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Scale-up Extrudate Swell of Rubber Compound on Capillary Rheometer to Extruder

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The relationship of extrudate swell occurring in capillary rheometer and extruder is the aim of this study. Three assumptions used are: (1) the molten polymer is sheared and given the largest deformation at the entrance of the die; (2) the retraction of an elastic part occurs during the molten flow in the die, and (3) the total retraction of the melt appears at the exit of the die. The largest deformation at the entrance of the die depends linearly on shear rate and the retraction in the die depends on both shear rate and die dimensions (diameter and length), which is defined as a retraction function. Separation of these two effects on the largest deformation and the retraction function is proposed by defining a 'extrudate swell rate function, K' as $(\sqrt{(\chi)^2 - 1}) / \dot{\gamma}_w$, *where* χ *is the extrudate swell ratio and* $\dot{\gamma}$ *is the shear rate. This indicates that* K *is a function of shear rate, die dimension and melt properties, similar to the retraction function.*

Extrudate swell of natural rubber compounds mixed with carbon black and calcium carbonate were examined with a capillary rheometer and an extruder at processing temperature of 100°C. The die used had various sizes varying from 1.5 mm – 5.5 mm in diameter and 16 mm – 30 mm in length while shear rate is varied from 1 – 2000 s^{-1}. *The results show that the empirical extrudate swell rate function* K *is equal to* $A\dot{\gamma}_w^n$, *where* n *is a constant varying from –0.70 to –0.89 depending on the type of compounds and* A *is a constant varying from 0.5 to 0.8 depending on the die length. An agreement between the extrudate swell rate functions* (K) *obtained either from the capillary rheometer and or with the extruder is discovered. Therefore, extrudate swell behaviour observed in laboratory equipment as a capillary rheometer can be used to explain the behaviour in an industrial extruder.*

Polymer melt is deformed under shear and extensional force during its flow through an extrusion die. Elastic recovery of polymer melt has been described to be the main reason for extrudate swell. Tanner[1] proposed that first normal stress difference (N_1) and wall shear stress (τ_w) of capillary die played an important role in the occurrence of extrudate swell, as shown in *Equation 1*.

$$\chi = \frac{D_f}{D} = \left[1 + \frac{1}{8}\left(\frac{N_1}{\tau_w}\right)^2\right]^{\frac{1}{6}} \qquad \ldots 1$$

where

χ = extrudate swell ratio

D_f and D = diameter of extrudate and die, respectively.

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Tanner's theory is satisfied only with experiments where the ratio of die length to diameter (L/D) is greater than 30 and low volumetric flow rates. Baird[2] showed that Tanner's equation does not take the factor of die dimension into account. Also it does not correlate well with the measurement of extrudate swell in normal polymeric extrusion. However, Tanner's principle has been widely accepted and still used by a number of workers. Han[3] and Guillet[4] developed Tanner's principle by adding dimensional effects into a previous equation. Cogswell[5,6] studied flows in the entrance region of capillary die. He concluded that not only shear flow but also extensional flow exist in this region. Cogswell showed that extensional deformation and extrudate swell related to entrance angle of dies. Decreasing entrance angle will decrease swell. Eggen[7] calculated extrudate swell by using Tanner's and Cogswell's principles, as shown in *Equation 2*:

$$\chi = \frac{D_f}{D} = 0.13 + K\left(1+\frac{D}{D+L}\right)\left\{\frac{\lambda_0^3 G(t_e+t_c)+\lambda_0[G(0)-G(t_c)]}{G(t_e+t_c)+\lambda_0[G(0)-G(t_c)]} + \frac{1}{2}\left(1+\frac{D}{D+L}\right)^6\left(\frac{\psi_1\dot{\gamma}^2}{2\eta\dot{\gamma}}\right)\right\}^{\frac{1}{6}} \quad \ldots 2$$

where

$G(0)$ = relaxation modulus before the entrance region
$G(t_c)$ = relaxation modulus at the end of entrance region
$G(t_e+t_c)$ = relaxation modulus at die exit region
ψ_1 = normal stress coefficient
λ_0 = extension ratio, the radius ratio of barrel and die
D and L = die diameter and die length, respectively
η and $\dot{\gamma}$ = viscosity and shear rate, respectively
K = scaling coefficient.

As shown in this equation, the control parameter is extrusion shear rate. Eggen[7] showed that his equation is well correlated with experiments. However, his equation uses too many parameters. Therefore, it is not practical for use in a real extrusion process. Both Tanner and Cogswell's theories are based on steady flow. Hence, both theories meet swell experiments with polymer melt flow at low viscosities and low elastic effects. They can not be used to explain extrudate swell of rubber compounds, which exhibit high viscosity and strong elastic effects. Bernart[8] showed that the extensional rate ($\dot{\varepsilon}_a$) at the entrance region is a linear function of volumetric flow rate (Q) as indicated in *Equation 3*:

$$\dot{\varepsilon}_a = Q(2-\cos\theta) / (\pi R_0^3) \quad \ldots 3$$

where
Q = flow rate
θ = entrance angle
R_0 = die radius.

This work aims to evaluate the relationship of extrudate swell occurring in capillary rheometer and extruder, by considering three hypotheses : firstly, the molten polymer is given the largest deformation at the die's entrance, secondly, there is some elastic retraction during flows in the die; and finally, the complete retraction of the melt appears at the die's exit. In a simple way, the extensional and shear deformations in the entrance region can not be separated, since the normal and shear components might be inversely transformed. Therefore, shear and extensional deformations are assumed to correspond to one apparent shear deformation.

A correlation of extrudate swell ratio (χ) and shear strain (λ_a), as defined by McIntosh[9], is used, as shown in *Equation 4*:

$$\chi = \frac{R_s}{R} = \sqrt{1+\gamma_a^2} \qquad \text{... 4}$$

where

R_s and R are radius of extrudate and die, respectively.

The shear strain (γ_a) at the die exit can be expressed as a function of the maximum shear strain at die entrance ($\gamma_{(max)}$) and of the strain relaxation function in the die $F(t, \lambda)$, as follows:

$$\gamma_a \propto \gamma_{(max)} F(t, \lambda) \qquad \text{... 5}$$

where

$\gamma_{(max)}$ = maximum shear strain at the die entrance

$F(t, \lambda)$ = strain relaxation function in the die which depends on residence time (t) in die and relaxation time (λ).

Residence time (t) depends on die wall shear rate, but the relaxation time (λ) is rather constant. Therefore, $F(t, \lambda)$ is a function of wall shear rate and die geometry. Maximum shear strain $\gamma_{(max)}$ and strain relaxation function $F(t, \lambda)$ are separated by estimating the maximum shear strain as a linear function of wall shear rate ($\dot{\gamma}_w$) as $\gamma_{(max)} = k\dot{\gamma}_w$ where k is an arbitary constant. Therefore *Equation 5* is then rewritten as :

$$\gamma_a \propto k\dot{\gamma}_w F(\dot{\gamma}_w, L, R) \qquad \text{... 6}$$

From the *Equations 4* and *6*, the relationship between the strain relaxation function and extrudate swell ratio is concluded as follows:

$$K = (\sqrt{(\chi)^2 - 1}) / \dot{\gamma}_w \propto F(\dot{\gamma}_w, L, R) \qquad \text{... 7}$$

where

K = extrudate swell rate function.

K value depends on the relaxation function $F(\dot{\gamma}_w, L, R)$. Extrudate swell rate function (K) is a function of die wall shear rate and die dimension (L and R). Correlation between K and wall shear rate is our main interest in this work. Moreover, the influence of geometry for extrusion die is another investigation.

MATERIALS AND METHODS

The best grade of natural rubber, TTR 5L (produced by Thavon Rubber Block Industry Co. LTD., Thailand) was used as base polymer and the two types of fillers used are carbon black (N-330), produced by Thai Carbon black Co. LTD., Thailand) and calcium carbonate (Surin Omya Co. LTD., Thailand). A single bore capillary rheometer, model RH-710, (Rosand Precision Co. LTD., England.) and a cold feed extruder with a barrel diameter of 38.4 mm (Farrel Co. LTD., England) were the two main types of equipment used in the study.

Various quantities of fillers (carbon black and calcium carbonate) were mixed into natural rubber using a two-roll mill at a mixing temperature of 70°C. Mooney viscosity, ML (1+4) 100, of the compound was in the range of 45±5. Extrudate swell was measured with the Rosand capillary rheometer and Farrel coldfeed extruder at a temperature of 100°C. Extrusion dies are capillary dies with 1.5 mm diameter and 16, 23 and 30 mm die length for capillary rheometer extruder and 2.5 mm and 5.5 mm with diameter 16 mm die length for the cold feed extruder. Tests were carried out at capillary wall shear rates of 5 –1800 s^{-1}.

Extrudate swell ratio is obtained from the ratio of L/D [length of screw (L) and diameter of die head (D)]. Average extrudate diameters was measured after 5 min of extrusion, without any haul off operation. The relationship between extrudate swell rate function (K) (as shown in *Equation 7*) and capillary wall shear rates were investigated .

RESULTS AND DISCUSSIONS

Extrudate swell ratio, swell rate functions (K) and die wall shear rates ($\dot{\gamma}_w$) for rubber compound mixing with $CaCO_3$ contents of 15, 30 and 45 p.h.r. are given in *Table 1*. It is clear that extrudate swell ratio increases with increasing wall shear rate, whereas extrudate swell rate function (K) decreases.

Log-log scale plot of extrudate swell rate function (K) and capillary wall shear rate of 15 p.h.r. $CaCO_3$ mixed compound are showed in *Figure 1*. This relationship shows a straight line, which can be proposed as a power equation:

$$K = A\,\dot{\gamma}_w^n \quad \text{... 9}$$

where
A = coefficient constant and n = power index.

The coefficient constant, power index and correlation coefficient for various cases were calculated mathematically and given in *Table 2*. For all cases of 3 different quantities of $CaCO_3$ and 3 different types of die geometry,

TABLE 1. EXTRUDATE SWELL RATIO AND SWELL RATE FUNCTION (K) AT VARIOUS WALL SHEAR RATE AND 3 DIFFERENT DIE LENGTHS FOR 3 LEVELS OF $CaCO_3$ MIXED COMPOUNDS

$CaCO_3$ (p.h.r.)	Shear rate (s^{-1})	χ extrudate swell ratio 16 mm	23 mm	30.75 mm	Extrudate swell rate function (K) 16 mm	23 mm	30.75 mm
15 p.h.r.	4.44	1.48	1.45	1.29	0.2425	0.2354	0.1824
	17.8	1.46	1.44	1.30	0.0594	0.0581	0.0470
	44.0	1.54	1.46	1.34	0.0265	0.0243	0.0202
	178	1.66	1.65	1.49	0.0074	0.0074	0.0062
	444	1.93	1.79	1.61	0.0033	0.0033	0.0028
	889	2.08	1.96	1.76	0.0021	0.0019	0.0016
	1728	2.33	2.17	1.98	0.0012	0.0011	0.0010
30 p.h.r.	4.44	1.38	1.35	1.22	0.2148	0.2032	0.1584
	17.8	1.38	1.35	1.25	0.0538	0.0508	0.0418
	44.0	1.42	1.38	1.28	0.0231	0.0214	0.0180
	178	1.60	1.52	1.41	0.0070	0.0065	0.0056
	444	1.80	1.74	1.55	0.0034	0.0032	0.0027
	889	2.02	1.87	1.71	0.0020	0.0018	0.0016
	1728	2.19	2.04	1.86	0.0011	0.0010	0.0009
45 p.h.r.	4.44	1.38	1.36	1.24	0.2150	0.2074	0.1633
	17.8	1.36	1.34	1.24	0.0513	0.0502	0.0409
	44.0	1.35	1.33	1.25	0.0207	0.0201	0.0171
	178	1.43	1.42	1.32	0.0058	0.0056	0.0049
	444	1.56	1.54	1.44	0.0027	0.0026	0.0023
	889	1.74	1.65	1.50	0.0016	0.0015	0.0013
	1728	1.93	1.80	1.63	0.0009	0.0008	0.0008

the coefficient of correlation (r^2) approaches unity which means that A and n values are accurate empirical data.

The relationship of extrudate swell rate function (K) and wall shear rate of carbon black mixed compounds shows also good agreement with *Equation 9*. The coefficient constant (A), power index (n) and coefficient of correlation (r^2) are given in *Table 3*. The coefficient constants (A) are similar to that of $CaCO_3$ mixed compounds, while the power indexes (n) are slightly higher.

The relationship between the coefficient constant (A) and die length of $CaCO_3$ and carbon black mixed compounds are showed in *Figures 2* and *3*, respectively. It is clear that the coefficient

TABLE 2. COEFFICIENT CONSTANT (A), POWER INDEX (n) AND CORRELATION COEFFICIENT (r^2) OF $CaCO_3$ MIXED COMPOUNDS OF 3 DIFFERENT DIE LENGTHS

$CaCO_3$ (p.h.r.)		L = 16 mm	L = 23 mm	L = 30 mm
15	A	0.80	0.79	0.60
	n	–0.88	–0.89	–0.87
	r^2	–0.99	–0.99	–0.99
30	A	0.69	0.66	0.51
	n	–0.87	–0.87	–0.86
	r^2	–0.99	–0.99	–0.99
45	A	0.71	0.71	0.56
	n	–0.90	–0.91	–0.90
	r^2	–0.99	–0.99	–0.99

TABLE 3. COEFFICIENT CONSTANT (A), POWER INDEX (n) AND CORRELATION COEFFICIENT (r^2) OF EXTRUDATE SWELL RATE FUNCTION AND SHEAR RATE FOR CARBON BLACK MIXED COMPOUNDS OF 3 DIFFERENT DIE LENGTHS

Carbon black (p..h.r.)		L = 16 mm	L = 23 mm	L = 30 mm
15	A	0.72	0.72	0.60
	n	–0.88	–0.88	–0.87
	r^2	–0.99	–0.99	–0.99
30	A	0.72	0.71	0.57
	n	–0.90	–0.91	–0.90
	r^2	–0.99	–0.99	–0.99
45	A	0.76	0.77	0.61
	n	–0.92	–0.93	–0.94
	r^2	–0.99	–0.99	–0.99

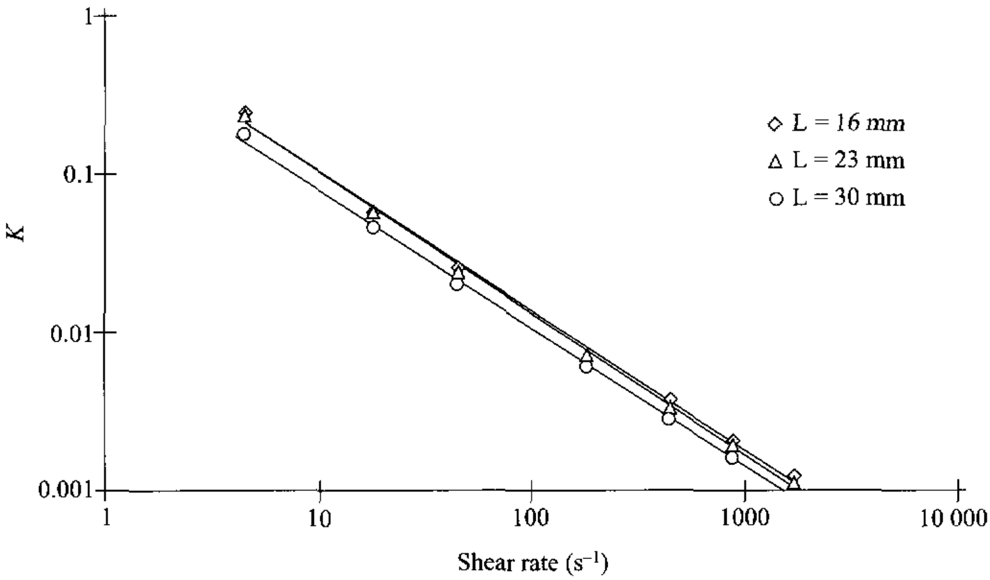

Figure 1. Log-log scale plot of extrudate swell rate function and shear rate of 15 p.h.r. $CacO_3$ mixed compound

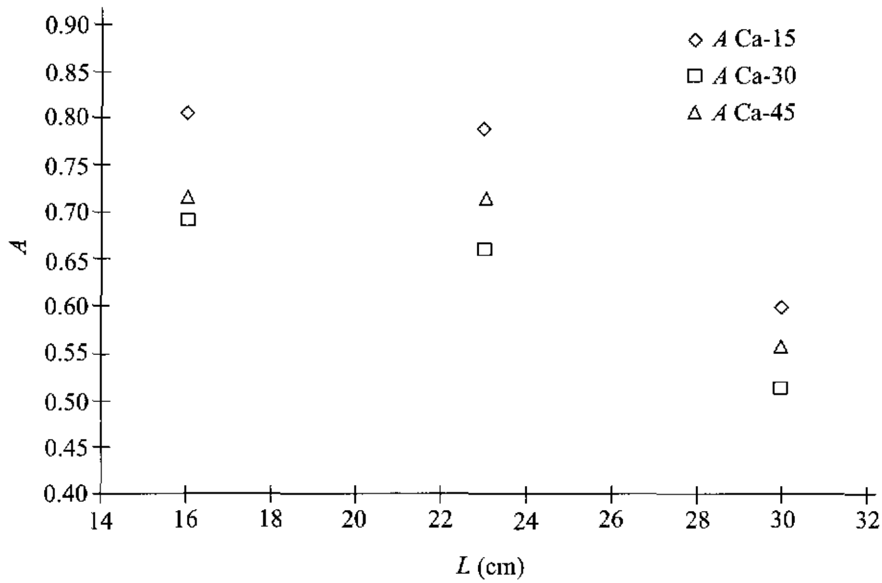

Figure 2. The correlation of coefficient constant (A) and die length of $CacO_3$ mixed compounds.

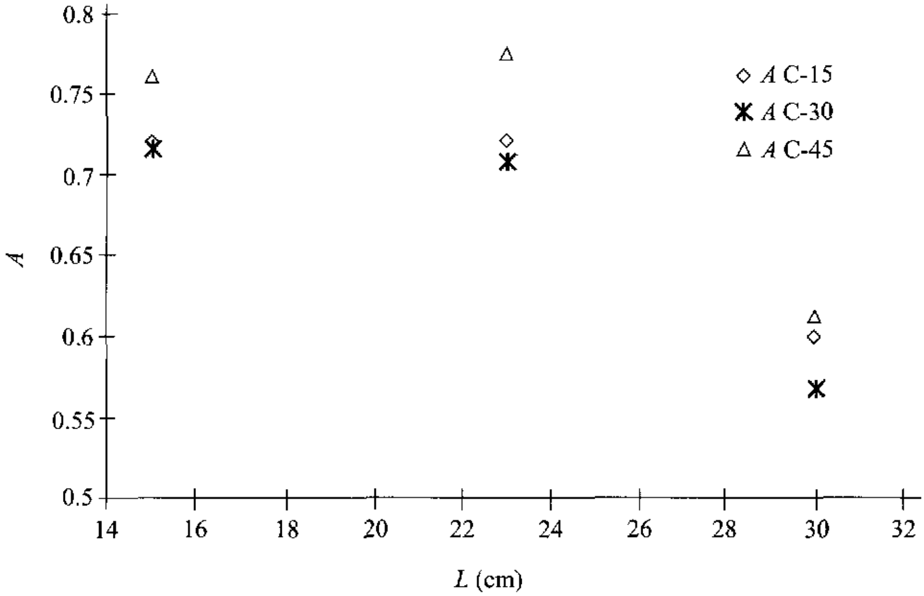

Figure 3 The correlation of coefficient constant (A) and die length of carbon black mixed compounds.

constant (A) decreases with increasing die length, but slightly increases with increasing filler level. It can be said that die length has a strong effect on the value of coefficient constant (A), but that filler level has little effect.

The relationship of power index (n) and die length are illustrated in *Figures 4* and *5*. Similar values of power index (n) at various die lengths are observed. However, power index increases with increasing filler level.

Extrudate swell in capillary extruder and cold-feed extruder was compared. Both fillers ($CaCO_3$ and carbon black) were used in the quantities of 15, 30 and 45 p.h.r. The compounds were extruded using capillary extruder with die having 1.5 mm diameter and 16 mm die length. The same batch compounds were extruded using cold-feed extruder with dies having 2.5 mm and 5.5 mm of diameter and 16 mm die length. Extruding temperature set for both cases was 100°C. *Figures 6* and *7* show a plot on log-log scale of the relationship between extrudate swell rate function (K) and wall shear rate ($\dot{\gamma}_w$). It was found that in all cases the swell rate function (K) was well fitted with *Equation 9* since a straight line was obtained. This relationship with different die diameters had the same trend and was slightly superimposed. For die diameter 5.5 mm it was slightly lower than the results of those 1.5 mm and 2.5 mm dies. This could be the influence of the die entrance angle and the difference in barrel diameter (50 mm for cold feed extruder and 10 mm for capillary rheometer). The die entrance angle of capillary rheometer was 180 degrees, while that for cold feed extruder was 90 degrees.

CONCLUSION

The power law relationship $K = A\,\dot{\gamma}_w^n$ between extrudate swell rate function (K) and die wall

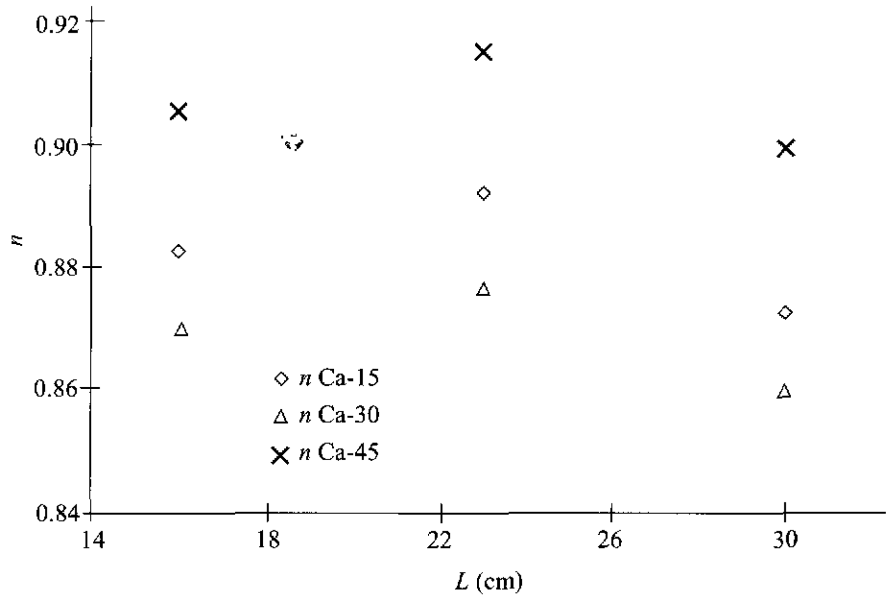

Figure 4 The correlation of power index (n) and die length of $CaCO_3$ mixed compounds.

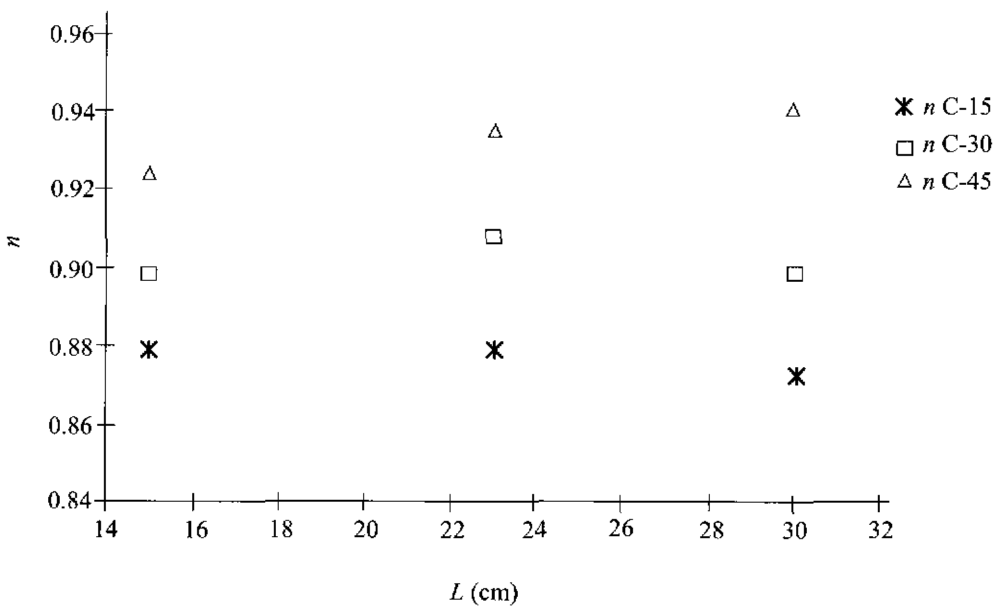

Figure 5 The correlation of power index (n) and die length of carbon black mixed compounds.

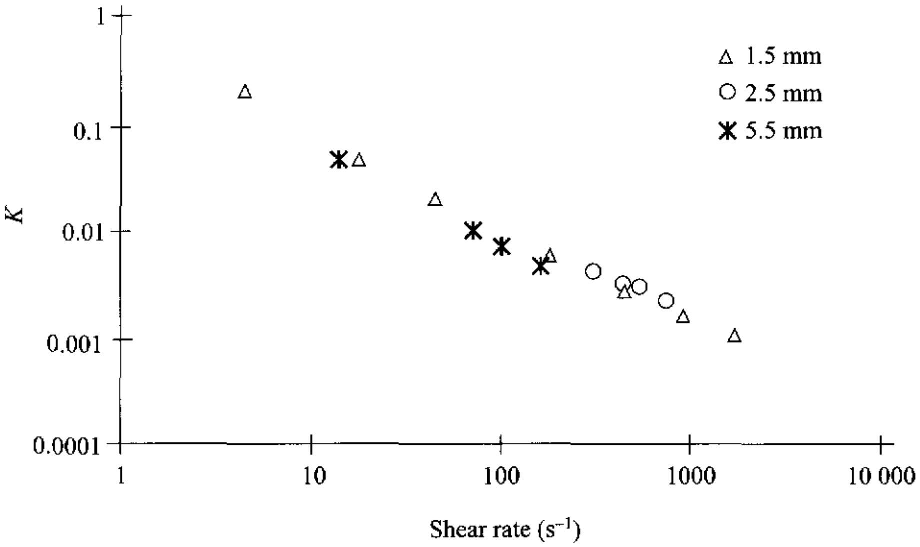

Figure 6. Extrudate swell rate function (K) and shear rate for 45 p.h.r. $CaCO_3$ mixed compounds extruded by capillary rheometer (ϕ 1.5 mm) and cold feed extruder (ϕ 2.5 mm and 5.5 mm).

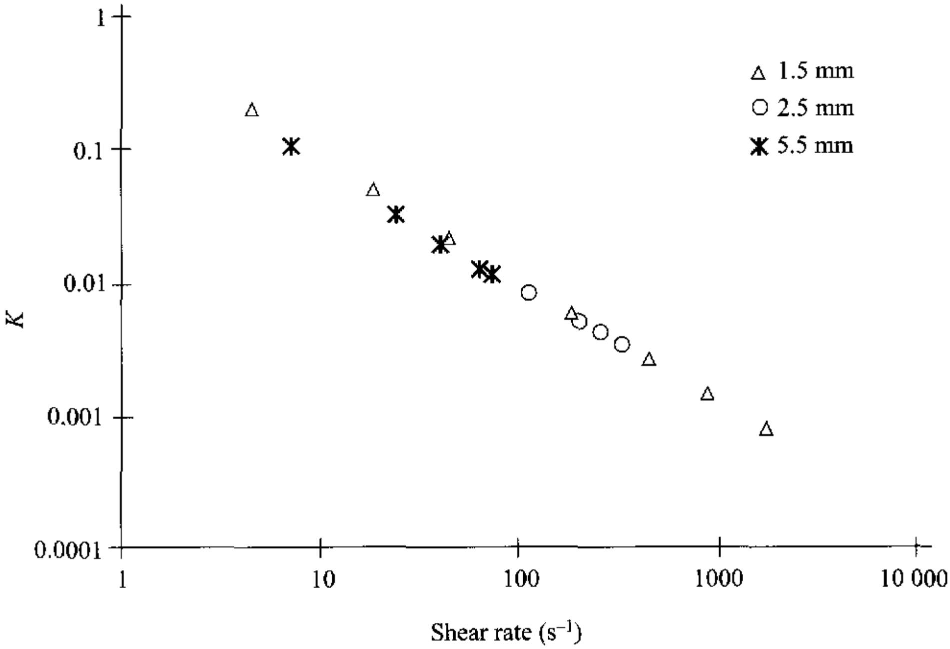

Figure 7. Extrudate swell rate function (K) and shear rate for 45 p.h.r. carbon black mixed compounds extruded by capillary rheometer (ϕ 1.5 mm) and cold feed extruder (ϕ 2.5 mm and 5.5 mm).

shear rate ıs lınear on log-log scale for both $CaCO_3$ and carbon black filled compounds The coeffıcient constant (A) decreases as dıe length ıncreases, but ıs not sıgnıficantly dependent on filler levels The power ındex (n) ıncreases wıth the ıncreasıng of the level of fillers and ıt ıs not affected by dıe length

The relatıonshıp of extrudate swell rate functıon and dıe wall shear rate does not depend on the type of extruder, barrel dımensıon or dıe dıameter A plot of thıs relatıonshıp on log-log scale for dıfferent extruder and dıfferent dıe dıameter havıng the same length of dıe, shows the same trends Therefore, ıt ıs lıkely that thıs relatıonshıp assessed from experıments wıth a capıllary rheometer could be practıcally used to predıct the extrudate swell wıth larger ındustrıal extrusıon dıes

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