The Influence of Elongational Viscosity on the Quality of Extruded Films

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Introduction and Problem

Knowledge of the elongational viscosity and its influence on the extrusion process is becoming more and more important. Processing a polymer by extrusion as in film casting, film blowing, fiber spinning or thermoforming is affected by the shear and the elongational viscosity of the material simultaneously. While the influences of shear viscosity of polymer melts on the extrusion process is known, detailled work, which describes the influence of the elongational viscosity on the extrusion process, is very rare. As Kurzbeck /KUR99/ reported, polyethylene which gets harder during extension, shows a homogenous deformation of the sample when a film is blown. Considering a fiber spinning process, polymers which have a smaller elongational viscosity have a better draw-down ability /HAN83/, /MEI72/. These authors only describe the influence of the elongational viscosity on an extrusion process after the melts left the die. They do not describe in what way the elongational viscosity affects the melt distribution in an extrusion die and thereby the product quality.

The main reason for the poor number of data at hand is the fact that measurements of the elongational viscosity are more complicated in comparison with measurements of shear viscosity. Moreover the elongational viscosity is considered as uniaxial, planar or equibiaxial viscosity.

Uniaxial elongational viscosity can be determined with tensile tests, in which a sample is pulled /MEI72/. The elongational viscosity is calculated from the resulting stress and strain rate. This method is generally referred as the direct method, because the transient viscosity can be measured with constant strain rate.

The determination of the elongational viscosity from the intake pressure loss resulting from polymer flow through a narrowing section is called the indirect method. In capillary rheometry, the intake pressure loss is determined using the Bagley correction /MAC94/. The intake pressure loss can be seen as a by-product, from which the apparent uniaxial elongational viscosity can be computed. In this experiment the strain rate is not constant. Therefore a mean value is used. The Cogswell approach /COG72/ is in this case the simplest and the most used method to calculate the elongational viscosity. Another approache has been developed e.g. by Binding /BIN88/, Gibson /GIB89/ or Zatloukal /ZAT02/. However different absolute values for the uniaxial elongational viscosities are calculated by the three approaches.

It must be mentioned that both, the direct and the indirect methods, have flaws. While the indirect method works with averaged strain rates and therefore with apparent elongational viscosities, in the direct method the strain rate cannot be regarded as constant throughout the sample.

In extrusion process the effect of the elongational viscosity on the melt distribution in a flat film die and therefore on the resulting film thickness is hardly known. For that reason the goal of the following work is to state, whether and in what way the film thickness distribution and the uniaxial elongational viscosity are linked and whether it is possible to predict the film thickness distribution for a new polymer only with the determination of its shear and elongational viscosity. This is a more detailed work of a report, which will be presented on ANTEC 2004 /GAB04/.

Methods

Rheometer

Laun et al. /LAU89/ published in 1989 a comparison between the direct and the indirect method. To calculate the elongational viscosity with the indirect method they used the Cogswell approach /COG72/. They es-

tablished an agreement between these methods for average strain rates of approximately $\dot{\varepsilon} = 1$ for a limited number of polymers. In the area of low strain rates the elongational viscosities deviated enormously.
With the direct method it is not possible to determine high elongational rates and to measure viscosities at high temperatures. Therefore in this work the elongational viscosity was determined from the intake pressure loss, which is common in capillary rheometry. A twin bore capillary rheometer TBCR (Rosand RH-7, Bohlin) was used (Fig. 1). The corrected intake pressure loss was determined by means of a zero die. The apparent elongational viscosity results from the pressure loss $P_{0,cor}$, the shear viscosity η, the strain rate $\dot{\gamma}$ and n as parameter of the power law /COG72/:

$$\mu = \frac{9}{32}\frac{(n+1)^2}{\eta}\frac{P_{0,cor}^2}{\dot{\gamma}^2} \quad (1)$$

The mean strain rate is calculated as:

$$\dot{\varepsilon} = \frac{4}{3}\frac{\eta}{n+1}\frac{\dot{\gamma}^2}{P_{0,cor}} \quad (2)$$

To describe the differences between the shear and elongational viscosities for the different polymers at high deformation rates the Trouton ratio is used. It is defined as:

$$Tr = \frac{\mu}{\eta} \quad (3)$$

For low deformation rates the value of the Trouton ratio is 3 in uniaxial extensions. It is not a material but a geometrical constant. For instance the Trouton ratio at low deformation rates for an equibiaxial extension equals 6 /MAC94/.
The viscosity $|\eta^*|$ at low shear rates was measured with a cone and plate rheometer (ARES, Rheometrics Scentific, D = 25 mm). The relation between shear viscosity η and complex viscosity $|\eta^*|$ was obtained owing to the Cox/Merz relation /MAC94/. It should be mentioned that the influence of wall slip and compressibility on the viscosity will not be further examined.

Extrusion line

In the film extrusion process a coat-hanger manifold was employed as a flat film die (Fig. 2). It was designed to be independent of the point of operation of the process and yielding constant shear rates in the whole flow channel. The output channel of the die is 150 mm long and 1 mm high. The simulation of the velocity field with the shear viscosity shows different velocities along the manifold, at crossing from manifold to the island and at the exit (Fig. 3). The melt is accelerated and elongated several times along any flow path, so that the elongational viscosity is bound to influence the velocity distribution. When initially the flat die was designed for the coextrusion of three layer films, it was used only for the extrusion of one-layer films by closing other entrances with a dummy plug. The extruded polymer melt cooled on the chill-roll of the haul-off unit. The haul-off velocity was equal the melt exit velocity. The melt temperature amounted T_M = 240 °C, the temperature of the chill-roll T_C = 90 °C.

Material

Detailed work about the influence of shear viscosity on the velocity distribution in a flat film extrusion dies and in a spiral mandrel dies leaded /STI01/. In this work, the velocity profile was computed with the software Polyflow for different flat film extrusion die geometries. The Carreau model was used as material model. A considerable influence of the reciprocal shear rate K_1 on the velocity distribution in the die was observed. At same zero viscosity η_0 and with higher getting reciprocal shear rate K_1, i.e. polymer melts become more non-newtonian for lower shear rates, the velocity distribution in the die gets irregular. The melt velocity is lower at the boundaries and higher in the middle of the flat film die. The gradient n, being measure of the pseudoplastic behaviour, has however a scarce influence on the velocity distribution. In /STI01/ no further research on the influence of elongational viscosity on the velocity distribution was done.

Detailed results about elongational viscosity properties of polymer melts are presented in /KUR99/. Polymer melts with much chain branching develop a strong strain hardening (PE-LD, branched PP) at extension rate as law as $\dot{\varepsilon} = 0{,}01\,s^{-1}$. In polymers with linear chain structure (PE-LLD, PE-HD) a strain hardening occurs only below extension rates of $\dot{\varepsilon} = 0{,}01\,s^{-1}$ or is not observed.

After successive steps of calculation, shear rate in the flat film extrusion die increases to $\dot{\gamma} = 60\ s^{-1}$ and extension rate to $\dot{\varepsilon} = 2\ s^{-1}$. Based on work done by /KUR99/, /STI01/ several polymers were chosen for the investigation presented here (Table 1). Three different types of PE-LD are considered, as high strain hardening was expected. Then a PE-LLD type was chosen, because no strain hardening was expected. Additionally an ABS with marked yield point was chosen, to investigate the influence of the yield point on the film thickness distribution.

Table 1: Characterization of the investigated samples.

polymer	**trade name**	**producer**	**Mw / kg/mol**	**M_w/M_n**
PE-LD	Lupolen 3020D	Basell Polyolefine GmbH	337.5	13.5
PE-LD	Lupolen 3026F	Basell Polyolefine GmbH	242	11
PE-LD	Lupolen 3220F	Basell Polyolefine GmbH	207	9
PE-LLD	Lupolex 18KFA	Basell Polyolefine GmbH	142.8	4.2
ABS	Terluran EGP-7	BASF AG	180	2.6

Results and Discussion

Flow model

The measured viscosities for the used polymers are shown in Fig. 4. To get the Trouton ratio at high deformation rates the description of the measured viscosities is made with the White-Metzner model / BAR92/. The second invariant II_D which describe the deformation rate is equal shear rate $\dot{\gamma}$ in shear flow and in simple uniaxial extensional flow equal to root-three times the strain rate $\sqrt{3}\,\dot{\varepsilon}$. Hence the second invariant II_D will be used instead of the shear and strain rate.

The elongational viscosity is described with:

$$\mu = 2\eta/(1-\frac{2}{3^{0,5}}\lambda II_D) + \eta/(1+\frac{2}{3^{0,5}}\lambda II_D) \qquad (4)$$

where

$$\eta = \frac{\eta_0}{1+(K_1 II_D)^n} \qquad (5)$$

is the shear viscosity and

$$\lambda = \lambda_0/(1+K_2 II_D) \qquad (6)$$

the relaxation time.

The determination of the parameters of eq. (5) and (6) was made gradually. At first parameters η_0, K_1 and n of the shear viscosity were computed. Thereafter λ_0 and K_2 were derived. An important condition of the model is the assumption of the same pseudoplasticity for the elongational and the shear viscosity. As if the molecular chains are completely extended, nothing more can change in its pseudoplastic behaviour /BAR92/.
The determination of λ_0 and K_2 turned out to be problematic because of the formerly presented measurement methods, in which the beginning of the strain hardening and the level of the maximal elongational viscosity could not be measured with the used rheometers. Therefore for low strain rates, values for the elongational viscosity were taken from the literature /LAU89/, /BAS01/ for materials with the same or approximately the same molecular weight respectively, to determine the parameters λ_0 and K_2, which are necessary for calculating the Trouton ratio. It is well known, that the use of the literature data results only in approximate values of λ_0 and K_2, however here these parameters are based only to compute the Trouton ratio at high stress velocity and therefore their absolute values are insigificant. The computed Trouton ratio depends only on the λ_0-K_2-ratio and not on its absolute values.
/BAS01/ showed elongational viscosities for PE-LD 3020D at T = 170 °C for low deformation rates, which are could not be realized with the twin bore capillary rheometer. Thus the temperature shift factor a_T /MAC94/ was used to calculate the data for T = 240 °C and to apply them in our case. The temperature shift factor a_T was derived from the zero shear viscosities at T = 170 °C and T = 240 °C as:

$$a_{T,240°/170°} = \eta_{0,240°} / \eta_{0,170°} \quad (7)$$

Following the elongational viscosity at T = 240 ° was calculated with:

$$\mu_{240}(\dot{\varepsilon}) = a_{T,240°/170°}\, \mu_{170}(\dot{\varepsilon}) \quad (8)$$

Fig. 4 shows a good agreement between measured and computed data for PE-LD 3020D. The Trouton ratio and thereby the strain hardening increases at low deformation rates of $II_D = 0{,}001\ s^{-1}$. For $II_D = 0{,}1\ s^{-1}$ it is nearly constant. This means, that in the extrusion process a strain hardening is observed, because elongational rates up to $\dot{\varepsilon} = 2\ s^{-1}$ are expected.
Only for ABS it was not possible to determine the White-Metzner parameters, because ABS shows a yield point (Fig. 6). Thus the Trouton ratio was calculated by graphical movement of the elongational viscosity on the shear viscosity.

Film thickness ratio

The extruded melt was deposited on the chill-roll of the haul-off system for cooling. However, it was not squeezed between the rolls. Through necking of the film, the film width decreased by 10 mm. A typical film thickness distribution for the tool used here (Fig. 2) is shown in Fig. 5, where the film thickness was standardised for different materials.
The film thickness was standardised to correct differences of the total film thickness, which can arise through the pulsation of the extruder or through the irregular movement of the haul-off system. For each polymer 5 measurements of the film thickness distribution along the film width were done to calculate its mean value.
The film thickness plotted versus the film width resembles a Gaussian. The dimensionsless film thickness decreases from the middle to the side edges and rises at the edges.
To evaluate the film thickness distribution a maximum-minimum ratio was employed. It is given by the maximum value d_{max} in the middle of the film and the minimum values d_{min1} and d_{min2} at the film edges:

$$Q = \frac{d_{max}}{(d_{min1} - d_{min2})/2} \quad (9)$$

The different minima are caused by discrepancies in the die geometry.

Furthermore the neck-in behaviour of the polymer melt must be mentioned. The necking is a contraction of the melt perpendicular to the extrusion direction after leaving the die. The intensity of the necking increases with the haul-off velocity. The necking is also influenced by the cooling conditions and by the material properties like the elasticity or the relaxation spectrum. The polymer chains are stretched in the flow channel during the extrusion, where the relaxation can arise already, but it has not to be finished, so that the polymer chains also relax after leaving the die. The dependence of the extrusion parameters and the material properties is unknown so far.

Correlation of the film thickness ratio with the shear and elongational viscosity

A comparison of the film thickness ratio with the data of shear and elongational viscosity, i.e. zero viscosity, Trouton ratio and reciprocal shear rate is shown in Fig. 6 for the investigated polymers. The film thickness ratio corresponds to the reciprocal shear rate K_1. From there the theoretic work of /STI01/ can be proved by this experimental research. No correlation is observed between the film thickness ratio and the zero viscosity or the Trouton ratio. Also the assumption can be excluded, that high shear viscosity causes a deformation in the middle of the flat film die. A widened die would lead to an unsteadily film thickness distribution. But the shear viscosity of the PE-LLD 18KFA is always higher than the shear viscosities of PE-LD 3026F and PE-LD 3220F and its film thickness distribution is more equably.

Fig. 6 also backs the hypothesis, that the film thickness ratio does not depend on the elongational viscosity, which was described with the Trouton ratio as the measure of the strain hardening at high deformation rates. This is made clear with the comparison by the PE-LD 3026F and PE-LD 3220F. Both polymers have nearly the same shear viscosity but the PE-LD 3026F shows a much higher strain hardening. However, the higher strain hardening does not influence the film thickness distribution. Parameter n is a measure of the pseudoplasticity. For the investigated polymers the value $n \sim 0.6$ was similar and therefore negligible.

In reason of the specified results, a statement can be made, that the elongational viscosity has insignificant or not influence on flow distribution in the used flat film die and therefore on the film thickness distribution. Generally, a film thickness distribution of a polymer can be predicted only through the consideration of the reciprocal shear rate K_1.

This assumption is confirmed when looking at the film thickness distribution of the ABS with marked yield point. A comparison of the film thickness distribution of ABS film with the PE-LD 3020D shows a nearly pyramid-shaped film thickness distribution (Fig. 5) which is caused by the yield point as a kind of strong pseudoplasticity.

As already mentioned, irregularities seem to be present in the tool geometry, which are responsible for the observed asymmetrical film thickness distributions. Checking the gap with a gauge resulted in a difference of approx. 20 μm, with the exit slit being smaller on the right side (positive values in Fig. 5), so that a displacement of the flow to the left half of the slit should be the consequence.

A simulation of the flow process including the extension viscosity led /GUP03/. The result was a hardly changed velocity profile when using the elongational and shear viscosity simultaneously, instead of shear viscosities only. The findings in /GUP03/, too, back the experimental results presented here as well as their interpretation. The main found in /GUP03/ when using elongational viscosity data, was in pressure consumption. In this case the outcome of the simulation was a rise of the maximal pressure in the die by about 17 % compared to the pressure calculated by using shear viscosities only. Therefore a higher mechanical strength of the die has to be considered during designing.

Conclusion

The shear and elongational viscosities of different PE-LDs, a PE-LLD and an ABS have been determined with a twin bore capillary rheometer The zero shear viscosity was measured with a cone and plate rheometer. The viscosities have been described with a White-Metzner model to calculate the Trouton ratio as a measure for strain hardening. The strain hardening depends on the deformation rate and becomes constantly at high deformation rates. With a flat film die different films were extruded and the film thickness distribution along the film width was measured.

The film thickness ratio was compared to the flow characteristics of the polymers. The result is a dependence of the film thickness ratio only on the reciprocal shear rate K_1 and not on the elongational viscosity. For the prediction of the film thickness distribution of a new polymer it is sufficient to consider the reciprocal shear rate K_1. The elongational viscosity contributes only to a higher pressure in the flat film die, which has to be considered during the die designing.

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Key Words

Shear viscosity, elongational viscosity, White-Metzner model, flat film die, film thickness distribution

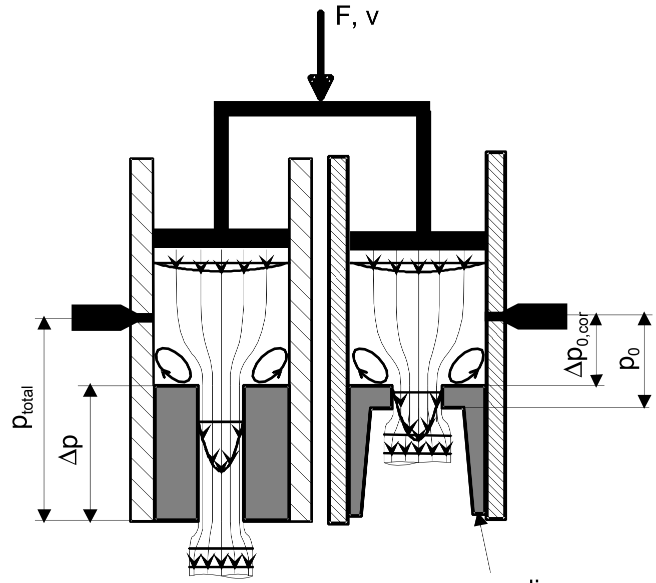

Fig. 1: Schematic drawing of a twin bore capillary rheometer (TBCR).

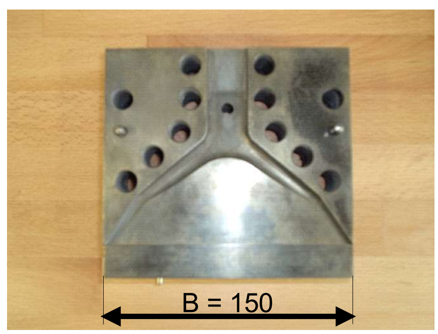

Fig. 2: The flat film die with coat-hanger manifold.

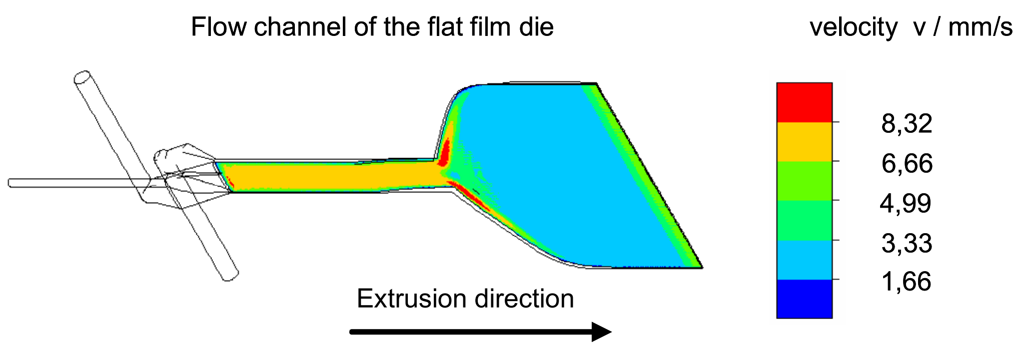

Fig. 3: Velocity distribution in the flat film die.

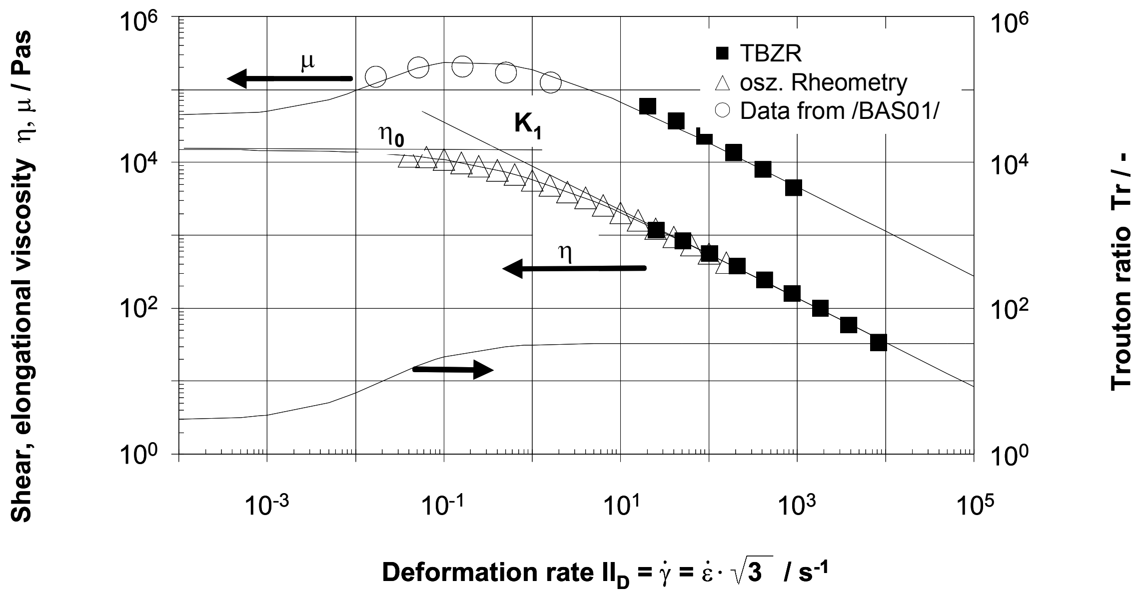

Fig. 4: Calculation of the viscosities with the White-Metzner model for PE-LD 3020D.

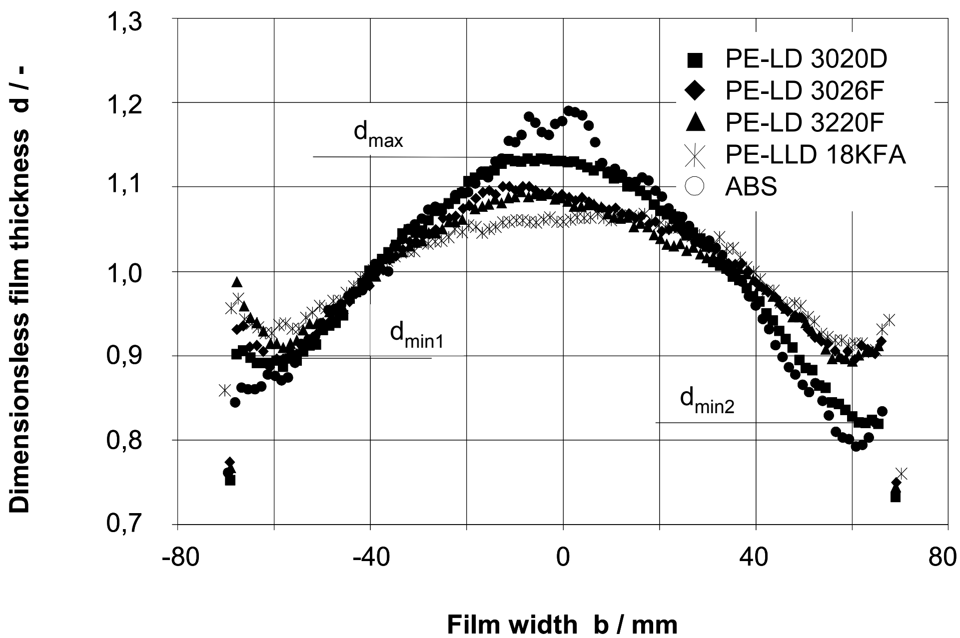

Fig. 5: Film thickness distribution; d_{max} and d_{min} given for PE-LD 3020D.

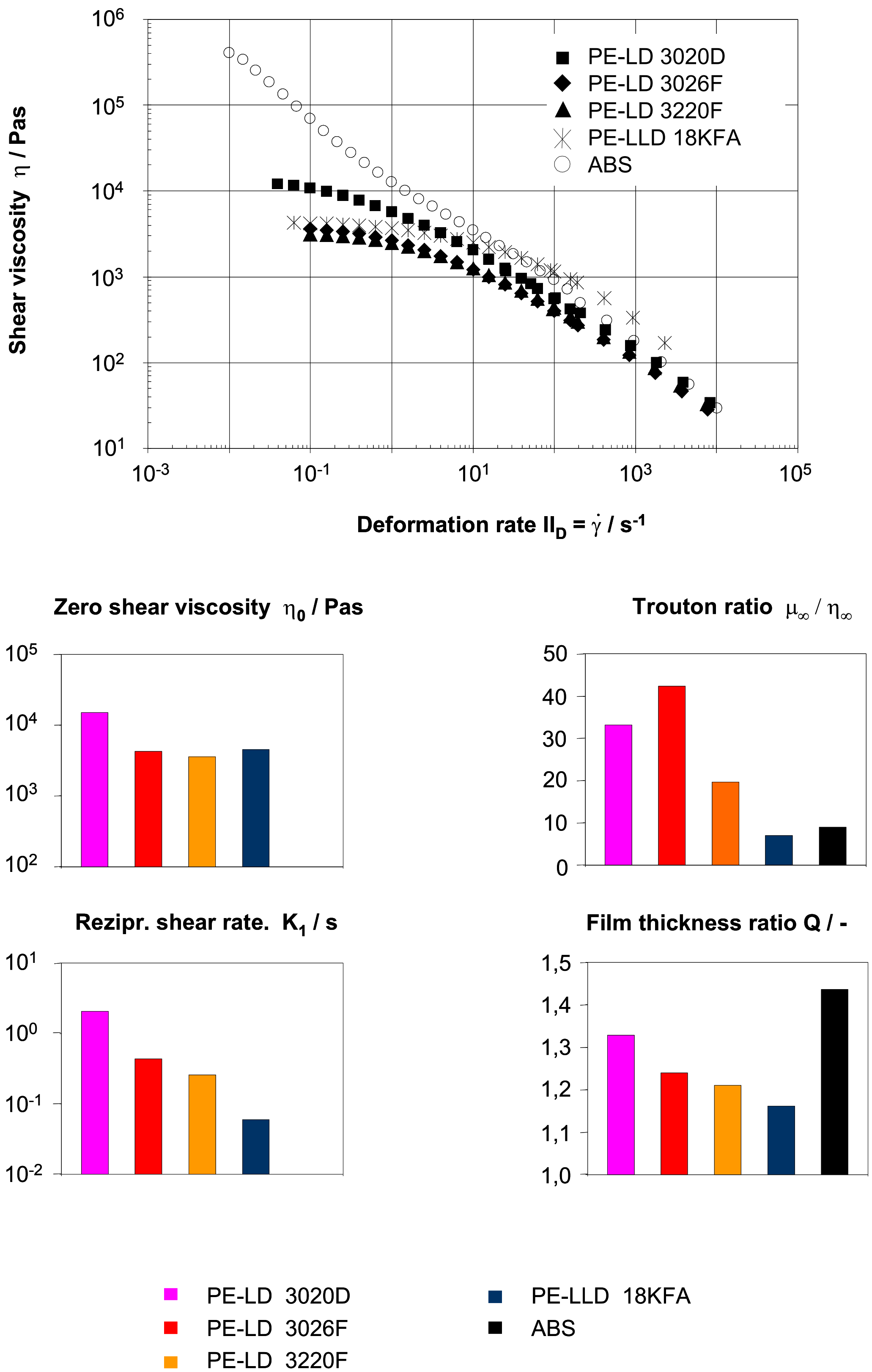

Fig. 6: Correlation of the film thickness ratio with the shear and elongational viscosity.