CHEMICAL ENGINEERING TRANSACTIONS
VOL. 56, 2017
A publication of AIDIC
The Italian Association of Chemical Engineering
Online at www.aidic.it/cet
Guest Editors: Jiří Jaromír Klemeš, Peng Yen Liew, Wai Shin Ho, Jeng Shiun Lim

ISBN978-88-95608-47-1; **ISSN** 2283-9216
DOI: 10.3303/CET1756202

Effect of Fly Ash from Thermal Power Plant on the Dielectric Properties of Polymer Composites Materials based on Matrix Epoxy DER 331

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Fly ash (FA) is a low cost, inorganic waste product obtained from thermal power plants. This waste product causes significant environmental problem but it possesses good mechanical properties due to the constituent phases contained in it i.e. silica and alumina. In this paper, epoxy resin matrix, a good insulating polymers, combined with FA was used to manufacture composite polymer materials. The effects of the compositions of fly ash witht and without treatment on the dielectric properties of polymer composite (PC) materials based on epoxy resin DER 331 were studied. The results show that the samples with modified fly ash using 2 % stearic acid and 2% silane GF80has significantly improved the insulating properties of the material compared to the non-modified composite samples.

1. Introduction

Fly ash, a waste product from the thermal power plants, is increasing day by day. This material causes significant economic and environmental problems. To mitigate the issues, the researchers proposed the use of fly ash for various purposes such as in chemical, agricultural, cement, constructional and polymer industries. In the polymer industry, FA filler has improved the mechanical properties of the material matrix such as tensile strength, impact resistance and brittleness (Pardo et al., 2010).
Epoxy, one of the best insulating materials, is applied in the power and electronics industries (Ahmad, 2012). The use of FA as filler in combination with epoxy resin to manufacture composite materials can help to re-use the waste product and reduce the waste treatment cost. However, untreated fly ash (UFA) has smooth surface, which causes poor interface adhesion to the polymer matrix. The role of the polymer matrix is to transfer the load to the filler to enhance the properties of the composites. The FA should be modified to improve its dispersion into the polymer matrix using coupling agents (Yang et al., 2006). This is evident in the previous researches done to investigate the effects of coupling agents on fly ash polybutadiene rubber (Alkadasi et al., 2004) and recycled poly(ethylene terephthalate)/fly ash composites.
In this study, the coupling agents, silane GF80 (3-glycidoxypropyltrimethoxysilane) and SA (stearic acid) provide bonded coupling between the fly ash particles and the epoxy resin (EP). The GF80 epoxy functional groups and carboxyl group of the SA assist to bridge the link between the hydroxyl groups of fly ash with epoxy functional groups of the EP. The modified fly ash can improve the electrical properties of the materials in which it is dispersed into.

2. Experimental

2.1. Materials

Fly ash was collected from Pha Lai (Vietnam) thermal power station. It consists of a mixture of solid and hollow spherical particles of varying size (from 1 μm to 100 μm, medium size: 28 μm). The chemical composition and morphological structure of the fly ash particles listed in Table 1 was experimentally determined through X-Ray Fluorescence (Viet Space 5,006-HQ02) and SEM (JSM – 6,490 - Japan).

Please cite this article as: Bachtrong P., Nguyenthanh L., Phamthi H., 2017, Effect of fly ash from thermal power plant on the dielectric properties of polymer composites materials based on matrix epoxy der 331, Chemical Engineering Transactions, 56, 1207-1212 DOI:10.3303/CET1756202

The characteristics of epoxy resin matrix, curing agent and coupling agents are presented in Table 2

Table 1: Chemical composition and morphological structure of fly ash

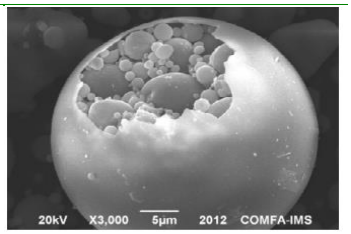
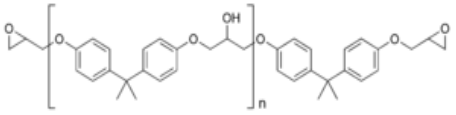
Chemical composition	wt%	Morphological structure
Al_2O_3	23.615	
SiO_2	50.503	
Fe_2O_3	11.894	
MgO	1.219	
CaO	1.064	
SO_3	0.409	
Other elements	11.296	

Table 2: Characteristics of epoxy resin matrix, curing agent and coupling agents of various materials

Materials	Provider	Characteristics
Epoxy DER 331	Dow (American)	 d: 1.16 g/cm^3, epoxy content: 22.0 - 22.6 %
Curing agent DETA Diethylentriamine	Dow (American)	$HN(CH_2CH_2NH_2)_2$ d: 0.95 g/cm^3
GF80 (3- glycidoxypropyltrimethoxysilane)	Momentive (Germany)	$C_9H_{20}O_5Si$; d: 1.070 g/cm^3; Epoxy group content: 17.50 %.
SA stearic acid	China	CH_3-$(CH_2)_{16}$-COOH d: 0.839 g/cm^3, M = 284.48

2.2 Methods

2.2.1 Surface treatment of fly ash with silane coupling agent GF80

The silane used was 2 wt% of the fly ash. 2 g of silane GF80 was mixed with 100 mL of ethanol solution and stirred for 30 min at 40 °C. 100 g of UFA was added into the solution and stirred for 4 h at 40 °C. The treated fly ash was then filtered, washed and dried at 100 °C in the oven for 12 h to remove the solvents. The silane-treated fly ash was labelled as FASGF80.

2.2.2 Surface treatment of fly ash with SA

The surface of FA particles (100 g in each case) was coated by immersing them in the solutions of SA (2 g of SA in 100 mL of acetone and toluene at the ratio of 3 : 1) separately under constant stirring for 30 min and dried for 24 h at room temperature. Then, it was vacuum dried to remove the solvents. The surface coated fly ash particles were designated as FASA.

2.2.3 Preparation of EP(DER 331)/silane and SA treated and untreated FA composites

To prepare polymer composite materials, silane and SA treated and untreated fly ash particles were mixed into the EP DER 331 with parts volume of the mixture (consist of fly ash and epoxy resin). After the mixtures were homogenously mixed, the curing agent DETA (amount of curing agent was calculated by epoxy content of epoxy resin) was added and the mixtures were moulded for curing. The mould was coated with a uniform thin film of silicone – a releasing agent for easy removal of cured specimen. The samples were cured at room temperature for about 24 h and further cured at 80 °C in the laboratory oven for 3 h. Then, the samples were removed from the mould and the electrical properties were inspected.

2.2.4 Methods for determining properties of materials

To compare the differences between the untreated and treated fly ash samples, the contact angle (Yuan and Lee, 2013) and IR spectroscopy of the samples were determined using the Thermo Cahn - Radian - Series 300 and IR 27 Brucker Tensor equipment.

2.2.5 Dielectric property measurements

The dielectric properties of the epoxy composites were measured using standard ASTM test procedures. The experiments were performed at a temperature of 25 °C and relative humidity of 60 %. Dielectric

constant and tan delta measurements in the frequency range of 4 x 10^2 - 10^6 Hz were performed using Agilent, USA and an average value of 5 samples was taken. The error in the measurement is within 2 %.
The dielectric strength measurement in the study was performed using a breakdown test cell designed using the appropriate electrodes according to ASTM D149 and the test was carried out in a medium of transformer oil (Figure 1). The electrodes (both top and bottom) are cylindrical electrodes of 25 mm diameter with edges rounded to 3.2 mm. The sample was placed between the electrodes and the AC (50 Hz) voltage was continuously increased at a speed of 500 V/s till the sample was broken down. The breakdown voltage, V (kV), of the sample was recorded and the dielectric strength, E (kV/mm) was calculated as E = V/t, where t is the thickness of the sample in millimeters.

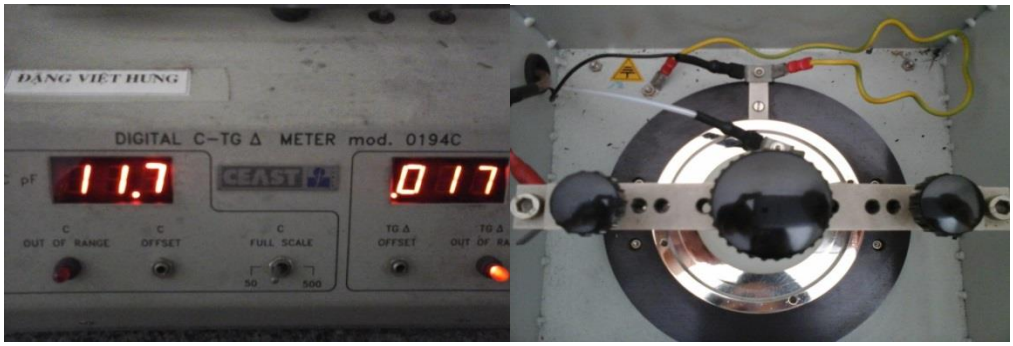


Figure 1: Measuring equipment electrical properties

3. Results and discussion

3.1. Characteristics of fly ash after treatment with GF80 and SA

The differences on the surfaces of untreated and treated FA samples were analysed using IR spectroscopy. Results are shown in Figures 2a and 2b.

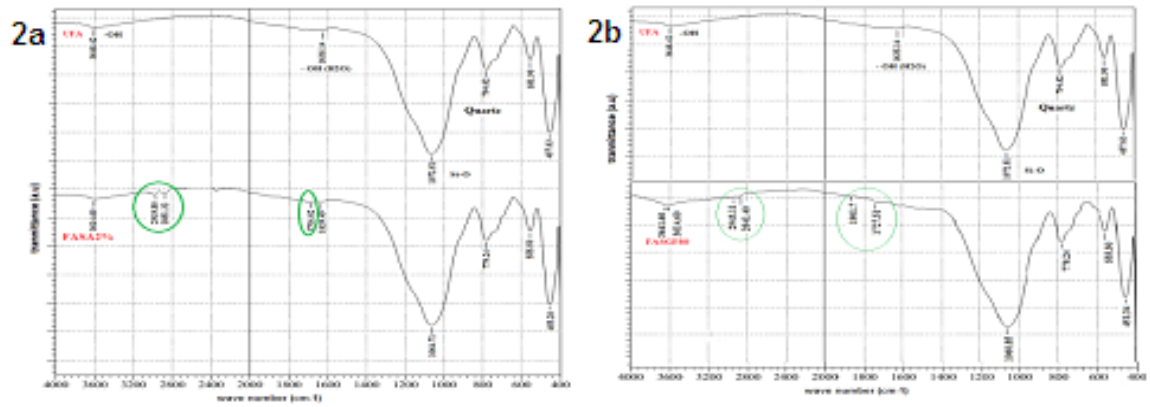


Figure 2: IR spectra of UFA, FASA 2 % (2a) and FAGF80 2 % (2b)

From Figure 2a, the IR spectra of the modified fly ash (FASA 2 %) display new peaks at 2,919 cm^{-1}; The wave number 2,851 cm^{-1} is typical peak for CH_2 and CH_3 groups and 1,704 cm^{-1} is the characteristic for C=O group of SA molecule. With sample FASGF80 2 % (Figure 2b), the IR spectra also show new peak at 1,727 cm^{-1}, which is the wave number specific to silane functional groups. This demonstrates the presence of these functional groups on the surface GF80 SA and fly ash.
The purpose of the modified method by SA and GF80 fly ash is to increase the activity of the particle surface or increase the wet ability of the particles with a polymer matrix. The fly ash particles will likely agglomerate and provide better interfacial adhesion with organic plastic matrix. The contact angle of the fly ash samples treated with SA and GF 80 in different liquid environments with same operating conditions was measured. The results are shown in Table 3.

Table 3: Contact angle of test liquids on silane and SA treated and untreated fly ash

Fly ash	Liquid environment	
	Water	Ethylene glycol
UFA	77.62°	61.66°
FASA 2 %	102.42°	100.69°
FASGF80 2 %	109.95°	100.64°

From Table 3, the contact angle of the samples FASA 2 % and FAGF80 2 % are higher in both polarised environments compared to UFA. This is because the silane and SA molecules attached onto the surface of the fly ash have transformed the surface of reinforcing fillers from hydrophilic to hydrophobic transfer over. The modified properties allow the fly ash inorganic particles to interact better with the polymer matrix.

3.2 Influence of fly ash to the electrical properties of polymer composite materials

3.2.1 Influence of fly ash content to volume resistivity of DER 331 epoxy composite materials/fly ash

The values of the resistivity of the composite samples are shown in Table 4. Results in Table 4 show that the volume resistivity of the DER 331 epoxy composite materials/fly ash has a value of about 10^{13} Ω.cm. The volume resistivity of composite materials decrease with increasing fly ash content in all composite samples (both with untreated and treated fly ash samples). The volume resistivity of the composite samples with modified fly ash are higher than the composite samples of untreated fly ash at all volume fractions. This is because the modified fly ash has improved the interfacial adhesion of the inorganic particles with resin matrix and simultaneously limit the polarisation of OH groups, which favour water-absorbing particles on the surface of fly ash. This results in the improvement of the volume resistivity of the material compared to the non-modified fly ash samples.
FAGF80 2 % samples have higher resistivity compared to the FASA 2 % samples due to the interaction of epoxy functional groups in the molecule silane GF80 with OH groups on the surface of fly ash and amino groups of DETA in the process of curing composite materials.

Table 4: Influence of fly ash content tovolume resistivity of DER 331 epoxy composite materials / fly ash

Fly ash (parts volume)	Volume resistivity (Ω.cm)		
	EP/UFA	EP/FASA 2 %	EP/FASGF80 2 %
0	43.1×10^{13}	43.1×10^{13}	43.1×10^{13}
20	24.9×10^{13}	30.0×10^{13}	36.9×10^{13}
30	16.5×10^{13}	18.6×10^{13}	25.7×10^{13}
40	6.7×10^{13}	9.1×10^{13}	15.4×10^{13}
50	5.6×10^{13}	6.8×10^{13}	10.2×10^{13}
60	5.4×10^{13}	6.6×10^{13}	8.6×10^{13}

3.2.2 Influence of fly ash content to surface resistivity of DER 331 epoxy composite materials /fly ash

Figure 3 shows that the surface resistivity increase in all samples at levels of 20 parts volume fly ash. Further increasing the fly ash content decreases the surface resistivity of samples. The surface resistivity of EP/UFA sample has largest reduction while EP/FAGF80 sample has the least reduction. This proves that the modified fly ash improves the surface resistivity of the composite materials, which is due to the good interaction between the modified fly ash with resin matrix.

3.2.3 Influence of fly ash content to dielectric constant and dielectric loss of DER 331 epoxy composite materials /unteated fly ash

To determine the ddielectric constant and dielectric loss, experiments has been conducted on the samples at room temperature with humidity of 60 % and 106 Hz frequency. Results of the dielectric constant and dielectric loss of DER 331 epoxy composite materials/unteated fly ash with different fly ash content are presented in Table 5.
From Table 5, it can be seen that all composite fly ash samples with epoxy resin have higher dielectric constants than the untreated fly ash. These values increase with increasing fly ash content. This is because the fly ash components contain metal oxides such as SiO_2, Al_2O_3, and Fe_2O_3.

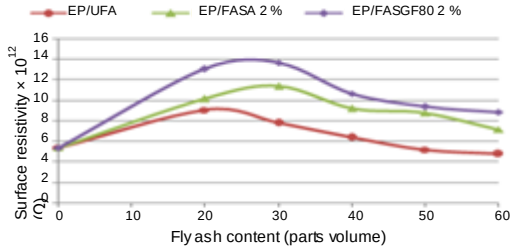


Figure 3: Influence of fly ash content to surface resistivity of DER 331 epoxy composite materials/fly ash

Table 5: Influence of fly ash content to dielectric constant and dielectric loss of DER 331 epoxy composite materials/unteated fly ash

Sample	Dielectric constant, ε	Dielectric loss, tan δ
EP	3.46	0.017
EP/UFA20	3.58	0.017
EP/UFA30	3.63	0.018
EP/UFA40	3.78	0.020
EP/UFA50	3.98	0.025
EP/UFA60	4.04	0.025
Sample	Dielectric constant, ε	Dielectric loss, tan δ
EP	3.46	0.017
EP/UFA20	3.58	0.017
EP/UFA30	3.63	0.018
EP/UFA40	3.78	0.020
EP/UFA50	3.98	0.025
EP/UFA60	4.04	0.025

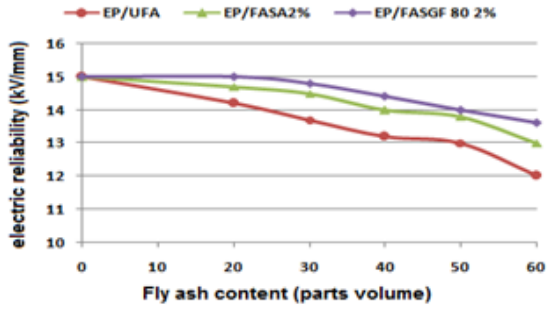


Figure 4: Influence of fly ash content to reliable electricity of DER 331 epoxy composite materials/fly ash

3.2.4 Influence of fly ash content to reliable electricity of DER 331 epoxy composite materials /fly ash

Empirical survey was conducted to investigate the electrical durability of DER 331 epoxy composite samples/fly ash at room temperature with an AC voltage, frequency 50 Hz, and thickness of the test sample of 2 mm. The results, which measure the effect of fly ash content on the electricity reliability of EP/FA composite materials are shown in Figure 4.

Electrical reliability of all composite samples decrease with increasing fly ash content. The values of the electrical reliability of the composite samples are smaller than the epoxy resin because of the oxide components in fly ash. Using the same amount of fly ash, the fly ash composite samples are modified to achieve higher electric reliability than the composite sample have not modified fly ash. Specific values of the electric reliability of the samples are presented in Table 6.

Table 6: Effect of fly ash content to reliable electricity samples of DER 331 epoxy composite materials/fly ash

Fly ash (parts volume)	Electric reliability (kV/mm)		
	EP/UFA	EP/FASA 2 %	EP/FASGF80 2 %
0	15.0	15.0	15.0
20	14.2	14.7	15.0
30	13.7	14.5	14.8
40	13.2	14.0	14.4
50	13.0	13.8	14.0
60	12.0	13.0	13.6

4. Conclusion

Results show that the samples with fly ash modified by SA 2 % and silane GF80 2 %has significantly improved insulating properties compared to the non-modified fly ash samples. Similarly, the treated fly ash samples produce highed dielectric constant and loss compared to the untreated fly ash samples.

Acknowledgement

The authors would like to thank the National Key Laboratory for Polymer and Composite Materials - Hanoi University of Science and Technology for helping during this study.

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