

**HIGH PRESSURE CAPILLARY RHEOMETRY ON WOOD PLASTIC COMPOSITES WITH VARIATION OF WOOD CONTENT AND MATRIX POLYMER***M.Eng. N. Laufer<sup>1</sup>, Prof. Dr.-Ing. H. Hansmann<sup>1,2</sup>, Univ.-Prof. Dr.-Ing. M. Koch<sup>3</sup>*<sup>1</sup>Institut für Polymertechnologien e. V.<sup>2</sup>Hochschule Wismar, FG Kunststofftechnik<sup>3</sup>Technische Universität Ilmenau, FG Kunststofftechnik**ABSTRACT**

Wood Plastic Composite (WPC) is a composite material made from wood fibers (fine chip, wood shavings or wood flour) and a thermoplastic matrix polymer. Matrix polymers are mainly polyethylene (PE), polypropylene (PP) and polyvinyl chloride (PVC). The wood content of WPC varies, depending on the applied method of processing, between 50 and 75 wt%. The flow behaviour of WPC melts is of decisive importance for their processability. Understanding rheological behaviour shall help to increase the efficiency of processing of WPC products due to increased material throughput. Furthermore it helps to simplify the development of new WPC products and extrusion tools respectively.

This paper describes how various WPC formulations were examined on the basis of high pressure capillary rheometry (HKR). It could be shown that the flow behaviour of these WPC melts can be well described by functions based on the power law. Furthermore the measurements also confirmed a good correlation between flow exponent and consistency factor.

**1. FUNDAMENTALS OF SUSPENSION RHEOLOGY**

A suspension is a system in which denser particles are distributed throughout a less dense fluid. In consideration of filled polymer systems, the fillers form a disperse phase and the molten polymers form the continuous phase. [1]

WPC melts constitute this very kind of heterogeneous solid-fluid mixtures which is why they are called suspensions. The flow behaviour of unfilled and filled polymer melts can be characterized by their viscosity. [2]

In physical terms viscosity is a constant of proportionality between the shear stress acting on the interface of a fluid element and the resulting shear rate:

$$\frac{\tau}{\dot{\gamma}} \quad (1)$$

	viscosity	[Pa s]
$\tau$	shear stress	[Pa]
$\dot{\gamma}$	shear rate	[1/s]

Rheological tests on wood-filled LDPE show that flow functions of these WPC melts are shifted to higher values in comparison to the flow curve of the matrix polymer. The extent of this displacement depends on both the wood concentration and the shear rate (*Figure 1*).

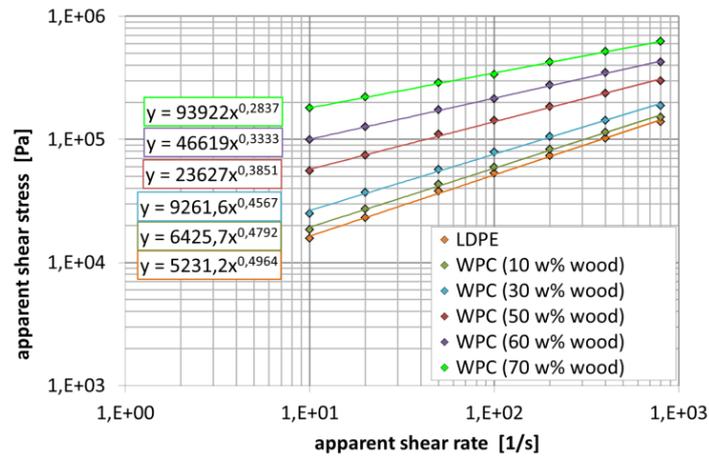


Figure 1 flow functions of low-density polyethylene (LDPE) and LDPE-based WPC melts with varying wood contents ( $T = 190 \text{ }^\circ\text{C}$ )

The increase of flow functions of suspensions against to those of the matrix can be explained vividly on the basis of the concept of shear-stress-equivalent inner shear rate according to W. Gleißle and M. K. Baloch. According to this model (Figure 2), the hydraulic width of the gap between the plates is reduced, due to the volume content of the filler (solid particles idealized as rigid plates). This leads to an increasing internal shear rate of the pure matrix while the external shear rate remains constant. [3], [4]

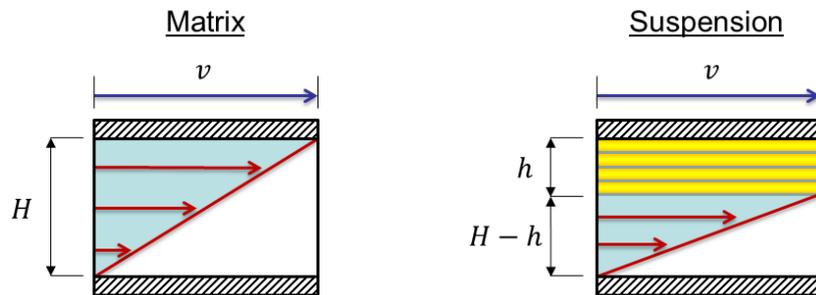


Figure 2 Idealization of flow processes in suspensions according to the concept of the shear-stress-equivalent inner shear rate

external shear rate

$$\dot{\gamma}_0 = \frac{v}{H} \quad (2)$$

inner shear rate

$$\dot{\gamma}_c = \frac{v}{H-h} \quad (3)$$

The relative increase of inner shear rate is expressed by shift factor  $B$ .

$$B = \frac{\dot{\gamma}_c}{\dot{\gamma}_0} = \frac{H}{H-h} \quad (4)$$

The flow process of suspensions in round capillaries can also be idealized with the concept of shear-stress-equivalent inner shear rate (Figure 3).

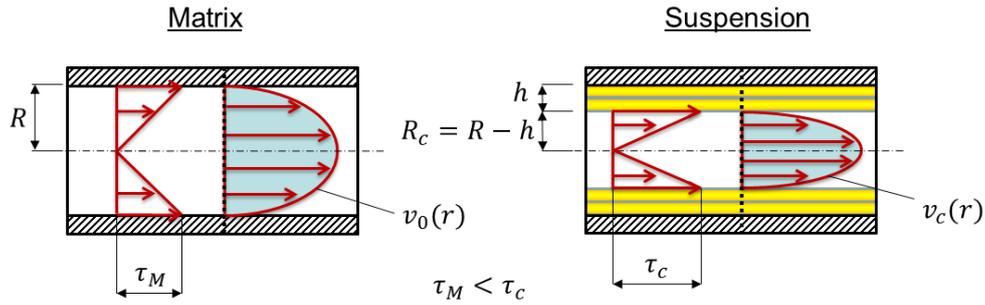


Figure 3 Idealization of flow processes of suspensions in round capillaries

external shear rate

$$\dot{\gamma}_0 = \frac{4 \cdot \dot{V}}{\pi \cdot R^3} \quad (5)$$

inner shear rate

$$\dot{\gamma}_c = \frac{4 \cdot \dot{V}}{\pi \cdot R_c^3} \quad (6)$$

shift factor

$$B = \frac{\dot{\gamma}_c}{\dot{\gamma}_0} = \frac{R^3}{R_c^3} \quad (7)$$

According to the concept of shear-stress-equivalent inner shear rate, the increase of flow functions of suspensions against to those of the matrix is attributed only to the volume content of the filler particles. This means that interactions between the particles have to be negligibly small.

Generally speaking, the matrix can be considered as a shear thinning fluid (polymer melt) whose flow behaviour can be described by the power law (defined by consistency  $K$  and flow index  $n$ ) of Ostwald/de Waele (Figure 4). [5] Provided that particle-particle interactions are negligibly small ( $n_c = n_0 = n$ ), shift factor  $B$  is dependent only on the volumetric filler concentration ( $c_v$ ) and a higher value is shown for the shear stress of the suspension in accordance with shift factor  $B$  and depending on the flow index  $n_0$  of the matrix.

$$\tau_c = K_0 \cdot \dot{\gamma}_c^{n_0} = \left( \frac{\dot{\gamma}_c}{\dot{\gamma}_0} \right)^{n_0} \cdot \tau_0 = B^{n_0} \cdot \tau_0 \quad (8)$$

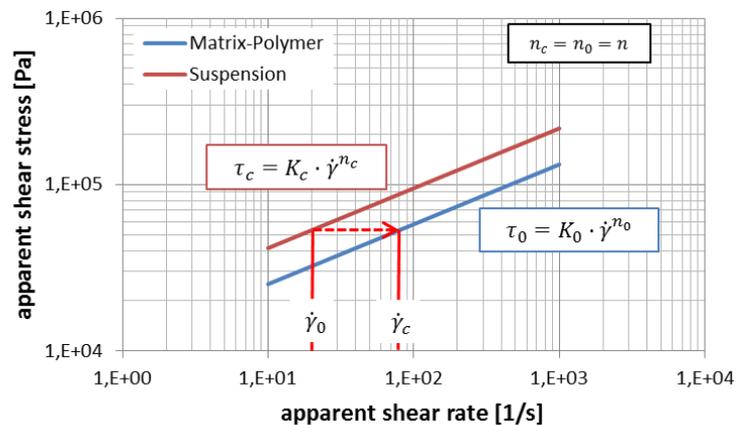


Figure 4 Comparison of flow functions (matrix-polymer and suspension) when no particle-particle interactions occur

## 2. HIGH PRESSURE CAPILLARY RHEOMETRY ON WPC

### 2.1 Materials and Methods

The materials used in this study are low-density polyethylene (LDPE, Lupolen 2420 K) as the matrix polymer and softwood flour (Lignocel® BK 40/90, particle size 300 – 500 µm) as wood filler (*Figure 5*).



*Figure 5 fiber structure of Lignocel® BK 40/90*

The preparations of all WPC formulations were carried out by compounding on the co-rotating twin screw extruder ZSE 27 MAXX (Leistritz Extrusionstechnik GmbH). The rheological tests were performed on a high pressure capillary rheometer (HKR) Smart RHEO 5000 twin bore (CEAST - Instron). For all measurements, an extruder die was used with a diameter of 2 mm and a length of 40 mm. Before the measurements began, the WPC was dried in an oven at 105 °C for at least 3 hours.

The measured data were analysed on the basis of the apparent shear stress and shear rates. This means that forecasting models for estimating the flow behaviour of WPC melts of any wood concentrations are not transferable to other flow channel geometries. The problem of employing the Bagley correction method (which takes into account the input and output pressure losses and corrects the shear stress) and the Weissenberg/Rabinowitsch correction method (which takes into account the deviation from the Newtonian flow behaviour and corrects the shear rate) is that these correction methods will deliver correct results only if the condition of wall adhesion is considered. With regard to WPC melts this is not the case due to their strong tendency towards wall slippage especially when wood concentrations and volume flows are high. With the help of the Mooney correction method wall slippage effects can be taken into consideration so as to split up the total volume flow into a slippage and a shear proportion. [2]

### 2.3 Results and Discussion

As mentioned before, the flow functions of wood-filled LDPE are shifted to higher values in comparison to the flow curve of the unfilled polymer (*Figure 6*). Not only the content of volume but also the fibrous structure of the wood particles (*Figure 5*) has a large influence on the flow behaviour of WPC melts. Due to their geometric shape, these particles have a great tendency to interact with one another. The decreasing flow index with increasing wood content shows that interactions between particles are by no means negligibly small.

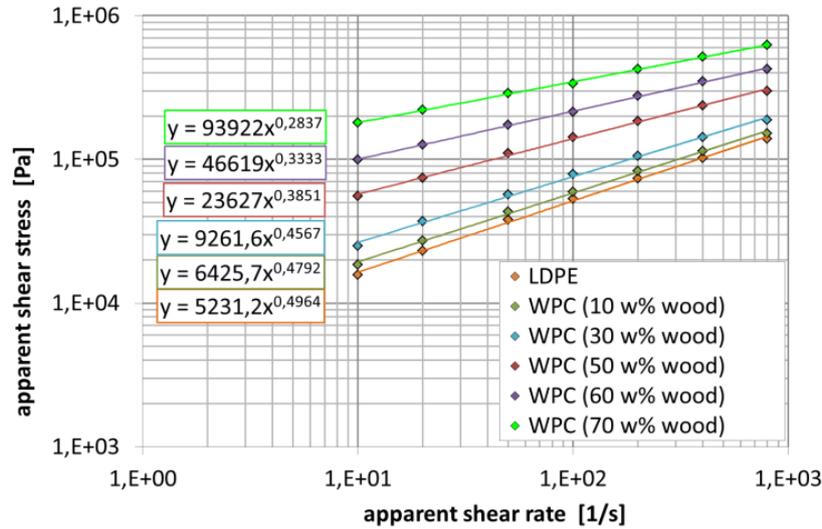


Figure 6 Flow functions of LDPE and LDPE-based WPC melts with varying wood contents

For this reason, the shift factor  $B$ , according to the concept of shear-stress-equivalent inner shear rate, becomes dependent on both the volumetric filler concentration and the applied shear stress.

$$B(\tau)^{n_c} = \frac{K_c}{K_0} \cdot \tau^{\left(\frac{n_c}{n_0} - 1\right)} \quad (9)$$

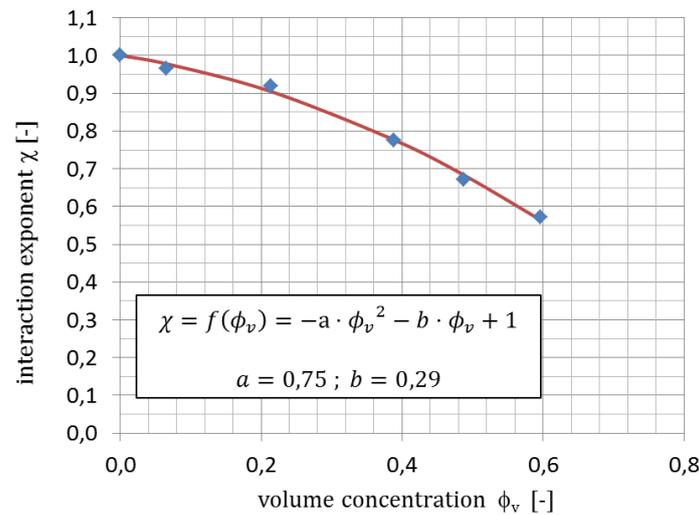
According to equation (9), exponent  $\frac{n_c}{n_0}$  expresses the intensity of interparticular interactions. Therefore, it can be referred to as *interaction exponent*  $\chi$ . [2]

$$B(\tau)^{n_c} = \frac{K_c}{K_0^\chi} \cdot \tau^{(\chi-1)} \quad (10)$$

For the borderline case of negligible interparticular interactions the interaction exponent is equal to one and equation (10) is reduced to the familiar shear-stress-independent form for the calculation of shift factor  $B$ .

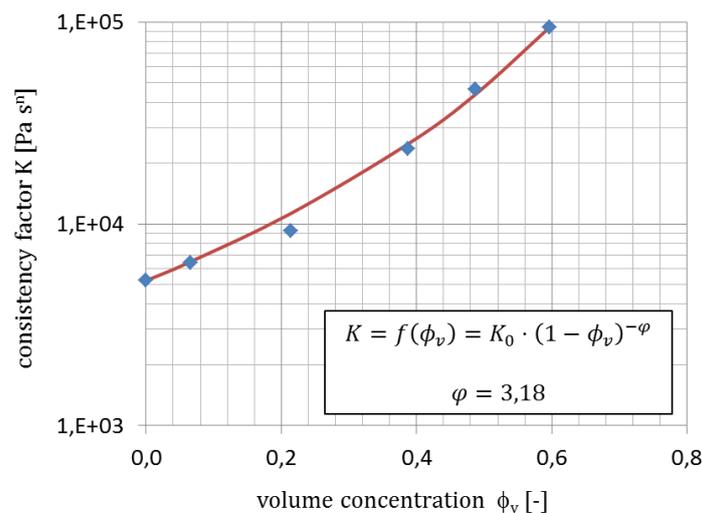
*Figure 6* illustrates that interaction exponent  $\chi$  as well as the consistency of the suspension  $K_c$  is dependent on the volumetric wood content. In order to examine the quantitative impact of wood concentrations on these parameters correlation graphs have been derived on the basis of measured values.

For wood-filled LDPE a quadratic correlation between interaction exponent and wood concentration was found, with reasonable accuracy (*Figure 7*).



*Figure 7* dependency between interaction exponent and wood content of LDPE-based WPC ( $T = 190$  °C)

In addition, the influence of the wood concentration on the consistency factor can also be described mathematically with adequate accuracy (*Figure 8*).



*Figure 8* dependency between consistency factor and wood content of LDPE-based WPC ( $T = 190$  °C)

On the basis of these relationships, the influence of the volumetric filler concentration and the applied shear stress on the shift factor B can now be described mathematically for LDPE-based WPC.

$$B(\phi_v, \tau) = \left(\frac{K_0}{\tau}\right)^{\frac{a\phi_v^2 + b\phi_v}{n_c}} \cdot (1 - \phi_v)^{-\frac{\varphi}{n_c}} \quad (11)$$

Figure 9 shows the results graphically. The points in *Figure 9* represent the shift factor B based on the test results, while the solid lines represents the shift factor according to equation 11.

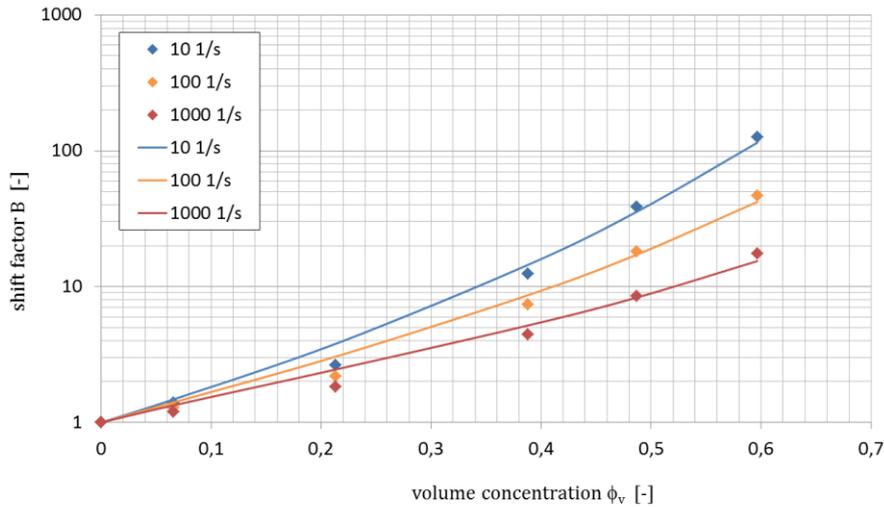


Figure 9 shift factor  $B$  for LDPE-based WPC for different apparent shear rates

Figure 9 shows, that in the region of medium volumetric wood concentrations, the calculated approximation functions for shift factor  $B$ , according equation 11, deviate from the measured values.

A better consistency, especially in the region of high volumetric wood concentrations can be achieved by direct approximation of the measured shift factors (Figure 10), according to the following equation:

$$B(\phi_v, \tau) = \left[ 1 + \left( \frac{\phi_v}{m} \right)^a \right]^{\frac{\phi}{a}} \quad (12)$$

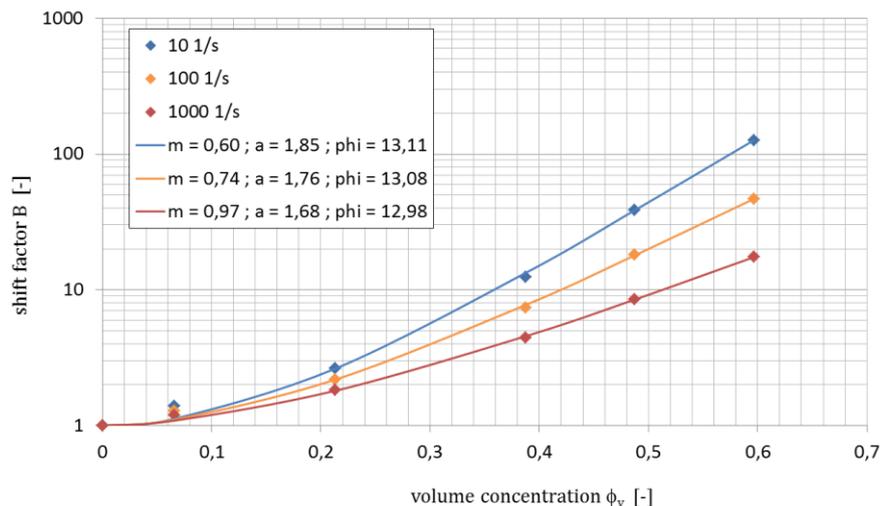


Figure 10 direct approximation of shift factor  $B$  for LDPE-based WPC

The mathematical model used for this approximation is based on the Carreau-Yasuda equation, which describes the viscosity characteristics of pseudoplastic fluids with transition to zero viscosity. The functional parameters  $m$ ,  $a$  and  $\phi$  are determined iteratively. This approach is of greater practical importance due to the good consistency to the measured values, especially in the region of technically relevant volumetric wood concentrations.

Furthermore, a functional correlation was found between the consistency factor and the interaction exponent. This correlation is also valid for wood-filled (Lignocel® BK 40/90) polypropylene (PP) (Figure 11). Each point in Figure 11 represents a certain volumetric wood concentration.

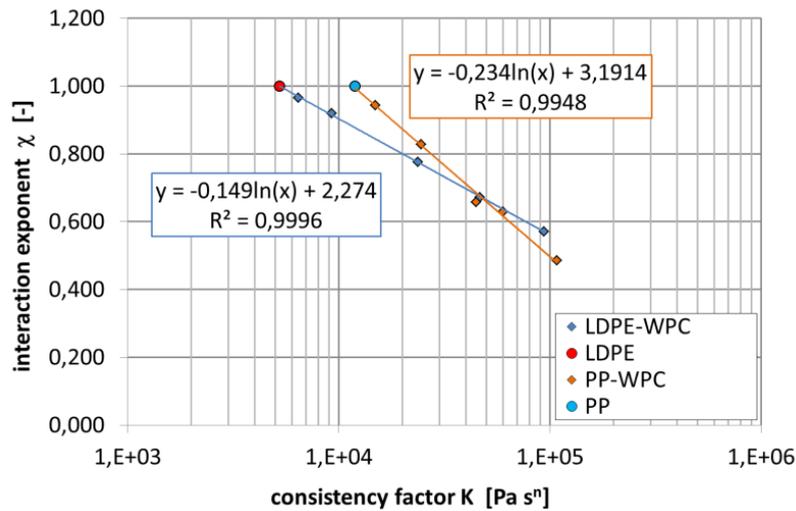


Figure 11 correlation between interaction exponent  $\chi$  and consistency factor  $K$  for different WPC

By means of this correlation function, the impact of the wood particles on the flow behaviour of WPC can be directly visualized, independently from the applied shear stress. Also Figure 11 shows clearly that the interaction effects of the same wood filler (Lignocel® BK 40/90) with the same mixing ratio may differ depending on the polymer matrix. The molecular structure of the polymer as well as its unfilled rheological characteristics apparently affects the flow behaviour of WPC.

Whether these correlation functions describe a general relationship between consistency and interaction exponent, independently of the WPC formulation, is subject of further investigations.

This approach for determining the shear stress independent correlation functions is of great practical importance, since the flow behaviour of WPC melts can be estimated as a function of the filler content in a simple manner. To demonstrate this, the flow curves for two different PP-based WPC formulations have been estimated. The estimated flow curves show a good consistency with the measured flow characteristics.

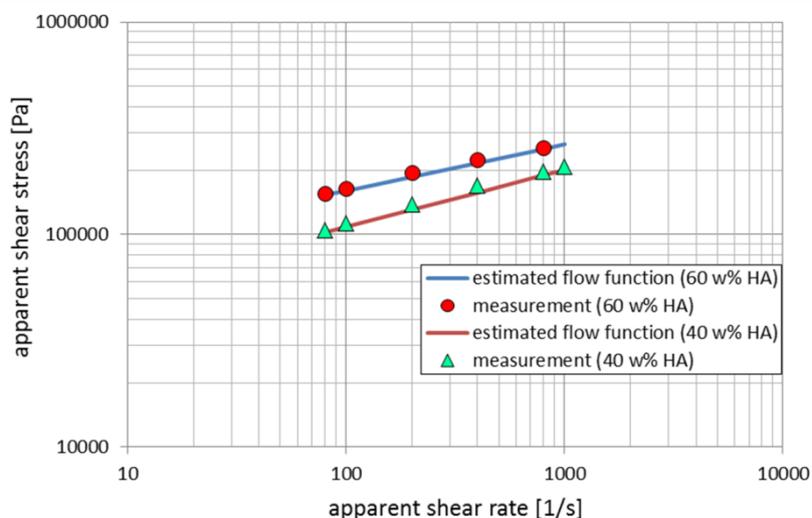


Figure 12 comparison of estimated and measured flow functions of PP-based WPC

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