Processibility and Mechanical Properties of Micronized Polytetrafluoroethylene Reinforced Silicone Rubber Composites

Eun-Soo Park

Youngchang Silicone Co., Ltd., 481-7, Gasan-Dong, Kumchun-Gu, Seoul 153-803, Korea

ABSTRACT: Various micronized polytetrafluoroethylene (PTFE) powders were compounded with silicone rubber (MQ) and mechanical properties of the MQ/PTFE composites were evaluated. The fracture surface morphologies of prepared composites were also investigated using scanning electron microscopy (SEM). At a level of only 5 wt %, the fractured surface of MQ/PTFE composites show layered structure morphology. This structure effectively improves the tear strength of MQ but it also led to lower the tensile properties of the composites. The addition of fluorosilicone rubber (FMQ) as compatibilizer, tensile and tear strength of the composites improved considerably. However, tensile properties of the MQ/solution of sodium in liquid ammonia treated PTFE composite decreased compared with those of the untreated one. To investigate the production potential of extrusion process, an electric wire was extruded with MQ/PTFE/FMQ composites. During the curing process, volatile molecules lead to bubble and void formation of extruded layer depending on the filler shapes. The spherical PTFE powder was suitable for extrusion process. © 2007 Wiley Periodicals, Inc. J Appl Polym Sci 107: 372–381, 2008

Key words: composites; morphology; filler; polytetrafluoroethylenel silicone rubber

INTRODUCTION

Silicone elastomers cover a large group of materials in which vinyl-methyl-silicone is often the central ingredient. The strong silicon-oxygen chemical structure of silicone gives the elastomer its unique performance properties, including biocompatibility, superior temperature and chemical resistance, good electrical properties, and oxidation resistance.[1–12] Their resistance to moisture makes them good for outdoor applications. However, silicone rubber (MQ) and fluorosilicone rubber (FMQ) have relatively low tear strength and abrasion resistance, and therefore, are generally not suited to dynamic applications.

The improvement in tear strength of silicone elastomer is important in reducing production costs by increasing output, since complex parts can be removed from hot molds with reduced scrap rate. The use of polytetrafluoroethylene (PTFE) reinforcing additives effectively improve the tear strength of silicone elastomers.[1–5] It also decreases mold fouling and sticking, while having no adverse effect on adhesion of elastomers to metal.[2] When the high molecular weight PTFE is added as a filler, the shear developed during compounding into an elastomer fibrillates the filler into a continuous network of nodes and fibers. This network structure effectively reinforces elastomers but it also leads to distortion of finished parts and unacceptably high hardness and modulus of vulcanizates.[2] These problems limit the practical level of addition of high molecular weight PTFE powders and, therefore, tend to limit their usefulness to applications. Special compounding processes have been developed to reduce the effects of fibrillation and aid in the dispersion of high molecular weight PTFE into elastomers.[9] Techniques to modify the surface of PTFE powder to improve compatibility with hydrocarbon elastomers and reduce fibrillation have also been reported.[8]

PTFE is distinguished by its complete resistance to attack by virtually all chemicals and by its slippery surface. It maintains its physical properties over a large temperature range from −270 to 380°C. These properties make it especially useful for components that must operate under harsh chemical conditions and at temperature extremes.[6,11,12] However PTFE is not a melt processable material and consequently does not lend itself to secondary recycling. The key technology of PTFE recycling is micronization. Specialized milling is used to avoid particle edges and assure a round shape in the final particle. The result can actually vary from particles at 50 μm in mean diameter down to the more common 10 μm and less. In this study, PTFE composites had been prepared using micronized PTFE as reinforcement with MQ as matrices. The prepared specimens were subjected to

Correspondence to: E.-S. Park (t2phage@hitel.net).

thermal and mechanical tests and the fractured surfaces were observed using scanning electron microscopy (SEM). The effect of surface treatment of PTFE powder and addition of FMQ as a compatibilizer into MQ/PTFE composites were also investigated.

EXPERIMENTAL

Materials

Silicone rubber (MQ, NT6802, NanTai Hardware and Chemical Co., Ltd, China), fluorosilicone rubber (FMQ, FSE7160, Dong Yang Silicone Co., Korea) and dicumyl peroxide (DCP, Hae Ryong Silicone, Korea) were used as received. 2,4-dichlorobenzoyl peroxide (DCBP, DCLBP-50-PSI, 50% paste in silicone oil) was purchased by Akrochem Co. (OH, USA). Virgin PTFE powder [L5 (MPTFE1)] was donated by Daikin Industries Fluoroplastics. Micronized PTFE powders [SP1003 (MPTFE2), SP1040 (MPTFE3), 14946 (MPTFE4), 14948 (MPTFE5), Bogochem, Korea] were used as received. All fillers were dried in a convection oven at 60°C for 12 h to remove any moisture from the powders before compounding. The characteristics of the micronized PTFE fillers and the structure of the peroxide curing agent used in this study were shown in Table I.

Instrumentation

Thermal stability of composites was determined by Thermogravimetry (TG, Perkin–Elmer TGS-2, Norwalk, CT). The TG curves were obtained under an N_2 atmosphere at a flow rate of 4 mL min^{-1} and a scanning from 20 to 900°C with the heating rate of 20°C min^{-1}.

Scanning electron microscopy (SEM) observations of the samples were performed on a Hitachi *S*-4200 model. The fractured surfaces of the composites were prepared by using cryogenic fracturing in liquid nitrogen followed by a coating with gold in an SPI sputter coater. The morphology was determined using an accelerating voltage of 10 kV.

Hardness of the specimens was measured with a Shore A hardness tester (GS-706, Teclock Co., Japan) and the readings were averaged. Five locations were measured for each specimen and surface.

Etching of the PTFE powder

PTFE powder was immersed a solution of sodium in liquid ammonia (Na/NH$_3$, 1M) for 2 min. The etched powder were filtered, washed with hot water and acetone, and dried in a drying oven at 60°C until constant weight.
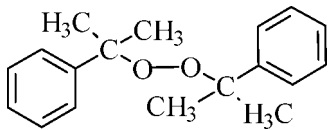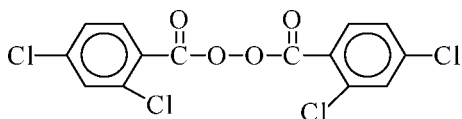
Compounding of MQ/PTFE composites

The silicone compound (95 g) and PTFE (5 g) were premixed using a two-roll mill at room temperature for 10 min followed by peroxide curing agent (3 g, for compression molding composites: DCP; for extruding composites: DCBP) was slowly added to the flux roll. The compound band was then sheeted, folded and rebanded for mixing. This process was repeated 30 times to ensure thorough mixing.

Compression molding of MQ/PTFE composites

The milled rubber sheet (180 × 180 × 2.5 mm^3) was fed into a compression mold cavity (200 × 200 × 2 mm^3).

TABLE I
Characteristics of the Micronized PTFE Powder and Structure of Peroxide Curing Agents

Properties	MPTFE1	MPTFE2	MPTFE3	MPTFE4	MPTFE5
Color	White	White	White	Off white	Off white
Particle size (μm)	5	10	40	40	10
Particle shape	Sphere	Sphere	Flake	Irregular	Sphere
Specific gravity (25°C)	2.20	2.15	2.15	2.20	2.23
Type	Virgin	Micronized	Micronized	Micronized	Micronized

Material	Name	Abbreviation	Structure		
Peroxide curing agents	Dicumyl peroxide	DCP			
	Di(2,4-dichlorobenzoyl) peroxide	DCBP			
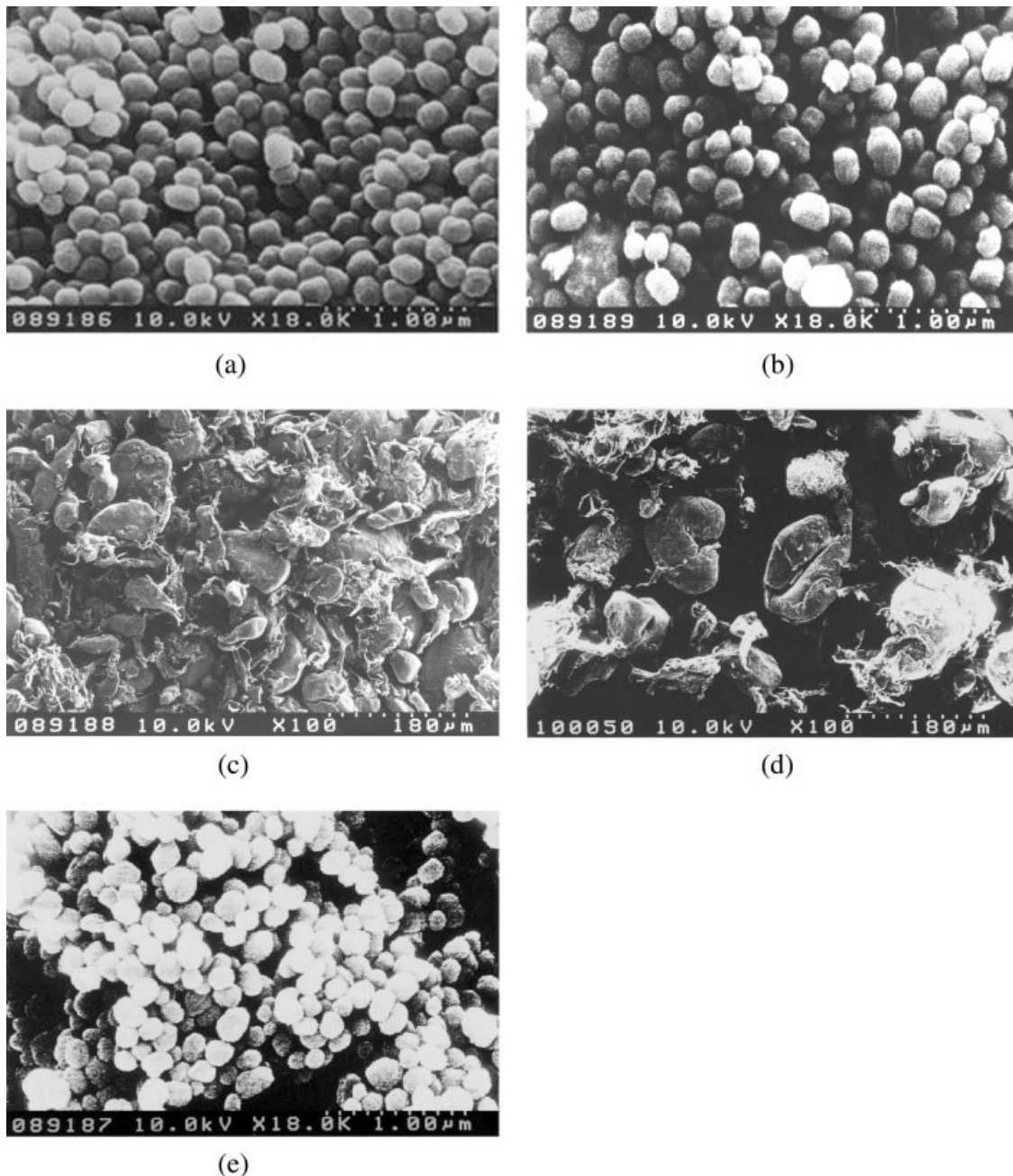
Figure 1 SEM micrographs of the micronized PTFE powder. (a) MPTFE1, (b) MPTFE2, (c) MPTFE3, (d) MPTFE4, (e) MPTFE5.

The molder was placed between two stainless-steel platens and heated in a 200°C hot press at a pressure of 10 atm for 10 min. When the curing process finished, the platens and molder were cooled to room temperature.

Extrusion of MQ/PTFE composites

The extruded electric wire was composed of tinned annealed copper conductor and outer insulation. The insulation is applied to the copper conductor by extrusion with MQ/PTFE/FMQ composites. All test specimens were extruded at a speed of 100 rpm using a single screw extruder [SH2001-100 (maximum loading capacity: 70 kg h^{-1}), Sung Heuy Industry Co., Korea] equipped with a 2.0-mm-diameter die and a 1 : 10 compression ratio screw. Extruded samples were cured by passage through a heating box at 500°C with a rate of 100 m min^{-1}.

Breakdown voltage measurement

The measurements of the breakdown voltage (BDV) were carried out at 60 Hz AC voltage by using a

TABLE II
Mechanical Properties of Compression-Molded MQ/PTFE Composites

| Sample code | MQ/PFTE composition (wt %) | Tensile properties | | Tear strength (kN m^{-1}) | Hardness (Shore A) |
		Tensile strength (MPa)	Elongation at break (%)		
MQ	100	4.5 ± 0.1	142 ± 1	15.9 ± 1.5	70
MQ/MPTFE1-05	95/05	4.9 ± 0.2	148 ± 9	17.2 ± 2.4	74
MQ/MPTFE2-05	95/05	4.9 ± 0.2	143 ± 8	17.5 ± 2.6	75
MQ/MPTFE3-05	95/05	6.6 ± 0.5	138 ± 5	18.0 ± 3.5	76
MQ/MPTFE4-05	95/05	4.3 ± 0.1	118 ± 8	20.1 ± 6.1	79
MQ/MPTFE5-05	95/05	4.8 ± 0.4	150 ± 3	17.8 ± 6.8	76
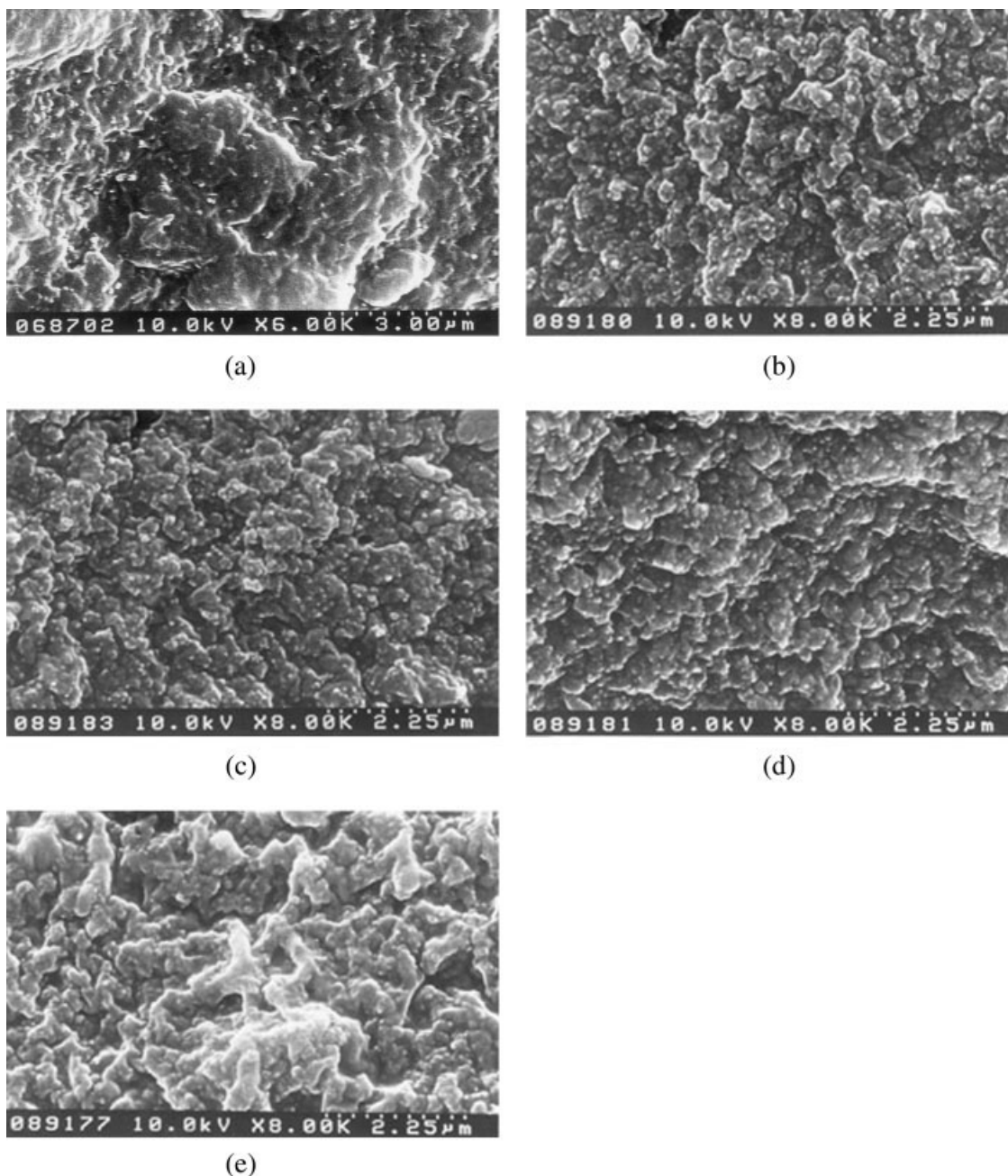
Figure 2 SEM micrographs of the compression-molded MQ/PTFE composite. The content of the PTFE powder was fixed at 5 wt %. (a) MQ, (b) MQ/MPTFE1-05, (c) MQ/MPTFE3-05, (d) MQ/MPTFE4-05, (e) MQ/MPTFE5-05.

high voltage tester (SM-40PT08, Sungmin Instruments, Korea) with a rate of 0.5 kV s^{-1} in air. The test sample consists of copper conductor (diameter of 0.6 mm) and outer insulator (insulation thickness of 1 mm). Five samples are tested and the average voltage breakdown is calculated.

Tensile and tear resistance test procedure

Dumbbell specimens for tensile tests were prepared in accordance with ASTM D412 specification. The test specimens were preconditioned to 20% relative humidity and $(20 \pm 1)°C$ to standardize test conditions. Tensile properties of samples were determined with a universal test machine (UTM, Model DECMC00, Dawha test machine, Korea) at a cross head speed of 50 mm min^{-1} at $(20 \pm 1)°C$. Die C specimens for tear resistance tests were prepared in accordance with ASTM D624. Tear strength of samples were determined with UTM at a cross head speed of 20 mm min^{-1} at $(20 \pm 1)°C$. The mean value of at least five specimens of each sample was taken, although specimens that broke in an unusual manner were disregarded.

RESULTS AND DISCUSSION

Figure 1 shows the SEM image of PTFE powders. MPTFE1, MPTFE2, and MPTFE5 were of spherical shaped particles with average particle sizes of 5 μm, 10 μm, and 5 μm, respectively. MPTFE3 with platy particles had a diameter of 40 μm. MPTFE4 was irregular shape with ∼ 40 μm in length.

MQ, peroxide curing agent and PTFE powder were mixed using a two-roll mill at room temperature and compression-molded in a hot press at 200°C for 10 min. Free radical cure systems employ peroxides that are either vinyl specific or nonspecific in nature. Nonspecific peroxides such as DCBP and

benzoyl peroxide do not require the presence of vinyl or other unsaturated alkyl species in the polymers making up the elastomer formulations. They are commonly used in extrusion process. DCP is vinyl specific initiator and requires the presence of vinyl or other alkenyl species in the polymers contained in the elastomers. This vinyl specific initiator does not perform well in extrusion applications, and is thus limited to use in molding. Organic peroxides undergo homolytic cleavage at elevated temperature. The resulting free radicals induce hydrogen abstraction of hydrogen atoms from the polymer backbone. These in turn create free radicals on the polymer backbone, which allow the chains to form carbon–carbon crosslinks with one another.

To gain some basic knowledge of the MQ/PTFE system, the mechanical behavior was examined by using compression-molded composites with 5 wt % PTFE filler (Table II). The abbreviation of the sample code in Table II, MQ/MPTFE3-05, for example, means that the content of MPTFE3 in the MQ was 5 wt %. Mechanical property measurements show that incorporation of MPTFE1, MPTFE2, MPTFE3, and MPTFE5 slightly increased both the tensile strength and the elongation at break, whereas for irregular shaped MPTFE4 led to lower the elongation at break of composites significantly. Irregular shaped particles occupy larger effective volume than particles with spherical shape. As a result the composites with irregular shaped fillers have lower elongation at break compared to the composite with spherical fillers. It can be also seen that tear strength and hardness of all composites increased significantly. Figure 2 represents SEM images of fractured surfaces of compression-molded MQ/PTFE composites. It can be seen that at a level of only 5 wt %, all composites show the layered structure morphology without regard to filler size or shape. Morgan et al.[2] reported that when a high molecular weight PTFE is mill

TABLE III
Mechanical Properties of Compression-Molded MQ/PTFE Composites

		Tensile properties			
Sample code	MQ/PFTE composition (wt %)	Tensile strength (MPa)	Elongation at break (%)	Tear strength (kN m^{-1})	Hardness (Shore A)
MQ/MPTFE1-05	95/05	4.9 ± 0.2	148 ± 9	17.2 ± 2.4	74
MQ/MPTFE1-10	90/10	4.5 ± 0.2	130 ± 8	18.6 ± 5.3	75
MQ/MPTFE1-15	85/15	4.0 ± 0.1	124 ± 7	19.7 ± 5.2	76
MQ/MPTFE1-30	70/30	3.3 ± 0.2	115 ± 6	22.8 ± 8.3	78
MQ/MPTFE3-05	95/05	6.6 ± 0.5	138 ± 5	18.0 ± 3.5	76
MQ/MPTFE3-10	90/10	6.5 ± 0.2	113 ± 2	20.5 ± 8.1	77
MQ/MPTFE3-15	85/15	6.2 ± 0.1	96 ± 1	21.0 ± 3.8	78
MQ/MPTFE3-30	70/30	5.9 ± 0.7	80 ± 1	18.8 ± 1.0	80
MQ/MPTFE5-05	95/05	4.8 ± 0.4	150 ± 3	17.8 ± 6.8	76
MQ/MPTFE5-10	90/10	4.6 ± 0.3	143 ± 7	20.6 ± 4.2	78
MQ/MPTFE5-15	85/15	4.2 ± 0.5	135 ± 8	21.7 ± 5.5	79
MQ/MPTFE5-30	70/30	3.8 ± 0.6	125 ± 5	23.2 ± 7.2	82
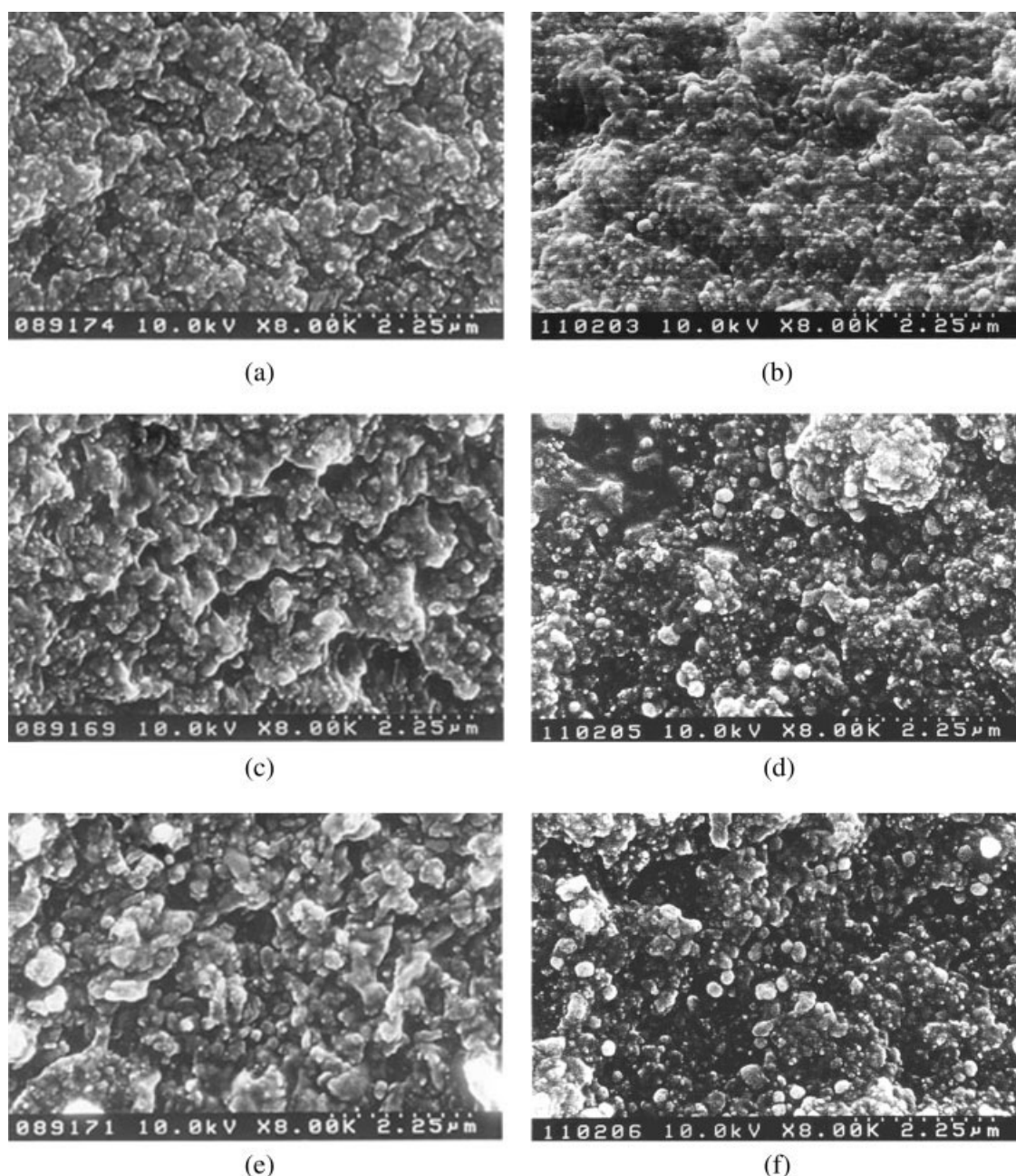
Figure 3 SEM micrographs of the compression-molded MQ/PTFE/FMQ composite. (a) MQ/MPTFE1-10, (b) MQ/MPTFE1-10/FMQ-05, (c) MQ/MPTFE1-15, (d) MQ/MPTFE1-15/FMQ-05, (e) MQ/MPTFE1-30, (f) MQ/MPTFE1-30/FMQ-05.

mixed into MQ, even at very low levels, an anisotropic fibrillated layered structure is obtained. This network structure effectively reinforces elastomers, but it also leads to unacceptably high hardness and modulus of composites.

Table III demonstrates the effect of PTFE concentration on the mechanical properties of composites. As the concentrations of MPTFE1, MPTFE3, and MPTFE5 were increased, both the tensile strength and the elongation at break were decreased. The tensile properties of MQ/PTFE composites decreased with the presence of filler that indicates an interfer-

ence[13] by the filler in the mobility[14] or deformability of the matrix. An increase in weight percentage of filler reduced the deformability of the matrix, and, in turn, reducing the ductility in the skin area so that the composite tended to form a weak structure. On the other hand, all composites show a significant increment in tear strength without regard for its tensile strength value. An $\sim$ 8% overall increase in the tear strength from 17.2 to 23.2 kN m^{-1} was observed addition of PTFE. This indicated that the layered structure more effectively reinforces lateral direction of compression-molded MQ/PTFE composite than

TABLE IV
Mechanical Properties of Compression-Molded MQ/PTFE/FMQ and MQ/Etched PTFE Composites

| Sample code | MQ/PTFE/FMQ blend ratio[a] (phr, 100 g/g) | Tensile properties | | Tear strength (kN m^{-1}) | Hardness (Shore A) |
		Tensile strength (MPa)	Elongation at break (%)		
MQ/MPTFE1-10/FMQ-01	90/10/1	4.8 ± 0.3	153 ± 8	18.6 ± 0.4	75
MQ/MPTFE1-10/FMQ-05	90/10/5	4.6 ± 0.1	147 ± 13	21.1 ± 0.4	74
MQ/MPTFE1-10/FMQ-10	90/10/10	4.6 ± 0.4	152 ± 1	21.8 ± 0.6	73
MQ/MPTFE1-10/FMQ-25	90/10/25	4.7 ± 0.1	126 ± 1	18.8 ± 1.4	73
MQ/MPTFE1-15/FMQ-05	85/15/5	4.3 ± 0.3	126 ± 10	22.5 ± 4.3	75
MQ/MPTFE3-10/FMQ-05	90/10/5	6.7 ± 0.1	114 ± 8	22.8 ± 0.8	73
MQ/MPTFE3-15/FMQ-05	85/15/5	6.3 ± 0.1	104 ± 21	24.4 ± 1.0	74
MQ/MPTFE5-10/FMQ-05	90/10/5	4.8 ± 0.4	150 ± 12	23.0 ± 0.8	76
MQ/MPTFE5-15/FMQ-05	85/15/5	4.6 ± 0.2	142 ± 10	23.5 ± 1.0	77
MQ/MPTFE1-10E[b]	90/10/0	3.4 ± 0.4	92 ± 4	9.9 ± 2.5	73
MQ/MPTFE1-15E[b]	85/15/0	2.3 ± 0.4	76 ± 4	10.2 ± 3.0	84
MQ/MPTFE5-10E[b]	90/10/0	4.5 ± 0.5	135 ± 10	19.7 ± 2.5	78
MQ/MPTFE5-15E[b]	85/15/0	4.0 ± 0.6	130 ± 4	20.2 ± 3.0	88

[a] 100 g of MQ/PTFE composites/g of FMQ.
[b] MQ/solution of sodium in liquid ammonia treated PTFE composites.

compressed direction. When a compressive force is applied to the mold, the milled rubber compound in mold cavity is compressed along thickness direction and expands along the lateral direction so that PTFE networks are produced along the rubber flow. An increase in filler content enhances the layered structures along the lateral direction (Fig. 3) and significantly increase the force required to make an initial tear.

Table IV indicates tensile and tear properties of compression-molded MQ/PTFE/FMQ and MQ/solution of Na/NH$_3$ treated PTFE composites. Remark-

able increases in tear strength are obtained when FMQ is compounded into MQ/PTFE composites. The composites also show significant increases in tensile strength and elongation at break with a slight increase in hardness. FMQ acts as a compatibilizer, lowering the particle-to-particle interaction of PTFE and promoting interfacial adhesion between the dispersed phase and the polymer matrix. The increase of FMQ contents above 10 phr did not lead to a further improvement. Figure 3 represents the morphology of the compression-molded MQ/PTFE and MQ/PTFE/FMQ composites. When 5 phr of FMQ added in MQ/PTFE composite, a layered structure has disappeared and PTFE filler becomes more uniformly dispersed in the matrix. SEM observations confirmed, after the addition of FMQ, improvement of the interfacial adhesion between the dispersed phase and the polymer matrix.

In contrast, the tensile properties and tear strength of the MQ/solution of Na/NH$_3$ treated PTFE com-
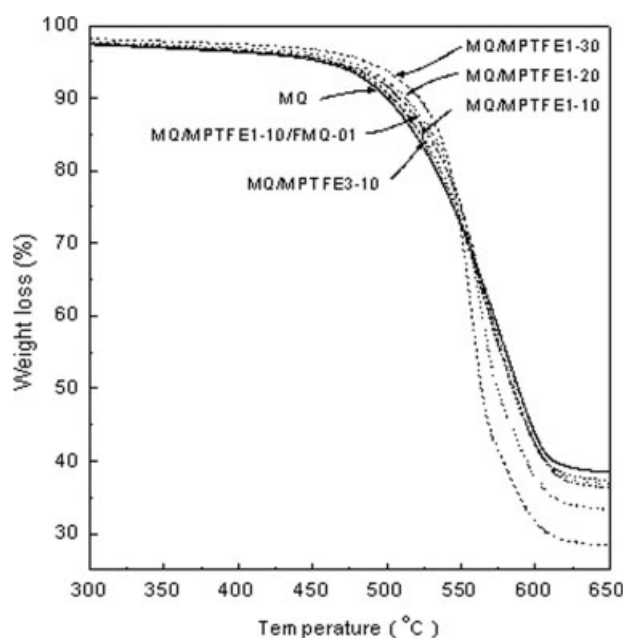

Figure 4 Thermogravimetry analysis (TGA) of the compression-molded MQ/PTFE composites.

TABLE V
Breakdown Strength of the Extruded Samples

| Sample | Breakdown strength (kV/mm) | |
	Before thermal ageing	After thermal ageing[a]
MQ	16.5	14.5
MQ/MPTFE1-10/FMQ-05	17.2	15.7
MQ/MPTFE3-10/FMQ-05	8.9	5.4
MQ/MPTFE4-10/FMQ-05	6.7	4.7
MQ/MPTFE5-10/FMQ-05	16.7	14.9

[a] Ageing temperature: 200°C for 30 min.

(a) MQ

(b) MQ/MPTFE1-10/FMQ-05

(c) MQ/MPTFE3-10/FMQ-05
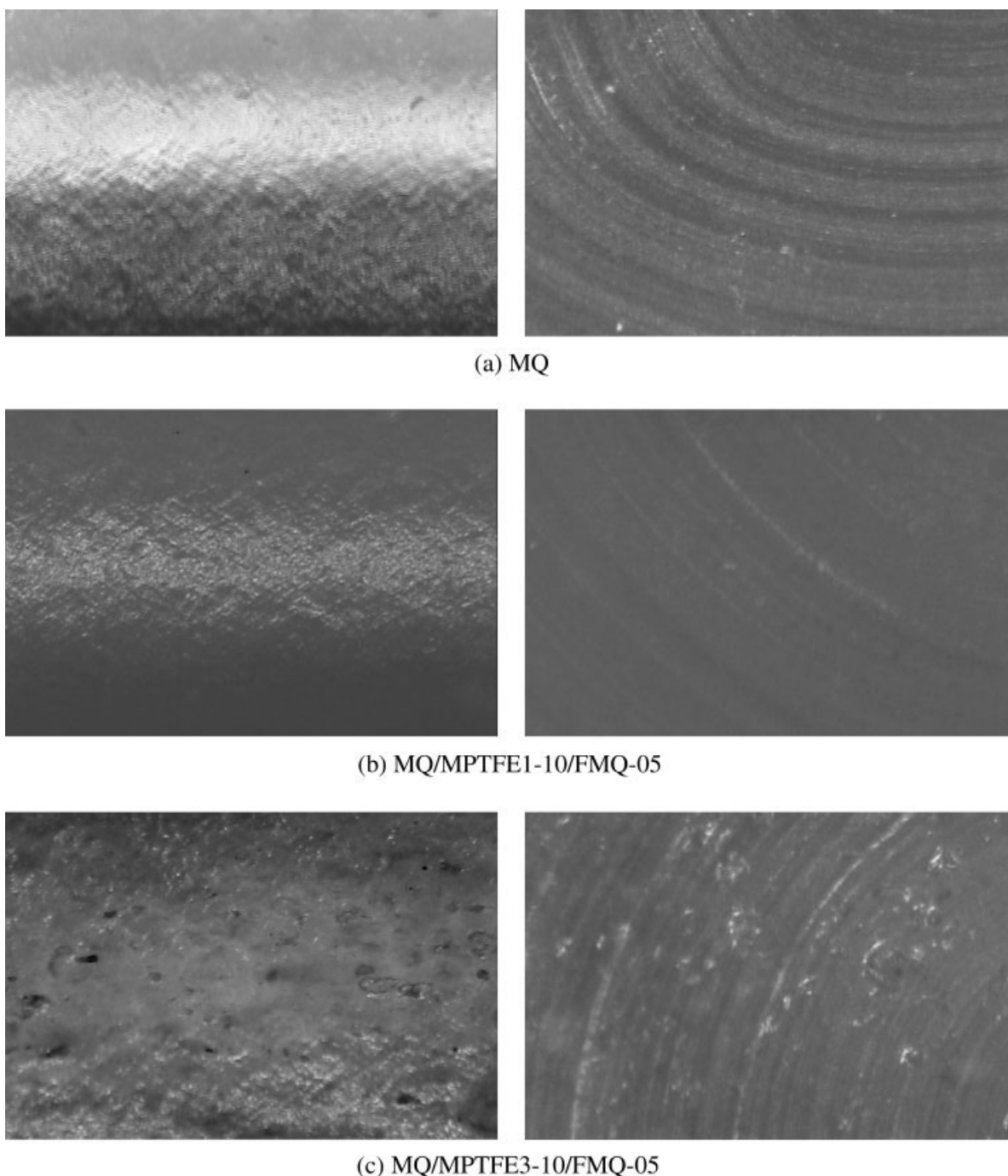
Figure 5 Optical photography of the extruded MQ/PTFE/FMQ composite (×300).

posite decreased compared with those of the untreated one. Surface etching is a chemical process of surface treatment to material to allow for adhering to another surface. When the fluorine atom is removed from PTFE surface by the etching process, the electron shortage is made up by oxygen, hydrogen, and water vapor. This forms polar functional groups, such as those of the hydroxyl group, carboxyl group, or carbonyl group. In practice, it is difficult to obtain uniform mixtures of MQ and organic

polymer having polar groups through milling because of the poor compatibility between the two components. The surface etched PTFE exhibit a slightly more polar characteristic and lower the compatibility of the silicone moieties with the hydrophobic domain.

Figure 4 shows the TG results of compression-molded MQ/PTFE and MQ/PTFE/FMQ composites. The virgin MQ shows an initial degradation temperature at 317°C which is the lowest of all investigated
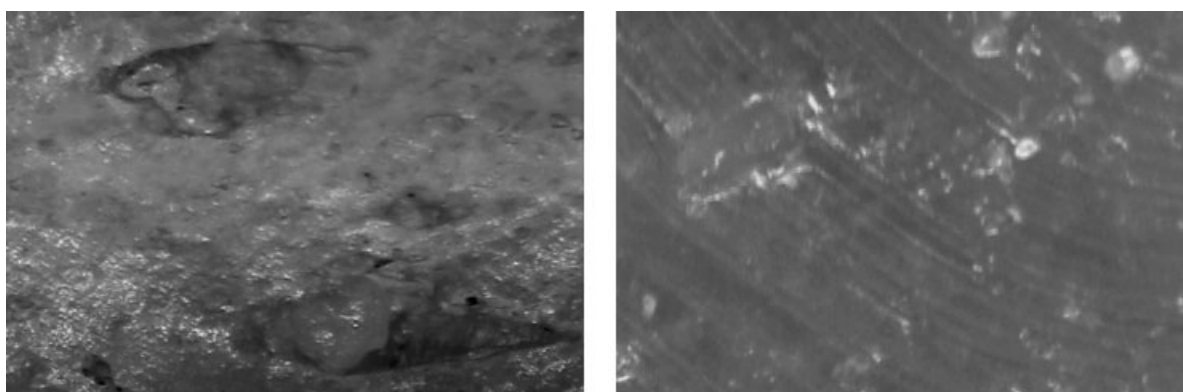
(d) MQ/MPTFE4-10/FMQ-05
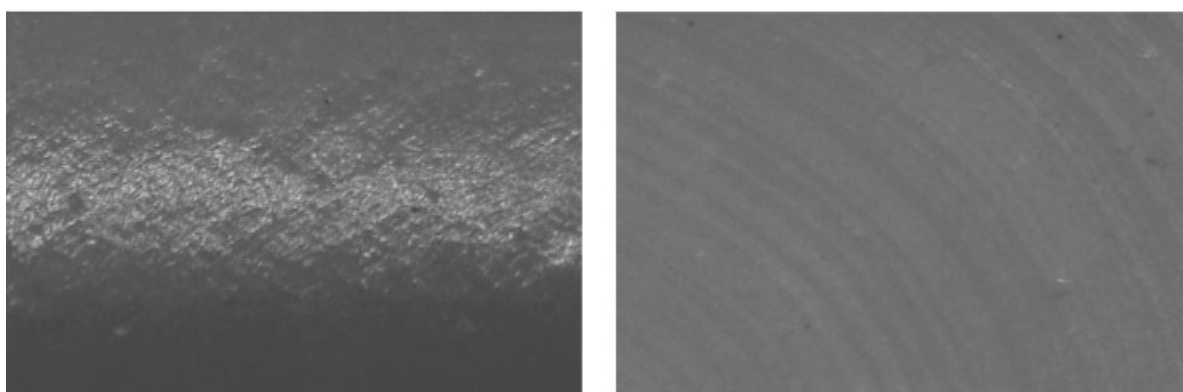
(e) MQ/MPTFE5-10/FMQ-05

Figure 5 (*Continued from the previous page*).

samples. It can be due to lower bond energy of Si—C bonds (Si—C bond dissociation energy 306 kJ mol^{-1}) in MQ compare to bond energy of C—C (346 kJ mol^{-1}) in other polymers.[10] When the PTFE content increased, TG traces show a significant shift of the weight loss towards higher temperature for MQ/PFTE composites than unfilled MQ. The high bond dissociation energy of the C—F bond (413 kJ mol^{-1}) in PTFE[11] is the main reason for increasing the thermal stability of MQ/PTFE composites. The addition of FMQ into MQ/PTFE composite resulted in a slight increase in thermal stability. This improvement is likely associated with the interaction increase between SR matrix and PTFE.

To investigate the production potential of extrusion processing method, MQ/PTFE/FMQ composites were extruded on the conductor and cured at heating rate of 100 m min^{-1} at 500°C. Table V shows dielectric breakdown strength of extruded samples before and after thermal aging at 200°C for 24 h. The dielectric strength of spherical PTFE filled composites is higher than other samples, but it has also the highest breakdown strength after thermal aging.

This implies that the size and shape of fillers significantly affect the breakdown strength of the composites.

Figure 5 represents microphotography of side surface and cross section surface of extruded MQ/PTFE/FMQ composites. MQ/MPTFE3-10/FMQ-05 and MQ/MPTFE4-10/FMQ-05 composite is covered with small bubbles and agglomerates that leave a bumpy texture on the surface. During the curing, volatile molecules should be produced and this can lead to gas bubble and void formation depending on the filler shapes. The presence of gas bubbles on the surface of insulation significantly reduces the breakdown voltage. This is due to a combination of two factors: (a) air has lower breakdown strength than polymers; and (b) presence of a void creates local electric field strength irregularities, with the lower permittivity of air causing an intensification of the electric field at the solid/void interface. Consequently, breakdown preferentially occurs first at a solid/void interface has the consequence of disrupting the polymer structure and increasing the void size.[15]

CONCLUSIONS

Micronized PTFE powders were compounded with MQ using a two roll mill at room temperature and the composites were compress-molded at 200°C. At a level of only 5 wt % of micronized PTFE powder made a continuous fiber network during compounding. This network structure effectively reinforces the MQ/PTFE composites but it also leads to unacceptably high brittleness of composites. The addition of FMQ, tensile and tear properties of the composites improved considerably. FMQ acts as a compatibilizer, lowering the particle-to-particle interaction of PTFE and promoting interfacial adhesion between the dispersed phase and the polymer matrix. In contrast, the tensile properties and tear strength of solution of Na/NH_3 treated PTFE composite decreased compared with those of the untreated one. This indicated that the surface etching process enhance the polar characteristics of the PTFE surface and lower the compatibility between the two components.

To investigate the production potential of extrusion processing method, MQ/PTFE/FMQ composites were extruded an electric wire. During the peroxide curing, volatile molecules lead to bubble and void formation of extruded layer except for spherical PTFE powder filled composites. The dielectric strength of spherical powder filled composites is also higher than other samples. It can be said that spherical PTFE powder leads to a better electrical behavior of the employed MQ/PTFE composites than flake or irregular powder filled composites.

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