

# Blending Characteristics and Viscoelastic Properties of Plastic Composites

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## Abstract

An experimental study was carried out on the blending process of thermoplastics, mixed with various fillers. Many characteristics on blending torque versus time relation were obtained using a Brabender plastograph, and then dimensionally analysed, where the temperature, weight percentage of the filler, length of the fibrous filler and the blending time were changed as the principal variable quantities.

Furthermore, the effect of the filler on die swell was studied, because die swell demonstrates clearly the viscoelastic property of the thermoplastic melt.

## 1. Introduction

Although the variety of polymers is considerable, as far as physical or chemical properties are concerned, it is often difficult to meet a particular specification with any single material. However, in such a case, it may be possible to find a material better suited for a given specification by designing a plastic composite, in which some kind of plastic is combined with another material.

Where such a plastic composite is used, we find that although the properties of the final product under real conditions of application are important for the user, rheological properties exhibited during the forming process of the product are very important for the processor.

For testing the rheological properties, the Brabender plastograph is convenient.

We applied the Brabender plastograph to study the blending characteristics of thermoplastic mixed with fibrous fillers.

During the forming process, the plastic melt exhibits various problematic tendencies caused by its viscoelastic property.

For testing the viscoelasticity, it is best to measure die swell by extruding the plastic melt through a screw extruder die, since die swell is one of the best representative phenomena exhibited by viscoelastic material.

We applied a screw extruder to study the effect of filler added to the plastic melt on the viscoelasticity by measuring die swell.

## 2. Blending Characteristics of Plastic Composites by the Brabender Plastograph

The Brabender plastograph is an experimental apparatus for testing blending characteristics of pasty or sticky substances. It records automatically a characteristic curve

representing a relation between the torque and the blending time as shown in Fig. 1. This figure shows a case where polyethylene pellet material was blended at 150°C. At first the torque increases rapidly until it reaches the maximum value, during the time the pellets are melted by heat transfer and stick to each other, increasing the size of the lumps. After getting over the summit, the torque gradually decreases until it reaches asymptotically the stable value during which time the material becomes well blended increasing its homogeneity.

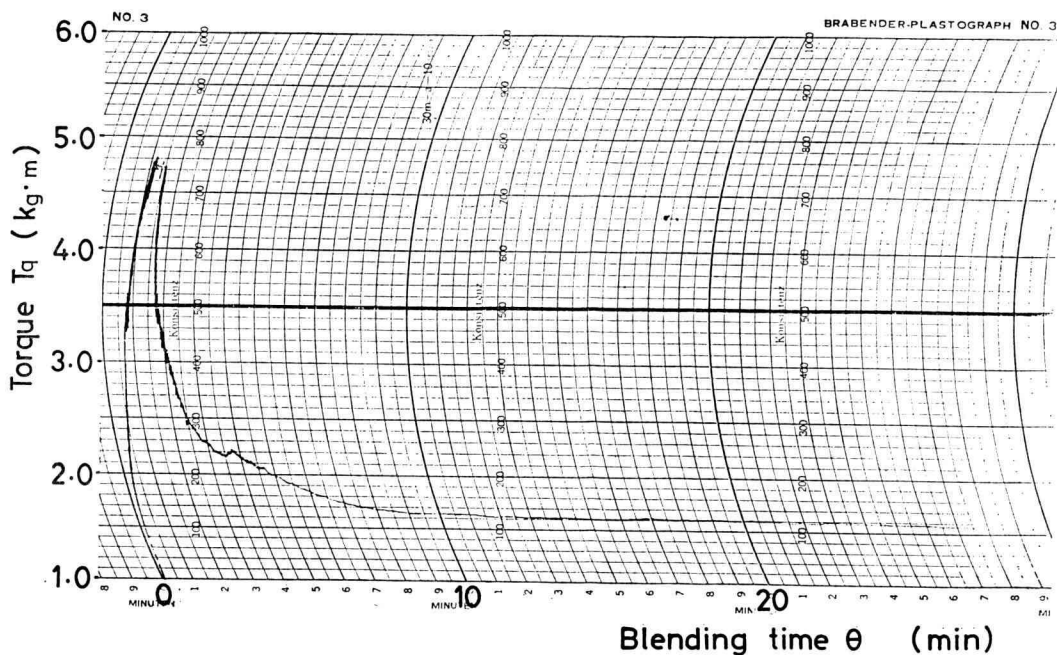


Fig. 1 Torque characteristic of the Brabender Plastograph

## 2-1 Dimensional Analysis

For our purpose of studying a general view of the blending characteristics of plastic composites, the principal factors are as follows:

- (1) effect of blending temperature  $t$  on the torque  $T_q$
- (2) effect of weight percentage of filler  $F$  on the torque  $T_q$
- (3) effect of length of fibrous filler  $l$  on the torque  $T_q$
- (4) effect of the blending time  $\theta$  on the torque  $T_q$

For the purposes of (1), (2) and (3), the torque  $T_q$  is defined as it is represented by the lowest stable value in the state of having been sufficiently blended.

Concerning the effects of these factors on the torque, we can analyse as follows:

Representing the factors by the symbols of the quantities, mass  $M$ , length  $l$ , and time  $T$ ,

$T_q$  = blending torque  $[ML^2T^{-2}]$

$\eta$  = viscosity of plastic melt  $[ML^{-1}T^{-1}]$

$F$  = weight % of filler  $[ML^{-2}T^{-2}]$

$l$  = length of fibrous filler  $[L]$

$\theta$  = blending time  $[T]$

then, writing

$$Tq = C_1 \eta^a F^b l^c \theta^d \quad (1)$$

and substituting the dimensions gives

$$\begin{aligned} [ML^{-2}T^{-2}] &= [ML^{-1}T^{-1}]^a [ML^{-2}T^{-2}]^b [L]^c [T]^d \\ b &= 1-a, \quad c = 4-a, \quad d = -a \\ \left[ \frac{Tq}{Fl^4} \right] &= C_2 \left[ \frac{\eta}{Fl\theta} \right]^a \end{aligned} \quad (2)$$

The dimensions of each of the two bracketed groups in Equation (2) are zero, and the groups are dimensionless. In this equation, because  $F$  corresponds to density of the blending material  $\rho$ , and can be written as  $F = \rho g_c$ ,  $Fl\theta = \rho(g_c\theta)l = \rho vl$ , representing  $g_c$  for the correction factor of gravity,  $v$  for velocity,  $Fl\theta/\eta = \rho vl/\eta$  corresponds to the Reynolds Number  $Re$ . Therefore, the equation (2) can be written as

$$\frac{Tq}{Fl^4} = \phi(Re) \quad (3)$$

Now, in the case of blending plastic composites by the Brabender plastograph, the viscosity is not easy to measure, but can be well represented by a function of the temperature  $t$  as the following Arrhenius equation tells.

$$\eta = \eta_0 e^{\frac{E}{Rt}} \quad (4)$$

where,  $E$  = activation energy

$R$  = gas constant

Introducing eq. (4) in eq. (1), we get

$$\log Tq = \frac{C_1}{t} + C_2 \log F + C_3 \log l + C_4 \log \theta + C \quad (5)$$

where  $C, C_1 \sim C_4$  are constants, which can be decided by experiments.

## 2-2 Experiments

On the blending of plastic composites by the Brabender plastograph we did extensive experiments. The materials used are as follows:

### Plastics

A, Low density polyethylene	Sumitomo F-704-1
B, Polypropylene	Mitsue J-700

### Fillers

C, D, E, Carbonfibers	Kureha Co.
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Symbols		Diameter $d \mu$	length $l$ mm
C	M-101	14.5	0.1
D	C-103	10.5	3.0
E	C-106	10.5	6.0

F, Calcium Carbonate

G, Asbestos

From Fig. 2 to Fig. 5, 4 figures show the representative examples of the experimental results obtained. The dimensions used are shown in the figures.

Fig. 2 shows the characteristics of torque versus temperature with the parameter of weight % of filler. The matrix is A, polyethylene, and the filler is E carbon fiber C-106.

Linearity of the characteristics is fairly good. This means that the Arrhenius equation holds good here.

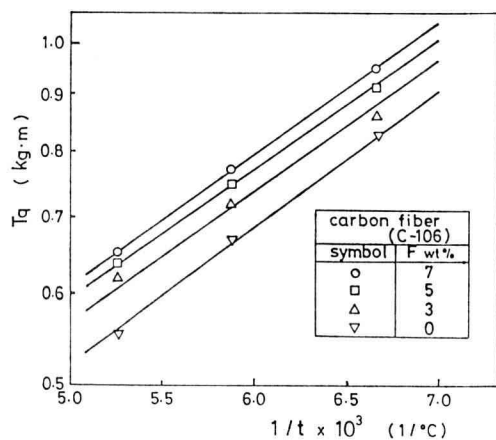


Fig. 2 Torque  $T_q$  versus temperature  $t$

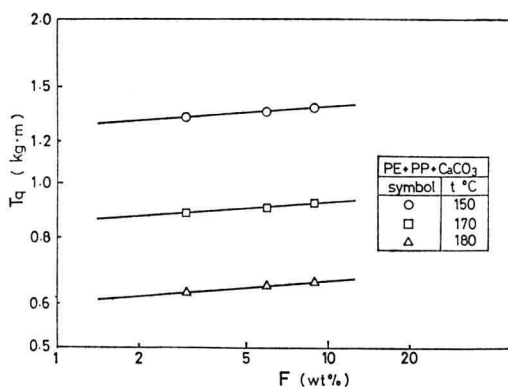


Fig. 3 Torque  $T_q$  versus % of filler  $F$

Fig. 3 shows the characteristics of torque versus weight % of filler with the parameter of temperature. The matrix plastic is a mixture of 90% polypropylene B with 10% polyethylene A, and the filler is Calcium carbonate F. The linearity is very good.

Fig. 4 shows the characteristics of torque versus length of fiber. In the case of different diameters of fiber, the equivalent length was calculated under the assumption that the surface area and the diameter are equal. Here also the linearity is good.

Fig. 5 shows the characteristics of torque versus blending time. These log-log plots seem to be divided into three regions, from 1 to 5 min. with a sharp inclination, from 5 to 40 min. with medium inclination, and above 40 min. with a horizontal line.

From the experimental results shown in the above explained figures, we could calculate all constants in the equation (5), as shown in Table 1.

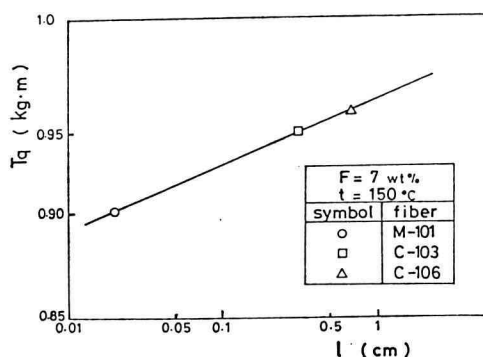
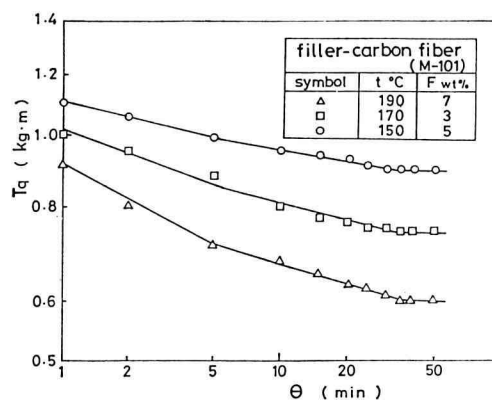
Fig. 4 Torque  $T_q$  versus length of fiberFig. 5 Torque  $T_q$  versus blending time

Table 1. Values of Constants in eq. (5)

Constant Composite	$C_1$	$C_2$	$C_3$	$C_4^*$	$C$
A+D	233	0.164	0.0197	-0.1020	-1.80
A+E	238	0.120	0.0197	-0.1035	-1.89
A+C	248	0.097	0.0197	-0.0612	-1.89
A+G	345	0.154	0	-0.0730	-2.16
B(90%)+A(10%)+F	631	0.040	0	-902080	-3.19

\* Column  $C_4$  was calculated from the medium inclination regions in Fig. 5. From these Figures and Table 1, we may say that the torque characteristics of the Brabender plastograph for blending plastic composites are well expressed by the equation (5).

### 3. Effect of Filler mixed in Plastic on the Viscoelasticity

By mixing fibrous or particulate fillers in the plastic melt, the viscoelastic deformability of the melt will decrease, because the filler will act to restrict the deformation of the high polymer molecules in the melt. To study this problem, we performed experiments on the die swell of the plastic composites.

#### 3-1 Experiments on the die swell

For the experiments on the die swell, a single screw extruder was used. Dimensions of the dies used are as shown in Table 2.

Table 2. Dimensions of dies

No. of die	1	3	5	6	7
Dia. of capillary $d_1$ mm	4	4	4	4	4
Length of Capillary $L$ mm	2	8	4	4	4
Dia. of Reservoir $d_2$ mm	8	8	10	15	20

The materials used for the die swell experiments are as follows,

Plastic : High density polyethylene (6009-ELCLOR) MI=0.9 g/10 min.

Filler : Diatomaceous earth obtained in Rio Grande de Norte in Brazil

From Fig. 6 to Fig. 9, 4 figures show the representative examples of the experimental results obtained.

In the figures, the symbols used are as follows:

$$B = \text{Swelling ratio} = \frac{d_e}{d_1} = \frac{\text{dia. of extrudate}}{\text{dia. of capillary}}$$

$$\dot{\gamma} = \text{Rate of shear} = 32Q/\pi d_1^3 \quad [\text{sec}^{-1}]$$

$$Q = \text{Volumetric rate of flow} \quad [\text{cm}^3/\text{sec}]$$

$$\tau_R = Pd_1/4L \quad [\text{kg}/\text{cm}^2]$$

$$P = \text{Pressure} \quad [\text{kg}/\text{cm}^2]$$

Fig. 6 shows the characteristics of swelling ratio versus rate of shear with the parameter of weight % of filler. The swelling ratio  $B$  increases gradually as the rate of shear  $\dot{\gamma}$  increases. But  $B$  decreases as the % of filler  $F$  increases. It is surprising that  $B$  hardly changes by increasing  $\dot{\gamma}$  in the cases of  $F=30$  and  $40\%$ .

Fig. 7 shows the characteristics of the swelling ratio  $B$  versus shear stress  $\tau_R$ . This figure is comparable with Fig. 6.

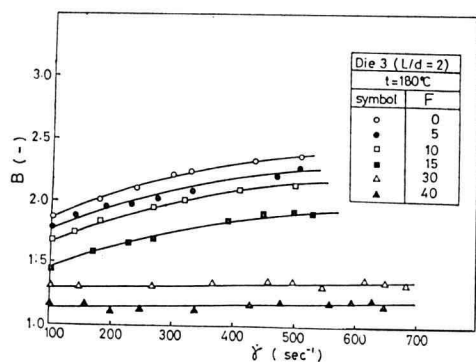


Fig. 6 Swelling ratio  $B$  versus rate of shear  $\dot{\gamma}$

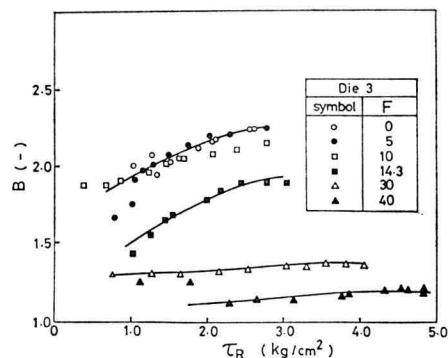


Fig. 7 Swelling ratio  $B$  versus shear stress  $\tau_R$

Fig. 8 shows the characteristics of  $B$  versus  $L/d_1$ . This figure shows the effect of the length of capillary on the die swell. By increasing the length of capillary, the die swell  $B$  decreases gradually. This phenomenon can be explained by considering the stress relaxation occurring in the capillary. In this figure, the effect of filler on die swell shows very clearly that  $B$  decreases as  $F$  increases.

Fig. 9 was obtained from Fig. 6. This figure shows the characteristics of die swell  $B$  versus % of filler  $F$  with the parameter of rate of shear  $\dot{\gamma}$ . These curves show clearly how  $B$  decreases with increasing  $F$ . This figure shows also that each of the swelling ratio curves converge to the point at  $F=40\%$ .

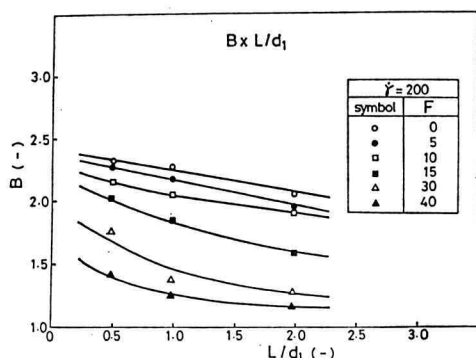


Fig. 8 Effect of length of capillary  $L/d_1$  on swelling ratio B

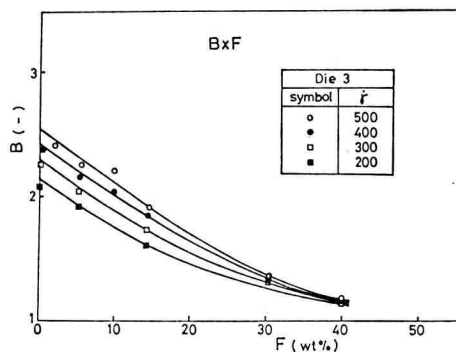


Fig. 9 Swelling ratio B versus % of filler F.

### 3-2 Effect of filler mixed in plastic on recoverable elongational strain

By experimentation, we have made it clear what effect mixing filler in the melt has on die swell. Next we should analyse the problem further.

Many reports have been published concerning die swell. However, because the mechanism of the phenomenon is not clearly explained by those reports, a new explanation is proposed here.

When a plastic melt is extruded through a circular hole die, the driving pressure gradient, along the direction of the axis, gives a shear strain to a flow unit, which is a small cubical element imagined in the melt. During the shear flow, a long chain molecule will be elongated in the flow direction, by a frictional force produced by neighbouring molecules. The elongational strain  $\epsilon$  of the flow element is a function of rate of shear  $\dot{\gamma}$  or shear stress  $\tau$ . Long chain molecules receiving this stretching due to the shear flow tend, at the same time, to contract, due to their relaxation characteristics. As the result, if the duct is long enough, the elongational strain comes to an equilibrium value. This equilibrium value may be called "recoverable elongational strain," because the molecule has a capability to return to its original random coil dimension, when it is completely released from the loaded stress, and has enough time for this relaxation.

When the melt comes out of the die exit, this state is realized, and the molecule begins to contract gradually until the recoverable strain disappears. The volume of the plastic melt generally changes very little. Therefore, the diameter of the extrudate must increase gradually, corresponding to the gradual contraction in the axial direction. The die swell is caused like this.

From this point of view, the effect of filler on die swell, can be explained as follows.

Fibrous or particulate solid filler can not deform so much as the long chain molecule of high polymers. Therefore, if some volume % of filler is added to the matrix melt, the remaining part of the flow unit, must receive the more severe shearing action compared with the case of filler not being added. So, long chain molecules can not be elongated so much

corresponding to such a severe shearing action by the friction of neighbouring molecules, because the capability of the being elongated must have some upper limit, and if this limit is exceeded, some slippage may occur. In other words, the recoverable elongational strain can not increase as much as the rate of shear in the matrix when the % of filler is increased. As the total effect, the swelling ratio  $B$  or the recoverable elongational strain  $\varepsilon(\dot{\gamma})$  for the same rate of shear, may be decreased by increasing the weight % of filler  $F$ . This is our explanation for the characteristics of the swelling ratio  $B$  versus % of filler  $F$  shown in Fig. 6. or Fig. 9.

Advancing our way of thinking on the mechanism of die swell, we can obtain analytically the recoverable elongational strain, using the experimental data shown in Fig. 6. as follows.

Now, representing the recoverable elongational strain of the flow element in the die as a function of shear stress  $\tau$  acting on the element by  $\varepsilon(\tau)$ , the length of the element in the die which corresponds to a unit length of the element in the extrudate completely relaxed, can be written as  $(1+\varepsilon(\tau))$ .

Then, considering that the volumetric change of the melt due to the pressure change is negligible, and integrating the volume of the element over the whole cross section in the die and in the extrudate, we can write,

$$\begin{aligned} \int_0^R (1+\varepsilon(\tau)) 2\pi r dr &= \int_0^{Re} 1 \times 2\pi r dr = \pi R_e^2 \\ 2 \int_0^R (1+\varepsilon(\tau)) r dr &= R_e^2 \end{aligned} \quad (6)$$

where  $R$ =Radius of the die

$Re$ =Radius of the extrudate

Considering the shear stress distribution in the radial direction

$$\frac{\tau}{\tau_R} = \frac{r}{R} \quad (7)$$

where

$$\tau_R = \frac{PAD}{4L} \quad (8)$$

$\Delta P$ =Pressure drop in the die.

We can change the variable  $r$  into the shear stress  $\tau$  in equation (6), and we get

$$2 \left( \frac{R}{\tau_R} \right)^2 \int_0^{\tau_R} (1+\varepsilon(\tau)) \tau d\tau = R_e^2$$

Here, as the swelling ratio  $B$  is  $Re/R$ , we can write

$$B^2 = \frac{2}{\tau_R^2} \int_0^{\tau_R} (1+\varepsilon(\tau)) \tau d\tau \quad (9)$$

Differentiating equation (9) with respect to  $\tau_R$  we get,

$$\varepsilon(\tau_R) = B^2 + \frac{1}{2} \frac{dB^2}{d \ln \tau_R} - 1 \quad (10)$$



Assuming that the flow characteristic of the melt can be expressed by the power law,

$$\dot{\gamma} = \frac{\tau^n}{\eta} \quad (11)$$

$$\text{then, as } \frac{d\dot{\gamma}}{d\tau} = n \frac{\tau^{n-1}}{\eta} = n \frac{\dot{\gamma}}{\tau}, \quad \frac{dB^2}{d \ln \tau} = \frac{dB^2}{d \dot{\gamma}} n \dot{\gamma}$$

we get

$$\varepsilon(\dot{\gamma}) = B^2 + \frac{n}{2} \frac{dB^2}{d \ln \dot{\gamma}} - 1 \quad (12)$$

Therefore from the index  $n$  of the power law of the melt, and the characteristic of the swelling ratio  $B$  versus rate of shear  $\dot{\gamma}$ , we can calculate  $\varepsilon(\dot{\gamma})$  by eq (12).

As an example of this analysis, we calculated the  $\varepsilon(\dot{\gamma})$  in the case of Fig. 6, and got the result as shown in Fig. 10. In this case, the power law index  $n$  was 3.1. From the log-log plot of Fig. 10., for the cases of the filler  $F=0\sim 15$  weight %, we can write

$$\varepsilon(\dot{\gamma}) \doteq C \dot{\gamma}^{1/3} \quad (13)$$

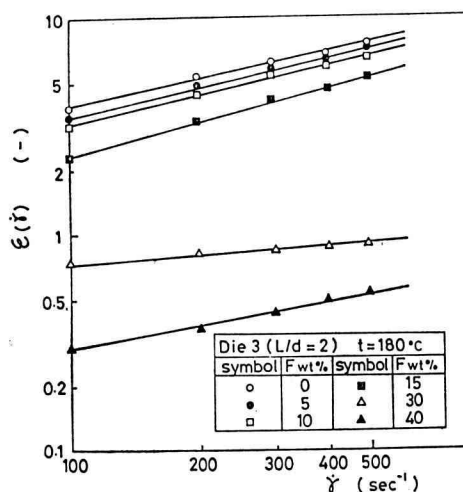


Fig. 10 Recoverable elongational strain  $\varepsilon(\dot{\gamma})$  versus rate of shear  $\dot{\gamma}$

#### 4. Conclusion

Concerning plastic processing, we studied the rheological properties of plastics mixed with various fillers, using the Brabender plastograph and screw extruders. From the results of the study, we can deduce the following.

The troque characteristics of the Brabender plastograph can be expressed well by eq. (5) as a function of the variables of temperature  $t$ , weight % of filler  $F$ , length of fibrous filler  $l$ , and blending time  $\theta$ .

Concerning screw extruding, die swell increases when rate of shear is increased and decreases when the weight % of filler is increased, but if the % of filler is over 30%, the swelling ratio does not change when the rate of shear is changed.

Further, as the result of our study on die swell, we can say that the mechanism of why the swelling phenomena occur, can be explained well by assuming that the recoverable elongational strain is caused in the flow unit by shear flow. Using this conception, we can well explain the reason why swelling ratio decreases when the weight % of filler is increased. Equation (12) was derived analytically for expressing the recoverable elongational strain  $\varepsilon(\dot{\gamma})$ , as a function of the swelling ratio  $B$ . As an example of application of the equation (12), we could get an equation (13) of  $\varepsilon(\dot{\gamma})$  as a function of the rate of shear  $\dot{\gamma}$ .

### Notation

$B$ =swelling ratio= $R_e/R$	$r$ =radius
$d_1$ =diameter of die hole	$Tq$ =torque
$d_2$ =diameter of reservoir	$t$ =temperature
$F$ =weight % of filler	$\varepsilon(\dot{\gamma})$ =recoverable elongational strain
$L$ =length of die hole	$\dot{\gamma}$ =rate of shear
$l$ =length of fibrous filler	$\theta$ =blending time
$R$ =radius of die hole	$\tau$ =shear stress
$R_e$ =radius of extrudate, or Reynolds number	$\eta$ =viscosity

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