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Utilization of Fly Ash Acid Residue as a Reinforcing Filler in Ethylene Propylene Diene Monomer Rubber

Feng Luo, Shujuan Wang, Bing Xue, Yinshan Jiang, Cundi Wei
Department of Materials Science and Engineering, Key Laboratory of Automobile Materials of Ministry of Education, Jilin University, Changchun 130025, People's Republic of China
Correspondence to: C. Wei (E-mail: Weicd@jlu.edu.cn)

ABSTRACT: Fly ash acid residue (FAAR), a by-product of circulating fluidized bed fly ash extracted Al_2O_3 by acid leaching method, has been posing problems because of its disposal. The major chemical components of FAAR are amorphous SiO_2 (66.38 wt %) and unburned carbon (20 wt %). Attempts were made for its application as a reinforcing filler for ethylene propylene diene monomer (EPDM) rubber in this article. Surface modification for FAAR by silane coupling agent (Si69) was carried out. The effect of surface modification and unburned carbon existing in FAAR on the performance of FAAR was characterized by Fourier transform infrared and dispersibility test. The results indicated that surface modification could reduce the hydrophilicity of FAAR and unburned carbon also had positive effect on the dispersion of FAAR particles in kerosene. The effect of partial replacement of carbon black by FAAR on the curing behavior, mechanical properties, and morphological characteristics of EPDM rubber was also studied. It was proved that with partial replacement of carbon black by FAAR, the cure time (t_{90}) and maximum torque (M_H) of EPDM composites increased with the content of FAAR. The mechanical properties were significantly improved when 15 wt % of carbon black was replaced by FAAR. SEM micrographs confirmed that surface modification can improve the compatibility between FAAR and rubber matrix. Unburned carbon existing in FAAR was also beneficial to the interface bonding. © 2012 Wiley Periodicals, Inc. J. Appl. Polym. Sci. 129: 1053–1059, 2013

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INTRODUCTION

In general, rubber is always filled with appropriate type of filler to improve the mechanical properties, process ability.[1] Carbon black (CB) is the most important reinforcing filler used so far owing to its superior rubber–filler interaction owing to the larger surface area and higher surface activity. Apart from CB, silica is widely used as a nonblack reinforcing filler because of its fine particle size (high-specific surface area), and performance in improving the mechanical properties of the rubber, particularly tensile strength, tear resistance, abrasion resistance, and hardness.[2] Some inorganic particles such as clay,[3,4] nanokaolin,[5] lignin,[6] pulverized polyurethane foam particles,[7,8] cellulose fiber,[9] Halloysite Nanotubes,[10] Rattan,[11] and bentonite[12] have been investigated as the reinforcing filler for rubber. Recently, many attempts have been made to use silica from by-product, such as black rice-husk ash,[13,14] and white rice-husk ash[15,16] as the reinforcing filler for rubber because of cost savings, good mechanical properties, better dimensional stability, and environmental issues. Much research[17–28] has been carried out on fly ash as a reinforcing filler for rubber because of its high silica content. However, there are several disadvantages when fly ash is used as a reinforcing filler. For example, there are about 25–40 wt % Al_2O_3 apart from silica; the highly crystalline structure usually results in the low activity of fly ash; the surface of fly ash is always smooth, which induces the poor compatibility between fly ash and rubber matrix.

However, extracting Al_2O_3 from circulating fluidized bed fly ash has been carried out in China because of the absence of aluminum sources. At the same time, a large amount of fly ash acid residue (FAAR) is generated in the process of extracting Al_2O_3. According to our previous description mentioned above, FAAR has more several advantages than fly ash when used as a reinforcing filler. As far as the chemical composition is concerned, FAAR contains about 70 wt % silica, which is higher than fly ash and 20 wt % unburned carbon, which may improve the compatibility with rubber matrix. Moreover, FAAR has higher surface activity than fly ash because of its amorphous phase and the surface is irregularity, which provides better adhesion between FAAR and rubber matrix. It is noticed that FAAR contains 20 wt % unburned carbon and this content is much more than original fly ash. A mass of unburned carbon in FAAR may affect some properties of rubber. However, there is rare research article about this issue.

In this study, FAAR was first treated by silane coupling agent, *bis*-(3-triethoxysilylpropyl) tetrasulfane (Si69) because its surfaces have hydrophilic siloxane and silanol groups, resulting in strong filler–filler interaction by hydrogen bonds. After surface modification, FAAR was used as a reinforcing filler for EPDM rubber. The effect of partial replacement of CB by FAAR on the curing characteristic, mechanical properties, and morphological properties of EPDM composites was studied. The effect of unburned carbon on the properties of EPDM was also studied.

EXPERIMENTAL

Raw Materials

FAAR was supplied by the laboratory of Jilin University, China. Prior to use, FAAR was ground with a ball mill. A successive washing was carried out to remove the unreacted acid and later filtered. The filtered sample was dried at 105°C in an oven and was used for surface modification. Ethylene–propylene–diene monomer rubber (J-3080) was supplied by China National Petroleum, Jilin Cty, Jilin Province. CB (N330) was supplied by TongWang Chemical Industry, Changzhou, China. *Bis*-(3-triethoxysilylpropyl) tetrasulfane (designated as Si69), $[(C_2H_5O)_3{-}Si{-}(CH_2)_3{-}S_4{-}(CH_2)_3{-}Si{-}(C_2H_5O)_3]$, was supplied by NanJing XinHuai Scientific, Nanjing City, Jiangsu Province. Formulation of ethylene propylene diene monomer (EPDM) rubber compounds, supplied by sealing rubber factory, was as follows: 100 phr rubber, 65 phr CB, 5.0 phr zinc oxide (ZnO), 1.5 phr stearic acid, 3.5 phr antioxidant, 45 phr liquid paraffin, 1.5 phr sulfur, 6 phr dicumyl peroxide (DCP), 1.5 phr tetramethylthiuram disulphide (TMTD), 1.0 phr accelerator M (MBT).

Surface Treatment of FAAR

Two methods were used for surface modification; (1) FAAR that was treated directly by silane couple agent (Si69), namely FAAR-Si69; (2) FAAR that was first subjected to calcine at 700°C for 2 h until unburned carbon disappear, namely C-FAAR. Then, C-FAAR was treated by Si69 and the final product was called C-FAAR-Si69. The surface modification process commenced as follows: an appropriate amount of Si69 (different weight percentage of FAAR from 0 to 4%) and 50 mL ethanol was added in the beaker, and then stirred for 20 min. A sample of 20 g of FAAR or C-FAAR was then added into the solution while stirring for a further period of 1 h. The sample was then dried at 100°C for 10 h in an oven till constant weight.

Particle Dispersion Experiment

A sample of 0.5 g solid sample (FAAR, FAAR-Si69, or C-FAAR-Si69) and 10 mL solvent (kerosene) were introduced into a dried and clean graduated test tube. The whole system was oscillated for 1 min. The turbid solution formed and then the sedimentation of solid particles began. The liquid–solid interface appeared in the sedimentation process. When the interface reached the position of 5 mL scale of test tube, the time it costed was defined as $T_{1/2}$ here. A big $T_{1/2}$ value will display a better dispersibility for particles in solvent. According to the result of particle dispersion test, the solid particle which had best dispersibility in solvent was used as a reinforcing filler.

Preparation of EPDM/CB/FAAR Composites

In the following experiments, three kinds of FAAR particles (FAAR, FAAR-Si69, and C-FAAR-Si69) were substituted for CB as the reinforcing filler for EPDM rubber. The rate of substitution was from 0 to 20%. There were two steps for making the rubber samples for property evaluation. In the first step, rubber was masticated with fillers (CB and FAAR), ZnO, stearic acid, antioxidant, and liquid paraffin in a laboratory two-roll mill (Education apparatus, Changchun, China) for 20 min at a rotor speed of 40 rpm. The samples were kept for 24 h in room temperature. In the second step, the rubber mixture was compounded with vulcanization chemicals (sulfur, DCP, TMTD, and MBT) on the two-roll mill for a further 20 min, and then cured in an electrically heated hydraulic press at 160°C under a pressure of 10 MPa for the respective optimum cure time.

Sample Analysis and Measurement

An oxide analysis of FAAR was obtained by chemical analysis and the major crystalline phases determined by powder X-ray diffraction (XRD) analysis. Surface morphology was investigated using S-570(HITACHI) scanning electron microscopy. Fourier transform infrared (FTIR) of the sample was carried out on a Nexus 670 auto FTIR spectrometer. The particle size was determined by JL9200 laser particle size analyzer (Weina, Jinan, China). The curing characteristics of the rubber compounds at 160°C were determined by a Moving Die Rheometer (MDR-2000E). Tensile strength of the specimen was determined according to ASTM D 412-06a. Tear strength of the specimen was determined according to ASTM D 624-00.

RESULTS AND DISCUSSION

Characteristics of FAAR

Chemical and Crystalline Compositions. The chemical composition of FAAR determined by chemical analysis is summarized in Table I. It can be readily seen that the chemical composition of FAAR was silica (66.38 wt %), unburned carbon (20.20 wt %), aluminum oxide, and other minor metal oxides. Silica is the important reinforcing composition. Hence, FAAR had the potential application as a reinforcing filler for rubber. It

Table I. Chemical Composition of FAAR

Oxide	Mass (%)
SiO_2	66.38
Al_2O_3	6.62
TiO_2	2.36
MgO	1.24
Fe_2O_3	0.19
CaO	0.23
FeO	0.10
K_2O	0.10
LOSS	20.20

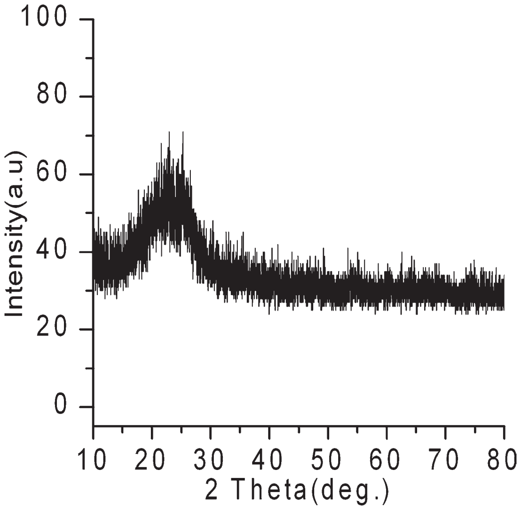

Figure 1. XRD pattern of FAAR.

is worth noticing that the amount of unburned carbon was much more than fly ash. Unburned carbon was expected to enhance the compatibility with rubber because of its nonpolarity.

Figure 1 shows the XDR pattern of the FAAR. The broad diffraction scatter at the beginning of the XRD trace indicated a high degree of disorder within the phases. This indicated silica had an amorphous structure, which was similar with precipitated silica. As it is well known, the structure of fly ash is highly crystalline, leading to low activity. However, FAAR had higher activity than fly ash because of its amorphous structure, which was beneficial to FAAR as a reinforcing filler.

Morphology and Particle Size. The morphology and the particle size distribution of the FAAR are shown in Figures 2 and 3, respectively. From the SEM micrograph, it can be seen that FAAR was irregular with coarse surface. The average particle diameter determined by laser particle size analyzer was 3.55 μm.

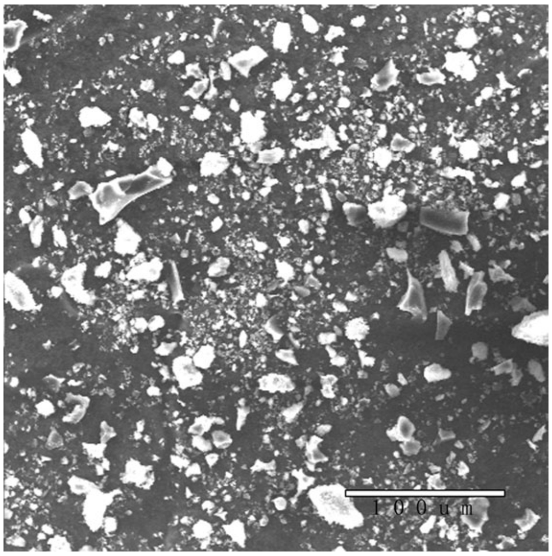

Figure 2. SEM micrograph of FAAR.

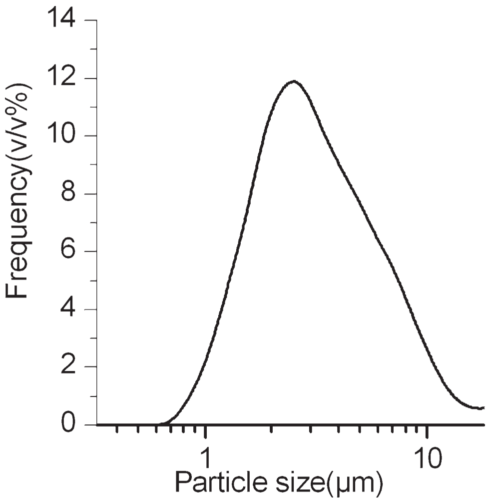

Figure 3. Particle size distribution of FAAR.

It is worth noticing that the particles of fly ash were globular with smooth surface, whereas FAAR was irregular. One would expect to obtain higher interface bonding between rubber and FAAR particles and improved mechanical properties of FAAR/EPDM composites as a result of rougher physical surface.

Surface Modification of FAAR

FTIR Analysis. The FTIR spectral characteristics of FAAR, FAAR-Si69, and C-FAAR-Si69 particles are shown in Figure 4. The peaks at 3446 and 1640 cm^{-1} were attributed to the dissymmetry stretching vibration and bending vibration of —OH, respectively. The peaks at 1097 and 806 cm^{-1} were assigned to the dissymmetry stretching vibration and symmetry stretching vibration of Si—O—Si, respectively. As shown in Figure 4, FTIR spectra of FAAR were similar with precipitated silica.[25] After modification by Si69, the intensity of —OH band reduced. At the same time, two sharp bands at 2921 and 2852 cm^{-1} appeared, which were attributed to the asymmetric and symmetric CH_2 stretch, respectively.[29] It is concluded that the hydroxyl group of FAAR has been successfully reacted with Si69 and Si—O—R bond formed, where *R* was

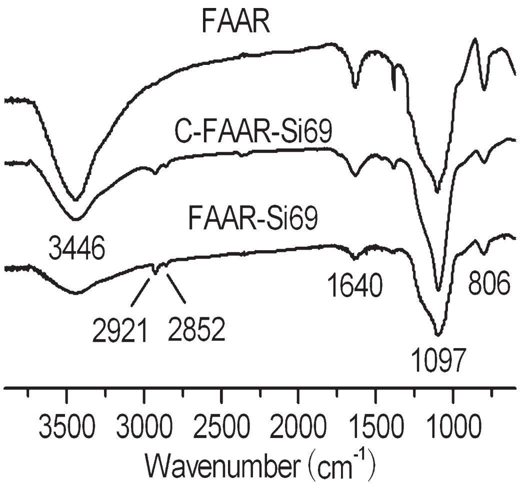

Figure 4. FTIR spectra of FAAR, C-FAAR-Si69, and FAAR-Si69.

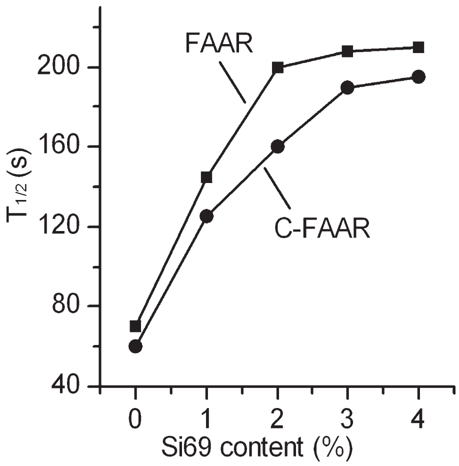

Figure 5. Dispersion experiments of FAAR and C-FAAR-Si69 modified by different Si69 content.

$(C_2H_5O)_2—Si—(CH_2)_3—S_4—(CH_2)_3—Si—(OC_2H_5)_3$. Surface modification decreased the hydrophily of FAAR surface, which was expected to enhance the compatibility with rubber. The weak peak of dissymmetry stretching vibration of —OH existing in FAAR-Si69 and C-FAAR-Si69 indicated that not all Si—OH groups could react with Si69. The reason could be owing to the longer chains of Si69, which consisted of tri-ethoxysilylpropyl groups on their molecules. These groups probably caused a steric hindrance for reaction between Si69 and Si—OH.

Particle Dispersion Analysis. To a certain degree, the dispersibility of inorganic filler in organic solvents shows the compatibility between filler and rubber matrix. Figure 5 shows the dispersibility of FAAR and C-FAAR particles modified by silane coupling agent at different contents in apolar kerosene. It can be seen that the surface modification largely increased the dispersibility of FAAR in nonpolar kerosene. $T_{1/2}$ values of the FAAR increased gradually with the increase of the content of Si69. When the content of Si69 exceeded 2 wt %, the effect of surface modification was no longer evident. The optimal surface modification for C-FAAR appeared when the Si69 content was 3 wt %. This result may be explained by the polarity theory. FAAR is polar and hydrophilic but kerosene is apolar and hydrophobic. Hence, FAAR was not well dispersed in organic solvents. FAAR became apolar and hydrophobic after surface modification by Si69 and thus enhanced the dispersibility in kerosene. Meanwhile, as shown in Figure 5, $T_{1/2}$ values of the FAAR were higher than that of C-FAAR when the same content of Si69 was used. The possible reason is that unburned carbon adhered to the surface of FAAR, which reduced the hydrophilic of particle and thus enhanced the dispersibility of FAAR in organic solvent.

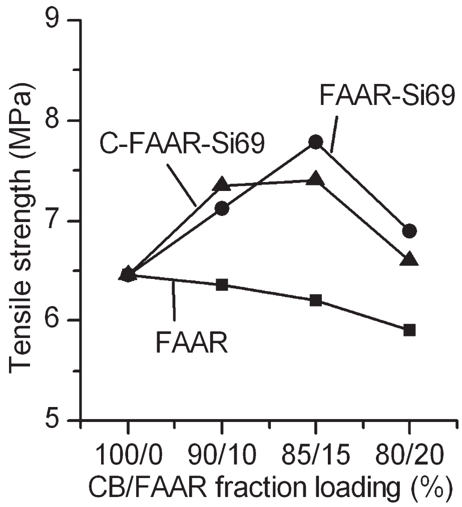

Figure 6. Comparison of tensile strength of EPDM composite with different reinforcing fillers.

Performance of EPDM Rubber

Cure Characteristics. As summarized in Table II, the replacement of CB by three kinds of FAAR mentioned above had minor effect on scorch time (t_{10}) and minimum torque (M_L), but had strong effect on cure time (t_{90}) and maximum torque (M_H) of EPDM rubber. The cure time of EPDM rubber increased as FAAR replacement content increased. It is worth noting that the cure time of rubber filled with FAAR-Si69 and C-FAAR-Si69 showed shorter value than that of filled with FAAR at similar blend ratio. The following text can explain these results. When FAAR was used as a reinforcing filler, the presence of silanol groups retarded curing in sulfur-cured vulcanizates because of the absorption of curatives and cure activators on silica surfaces.[30] The silanol group increased with the FAAR content increase, which resulted in longer cure time. In addition, the surfaces of FAAR were polar and hydrophilic. There was a strong tendency to absorb moisture, which adversely influenced cure and properties of rubber vulcanizates.[31] When FAAR was treated by Si69, the silanol groups existing in FAAR surface decreased in a largely scale. Hence, the cure time of the rubber filled with FAAR-Si69 and C-FAAR-Si69 showed shorter value than that of the rubber filled with FAAR.

Table II. Cure Behavior of EPDM Composites with Different Reinforcing Fillers

		CB/FAAR (%/%)			CB/FAAR-Si69 (%/%)			CB/C-FAAR-Si69 (%/%)		
Sample	CB 100%	90/10	85/15	80/20	90/10	85/15	80/20	90/10	85/15	80/20
t_{10}	60″	63″	63″	63″	60″	64″	65″	60″	60″	64″
t_{90}	8′21″	8′58″	9′04″	9′15″	8′10″	8′54″	9′30″	8′45″	8′57″	9′10″
M_H (N.m)	1.19	1.24	1.26	1.30	1.28	1.31	1.33	1.30	1.33	1.35
M_L (N.m)	0.04	0.05	0.04	0.04	0.05	0.04	0.05	0.05	0.05	0.05

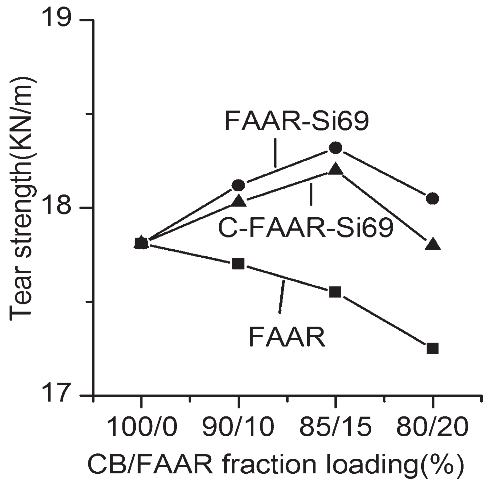

Figure 7. Comparison of tear strength of EPDM composite with different reinforcing fillers.

It is noted that the cure time of the rubber filled with C-FAAR-Si69 showed shorter value than that of the rubber filled with FAAR-Si69. This was because C-FAAR-Si69 had more Si69 content than FAAR-Si69 at similar filler loadings and thus the cure time of composite was shorter. The *S*-atom existing in Si69 can react with EPDM and enhance the cure rate simultaneity. Thus, the cure time of composite was shorter.

As summarized in Table II, the maximum torque (M_H) for three blends increased with an increase in FAAR replacement content. This probably resulted from the rigid and hard particle of FAAR, which tended to immobilize the movement of macromolecular rubber chains. As a result, EPDM rubber became harder and stiffer. However, the M_H of rubber filled with FAAR-Si69 and C-FAAR-Si69 exhibited higher value than that of filled with FAAR at similar blend ratio. The result indicated surface treatment was beneficial to rubber–filler interaction, leading to additional crosslinks in the network structure. The improvement of M_H resulted from the increased interaction between filler and rubber matrix, particularly with the presence of Si69.[32]

Mechanical Property. Figures 6 and 7 show the mechanical properties of EPDM rubber. It is clearly seen that the tensile strength and tear strength of rubber exhibited the similar trend. The mechanical properties decreased when FAAR partially replaced CB as the reinforcing filler. This result was owing to the poor filler–rubber interaction. However, the tensile strength and tear strength of EPDM rubber increased gradually when FAAR-Si69 and C-FAAR-Si69 partially replaced CB as the reinforcing filler. The maximum of the tensile strength and tear strength

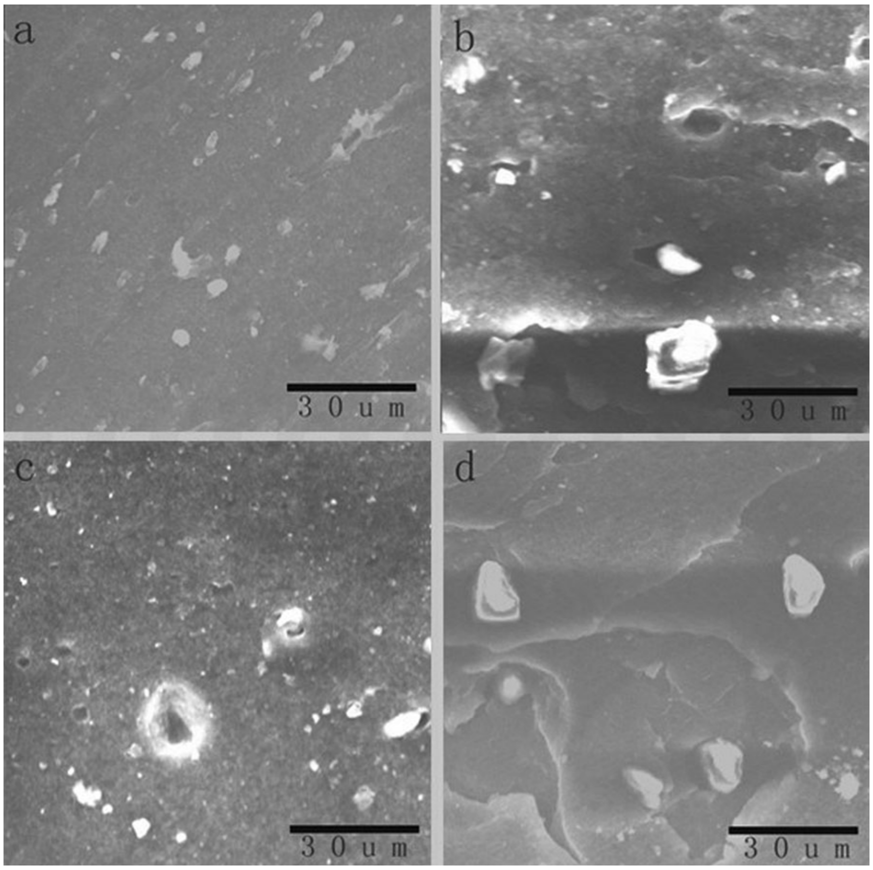

Figure 8. SEM micrographs for EPDM composites filled with different reinforcing fillers. (a) CB, (b) CB+FAAR, (c) CB+FAAR-Si69, and (d) CB+C-FAAR-Si69.

appeared when the rate of substitution was 15%. When FAAR was treated by Si69, a stable Si—O—R chemical bond formed between the Si69 and the filler particle surface. There was a decrease in mechanical properties when the rate of substitution was beyond 15%. This was because larger particles of FAAR caused stress concentration point and resulted in a decrease in mechanical properties. Worth noting was that EPDM blends filled with FAAR-Si69 exhibited higher mechanical properties than that of filled with C-FAAR-Si69. This result was similar to the dispersion result obtained above. SEM micrograph of rubber can also explain this result in the following section.

Morphological Characteristics. Figure 8(a) shows the tensile fracture surface of EPDM composites when CB as the only reinforcing filler. Figure 8(b–d) shows the fracture surface of the EPDM composites when the CB was replaced by different types of FAAR fillers. As shown in Figure 8(a), adhesion between CB and EPDM matrix was very tight. The CB particles were found to be wrapped up in the rubber matrix and no hole existed in matrix. This was the reason why the CB was the most important reinforcing filler for rubber. As shown in Figure 8(b), the compatibility between FAAR and EPDM matrix was very poor. The graph shows that gaps existing around FAAR were very evident. The reason was that the large holes formed when stress was applied. This can explain the reason why the mechanical properties decreased when FAAR was added. The compatibility between FAAR-Si69 and EPDM matrix is shown in Figure 8(c). The graph shows that there were no evident larger cavities existing in matrix. Only narrow gap appeared when stress was applied. This indicated that the compatibility between rubber and filler was improved when FAAR was treated by Si69. This was coupled with an improvement in mechanical properties of EPDM composite with FAAR-Si69. Figure 8(d) shows the compatibility between C-FAAR-Si69 and EPDM matrix. The gap was seems to be larger between C-FAAR-Si69 and matrix than FAAR-Si69 and matrix. This indicated that the compatibility between matrix and FAAR-Si69 was better than matrix and C-FAAR-Si69. It can explain the reason why the mechanical properties of EPDM rubber filled with FAAR-Si69 were better than that of EPDM rubber filled by C-FAAR-Si69. It is concluded that unburned carbon existing in FAAR was beneficial to the compatibility of the FAAR particle with the rubber matrix.

CONCLUSIONS

FAAR modified by silane coupling agent was utilized as a reinforcing filler for EPDM rubber. Surface modification reduced the hydrophilic of FAAR and thus enhanced the compatibility of FAAR particles with rubber matrix. Unburned carbon was beneficial to compatibility between FAAR and rubber matrix. The mechanical properties of EPDM composites increased gradually with the replacement of CB by FAAR modified by Si69. The maximum of the tensile strength and tear strength of EPDM rubber appeared when the rate of substitution was 15%.

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