RELAXATION ZONES IN EXTRUSION DIES

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Abstract

When designing extrusion dies very often only the flow in the die is balanced. To avoid polymer depositions in the die the wall shear rate and the wall shear stress are calculated. For minimal internal stresses in the extrudate a relaxation zone upstream of the land length is helpful. The characteristic values for designing the relaxation zone are the relaxation time and the critical shear rate. The calculation used is the finite volume element method.

Introduction

In the design of extrusion dies, mostly only the viscous flow of polymers is considered. Very often, only the throughput and pressure drop are calculated. This is unsatisfactory because the cleaning of the die surfaces is not considered. To avoid material depositions the wall shear rates and the wall shear stresses have to be large enough to clean the surfaces properly. For that so-called land length there are empirical values for the ratio of length to gap. The dimensions of the product are essentially determined by the gap. The critical shear rate for relaxation and the residence time must be reached independently.

It makes sense to take into account material functions and their characteristic values for the designing the relaxation zone upstream of the land length. Residence time and shear rate - taking account of decomposition and cleaning - can be chosen freely. For a material-oriented design of the relaxation zones in dies the characteristic relaxation time and the critical shear rate at which the macromolecules reset from stretching have to be known. The methods for determining the characteristic rheological material functions and characteristic values are explained and numerical values for various polymers will be given. As an example, a micro tube extrusion die is calculated, and improved. Originally the die had only a short production time. The wall shear rates and the residence times can be calculated using FVE (finite volume element) method.

Viscoelastic Properties of Polymer Melts

The specific rheological properties of the melt result from their macromolecular structure. The macromolecules of a polymer are formed by a flexible polymer chain with short chain segments (Figure 1a). Each segment has ten monomer units of the polymer which are combined and characterized by an arrow. Its length and orientation to adjacent arrows can change arbitrarily. At rest these segments form a three-dimensional random coil which can be stretched and tensed by macroscopic deformations in a shear or an elongational flow.

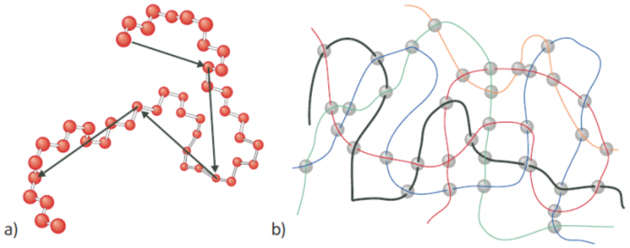


Figure 1. Schematics for macromolecules: a) schematic representation of segments of a macromolecule in a polymer melt, and b) schematic representation of an entangled polymer network [1-3].

In a concentrated polymer solution or melt, many polymer chains are adjacent. Entanglements occur through the micro-Brownian movements and the diffusion of the flexible chains (Figure 1b). The entanglements of the macromolecules generate a three-dimensional polymer network with temporary physical knots that determines the deformation behaviour of the polymer melt. The polymer network has elastic and viscous properties. The elastic deformation behaviour dominates at short loading times with high strain rates the viscous flow predominates at long loading times with low strain rates.

The deformation of the macromolecules leads to stresses which relax after the cessation of the loading. The relaxation process in the polymer network is complex. The characteristic material parameter for this is the relaxation time.

A polymer melt has not a uniform relaxation time but has a relaxation time spectrum which results from different spatial configurations and points of attachment of the macromolecules. In order to illustrate this one introduces a model for the elastic stretching and the viscous flow of the polymer network. The Maxwell model describes the spring-dashpot behaviour of this network (Figure 2).

The spring describes the elastic behaviour for stress during short-time loading. The dashpot describes the viscous flow during long-time loading. For a real melt one has to turn more spring-dashpot models parallel switched to include the influence of the broad distribution of different molecular configurations (like molecular weight distribution and long chain branching).

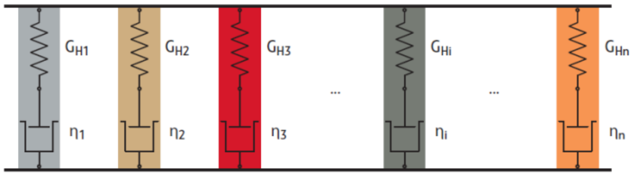


Figure 2. Parallel switched Maxwell models.

Figure 2 shows n parallel switched Maxwell models with the individual spring constant G_{Hi} and the relaxation time $\lambda_i=\eta_i/G_{Hi}$. If the polymer melt flows through a die, the molecules are stretched more or less according to the loading and the entanglements sliding on each other. According to the model the viscosity stands for sliding of the entanglements in the network. If one reduces the stress the macromolecules turn back and achieve the coil state. The complex theory is simplified and the correlations in the stationary shear flow will be demonstrated. The polymer melt is primarily sheared stationary in an extrusion die. The flow is rheological fully developed. The relationship between viscosity and shear rate is described with the viscosity curve (Figure 3).

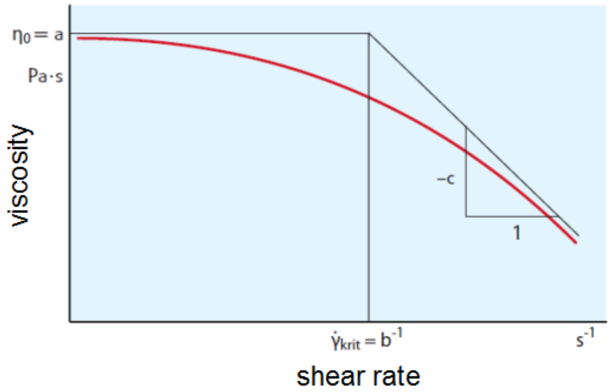


Figure 3. Viscosity curve of a polymer melt, approximated with the Carreau-formula [3].

At very low shear rates, the viscosity has the highest value. The entanglement can develop fully. With increasing shear rate the macromolecules are stretched and the entanglement cannot be developed fully after slipping on each other. The melt viscosity of the polymer decreases.

The viscosity at low shear rate is defined as the zero shear viscosity. It is obtained from the viscosity function with the plateau value of shear modulus G_0 and a characteristic relaxation time according to the equation (2) and (3).

$$\eta_0 \approx G_0 \cdot \lambda_{char} \tag{1}$$

Eq. (1) is used to determine the characteristic relaxation time of the polymer melt

$$\lambda_{char} \approx \frac{\eta_0}{G_0} \tag{2}$$

The shear viscosity function of a polymer melt as shown in Figure 3 is described for calculations with the Carreau formula:

$$\eta(\dot{\gamma}) = \frac{a}{(1 + b \cdot \dot{\gamma})^c} \tag{3}$$

The viscosity curve shows the shear rate, where the macromolecules are stretch reversible. The intersection of the horizontal tangent in the Newtonian flow region at $\eta(g=0)=\eta_0=a$ and of the asymptote with the slope -c for the shear thinning region is g_{krit}^{-1} and characterized by the parameter b. This value represents the transition from the Newtonian to the pseudo plastic flow behaviour (shear thinning) and represents a shear rate, which should not be exceeded if the shear-induced molecular orientation is compensated by entanglements. In the area $g_w \leq g_{krit}$ the entanglements arise through permanent dissolving and developing of physical knots in the polymer network exclusively over time. This is called the linear visco elastic deformation behaviour of the melt.

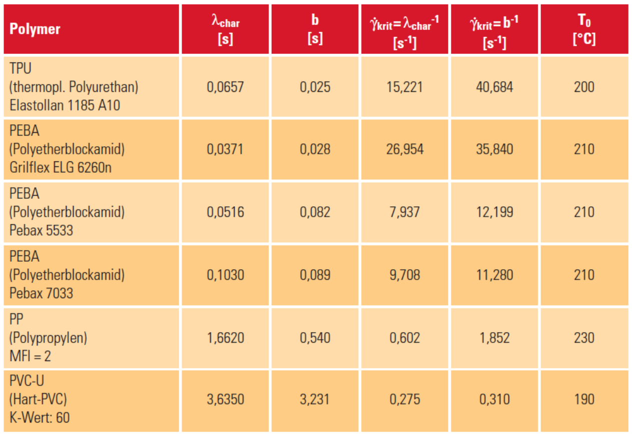
The critical shear rate g_{krit} has the dimension of s^{-1}. Its inverse g_{krit}^{-1} can be considered as a kind of characteristic relaxation time of the polymer melt. In the relaxation zone of a die at their exit a uniform stress and orientation state of the polymer network should be reached. This time for relaxation is required. This relaxation time is correlated with the characteristic relaxation time λ_{char} obtained from oscillatory experiments. The parameter b is used, as well as λ_{char}, as limiting criterion of wall shear rates and mean residence times in relaxation zones.

Characterization of the Flow Behaviour and the Viscoelastic Properties of Selected Polymer Melts

Sophisticated products - such as micro tubes in medical technology - must have a uniform state of orientation of the macromolecules. For the design of the relaxation zone for the extrusion die one needs quantities for the relaxation time and the limitations of the shear rate.

Table 1 contains the characteristic relaxation time λ_{char} - determined from the discrete relaxation time spectrum [4-6] - and the parameter b of the Carreau formula [3] for six polymers.

Table 1. Characteristic relaxation time λ_{char}, Carreau parameter b and resulting critical shear rates $\mathbf{g}_{krit}$ for six polymers.



Polymer	λ_{char} [s]	b [s]	$\dot{\gamma}_{krit}=\lambda_{char}^{-1}$ [s^{-1}]	$\dot{\gamma}_{krit}=b^{-1}$ [s^{-1}]	T_0 [°C]
TPU (thermopl. Polyurethan) Elastollan 1185 A10	0,0657	0,025	15,221	40,684	200
PEBA (Polyetherblockamid) Grilflex ELG 6260n	0,0371	0,028	26,954	35,840	210
PEBA (Polyetherblockamid) Pebax 5533	0,0516	0,082	7,937	12,199	210
PEBA (Polyetherblockamid) Pebax 7033	0,1030	0,089	9,708	11,280	210
PP (Polypropylen) MFI = 2	1,6620	0,540	0,602	1,852	230
PVC-U (Hart-PVC) K-Wert: 60	3,6350	3,231	0,275	0,310	190

Examples are given for a thermoplastic polyurethane (TPU), three polyether block amides (PEBA), a polypropylene (PP) and a rigid PVC (PVC-U). The $\mathbf{g}_{krit}$ -values calculated from λ_{char} and b are in the same order. So one can use $\mathbf{g}_{krit}$ calculated from the steady shear viscosity function for a rheological correct design of the relaxation zones.

Figure 4a shows the storage (G') and the loss modulus (G'') and the complex shear viscosity (η^*) of a rigid PVC.

Since PVC compounds decompose rapidly, the measurement times for the rotational rheometer must be restricted. The frequency range ω=1-100 rads^{-1} was determined for this rigid PVC and the measurement - including melting and tempering of the sample - in seven minutes performed. λ_{char}=3.635 s for these PVC-U which corresponds to a critical shear rate $\mathbf{g}_{krit}$ = 0.275 s^{-1}.

The complex shear viscosity η^* in Figure 4b can be interpreted as steady shear viscosity of the PVC melt for $\omega=\mathbf{g}$ [7] and approximated with the Carreau formula [3]. The parameter b=3.231 s is in this approach. The reciprocal value of this gives a critical shear rate is $\mathbf{g}_{krit}$=0.31 s^{-1}. For the design of relaxation zones in extrusion dies shear rates and mean residence times should be restricted to these values (except a thin rand zone near the wall of the die).

Design of a Micro Tube Extrusion Die

Micro tubes used for medical and technical applications have very high requirements in terms of their structure, surface quality and dimensional accuracy. They can fulfil this only if the micro tubes are produced from a high quality polymer with a uniform orientation of the macromolecules in extrusion and circumferential direction. The example of a micro tube extrusion die is based on a die design concept, in which the wall shear stresses are high enough to clean the flow channel walls and the macromolecules relax before the land length.

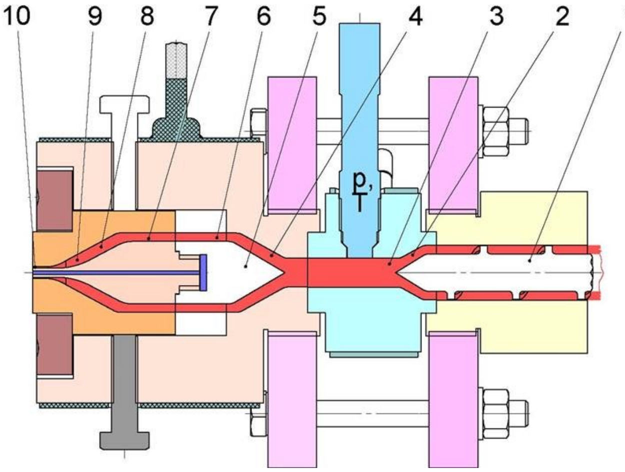


Figure 5. Micro tube extrusion die: 1) Extruder, 2) manifold, 3) measuring position for pressure and temperature, 4) divergent-conical annular gap around the mandrel tip, 5) mandrel, 6) spider-holder for the mandrel, 7) cylindrical annular gap after the spider, 8) relaxation zone, 9) transition zone ring with decreasing gap height, and 10) land length.

Micro tubes for medical applications are often made of PVC-U with a miniaturized die as shown in Figure 5. The melt processed by the extruder flows through a well-rounded adapter for cleaning of the screw tip and a measuring position for pressure and temperature with a rectangular cross section into the pipe head. The tube tool consists of few parts as possible in order to reduce the number of joints. In the land length, the gap must be completely cylindrical. Already a very small concentricity of the channel will distort the pipe with smallest wall thickness at the die exit. Mandrel tip and spider-holder are in one piece. Upstream of the land length a relaxation zone is positioned in which a sufficient residence time is given for relaxing of the macromolecules (Table 1). The critical shear rate should not be exceeded. The volume of this relaxation zone is low compared with the total volume of the flow channel. The length of the land length is extremely short compared to conventional designed dies. The pressure is considerably lower than usual. The channel heights are chosen so that the wall shear rates in each zone upstream of the land length are constant. For the required flexibility in throughput the shear rate is chosen between g_w=8 s^{-1} and 20 s^{-1}. The extrusion die has a sufficiently wide operation range.

Verification of the Design Concept for a Micro Pipe Extrusion Die

Two different die were analysed (Figure 5). The original version (version 1) had deposits in the flow channel and the extruded tubes were deformed because of unequal internal stresses. The modified tool (version 2) was improved by consideration of rheological view points, which has been proven in the processing praxis. The design was carried out analytically. With the presented calculation method one can optimize the flow channels.

With help of the software package INIX the flow channels for a polypropylene melt were calculated numerically in the zones of the extrusion die. The discretization of the different flow channels was performed using the finite volume element method (FVE). Local velocities, shear rates, wall shear stresses, residence times and pressure drop in the flow channels of the extrusion die were calculated three-dimensionally.

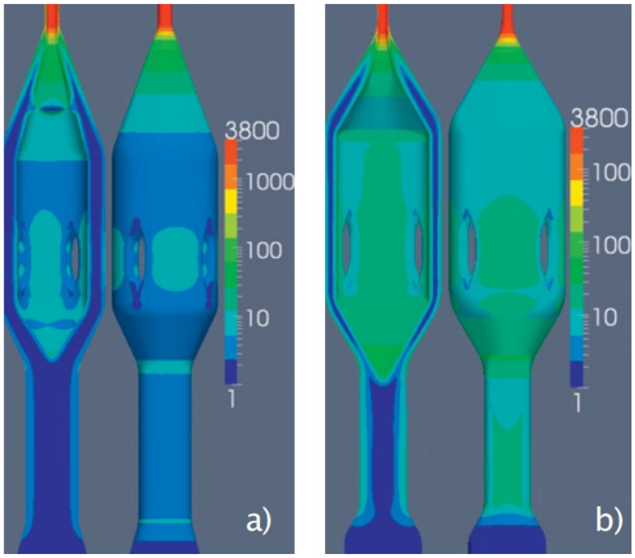
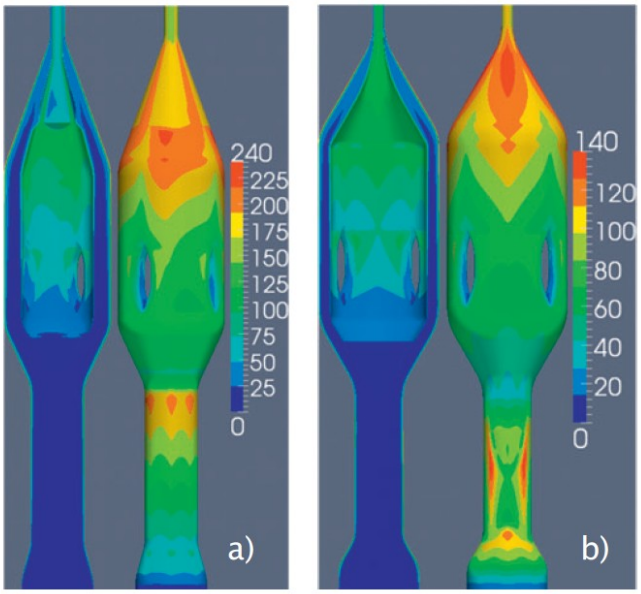


Figure 6. Shear rate distribution over the cross section (left) and on the surface (right) for a) the original version 1 and b) the modified version 2.

Figures 6a and 6b show the distribution of the shear rates for the two versions of the die. The wall shear rates are too low in the original version (Figure 6a, with 5 mm channel height). The surfaces are not sufficiently cleaned (insufficient cleaning flow). The modified version (Figure 6b) has a significantly higher of shear level because the flow channels have 3 mm heights. The computational method can be used for optimizing the radii, the mandrel tip and the spider.

The mean residence time is shown in Figures 7a, b. The modified die has a significantly shorter mean residence time. On closer analysis of the local residence time one can see stagnation zones. The analysis of the local residence time shows stagnation zones. Here is the information of the residence much more precise than the shear rate. The red coloured area shows very long residence time at the entrance of the relaxation zone. The radius was chosen to small at this position with 12 mm. In version 3 the radius of 16 mm avoids this error.

The difference in the two versions shows a direct comparison of the residence times and the shear rates over the die length (Figure 8). The residence times in the improved version 2 are significantly shorter, and the shear rate range is controlled higher. The very high shear rates in the land length are unavoidable because of the requested geometry of the tube.



Figures 7a, b. Residence time over the cross section (left) and on the surface (right) for a**)** the original version 1, and b**)** the modified version 2.

Summary

The flow channels are calculated by finite volume element method. The process optimization is performed by a material-related, rheological correct design of relaxation zones of the channels. The characteristic relaxation time λ_{char} of a plastic melt can be obtained either from the relaxation time spectrum determined in oscillatory shear experiments or from steady shear viscosity function measured using capillary rheometer. The goal is to design a relaxation zone. The melt entering the land length die should be a completely relaxed polymer network with entangled macromolecules. The required uniform molecular orientation in the extrudate is ensured at the extrusion die exit and semi-finished products can be produced with less warpage and more homogeneous structure. Characteristic relaxation times for a number of polymers are specified. It is shown how the wall shear rates, the wall shear stresses and the mean residence times can be calculated with the FVE method. The flow channel geometry is rheological correct with respect to the shear rates.

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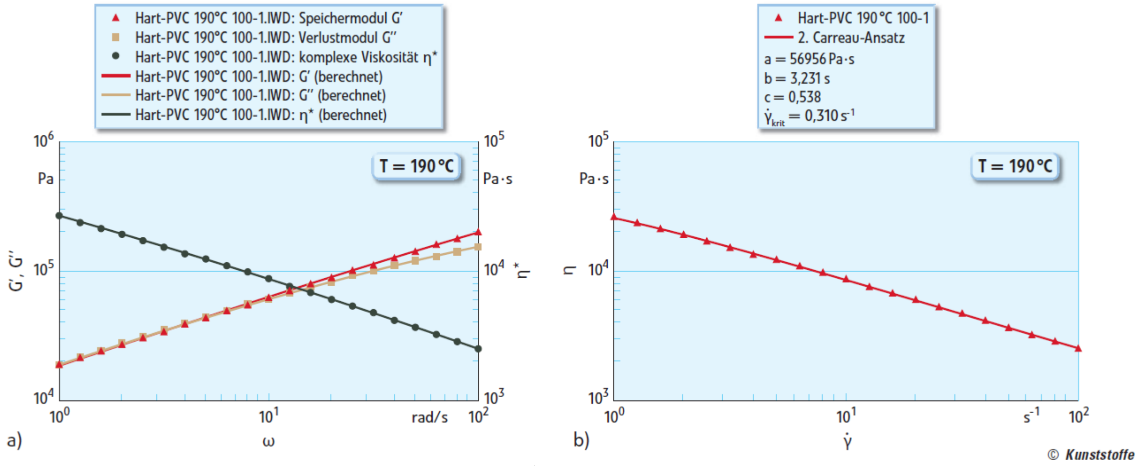

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Figures 4PV-melt rheology: a) $G'(\omega)$, $G''(\omega)$, and $\eta^{*}(\omega)$, and b) $\eta(g)$ for the PVC-U melt [7].

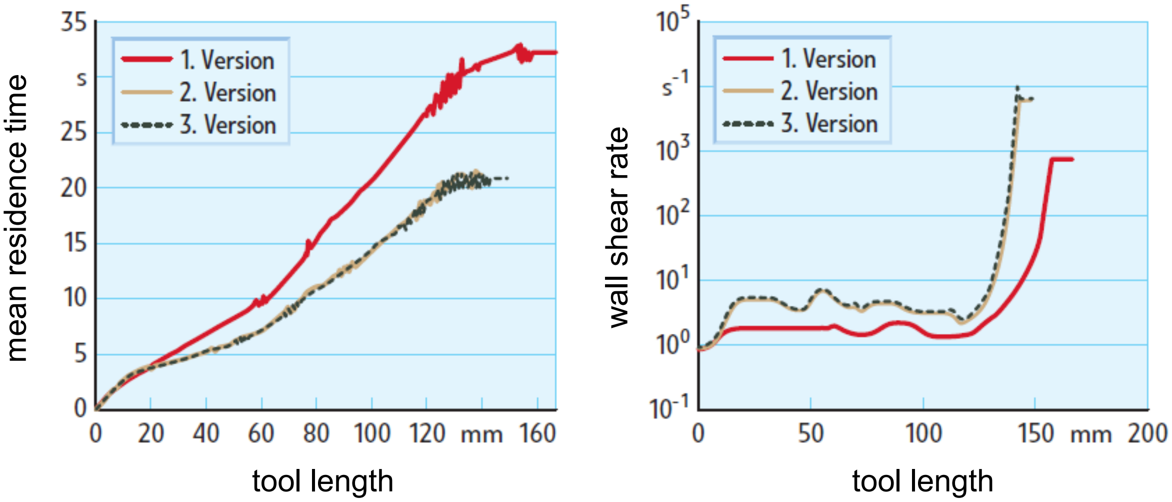

Figure 8. The mean residence times and the shear rates along the die length for tree different versions of the tube die.