

Elastic Effects of Some Polyolefines

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Due to the elastic behaviour of the melting at the extrusion head exit, the product changes its dimensions (swelling). This phenomenon depends on the L/d ratio of the capillary or the extrusion die. Experimental determinations have been undertaken in order to establish the swelling phenomenon for various polymers.

Keywords: elastic behaviour, extrusion head exit, swelling phenomenon

After exiting the capillary many polymeric products have a larger diameter than the capillary. This phenomenon was noticed for the first time by Barus in 1893, being mentioned in the scientific literature under various names: extrudate swelling, swelling memory, Barus effect, Merrington effect etc. [1]

The swelling phenomenon is the result of the symptom of normal components of the tension tensor which appear during the material flow through the capillary. It is thus a symptom of the elastic memory of the material.

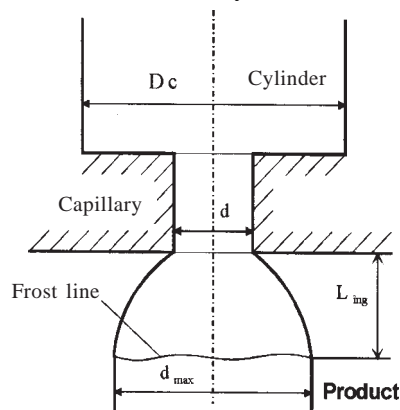


Fig. 1. Definition of swelling: D_c - cylinder diameter; d - capillary diameter; d_{max} - maximum diameter of the extruded product; L_{ing} - distance to the "frost line"

The swelling is quantitatively defined by the ratio between the product diameter and the capillary diameter (fig. 1):

$$B = \frac{d_{max}}{d} \quad (1)$$

In the case of the Poiseuille flow through a capillary the normal components of the tensions tensor have the following expressions in cylindrical coordinates [1; 2]:

$$\left. \begin{aligned} \tau^{rr} &= -\frac{1}{\varphi} \cdot \sigma_1 \cdot \left(f \cdot \frac{\varphi}{2} \right) \cdot d\varphi + z \cdot f + \rho \cdot \psi + q(t) \\ \tau^{zz} &= \tau^{rr} + \sigma_2 \cdot \left(f \cdot \frac{r}{2} \right) - \sigma_1 \cdot \left(f \cdot \frac{r}{2} \right) \end{aligned} \right\} (2)$$

in which: φ is the coordinate in the axial direction, dependent on time ($\varphi(t) = z$); f - driving force specific to the flow; ρ - density of the material; $\psi = g \cdot z$ - according to the potential depending on the forces exercised by the external environment on the material; $q(t)$ - arbitrary function of the current ray, r and time, t ; σ_2 ; σ_1 - modified

functions of normal tension components, which are experimentally determined.

If the swelling factor is defined as a ratio of an element after (index 2) and before (index 1) the full recovery of the accumulated elastic deformations, then for the relative protraction between the two statuses can be written [3]:

$$\varepsilon_R = \ln \frac{L_2}{L_1} \quad (3)$$

If it is accepted that the recovery takes place at a constant volume, for a rectangular profile:

$$W_1 \cdot H_1 \cdot L_1 = W_2 \cdot H_2 \cdot L_2 \quad (4)$$

in which: W is the width; H - height and L - length for the analysed volume element.

A recovery of longitudinal deformations leads to swelling factors according to the width [3]:

$$B_B = \exp(K^* \cdot \varepsilon_R) \quad (5)$$

and to thickness:

$$B_H = \exp(K \cdot \varepsilon_R) \quad (6)$$

where $K + K^* = 1$. The K and K^* coefficients indicate the anisotropy of the swelling values. In general, coefficients are different and depend on the form of the extrudate section; for the square section they are equal. Relations (5) and (6) show that the swelling factors can be correlated with the average status of reversible deformation determined by calculation.

The swelling that can be noticed at the extrusion die cannot be attributed only to the elastic properties of the melting. At the extrusion die, a redistribution of the speeds profile is added, more or less parabolic, for a cork type of profile. In the case of melting adhering to the wall, a protraction of the strata close to the wall takes place, while the strata in the middle of the melting are compressed [4]. This leads to the modification of the section form, which cannot be calculated. The effects over the extrudate section can be very important.

In figure 2 the influence of the flowing channel geometry over the accumulation / the contraction of reversible deformation is presented (the channel has the shape of a broad slot of a variable height). One can ascertain how important the influence of the convergence sections of the channel is. The shear deformations are accompanied by protractions with a greater width from the point of view of reversible deformations. In the channel portions following

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the converging ones one reaches to a contraction of elastic deformations. The effects of so-called "smoothing" areas are accentuated at the extrusion dies.

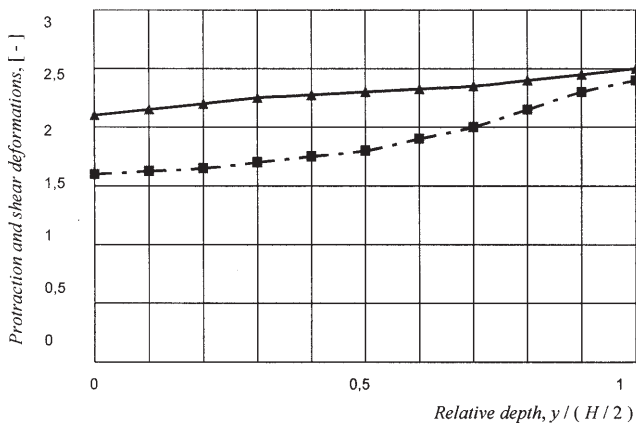


Fig. 2. Variation of protraction and shear deformations due to the speed differences following the flow direction and the shear direction, according to the relative depth $y / (H / 2)$ [3]:
▲ – protraction deformations; ■ – shear deformation

The relatively accentuated increase in the values of deformations near the wall must be attributed to the high rheo-slopes from the last portion, which is narrower. It results that a "smoothing" area has no positive effect over contracting reversible deformations near the wall.

When analysing the accumulation of reversible elastic deformations in a circular channel, one must also consider the longitudinal and circumferential deformations in the convergent / divergent portions of annular slots [5].

The influence of geometry over the accumulation or the contraction of reversible deformations, for a rectangular section channel with of a variable height, is presented in figure 3. In the area of convergence fields, shear deformations are accompanied by protraction with a greater width from the point of view of reversible deformations. In the channel portions following the convergent ones a contraction of elastic deformations is reached. This means that the effects of so-called "smoothing" areas in the extrusion dies are accentuated. Geometries I and II show that direct deformations at the extrusion die are more quickly efficient than those applied in the preceding areas. For geometry III one can notice lower ϵ_R values are obtained. The relatively pronounced increase of ϵ_R values near the wall must be attributed to the high rheo-slopes from the last portion of geometry III. It results that a smoothing area has no positive effect on contracting reversible deformations near the wall. The geometry of the exit slot influences reversible deformations and implicitly the swelling of the product. Thus one can quantitatively evaluate the swelling behaviour according to the variation of regime parameters (flow, temperature) and geometry of the flow channel. The parallel channel areas lead to a contraction of reversible elastic deformation. The die, which has a relatively high length, imposes great pressure falls.

The parameters influencing swelling are: rheo-slope, temperature, L / d ratio of the die, D_c / d contraction ratio, intrusion die angle, die geometry (convergent or divergent), effect of the molecular structure, ingredients, thermal treatment.

Accomplished experimental results [5; 7] for a high density polyethylene shows that the swelling increases simultaneously to the contraction ratio. This can be explained by taking into account the accumulation by the polymeric material of an elastic energy (fig. 4). The

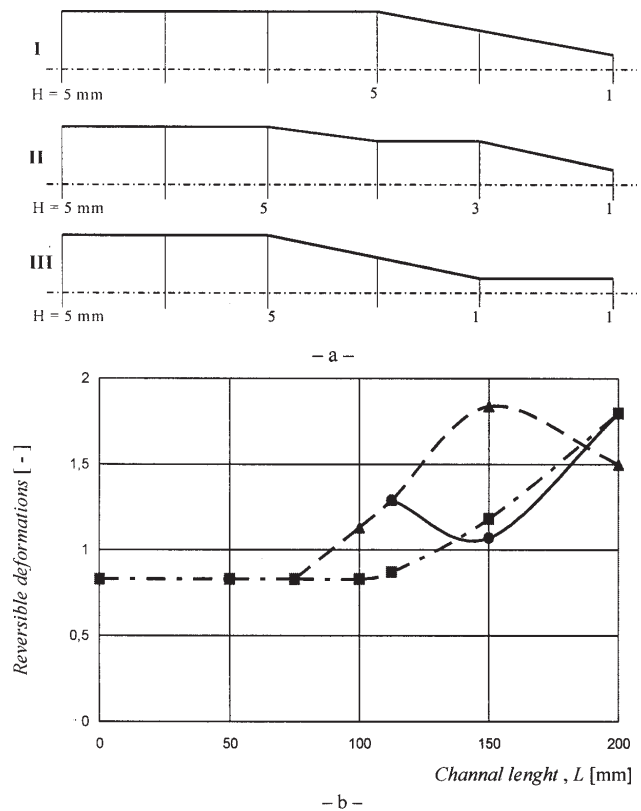


Fig. 3. Medium reversible deformations [3]:
a) – various geometries for the flow channel (variable H channel height);
b) – reversible deformations:
■ – geometry (I); ● – geometry (II); ▲ – geometry (III)

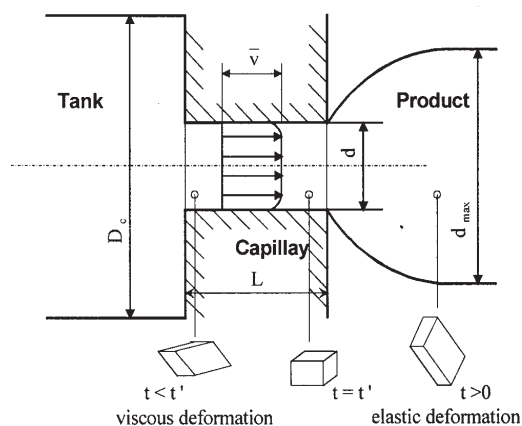


Fig. 4. Elastic recovery of a volume element after viscous deformation [6]

influence of this elastic energy over the swelling disappears when the reservoir diameter has certain values, for which the current lines at the materials intrusion into the capillary do not depend on the geometry of the reservoir.

The study of the "swelling" phenomenon led to the following results:

- for PEId, the swelling increases simultaneously with the D_c / d ratio;
- the swelling decreases if the intrusion angle is $< 60^\circ$;
- for PS, the swelling for a convergent nozzle is less than for a divergent nozzle;
- the phenomenon is influenced both by the molecular structure and by the ingredients used.

A series of researchers have tried to explain the swelling phenomenon and proposed various models presented in table 1.

Table 1

$\sigma(t) = \int_{-\infty}^t M(t-t') C_r^{-1}(t') dt'$	Lodge [8]
$s_w = \frac{(\sigma_{11} - \sigma_{22})_w}{2 \cdot \sigma_{12,w}} = \frac{1}{f} \left[\frac{1+n}{n} \cdot \frac{1+3 \cdot n}{1+2 \cdot n} \cdot \frac{1}{n \cdot B^2} \cdot \left(1+n - \frac{\partial \log B}{\partial \log \left(\frac{8 \cdot v}{D} \right)} \right) \right]$	Metzner ș. a. [9]
$\frac{B - B_\infty}{B_0 - B_\infty} = \exp(-k \cdot t_r)$	Bagley ș.a. [10]
$\frac{W - W_\infty}{W_0 - W_\infty} = \exp\left(-\frac{t_r}{\lambda_a^N}\right)$	Nakajima [11]
$B = \left[1 + \frac{1}{2} \cdot \left(\frac{N_1}{2 \cdot \sigma_w} \right)^2 \right]^{\frac{1}{6}}$	Tanner [6, 12]

In paper [13] there is an extended analysis of the issue of polymeric material melting elasticity and its effects on the quality of the extruded product.

Experimental device

For experimentally determining the behaviour of polyolefins melting a rheo-meter with a capillary was used (piston plastometer, Polireo – 01, fig. 5), designed and made by the Process Equipment department of the Politehnica” University of Bucharest [14].

The device is made up of two distinctive parts:

- mechanical (I);
- automation (II).

The mechanical part of the device achieves the transformation of the granulated or pulverised polymeric material, its extrusion through a capillary and the sampling task. The automation component insures the temperature measurement and adjustment, the establishment of the sampling interval and the command for activating the severing apparatus.

The main components of the mechanical parts and also of the automation part result from figure 5.

On the external cylinder four heating electric fuses are installed, for bringing the material from solid state to melting.

The internal cylinder, in which the polymeric material is introduced, has the following dimensions: internal diameter $D = 9.5_0^{+0.015}$ mm and length $L_c = 171.5 \pm 0.063$ mm. Nozzles were used with the following values of the capillary diameter: $d = 1.27$ mm, $d = 2.16$ mm and $d = 3.63$ mm. Nozzles have different L/d relative lengths.

Operating method

For accomplishing determinations the cylinder is filled with a polymeric material charge. The material is compacted with the help of an auxiliary instrument.

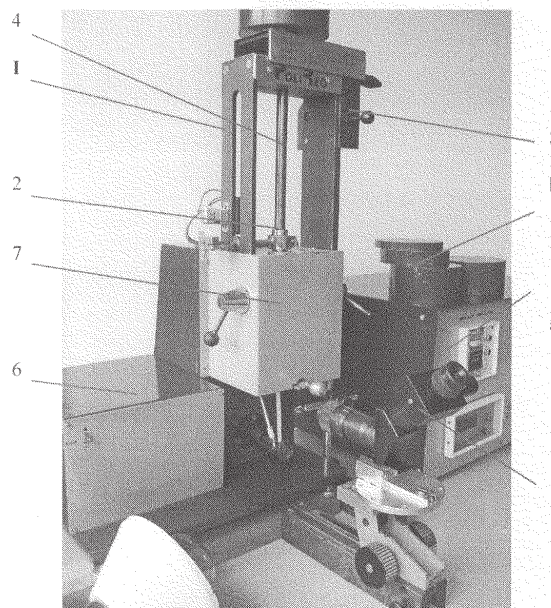
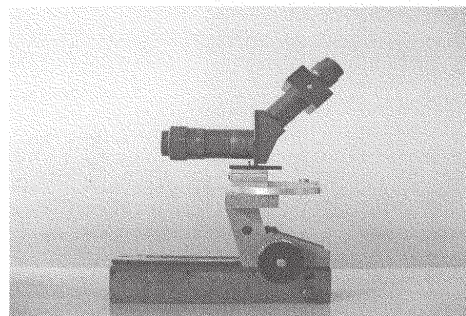


Fig. 5. Piston plastometer (Polireo – 01).

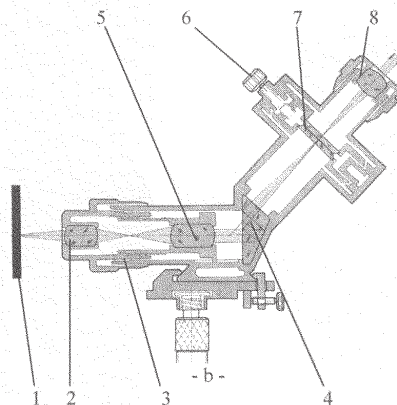
I – mechanical part; II – automation part 1 – crane; 2 – oven; 3 – working weights; 4 – piston; 5 – telescope; 6 – lamp; 7 – ruler for measuring length

Corresponding stages of the operating method are included in papers [14; 15]. The samples for which the swelling corresponding diameter is measured are those corresponding to the standard trip (30 mm) marked by the piston. For the rest of the trip the polymeric material melting is not uniform (the flow is not stabilized).

With the help of an optic device (telescope – picture 5 – fig. 5, fig. 6) a visualisation of the behaviour of the material at the exit from the nozzle was performed (fig. 7), measuring the product diameter and the length of the frost line (3 divisions = 1 mm).



- a -



- b -

Fig.6. The optic device (telescope) used for visualising the behaviour of the material at the exit from the nozzle (picture 5 – Fig. 5: a – view, b – section): 1 – object plan; 2 – objective 1 (6.3 x 0.16); 3 – mechanism for inducting objectives; 4 – deviating prism; 5 – objective II (4 x 0.14); 6 – optical interpolator regulation screw (reticule); 7 – reticule; 8 – ocular

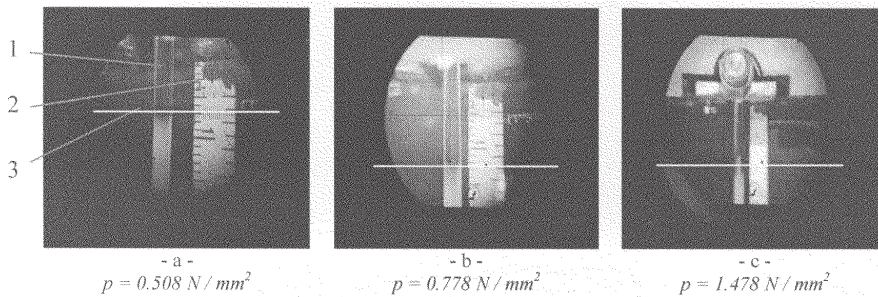


Fig. 7 Profiles of the extruded product, according to pressure, p , for 5200 B high density polyethylene ($d = 2.16$ mm; $L/d = 3.7$; $T = 200^\circ\text{C}$). 1 – profile of the extruded product; 2 – ruler for measuring “frost length”; 3 – position of the “frost line” [16].

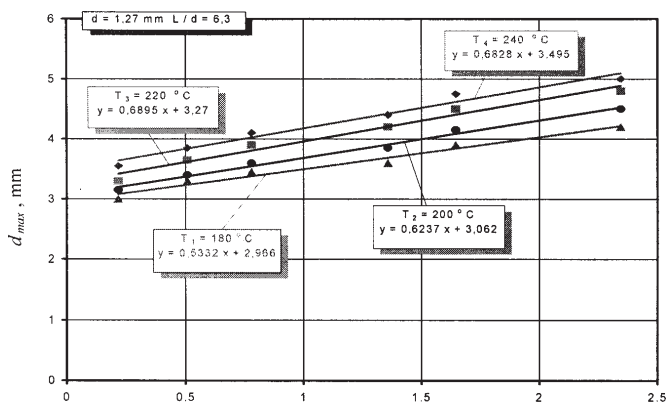


Fig. 8. Variation of the maximum diameter d_{max} after exiting the capillary, according to pressure, p , for 5200 B high density polyethylene. Temperatures: $T = 180; 200; 220$ and 240°C ($d = 1.27$ mm; $L/d = 6.3$) [16]

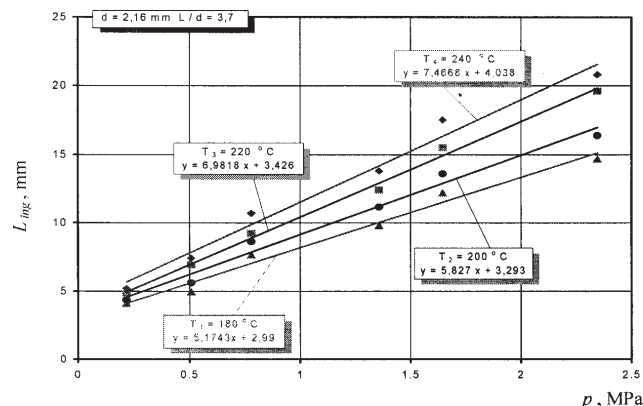


Fig. 10. Variation of the “distance to the frost line”, L_{ing} , according to pressure, for 5200 B high density polyethylene. Temperatures: $T = 180; 200; 220$ and 240°C ($d = 2.16$ mm; $L/d = 3.7$) [16]

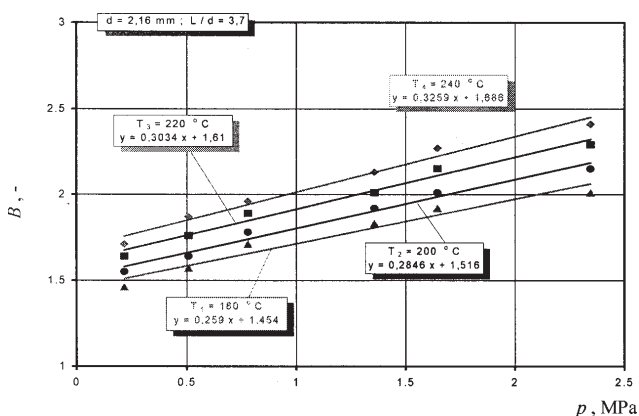


Fig. 9. Variation of the swelling factor, B , after exiting the nozzle, according to pressure, for 5200 B high density polyethylene. Temperatures: $T = 180; 200; 220$ and 240°C ($d = 2.16$ mm; $L/d = 3.7$) [16]

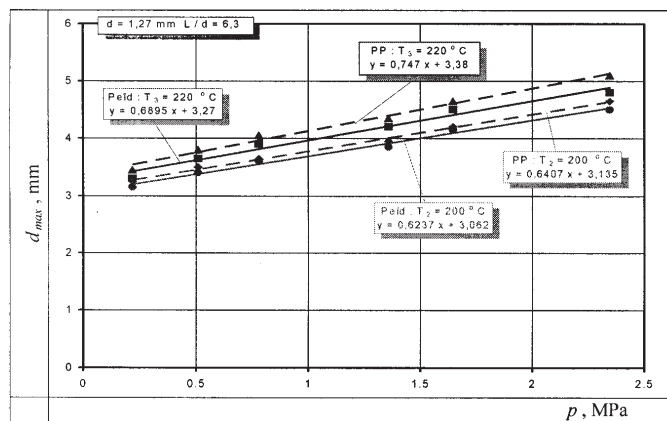


Fig. 11. Variation of the maximum diameter d_{max} after exiting the capillary, according to pressure, p , for FD 620 polypropylene and 5200 B high density polyethylene. Temperatures: $T = 200$ and 220°C ($d = 1.27$ mm; $L/d = 6.3$) [16]

The maximum diameter, d_{max} (fig. 8), and the swelling degree, B , were established, according to the pressure from the capillary for PEid 5200 B, at various temperatures (fig. 9), using 3 types of capillaries with diameters, d , and different L/d ratios (2,3; 3.7; 6.3).

Passing from the melting state to the solid state takes place in the cooling area of the extrudate, limited by the so-called frost line (fig. 8). With the help of the telescope measurements were made in order to determine the variations of the frost line position according to the pressure for various temperatures. For all used capillaries the distance up to the frost line increases simultaneously with increasing pressure. If p increases, then the melting speed increases, which makes L_{ing} increase (fig. 10).

For the two materials analysed (PEid 5200 B and PP FD 620) there was a comparative representation of the maximum diameter, d_{max} , variation, according to the capillary pressure, for two temperatures (fig. 11), at various pressure levels. It was noticed that the swelling, d_{max} , for polypropylene is greater than in the case of polyethylene. It was highlighted the fact that in the case of polypropylene the elastic component is more important than for polyethylene.

Conclusions

It was observed that for a $L/d \geq 60$ any melting stops suffering that phenomenon. But such a fairly high ratio would impose a higher action power for pushing the melting and a reduced speed for avoiding the break-up within the

melting at the surface of the product. For this reason during the building of extrusion dies a compromise is made which leads to a controlled swelling.

During the experiments the following were noticed: in all cases the swelling is linearly increasing simultaneously with the increase in the extrusion pressure; no thermal degradation of the materials was noticed even at relatively high temperatures and high retaining durations.

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