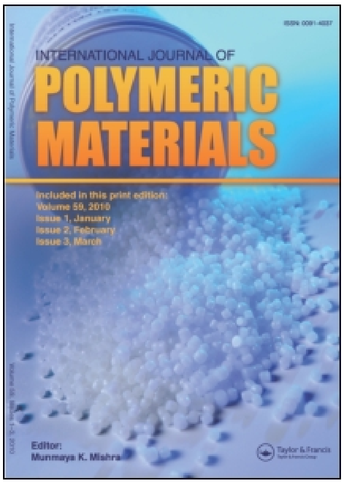

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Die-Swell of Mica-Filled Styrene-Butadiene Rubber Compounds

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Die-Swell of Mica-Filled Styrene–Butadiene Rubber Compounds

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Die-swell of mica-filled styrene-butadiene rubber has been correlated with the recoverable deformation, elastic energy and shear modulus of the melt at different shear-rate and at various length to diameter ratio of the capillary. The Reynolds number and power consumption of melt flow have been correlated with the die-swell and shear-rate of the melt flow. The inverse relationship between shear-viscosity and shear-rate will also correlate power consumption with shear viscosity.

Keywords: Mica, Die-swell, Recoverable deformation, Elastic energy, Reynolds number and Power consumption.

1. INTRODUCTION

The ratio of extrudate diameter (after recovery) to that of the capillary (die) diameter is known as the die-swell. The die-swell is the consequence of the elastic recovery when the extrudate comes out of the die.

Consideration of die-swell is very important from engineering stand point and most crucial for items having non-symmetrical cross-section and where specifications are to be satisfied within close tolerances. The effect of compounding, temperature,[1] length to diameter ratio,[2] shear-rate,[3] molecular weight and molecular weight-distribution[4] on die-swell of visco-elastic materials have already been studied for different polymeric systems. Dannenberg *et al.*[5] found die-swell as a measure of carbon black structure. Die-swell of black-filled rubber compounds could be correlated with the structure and dibutyl phalate absorption of carbon blacks.[6]

The flow behaviour of polyolefins filled with mica-powder as well as mica flakes having high aspect ratio has been reported by several authors.[7-9] Scrutiny of the available literature reveals that no work has been reported on the flow-behaviour and die-swell characteristics of mica-filled elastomeric system. Das *et al.*[10] correlated die-swell with recoverable deformation, elastic energy and relaxation time.

The previous papers[11-12] in this series deal with the viscosity and die-swell of mica-filled styrene-butadiene rubber. This paper deals with the correlation between the die-swell and various parameters like recoverable deformation, recoverable elastic energy, shear-modulus of the melt and Reynolds number of melt flow. Power consumption of melt flow has been correlated with the shear-rate. The relationship between shear-rate and viscosity will also correlate power consumption and viscosity.

2. EXPERIMENTAL

2.1. Materials used

The materials characteristics of the experiments and formulations of the mixes are shown in Table I.

TABLE I
Formulations of the compounds

Mix no.	C_{00}	C_{10}	C_{40}
SBR-1502[a]	100.0	100.0	100.0
Zinc Oxide[b]	—	5.0	5.0
Stearic Acid[b]	—	2.0	2.0
Mica[c]	—	10.0	40.0

[a] Styrene-butadiene rubber containing 23% bound styrene, Manufactured by Synthetics & Chemicals Ltd., Bareilly, India.
[b] Zinc oxide and stearic acid are of commercial grade.
[c] 3-micron muscovite mica, supplied by Mica Trading Corporation of India, Patna, India.

2.2. Preparation of the compounds

Mixing was done on an open two-roll mill with a nip gap of 2.5×10^{-3} m and at a friction ratio of 1:1.10 at 70°C. The mixing parameters (nip gap, temperature and time) were kept constant from mix to mix so that the change in viscosity is same for all the compounds.

2.3. Rheological measurements

The details of viscosity measurements with capillary Rheometer model 3210 and calculations with and without Bagley Corrections[13–14] have already been described elsewhere.[11]

2.4. Determination of die-swell of extrudate

During extrusion when the shear-stress attains equilibrium value for any shear-rate, the corresponding extrudate is allowed to support on an aluminium foil to avoid the gravity effect and mechanical damage out of handling. In most cases, particularly at higher shear-rates ($>20\,s^{-1}$), the extrudate surface becomes rough, average extrudate diameter was obtained by weighing a known length of extrudate and the knowledge of its density.

2.5. Calculations of parameters

Recoverable deformation is directly related to the die-swell of the extrudate and is obtained with the help of the Eq. (1).

$$v_r = 0.707[C^{-1}(\alpha^4 + 2\alpha^{-2} - 3)]^{1/2} \tag{1}$$

where, α = die-swell, d/D_0 and C is defined by

$$C = \frac{3(n'^4) + 1}{4(5n' + 1)}$$

when n' is flow behaviour index of the melt.

Recoverable elastic energy is related to the recoverable deformation of the extrudate. It is calculated from Eq. (2).

$$E = Cv_r\tau \tag{2}$$

where τ = shear-stress and C and v_r are as defined previously.

Shear-modulus of the melt, Reynolds number and power consumption of melt flow are defined by Eqs. (3), (4) and (5) respectively.

$$\text{Shear-modulus } (G) = \frac{\tau}{v_r} \tag{3}$$

$$\text{Reynolds number } (NR_e) = \frac{D_0 U \rho}{\mu} \tag{4}$$

$$\text{Power Consumption } (W) = U \times \mu \times v \times A \tag{5}$$

where D_0 = diameter of the capillary, U = velocity of the melt in the capillary, ρ = density of the melt, μ = viscosity of the melt, v = shear-rate at the wall and A = surface area of the capillary.

3. RESULTS AND DISCUSSIONS

3.1. Effect of L_0/D_0 ratio of capillary and mica concentration of the melt

Die-swell as a function of shear-rate for various length to diameter ratio of the capillary and at a temperature of 120°C are shown in

TABLE II

Die-swell of various length to diameter ratio of the capillary at 120°C

Shear rates s^{-1}	Mix. no.	Die-swell L_0/D_0 ratio 20.0 C_{00}	C_{10}	C_{40}	40.0 C_{00}	C_{10}	C_{40}
2.97		1.50	1.40	1.20	1.44	1.38	1.17
11.86		1.52	1.45	1.20	1.48	1.40	1.15
29.86		1.55	1.50	1.23	1.52	1.45	1.21
118.60		1.60	1.53	1.27	1.57	1.52	1.25
296.60		1.65	1.60	1.31	1.62	1.57	1.29
593.30		1.68	1.63	1.37	1.65	1.61	1.30
1186.50		1.75	1.73	1.42	1.73	1.70	1.38
2963.30		1.81	1.80	1.46	1.80	1.81	1.43

TABLE III
Die-swell at various length to diameter ratio of capillary at 120°C

Shear rare s^{-1}	Mix. no.	Die-swell L_0/D_0 ratio 33.3 C_{00}	C_{10}	C_{40}	66.7 C_{00}	C_{10}	C_{40}
1.72		1.46	1.40	1.19	1.44	1.38	1.16
6.864		1.50	1.45	1.17	1.49	1.41	1.15
17.2		1.55	1.47	1.22	1.52	1.44	1.22
68.60		1.60	1.53	1.26	1.55	1.52	1.25
171.6		1.65	1.58	1.30	1.65	1.56	1.29
343.2		1.68	1.62	1.35	1.68	1.60	1.30
686.4		1.74	1.72	1.40	1.71	1.70	1.38
1716.0		1.81	1.81	1.45	1.80	1.80	1.47

Tables II and III. As expected die-swell increases with shear-rate,[3] because the higher deformation at higher shear-rate induces faster relaxation rate and higher percentage of recovery of original elastic stress. The decrease of die-swell with mica concentration is associated with the reduced flexibility and recovery.[5] It is also clear from Tables II and III that die-swell decreases with length to diameter ratio of the capillary. This has been attributed to the higher residence time of the melt in the capillary. At higher residence time in the capillary, relaxation rate becomes faster (due to higher temperature) and a greatest part of elastic stress decreases within the capillary and reduces the die-swell.

3.2. Effect of recoverable deformation of melt

Figure 1 shows the die-swell as a function of recoverable deformation (at various shear-rate) of the extrudate for $L_0/D_0 = 40.0$ and at a temperature of 120°C. The die-swell is a qualitative manifestation of the recoverable deformation of the melt after the deforming force was withdrawn. More and more is the recoverable deformation more is the die-swell.[10] It is clear from Figure 1 that recoverable deformation decreases with filler loading. The filler acts as hindrance to the mobility of the polymer chains and the percentage recovery of deformation decreases. The decreased

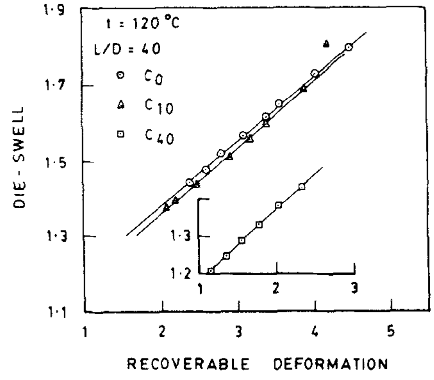

FIGURE 1 Plots of die-swell as a function of recoverable deformation of the melts for $L_0/D_0 = 40.0$ and at a temperature of 120°C.

percentage recovery of the deformation accounts for the decreased die-swell of the filled compounds.[5]

3.3. Effect of recoverable elastic energy of melt

It has been shown that recoverable elastic energy is a function of recoverable deformation. It is also known that relaxation of elastic stress (normal and radial) causes die-swell of non-Newtonian viscoelastic fluid.[15–16] Hence, die-swell increases with the increase of recoverable elastic energy (Figure 2). Moreover, the same figure shows that at any shear-rate (the points are at different shear-rates, varying from 1.72 to $2966.0\,\mathrm{sec}^{-1}$) recoverable elastic energy decreases with filler loading and consequently the die-swell.

3.4. Effect of shear modulus of melt

Figure 3 shows plots of die-swell as a function of shear modulus of the melt at $L_0/D_0 = 40.0$ and at a temperature of 120°C. Shear-modulus passes through a maximum with die-swell and then decreases. This decrease is possibly attributable to the yielding phenomenon. Beyond the maximum shear-modulus die-swell increases at a faster rate than before the maximum. Similar observation was also found in case of die-swell shear-rate plots where

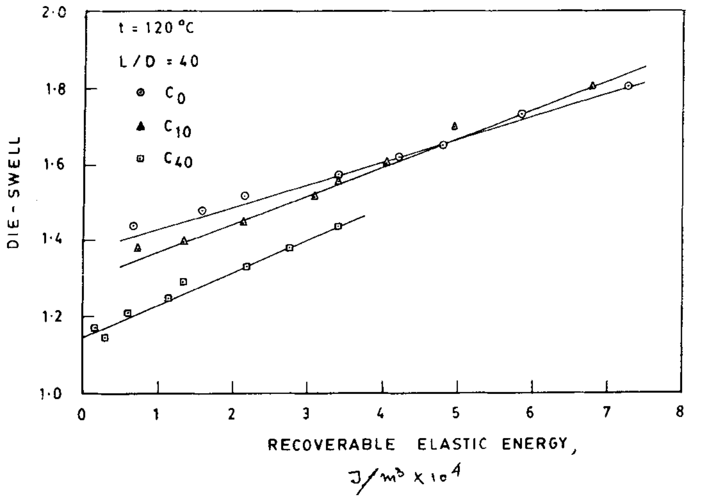

FIGURE 2 Plots of die-swell as a function of recoverable elastic energy of the melt for $L_0/D_0 = 40.0$ and at a temperature of 120°C.

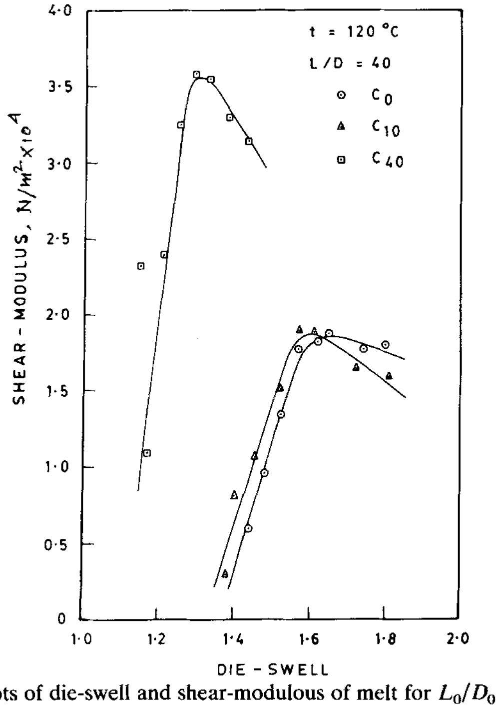

FIGURE 3 Plots of die-swell and shear-modulous of melt for $L_0/D_0 = 40.0$ and at a temperature of 120°C.

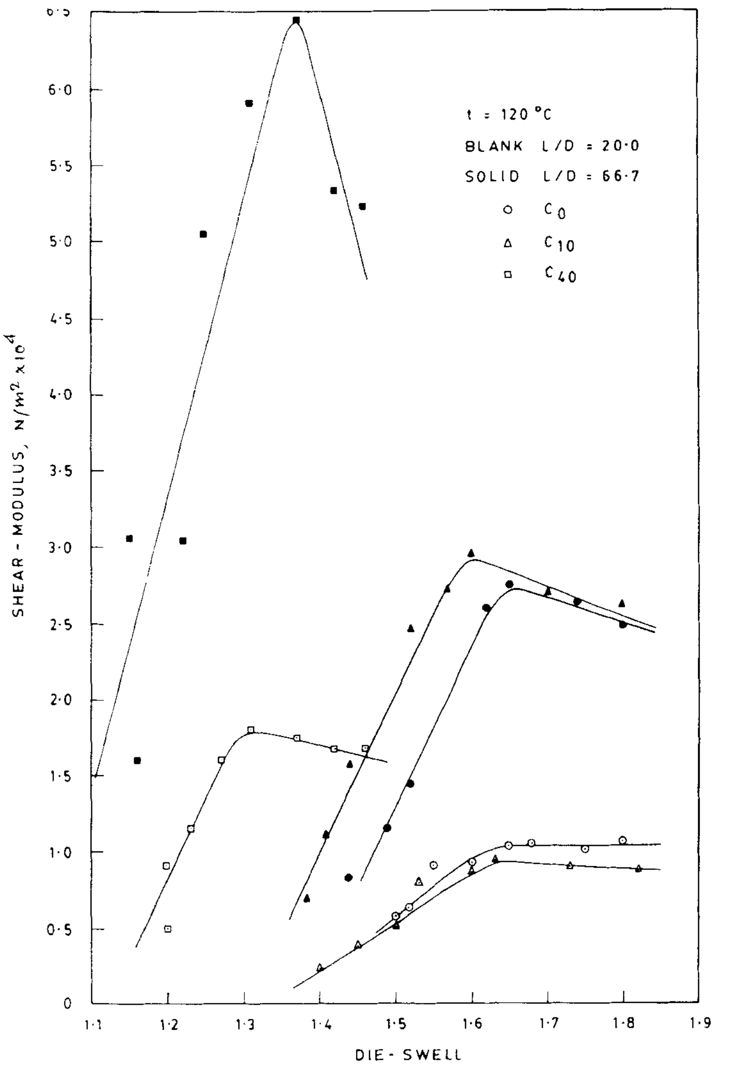

FIGURE 4 Plots of die-swell and shear-modulous at a temperature of 120°C for various length to diameter ratio of the capillary.

die-swell registers a faster increment beyond a certain shear-rate.[12] The decrease of die-swell with mica incorporation is due to the higher shear-modulus of the melt which imposes a resistance against recovery and hence die-swell.

In Figure 4 shear-modulus and die-swell have been plotted at different length to diameter ratio of the capillary at 120°C for various concentration of mica in the melt. Special feature to note here is the very rapid increase of shear-modulus at very high L_0/D_0 ratio (66.7), however, die-swell does not decrease accordingly.

3.5. Effect of Reynolds number (NR_e)

Die-swell has been correlated with viscosity and shear-rate. Reynolds number of fluid is a function of viscosity and velocity. So, correlation of die-swell with Reynolds number will include the combine effects of viscosity and shear-rate (shear-rate is a function of velocity). Figure 5 shows that die-swell increases with the increase of Reynolds number. Probably this higher die-swell at higher Reynolds number is associated with the transition between the stream-line and turbulent flow of the melt, though the concept of critical Reynolds number is completely different for polymer melt than in case of simple liquids.[17–18]

Die-swell-Reynolds number plots show a break of continuity at a Reynolds number of 10^{-6} to 10^{-5}. This might be due to the existence of a critical Reynolds number. On either side of this critical value the rate of change of die-swell changes with the change of Reynolds number.

3.6. Power consumption

Figure 6 shows the plots of power consumption of the plunger as a function of shear-rate for different length to diameter ratio of the capillary at 120°C. Power consumption increases directly with the increase of shear-rate. At any constant shear-rate mica-generally increases the power consumption. Length to diameter ratio also increases the power consumption. However at very high L_0/D_0 ratio (66.7) the difference between the mica-filled and unfilled systems becomes very insignificant. This is possibly due to the similarity of viscosity of unfilled and mica-filled systems for capillary having very

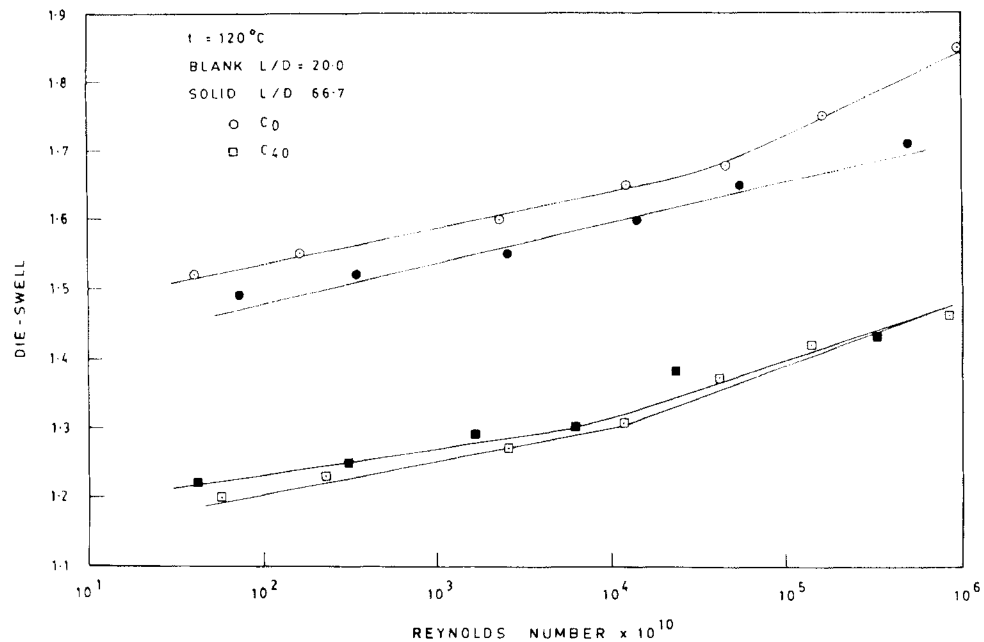

FIGURE 5 Plots of die-swell as a function of Reynolds number of flow for $L_0/D_0 = 20.0$ and 66.7 and at a temperature of 120°C.

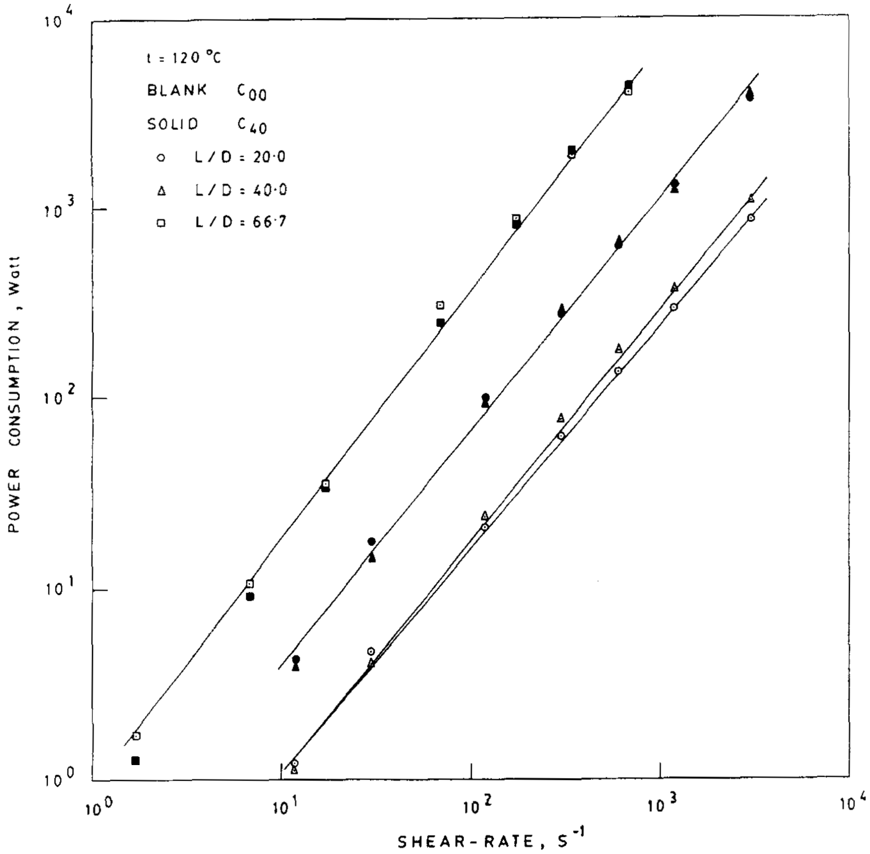

FIGURE 6 Plots of power consumption as a function of shear-rate at a temperature of 120°C for various length to diameter ratio of the capillary.

high L_0/D_0 ratio.[11] From this analogy and with the inverse relationship of viscosity—shear-rate, it can be said that power consumption increases with the decrease of viscosity when viscosity decreases by higher shear-rate only. Apparently it seems to be paradoxical. The exact explanation is that the increase in energy requirement for the generation of higher shear-rate is more than the savings in energy requirement due to decreased viscosity. However, if viscosity decreases due to thermal effects (at higher temperature) then obviously power consumption will be lower at lower viscosity. In the present system viscosity is very insensitive to the thermal effects, at least in the temperature range studied (90–120°C), which restricts us to prove the supposition at constant shear-rate and high temperature.

The correlation among the power consumption, shear-rate and length to diameter ratio of the capillary suggests for optimum shear-rate and L_0/D_0 ratio of the capillary for optimum power consumption.

CONCLUSIONS

The salient features of this article are

1) Die-swell increases with the increase of recoverable deformation of melt.

2) Die-swell increases also with the increase of recoverable elastic energy of the melt.

3) Die-swell decreases with the rise of shear modulous of melt. Shear-modulous shows a maximum with shear-rate, beyond which it decreases again. It might be related to the yielding of the melt.

4) Die-swell increases with the rise of Reynolds number of melt flow.

5) Power consumption increases with the rise of shear-rate. Shear-rate being inversely related to the viscosity, power consumption increases with the decrease of viscosity of the melt when viscosity is decreased by increased shear-rate only.

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