Mill Processability of Brominated Isobutylene-*co*-paramethylstyrene and Its Blends with EPDM

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ABSTRACT: Milling behavior of brominated isobutylene-*co*-paramethylstyrene (BIMS) and its blends with ethylene propylene diene terpolymer (EPDM) rubber, was investigated over a range of temperatures and friction ratios in a drop mill operation. BIMS showed striking changes, that is, from a loose nervy band to a tight elastic band, as the temperature of the rolls was increased from 30°C to 90°C. For EPDM a loose band was observed at all temperatures and friction ratios studied. For the blends of BIMS and EPDM, the milling behavior changed from a tight elastic band to a loose bagging band on increasing the EPDM content. The critical nip gap (CNG), at which the front-to-back roll (F–B) transition occurred, was also measured. BIMS showed a much higher value of CNG than that of EPDM, indicating that the former had a significantly higher tendency for F–B transition than the latter material. For different blends of BIMS and EPDM, the CNG decreased on increasing the EPDM content, indicating a decrease in the tendency for F–B transition. The results were explained in terms of the rubber-to-metal adhesion and the viscosity of the polymers. © 2001 John Wiley & Sons, Inc. J Appl Polym Sci 82: 1483–1494, 2001

Key words: critical nip gap; mill band-formation indices; front-to-back roll transition; processability

INTRODUCTION

Mill Processability

Milling is one of the important operations in rubber processing. Two-roll mills are used for dispersing fillers and other ingredients in rubber as well as for sheeting and warming up operations of compounded rubber before calendaring and extrusion. The significant advantage of two-roll mills is that it represents an open process to which additives can be easily introduced. In addition, the stock temperature can be held within narrow limits with the cooling arrangement. Rubber formulations must be homogenized with very high shear forces at a controlled temperature to prevent premature crosslinking.

Flow of rubbers during milling is essentially the same as flow between the rotor tip and wall in an internal mixer as well as flow between rotors. Because of the simpler geometry of a mill, the analytic solution to optimization of mixing conditions can be obtained, which enables quantitative estimation of the effect of material and process variables. It is difficult to understand these variables in an internal mixer because of the complex flow geometry. Furthermore, the influence of a single variable can be isolated in model calculations in mill processing, whereas it might be dif-

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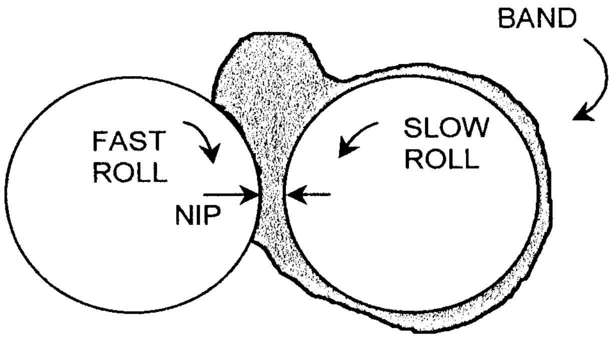

Figure 1 Schematic drawing showing the two-roll mill with banded elastomer.

ficult to alter the variables independently in an internal mixer.

During normal processing operations, the rubber compound is dumped onto a two-roll mill and then banded on the slower front roll for addition and dispersion of ingredients by cutting and folding operations (Fig. 1). Different rubbers and rubber compounds show different milling behaviors depending on their chemical structure, molecular weight and molecular weight distribution of the rubber, particle size and structure of particulate fillers and their amount, and the affinity of the filler for rubber.[1,2] Machine parameters such as speed, friction ratio, nip gap, diameter, length, and temperature of the rolls also influence the milling behavior of a rubber compound.[1,2] Some rubber compounds band well on the slower roll (ideal condition), whereas others either bag or go to the fast roll. Formation of a band on the front roll is desired not only for safety reasons but also for better efficiency of mixing. When a rubber compound does not band on the slower roll and has a tendency to go to the back roll, it delays mixing and disrupts subsequent processing operations, thus lowering productivity. In actual practice, this difficulty is overcome by application of soaps or stearic acid on the back roll. Another way is to optimize machine parameters such as friction ratio, nip gap, and temperature of the rolls so that a smooth band is formed on the front roll and the tendency to go to the back roll is either eliminated or at least minimized.

Several authors analyzed the flow behavior of rubber at the nip area of a two-roll mill.[1–5] Bergen[4] and Pearson[6] published general reviews on the subject. Pasley[5] studied the flow of nonlinear viscoelastic fluids through the mill rolls. Chong[1] and Tokita and White[2] 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 (Fig. 2):

Region 1: The rubber remains mainly on the bank of the mill and only a small quantity falls through the nip gap.
Region 2: The rubber forms a tight elastic band adhering to the roll.
Region 3: The rubber compound is torn and granulated at the nip region, resulting in formation of a loose band (bagging).
Region 4: The 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, that is, the behavior under Region 2.

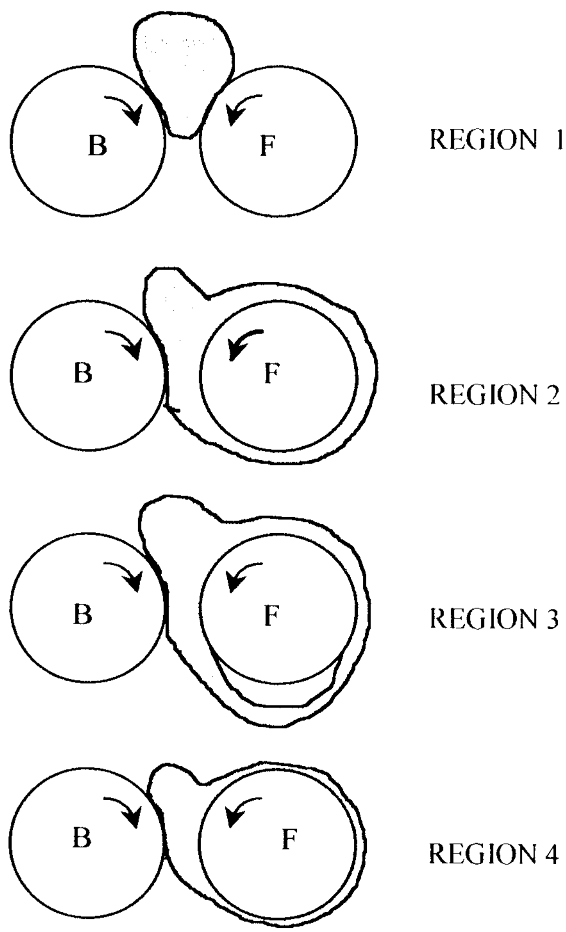

Figure 2 Schematic drawing showing the four different regions of mill behavior.[9] B = back roll; F = front roll.

Tokita[10] also introduced dimensionless groups known as mill band-formation indices (MBI), which describe the milling behavior of a rubber compound with respect to machine parameters such as friction ratio, nip gap, roll diameter, speed, and process time. Hence, the influence of the machine parameters on milling behavior of a rubber compound can be analyzed with MBI. However, in his analyses Tokita simplified the analysis considerably by making certain assumptions. Bhowmick et al.[11–14] extended Tokita's approach by introducing additional variables, thus resulting in an improved understanding of band-formation behavior.

Brominated isobutylene-*co*-paramethylstyrene (BIMS) has recently been marketed by Exxon-Mobil Chemical Co. (Houston, TX). The rubber has superior thermal stability and better cure compatibility with other rubbers. However, the processing behavior of the rubber is not fully understood. Kumar et al.[16] studied the adhesion of BIMS with natural rubber (NR) and ethylene propylene diene terpolymer (EPDM). Waddell and Poulter[17] reported phase mixing of BIMS-based tire tread compound. The tread formulation consisted of BIMS, *cis*-polybutadiene rubber (BR), and polystyrene-*co*-butadiene rubber (SBR) along with reinforcing fillers such as carbon black and precipitated silica. BIMS blends were previously discussed by Potluri et al.,[18] McElrath and Tisler,[19] Kruse et al.,[20] and Flowers et al.[21] These authors discussed specific mixing protocols for obtaining an advantageous morphology.

In the present investigation, the mill band-formation behavior of BIMS and the effect of blending with EPDM was studied over a range of temperatures (30, 50, 70, and 90°C) and friction ratios (1 : 1.2, 1 : 1.5, and 1 : 1.8). Furthermore, the critical nip gap, at which the front-to-back roll (F–B) transition occurs was also determined, using a drop mill operation as described by Tokita.[10] The mill band-formation indices (MBI) and the mill band-formation exponent (MBE) were calculated from the critical nip gap, following the theoretical model of Tokita[10] and subsequently modified by Bhowmick et al.[11,12]

Analyses

Studies by Tokita[10] revealed that a number of parameters influence the band formation behavior in drop mill operations. Because the number of parameters is large, dimensional analysis approach was applied[10–15] and the following dimensionless equation was obtained:

$$\left(\frac{2H_0}{t_pU_-}\right) = k_1\left(\frac{F_at_p}{\eta}\right)^b\left(\frac{Pt_p}{\eta}\right)^c\left(\frac{R_0}{t_pU_-}\right)^d \qquad (1)$$

where $2H_0$ is the critical nip gap at which the front-to-back roll transition occurs; t_p is the process time taken by the rubber in passing through the nip area during the milling operation; U_- is the speed of the slower roll; F_a is the force of adhesion between the rubber and the roll surface; η is the viscosity of the rubber; P is the pressure buildup at the nip gap; R_0 is the radius of the rolls; and k_1, b, c, and d are constants. The process time (t_p) is defined[10] as

$$t_p = 2\,\frac{(2H_0R_0)^{1/2}}{(U_-(f+1))} \qquad (2)$$

where f is the friction ratio defined as the ratio of speed of the front roll to that of the back roll.

In eq. (1), the two factors (R_0/t_pU_-) and ($2H_0/t_pU_-$) contain no material properties, but only machine parameters. Following Tokita[10] and Bhowmick et al.[11–14], substituting for t_p in these expressions one obtains

$$\left(\frac{2R_0}{t_pU_-}\right) = \frac{\left[(f+1)\left(\frac{R_0}{2H_0}\right)^{1/2}\right]}{2} \qquad (3)$$

$$\left(\frac{2H_0}{t_pU_-}\right) = \frac{\left[(f+1)\left(\frac{2H_0}{R_0}\right)^{1/2}\right]}{2} \qquad (4)$$

From the right side of eqs. (3) and (4), the following dimensionless numbers may be defined as

$$N_1 = \left(\frac{2H_0}{R_0}\right)^{1/2}\left(\frac{1}{f+1}\right) \qquad (5)$$

$$N_2 = \left(\frac{2H_0}{R_0}\right)^{1/2}(f+1) \qquad (6)$$

where N_1 and N_2 are known as the mill band-formation indices (MBI), which are of practical importance and provide the information on the conditions for the band formation on the front roll as well as the switchover of the band from the front roll to the back roll during the milling operation. The analysis also shows that the mill band-formation indices (N_1 and N_2) are dependent on critical nip gap, process time, and mill radius.

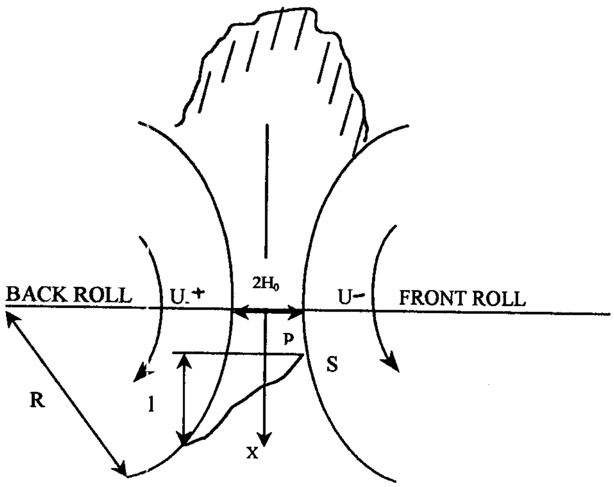

Figure 3 Moment balance at the nip section. U_+ and U_- are the speed of the back and front roll, respectively; $2H_0$ is the critical nip gap; S is the point on the front roll where the band separates from the roll; and l is the difference in length at the roll surface attributed to the difference in the speed of the roll.[10]

Tokita[10] described the mill behavior in terms of the front-to-back roll transition as a consequence of imbalance between the moments attributed to adhesive and shear forces acting at the point of separation of the band from the roll (i.e., point "S" in Fig. 3). The ratio of moments of balance is defined as

$$\frac{M_a}{M_s} = \frac{F_a t_p}{\eta} \tag{7}$$

where M_a and M_s are the moments of balance ascribed to adhesive forces 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:

$$\frac{F_a t_p}{\eta} \begin{cases} =1, \text{ no band formation} \\ >1, \text{ band formation on back roll} \\ <1, \text{ band formation on front roll} \end{cases} \tag{8}$$

The front roll to back roll transition of the band (F–B transition) would take place if the torque acting on the material at the point of separation at the back roll is greater than that acting toward the front roll. Hence, the moment balance at the nip area would provide a necessary condition for the F–B transition using the dimensional analysis approach (Fig. 3).

The adhesive force per unit area is assumed to be directly proportional to the maximum pressure P at the nip during process time and inversely proportional to the separation time τ (the time taken for one revolution of the roll).

Thus,

$$F_a = \frac{K_2 P t_p}{\tau} \tag{9}$$

where K_2 is a dimensionless number and depends on the properties of the material.

Given that $\tau = 2\pi R_0/U_-$, eq. (9) then reduces to

$$F_a = \frac{K_2 P t_p U_-}{2\pi R_0} \tag{10}$$

Therefore, at the transition point, by combining eqs. (8) and (10), one obtains

$$\frac{F_a t_p}{\eta} = (K_2/2\pi)(Pt_p/\eta)(t_p U_-/R_0) = 1 \tag{11}$$

or

$$\frac{Pt_p}{\eta} = (2\pi/K_2)(R_0/t_p U_-) \tag{12}$$

By substituting $F_a t_p/\eta = 1$, the condition for no band formation, and eq. (12) in eq. (1), one gets the following relation for the F–B transition:

$$\left(\frac{2H_0}{t_p U_-}\right) = K_1\left(\frac{2\pi}{K_2}\right)\left(\frac{R_0}{t_p U_-}\right)^c\left(\frac{R_0}{t_p U_-}\right)^d \tag{13}$$

Furthermore, by combining eqs. (4) and (13) one obtains the following expression:

$$\left(\frac{2H_0}{t_p U_-}\right) = K_1\left(\frac{2\pi}{K_2}\right)^c\left(\frac{1}{2}\right)^e\left[\frac{(f+1)^e}{\left(\frac{2H_0}{R_0}\right)^{e/2}}\right] \tag{14}$$

where $e = c + d$.

On simplification, one gets

$$\frac{2H_0}{R_0} = K(f+1)^{2\varepsilon} \tag{15}$$

where

$$K = \left(2K_1\left(\frac{2\pi}{K_2}\right)^c\left(\frac{1}{2}\right)^e\right)^{2/(e+1)} \tag{16}$$

and

$$\varepsilon = \frac{e-1}{e+1}$$

From eq. (15), the following cases may be visualized:

Case I. If ϵ is positive, $(2H_0/R)$ increases with increasing $(f + 1)$, that is,

$$(2H_0/R)/(f+1)^{2\varepsilon} = \text{constant} \tag{17}$$

Case II. If ϵ is negative, $(2H_0/R)$ decreases with increasing $(f + 1)$, that is,

$$(2H_0/R)(f+1)^{2\varepsilon} = \text{constant} \tag{18}$$

Case III. If ϵ is zero, $(2H_0/R)$ is independent of $(f + 1)$, that is,

$$(2H_0/R) = \text{constant} \tag{19}$$

The parameters K and ϵ in eq. (15) are material parameters. These parameters can be considered to be the material characteristics and one of them should be constant for a particular compound. The value of ϵ can be calculated from the slope of $\log(2H_0/R_0)$ versus $\log(f + 1)$ plots and describes the effect of friction ratio on the material behavior. A positive value indicates a tendency for the F–B transition to increase on increasing the friction ratio, whereas a negative value indicates a tendency for the F–B transition to decrease on increasing the friction ratio. It was previously reported[12,13] that ϵ has a positive value for polychloroprene and acrylic rubbers and a negative value for natural rubber and styrene–butadiene rubber (SBR).

EXPERIMENTAL

Materials

Brominated isobutylene-*co*-paramethylstyrene (BIMS-7745, paramethyl 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; diene content, 4–7 wt. %; ML_{1+8} 125°C, 26) were supplied by ExxonMobil Chemical Company.

Preparation of Samples

BIMS and EPDM were passed through a Brabender Plasticorder (Model PLE 330, capacity 65 mL) at 80°C and 60 rpm. 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 also prepared in a Brabender Plasticorder. The mixing time was kept constant (4 min) in all cases.

Mixing Mill

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

Determination of Critical Nip Gap

A procedure similar to the one used by Tokita[10] was followed. About 100 g of the material (after mixing in the Brabender) was dropped vertically on the bank of the roll of the mill. If the material went to the back roll, the nip distance was reset a little wider. This was repeated until the material tended to band on the front roll. At this point, the nip distance was reset slightly narrower. This process was repeated until the material would not band on either roll. This nip gap distance was defined as the critical nip gap ($2H_0$). Analysis of band formation behavior was done above the critical nip gap, that is, the nip gap above which the polymer forms a band on the front (slower) roll.

Determination of Rubber-to-Metal Adhesive Force

The joint strength between rubber and metal was measured using an 180° peel adhesion test specimen. A rubber sheet of 3 mm thickness backed by a cotton fabric was pressed on the metal strip with an application of 35 MPa of pressure. The adhesion test was immediately done at a jaw separation rate of 50 mm/min. The adhesion force (G_a) was calculated using the following formula:

$$G_a = \frac{2F}{w} \tag{20}$$

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

Table I Critical Nip Gap ($2H_0$) (mm) at Different Friction Ratios and Different Temperatures

	30°C			50°C			70°C			90°C		
	Friction Ratio			Friction Ratio			Friction Ratio			Friction Ratio		
BIMS : EPDM	1.2	1.5	1.8	1.2	1.5	1.8	1.2	1.5	1.8	1.2	1.5	1.8
100 : 00	1.60	2.00	2.70	2.05	2.30	3.70	1.65	1.85	2.30	1.85	1.95	2.40
90 : 10	1.50	1.65	1.90	1.85	2.00	2.10	1.55	1.75	1.80	1.65	1.80	1.95
80 : 20	1.20	1.40	1.65	1.30	1.60	1.70	1.50	1.55	1.65	1.55	1.70	1.80
70 : 30	1.10	1.30	1.50	1.10	1.40	1.60	1.45	1.50	1.60	1.45	1.50	1.60
60 : 40	1.05	1.25	1.40	1.00	1.20	1.35	1.25	1.42	1.47	1.35	1.45	1.55
50 : 50	1.00	1.10	1.25	1.00	1.15	1.25	0.90	0.90	1.00	0.97	0.97	1.05
40 : 60	0.85	0.95	1.10	0.90	1.00	1.10	0.65	0.67	0.68	0.65	0.70	0.75
30 : 70	0.60	0.70	0.80	0.80	0.90	1.05	0.55	0.57	0.60	0.57	0.65	0.70
20 : 80	0.50	0.50	0.65	0.47	0.50	0.55	0.45	0.45	0.45	0.47	0.57	0.61
10 : 90	0.45	0.45	0.45	0.40	0.40	0.40	0.40	0.40	0.40	0.40	0.40	0.40
00 : 100	0.40	0.40	0.40	0.40	0.40	0.40	0.40	0.40	0.40	0.40	0.40	0.40

RESULTS AND DISCUSSION

Milling Behavior of BIMS, EPDM, and Their Blends

Table I displays the results of the critical nip of BIMS, EPDM, and their blends at different friction ratios and temperatures. The critical nip gap increased with friction ratio (f) except for 10 : 90 BIMS : EPDM and 0 : 100 BIMS : EPDM blends. This increase lies between 10 and 20% depending on the blend ratio and temperature. For example, the critical nip gap for BIMS changed from 2.05 (at $f = 1:1.2$) to 3.70 (at $f = 1:1.8$) at 50°C. However, there was no particular trend of these values when the temperature of the mill was varied. The $2H_0$ value is lower at higher EPDM content at all temperatures and friction ratios. It is interesting to note that the value of $2H_0$ for EPDM is independent of friction ratio and temperature.

Analysis of band formation behavior was done above the critical nip gap, that is, the nip gap above which the polymer forms a band on the front (slower) roll (friction ratio, 1 : 1.2, Table I). BIMS showed striking changes in its milling behavior on the mill when the temperature of the rolls was raised from 30°C to 90°C. At 30°C and a friction ratio of 1 : 1.2, BIMS showed a milling behavior corresponding to Region 1 (Fig. 2), that is, a nervy wedge of polymer was observed at the bank of the rolls with only strands of polymer moving through the nip gap. These polymer strands were observed to exist on both the slow and fast rolls, although usually predominantly on the slower front roll. On increasing the temperature to 50°C, BIMS formed a very irregular and nervy band, which recoiled after becoming loose from the front roll. This behavior may correspond to the transition zone between Region 1 and Region 2, in which the polymer still has high nerve but undergoes partial deformation. On a further increase of the temperature to 70°C, BIMS formed a tight regular elastic band, which showed much lower nerviness than that observed at 50°C. Also, the band was much smoother and uniform in character than the band formed at 50°C. At 90°C, a band with little or no nerviness, similar to that formed at 70°C, was observed. Changing the friction ratio from 1 : 1.2 to 1 : 1.8 had no effect on the milling behavior of BIMS at different temperatures.

In the case of EPDM, a bagging and crumbing band was observed at all temperatures and friction ratios studied, which corresponded to Region 3 in Figure 2. Also, the tendency to bag increased on increasing the temperature and the friction ratio.

Figure 4 reveals the band formation behavior of various blends. Region 2 (Fig. 2) of mill processing behavior was observed for different blends of BIMS and EPDM containing up to 20 parts of EPDM at all temperatures and friction ratios. For the blends containing EPDM greater than 20 parts but less than 50 parts, a shift from Region 2 to Region 3 (i.e., from tight elastic band to loose band/bagging) was observed either at high friction ratio (friction ratio, 1 : 1.8; temperature, 30°C and above) or at high temperature (temperature, 70°C; friction ratio, 1 : 1.2 and above). For blends containing EPDM greater than 50 parts of

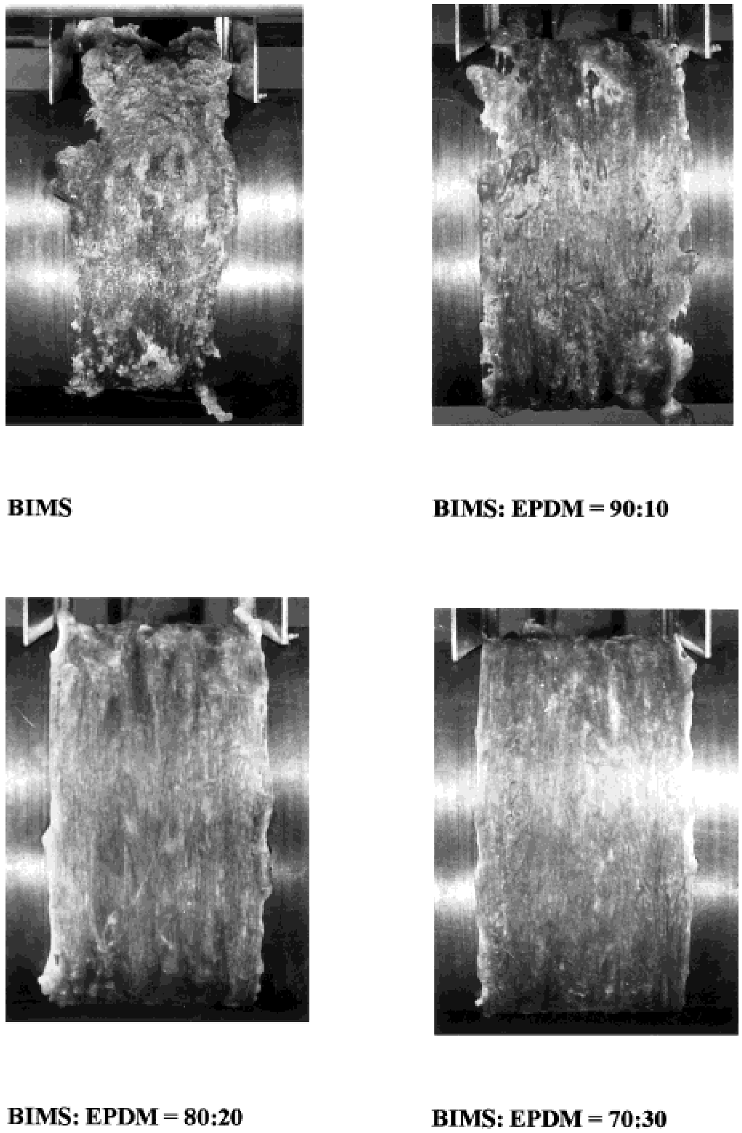

Figure 4 Milling behavior of BIMS and its blends with EPDM at 30°C and friction ratio 1 : 1.2.

EPDM, Region 3 of the mill processing behavior was observed at all temperatures and friction ratios studied. As the EPDM content was further increased, a higher tendency to form a loose band (bagging) was observed (although the band on the front roll became more regular and smooth).

Therefore, the results showed that to improve the processability of BIMS, the EPDM content should be kept below 50 parts.

Mill Band-Formation Indices

Values of the mill band-formation indices (MBI) N_1 and N_2 for BIMS, EPDM, and their blends at different temperatures and friction ratios were calculated using the critical nip gap $2H_0$ (Table I) and eqs. (5) and (6). Figure 5 shows the plots of MBI versus blend composition.

Effect of Blend Composition

As the EPDM content was increased, the values of the critical nip gap and MBI, N_2, decreased, thus indicating a decrease in the tendency of BIMS to go to the back roll with the addition of EPDM (Fig. 5; Table I). In addition, the band on the front roll becomes more regular and smooth (Fig. 4). Furthermore, as soon as 10 parts of EPDM was added, N_2 dropped sharply. For the blends containing EPDM between 10 and 50 parts, the de-

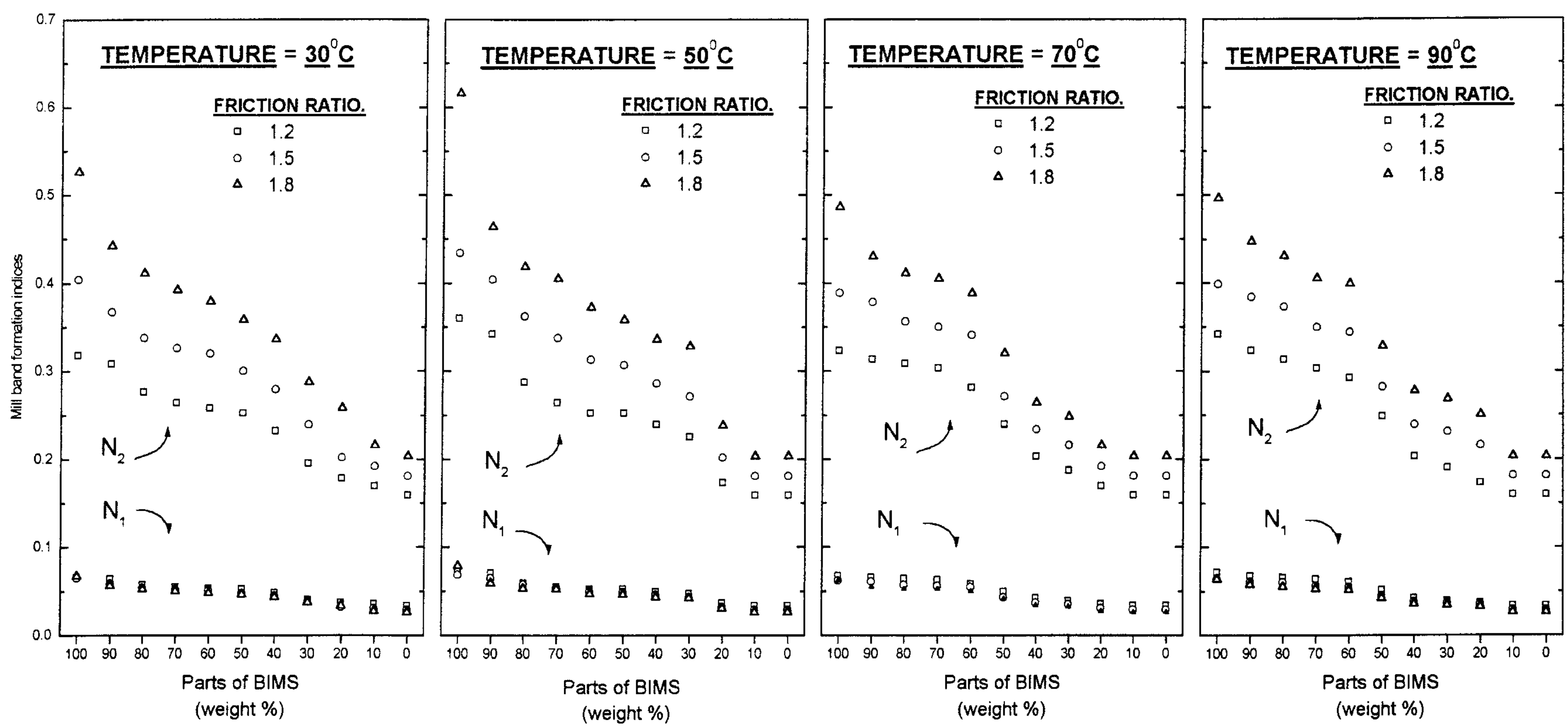

Figure 5 Effect of temperature and friction ratio on mill band-formation indices N_1 and N_2 for BIMS, EPDM, and their blends.

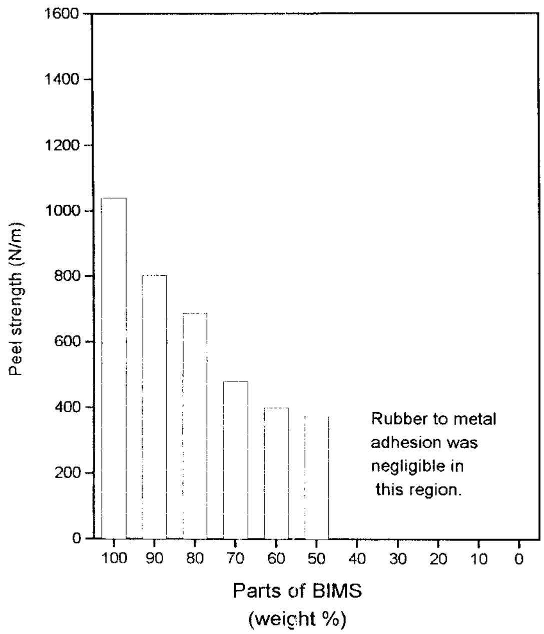

Figure 6 Rubber-to-metal adhesion of different BIMS–EPDM blends.

crease in the critical nip gap as well as N_2 was gradual, showing the dominant behavior of the BIMS phase. However, for blends containing

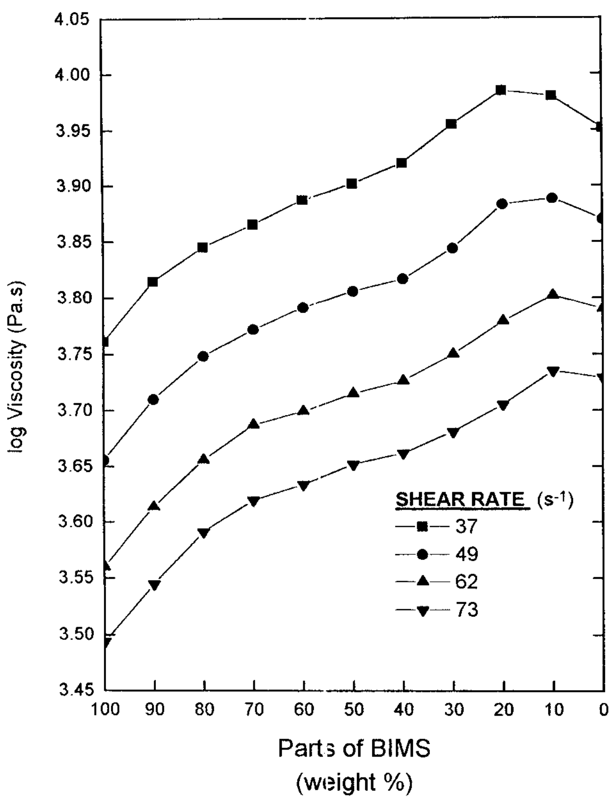

Figure 7 Viscosity of BIMS–EPDM blends.

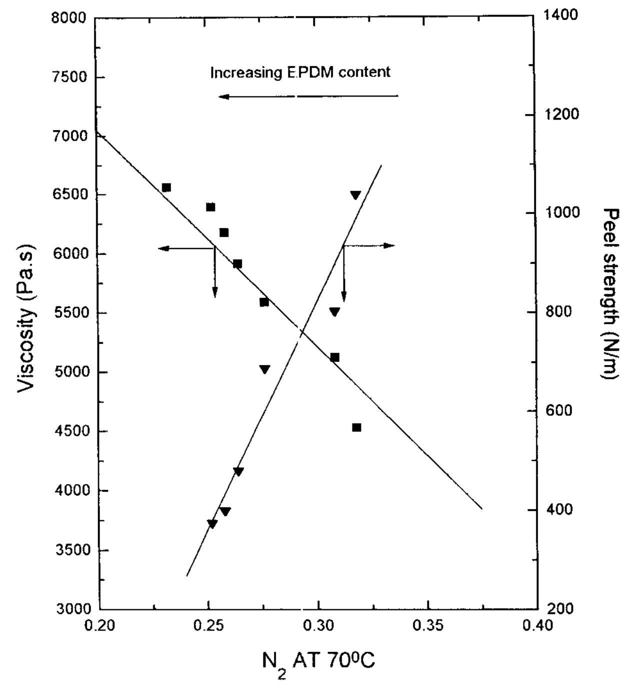

Figure 8 Rubber-to-metal adhesion versus mill band-formation index N_2 and viscosity versus mill band-formation index N_2 at 70°C.

greater than 50 parts EPDM, a sharp decrease in the critical nip gap and N_2 occurred, which is ascribed to the EPDM rubber becoming the major phase. The other MBI, N_1, showed a continuous decrease with the addition of EPDM to BIMS (Table I; Fig. 5).

To explain these results, the adhesion between the rubber and the metal as well as the viscosity of BIMS and its blends with EPDM were measured. It was observed that on increasing the EPDM content, adhesion strength decreased, whereas the viscosity increased (Figs. 6 and 7). This caused an overall decrease of the $F_a t_p/\eta$ or M_a/M_s factor, which in turn decreased the tendency of the material to go to the back roll. As a result the critical nip gap is shifted toward lower values (Fig. 5; Table I). It was also observed that, above 50 parts of EPDM, the blends completely failed to stick to the metal surface (Fig. 6).

A plot between N_2 and peel strength for a variety of BIMS–EPDM blends showed a linear relationship between the two quantities (Fig. 8). Similarly, a plot between N_2 and viscosity for different blends showed a linear relationship (Fig. 8). Thus, it can be concluded that the tendency to go to the back roll for BIMS and it blends with EPDM is related to adhesion with the rolls of the mill and viscosity. These two plots (Fig. 8) also verify eq. (1), as shown by Tokita[10] for the front-to-back roll transition.

Effect of Friction Ratio

For BIMS and its blends with EPDM it was observed that at all temperatures, the critical nip gap and N_2 increased on increasing the friction ratio (Table I; Fig. 5), although N_1 showed no change. The increasing tendency for a front-to-back roll transition with friction ratio can be understood in terms of the interrelationship between the friction ratio and the properties of the material. During the drop mill operation, the point of separation of the polymer at the back roll is more advanced than that at the front roll (distance "l" and point "S" in Fig. 3). This results in greater contact area between the rubber and the metal surface at the back roll compared to that between the rubber and the front roll. This difference increases as the friction ratio is increased. Given that rubber-to-metal adhesion is significant for BIMS and its blends with EPDM (i.e., BIMS content greater than 50 parts) (Fig. 6), the tendency of the material to go to the back roll is enhanced with an increase in the friction ratio. Thus, the system studied is similar to Case I, as defined by Bhowmick et al.,[11–14] who observed that processing would be more facile at lower friction ratios for such materials (Fig. 5; Table I). It is noteworthy that EPDM showed no tendency to back roll. This was shown by its very low value of critical nip gap compared to that of BIMS.

Effect of Temperature

For BIMS the critical nip gap increased on increasing the temperature of the rolls from 30°C to 50°C at $f = 1.2$. However, a further increase in the critical nip gap showed a decrease in value with an increase in temperature to 70°C. In addition, a more regular and smoother band on the front roll was obtained, thus indicating an improvement in processability at 70°C (Table I). A further increase in temperature (90°C) resulted in a slight increase in the critical nip gap, although this value was still lower than the value found at 50°C. A possible explanation could be that with an increase in temperature both the viscosity and the green strength of the polymer decrease. The decrease in viscosity increases the tendency of polymer to wet the metal surface of the rolls, which would enhance the polymer-to-metal adhesion. On the other hand, a decrease in the green strength with temperature tends to decrease rubber-to-metal adhesion. Thus interplay between the viscosity and rubber-to-metal adhesion dictates the processing behavior of elastomers on the mill. Hence, it can be concluded that with an increase in temperature (≤50°C), the adhesion force increases and the viscosity decreases, resulting in an increase of the $F_a t_p/\eta$ factor, which in turn causes shifting of the critical nip gap toward a higher value. At temperatures of 50°C or greater, the adhesive force decreased because of the lowering of green strength, producing a decrease in the value of $F_a t_p/\eta$, which in turn causes a shift of the critical nip gap toward a lower value. However, there is no particular trend observed in the blends because of a lack of information on the surface structure and its associated change with temperature.

An interesting trend was observed from the plot of N_2 versus blend composition at different temperatures. As discussed earlier there is a critical blend composition in the range of 60–40 BIMS content where there is a sudden drop in the value of N_2. This critical value shifted toward the lower BIMS content on increasing the temperature of the mill from 30°C to 50°C. This means that there is an increase in the tendency to back roll. With further increases in temperature (≥70°C), the plot shifts toward higher BIMS content, which indicates a lowering of the tendency for F–B transition (Fig. 5).

Mill Band-Formation Exponent

The value of ϵ for different systems was calculated from the slope of $\log(2H_0/R_0)$ versus $\log(f + 1)$ plots (Fig. 9). The results are summarized in Table II. In the case of BIMS a positive value of ϵ was observed, which corresponds to Case I of mill band-formation analyses. This indicated that the tendency of BIMS to back roll increases on increasing the friction ratio. It was also observed that ϵ showed a maximum at 50°C and subsequent minima at 70°C, which indicated that the tendency to back roll was greatest at 50°C and minimized at 70°C. In the case of EPDM the ϵ value was equal to zero, which indicated that the tendency to back roll was independent of friction ratio. This corresponded to Case III of mill band-formation analyses. In the case of BIMS–EPDM blends, it was observed that at all the temperatures, ϵ decreased with the addition of EPDM, thereby indicating that the tendency to back roll decreases.

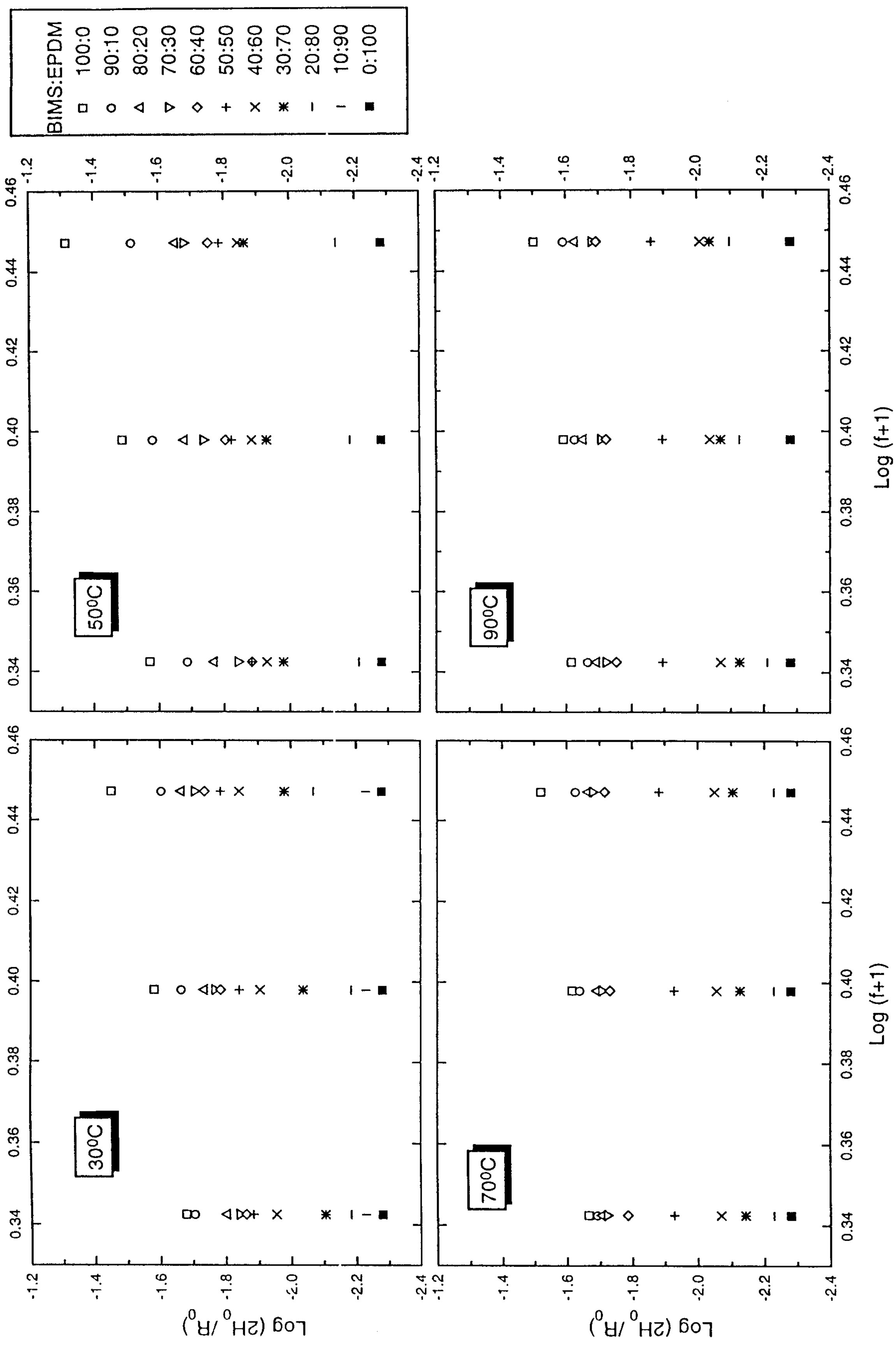

Figure 9 $(2H_0/R_0)$ as a function of $(f + 1)$ for BIMS–EPDM blends at different temperatures.

Table II Mill Band Formation Exponent ε for Different Blends at Different Temperatures

BIMS : EPDM	30°C	50°C	70°C	90°C
100 : 00	1.08	1.21	0.53	0.68
90 : 10	0.78	0.81	0.31	0.35
80 : 20	0.66	0.76	0.20	0.31
70 : 30	0.64	0.78	0.20	0.30
60 : 40	0.60	0.62	0.20	0.29
50 : 50	0.56	0.46	0.21	0.26
40 : 60	0.53	0.46	0.19	0.24
30 : 70	0.59	0.42	0.18	0.23
20 : 80	0.53	0.32	0	0.15
10 : 90	0	0	0	0
00 : 100	0	0	0	0

CONCLUSIONS

In the present investigation the milling behavior of BIMS and its blends with EPDM was investigated over a range of friction ratios and temperatures and the results were analyzed in terms of the theoretical model developed by Tokita[10] and Bhowmick et al.[11–14] In addition, an attempt was made to optimize the mill parameters, to obtain a smooth and regular band. The conclusions from the investigation are as follows:

1. Milling behavior of BIMS changes from a loose nervy band to a tight elastic band, as the temperature of the rolls increased from 30°C to 90°C. For EPDM a loose band was observed at all temperatures and friction ratios studied. For the blends of BIMS and EPDM, the milling behavior changes from a tight elastic band to a loose bagging band on increasing the EPDM content.
2. Addition of EPDM to BIMS resulted in a decrease of the critical nip gap ($2H_0$) and the mill band-formation indices (N_1 and N_2), thus indicating an improvement in processing.
3. The critical nip gap and the mill band-formation index N_2 increased on increasing the friction ratio, whereas N_1 showed no corresponding change in value.
4. For BIMS and its blends with EPDM, the tendency to back roll is related directly to the adhesion strength with the mill roll surface and inversely proportional to the material viscosity.

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