

PREPARATION AND CHARACTERIZATION OF SILICA FROM SUGARCANE BAGASSE ASH AS A FILLER IN NATURAL RUBBER

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Abstract

In this study, silica particles were prepared from sugarcane bagasse ash (SBA) by the acid precipitation method. The chemical composition, chemical structure, morphology, and particle size distribution of the SBA, extracted silica, and commercial silica were characterized. The silica content in the SBA was 64.8%. After the extraction process, the silica content increased to 91.4%. X-ray diffraction analysis indicated that the extracted silica was amorphous. The Fourier transform infrared spectrum of the extracted silica confirmed the silica's characteristics. The results from a scanning electron microscope and particle size analyzer revealed that the extracted silica had an irregular shape with a particle size of $26.97 \pm 18.43 \mu\text{m}$. The specific surface area of the commercial silica was higher than that of the extracted silica. The effect of the extracted silica content on the cure characteristics, mechanical properties, and morphology of extracted silica/natural rubber (NR) composites were studied. Moreover, at a silica content of 20 phr, the properties of the extracted silica/NR composite were compared with the commercial silica/NR composite. The cure time of the extracted silica/NR composites increased with increasing the extracted silica content due to the disturbance of the vulcanization process by the silica surface. The extracted silica/NR composite showed a shorter scorch time and cure time than the commercial silica/NR composite due to acceleration of the vulcanization process by the residue metal oxide in the extracted silica. The modulus and hardness of the NR composites were increased while the elongation at break, tensile strength, and tear strength were decreased with increasing the extracted silica content. The mechanical properties of the commercial silica/NR composite were higher than those of the extracted silica/NR composites because the commercial silica had a smaller particle size than the extracted silica. The incorporation of a silane coupling agent (Si-69) resulted in reduced scorch time

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and cure time but improved the mechanical properties of the composites filled with 20 phr of both types of silica.

Keywords: Silica, sugarcane bagasse ash, natural rubber, composite

Introduction

Natural rubber (NR) composites have been extensively studied over a long period of time because they have been used in various applications. The properties and performances of NR compounds depend mainly on the right compounding ingredient combination, including rubber additives and reinforcing fillers. Different types of reinforcing fillers have a significant effect on the properties of NR compounds (Ahmed *et al.*, 2014; Sengloyuan, 2015). Silica has been used as a reinforcing filler in the natural rubber industry for improving the mechanical properties of natural rubber composites such as the tensile properties, hardness, abrasion resistance, and rolling resistance (Rattanasom *et al.*, 2017). However, the production of silica particles for commercial applications is still high in cost and energy consumption and is hazardous to the worker (Faizul *et al.*, 2013). It has been reported that silica particles can be obtained from agricultural wastes e.g. rice husk (Kalapathy *et al.*, 2000), rice straw (Nandiyanto *et al.*, 2016) and sugarcane bagasse (Norsuraya *et al.*, 2016). The deposition of silica in the agriculture residues is influenced by the quantity and availability of silicon in the soil (Norsuraya *et al.*, 2016). Sugarcane bagasse ash (SBA) is an interesting agricultural waste because it produces silica due to its high content of silicon, is low cost, and is abundantly available in an agricultural country. SBA is a waste in the sugar industry and is a by-product from bio-power stations. It needs to be disposed of properly, otherwise it may cause a major environmentally sustainable issue. Using SBA as a source of silica for natural rubber composite applications not only provides a solution for waste disposal but also recovers a valuable asset from agricultural waste. The preparation of silica from SBA with different processes has been reported by Worathanakul *et al.* (2009), Rahman *et al.*

(2015), Norsuraya *et al.* (2016), Alves, *et al.* (2017), and Huabcharoen *et al.* (2017).

In this study, silica particles were extracted from SBA. The extracted silica and commercial silica were characterized in terms of their chemical composition, chemical structure, particle size distribution, surface properties, and morphology. The effect of the extracted silica content on the cure characteristics, mechanical properties, and morphology of the NR composites were studied. Moreover, the properties of the NR composites filled with 20 phr of extracted silica and commercial silica with and without a silane coupling agent (Si-69) were compared.

Materials and Methods

Materials

SBA was supplied by Khonburi Power Plant Co., Ltd. (Khonburi, Nakhon Ratchasima, Thailand). Natural rubber (NR, STR XL) was purchased from Natural Art and Technology Co., Ltd. (Pluak Daeng, Rayong, Thailand). The chemicals used in the silica preparation were hydrochloric acid (HCl) and sodium hydroxide (NaOH). Commercial silica (silica, Ultrasil VN2), bis[3-(triethoxysilyl)propyl]tetrasulfide (Si-69) and other ingredients which were used to vulcanize natural rubber including stearic acid, zinc oxide, N-cyclohexyl-2-benzothiazole-2-sulphenamide (CBS), and sulfur were supplied by PI Industry Co., Ltd. (Bangkok, Thailand).

Preparation of Silica from SBA

One hundred grams of SBA was firstly washed with distilled water and dried at 80°C for 24 h and then refluxed with 1 molL⁻¹ HCl at 100°C for 4 h to remove dirt and metal oxide impurities. The mixture was cooled down to room temperature and filtered using filter

paper (Whatman No.41). The remaining solid portion was continuously washed with distilled water until pH 7 was obtained and then filtered and dried. The acid-leached SBA was dispersed and vigorously stirred in 50 ml of 3 molL⁻¹ sodium hydroxide (NaOH) for 4 h at 80°C to produce sodium silicate solution (Norsuraya *et al.* 2016). The suspension was cooled down and filtered in order to remove solid residue. Then, the solution was precipitated with 1 molL⁻¹ HCl dropwise until pH 8 was obtained. After aging for 1 h at room temperature, the gel was gently broken, washed with water, filtered, and freeze-dried in a deep vacuum overnight to remove water.

Characterization of SBA, Extracted Silica and Commercial Silica

The chemical compositions of the SBA, extracted silica, and commercial silica were analyzed using a wavelength dispersive X-ray fluorescence spectrometer (XRF, Axios, Malvern Panalytical, Malvern, UK).

The FTIR spectra of the SBA, extracted silica, and commercial silica were performed in the attenuated total reflection mode at a resolution of 4 cm⁻¹ in the range of 400-4,000 cm⁻¹ using a Fourier transform infrared spectrometer (FTIR, Bruker T27/ Hyp2000, Bruker Corp., Billerica, MA, USA).

X-ray diffraction pattern of all samples were carried out with an X-ray diffractometer (Bruker D2 PHASER, Bruker Corp., Billerica, MA, USA) in the range of 2θ = 10-80°.

The particle size distribution of the extracted silica and commercial silica was

measured by using a laser scattering particle size distribution analyzer (Horiba LA- 950, Horiba Ltd., Kyoto, Japan).

The morphologies of the extracted silica and commercial silica were examined using a scanning electron microscope (SEM, Hitachi S3400N, Hitachi Ltd., Tokyo, Japan) at 10 keV. The samples were coated with gold before examination.

The surface properties of the extracted silica and commercial silica were characterized using the Brunauer-Emmett-Teller method (BELSORP-mini II, Microtrac Inc., Montgomeryville, PA, USA).

Preparation of NR Composites

Formulations of the natural rubber composites are shown in Table 1. The compounds were mixed using an internal mixer (CT, Chareon Tut Co., Ltd., Samut Prakarn, Thailand) at 60°C. The total mixing time was 25 min. Test specimens were prepared using a compression molding machine (Labtech LP20-B, Labtech Engineering Co. Ltd., Samut Prakarn, Thailand) under a pressure of 150 MPa and a temperature of 150°C.

Characterization of NR Composites

Cure Characteristics

The maximum torque (MH), minimum torque (ML), scorch time (t_{s1}), and cure time (t₉₀) of the NR and NR composites were investigated with a moving die rheometer (MDR, GOTECH/GT-M200F, Gotech Testing Machines, Inc., Taichung City, Taiwan) at 150°C.

Table 1. Formulation of natural rubber compounds

Materials	Content (phr)					
Natural rubber	100	100	100	100	100	100
ZnO	5	5	5	5	5	5
Stearic acid	2	2	2	2	2	2
CBS	1.5	1.5	1.5	1.5	1.5	1.5
Sulfur	1	1	1	1	1	1
Extracted silica	-	1	5	10	20	-
Commercial silica	-	-	-	-	-	20
Si-69					2	2

Mechanical Properties

The tensile properties of the NR and NR composites were tested according to ASTM D412 using a universal testing machine (UTM, INSTRON/5565, Instron, Norwood, MA, USA) with a load cell of 5 kN, a crosshead speed of 500 mm/min, and a gauge length of 33 mm. The modulus at 100% elongation, modulus at 300% elongation, tensile strength, and elongation at break of the NR and NR composites were measured.

The tear properties of the NR and NR composites were tested according to ASTM D624 using the UTM, INSTRON/5565 with a load cell of 5 kN at a crosshead speed of 500 mm/min.

The hardness of the NR and NR composites were determined according to ASTM D2240 using a shore A durometer (Type BS61 II, Bareiss Prüfgerätebau GmbH, Oberdischingen, Germany).

Morphological Properties

Tensile fractured surfaces of NR and NR composites were examined using a scanning electron microscope (SEM, Hitachi S3400N) at 5 keV. The samples were coated with gold before examination.

Results and Discussion

Characterization of SBA, Extracted Silica and Commercial Silica

The chemical composition of the SBA, extracted silica, and commercial silica from the XRF results are tabulated in Table 2. The SBA contained 64.80% of SiO_2 and the major metal oxide impurities were K_2O , Fe_2O_3 , CaO ,

and TiO_2 with contents of 15.20%, 8.90%, 5.20%, and 0.90%, respectively. After the extraction process, the silica content increased to 91.41% and various metal oxide contents were reduced. This indicated that the process was useful for removing some of the metallic impurities and resulted in improved purity of the silica (Lee *et al.*, 2017). However, the commercial silica contained high purity of silica (98.69%).

The FTIR spectra of the SBA, extracted silica, and commercial silica are shown in Figure 1. The spectrum of the extracted silica was the same as the spectrum of the commercial silica. The vibration peaks around 1,070, 780, and 460 cm^{-1} were attributed to the asymmetric stretching, symmetric stretching, and symmetric bending of the Si-O-Si bands, respectively (Ghorbani *et al.*, 2013; Huabcharoen *et al.*, 2017). These 3 peaks were the main indices of the silica materials. Moreover, the O-H stretching of the hydroxyl functional group at the silica surface was detected at of 960 cm^{-1} .

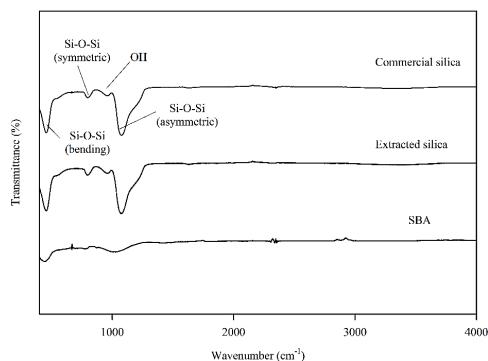


Figure 1. FTIR spectra of SBA, extracted silica, and commercial silica

Table 2. Chemical compositions of RHA and commercial silica

Compound	SBA	Extracted silica	Commercial silica
SiO_2	64.80	91.41	98.70
Cl_2	-	0.53	0.06
K_2O	15.20	3.10	0.02
Fe_2O_3	8.90	1.26	1.06
TiO_2	0.90	0.23	0.45
CaO	5.20	1.87	0.23

The raw SBA showed a sparse peak at 1,030 and 440 cm^{-1} that referred to the asymmetric stretching and symmetric bending of the Si-O-Si, respectively. This peak represented the existence of silica in the SBA.

As shown in Figure 2, the broad X-ray diffraction pattern of the commercial silica and extracted silica in the range of 15°-30° was a typical peak for amorphous solids. However, the extracted silica and SBA showed narrow peaks which represented some of metal oxide residues in the samples (Affandi *et al.*, 2009; Lee *et al.*, 2017).

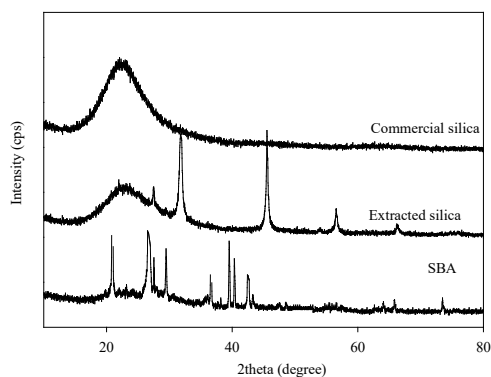


Figure 2. XRD patterns of SBA, extracted silica, and commercial silica

The particle size distribution and surface properties of the extracted silica and commercial silica are shown in Figure 3 and Table 3, respectively. The extracted silica showed a larger particle size and broader particle size distribution but lower specific surface area, pore volume, and pore area when compared to the commercial silica. This may be because the agglomeration occurred during the extraction process such as the aging and drying process. The carefully controlled aging

and drying process may lead to the formation of non-agglomeration of the silica particles (Hench and West, 1990).

The SEM images of the extracted silica and commercial silica at a magnification of 300X are shown in Figure 4. It can be seen that both of the silica had an irregular shape and that the aggregate of the extracted silica was larger than the commercial silica.

Cure Characteristics

The MH and ML of the NR and NR composites are displayed in Figure 5 and Figure 6, respectively. The MH and ML of the composites continuously increased with increasing the extracted silica content due to an increase in the stiffness of the composites and a reduction of the deformation of the NR molecules. With the presence of 20 phr of silica, the commercial silica/NR composite showed higher MH and ML when compared to the extracted silica/NR composite. The commercial silica had a smaller particle size than the extracted silica that resulted in a better filler-rubber interaction (Pongdong *et al.*, 2015). The incorporation of Si-69 into the NR

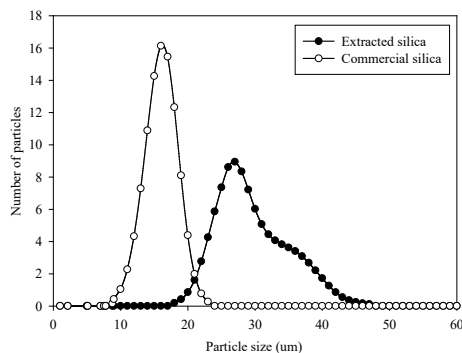


Figure 3. Particle size distribution of extracted silica and commercial silica

Table 3. Surface properties and particle size of extracted silica and commercial silica

Properties	Extracted silica	Commercial silica
BET surface area (m^2g^{-1})	52	110
Pore area (m^2g^{-1})	87.42	91.54
Pore volume (cm^3g^{-1})	0.9846	1.2242
Average particle size (μm)	26.97 ± 18.43	16.12 ± 6.37

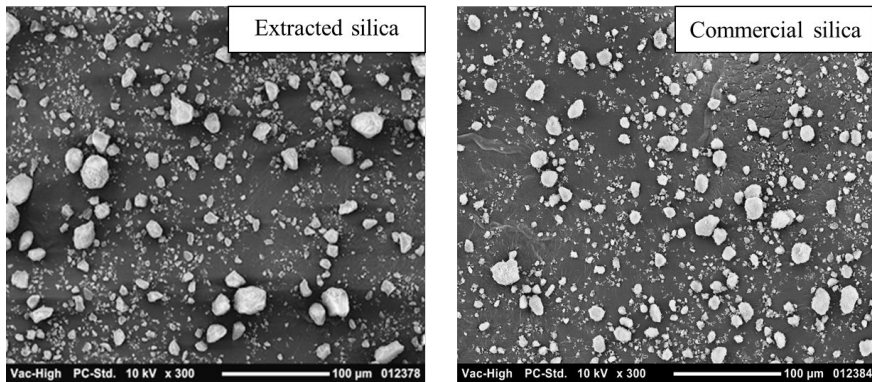


Figure 4. SEM images at 300X of extracted silica and commercial silica

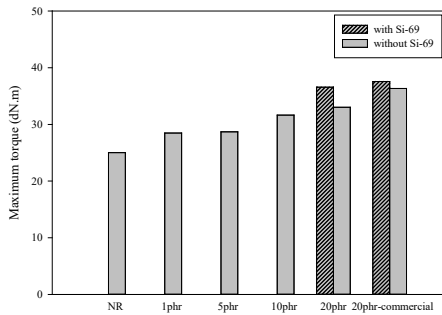


Figure 5. Maximum torque of NR and NR composites

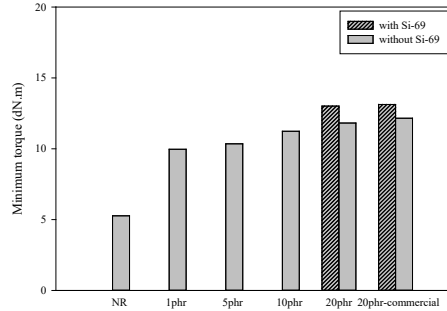


Figure 6. Minimum torque of NR and NR composites

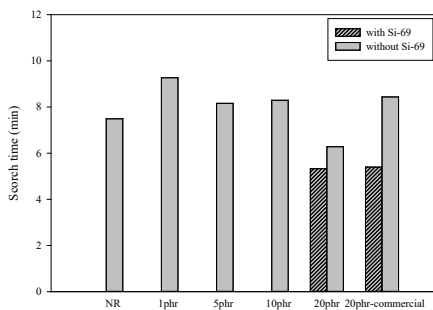


Figure 7. Scorch time of NR and NR composites

composites increased the MH and ML of the composites due to improved compatibility between the silica and NR matrix. The scorch time (t_{s1}) and cure time (t_{90}) of the NR and NR composites filled with the extracted silica and commercial silica are shown in Figure 7 and

Figure 8, respectively. The cure time of the extracted silica/NR composites increased with increasing the extracted silica content. This may be because hydroxyl groups on the silica surfaces can interact with the accelerator in the vulcanization process. Thus, the vulcanization process was retarded (Pongdong *et al.*, 2015; Mathew and Narayanankutty 2010), whereas the extracted silica content showed an insignificant effect on the scorch time of the composites. The extracted silica contained a higher content of metal impurities than the commercial silica. These metal oxide impurities can accelerate the vulcanization process (Sombatsompop *et al.*, 2004). Thus, the extracted silica/NR composites showed a shorter scorch time and cure time than the commercial silica/NR composites. Adding Si-69 led to the reduction of the scorch time

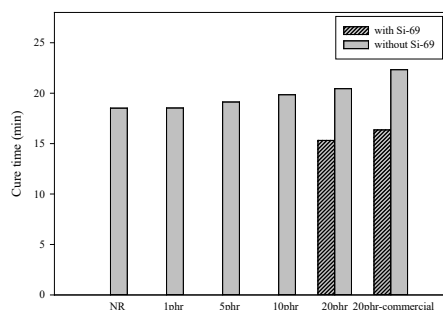


Figure 8. Cure time of NR and NR composites

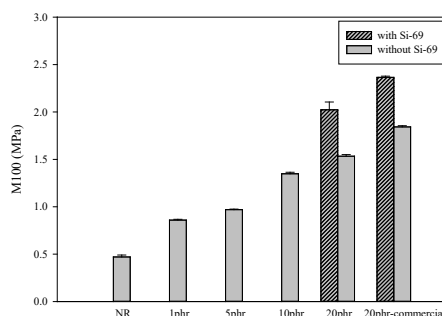


Figure 9. Modulus at 100% elongation of NR and NR composites

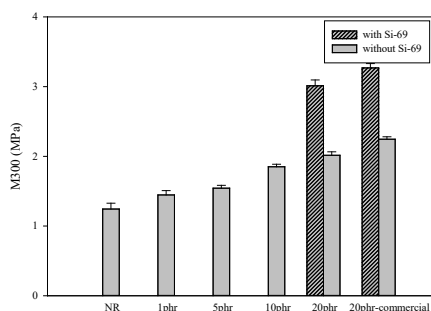


Figure 10. Modulus at 300% elongation of NR and NR composites

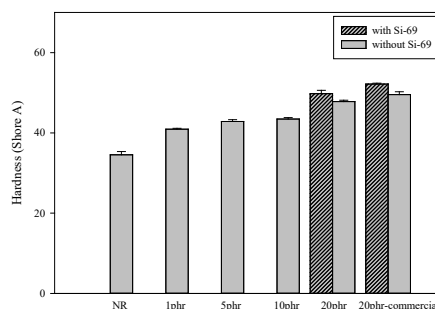


Figure 11. Hardness of NR and NR composites

and cure time of the NR composites as shown in Figure 7 and Figure 8, respectively. The ethoxy groups of Si-69 were hydrolyzed to form a hydroxyl group which underwent a condensation reaction with the silanol groups on the silica surface resulting in less adsorption of the accelerator. Thus, shorter scorch and cure times of the silica/NR/Si-69 composites were found (Li *et al.*, 2014; Katueangngan *et al.*, 2016).

Mechanical Properties

The modulus at 100% elongation (M100), modulus at 300% elongation (M300), and hardness of the NR and NR composites filled with extracted silica and commercial silica are shown in Figure 9, Figure 10, and Figure 11, respectively. The incorporation of the extracted silica increased the M100, M300, and hardness of the NR and the properties

continuously increased with the silica content due to reduced elasticity of the NR molecular chain and increased stiffness of the composites (Sarkawi and Aziz, 2003). The addition of the extracted silica into the NR resulted in a reduction of the elongation at break due to the reduction of the elasticity of the NR molecular chains as shown in Figure 12. Increasing the extracted silica content led to a slight decrease in the elongation at break of the composites. At a silica content of 20 phr, the extracted silica/NR composite showed a lower M100, M300, and hardness than the commercial silica/NR composite. This may be because the particle size of the commercial silica was smaller than the extracted silica. The smaller particle size resulted in a higher specific surface area of the silica, thus improving the filler-rubber interaction (Yin *et al.*, 2013).

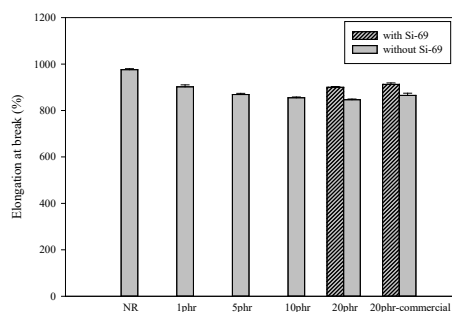


Figure 12. Elongation at break of NR and NR composites

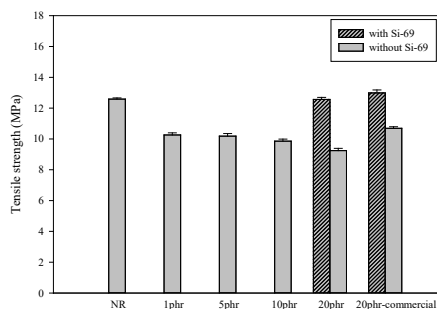


Figure 13. Tensile strength of NR and NR composites

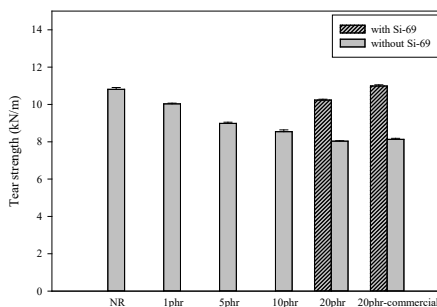


Figure 14. Tear strength of NR and NR composites

content due to the incompatibility between the silica and NR chains (Da Costa *et al.*, 2000; Sae-Oui *et al.*, 2005). The tensile strength of the extracted silica/NR composite was lower than that of the commercial silica/NR composite due to the smaller particle size and better filler-rubber interaction of the commercial silica (Prasertsri and Rattanasom, 2012). However, the tear strength of the extracted silica/NR composite was an insignificantly different when compared to that of the commercial silica/NR composite. The mechanical properties of the silica/NR composites with Si-69 were better than those of the silica/NR composites without Si-69. This was because the Si-69 enhanced the compatibility between the silica and natural rubber (Sae-oui *et al.*, 2005, Rahman and Padavettan, 2012).

Morphology

The tensile fractured surfaces of the NR, extracted silica/NR, and commercial silica/NR composites are shown in Figure 15. The NR showed a smooth fractured surface which indicated the simple rupture of the material. However, the agglomerate formation in the NR matrix was found in the NR composites due to the high filler-filler interaction. Moreover, many of the micropores were observed in all of the silica/NR composite samples. This corresponded to the decreased tensile strength and tear strength of the composites. Moreover, at a high content of silica (20 phr), larger aggregates of silica were found due to the induced filler-filler interaction. The composites with Si-69 showed less silica agglomeration and a more complex fractured surface than that of the composites without Si-69 due to their strong interaction between the silica and NR matrix (Bailly *et al.*, 2010).

Conclusions

The tensile strength and tear strength of the NR and NR composites filled with the extracted silica and commercial silica are shown in Figure 13 and Figure 14, respectively. The tensile strength and tear strength of the NR composites decreased with the extracted silica

Silica particles in the size range of $26.97 \pm 18.43 \mu\text{m}$ were prepared from SBA. The XRD pattern of the extracted silica indicated the amorphous structure. The extracted silica had a larger particle size and broader particle size distribution but lower

surface area, pore area, and pore diameter than the commercial silica. The FTIR spectrum of the extracted silica was similar to that of the commercial silica. Increasing the extracted silica content led to the increment of the ML and MH of the NR. The cure time of the

extracted silica/NR composites increased with the extracted silica content because of the disturbance of the vulcanization process by the silica surface. The extracted silica/NR composite had a shorter scorch time and cure time than the commercial silica/NR composite

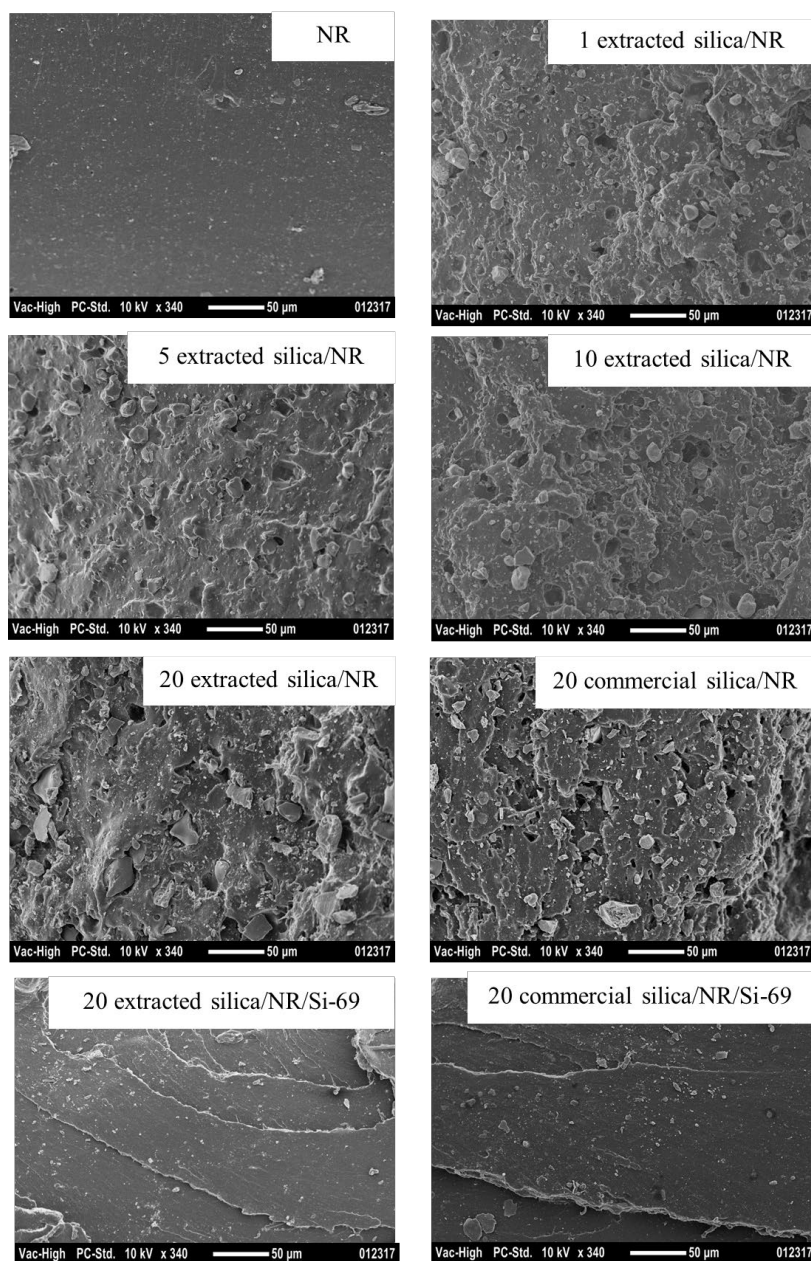


Figure 15. SEM images at 340X of NR and NR composites

because the residual metal oxides in the extracted silica accelerated the vulcanization process. With increasing the extracted silica content, the M100, M300, and hardness of the composites increased while the elongation at break reduced because the rigidity of the composites was enhanced. The reduction of the tensile strength and tear strength of the composites was observed with increasing the silica content due to poor interfacial adhesion between the silica and the matrix. The tensile strength, modulus, hardness, ML, and MH of the commercial silica/NR composite were higher than those of the extracted silica/NR composite due to the smaller particle size of the commercial silica which resulted in the better filler-rubber interaction. The addition of Si-69 improved the compatibility between the silica and NR matrix leading to the increment of the mechanical properties of the composites.

Acknowledgements

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