Effect of Filler on the Mechanical, Dynamic Mechanical, and Aging Properties of Binary and Ternary Blends of Acrylic Rubber, Fluorocarbon Rubber, and Polyacrylate

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ABSTRACT: We studied the effects of fillers on the mechanical, dynamic mechanical, and aging properties of rubber–plastic binary and ternary blends derived from acrylic rubber, fluorocarbon rubber, and multifunctional acrylates. The addition of fillers, such as carbon black and silica, changed the nature of the stress–deformation behavior with a higher stress level for a given strain. The tensile and tear strengths increased with the addition of the fillers and with loading, but the elongation at break decreased, and the tension set remained unaffected. The aging properties of carbon-black-filled blends were better because of the thermal antioxidant nature of carbon black. The swelling resistance of the binary and the ternary blends in methyl ethyl ketone increased with the incorporation of fillers. From dynamic mechanical thermal analysis, we concluded that the filler altered the height and half-width of the damping peak at the glass-transition temperatures. There was little change in the loss tangent values at higher temperatures. A higher loading of the filler increased the storage modulus at all of the temperatures measured. © 2003 Wiley Periodicals, Inc. J Appl Polym Sci 90: 278–286, 2003

Key words: fluoropolymers; blends; fillers; mechanical properties; ageing

INTRODUCTION

Polymer blends offer versatile industrial applications through property enhancement and economic benefits. The blending of two or more polymers of similar or dissimilar natures has been practiced for many years.[1] Rubber–rubber blends were reviewed by Corish and Powel[2] and Roland.[3] Among many the rubber–rubber blends, only few are miscible because of different physicochemical parameters, including the high-molecular-weight nature of the rubbers.[4] Elastomeric rubber–plastic blends have become technologically important as thermoplastic elastomers because of their superiority in processing, their properties, and their lower cost of production over conventional rubbers.[5,6] The versatility in the adjustment of any property by a mere change in the blend ratio, the incorporation of an additive, and the processing conditions make this class of materials more cost-effective. A large number of rubber–plastic blends, as listed in the literature, have been obtained from blending polyolefins with many diene rubbers.[7–9] The blending of a polar rubber with polar plastics, such as nitrile rubber (NBR)–nylon,[10] poly(vinyl chloride)–NBR,[9] and chlorinated polyethylene–acrylic rubber (ACM),[11] to achieve specific properties, such as oil, fuel, and solvent resistance and high-temperature resistance, has also been reported in the literature. Jha and Bhowmick described novel heat- and oil-resistant rubber–plastic blends from ACM and nylon and the influence of filler and plasticizer on the performance of such plastic–rubber blends.[12,13] Recently, we reported on the development and properties of binary and ternary blends from ACM, fluorocarbon rubber (FKM), and acrylate plastics.[14] The morphology, rheological properties, thermal aging, and swelling behavior of these blends were also discussed.[15–17] In this investigation, the effects of filler on the mechanical and dynamic mechanical thermal properties of these blends were studied. The samples after heat aging were also investigated.

Polymers are rarely used in their pristine form in many applications. They are often mixed with fillers to improve their processability and mechanical strength and to reduce costs. For this purpose, many rubber–rubber blends are filled with fillers, such as carbon black, silica, and clay. The addition of reinforcing fillers is essential for the many synthetic rubbers whose strength properties are poor. The reinforcement of rubbers has been reviewed in the literature.[18] ACM shows poor gum strength because of the presence of a polar group in the pendant position. The rheological and mechanical properties of filled ACM/FKM blends

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show improvement in both strength and processability.[19] However, the inclusion of a filler in a rubber–plastic blend may sometimes lead to a reduction in key properties because of phase-separated microstructures. Moreover the hard plastic phase acts like a filler under normal usage. Studies have shown that the addition of a small amount of clay to a nylon–NBR blend and of carbon black to an ethylene–propylene–diene rubber/polypropylene blend has little effect on stiffness and strength.[10] However, in this study, we investigated the effect of fillers on mechanical properties and dynamic mechanical thermal properties for the following reasons:

- The strength of ACM can greatly be improved by the addition of carbon black.
- The oil and fuel resistance of ACM and FKM can be increased.

EXPERIMENTAL

Materials

ACM (Nipol AR 51), derived from the polymerization of ethyl acrylate with a small percentage of a proprietary epoxy cure site monomer [density = 1100 kg m^{-3} at 25°C, Mooney viscosity ML(1 + 4) at 100°C = 55] was obtained from Nippon Zeon Co., Ltd. (Tokyo). FKM (Viton B 50), a terpolymer of vinylidene fluoride, hexafluoropropylene, and tetrafluoroethylene [density = 1850 kg m^{-3}, Mooney viscosity ML(1+10) at 121°C = 56, 68% F, 1.4% H], was supplied by DuPont Dow Elastomers (Freeport, TX). The acrylate monomers, (1) p-hexanediol diacrylate (p-HDDA; density = 1050 kg m^{-3}) and (2) p-trimethylolpropane triacrylate (p-TMPTA; density = 1110 kg m^{-3}), were procured from UCB Chemicals (Brussels, Belgium). The molecular structures of ACM, FKM, and the multifunctional acrylates were given in our earlier publication.[14] Carbon black (fine extrusion furnace (FEF), N550) was supplied by Phillips Carbon Black, Ltd. (Durgapur, India). Silica (Ultrasil VN$_3$; particle size = 20–100 nm) was provided by Bayer, Ltd. (Mumbai, India).

Preparation of the samples

ACM and FKM were mixed along with the required polyfunctional acrylate monomer in a Brabender plasticorder (PLE 330) (Akron, OH) at 100°C at a rotor speed of 60 rpm. The compositions of the blends are given in Table I. The mixing sequence follows. FKM was first charged into the Brabender plasticorder followed by ACM. The blend was mixed for 5 min. The liquid monomer containing 0.1% benzoyl peroxide (the polymerization initiator) was then added slowly into the mixer at the same temperature, and mixing

was continued until the mass became homogeneous after about 3 min. The required amount of the filler was then added, and mixing was continued for another 2 min. After mixing, the mass was sheeted out at 30°C with a laboratory two-roll mill. (6 × 13 in., Schwabenthan, Berlin). The samples were then molded at 170°C and at a pressure of 5 MPa between Teflon sheets in a single daylight two-plate hydraulic compression press (Moore Press, UK) provided with cooling facility. The samples were cooled in the press itself at 50°C for 5 min.

Testing

Mechanical tests

Tensile and tear specimens were punched out from the molded sheet with an ASTM Die-C and an ASTM angle-tear specimen die, respectively. Mechanical tests were carried out as per ASTM D 412-99 in a universal testing machine (UTM-ZWICK-1445, Ulm, Germany) at a crosshead speed of 500 mm/min at 25°C. The average values of three tests for each sample are reported here.

Dynamic mechanical thermal analysis (DMTA)

Dynamic mechanical thermal properties were evaluated on a Rheometric Scientific DMTA IV (New Jersey) under tension mode. The experiments were carried out at a frequency of 1 Hz. The measurements were taken from −50 to 100°C at a heating rate of 2°C min^{-1}. The storage modulus (E') and loss tangent (tan δ) were measured for all of the samples under identical conditions. The data were analyzed with RSI Orchestrator application software (New Jersey) on an Acer computer attached to the machine

Aging studies

The aging of the samples was performed in an air-circulating oven operated at 150°C for 36 h. Tensile testing (ASTM D 412-99) was carried out on tensile dumbbells before and after aging to estimate aging resistance.

Swelling

Circular test pieces about 20 mm in radius were die-cut from a molded sheet 1.5–2.0 mm thick. The accurately weighed samples were immersed in solvents with different solubility parameters at 30°C for 7 days, and the swollen samples were weighed accurately and quickly in a glass-stoppered bottle after blotting with filter paper. The percentage volume swelling was calculated with the following equation:

TABLE 1
Physical Properties of the Filled Rubber–Plastic Blends Before and After Aging

|	$BF2_{30}$	$AFT30$	$AT30F_{30}$	AFT $(70/30)F_{30}$	$AFT30F_{30}$	AFT $(30/70)F_{30}$	$FT30F_{30}$	$AFT30F_{10}$	$AFT30F_{50}$	$AFH30F_{30}$	$AFT30S_{30}$
Composition (w/w)											
ACM	50	50	100	70	50	30	0	50	50	50	50
FKM	50	50	—	30	50	70	100	50	50	50	50
HDDA	—	—	—	—	—	—	—	—	—	30	—
TMPTA	—	30	30	30	30	30	30	30	30	—	30
FEF (N550)	—	—	30	30	30	30	30	10	50	30	—
Silica	—	—	—	—	—	—	—	—	—	—	30
Physical properties											
100% modulus (MPa)	1.4	7.8	5.5	6.5	10.9	—	4.8	8.4	—	9.2	9.6
Tensile strength (MPa)	2.3	9.2	9.4	7.7	12.0	7.8	5.2	9.7	12.8	11.5	10.8
Elongation at break (%)	1600	138	378	113	107	75	106	127	91	137	113
Tear strength (N/mm)	17.5	56	47	45	58	42	43	56	58	46	52
Tension set (%)	—	8	8	8	8	8	22	8	10	16	8
Swelling in MEK (%)	Soluble	134	145	—	118	—	66	128	104	128	127
Physical properties after thermal aging at 150°C for 36 h											
Tensile strength (MPa, % change)	—	9.1 (−1.0)	5.2 (−44.7)	5.1 (−33.3)	11.4 (−5.0)	7.1 (−9.0)	4.8 (−7.7)	9.2 (−5.2)	12.1 (−5.5)	11.1 (−3.5)	10.4 (−3.7)
Elongation at break (% change)	—	96 (−30.4)	81 (−78.6)	78 (−31.0)	84 (−21.5)	66 (−12.0)	99 (−6.4)	92 (−27.6)	64 (−29.7)	103 (−24.9)	84 (−25.7)
Tear strength (N/mm, % change)	—	43 (−23.2)	28 (−40.4)	31 (−31.2)	44 (−24.2)	36 (−14.2)	37 (−14.0)	42 (−25.0)	45 (−22.4)	35 (−24)	39 (−25.0)
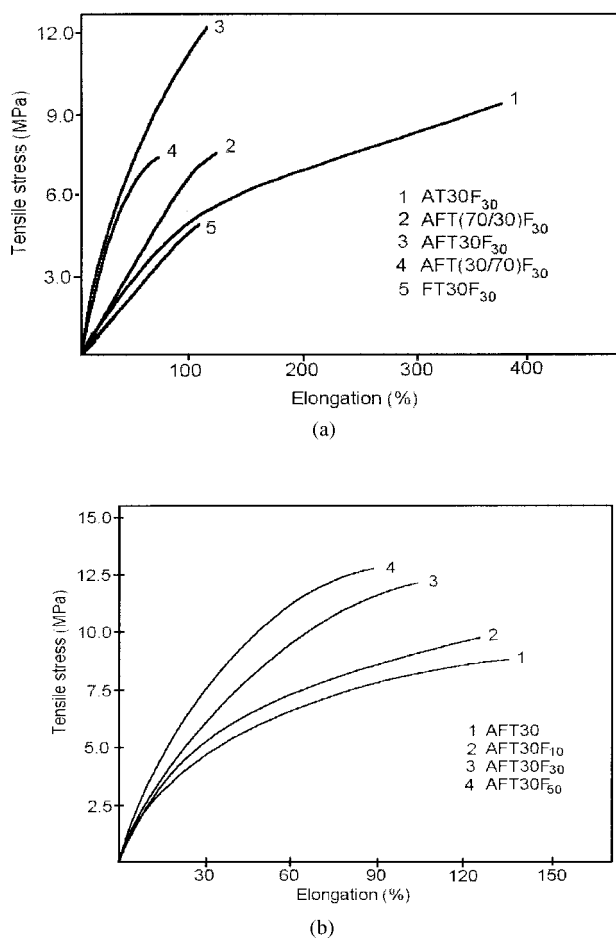
Figure 1 Stress–strain curves of (a) 30-phr N550-filled 50/50 (w/w) ACM/FKM/polyacrylate blends containing different proportions of ACM and FKM and (b) 50/50/30 (w/w) ACM/FKM/polyacrylate blends filled with different levels of carbon black.

$$q - 1 = \left(\frac{W_2}{W_1} - 1 \right) \frac{\rho_b}{\rho_s} \qquad (1)$$

where q is the ratio of the swollen volume to the original volume, $(q - 1)$ is the percentage volume swell divided by 100; W_1 and W_2 are the weights of the specimen before and after swelling, respectively; and ρ_b and ρ_s are the densities of the specimen and the test solvent, respectively. The densities of the polymers and solvent were taken from the standard book for the calculation of the volume swell.

RESULTS AND DISCUSSION

Stress–strain properties

The stress–strain curves of representative samples of 30-phr N550-filled ACM/FKM/p-TMPTA blends with varying compositions are represented in Figure 1. The composition was varied only for ACM and FKM at a constant level (30 phr) of p-TMPTA. By increasing the level of ACM in the blend, the modulus and the stress at break increased until the composition, AFT30F$_{30}$, was reached. This indicates that the compatibility increased with increasing ACM in the blend. Beyond 50 phr of FKM in the blend, the stress at break decreased because of the incompatible nature with p-TMPTA. FT30F$_{30}$ showed the lowest values of stress level and modulus at 100% strain level. When the stress–strain properties of the unfilled and the filled blends of the same composition were compared, the blends containing higher proportions of ACM showed considerable improvement in stress–strain properties on incorporation of carbon black. The effect of carbon black on the blends containing higher proportions of FKM was minimal. This indicates a moderate reinforcing effect of carbon black with FKM, possibly because of the presence of the symmetric polytetrafluoroethylene in FKM.

The stress–strain behavior of filled 50/50/30 (w/w) ACM/FKM/p-TMPTA [Fig. 1(b)] showed the reinforcing nature of carbon black at higher loading. The addition of a small quantity (10 phr) of N550 did not alter the stress–strain behavior of AFT30 considerably. When the level was increased, there were increases in modulus and stress at break with a reduction in extensibility. This effect was very similar to that observed for the filled elastomer. However, at a still higher loading of the filler, it was expected that there would be a dilution effect with the formation of filler aggregates through clustering, which would generate a weaker link between the filler phase and polymer chain, leading to a further reduction in extensibility.[20] The effect of the addition of silica was similar to carbon black, but the changes in the stress–strain properties were lesser in magnitude (AFT30S$_{30}$ and AFT30F$_{30}$ in Table I). All of these observations were in line with the results reported by Coran and Patel for a NBR–nylon blend.[10] We postulate that the filler remained in the rubber phase and enhanced the stiffness of the system, which is clarified more in the later discussion on dynamic mechanical transition.

The stress–strain behavior of the filled ternary blend was also influenced by the nature of the multifunctional acrylate. The binary blend of 30-phr N550-filled ACM/FKM (BF2$_{30}$) without polyacrylate showed a high level of elongation at a relatively low level of stress. The addition of polyacrylate to this blend caused a dramatic change in both stress level and extensibility. As the functionality of polyacrylate was 2, the stress at break increased from 2.3 to 11.5 MPa with a great loss in extensibility from greater than 1600% to 137%. This extensibility was further reduced when the functionality of polyacrylate was increased from 2 to 3. The polyfunctional acrylate restricted the mobility of the polymer chains by undergoing grafting and partial crosslinking, which led to an increased modulus and reduced elongation. The addition of
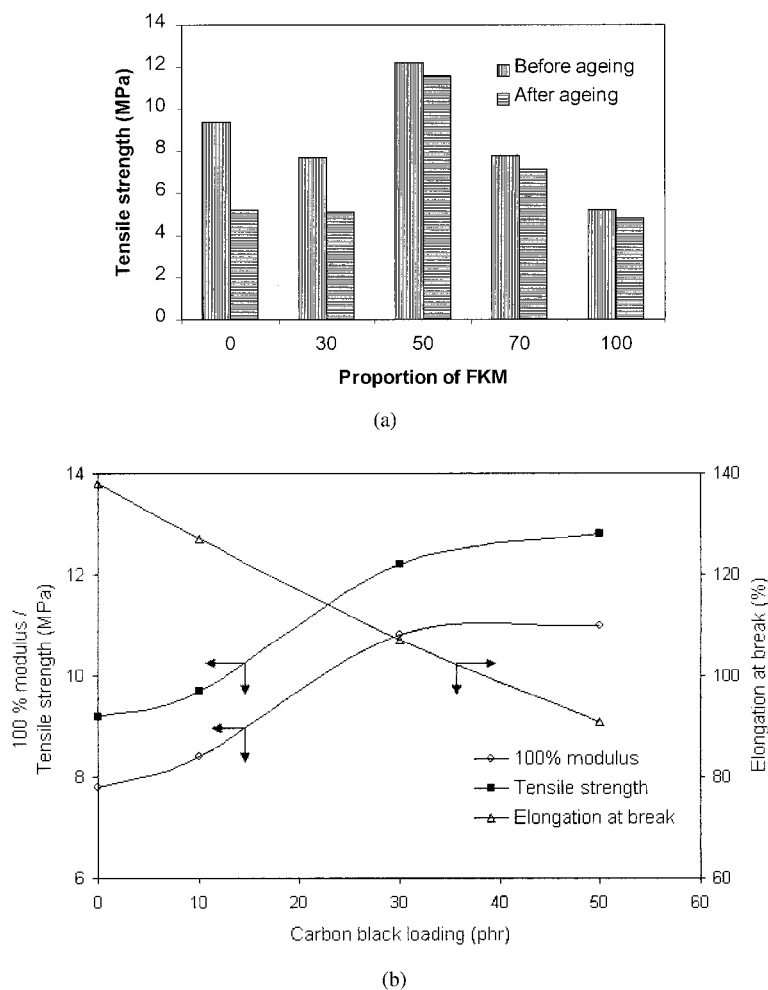
(a)

(b)

Figure 2 (a) Tensile strength of ACM/FKM/polyacrylate blends with varying compositions of ACM and FKM. (b) Effect of carbon black loading on the mechanical properties of 50/50/30 (w/w) ACM/FKM/p-TMPTA.

filler did not change this trend. Table I shows the numerical values of the important mechanical properties of the various filled blends under study. The mechanical properties of unfilled samples of the same blends were given in our earlier publication.[14] Evidently, the addition of carbon black increased the modulus, tensile strength, and tear strength and decreased the elongation at break of all the blends, regardless of their composition, but at different magnitudes. For example, the tensile strength of AFT30 increased from 9.1 MPa for the unfilled sample to 12.8 MPa for the filled one (AFT30F$_{30}$). At the same time, the elongation at break decreased from 138 to 107% for the same blend. Figure 2(a) depicts the effect of blend compositions of filled blends on tensile strength. The maximum improvement in tensile strength was observed for AFT30F$_{30}$. The effects of carbon black loading on the mechanical properties of AFT30 are given in Table I and Figure 2(b). As the carbon black loading increased, the tensile and tear strengths also increased, with a reduction in the elongation at break. However, the increases in tensile strength and modulus were

small in the range of 0–10 and 30–50 phr of carbon black loading and rose considerably when the loading was in the range 10–30 phr. So, we inferred that the optimum increase in properties of this filled rubber–plastic blend was achieved at a 30–50-phr loading of carbon black. Also, the addition of filler did not change the tension set of all of the blends, indicating a retention of the elastic recovery of the blends.

Effect of aging

Table I also shows the mechanical properties of the blends aged at 150°C for 36 h. The percentage change in properties of the blends after aging is indicated within the parenthesis. Regardless of their composition and nature, all of the blends showed negative values of percentage change in properties, indicating a deterioration of their properties with aging [Fig. 2(a)]. The addition of filler reduced the extent of heat aging by lowering the percentage change in tensile properties, especially the elongation at break. For example, the change in elongation at break of AFT30 decreased
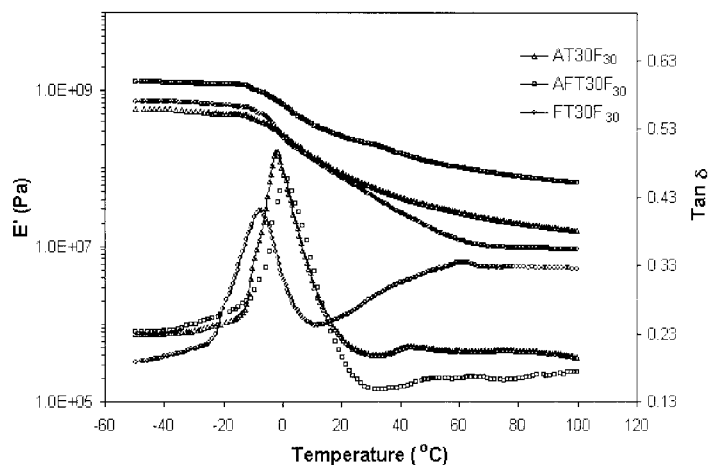
Figure 3 Dynamic mechanical thermal properties of N550-filled ACM, FKM, and 50/50/30 (w/w) ACM/FKM/*p*-TMPTA.

from −30.4% for the unfilled sample to −21.5% for the 30-phr N550-filled sample. The function of carbon black, in addition to reinforcement, was envisaged as a thermal antioxidant to control the aging of the blends to some extent. A higher loading of carbon black had the reverse effect in decreasing the percentage change in elongation at break (EB), % after aging. This may be attributed due to a reduction in the polymer volume to hold the filler material. We inferred that during aging, there was a cyclization and partial crosslinking of unused polyfunctional acrylate groups, resulting in an increased phase separation of the acrylate phase from the rubbery ACM/FKM phase. The cyclized product had lower mechanical properties, which led to a reduction in the overall properties.

Effect of swelling

The swelling of all of the filled blends was carried out in a representative solvent. Because methyl ethyl ketone (MEK) is a common solvent for both acrylic polymers and fluorocarbon polymers, the solubility of the blends were determined in this solvent for equilib-

rium swelling. Although the unfilled binary blend of ACM/FKM was completely soluble, the filled samples underwent disintegration rather than dissolution, indicating the presence of bound rubber. The unfilled ternary blend, AFT30, showed an equilibrium swelling of 134, which was lower than any of the binary and the ternary blends and as reported earlier.[14] The addition of carbon black decreased the swelling of all of the blends. This may have been due to a restriction to the penetration of solvent molecules into the bulk of the polymer by the filler–polymer network. The extent of the swelling of a blend in a solvent depends on the structure of the polymer phases and can be related to the properties of the polymer chains, such as molecular mobility, phase interaction, and so on.[21] As a filler preferentially reinforces the rubber phase, its introduction should reduce the volume swell of the rubber phase and, thus, reduce the overall volume swell of the blends in MEK.

DMTA

Figure 3 shows the temperature dependence of tan δ and E' for the blends with variations in blend ratios at

TABLE II
Dynamic Mechanical Properties of Filled ACM/FKM/Acrylate Rubber–Plastic Blends

					Composition (w/w)					
	$BF2_{30}$	AFT30	$AT30F_{30}$	AFT (70/30)F_{30}	$AFT30F_{30}$	AFT (30/70)F_{30}	$FT30F_{30}$	$AFT30S_{30}$	$AFH30F_{30}$	$AFT30F_{50}$
T_g (°C)	4.0	−0.3	−2.2	−1.0	0	−3.0	−8.0	0	−1.0	0
Tan δ at T_g	1.11	0.82	0.50	0.21	0.42	0.33	0.41	0.46	0.87	0.38
Tan δ at 25°C	0.25	0.22	0.21	0.14	0.19	0.15	0.27	0.16	0.18	0.19
Tan δ at 50°C	0.23	0.16	0.21	0.13	0.17	0.18	0.32	0.16	0.21	0.18
Tan δ at 100°C	0.28	0.15	0.20	0.12	0.18	0.15	0.33	0.17	0.25	0.18
Log E' at tan δ (Pa)	8.34	7.92	8.51	8.61	8.81	8.59	8.71	8.25	8.34	8.84
Log E' at 25°C (Pa)	6.46	7.22	7.84	7.99	8.35	8.05	7.77	7.47	7.50	8.40
Log E' at 50°C (Pa)	6.35	7.07	7.52	7.76	8.08	7.80	7.07	7.33	7.38	8.22
Log E' at 100°C (Pa)	6.31	6.98	7.20	7.52	7.81	7.59	6.96	7.18	7.13	7.97

a constant level of carbon black (30 phr). Table II shows the values of tan δ and E' at different temperatures for these systems. The binary blends of 30-phr filled ACM/p-TMPTA (AT30F_{30}) and FKM/p-TMPTA (AT30F_{30}) showed a maximum loss tangent value (tan δ_{max}) corresponding to the glass–rubber transition [glass-transition temperature (T_g)] at -2.2 and $-8.0°C$, respectively. Moreover, AT30F_{30} had a higher damping value (0.50) than FT30F_{30} (0.4) because of the more rubbery nature of AT30F_{30}. However, the E' below T_g of FT30F_{30} was higher than that of AT30F_{30}. When the change in E' with temperature of these two binary blends was compared, we saw that FT30F_{30} showed a dramatic change in E' above and below T_g, with the result that E' at 100°C was much less than that of AT30F_{30}. This could be attributed to incompatibility between the acrylate phase and FKM. This could be explained by the effect of temperature on phase stability. At higher temperatures, because of higher molecular mobility, there may have been a partial phase separation between FKM and the polyfunctional acrylate.

The filled ternary blends of ACM/FKM/polyacrylate with varying compositions of ACM and FKM showed interesting features in their dynamic mechanical properties. These blends showed a single T_g, representing the miscibility of ACM/FKM in the range $-3°C$ to 0 depending on the composition even in the presence of filler (Table II). As the level of FKM was increased form 0 to 100%, the tan δ_{max} peak shifted toward the positive temperature side with a maximum shift of 2°C, showing some kind of interaction as explained in our earlier publication.[14] The change in tan δ_{max} corresponding to damping characteristics did not follow any systematic trend, with the lowest value for AFT(70/30)F_{30} followed by that for AFT(30/70)F_{30}. This type of behavior could be explained in terms of the morphology of the blends, if it is assumed that the morphology did not change in the presence of filler. In our earlier publication, we explained the phase separation of FKM from ACM/polyacrylate phase with increasing levels of FKM.[15] This led to a change in the damping properties of the blends. However, the variation of tan δ values at higher temperatures, namely, 50 and 100°C, was significantly less because of the overall mobility of the polymer chain at higher temperatures. The variation of E' as a function of temperature for blends with different compositions of ACM/FKM is shown in Figure 3. As the FKM content increased, the E' value also increased up to 50% (AFT30F_{30}) and then decreased for AFT30/70F_{30}.

Variation of filler

The effects of the addition of carbon black (N550) and silica on the dynamic mechanical thermal properties of the blends are shown in Figure 4(a). The E' and tan
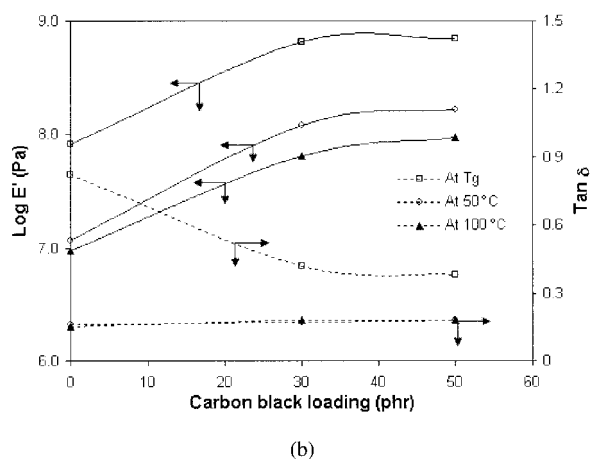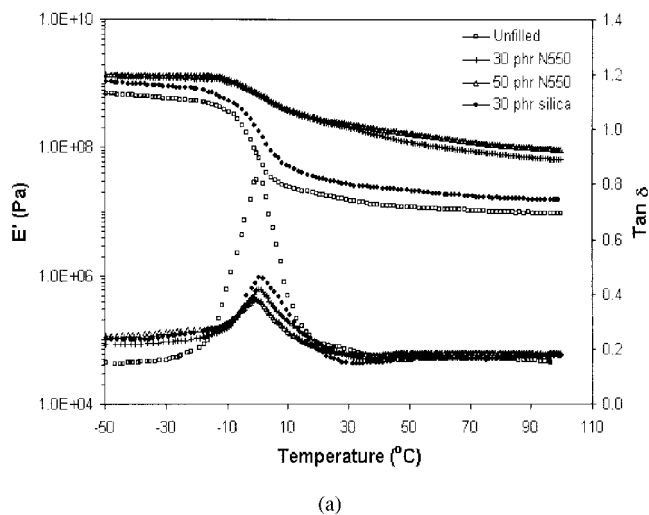

(a)

(b)

Figure 4   (a) Dynamic mechanical thermal properties of the blends of 50/50/30 (w/w) ACM/FKM/polyacrylate with silica and different levels of carbon black (N550). (b) Effect of carbon black loading on E' and tan δ of the 50/50/30 (w/w) ACM/FKM/polyacrylate blend.

δ values at different temperatures are given in Table II. The incorporation of filler into rubber–rubber and rubber–plastic blends decreased the damping characteristic of the blends by reducing the tan δ_{max} peak height. Moreover, the addition of filler broadened the half-width of the damping peak, indicating the reinforcing nature of filler with polymers.[22] The filler did not shift the T_g peak position of the rubber–plastic blend. However, there was a shift in T_g of the rubber–rubber blend (4°C) filled with 30-phr carbon black because of an increased restriction in the conformational mobility of the polymer chains by the filler–polymer network. Figure 4(b) shows the change in tan δ and E' values of the rubber–plastic blend (AFT30) filled with different levels of carbon black. The unfilled blend showed the highest tan δ_{max}, and the difference in tan δ_{max} between the unfilled and the 30-phr filled sample was larger than that between the 30- and 50-phr filled blends. However, there was not
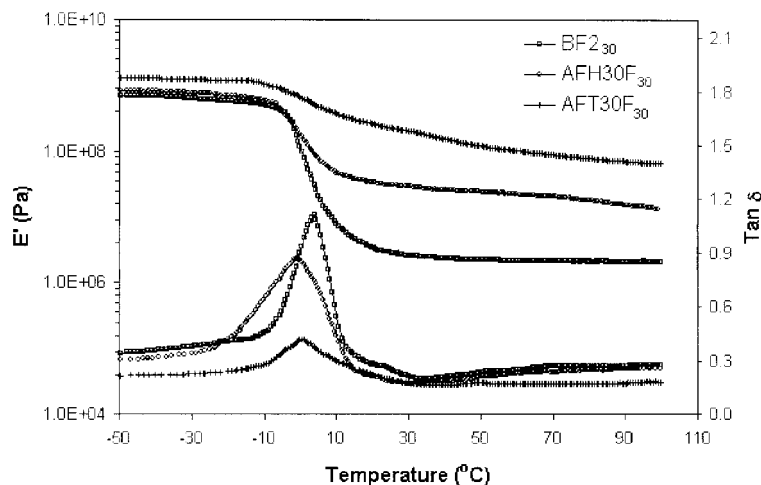
(a)
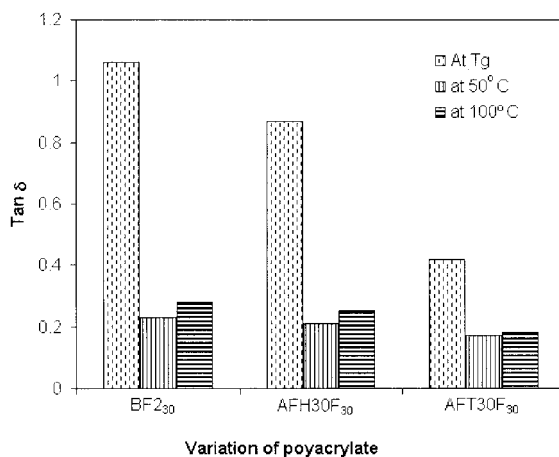
(b)

Figure 5 (a) Temperature versus log E' and tan δ of the blends of filled 50/50/30 (w/w/w) ACM/FKM/polyacrylate blends. (b) Effect of the variation of polyacrylate on tan δ of the filled 50/50/30 (w/w) ACM/FKM/polyacrylate blend.

much difference in the tan δ values at higher temperatures, namely, 50 and 100°C. This could be explained if one considers the segmental mobility of polymer chains at T_g. The unfilled polymer chains had more freedom of chain mobility and, thereby, showed higher damping characteristics. On the addition of reinforcing filler, the segmental motion was restricted by filler particles, which were bound to polymer chains either through secondary forces of attraction or by chemical interaction. The effect of silica in the reduction of tan δ_{max} and the broadening of the peak was almost similar to N550, indicating the reinforcing nature of silica. The trend in the variation of E' was almost similar to that of tan δ_{max} with increasing filler. The filler imparted more rigidity and stiffness to the polymer chains, and so, higher values of E' for the filled system were obtained.

Variation of the nature of acrylates

Figure 5(a) shows the influence of multifunctional acrylate on the dynamic mechanical thermal properties of the filled blend systems with and without polyacrylates. The filled ACM/FKM blend (BF2$_{30}$) without acrylate had a T_g of 4°C with a corresponding tan δ_{max} of 1.16. The unfilled and filled ternary blends (AFT30 and AFT30F$_{30}$, Table II) had a T_g value of 0°C with tan δ_{max} values of 0.82 and 0.42, respectively. The shift in T_g position for BF2$_{30}$ was attributed to filler–polymer interactions. However, the inclusion of polyacrylates, such as p-HDDA and p-TMPTA, did not change the T_g values but reduced the tan δ_{max} significantly. This indicated that polyacrylates reduced the damping properties by restricting the molecular mobility through either grafting or partial crosslinking. In doing so, the triacrylate (p-TMPTA) was more effective

than the diacrylate (*p*-HDDA), as shown by the higher tan δ_{max} for the diacrylate [Fig. 5(b)]. A comparison of E' (Table II) shows the increase in the dynamic modulus of the filled ACM/FKM blend by polyfunctional acrylates. A higher functional polyacrylate was more effective in increasing E' than one with a lower functionality.

CONCLUSIONS

We studied binary and ternary blends of ACM, FKM, and their blends with different polyacrylates to investigate the effects of fillers on their mechanical, dynamic mechanical, and aging properties. The following conclusions were drawn from these studies:

1. The incorporation of fillers into the binary and the ternary blends changed the nature of the stress–strain curves with increases in stress at break, indicating filler–polymer interactions. The tensile properties, such as tensile strength and tear strength, of all of the filled blends increased. However, there was a reduction in the elongation at break. The maximum increase in the tensile and tear strengths were observed at 30-phr carbon black loading. There was no change in the tension set of any of the filled blends, indicating the retention of elastic recovery properties.
2. The carbon-black-filled blends showed good retention of tensile properties after heat aging because of the thermal antioxidant nature of carbon black. However, the aging of the 50-phr N550-filled blend led to increased property deterioration because of a dilution effect.
3. The swelling of filled binary and ternary blends decreased with the inclusion of filler because of a restriction in solvent penetration into the bulk of the filled polymer.
4. The dynamic mechanical properties were influenced by the presence of filler. Although the position of T_g of the filled rubber–plastic blend was not altered, the damping characteristics (tan δ_{max}) at T_g were changed with a reduction in the height of the damping peak and a broadening of the peak. A further reduction in tan δ_{max} was observed for higher loadings of filler. The incorporation of filler increased the E' of the rubber–plastic blend at all of the measured temperatures.

References

1. Koning, C.; Duin, M. V.; Pagnoulle, C.; Jerome, R. Prog Polym Sci 1998, 23, 707.
2. Corish, P. J.; Powel, B. D. W. Rubber Chem Technol 1974, 47, 481.
3. Roland, C. M. In Handbook of Elastomers; Bhowmick, A. K.; Stephens, H. L., Eds.; Marcel Dekker: New York, 2001.
4. Robeson, L. M. In Polymer Compatibility and Incompatibility; Cole, K., Ed.; Harwood Academic: New York, 1982; p 177.
5. Kresge, E. N. In Polymer Blends; Paul, D. R.; Newman, S., Eds.; Academic: New York, 1978; Vol. 2, p 293.
6. Morris, H. L. Handbook of Thermoplastic Elastomers; Walker, B. M., Ed.; Van Nostrand Reinhold: New York, 1979; p 5.
7. Elliott, D. J. In Developments of Rubber Technology; Whelan, A.; Lee, K. S., Eds.; Elsevier Applied Science: Barking, UK, 1982; Vol. 3, p 203.
8. Gotoh, K. In Polymer Blends; Nikkon Kogyon Shinbun-sha: Tokyo, 1970; p 109.
9. Coran, A. Y.; Patel, R. Rubber Chem Technol 1981, 54, 892.
10. Coran, A. Y.; Patel, R. Rubber Chem Technol 1980, 53, 781.
11. Wu, C.; Yamagishi, T.; Nakamoto, Y.; Ishida, S. Polym Prepr Jpn 1999, 48, 4169.
12. Jha, A.; Bhowmick, A. K. Rubber Chem Technol 1997, 70, 798.
13. Jha, A.; Dutta, B.; Bhowmick, A. K. J Appl Polym Sci 1999, 74, 1490.
14. Kader, M. A.; Bhowmick, A. K. Rubber Chem Technol 2001, 74, 662.
15. Kader, M. A.; Bhowmick, A. K. J Mater Sci 2002, 37, 1503.
16. Kader, M. A.; Bhowmick, A. K. J Appl Polym Sci 2003, 89, 1442.
17. Kader, M. A.; Bhowmick, A. K. Polym Eng Sci 2003, 43, 975.
18. Donnet, J. B.; Bansal, R. C.; Wang, M. J. Carbon Black Science and Technology, 2nd ed.; Marcel Dekker: New York, 1993.
19. Kader, M. A.; Bhowmick, A. K. Polym Polym Compos 2001, 9, 263.
20. Patel, A. C. J Elast Plast 1999, 31, 213.
21. Klier, J.; Peppas, N. A. Polymer 1987, 28, 1851.
22. Maiti, S.; De, S. K.; Bhowmick, A. K. Rubber Chem Technol 1992, 65, 293.