Mill Processability of Brominated Isobutylene-Co-Paramethyl Styrene and Its Blends with Ethylene Propylene Diene Terpolymer (EPDM) in the Continuous Milling Operation

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ABSTRACT: The milling behavior of brominated isobutylene-co-paramethylstyrene (BIMS), ethylene propylene diene terpolymer (EPDM), and their blends was investigated over a range of temperatures and friction ratios using the continuous milling operation. At 30 °C, BIMS forms a loose nervy appearance and, as the temperature is increased to 90 °C, it gradually forms a tight elastic band. EPDM forms a loose band at all temperatures and friction ratios studied. For different blends of BIMS and EPDM, the milling behavior changes from a tight elastic band to a loose bagging band on increasing the EPDM content. Addition of different fillers results in lowering of the tendency to go to the back roll, even at lower temperatures. In all the filled systems, a smooth, regular, and tight elastic band is obtained at all the temperatures and friction ratios studied. © 2002 Wiley Periodicals, Inc. J Appl Polym Sci 85: 1484–1495, 2002

Key words: friction ratio; front-to-back roll transition; continuous milling; milling behavior; processability

INTRODUCTION

In the manufacture of rubber products, one of the most important processing operations is milling. The great advantage of two roll mills is that it represents an open process to which additives can be easily introduced. Until internal mixers replaced them, two roll mills have been the primary mixing equipment in the rubber industry. However, in many industries, mixing on two roll mill is still routinely carried out. In large rubber industries, mixing mills are mostly used in conjunction with Banbury (or internal mixers), calendars, or extruders during normal processing operations. Mills, although conjugational with internal mixers, are used to sheet off hot compounded rubber dumped onto them. This operation is necessary not only for instantaneous cooling of compounded polymer that is designed to avoid scorching and polymer degradation, but also for easier handling of the subsequent operations, such as mixing of curatives. In the case of extrusion operation, mixing mills are used to warm up the compounded rubber stock to obtain a uniform extrudate. Hence, it becomes very important that the compounded rubber stock forms a regular continuous band on the front roll (slower) to

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maintain continuity of the entire processing operation.

Milling behavior of natural rubber and synthetic rubbers has been studied by several researchers[1–10] with special reference to the effects of temperature and mixing conditions. It has been observed that some synthetic rubbers often show anomalous behavior, like back rolling and bagging. In the case of back rolling, the polymer tears across the length of the roll and then proceeds to band itself on the back (fast) roll, whereas in the case of bagging, the band separates from the roll. Consequently, the automatic knives are no longer in contact with the rubber. Therefore, the band must be manually cut, which is not considered a safe procedure under these conditions. This behavior delays the subsequent processing operation and adversely affects the product quality through air entrapment.

In actual practice, back rolling is overcome by the application of soaps or stearic acid on the back roll and bagging is controlled by changing the viscosity of the compounded rubber stock. It has been also reported that optimization of temperature and the mill parameters, such as friction ratio, nip gap, and roll speed, facilitate easy processing of these rubbers.

Results of studies on the milling behavior of a rubber aid in understanding the mixing behavior in an internal mixer. The flow of a rubber through the rolls of a mill is similar to the flow between the rotor tip and wall in an internal mixer as well as the flow between the rotors. Furthermore, the simpler geometry of a mill, allows an analytical solution for optimization of mixing conditions and understanding of the effects of material and process variables on mixing.

Several authors have analyzed the flow behavior of rubber at the nip area of a two-roll mill.[1–5] Bergen[2] and Pearson[6] published general reviews on the subject. Pasley[3] studied the flow of nonlinear viscoelastic fluids through the mill rolls. Chong[4] and Tokita and White[5] considered non-Newtonian viscoelastic behavior at the nip gap. The Reynolds lubrication theory of Newtonian hydrodynamics was used for the analysis.[7,8] White and Tokita[9] classified the mill processing behavior of a polymer in four regions in terms of mill band formation characteristics (Figure 1): Region 1, rubber remains mainly on the bank of the mill and only small quantity falling through the nip gap; Region 2, rubber forms a tight elastic band adhering to the front roll; Region 3, rubber compound torn and granulated at the nip region resulting in the formation of a loose band (bagging); Region 4, rubber compound forms a transparent fluid film band. The ease of processing, in the mill operation or the measure of good mill processability is determined by the capacity of the raw or compounded rubber to form a smooth elastic band on the front roll (i.e. the behavior under Region 2).

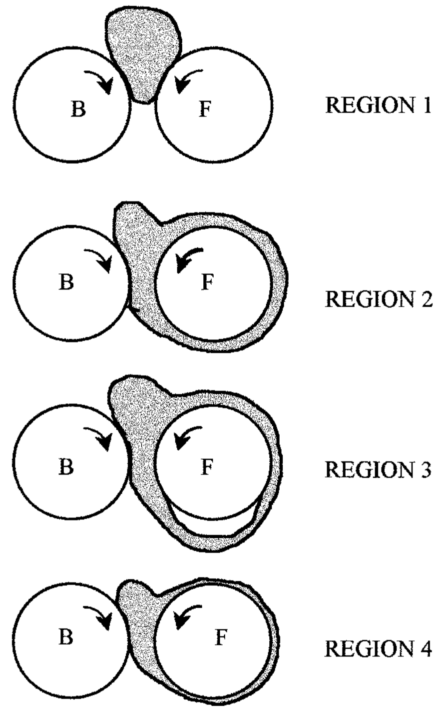


Figure 1 Schematic drawing showing the four regions of mill behavior.[9] Key: (B): back roll; (F) front roll.

Tokita[10] described the mill behavior in terms of the front-to-back roll transition as a consequence of imbalance between the moments due to adhesive and shear forces acting at the point of separation of the band from the roll (i.e., point 'S' in Figure 2). The ratio of moments of balance is defined as

$$\frac{M_a}{M_s} = \frac{F_a t_p}{\eta} \qquad (1)$$

where M_a and M_s are the moments of balance due to adhesive forces (F_a) and shear forces, respectively, F_a is the adhesive force per unit area, t_p is the process time (i.e., the time for which the material remains in the nip region), and η is the viscosity of the material being processed. As defined by Tokita,[10] the imbalance between the moments causes the material to band on either roll according to the following conditions:

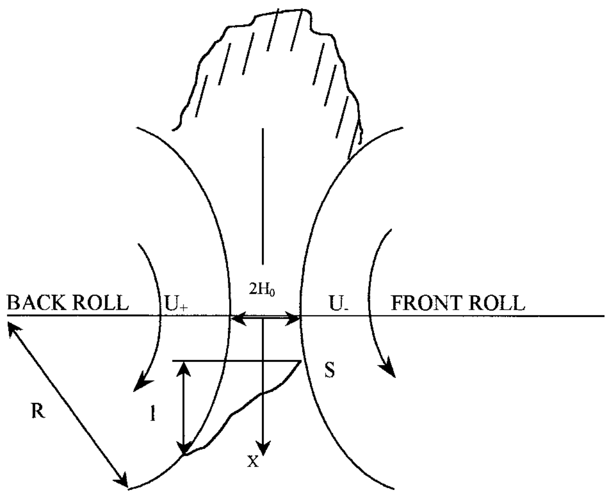


Figure 2 Moment balance at the nip section, where U_+ and U_- are the speeds of the back and front rolls, respectively, $2H_0$ is the critical nipe gap, S is the point on the front roll where the band separates from the front roll, and l is the difference in length of polymer contact at the roll surfaces due to the difference in the speed of the roll.[10]

$$\frac{F_a t_p}{\eta} \begin{cases} = 1, & \text{no band formation} \\ > 1, & \text{band formation on back roll} \\ < 1, & \text{band formation on front roll} \end{cases} \quad (2)$$

The front-to-back roll transition (F-B transition) of the band would take place if the torque acting on the material at the point of separation at the back roll is greater than that acting towards the front roll. Hence, the moment of balance at the nip area would provide a necessary condition for the F-B transition (Figure 2).

It has been reported earlier[11–13] that the viscosity of the filled rubber is considerably influenced by the structure and the particle size of the filler. Fillers having high structure and small particle size increase the viscosity to a greater extent than fillers with lower structure and larger particle size. Secondly, the elastic recoil after the shear flow reduces by addition of filler. It has also been reported that this elastic recoil decreases on increasing the structure of filler or filler loading and with decreasing particle size. It has been demonstrated that increasing the filler loading decreases the tendency toward melt flow instabilities in polymer processing, and addition of plasticizer to the filled rubber compounds influences their rheological behavior considerably and in general reduces the viscosity of the filled system.

Brominated isobutylene-co-paramethylstyrene (BIMS) is a new class of rubbers that possesses improved thermal stability and cure compatibility in blends compared with conventional butyl polymers. However, the processing behavior of the rubber is not fully understood. Recently, Kumar et al.[14,15] reported the mill processability of gum BIMS and its blends with ethylene propylene diene terpolymer (EPDM) by drop mill operation. Waddell and Poulter[16] reported phase mixing of BIMS-based tire tread compound. BIMS blends have been also discussed by Potluri et al.,[17] McElrath and Tisler,[18] Kruse et al.,[19] and Flowers et al.[20] These authors have discussed specific mixing protocols for obtaining an advantageous morphology. However, the influence of fillers and process oil on the milling behavior has not been reported. The present investigation deals with the milling behavior of BIMS, EPDM, and their blends with respect to the F-B transition in a continuous mill operation. The effects of carbon black (N330, N550, N774) and silica and processing oil on the milling behavior of the rubber compositions are reported.

EXPERIMENTAL

Materials

Polymers

Brominated isobutylene-co-paramethylstyrene (BIMS-7745; paramethylstyrene content, 7.7 wt %; bromine content, 1.2 mol %; ML_{1+8} 125 °C, 45) and ethylene propylene diene terpolymer (Vistalon-2504; ethylene content, 50 wt %; diene content, 4–7 wt %; ML_{1+8} 125 °C, 26) were supplied by Exxon Mobil Chemical Company, Houston, TX.

Fillers

Carbon blacks (grade, N330; particle size, 26–30 nm; grade, N550; particle size, 40–48 nm and grade, N774; particle size, 61–100 nm) were provided by Philips Carbon Black Ltd., Durgapur, West Bengal, India. Silica (grade, Ultrasil VN_3; particle size, 20–100 nm) was provided by Bayer India Limited, Mumbai, India.

Processing Oil

SUNPAR 2280, supplied by Sunoco, Inc., Philadelphia, PA, with aromatic carbon atoms (4%), naphthenic carbon atom (23%), and paraffinic carbon atoms (73%), was used.

Table I Milling Behavior of BIMS, EPDM, and Their Blends at Different Temperatures and Friction Ratios[a]

Blend Designation	Temperature (°C)											
	30			50			70			90		
	Friction Ratio			Friction Ratio			Friction Ratio			Friction Ratio		
	1 : 1.2	1 : 1.5	1 : 1.8	1 : 1.2	1 : 1.5	1 : 1.8	1 : 1.2	1 : 1.5	1 : 1.8	1 : 1.2	1 : 1.5	1 : 1.8
$B_{100}V_0$	B-R (4)	B-R (4.5)	B-R (5)	B-R (2.5)	B-R (2)	B-R (1.5)	F-R (8)	F-R (5)	F-R (4)	F-R (<1)	F-R (<1)	F-R (<1)
$B_{90}V_{10}$	B-R (4)	B-R (5)	B-R (5.5)	F-R (3)	F-R (2.5)	F-R (2)	F-R (12)	F-R (9)	F-R (6)	F-R (<0.5)	F-R (<0.5)	F-R (<0.5)
$B_{80}V_{20}$	F-R (10)	F-R (8.5)	F-R (5.5)	F-R (6)	F-R (5)	F-R (4)	F-R (>20)	F-R (15)	F-R (10)	F-R (<0.5)	F-R (<0.5)	F-R (<0.5)
$B_{70}V_{30}$	F-R (>20)	F-R (>20)	F-R (>20)	F-R (15)	F-R (10)	F-R (7.5)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (<1)	F-R (<1)	F-R (<1)
$B_{60}V_{40}$	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>1)	F-R (>2)	F-R (>2)
$B_{50}V_{50}$	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>5)	F-R (>5)	F-R (>5)
$B_{40}V_{60}$	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R* (>20)	F-R* (>20)	F-R* (>20)
$B_{30}V_{70}$	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R* (>20)	F-R* (>20)	F-R* (>20)
$B_{20}V_{80}$	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R* (>20)	F-R* (>20)	F-R* (>20)	F-R* (>20)	F-R* (>20)	F-R* (>20)
$B_{10}V_{90}$	F-R (>20)	F-R (>20)	F-R (>20)	F-R* (>20)	F-R* (>20)	F-R* (>20)	F-R* (>20)	F-R* (>20)	F-R* (>20)	F-R* (>20)	F-R* (>20)	F-R* (>20)
B_0V_{100}	F-R* (>20)	F-R* (>20)	F-R* (>20)	F-R* (>20)	F-R* (>20)	F-R* (>20)	F-R* (>20)	F-R* (>20)	F-R* (>20)	F-R* (>20)	F-R* (>20)	F-R* (>20)

[a] Key: (B-R) back roll; (F-R) front roll; (*) bagging; (values in bracket) duration (min) for which band stays on the roll.

Preparation of Samples

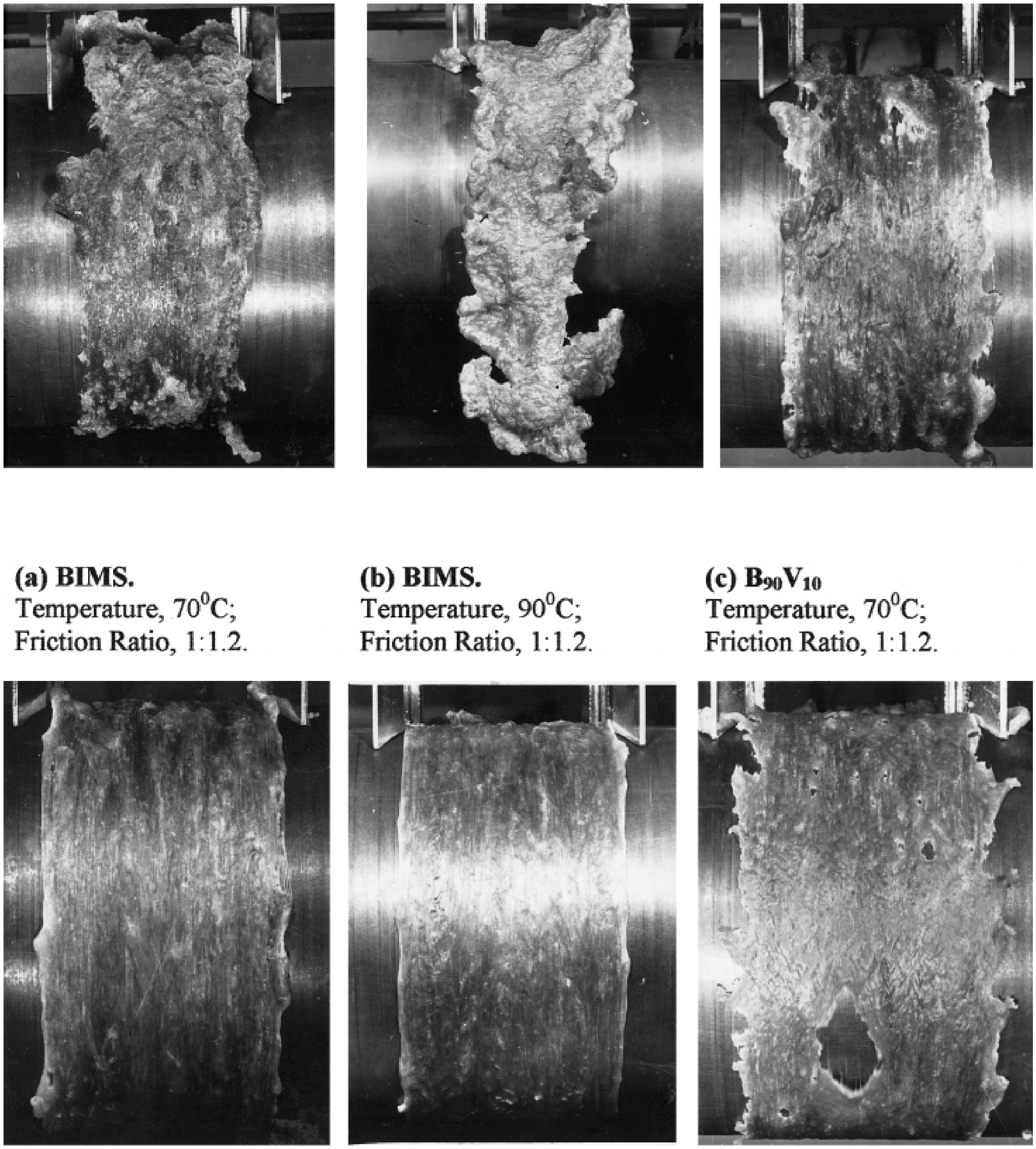
For studies of unfilled systems, blends of BIMS-7745 and VISTALON-2504 in the ratios of 90:10, 80:20, 70:30, 60:40, 50:50, 40:60, 30:70, 20:80, and 10:90 were prepared in a Brabender Plasticorder (model PLE 330, capacity 65 mL) at 80 °C and 60 rpm. For studies on filled systems, blends in the ratios of 90:10, 70:30, 50:50, and 30:70 were used. During mixing operation, the gum polymers were initially mixed for 2 min and then the filler was added. For mixes containing oil, half of the filler and full quantity of oil were added after 2 min and then the rest of filler was added after 4 min. The mixing time in all cases was kept constant at 6 min. Gum and filled blends were designated as B_nV_m were n and m are the weight percent of BIMS and EPDM in the blend, respectively. For filled systems, the type of filler and its loading has been mentioned.

Mixing Mill

An oil-heated, laboratory size, two-roll mixing mill (0.15 × 0.33 m) supplied by Farrell Bridge Ltd, U.K., was used for this work. The normal operating speed for the front roll was varied from 16.8 to 33.6 rpm, whereas the speed of the back roll was kept constant at 33.6 rpm.

Continuous Mill Operation

A procedure similar to the one used by Tokita[10] and Bhowmick et al.[21–24] was followed. About 100 g of the material (after mixing in the Brabender) was banded on the front roll of the mill at a particular nip gap (0.4 mm), and milling was continued for a certain time interval (20 min). The behavior of the band was observed with the time of milling. The time beyond which the band transferred itself to the back roll was noted. This time is defined F-B transition time.



(a) BIMS. Temperature, 70^0C; Friction Ratio, 1:1.2.

(b) BIMS. Temperature, 90^0C; Friction Ratio, 1:1.2.

(c) $B_{90}V_{10}$ Temperature, 70^0C; Friction Ratio, 1:1.2.

(d) $B_{90}V_{10}$ Temperature, 90^0C; Friction Ratio, 1:1.2.

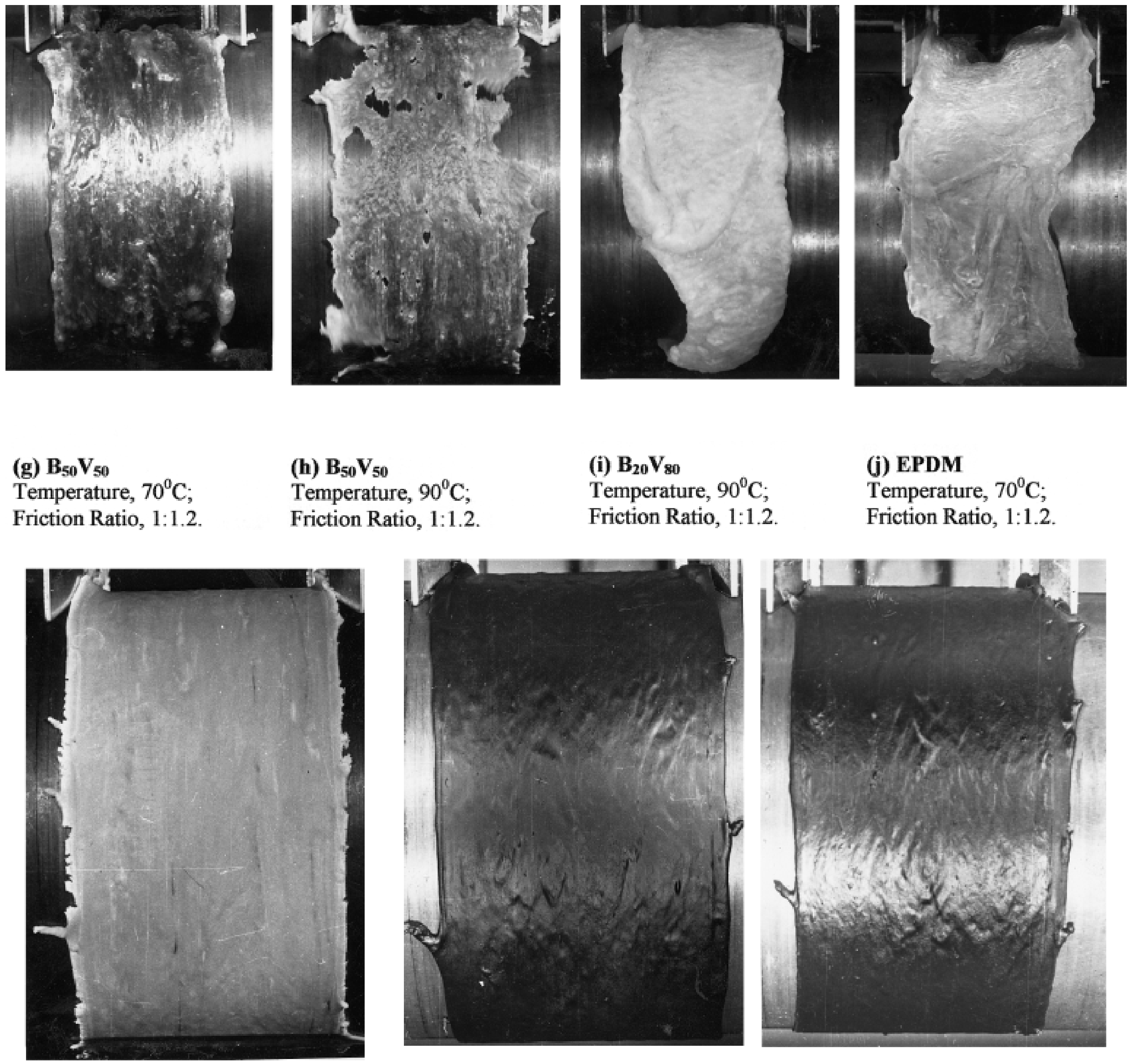
(e) $B_{70}V_{30}$ Temperature, 70^0C; Friction Ratio, 1:1.2.

(f) $B_{70}V_{30}$ Temperature, 90^0C; Friction Ratio, 1:1.2.

Figure 3 Milling behavior of gum and filled BIMS and BIMS–EPDM blends.

Viscosity Measurements

Viscosity measurements were made with a Monsanto Processability Tester (capillary rheometer) at a shear rate in the milling region (i.e., 36–73 s^{-1}) and temperature of 70 °C. The capillary length (30 mm) to diameter (1 mm) ratio (*L*/*D*) was 30. The preheat time for each sample was 5 min.



(g) $B_{50}V_{50}$
Temperature, 70⁰C;
Friction Ratio, 1:1.2.

(h) $B_{50}V_{50}$
Temperature, 90⁰C;
Friction Ratio, 1:1.2.

(i) $B_{20}V_{80}$
Temperature, 90⁰C;
Friction Ratio, 1:1.2.

(j) EPDM
Temperature, 70⁰C;
Friction Ratio, 1:1.2.

(k) BIMS
FILLER: Silica-30 phr.
Temperature, 70⁰C;
Friction Ratio, 1:1.2.

(l) BIMS
FILLER: N330 carbon black-30 phr.
Temperature, 70⁰C;
Friction Ratio, 1:1.2.

(m) BIMS
FILLER: N550 carbon black-30 phr.
Temperature, 70⁰C;
Friction Ratio, 1:1.2.

Figure 3 (*Continued from the previous page*)

Determination of Rubber-to-Metal Adhesive Force

The joint strength between rubber and metal was measured using a 180° peel adhesion test specimen. A rubber sheet of 3 mm thickness, backed by cotton fabric, was pressed on the metal (steel) strip under a pressure of 35 MPa. An adhesion test was done at a jaw separation rate of 50 mm/min at 25 °C within 48 h of making the rubber-to-metal bonded test sample. The adhesion force (G_a) was calculated using the following formula:

$$G_a = \frac{2F}{w} \tag{3}$$

where F is the force of separation in N and w is the width of the joint in meters.

RESULTS AND DISCUSSION

The milling behavior of BIMS, EPDM, and their blends by continuous milling operation is summarized in Table I. BIMS showed remarkable changes in milling behavior as the temperature of the roll was increased from 30 to 90 °C. At 30 °C, it formed a torn and 'nervy' band on the back roll only. This band (bagged) fell from the roll after some time and on increasing the friction ratio the time of fall from the rolls marginally increased (Table I). On increasing the temperature of the roll to 50 °C, BIMS again formed the band on the back roll, but this time the band was less 'nervy' and also the time of fall from the roll decreased compared with that at 30 °C. The time of fall from the roll decreased on increasing the friction ratio. The characteristic of the band at 30 and 50 °C is similar to Region 1 (Figure 1) of the mill processing behavior as classified by White and Tokita.[9] On further increasing the temperature of the roll to 70 °C, BIMS formed the band on the front roll (Figure 3a). The band formed was rough in appearance and elastic in nature. The characteristic of this band was intermediary to Region 1 and Region 2 of the mill processing behavior.[9] The time of fall from the roll decreased as the friction ratio was increased (Table I). At 90 °C, initially the band split into two parts before going to the back roll and then transferred completely to the front roll and formed a broken band with cuts on the side of band (Figure 3b). After some time it fell from the roll.

The $B_{90}V_{10}$ blend at 30 °C behaved in very similar manner to BIMS and formed a band on the back roll on being dropped on the mill. The time of the fall from the roll increased with the friction ratio (Table I). On increasing the temperature of the roll to 50 °C, it formed a regular band on the front roll with some cuts on the sides and some within the band, which increased in size with milling time. The time of the fall from the roll increased on increasing the friction ratio (Table I). This band was less 'nervy' in character compared with BIMS. On increasing the temperature to 70 °C, band formation became more regular with some cuts on the side of band (Figure 3c) and cuts within the band increased in size with milling time. At 90 °C, the behavior was very similar to BIMS though the cuts were larger (Figure 3d). The characteristic of band at all temper-

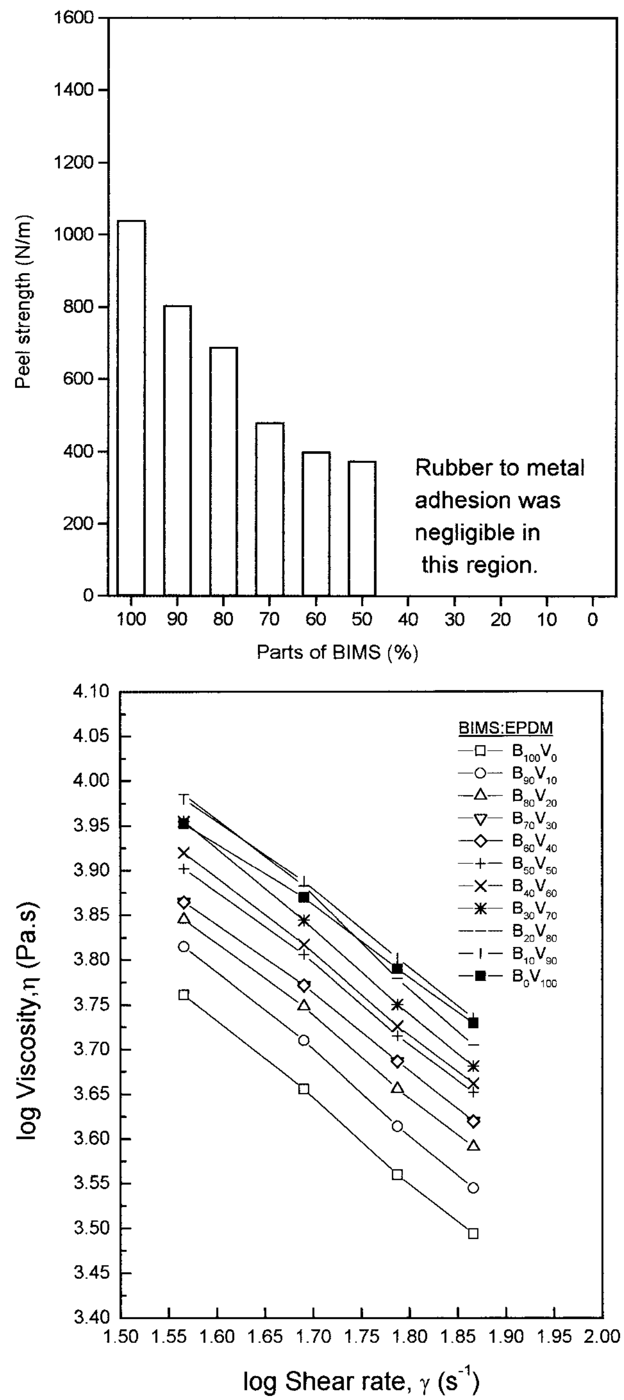


Figure 4 (a) Rubber-to-metal adhesion of different BIMS–EPDM blends. (b) Viscosity of BIMS, EPDM, and their blends.

Table II Milling Behavior of BIMS Loaded with 30 phr of Fillers at Different Temperatures and Friction Ratios[a]

	Temperature (°C)											
	30			50			70			90		
	Friction Ratio			Friction Ratio			Friction Ratio			Friction Ratio		
Filler Type	1 : 1.2	1 : 1.5	1 : 1.8	1 : 1.2	1 : 1.5	1 : 1.8	1 : 1.2	1 : 1.5	1 : 1.8	1 : 1.2	1 : 1.5	1 : 1.8
N330	B-R (>20)	B-R (>20)	B-R (>20)	B-R (>20)	B-R (>20)	B-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)
N550	B-R (>20)	B-R (>20)	B-R (>20)	B-R (>20)	B-R (>20)	B-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)
N660	B-R (>20)	B-R (>20)	B-R (>20)	B-R (>20)	B-R (>20)	B-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)
Silica	B-R (>20)	B-R (>20)	B-R (>20)	B-R (>20)	B-R (>20)	B-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R* (>20)	F-R* (>20)	F-R* (>20)

[a] Key: (B-R) back roll; (F-R) front roll; (*) bagging; (values in bracket) duration (min) for which band stays on the roll.

atures was similar to Region 2 of the mill processing behavior.[9]

The $B_{80}V_{20}$ blend formed a band at 30 °C on the front roll with some cuts inside the band, which increased in size with time. On increasing the temperature of the roll to 50 °C, it formed a smooth and regular band on the front roll with smaller cuts on the sides. The band stayed on the front roll for a longer time than neat BIMS or the $B_{90}V_{10}$ blend (Table I). On increasing the temperature to 70 °C, a more regular continuous band was formed on the front roll. On increasing the friction ratio between the rolls, the time of fall from the front roll decreased (Table I). At 90 °C, the blend again formed regular continuous band on the front roll. The characteristics of the band at all temperatures were similar to Region 2 of the mill processing behavior.[9]

The $B_{70}V_{30}$ blend, formed a regular continuous band at 30 °C on the front roll. On increasing the temperature of the rolls to 70 °C, the smoothness of the band increased (Figure 3e). At 90 °C, very large cuts were observed at longer milling time (Figure 3f).

The $B_{60}V_{40}$, $B_{50}V_{50}$, $B_{40}V_{60}$, and $B_{30}V_{70}$ blends formed a smooth and regular band on the front roll at all the temperatures studied (Figure 3g). For these blends, large cuts at the sides of the bands were observed at 90 °C at longer mixing time (Figure 3h). At temperatures <90 °C, the band stayed on the front roll for >20 min. The characteristic of the bands for these blends (containing up to 80 parts of EPDM) was similar to Region 2 of the mill processing behavior.[9]

For the $B_{20}V_{80}$ blend, some bagging (Region 3; Figure 3i) appeared at longer duration at higher temperature (90 °C), whereas for the $B_{10}V_{90}$ blend, it appeared within 2 min of milling at lower temperature (50 °C), indicating that the addition of EPDM at high loading increases the tendency to bag, and the mill band formation behavior changes from Region 2 to Region 3.

As expected, EPDM showed bagging even at lower temperature (Region 3; Figure 3j), and the tendency to bag increased on increasing the temperature.

To explain these results, rubber-to-metal adhesion and viscosity of BIMS and its blends with EPDM were measured. It was observed that on increasing the EPDM content, the rubber-to-metal adhesion decreased and beyond 50 parts of EPDM and the blends failed to stick to the metal (Figure 4a), whereas the viscosity increased (Figure 4b). This result caused an overall decrease of the $F_a t_p/\eta$ or M_a/M_s factor, which in turn decreased the tendency to go to the back roll.

Effect of Fillers on Mill Processing Behavior of BIMS

Mill processing behavior of BIMS changed with the addition of different fillers. Even at lower temperatures (30 °C), this effect was observable, and at the higher temperatures (>50 °C) it became prominent (Figures 3k– 3m). At 30 °C, filled

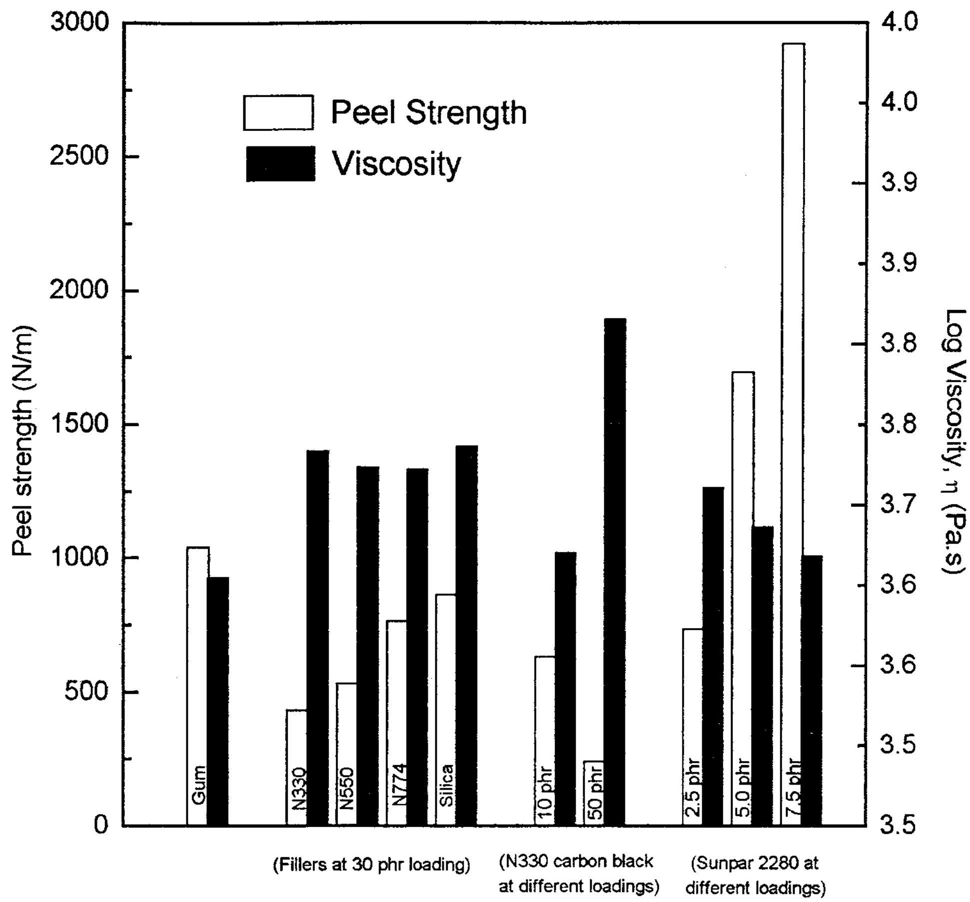


Figure 5 Effect of fillers and oil on the rubber-to-metal adhesion and shear viscosity at 49 s^{-1} (viscosity at other shear rates, not shown, follows the same trend).

BIMS showed a tendency to go to the back roll at higher friction ratio (at friction ratio 1:1.8), even though the band was much smoother and regular than that of the gum polymer (Table II). In addition, unlike gum polymer, the filled systems stayed on the front roll for a longer duration of time. The characteristics of the band for these systems were similar to Region 2 of the mill processing behavior, as described earlier.

On increasing the temperature of rolls to 50 °C, different filled systems showed a tendency to go to the back roll at a higher friction ratio 1:1.8. In the case of the silica filled system, the tendency for the back roll transition was somewhat higher, as BIMS formed a band on the back roll only (Table II). On further increasing the temperature of the rolls to 70 °C, different filled systems formed a tight elastic band on the front roll, even at a higher friction ratio (1:1.8). The bands were also smooth and regular in nature (Region 2). The elastic nature of the band could be observed by cutting it with the knife, which showed some contraction. In the case of carbon black N330 and silica, this contraction was much lower than that of carbon black N550 and N774. On further increasing the temperature of the rolls to 90 °C, different filled systems formed a tight elastic band on the front roll. The bands formed were smoother and more regular in nature than the bands at 70 °C, although they were somewhat less elastic in nature.

To explain these results, rubber-to-metal adhesion for filled BIMS systems was again measured (Figure 5). It was observed that with addition of different fillers, the rubber-to-metal adhesion decreased compared with that of the gum rubber. Further, it was observed that systems containing reinforcing filler (carbon black N330) showed lower value adhesion compared with systems containing low and nonreinforcing fillers (carbon black N550 and N774). In the case of silica-filled compounds, a somewhat higher value of rubber-

Table III Effect of Fillers (at 30 phr Loading) on Milling Behavior of BIMS, EPDM, and their Blends at Different Temperatures and at a Constant Friction Ratio of 1 : 1.2[a]

	N330 Carbon Black				N550 Carbon Black				N774 Carbon Black				Silica			
	Temperature (°C)				Temperature (°C)				Temperature (°C)				Temperature (°C)			
Blend Designation	30	50	70	90	30	50	70	90	30	50	70	90	30	50	70	90
$B_{100}V_0$	B-R (>20)	B-R (>20)	F-R (>20)	F-R (>20)	B-R (>20)	B-R (>20)	F-R (>20)	F-R (>20)	B-R (>20)	B-R (>20)	F-R (>20)	F-R (>20)	B-R (>20)	B-R (>20)	F-R (>20)	F-R* (>20)
$B_{90}V_{10}$	F-R (>20)	B-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	B-R (>20)	F-R (>20)	F-R (>20)	B-R (>20)	B-R (>20)	F-R (>20)	F-R (>20)	B-R (>20)	B-R (>20)	F-R (>20)	F-R* (>20)
$B_{70}V_{30}$	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R* (>20)
$B_{50}V_{50}$	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R* (>20)
$B_{30}V_{70}$	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R* (>20)	F-R* (>20)	F-R* (>20)	F-R* (>20)
B_0V_{100}	F-R* (>20)	F-R* (>20)	F-R* (>20)	F-R* (>20)	F-R* (>20)	F-R* (>20)	F-R* (>20)	F-R* (>20)	F-R* (>20)	F-R* (>20)	F-R* (>20)	F-R* (>20)	F-R* (>20)	F-R* (>20)	F-R* (>20)	F-R* (>20)

[a] Key: (B-R) back roll; (F-R) front roll; (*) bagging; (values in bracket) duration (min) for which band stays on the roll.

to-metal adhesion was observed (Figure 5). Furthermore, the viscosity of the compounds containing reinforcing fillers was higher than that of the low or non-reinforcing filler (Figure 5). Therefore, for reinforcing fillers, the factor M_a/M_s is lower than that of low or nonreinforcing fillers, which in turn lowers the tendency for the F-B roll transition.

Effect of Blend Composition of Filled Systems

The effect of the blend composition on the milling behavior of filled systems was studied by varying the EPDM content at a friction ratio of 1:1.2. For filled $B_{90}V_{10}$ and $B_{70}V_{30}$ systems, a tight, smooth, and regular elastic band on the front roll was observed at all temperatures. The characteristic of the band was similar to Region 2. The bands formed at 70 and 90 °C were smoother and more regular in nature than the bands at 30 and 50 °C; yet again, these were somewhat less elastic in nature (Table III).

For the filled $B_{50}V_{50}$ systems, a tight elastic band on the front roll was observed at all temperatures and friction ratios. The bands formed were much smoother and regular in nature, though some bagging was observed at higher temperatures (70 and 90 °C). Hence, the band characteristics changed from Region 2 to Region 3 on increasing the temperature and EPDM content (Table III).

For the filled $B_{30}V_{70}$ and EPDM systems, a loose band (bagging) on the front roll was observed at all temperatures and friction ratios. The bagging was much more prominent at higher temperatures (70 and 90 °C) compared with that at lower temperatures (30 and 50 °C). The band characteristic for these systems was similar to Region 3.

Effect of Filler Loading on Milling Behavior for Blends

The effect of the filler loading was studied by varying the carbon black N330 loading at a friction ratio 1:1.2. An increase in the filler loading decreased the tendency for the material to go to the back roll (Table IV). Blends containing 10 phr of carbon black and higher BIMS content showed a modest tendency to go to the back roll, whereas the system containing 50 phr of carbon black formed a band on the front roll only (Table IV). On increasing the EPDM content, the smoothness of the band increased and the mill band formation behavior changed from Region 2 to Region 3.

To explain these results, again, the rubber-to-metal adhesion and viscosity of these systems were measured, and it was observed that on increasing the filler content, the rubber-to-metal adhesion decreased, whereas the viscosity increased (Figure 5). This result caused an overall decrease of $F_a t_p/\eta$ or M_a/M_s factor, which in turn decreased the tendency to go to the back roll.

Effect of Oil Loading on Milling Behavior for Blends

The effect of oil content on milling behavior was studied by varying the Sunpar 2280 loading at a

Table IV Effect of Loading of N330 Carbon Black on Milling Behavior of BIMS, EPDM, and their Blends at Different Temperatures and at a Constant Friction Ratio of 1 : 1.2[a]

Blend Designation	Filler Loading (phr)							
	10				50			
	Temperature (°C)				Temperature (°C)			
	30	50	70	90	30	50	70	90
$B_{100}V_0$	B-R (>20)	B-R (>20)	B-R (>20)	B-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)
$B_{90}V_{10}$	B-R (>20)	B-R (>20)	B-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)
$B_{70}V_{30}$	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)
$B_{50}V_{50}$	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)
$B_{30}V_{70}$	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)
B_0V_{100}	F-R* (>20)	F-R* (>20)	F-R* (>20)	F-R* (>20)	F-R* (>20)	F-R* (>20)	F-R* (>20)	F-R* (>20)

[a] Key: (B-R) back roll; (F-R) front roll; (*) bagging; (values in bracket) duration (min) for which band stays on the roll.

friction ratio 1:1.2. An increase in the oil loading increased the tendency to go to the back roll for BIMS and $B_{90}V_{10}$ mixes (Table V). The blends containing up to 5.0 phr of oil formed a band on the front roll only, whereas the system containing 7.5 phr of oil and higher BIMS content showed

Table V Effect of Process Oil Loading on Milling Behavior for BIMS, EPDM, and their Blends Filled with 30 phr of N330 Carbon Black at Different Temperatures and at a Constant Friction Ratio of 1 : 1.2[a]

Blend Designation	Loading of Sunpar 2280 (phr)											
	2.5				5.0				7.5			
	Temperature (°C)				Temperature (°C)				Temperature (°C)			
	30	50	70	90	30	50	70	90	30	50	70	90
$B_{100}V_0$	B-R (>20)	B-R (>20)	F-R (>20)	F-R (>20)	B-R (>20)	B-R (>20)	B-R (>20)	F-R (>20)	B-R (>20)	B-R (>20)	B-R (>20)	B-R (>20)
$B_{90}V_{10}$	F-R (>20)	B-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	B-R (>20)	B-R (>20)	F-R (>20)	B-R (>20)	B-R (>20)	B-R (>20)	B-R (>20)
$B_{70}V_{30}$	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)
$B_{50}V_{50}$	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)
$B_{30}V_{70}$	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)	F-R (>20)
B_0V_{100}	F-R* (>20)	F-R* (>20)	F-R* (>20)	F-R* (>20)	F-R* (>20)	F-R* (>20)	F-R* (>20)	F-R* (>20)	F-R* (>20)	F-R* (>20)	F-R* (>20)	F-R* (>20)

[a] Key: (B-R) back roll; (F-R) front roll; (*) bagging; (values in bracket) duration (min) for which band stays on the roll.

some tendency to go to the back roll (Table V). On increasing the EPDM content, the smoothness of the band increased and the mill band formation behavior changed from Region 2 to Region 3.

To explain these results, rubber-to-metal adhesion and viscosity of these systems were measured. On increasing the oil content, the rubber-to metal adhesion increased (Figure 5), whereas the viscosity decreased (Figure 5), which caused overall increase of $F_a t_p/\eta$ or M_a/M_s factor, which in turn increased the tendency to go to the back roll.

CONCLUSIONS

In the present investigation, the milling behavior of BIMS, EPDM, and their blends was investigated over a range of temperatures and friction ratios using the continuous milling operation. The effects of fillers and process oil on the milling behavior of BIMS and its blends with EPDM were also investigated. The conclusions from this investigation are as follows:

1. As the temperature of the rolls is increased from 30 to 90 °C, the band characteristics of BIMS changed from a loose nervy band to a tight elastic band.
2. EPDM forms a loose band at all temperatures and friction ratios studied.
3. Milling behavior changes from a tight elastic band to a loose bagging band on increasing the EPDM content.
4. Addition of different fillers results in a lowering of the tendency to go to the back roll even at lower mill temperatures.
5. In the case of filled BIMS systems, a smooth, regular, and tight elastic band was obtained at all the temperatures and friction ratios studied.
6. The tendency for a F-B transition decreased on increasing the loading of N330 carbon black, whereas it increased with an increase in the loading of the processing oil.

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