# Advanced Rheological Characterisation for Thermal Sensitive Materials Using Shear Heating Device

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**Abstract.** In predictive engineering for polymer processes, the rheological data is the most important data to end up with good prediction of processing parameters. In case of materials which are highly temperature sensitive and have a low bulk density, one has to be sure that the obtained data is representative for the future processing. One should avoid applying unrealistic conditions to the material causing unacceptable degradation of the material. To cover this issue, an advanced capillary rheometer has been developed which is able to heat the material to be tested in an advanced way. Simply conductive heating is combined with some controllable shearing resulting in measurement times which are only some minutes. Using a standard capillary rheometer, measurement times are most often more than ten minutes. The main disadvantage of this fast measuring technique is the sample temperature. To illustrate the good functioning of this equipment together with the validity of the results, a comparative reference measurement is set up for a non thermal sensitive material. This approach also allows to investigate the influence of the different settings influencing the shearing within the sample preparation for the shear viscosity measurement. Based on these promising results, the advanced rheometer can be used to perform accurate measurements for thermal sensitive materials as there are PVC. PVC is typically provided in powder form as raw material. While processing this material, it is highly sensitive for slip at the wall. Some promising results are obtained using this equipment.

## INTRODUCTION

Within polymer processing, the products which are realized through injection moulding and extrusion are more and more challenging in the sense that they contain more and more innovative features. To realize this, more often numerical simulation tools are used to predict the processing behaviour. The quality of the numerical simulation results is highly depending on the input data for the material data. For the extrusion and the injection moulding process, the shear viscosity behaviour is crucial information to predict the processing behaviour. Next to the ever increasing novelty in geometrical innovations, also the materials which are used are becoming more and more challenging. Next to the typical powder form for rigid PVC extrusion, more and more bio-based materials are processed which are typically highly sensitive to high temperatures. Nowadays, also more and more materials are recycled. For this type of materials, the amount of thermal stabilization is also limited. From economical point of view, no extra additives will be applied if there is no strong request for it. During standard extrusion and injection moulding for these types of materials, they are only exposed to higher temperature for only short amount of time (some minutes). To provide accurate numerical simulation results, it is necessary to have representative rheological data. To cope with the large shear rate ranges valid within extrusion and injection moulding, typically a capillary rheometer is used. In this rheometer, the cold material is typically entered into a cylindrical cavity to be heated by conduction. A typical diameter of this cavity is 15mm which request between 5 and 10 minutes heating time to end up with an acceptable homogeneous temperature through the thickness of this cavity. If the raw polymer material is provided as a powder, it is almost impossible to perform accurate measurement, as the homogenized thermal conductivity of the powder is much lower due to the low bulk density. The compression of the material sample is

also only applied when all the material is loaded into the measurement cavity, which is too late in case of powder materials. This is causing material which will be exposed too long to high temperatures and entrapped air which on his turn also causes measurement problems. Using the RheoArt® equipment allows to cope with all these difficulties. A picture and schematic drawing of this equipment is shown in Fig. 1. The equipment is promising but there are also some pitfalls in using this equipment. Within this paper, a reference measurement is set up to compare the obtained rheological data with the data obtained using a standard capillary rheometer for a nonthermally sensitive material. Next to that, the influence of the different extra parameters on the RheoArt® [1] are investigated and presented. Finally, first results on the rheological characterisation for rigid PVC are illustrated together with the characterisation for slip at wall. One has to be aware that the characterisation of slip at wall is highly depending on the surface finish of the measurement geometry. The obtained data with this equipment is only indicating, to obtain the real slip behaviour for a material in combination with a tool, similar surface finish is requested which is most probably different from the surface quality within a capillary which is used within a capillary rheometer [2]. To end up with correct shear viscosity behaviour and slip at wall, one need to perform reverse engineering based on experimental results obtained from a die with the applied surface roughness. This roughness can also change as function of time due to high amount of hard fillers.

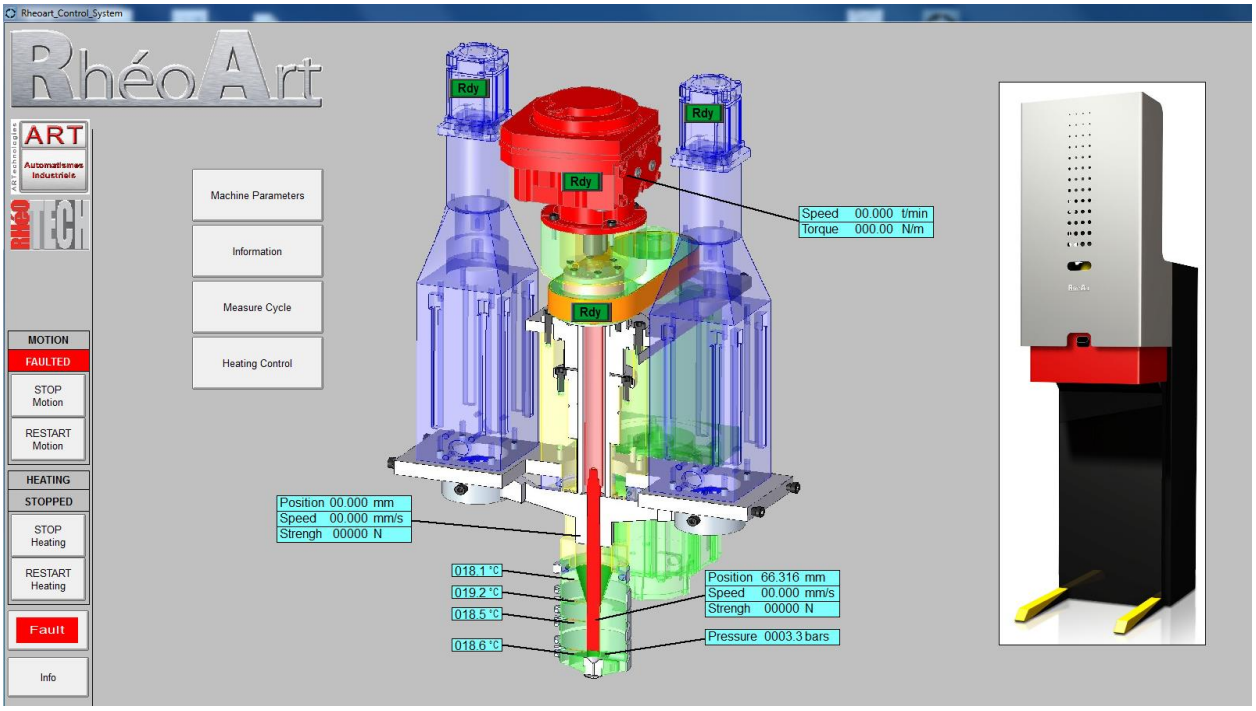


**FIGURE 1.** Initial screen of the RheoArt® software tool indicating the main parts within the equipment [3].

## MEASUREMENT STRATEGY

Figure 2 illustrates the different steps to perform a measurement with the RheoArt®. The picture on the left shows the starting position for the measurement. The operator has to load the test sample manually into the cavity below the plunger. During this loading, no compression is applied on to the material. The central image in Fig. 2 illustrates how the test sample is heated. The whole barrel is heated causing on one hand heating by thermal conductivity. On the other hand, the material is forced by the plunger through the gap indicated in Fig. 2. The speed of the plunger can be modified to adjust the applied shearing to the test sample. On top of the flow through a narrow gap, the piston is also rotating while the material is forced through the gap. Also, the rotation speed of the piston can be modified. By this transportation through a narrow gap, there is also a controllable heating of the sample by shear. This combination of heating allows fast heating, which is in favour to avoid too long measurement times for thermal sensitive materials, however, the temperature of the test sample is influenced by the amount of shear which is

applied. The right image of Fig. 2 illustrates the standard procedure for the measurement using a capillary of choice in the bottom of the equipment. During this measurement, the piston is moving down with a controlled speed. Within the RheoArt® software, different shear rates can be applied within one measurement. The shear viscosity data can be derived from the pressure sensor which is located at the entrance of the capillary. Application of the Bagley and Rabinowitsch corrections allows to define the shear viscosity based on different lengths for the same diameter of capillaries. If different diameters are used with the same *L*/*D* values, the wall slip behaviour can be defined using a Mooney analysis. In the labs of ProPoLiS in KU Leuven Bruges Campus, where this RheoArt® is installed, capillaries of 1 mm diameter and a length of 0, 10 and 20 mm are available. For 0 mm, 2 mm, 20 mm and 40 mm length are available. For 3 mm, a length of 0 mm and 30 mm is available. The 3 mm capillaries are mainly used for filled materials.

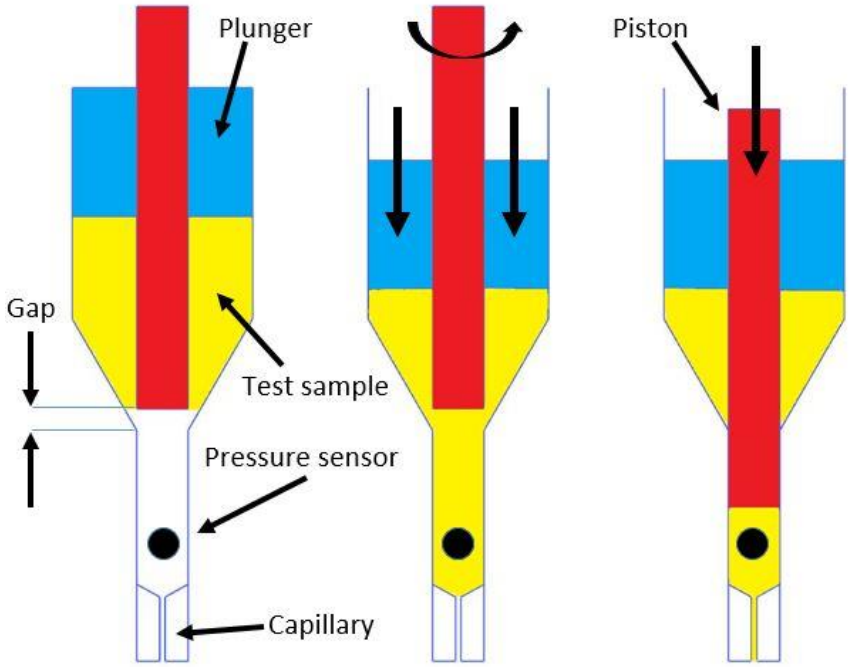


**FIGURE 2.** Different steps within a rheological measurement with the RheoArt® equipment.

## CORRELATION BETWEEN RHEOART® AND STANDARD CAPILLARY RHEOMETER

To illustrate the well function of the equipment, first of all, a reference measurement for a nonthermally sensitive material has been performed on a standard dual bore Ceast rheometer. The material used is an injection moulding grade of PP, namely Ineos 402 CB 12. The measurement was performed at 230°C in an apparent shear rate range between 10 and 5000 $s^{-1}$ using default values for the parameters influencing the shear heating during the preheating step, namely, a gap of 2 mm, a piston rotation speed of 50 rpm and a feeding speed of the plunger of 0,4 $mm.s^{-1}$. Capillaries with a diameter of 2 mm and a length of 20 and 40 mm are used. As can be seen in Fig. 3, rather good correlation is obtained, within capillary rheometry, and for sure with the standard Ceast equipment, there is some variability depending on the loading of the sample. For the RheoArt®, the repeatability of results is higher which is caused by the automated loading of the sample into the measuring volume. Figure 4 gives the measured shear viscosity data for the same *L*/*D* values equal to 10 for 1 and 2 mm diameter capillaries. This graph illustrates that for this material no slip is occurring.

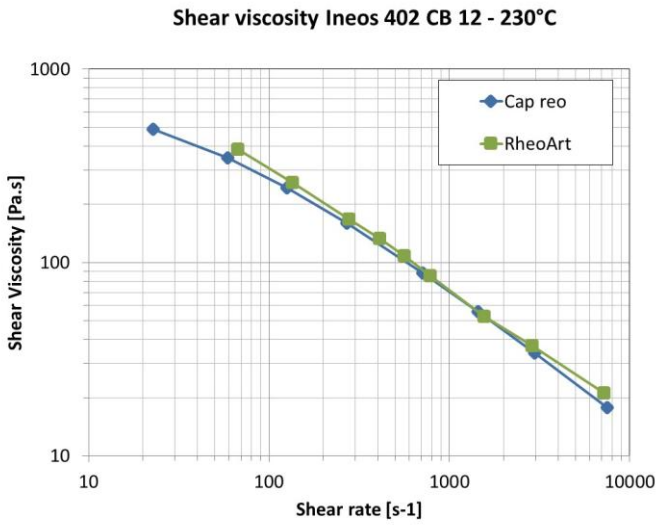


**FIGURE 3.** Shear viscosity data comparison for a standard PP based on experiments on a standard dual bore rheometer and the RheoArt®.

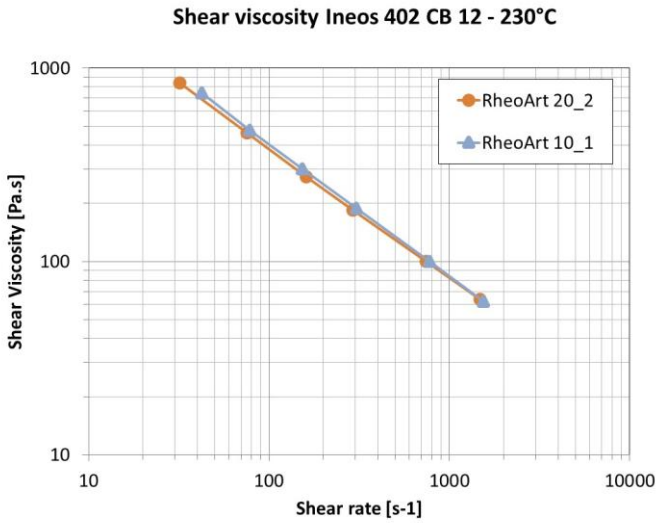


**FIGURE 4.** Shear viscosity data comparison for a standard PP for the same *L*/*D* values measured on the RheoArt®.

## INFLUENCE OF SHEAR PARAMETERS DURING PREHEATING OF THE TEST SAMPLE

As already earlier indicated, the absolute temperature value at which the viscosity measurement is performed is not known using the RheoArt® equipment. This is due to the application of shear heating of the sample to be measured. To illustrate the influence of the different parameters, some comparative measurements are performed. First of all, the reference PP as used before in this article has been measured using two different gap values during the loading phase. The measurement was performed at 230°C in an apparent shear rate range between 10 and 5000 $s^{-1}$ using default values for the other parameters influencing the shear heating, namely, a piston rotation speed of 100 rpm and a feeding speed of the plunger of 0,4 $mm.s^{-1}$. As can be seen in Fig. 5, on average a 5% higher pressure is obtained using a larger gap. This first parameter already illustrates the importance of this value. Figure 5 also indicates that the total measurement time is less than one minute in this case which is really fast to obtain stable pressure values for different shear rate conditions.

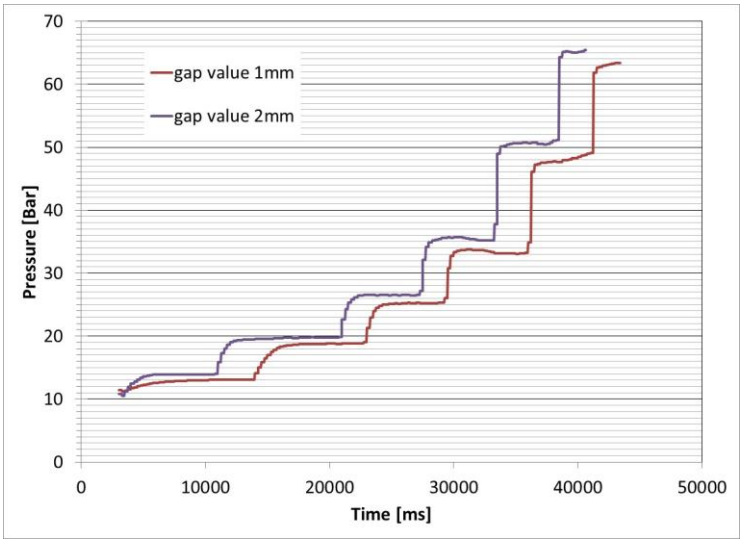


**FIGURE 5.** Pressure as function of time for a standard PP rheological measurement on the RheoArt®.

The influence of the other parameters influencing the shear heating, is checked based on measurements performed on a virgin rigid PVC material keeping the gap distance at 2 mm. The speed of the plunger has been varied from 0.4 to 0.2 $mm.s^{-1}$ and the rotational speed of the piston has been changed from 100 down to 50 rpm. The results at 195°C barrel temperature are shown in Fig. 6. Based on these measurements, changing the rotational speed of the piston does not influence the pressure values too much. Nevertheless, if one changes the feeding speed of the plunger, on average, more than 20% lower pressure value is registered if the feeding speed is lowered from 0.4 $mm.s^{-1}$ to 0.2 $mm.s^{-1}$. This is mainly caused by the longer residence time of the material within the volume where the shearing is applied. If a lower feeding speed is combined with a lower rotational speed, one ends up with similar data as the reference case (Feeding speed 0.4 $mm.s^{-1}$ and number of revolutions 100). Figure 7 indicates that a similar behaviour is noticed if a barrel temperature of 190°C is used. Figure 8 illustrates a change of pressure drop of 5 percent only changing the temperature with 5°C. The viscosity of the PVC material is highly temperature sensitive.

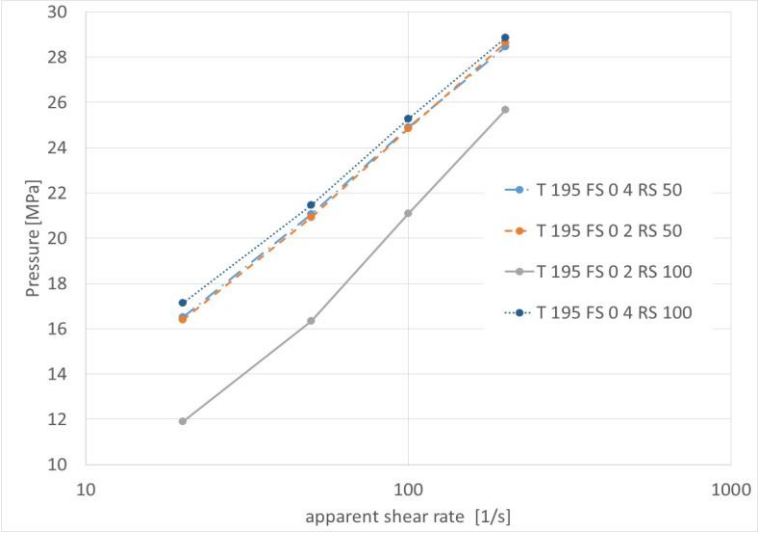


**FIGURE 6.** Pressure drop comparison for a Rigid PVC material for changing values influencing the shear behaviour during preheating at 195°C.

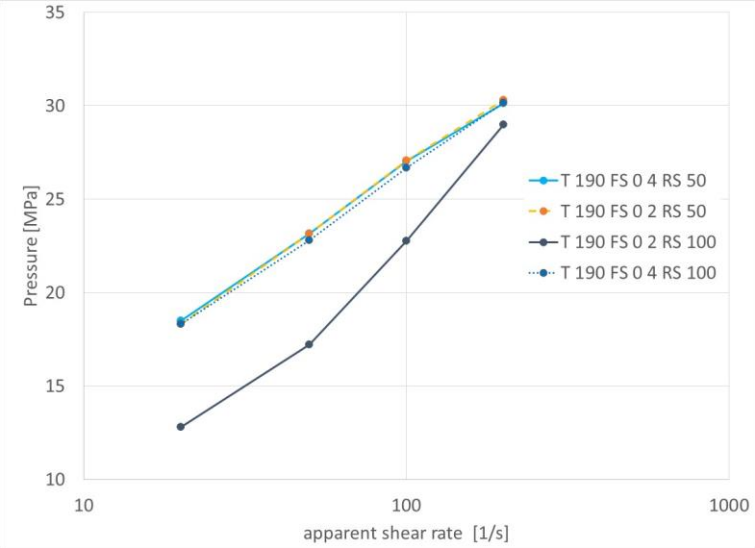


**FIGURE 7.** Pressure drop comparison for a Rigid PVC material for changing values influencing the shear behaviour during preheating at 190°C.

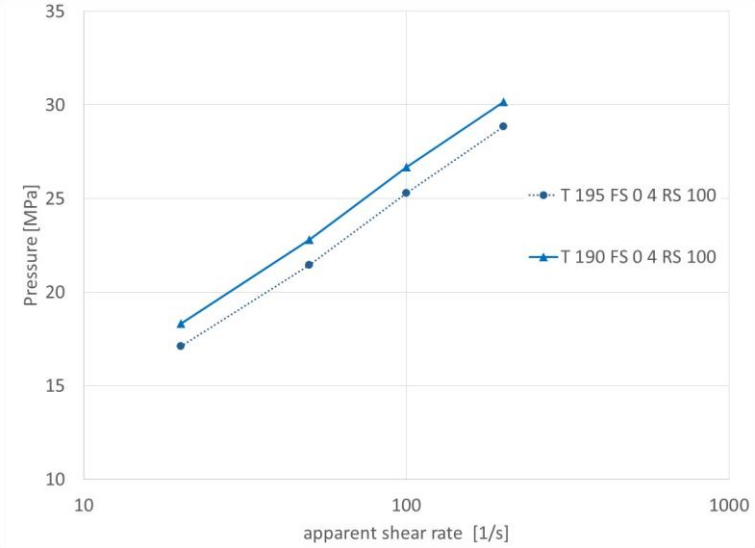


**FIGURE 8.** Pressure drop comparison for a Rigid PVC material at two temperatures.

## ROBUST MEASUREMENT AND CHARACTERISATION FOR SLIP AT WALL FOR RIGID PVC

Within tool design for extrusion of rigid PVC, one has to deal with the high sensitivity for slip at wall for rigid PVC material. The slip behaviour is of crucial importance to end up with a material distribution within the final profile as designed. During the preparation of the material, the processing company can add some ‘so called’ internal and external lubricants. With internal lubricants, one aims at a controlled shear thinning behaviour without too high viscosity, the external lubricant controls the slip at wall behaviour.

At first, one has to define the temperatures which are relevant for the processing of the material of interest. Next, it is necessary to define the shear rate range which has to be characterised. Extrapolation of data in this field should be avoided, as the phenomena has some inherently stochastic nature. Within the extrusion process, one should also

avoid shear stress levels beyond 300 kPa. From this stress level on, the material is mechanically highly loaded and pressure levels are unacceptable within extrusion tools. Due to these high pressures, a lot of shear heating will occur damaging the material. Typically, a measurement range between 20 and 500 $s^{-1}$ is in most cases enough for only low filled materials. In case of high filled materials (with potential higher slip at wall), a shear rate range between 5 and 100 $s^{-1}$ will do. Typically, the shear rate levels are logarithmically distributed to end up with equidistant points in a log-log graph which is typically used within rheology. During the material characterisation, one has to be sure one is covering the whole processing range with enough measurement points within the interval of interest. If necessary, one can always delete some not relevant data. The diameters of the capillaries used should also be appropriate for the material to be tested, in case of highly filled materials, a 1mm die is not recommended.

The data presented below is obtained for a standard rigid PVC used for window profiles. For each of the measurements, three measurements are performed, the repeatability is high due to the exclusion of the manual loading using the RheoArt® equipment. The coefficient of variation is typically below 3% which is acceptable within rheological measurements of this nature. All measurements are performed at a barrel temperature of 195°C which is a typical processing temperature for rigid PVC. The measurements were performed at shear rates 20, 50, 100, 200, 300, 400 and 500 $s^{-1}$. The values of 300 and 400 $s^{-1}$ were added to look for the extra data in the range of high shear stress values. For both capillary diameters (1 and 2 mm), three *L*/*D* values are measured, namely, 0, 10 and 20. For the length of 0 mm, an orifice die has been used as displayed in Fig. 9. Tolerances are not displayed due to confidentiality reasons.

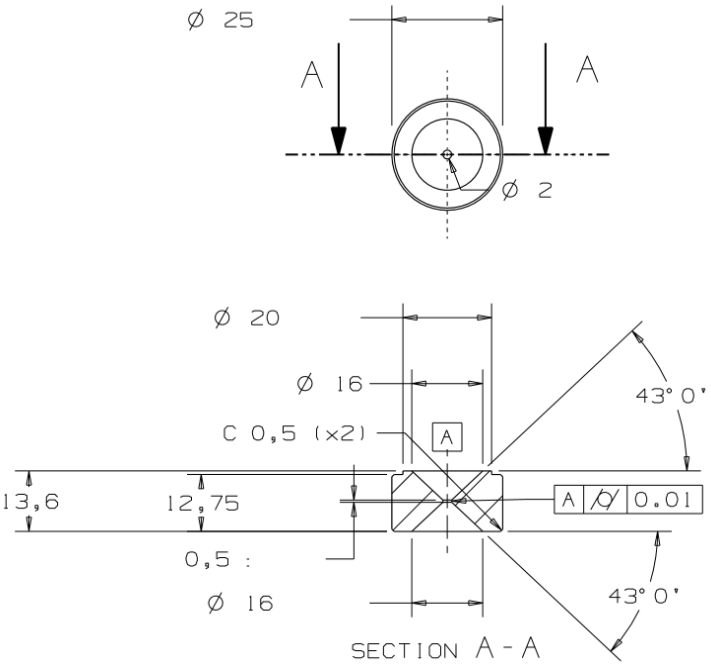


**FIGURE 9.** Dimensions of the orifice die used within this investigation.

Figure 10 shows the results obtained for the 1mm capillaries within a Bagley plot. On this plot, one can see that the pressure obtained at *L*/*D* equal to 20 are typically somewhat below the linear fit values. The temperature rise within the polymer material is seen as the main reason causing this deviation. To use all measured data, the linear fitted curve is used to generate the input values (pressure drop on particular capillary) for the slip at wall calculation. Based on this data, for the two capillary diameters, the shear stress as function of shear rate can be displayed. It can be clearly seen that the data for the 2 mm capillary is considerably higher than for the 1 mm capillary data indicating that non neglectable slip at wall is occurring. To apply the Mooney analysis, a power law has been fitted to the obtained shear stress data to be able to have robust interpolation taking into account all data. In that way, more physical possible data will be obtained, if only two neighbouring points are used, one ends up in some cases with nonrealistic data.

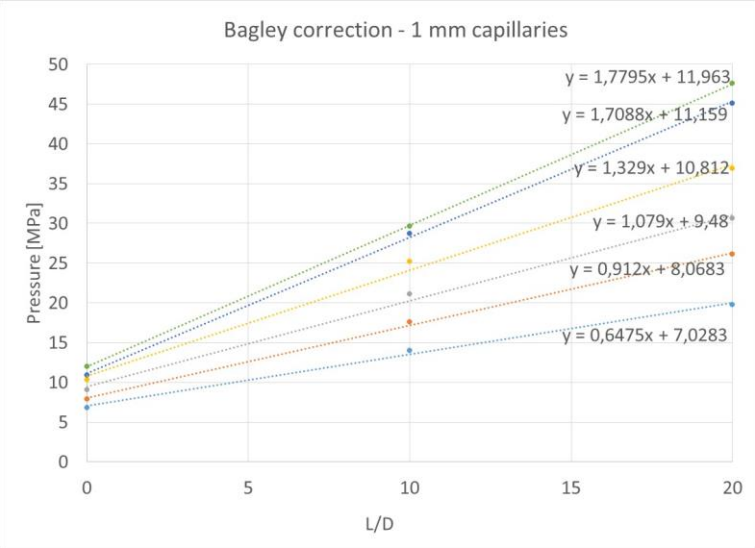


**FIGURE 10.** Bagley plot for capillaries 1 mm for shear rates 20, 50, 100, 200, 300, 400 and 500 $s^{-1}$.

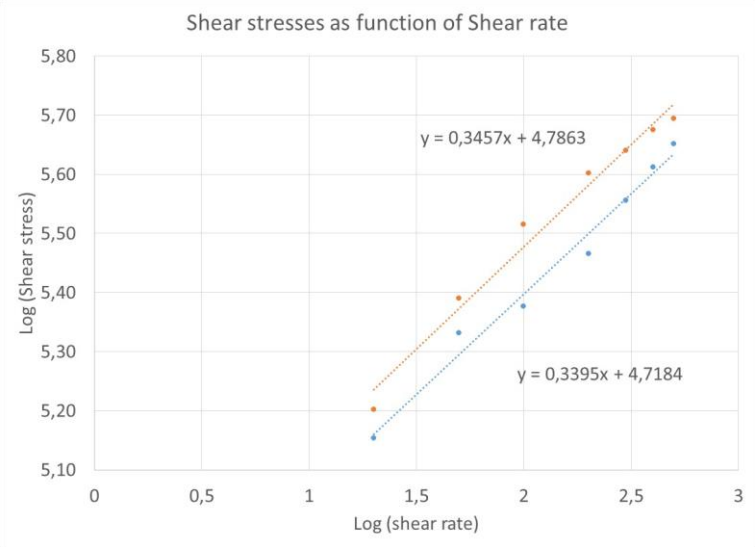


**FIGURE 11.** Shear stresses as function of apparent shear rate. The upper curve is for the 2 mm capillary, the lower one is for the 1 mm capillary.

Figure 12 shows the Mooney plot in which the shear stress data is transformed considering the results for the capillary diameter of 2 mm as a reference. This graph illustrates the nice results indicating the high slip at wall velocities. Figure 13 shows the fitting results for the obtained slip velocities as function of relative shear stress levels. The exponent of the power law equal to 3.02 is a high value and indeed shows that for higher shear stress levels, the resulting slip at wall velocity is really sensitive for a minor change in material and surface finish.

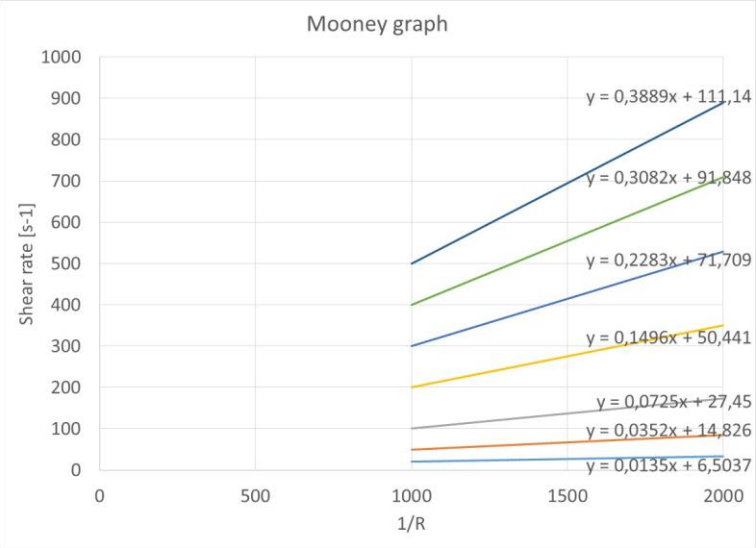


**FIGURE 12.** Shear stresses as function of apparent shear rate. The upper curve is for the 2 mm capillary, the lower one is for the 1 mm capillary.

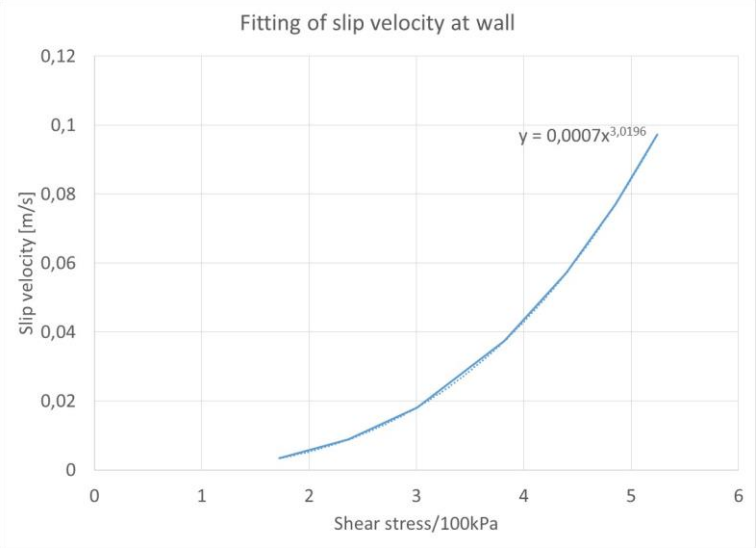


**FIGURE 13.** Fitting with power law for slip at wall in large shear stress interval.

Figure 14 shows the obtained shear viscosity data for the characterised rigid PVC data. On this graph, also the real shear viscosity is displayed if one neglect slip at wall velocities. As expected, to end up with similar pressure values if one neglect the slip at wall, the overall viscosity level is lower and the obtained slope is also indicating higher shear thinning behaviour. As long one is only using one capillary diameter based on the fact that one only produces products with constant thicknesses equal to the capillary diameter, this approach delivers reasonable results. In fact, this approach is nowadays still used in a lot of companies while it is tricky. As soon as dimensions are changing, the results will deviate considerably.

Within commercial numerical simulation tools, a lot of them already are capable to cope with slip at wall behaviour [4,5], however, in some cases, they are not really well suited to cope with large slopes in the slip behaviour causing non stable or non-converging behaviour. To avoid this, a small tool named Profile Balancing has been developed within the ProPoLiS research group to solve for simple geometries taking into account the slip

behaviour. It is designed as an educational tool to illustrate the influence of slip to the master students. It can also serve as a simple numerical simulation tool to design extrusion dies as long as they can be separated into rectangular and circular sections (or equivalent radius approach). For the shear viscosity description, a power law description is used combined with an Arhenius temperature dependency [6]. For the slip at wall behaviour, an power law model is used based on the shear stress at the wall [6]. Those functions are also fitted within Fig. 13 and Fig. 14.

Figure 15 shows the comparison between the obtained numerical simulation tool results and the experimental data. A nice correlation is shown, the average deviation is below five percent which is acceptable for this type of measurements.

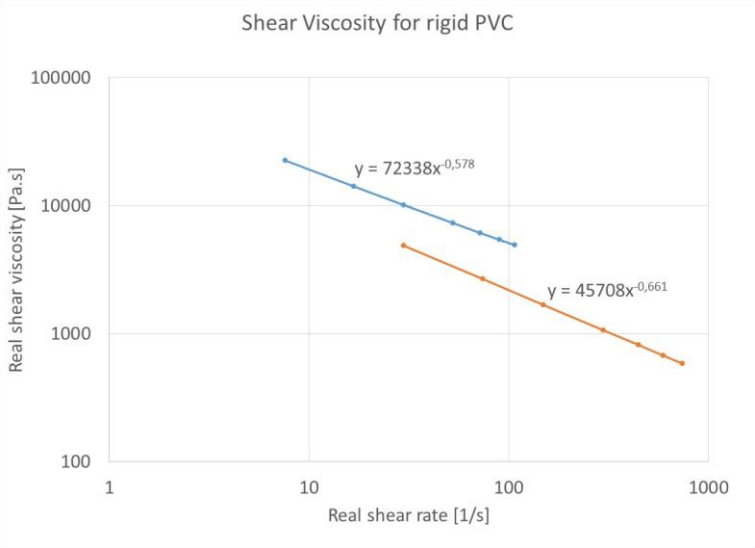


**FIGURE 14.** Real shear rate viscosity taking into account slip at wall (upper curve) and with neglecting slip at wall (lower curve).

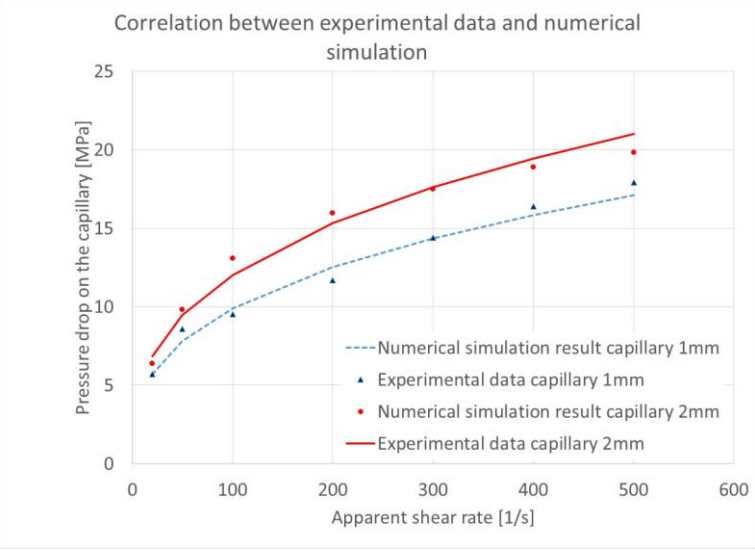


**FIGURE 15.** Comparison between numerical simulation results based on the characterised data and experimental data.

## CONCLUSIONS

This paper gives an scientific overview of the potential of the RheoArt® equipment. There are a lot of important advantages that can help to cope with industrial challenges. Some extra parameters to be defined using the RheoArt® equipment are investigated. The sensitivity has been shown for both tests on a PolyPropyleen material and also for rigid PVC. Using appropriate settings for measuring combined with enough high quality capillaries, allows to measure the slip at wall behaviour for rigid PVC. The obtained parameters for the shear viscosity behaviour and the slip at wall are compared with experimental data showing good correlation. This data can be used within a commercial numerical simulation tool to compare the data with experimental data. Based on this comparison, further optimization can be performed. This optimization cannot be avoided as the surface finish of an extrusion die is always different from the one applied to a capillary die.

## ACKNOWLEDGMENTS

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