

Mill Behavior of Elastomers: Scale-Up Criteria*

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Synopsis

An investigation into the phenomenon known as "bagging" on a rubber mill was made. Based upon the assumption that bagging is a manifestation of a viscoelastic hydrodynamic instability, a scale-up criterion for the onset of bagging was developed. The derived scale-up method was tested over wide ranges of elastomer compounds and mills and found to hold within the estimated experimental error.

INTRODUCTION

The milling of uncured elastomer compounds is an important operation in the processing of these materials. Although originally intended as a mixing device, the rubber mill is now primarily used as a warm-up and sheet-off device, with mixing functions largely taken over by the internal Banbury mixers. As with most other rubber-processing operations, milling remains more art than science.

A typical rubber mill is sketched in Figure 1. In normal operations a charge of polymer is dumped onto the mill and then banded around the slower of the two rolls. A continuous strip is usually fed off the mill to the next operation. The entire process is done automatically or semiautomatically.

The flow in the nip area has been analyzed by several authors.¹⁻³ The nip area may be loosely defined as that area in which the polymer makes contact with both rolls of the mill. Only Tokita and White⁷ and Chong⁸ have considered, in a meaningful fashion, nonlinear viscoelastic materials, while these two papers and Bergen's work⁵ are the only ones to consider nonequal roll speeds. General reviews of the subject have been given by Bergen⁵ and Pearson.⁹

In this paper we wish to investigate problems that have been dealt with only marginally in previous works. These are the closely related problems of "bagging" and "back rolling." By bagging is meant a looseness of the polymer band on the front (slow) roll. When the bag becomes severe, the band separates from the roll sufficiently so that the automatic knives no longer contact the polymer with their cutting edges and the takeoff strip is

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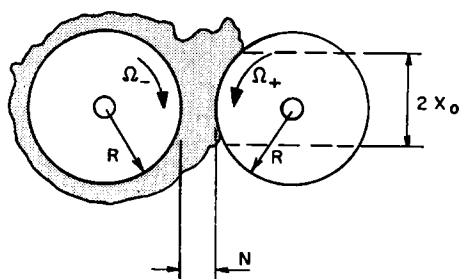


Fig. 1. Typical rubber mill.

lost. If it is to be handled at all, the polymer must then be cut by hand and under bagging conditions this is not considered to be a safe procedure. "Back rolling" occurs when the polymer tears across the length of the roll and then proceeds to band itself on the back (fast) roll.

Many processing plants cannot handle a compound on the back roll either through a lack of automatic cutting and takeoff equipment or, as is frequently the case, the mill is placed with this roll very close to a wall or other equipment and not enough room is available for working a stock on the back roll.

QUALITATIVE OBSERVATIONS

Tokita and White⁷ have observed what they classify as four regions of milling behavior for gum stocks: *region 1*—nervy wedge of polymer in bank; only occasional strands going through nip; *region 2a*—tight elastic band; *region 2b*—severe bagging and crumbling; *region 3*—bagging, tearing; *region 4*—smooth viscoelastic fluid. One progressed from region 4 down through region 1 by lowering the temperature. Regions 2a and 2b represent alternate possibilities and a polymer that entered region 2b would, upon further cooling, powder off the mill. Tokita and White varied only the temperature, using the same mill and keeping the nip setting constant. In our laboratory we have demonstrated the same sequence of behavior at constant temperature by varying the nip setting, going from region 4 to region 1 by narrowing the nip. This result is predictable, as will be discussed in the next section. We have also demonstrated this sequence for compounded elastomers, i.e., raw elastomers with some combination of oil, carbon black, and cure ingredients mixed in.

Since most mixing is now done in large internal mixers and most milling is done with these premixed compounds, we have focused our attention on the milling behavior of these compounds. If such a compound can be kept on the front (slow) roll of a mill, it is usually a simple matter to demonstrate all four milling regions by varying the nip setting. For those compounds that tend to switch to the back roll, however, a different sequence of events is observed. When the nip is narrowed on such a compound, it progresses from region 4 to bagging region 3. In such a case, however,

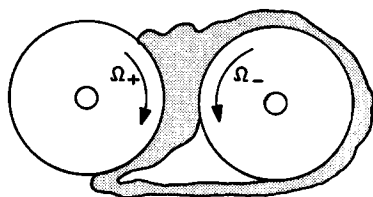


Fig. 2. Polymer bagging on mill.

instead of hanging free, the excess material in the bag is taken up on the back roll, as shown in Figure 2. As the nip is narrowed further, the material in the bag will be stretched until it reaches a point where a tear initiated in the bagging region will propagate across the width of the polymer strip. The polymer will then wind itself onto the back roll until all the material is banded on this roll and the front roll is left clean. The subsequent band on the back roll is indistinguishable in appearance from what we would call region 4 on the front roll. Indeed, if such a compound is initially banded on the back roll, it will maintain a region 4 appearance until region 1 is reached. The improved milling behavior of polymers on the back roll of a rubber mill has been previously reported by Staib.¹⁰ Those compounds with low tear strength will not exhibit bagging behavior at all, but will proceed directly from region 4 behavior to a band on the back roll.

It should be reemphasized here that not all compounds exhibit this roll switching behavior. There are many compounds that, when banded on the front roll, will proceed through the four regions of mill behavior and remain on the front roll.

All of the above information tends to support the hypothesis of Tokita and White that the transition from region 4 to region 3 is a viscoelastic hydrodynamic instability. This assumption will be taken as a working hypothesis in the material to follow.

THEORETICAL ANALYSIS AND DEVELOPMENT OF SCALE-UP CRITERIA

We now wish to investigate, in detail, the region 4–region 3 transition. One method of attack is to attempt the solution of the equations of motion and continuity for the fluid in the nip area. Since we have hypothesized this phenomenon to be a viscoelastic instability we must use a properly invariant viscoelastic constitutive relationship. Such an approach has been attempted previously using a second-order Rivlin-Ericksen fluid model,⁷ a three-constant Oldroyd model,⁸ and a modified second-order Rivlin-Ericksen model.⁸ None of these solutions has proceeded beyond the Newtonian terms obtained by Bergen.⁵

Rather than make another attempt at this type of solution, we apply the techniques of dimensional analysis. We assume that the fluid in question possesses a characteristic time s_0 and a characteristic viscosity μ_0 . The

process itself is characterized by a velocity V , length L , and time t_p . To complete the list of variables we add the fluid density ρ , velocity vector \mathbf{v} , modified pressure,

$$P = p + \rho\phi \quad (1)$$

where we have assumed that the external body force vector per unit mass \mathbf{f} , is representable by the gradient of a potential ϕ ,

$$\mathbf{f} = -\nabla\phi, \quad (2)$$

extra stress tensor,

$$\mathbf{T}_E = \mathbf{T} + p\mathbf{I}, \quad (3)$$

and the time t .

Application of the Buckingham pi-theorem to this list of variables then yields the following dimensionally invariant description of the process:

$$\frac{\mathbf{v}}{V} = f\left(\frac{t}{t_p}, \frac{s_0}{\mu_0} \mathbf{T}_E, \frac{s_0}{\mu_0} P, N_{Re}, N_{Wi}, N_{De}\right) \quad (4)$$

where the Reynolds number, Weissenberg number,^{11,12,13,15,22,23} and Deborah number^{14,15,22-25} are given by

$$N_{Re} = \frac{\rho LV}{\mu_0}, N_{Wi} = \frac{s_0 V}{L}, N_{De} = \frac{s_0}{t_p}. \quad (5)$$

It may be constructive to note here that there is no essential difference between the Deborah and Weissenberg numbers since L/V can always be considered as a second process characteristic time t_{p2} . Thus a fluid with n characteristic times undergoing a process with m independent characteristic times will require $(n+m-1)$ independent time ratios in addition to a dimensionless chronological time to provide a dimensionally invariant description of the process. These time ratios may be expressed as Deborah numbers. Thus, for example, a process with three characteristic times, t_{p1} , t_{p2} , and t_{p3} , operating on a fluid with three characteristic times, s_1 , s_2 , and s_3 , could be described by the five Deborah numbers

$$N_{De1} = \frac{s_1}{t_{p1}}, N_{De2} = \frac{s_1}{t_{p2}}, N_{De3} = \frac{s_1}{t_{p3}}, N_{De4} = \frac{s_2}{t_{p1}}, N_{De5} = \frac{s_3}{t_{p1}}. \quad (6)$$

It is, therefore, constancy of parameters of this form that should provide scale-up criteria for viscoelastic processes.

Needless to say, the Buckingham pi-theorem yields an infinity of equivalent dimensionally invariant descriptions of a process. Those dimensionless groups selected for eq. (4) were chosen for their familiarity, utility, and ease of interpretation. A careful nondimensionalizing of the equations of motion and continuity will, of course, yield equivalent information.¹⁵

USE OF SCALE-UP CRITERIA FOR PREDICTING THE REGION 4-REGION 3 TRANSITION

From our basic assumption of a viscoelastic instability for the region 4-region 3 transition and from the results of the previous section, we conclude that this phenomenon should be scalable by a Deborah number criterion.

For our characteristic process time we choose a measure of the time that the polymer spends in the nip area, i.e., from the point where the polymer band first makes contact with both rolls to the point where it separates from one of the rolls. Referring to Figure 1, the distance involved is given as $2x_0$. Experimental results of Gaskell, as quoted by Pasley⁶ and Finston,¹⁶ indicate that

$$x_0 \cong \frac{1}{2}(RN)^{1/2}. \quad (7)$$

Pearson⁹ shows clearly how a grouping of this functional form arises out of purely geometrical considerations. If we then take the average of the surface velocities of the two rolls as our velocity measure, we may define a characteristic process time

$$t_p = \frac{2x_0}{V} = \frac{(RN)^{1/2}}{2\pi R \frac{(\Omega_+ + \Omega_-)}{2}} = \frac{1}{\pi(\Omega_+ + \Omega_-)} \left(\frac{N}{R}\right)^{1/2} \quad (8)$$

The choice of a characteristic time of the fluid is less obvious. An incompressible Noll simple fluid¹⁷ is defined by

$$\mathbf{T}_E = \frac{\mu_0}{s_0} \overset{\infty}{\underset{\sigma=0}{\mathbf{H}}} (\mathbf{C}_{(t)} [t - s_0\sigma]) \quad (9)$$

where $\mathbf{C}_{(t)}$ is the right relative Cauchy-Green strain tensor and $\overset{\infty}{\underset{\sigma=0}{\mathbf{H}}}$ is a dimensionally invariant tensor-valued functional. Slattery¹⁵ points out that, for a series of fluids which exhibit the same functional $\overset{\infty}{\underset{\sigma=0}{\mathbf{H}}}$, any definition of a characteristic time appropriate to the fluid will yield a consistent correlation. Indeed, this will be true for any series of fluids whose properly invariant constitutive equations exhibit the same functional dependency.

In the absence of specific knowledge of the forms of the constitutive equations, however, one must choose a characteristic time which is appropriate not only to the fluid, but also to the process. Many possibilities for such a choice exist. Truesdell¹⁸ has proposed a natural time based on normal stress effects. In the context of the milling operation, Tokita and White⁷ have proposed the use of the maximum relaxation time as given by Tobolsky.¹⁹ Bird^{20,21} proposes a characteristic time involving only the viscosity function which, however, seems inappropriate to this process.

It is not yet apparent what the proper choice of a characteristic time of the fluid is for this operation. Fortunately, for an engineering application

of these concepts this information is not critical. If both the polymer used and the temperature of operation are maintained constant, the scale-up criterion becomes merely the constancy of the characteristic time of the process, t_p , as given by eq. (8). In order to be able to use a single such criterion for this process, one must also maintain the friction ratio f , constant, where

$$f = \Omega_+/\Omega_- \quad (10)$$

EXPERIMENTAL RESULTS AND DISCUSSION

Four mills and numerous polymer compounds have been used in testing eq. (8). The characteristics of the mills used are given in Table I. A typical set of data is given in Table II. These data were collected at 85°C on a sample of an ethylene-propylene terpolymer extended with 50 parts of carbon black and 75 parts of oil for each 100 parts of elastomer.

TABLE I
Mill Characteristics

Mill roll diam., in.	Ω_- , rpm	Ω_+ , rpm	f
3	16	22.3	1.39
4	18	25	1.39
6	19.7	26	1.32
8	Variable	Variable	1.40

TABLE II
 t_p for the Region 4-Region 3 Transition

Mill roll diam., in.	t_p , sec
3	0.079
3	0.075
4	0.069
4	0.073
4	0.074
6	0.063
6	0.061
8 ($\Omega_+ = 30$ rpm, $\Omega_- = 21.4$ rpm)	0.070
8 ($\Omega_+ = 40$ rpm, $\Omega_- = 28.6$ rpm)	0.068
8 ($\Omega_+ = 40$ rpm, $\Omega_- = 28.6$ rpm)	0.073
Average	0.070

In general, all sets of data showed a scatter around an average value of approximately $\pm 15\%$. Characteristic times varied from 0.045 sec to 0.125 sec. Samples studied included several different compounds of three ethylene-propylene terpolymers, one styrene-butadiene rubber (SBR 1500), and one fluoroelastomer (Viton A).

While mill roll sizes and speeds and nip settings could be measured with a great deal of accuracy, there is a good deal of uncertainty in determining the precise point of the region 4–region 3 transition. This determination depends upon a subjective judgment and, as such, is subject to a great deal of experimental uncertainty. It is estimated that the determination of nip setting at this transition point is no better than $\pm 15\%$.

In summary, we may say that the data, despite their fairly large scatter, do tend to support the theory and that a method for prediction of one aspect of industrial milling behavior has been developed. The author is in complete agreement with others^{13,15} who believe that the techniques of dimensional analysis hold great potential for application to a wide range of polymer processing operations. This investigation has covered only one small aspect of a field which has been largely neglected from the viewpoint of rational mechanics.

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References

1. G. Ardichvili, *Kautschuk*, **14**, 23 (1938).
2. D. D. Eley, *J. Polym. Sci.*, **1**, 529 (1946).
3. R. E. Gaskell, *J. Appl. Mech.*, **17**, 334 (1950).
4. J. M. McKelvey, *Polymer Processing*, Wiley, New York, 1963.
5. J. T. Bergen, in *Processing of Thermoplastic Materials*, E. C. Bernhardt, Ed., Reinhold, New York, 1959.
6. P. R. Pasley, *J. Appl. Mech.*, **24**, 602 (1957).
7. N. Tokita and J. L. White, *J. Appl. Polym. Sci.*, **10**, 1011 (1966).
8. J. S. Chong, *J. Appl. Polym. Sci.*, **12**, 191 (1968).
9. J. R. A. Pearson, *Mechanical Principles of Polymer Melt Processing*, Pergamon Press, New York, 1966.
10. J. H. Staib, *Rubber Age*, 374 (1965).
11. J. L. White, *J. Appl. Polym. Sci.*, **8**, 2339 (1964).
12. A. B. Metzner, J. L. White, and M. M. Denn, *A.I.Ch.E. J.*, **12**, 863 (1966).
13. A. B. Metzner, J. L. White, and M. M. Denn, *Chem. Eng. Progr.*, **62**, 81 (1966).
14. M. Reiner, *Physics Today*, **17**, 62 (1964).
15. J. C. Slattery, *A.I.Ch.E. J.*, **14**, 516 (1968).
16. M. Finston, *J. Appl. Mech.*, **18**, 12 (1951).
17. C. Truesdell and W. Noll, *Handbuch der Physik*, III/3, p. 81. Springer-Verlag, New York, 1965.
18. C. Truesdell, *Phys. Fluids*, **7**, 1134 (1964).
19. A. V. Tobolsky, *Properties and Structure of Polymers*, Wiley, New York, 1960.
20. R. B. Bird, *Chem. Eng. Progr. Sympos. Series*, **61**, 86 (1965).
21. R. B. Bird, *Can. J. Chem. Eng.*, **43**, 161 (1965).
22. G. Astarita, *Ind. Eng. Chem., Fundam.*, **5**, 548 (1966).
23. *Ibid.*, **6**, 257 (1967).
24. G. Astarita, *Can. J. Chem. Eng.*, **44**, 59 (1966).
25. G. Astarita, *Ind. Eng. Chem., Fundam.*, **4**, 354 (1965).

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