MEASURING FIRST NORMAL STRESS DIFFERENCE AT HIGH SHEAR RATES VIA CAPILLARY RHEOMETER

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ABSTRACT

First normal stress difference is commonly measured in cone-plate geometries up to the shear rates of around 10 s^{-1} with rotational rheometers. At higher shear rates the measurement is limited either by the torque or normal force threshold of the instrument or by some material related limitations e. g. edge fracture. A new Normal Stress die designed to simultaneously measure steady-state shear viscosity and first normal stress difference at higher shear rates (>>10 s^{-1}) via capillary rheometer is first introduced.

Measured data of first normal stress difference are then correlated to the onset of flow instabilities like shark skin of plastics and poor extrusion of rubber compounds detected by Garvey die. Here dimensionless numbers introduced to increase selectivity. First normal stress difference measurement also opens an easy and effective way to analyse the die drool effect.

NORMAL STRESS DIE DESCRIPTION AND VALIDATION

The normal stress die was developed in collaboration with Karlsruhe Institute of Technology (KIT), institute for Chemical Technology and Polymer Chemistry (ITCP)[1].

Normal Stress die is composed of two parts, slit part and radial part: See **Fig 1**. Steady-state shear viscosity is measured in the slit part and first normal stress difference is measured in the radial part.

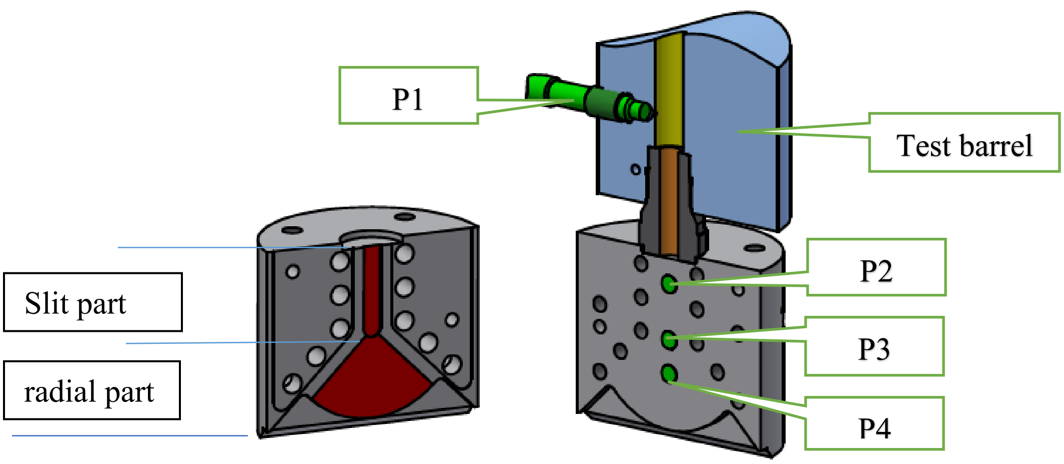

FIGURE 1: Open parts of the slit-radial die. (a) Channel half and (b) Sensor half.

Steady-state viscosity is calculated slit part: See Eq (**1**). The Bagley correction is not necessary since the pressure is measured with the pressure difference of the pressure transducers inside the die (P_3-P_2). The steady-state viscosity is obtained after Weissenberg-Rabinowitsch correction.

$$\eta_a = \frac{\sigma_a}{\dot{\gamma}_a} = \frac{\Delta P_{slit}}{L}\frac{H^3\,W}{12Q} \qquad (1)$$

Elongation viscosity is calculated using the Cogswell model from the entrance pressure loss measured between P1 in the barrel and the extrapolated pressure at the entry of the capillary extrapolated from pressure P2 and P3.
First normal stress difference is calculated in the radial part of the Normal Stress die by Eq (2).

$$\langle N \rangle = 0.1\left[-r_2\frac{P4-P3}{r_3-r_2}+\frac{(2n+1)}{n}\left[\frac{Q}{\pi h^2\,r_2}\frac{2n+1}{n}\right]^{n-1}\right] \qquad (2)$$

Where $\langle N \rangle$ is the average first normal stress, r_2 and r_3 are the radii in which the second and third pressure sensors are located, Q is the volumetric flux, P3 and P4 are the pressure values of third and fourth pressure sensors, h is the height of radial part and n is the power law index.

Steady-state first normal stress difference as a function of shear rate is obtained from transient shear experiment with 13 mm cone-plate geometry and Normal Stress die at 230 °C for polypropylene: see **Fig. 2**.

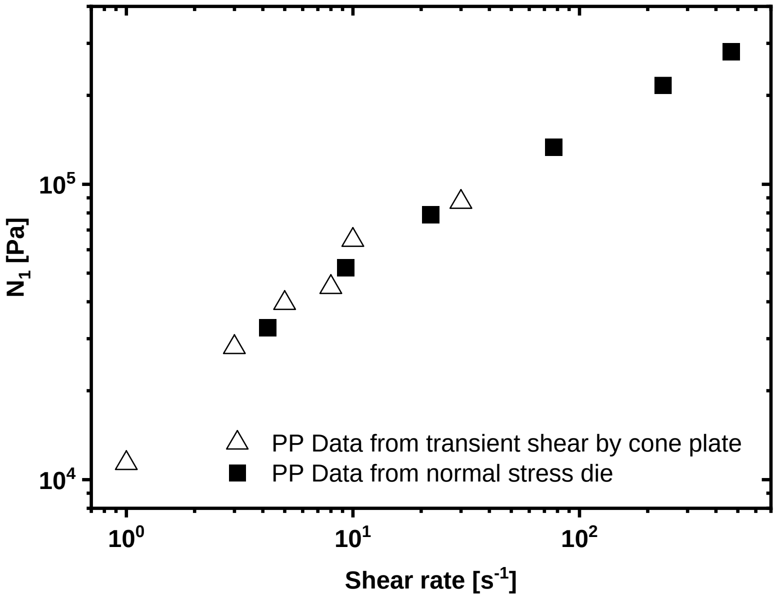

FIGURE 2: First normal stress difference from different methods vs shear rate for LDPEs at 230 °C.

FLOW INSTABILITIES AND FIRST NORMAL STRESS DIFFERENCE

The prediction of flow instabilities is one of the target tasks in extrusion quality. The effect of rheological properties like viscosity, entrance pressure loss/elongation viscosity and 1st normal stress difference is first illustrated using the measurement with normal stress die at 6 LDPE and detected flow instabilities. Flow instabilities are detected with the shark skin die developed in collaboration with KIT[2] mounted on a capillary rheometer with the geometry of 3/0.3/30 mm/mm/mm W/T/L slit die. Bagley and Weissenberg-Rabinowitsch corrected steady-state shear viscosity of the samples at 180 °C. is plotted versus shear rate: See **Fig 3**. Data of sample

3-4 and sample 5-6 show about the same behaviour. The onset of instabilities in a 3/0.3/30 mm/mm/mm W/T/L slit die at 180 °C is reported in **TABLE 1.**

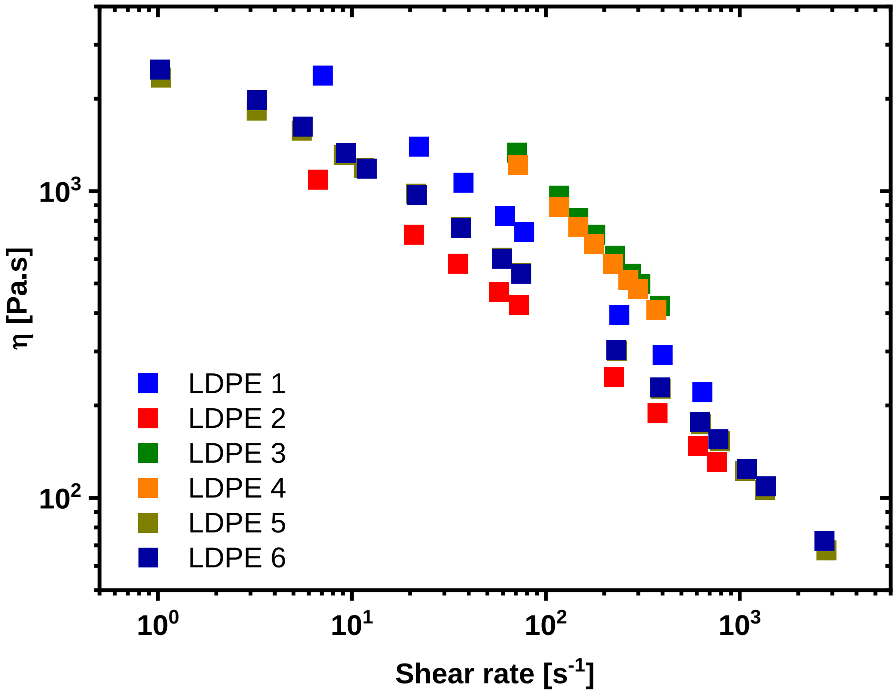

FIGURE 3: Steady-state shear viscosity vs shear rate of LDPEs obtained from slit part of the normal stress die at 180 °C.

TABLE 1: The onset of instabilities for different LDPEs at 180 °C.

Material	**Onset of instabilities (Shear rate) at 180 °C with slit die [s^{-1}]**	
LDPE 1	**250**	 
LDPE 2	**Not seen**	
LDPE 3	**20**	 
LDPE 4	**80**	 
LDPE 5	**Not seen**	
LDPE 6	**500**	 

Viscosity data cannot differentiate between samples in the same order as detected by shark skin analysis. Entrance pressure loss shows a better differentiation of the samples in regard of flow instabilities but cannot fully differentiate between the samples.

The measurement of 1st normal stress difference can differentiate between the samples. To enlarge the differences the data are divided by entrance pressure loss, which also influences instabilities. The number N_1/dP_e is dimensionless and plotted vs shear rate for the 6 LDPEs; See **Fig. 4**. The data show the same ranking as the data in table 1. The ratio of N_1/dP_e gets higher when the onset of instabilities happens at higher shear rates.

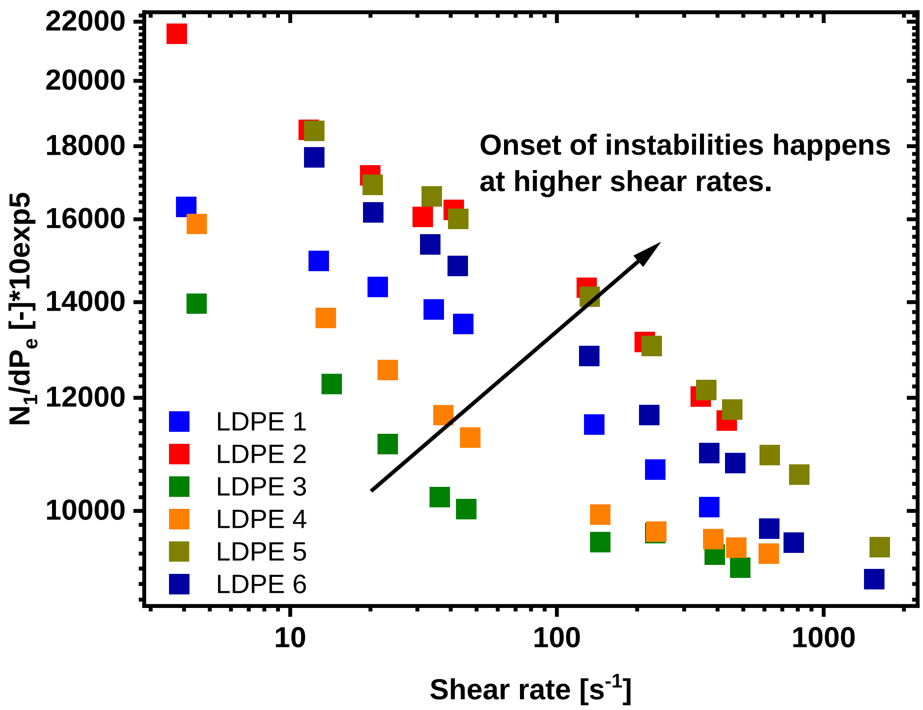

FIGURE 4 First normal stress difference divided by entrance pressure loss (dP_e) as a function of shear rare at 180 °C for different LDPEs.

NORMAL STRESS DATA COMPARED WITH GARVEY DIE EVALUATIONS

The quality of rubber extrusion compounds can be analyses by Garvey die measurement. The method is done in a manual operated time-consuming procedure with often even manual evaluation. This time-consuming evaluation of extrudate quality by Garvey die is performed on four different rubber compounds[3]: See **Fig.5**. Two pairs of material one with good and one with bad extrusion quality can be seen. The materials are then also measured by normal stress die in a much shorter time and evaluation just takes a mouse klick. The data of 1st normal stress difference divided by entrance pressure loss plotted versus shear rate and shows a similar tendency as the previous example, extrusion quality gets better with increasing ratio of N_1/dP_e: See **Fig6**. Garvey die measurement**: See Fig 5,** is made at one just one throughput and has to be repeated several times to evaluate different throughputs while the measurement performed by normal stress die: See Fig.6, is made at different shear rates within one filling of the test barrel.

Ratings:			45				
bad	1	Compound number	swell	edge	surf.	corn.	Sum
acceptable	2	4	1	1	1	1	4
good	3	7	3	3	4	4	14
excellent	4	11	1	2	1	3	7
		14	4	4	4	4	16

FIGURE 5: Gavey die evaluation from 4 rubber compounds at one throughput – shear rate

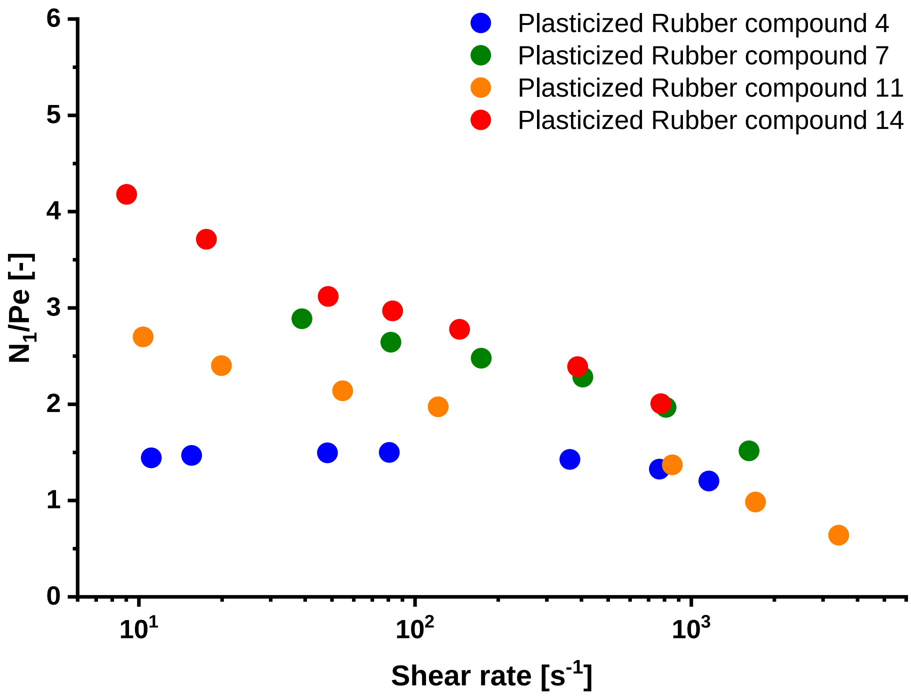

FIGURE 6: First normal stress difference divided by entrance pressure loss (dP_e) as a function of shear rare for four different rubber compounds

NORMAL STRESS DIFFERENCE AND DIE DROOL EVALUATIONS

Some polymers depose a part of the mass flow to the die lip. By a longer extrusion period the deposits grow and frequently some parts break and cause defects at the extrudate. This phenomenon is also called “die drool” or “die bleed”. The effect is also analyzed on some HDPE relating to their and molecular structure[4]. Differences in molecular structure also lead to differences in 1st normal stress difference.

Die drool is analyzed on 4 different HDPE and measured with Normal stress die: See **Fig. 7**. Die Drool was analyzed at Shear rate 500 1/s at 170°C for 2h extrusion. Sample 1 and 2 show nearly no die drool, while sample 3 shows slightly more. Only sample 4 clearly shows die drool under these conditions. 1st normal stress difference itself shows increasing values for increasing tendency of die drool build up, but sample 4 is no so much differentiated. There the data of 1st normal stress difference are also divided by entrance pressure loss. Now sample 4 clearly differs from the other sample: See **Fig.7**.

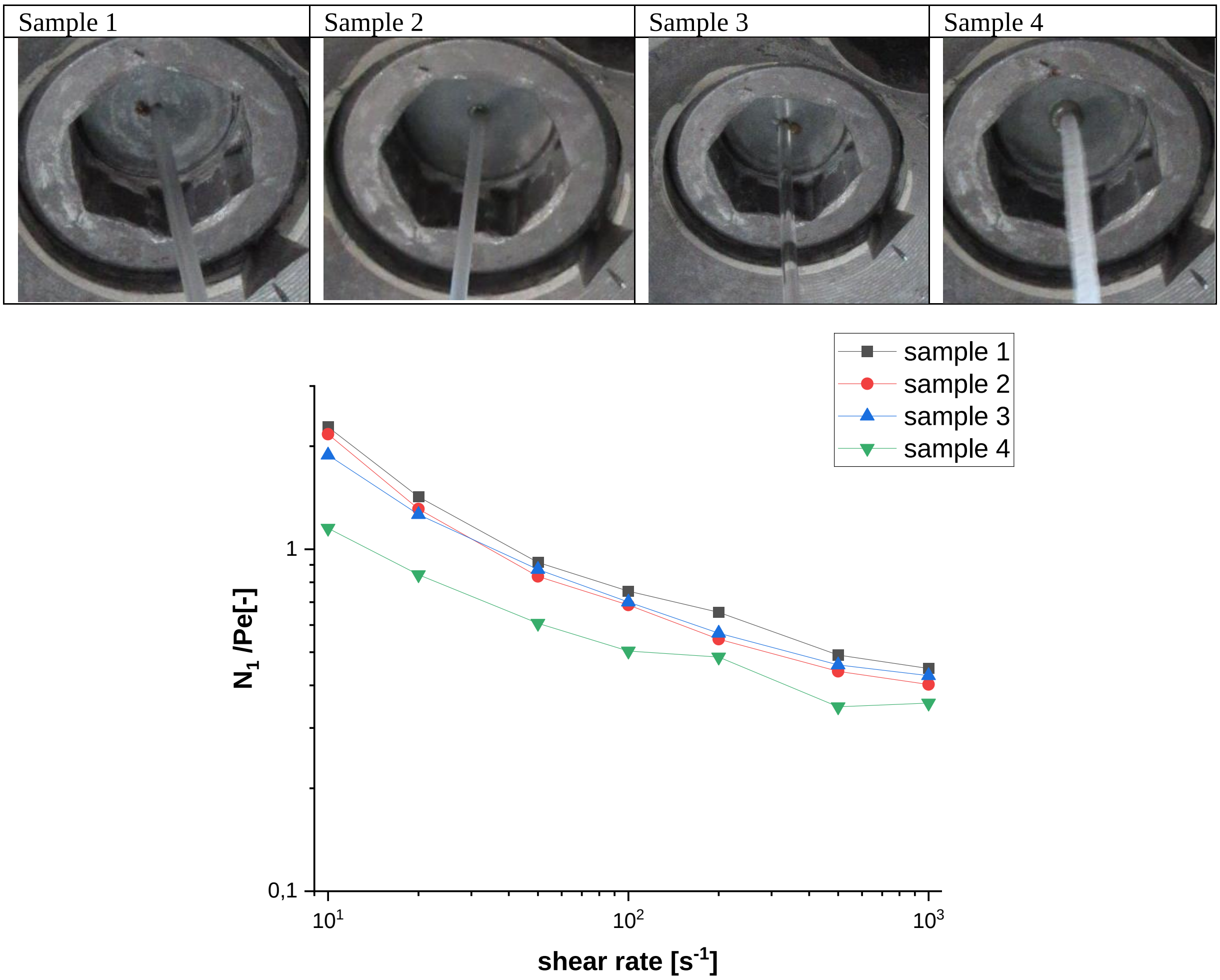

FIGURE 7: Die drool measurement at shear rate 500 1/s and first normal stress difference divided by entrance pressure loss (dP_e) as a function of shear rare for four HDPE samples

CONCLUSIONS

Normal Stress die can simultaneously measure steady-state shear viscosity, elongation viscosity and first normal stress difference. The viscosity obtained from the slit part of Normal Stress die after Weissenberg-Rabinowitsch correction matches the viscosity measured with capillary dies after Bagley and Weissenberg-Rabinowitsch corrections. Elongation viscosity is derived from the entrance pressure into the die. First normal stress difference obtained from Normal Stress die at high shear rates follows the same trend as transient shear data at low shear rates. Thus, a good accordance between the data of normal stress die and transient shear data is visible. Normal stress die allows an effective determination of normal stress at high shear rates and closer to the processing shear rates which was not possible with conventional technique.

Further the onset of instabilities is correlated to first normal stress difference and entrance pressure drop (graph of N_1/dP_e vs shear rate). The materials with lower N_1/dP_e for the same shear rate, show onset of instabilities at lower shear rates.
For rubber compounds extrudate defect measured by Garvey die are also first normal stress difference and entrance pressure drop (graph of N_1/dP_e vs shear rate). Materials with lower N_1/dP_e for the same shear rate, show an extrudate with more defects.
Finally, N_1/dP_e is correlated to die drool phenomenon. Materials with lower N_1/dP_e data show a higher tendency to die drool build up.

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