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Original published in:	ic-rmm2 : 2nd International Conference on Rheology and Modeling of Materials : October 5-9, 2015/Miskolc-Lillafüred, Hungary - [Bristol] : IOP Publishing. - (2017), art. 012017, 9 pp. ISBN 978-1-5108-3678-5 (Journal of physics. Conference Series ; 790)
Conference:	International Conference on Rheology and Modeling of Materials (ic-rmm) ; 2 (Miskolc-Lillafüred) : 2015.10.05-09
Original published:	2017
ISSN:	1742-6596
DOI:	10.1088/1742-6596/790/1/012017
[*Visited:*	2024-02-01]

International Conference on Recent Trends in Physics 2016 (ICRTP2016) IOP Publishing
Journal of Physics: Conference Series **755** (2016) 011001 doi:10.1088/1742-6596/755/1/011001

Rheological Characterisation of the Flow Behaviour of Wood Plastic Composites in Consideration of Different Volume Fractions of Wood

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Abstract. In this study, the rheological properties of wood plastic composites (WPC) with different polymeric matrices (LDPE, low-density polyethylene and PP, polypropylene) and with different types of wood filler (hardwood flour and softwood flour) have been investigated by means of high pressure capillary rheometry. The volume fraction of wood was varied between 0 and 60 %. The shear thinning behaviour of the WPC melts can be well described by the Ostwald - de Waele power law relationship. The flow consistency index K of the power law shows a good correlation with the volume fraction of wood. Interparticular interaction effects of wood particles can be mathematically taken into account by implementation of an interaction exponent (defined as the ratio between flow exponent of WPC and flow exponent of polymeric matrix). The interaction exponent shows a good correlation with the flow consistency index. On the basis of these relationships the concept of shear-stress-equivalent inner shear rate has been modified. Thus, the flow behaviour of the investigated wood filled polymer melts could be well described mathematically by the modified concept of shear-stress-equivalent inner shear rate. On this basis, the shear thinning behaviour of WPC can now be estimated with good accuracy, taking into account the volume fraction of wood.

Keywords: Wood Plastic Composites, High Pressure Capillary Rheometry, Rheology, Interaction Exponent, Flow Function, Interparticular Interaction Effects

1. Introduction

Flow and viscosity functions of wood filled polymer melts are shifted to higher values in comparison to those of the unfilled polymer melts (figure 1). The offset depends on the volume fraction of wood as well as the applied shear stress.

In general, the offset of the vertical shift of flow functions of suspensions can be well described by the concept of shear-stress-equivalent inner shear rate according to W. GleiBle and M. K. Baloch.

According to this concept (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 inner shear rate of the pure matrix while the external shear rate remains constant [1], [2],

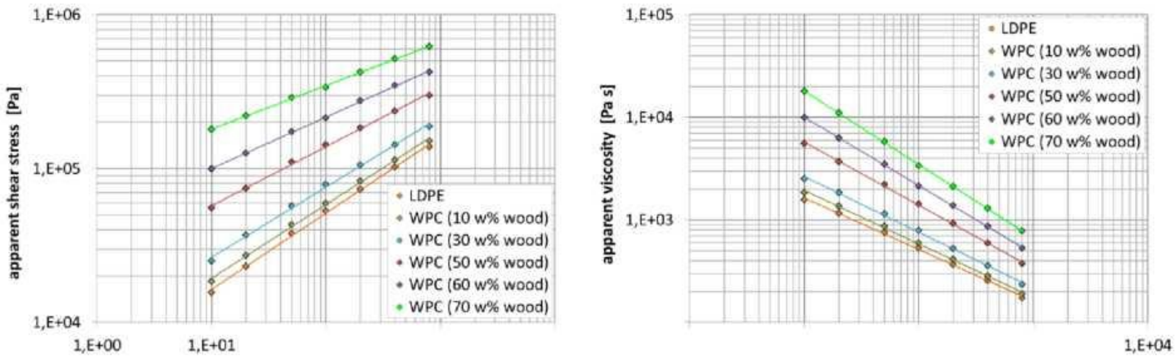

Figure 1. Flow and viscosity functions of LDPE-based WPC with varying wood content (T=190°C)

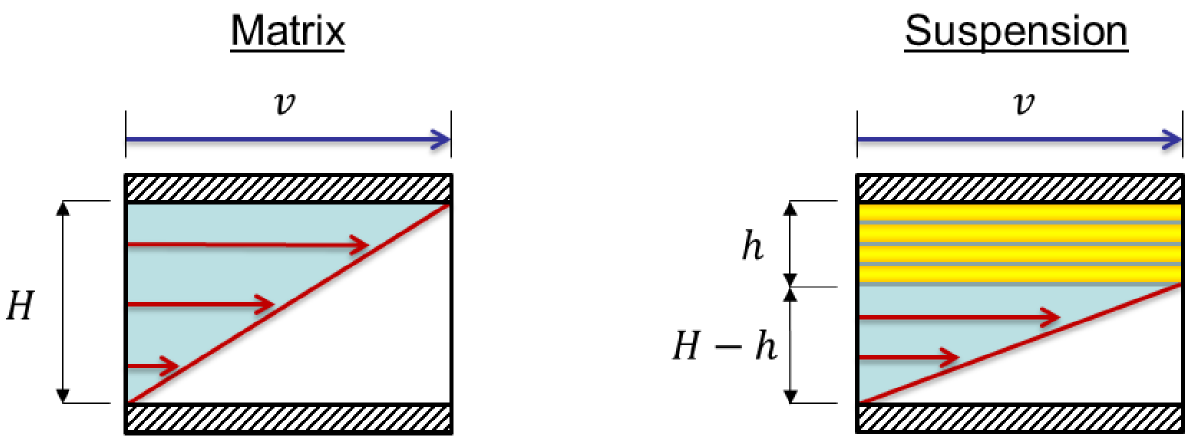

Figure 2. Idealization of flow processes in suspensions according to the concept of the shear-stress-equivalent inner shear rate

The external shear $\dot{\gamma}_0$ rate is calculated as:

$$\dot{\gamma}_0 = \frac{v}{H} \tag{1}$$

The inner shear $\dot{\gamma}_c$ rate is given by:

$$\dot{\gamma}_c = \frac{v}{H-h} \tag{2}$$

The relative increase of inner shear rate is expressed by the shift factor B:

$$B = \frac{\dot{\gamma}_c}{\dot{\gamma}_0} = \frac{H}{H-h} \tag{3}$$

The flow behavior of a shear thinning fluid, such as polymer melt, is generally described by the power law of Ostwald / de Waele (figure 3). According to the concept of shear-stress-equivalent inner shear rate, the relative increase of inner shear rate is attributed only to the volumetric content of the filler particles. Interaction effects of any kind need to be negligibly small ($n_c = n_0 = n$). On these conditions, the higher values for shear stresses of a shear thinning suspension can be calculated in accordance with shift factor as follows:

$$\tau_0 = K_0 \cdot \dot{\gamma}_c^{n_0} = \left(\frac{\dot{\gamma}_c}{\dot{\gamma}_0}\right) \cdot \tau_0 = B^{n_0} \cdot \tau_0 \tag{4}$$

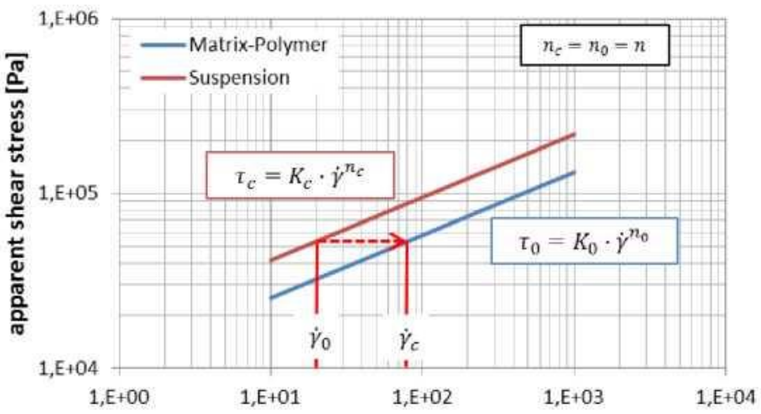

Figure 3. Comparison of flow functions (matrix-polymer and suspension) when no interactions effects

2. Materials and methods

2.1. Materials

In this study two different types of matrix polymers were used. On one hand the low-density polyethylene (LDEP, Lupolen 2420 K), which is highly branched and on the other hand the polypropylene (PP, Moplen HP 501L), which is a homopolymer with linear structure.

Softwood flour (Lignocel® BK 40/90) as well as hardwood flour (Lignocel® HB 120) were used as filler particles in this study (figure 4 and figure 5). The wood filler differ in particle size and compressibility.

Figure 4. Lignocel® BK 40/90 magnification: left: 50x, right: l00x

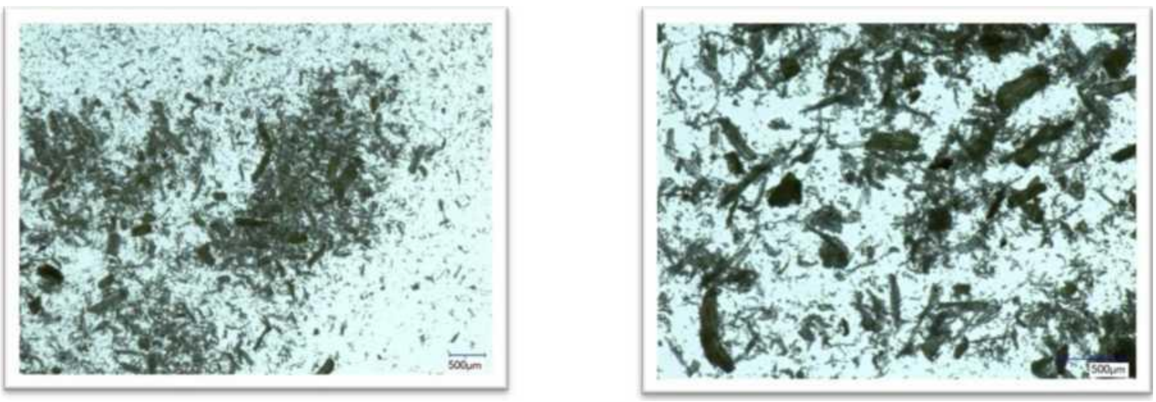

Figure 5. Lignocel® HB 120 magnification: left: 50x, right: 100x

2.2. Rheometry

All rheological measurements were performed on a high pressure capillary rheometer (CEAST Smart RHEO 5000, Instron®) using a round die with a diameter of 2 mm and a length of 40 mm. Before the measurements begun, 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. The apparent shear stress is calculated as:

$$\tau_{ap} = \frac{p \cdot r}{2 \cdot L} \tag{5}$$

with:

τ_{ap}	apparent shear stress	[Pa]
p	measured pressure	[Pa]
r	radius capillary	[mm]
L	length of capillary	[mm]

The apparent shear rate is given by:

$$\dot{\gamma}_{ap} = \frac{4 \cdot \dot{V}}{\pi \cdot r^3} = 4 \cdot \frac{R^2}{r^3} \cdot v \tag{6}$$

with:

$\dot{\gamma}_{ap}$	apparent shear rate	[l/s]
R	radius of cylinder/piston	[mm]
v	piston velocity	[mm]

3. Results and discussion

3.1. LDPE filled with softwood flour Lignocel® BK 40/90

For the unfilled LDPE and the LDPE filled with different content of softwood flour BK 40/90 the flow functions are displayed in figure 6. All flow functions show strongly pronounced shear thinning flow behaviour. The corresponding power law model parameters are reported in table 1.

It seems obvious that the fibrous structure of the wood particles has a great impact on the flow behaviour of WPC melts. Due to their eometric shape, these particles have a great tendency to interact with one another.

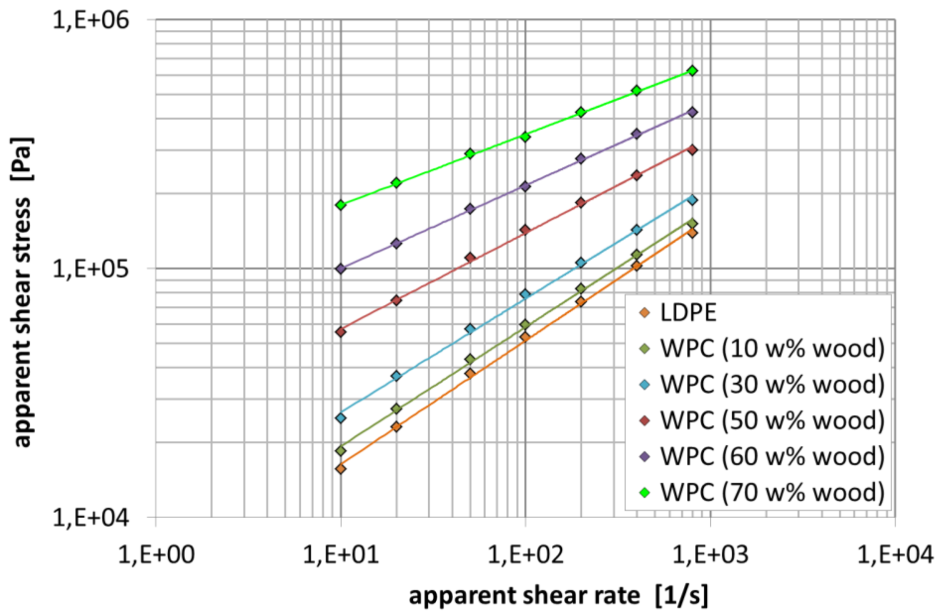

Figure 6. Flow functions of LDPE-WPC with varying wood contents (T = 190°C)

Table 1. Power law model parameters for LDPE filled with softwood flour BK 40/90

LDPE-WPC	softwood flour volumetric content [%]	K [Pa s^n]	n [-]
LDPE-BK0	0	5231	0.496
LDPE-BK10	5.9	6426	0.479
LDPE-BK30	19.4	9262	0.457
LDPE-BK50	36.0	23627	0.385
LDPE-BK60	45.7	46619	0.333
LDPE-BK70	56.7	93922	0.284

The decreasing flow index (n) with increasing wood content shows that interactions effects are by no means negligibly small. Therefore, the shift factor B, which represents the relative increase of the inner shear rate, becomes dependent on both the volumetric filler concentration and the applied shear stress. [3]

$$B(\tau)^{n_0} = \frac{K_c}{K_0^{\frac{n_0}{n_c}}} \cdot \tau^{\left(\frac{n_0}{n_c}-1\right)} \tag{7}$$

The term in the exponent of equation (7) expresses the intensity of interparticular interactions. For that reason, it can be referred to as interaction exponent χ.

$$B(\tau)^{n_0} = \frac{K_c}{K_0^{\chi}} \cdot \tau^{(\chi-1)} \tag{8}$$

Table 1 shows that the consistency (K) is dependent on the volumetric content of the wood filler. In order to describe the quantitative impact of the wood content on the consistency a mathematically correlation has been derived (figure 7).

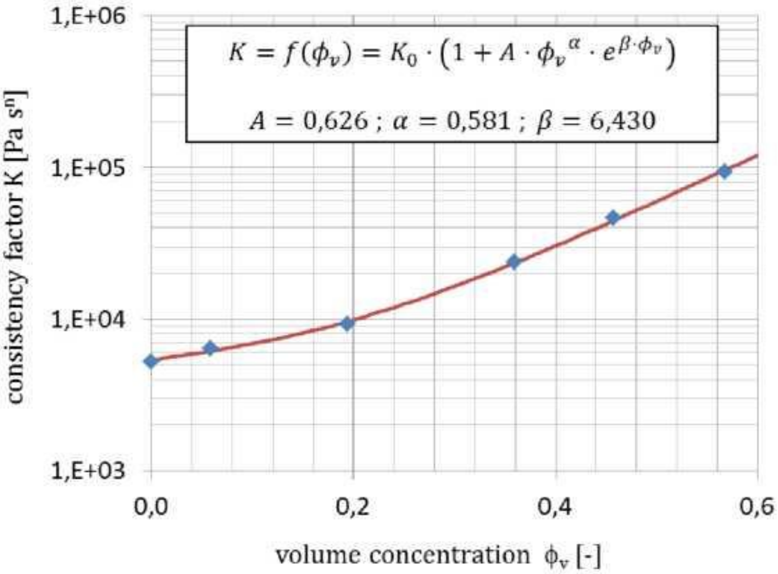

Figure 7. Dependency between consistency factor and wood content of LDPE filled with softwood flour BK 40/90 (T = 190°C)

In addition, a logarithmic correlation between interaction exponent *(x)* and consistency factor (K) was found (figure 8).

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 filled with softwood flour BK 40/90 with good accuracy (figure 9).

This approach is of great practical importance due to the good consistency to the measured values. Based on this approach the flow curves of WPC formulations based on LDPE, which is filled with softwood flour BK 40/90 can now be estimated in consideration of different volumetric contents of wood and the applied shear stress and shear rate, respectively (figure 10).

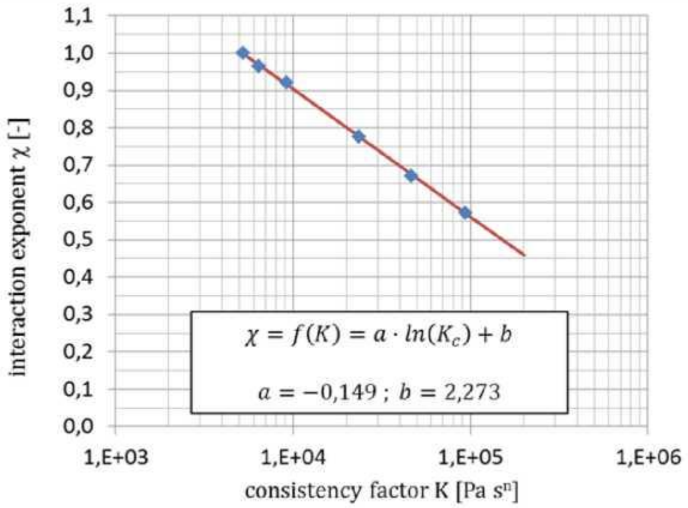

Figure 8. Dependency between interaction exponent and consistency factor for LDPE filled with softwood flour BK 40/90 (T = 190 °C)

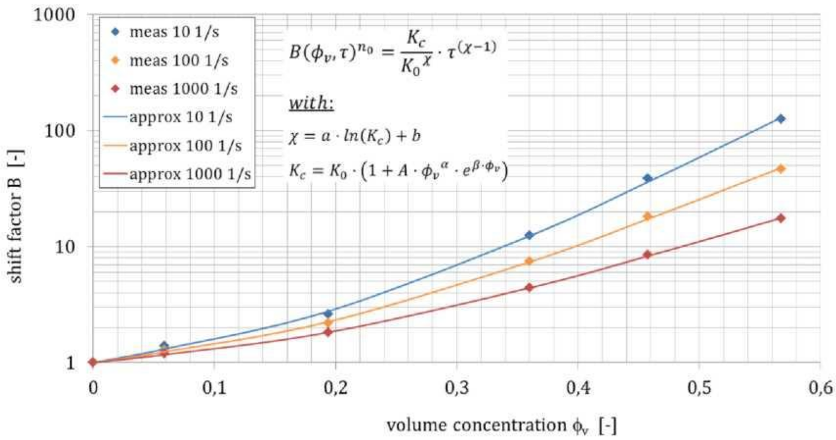

Figure 9. Shift factor B as a function of wood content and applied shear rate for LDPE filled with softwood flour BK 40/90 (T = 190 °C

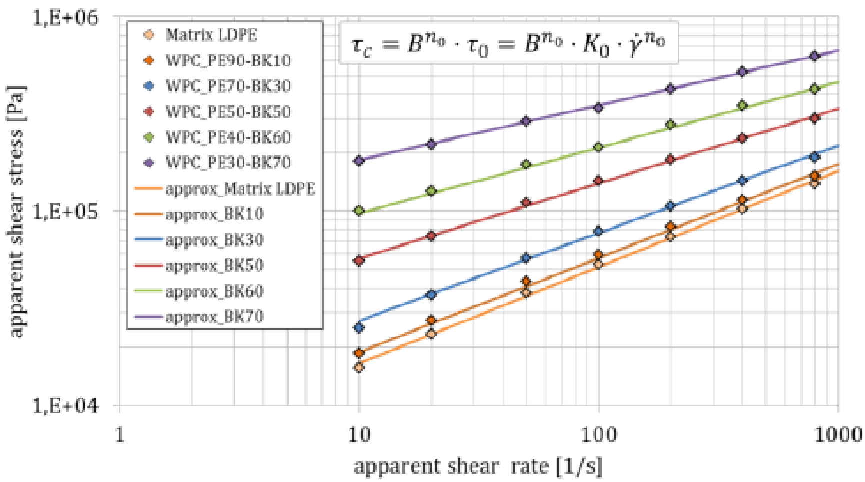

Figure 10. Comparison of estimated and measured flow functions of LDPE filled with softwood flour BK 40/90 (T = 190°C)

3.2. LDPE filled with hardwood flour Lignocel® HB 120

To investigate the applicability of this approach to wood particles with different particle sizes and compressibility, WPC formulations were prepared based on LDPE filled with different contents of hardwood flour HB 120.

Table 2 shows the corresponding power law model parameters of these WPC formulations.

Table 2. Power law model parameters for LDPE filled with hardwood flour HB 120

LDPE-WPC	hardwood flour volumetric content [%]	K [Pa s^n]	n [-]
LDPE-HB0	0	5231	0.496
LDPE-HB10	6.1	6396	0.481
LDPE-HB30	20.1	9553	0.454
LDPE-HB50	37.0	20205	0.405
LDPE-HB60	46.9	38596	0.345
LDPE-HB70	57.9	71810	0.30

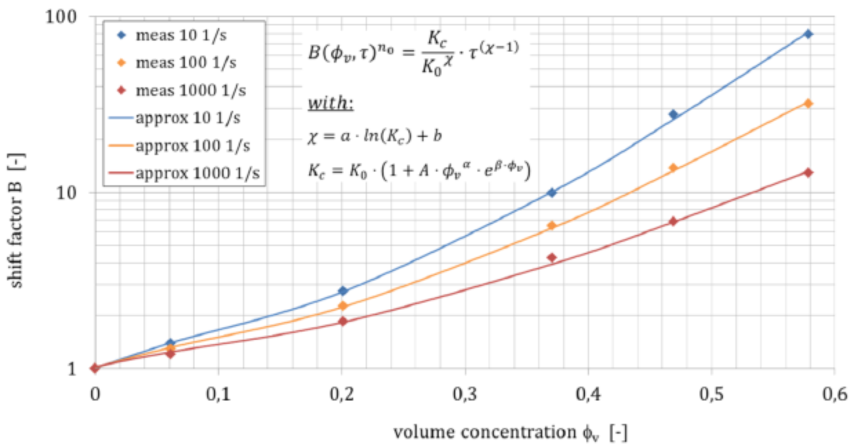

Figure 11. Shift factor B as a function of wood content and applied shear rate for LDPE filled with hardwood flour HB 120 (T = 190°C)

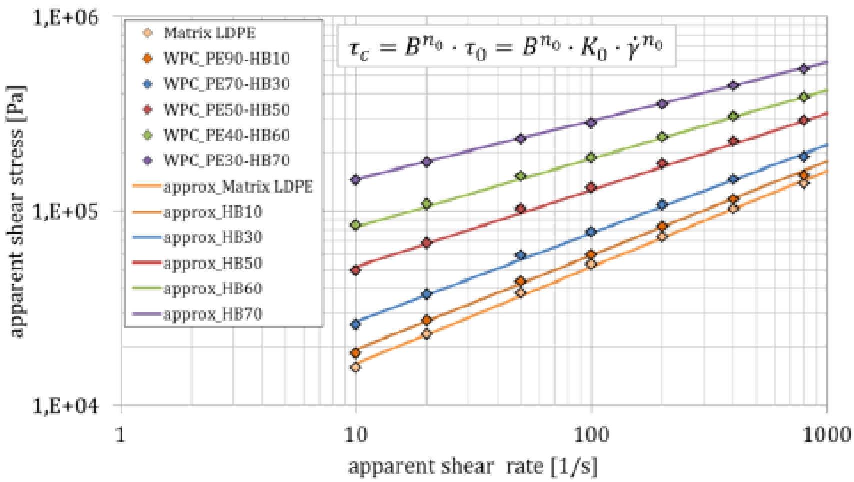

Figure 12. Comparison of estimated and measured flow functions of LDPE filled with hardwood flour HB 120 (T = 190°C)

It can be clearly seen in figure 11 and 12 that this approach is also appropriate to describe the influence of volumetric filler concentration and applied shear stress on the shift factor B for WPC formulations based on LDPE filled with hardwood flour HB 120. The estimated values correlate to the measured values with good consistency.

3.3. PP filled with softwood flour Lignocel® BK 40/90

To investigate the applicability of this approach to another matrix polymer, with different molecular structure and shear thinning flow behaviour, WPC formulations were prepared based on PP filled with different contents of softwood flour BK 40/90.

Table 3 shows the corresponding power law model parameters of these WPC formulations.

Table 3. Power law model parameters for PP filled with softwood flour BK 40/90

PP-WPC	softwood flour volumetric content [%]	K [Pa s^n]	n [-]
PP-BK0	0	12963	0.340
PP-BK10	5.9	14360	0.330
PP-BK30	19.5	21624	0.300
PP-BK50	36.1	49641	0.230
PP-BK60	45.9	70301	0.200
PP-BK70	56.9	109602	0.166

As a result of this investigation it can be concluded that this approach is also appropriate to calculate shift factor B for WPC formulations based on PP filled with softwood flour BK 40/90, by taking into consideration different volumetric filler concentrations and applied shear stresses (figure 13, 14).

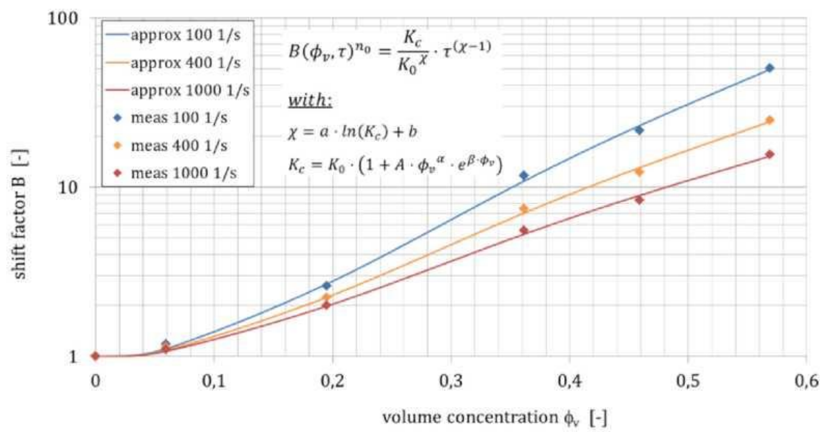

Figure 13. Shift factor B as a function of wood content and applied shear rate for PP filled with softwood flour BK 40/90 (T=200°C)

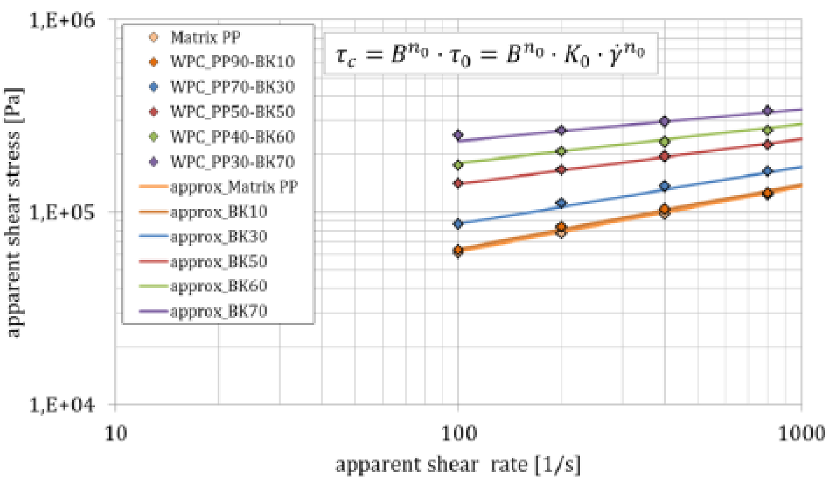

Figure 14. Comparison of estimated and measured flow functions of PP filled with softwood flour BK 40/90 (T = 200°C)

4. Conclusion

In this study, the flow behaviour of the investigated WPC formulations could be mathematically described by a modified approach, which is based on the concept of shear-stress-equivalent inner shear rate. It has been proven, that this approach is able to take account of interaction effects that typically occur in highly filled WPC formulations. On this basis, flow functions have been estimated with good consistency to the measured values, independently of the molecular structure of the matrix polymer (linear or highly branched structure) or the particle size and compressibility of the wood particles.

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