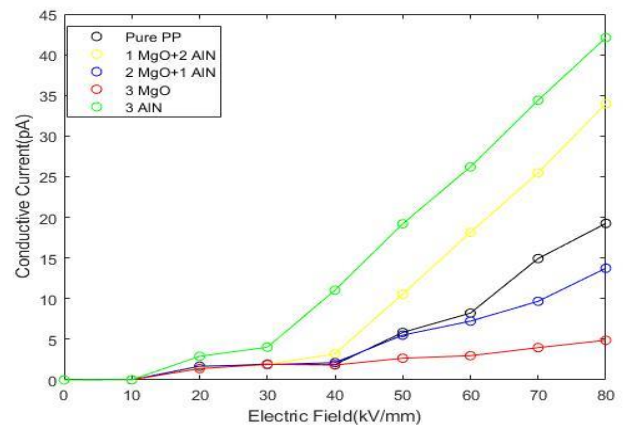


Fig.4. The conductive current under different electric field for all nanocomposites

IV. CONCLUSION

The addition of nano-AlN and nano-MgO modified with γ -methacryloxypropyltrimethoxysilane (MPS) have significant effect on the electrical properties on PP. The electrical properties would be worsened by introducing nano-AlN, while the nano-MgO could improve the electrical properties of nanocomposite. Even though nano-AlN could worsen the electrical properties of nanocomposites, introducing nano-MgO have significantly enhanced the breakdown strength and reduce conductivity of the nanocomposite. Also, there was not the rapid degradation of electrical performance which means the compatibility between nano-MgO and nano-AlN in terms of electrical performance was good and the interface relation between nano-AlN and nano-MgO does not have additional effect on electrical application under HVDC condition. Although, the breakdown strength and conductivity performance of the PP/MgO/AlN nanocomposite was reduced, this new nanocomposite still has good electrical properties and higher thermal conductivity, which means the effect electric field reversal on the HVDC application of the PP/MgO/AlN nanocomposites has been reduced. The results indicate that a nano-composite embedded with two types of nanoparticles complement each other and further investigation into the

other properties (chemical and electrical) would be done to fully assess the viability of this approach.

In the future work, more tests including thermal conductivity measurement, pulse electro-acoustic (PEA) test and dielectric characteristics will be done to do the further research on these nanocomposites' characteristics.

V. ACKNOWLEDGMENT

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VI. REFERENCE

- [1]. T. J. Lewis, "Nanometric Dielectrics," *IEEE Trans. Dielectr. Electr. Insul.*, Vol. 1, No. 5, pp. 812-825, 1994.
- [2]. T. Tanaka, M. Kozako, N. Fuse, and Y. Ohki, "Proposal of a multi-core model for polymer nanocomposite dielectrics," *IEEE Trans. Dielectr. Electr. Insul.*, Vol. 12, No.4, pp. 669-681, 2005.
- [3]. T. Tanaka, "Dielectric nanocomposites with insulating properties," in *IEEE Transactions on Dielectrics and Electrical Insulation*, vol. 12, no. 5, pp. 914-928, Oct. 2005.
- [4]. T. Tanaka, M. Kozako, N. Fuse, and Y. Ohki, "Proposal of a multi-core model for polymer nanocomposite dielectrics," *IEEE Trans. Dielectr. Electr. Insul.*, Vol. 12, No.4, pp. 669-681, 2005.
- [5]. Z. Li, W. Cao, G. Sheng, X. Jiang and M. G. Danikas, "Experimental study on space charge and electrical strength of MgO nanoparticles/polypropylene composite," in *IEEE Transactions on Dielectrics and Electrical Insulation*, vol. 23, no. 3, pp. 1812-1819, June 2016.
- [6]. T. J. Lewis, "Interfaces are the dominant feature of dielectrics at the nanometric level," *IEEE Trans. Dielectr. Electr. Insul.*, Vol. 11, No. 5, pp. 739-753, 2004.
- [7]. M. Roy, J. K. Nelson, R. K. Maccrone, L. S. Schadler, C. W. Reed, R. Keefe and W. Zenger, "Polymer nanocomposite dielectrics-The role of the interface," *IEEE Trans. Dielectr. Electr. Insul.*, Vol. 12, No. 4, pp. 629-643, 2005.
- [8]. J. Zha, Y. Wu, S. Wang, D. Wu, H. Yan and Z. Dang, "Improvement of space charge suppression of polypropylene for potential application in HVDC cables," in *IEEE Transactions on Dielectrics and Electrical Insulation*, vol. 23, no. 4, pp. 2337-2343, August 2016.
- [9]. Y. Zhou, J. Hu, X. Chen, F. Yu and J. He, "Thermoplastic polypropylene/aluminum nitride nanocomposites with enhanced thermal conductivity and low dielectric loss," in *IEEE Transactions on Dielectrics and Electrical Insulation*, vol. 23, no. 5, pp. 2768-2776, October 2016.
- [10]. IEC 62539, "Guide for the statistical analysis of electrical insulation breakdown data".
- [11]. T. Tanaka, A. Vaughan, "Tailoring of Nanocomposite Dielectrics". Pan Stanford Publishing, 2016.
- [12]. Y. Zhou *et al.*, "Temperature dependent electrical properties of thermoplastic polypropylene nanocomposites for HVDC cable insulation," in *IEEE Transactions on Dielectrics and Electrical Insulation*, vol. 26, no. 5, pp. 1596-1604, Oct. 2019.