Material behaviour

Measurement of the high rate flow properties of filled HDPE melts by capillary rheometer: Effects of the test geometry

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1. Introduction

Thermoplastic polymers reinforced with rigid particles or short fibres represent a very important class of engineering materials. They are processed with the same processes adopted for unfilled thermoplastics and, at the process temperature, they consist of suspensions of rigid particles in a polymer melt. The conversion of a polymeric raw material into a finished product, that is polymer processing, does not involve only shaping, but also material structuring [1] and, for polymers filled with rigid particles, structuring operates not only at a macromolecular scale, but also at a larger scale. For reinforced polymers, processing also controls the macro-structure of the system [2] (particle distribution and, for particles with aspect ratio higher than 1, also orientation), that governs the performance of the final product. The geometry of the channels through which the filled polymer melt is forced to flow during processing, with particular reference to extrusion and injection moulding, plays a role of great importance in the formation of such macro-structure, which in turn influences the rheological behaviour of the fluid.

Capillary rheometry is the reference technique for study of the rheological behaviour of polymer melts at processing rates. It allows the determination of the material steadystate flow properties by correlating the flow rate imposed (or measured) with the pressure measured (or applied) to drive the flow of a polymer melt into a capillary die from a wider reservoir. Also, in a capillary rheometer test, the flow geometry (test geometry) can influence the macroscopically observable rheological response for polymer melts filled with rigid particles or short fibres. Although the rheological behaviour of these materials has been the subject of many theoretical and experimental studies,

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summarized in [3], attention was rarely focussed on the experimental analysis of the test geometry effects on their rheological response in capillary rheometer tests [4–7].

In this work, a capillary rheometer was used for the measurement of the high rate steady-state flow properties of micro-filled high-density polyethylene (HDPE) melts, and the effects of the test geometry were investigated. More specifically, attention was focussed on the influence of the die diameter in tests carried out by a capillary rheometer equipped with cylindrical dies and operating at constant piston velocity. The flow properties examined were the localized pressure drop at the die entrance and shear viscosity. The micro-compounds examined, prepared via melt compounding, had the same filler content $(\approx 10 \text{ vol.}\%)$ in the melt state), but differed in the type of filler. All fillers were of commercial type: glass beads [of two different types differing both for dimensions (average diameter) and surface treatment (the one coated and the other uncoated)], discontinuous glass fibres and talc.

2. Experimental

2.1. Materials

The micro-compounds examined were based on a HDPE (blow moulding grade), having weight-average and number-average molar mass of 127,000 g/mol and 11,000 g/mol, respectively, and melt-mass flow rate, MFR, of 28 g/10 min (measured at 190 °C, with an extruding mass of 21.6 kg, according to ISO 1133 [8]). The details of the fillers used for the preparation of the various compounds and the actual filler volume contents are reported in Table 1. The compounding process was described in [9].

2.2. Capillary rheometry experiments

The tests were performed on a capillary rheometer Rheologic 5000 by Instron CEAST (Pianezza, Torino, Italy). In this instrument, the flow rate is imposed by a piston and the extrusion pressure monitored by a melt pressure transducer (pressure sampling time of 0.15 s) placed in the barrel at a distance of 15 mm from the die entry plane. The tests were performed at 180 °C using two sets of flat entrance circular dies with diameter. d. of 1 and 2 mm. respectively, made from Vanadis 30[™] steel. For each set, three dies characterized by different length to diameter (1/ d) ratios were used: dies with l/d = 10, 20 and 30 for the 1 mm diameter, and with l/d = 5, 10 and 20 for the 2 mm diameter (see Fig. 1). A barrel with diameter, D, of 15 mm was employed. The apparent shear rate at the die wall, $\dot{\gamma}_{app,w}$, ranged from 17.8 to 1780 s⁻¹ (with five different levels of $\dot{\gamma}_{app,w}$ per decade, equally spaced on a logarithmic scale). $\dot{\gamma}_{app,w}$ is related to the imposed flow rate, Q, and the die diameter, d, as follows:

$$\dot{\gamma}_{app,w} = \frac{32 \cdot Q}{\pi \cdot d^3} \tag{1}$$

Shear rates lower than 17.8 s⁻¹ were not considered because, for these rates, the time to reach the steady-state was too long and would probably have caused degradation problems. For the unfilled HDPE and GF compound, the experiment was repeated three times for each test geometry, to check repeatability.

In every test performed, the material, after being loaded into the instrument barrel, was subjected to a pre-heating operation characterized by a duration of 420 s and a maximum compacting force of 300 N.

3. Results and discussion

3.1. Die entrance pressure losses

Fig. 2a–e show Bagley plots constructed from the steady-state extrusion pressure, ΔP , data measured at various $\dot{\gamma}_{app,w}$ in the tests carried out on the unfilled and the

Table 1

Details of the fillers used for the preparation of the compounds (where not otherwise indicated, information from products' datasheets) and actual filler volume contents in the compounds.

Filler type (code used in the paper)	Commercial name (supplier)	Main characteristics	Filler volume content in the compound (melt state) ^d [%]
Uncoated glass beads (uGB)	0-45 (Potters Europe)	- Average diameter $^{\rm a}=39~\mu m$	9.84 ± 0.053
Coated glass beads (cGB)	5000 CP 0102 (Potters Europe)	- Average diameter $^a=8\ \mu m$ - Coated with a coupling agent for polyethylene	10.18 ± 0.072
Discontinuous glass fibres (GF)	Chop Vantage HP 3299 (PPG Fiber Glass Europe)	- Nominal diameter = 14 μm - Nominal fibre length^b = 4.5 mm - Coated with a coupling agent for polypropylene	9.85 ± 0.030
Talc	Extra A (Luzenac)	- Pure lamellar talc - Lamellae thickness ^c < 1 μ m - Lamellae width ^c \approx 10 μ m	9.51 ± 0.002

^a Measured from optical microscope photographs.

^b Datum before mixing. After blending, in the pellets of GF compound, weight-average and number-average fibre lengths of 640 and 470 μm, respectively, were evaluated as described in [9].

^c Estimated from scanning electron microscopy, SEM, photographs.

^d Calculated for each sample examined from the measured weight content (filler weight contents and experimental details concerning their measurement reported in [9]) by considering for polymer melt, glass fillers (beads and fibres), and talc a density of 0.760, 2.50 and 2.78 g/cm³, respectively; density values: for glass beads and talc, read from the datasheets; for the fibres, assumed equal to that of glass beads; for the polymer melt, measured by melt flow rate tests carried out by a Modular Melt Flow Indexer by Ceast, at 190 °C, with an extruding mass of 21.6 kg, according to ISO 1133 [8].



Fig. 1. Schematic representation of the flow channel geometries (dies) used in the capillary rheometer tests (scale drawing).

filled polymer melts, with 1 and 2 mm diameter dies. For the unfilled HDPE and GF compound, each pressure datum reported in the graphs is the average of three values measured in replicated experiments (standard deviation smaller than symbol size). The maximum value of $\dot{\gamma}_{app,w}$ indicated in each graph is the highest for which at least two valid ΔP data points were measured. For the unfilled and the glass-filled melts (uGB, cGB and GF compounds), the lack of valid steady-state extrusion pressure data at high levels of $\dot{\gamma}_{app,w}$ is due to the occurrence of flow instability of "stick-slip" type, which did not permit the achievement of a steady-state. For talc compound, at $\dot{\gamma}_{app,w} = 1780 \text{ s}^{-1}$, ΔP data could not be obtained in the test with the 2 mm dies since the reservoir had been emptied. In the tests for this compound with the 1 mm dies, valid ΔP data could not be measured since, with the longest die (1 = 30 mm) the maximum capacity of the pressure transducer (50 MPa) was reached, whereas with the other dies (l = 10 and)20 mm) the pressure signal showed some irregular fluctuations which did not allow the identification of a valid steady-state. These pressure fluctuations could not be directly related to the occurrence of flow instability of a clearly classifiable type, also in consideration of the fact that no evident distortion was observed on the extrudate surface. The melt fracture behaviour of the compounds examined is discussed in [9], and the effect of lamellar talc particles on the melt fracture behaviour of HDPE/talc compounds is at present under a more extended investigation by the authors.

For both the unfilled and the filled melts, at the levels of $\dot{\gamma}_{app,w}$ for which three valid data points were obtained, the dependence of the extrusion pressure, ΔP , on the die length to diameter ratio, l/d, is characterized by a high degree of linearity for both die entry geometries, as shown by the fact that the correlation coefficient of the least square regression straight line that interpolates the $\Delta P vs l/d$ data was not lower than 0.9995 at any given flow rate. The die entrance pressure drops, ΔP_{ent} , at each $\dot{\gamma}_{app,w}$ were determined, as usual, by extrapolating to zero the least square regression line that interpolates the experimental data. Among the series of $\Delta P vs l/d$ data composed of only two data points, in

three cases (cGB compound, 1 mm die; uGB compound, 2 mm die; GF compound, 2 mm die, $\dot{\gamma}_{app,w} = 1000 \text{ s}^{-1}$) a value of ΔP_{ent} considerably different from that expected from the trend outlined by ΔP_{ent} data evaluated at lower $\dot{\gamma}_{app,w}$ was obtained (regression straight line plotted as a dashed line in Fig. 2) and, since evaluated from ΔP data very close to the "stick-slip" region, disregarded.

In Fig. 3a–e, the die entrance pressure drops, ΔP_{ent} , for the unfilled and the filled melts are reported as a function of the true shear stress at the die wall (corrected for ΔP_{ent}), τ_w , in a double-logarithmic plot, from tests carried with 1 and 2 mm diameter dies. For each die diameter, at each $\dot{\gamma}_{app,w}$ explored, the value of τ_w is the average of the values from the dies with different l/d. Data points with ΔP_{ent} lower than 70 kPa, which deviate largely from the trend outlined by the data at higher levels of τ_w , have been disregarded as their values are comparable with the statistical error induced by the extrapolation of ΔP_{ent} from experimental data whose accuracy depends on that intrinsic of the pressure transducer. From this figure, it emerges that, within the explored range of shear stress, the dependence of ΔP_{ent} on τ_w appears quite linear (in the doublelogarithmic plot) for each material and die diameter, and the data of each set have been fitted by a power-law function. A power-law-type dependence within a finite range of shear stress is typically observed in unfilled polymer melts (see [10]), and it has been already seen in melts filled with micro- [11] or nano-particles [12]. In order to facilitate comparison between the unfilled polymer and the filled melts, the $\Delta P_{ent}{-}\tau_w$ power-law fitting curves (lines in a double-logarithmic plot) of the unfilled HDPE are reported also in all graphs of Fig. 3 that refer to a specific compound.

From Fig. 3, it clearly emerges that, with a fixed die entry geometry, the effects promoted by the presence of the rigid micro-particles on ΔP_{ent} depend on the type of filler dispersed in the HDPE melt. In particular, with respect to the unfilled polymer, particles with relatively high aspect ratio (lamellar particles of talc and discontinuous fibres) induce an increase in ΔP_{ent} , whereas particles with aspect ratio equal to 1 (glass beads) promote a decrease in ΔP_{ent} . It is known that when a polymer melt is forced to flow from a



Fig. 2. Bagley plots from experiments carried out with dies having diameter, d, of 1 mm (full symbols) and 2 mm (open symbols) for (a) unfilled HDPE, (b) uGB compound, (c) cGB compound, (d) GF compound, (e) talc compound. The least square regression straight line that interpolates the ΔP_{ent} vs l/d data, at each $\dot{\gamma}_{app,w}$ examined, is traced; where a ΔP_{ent} datum of questionable reliability was obtained (see text, Section 3.1) a dashed line is used.



Fig. 3. Die entrance pressure drops, ΔP_{ent} , as a function of the shear stress at the die wall, τ_{w} , for (a) unfilled HDPE, (b) uGB compound, (c) cGB compound, (d) GF compound, (e) talc compound, from experiments carried out with dies having diameter of 1 mm (full symbols) and 2 mm (open symbols). Power-law fitting curve is traced for each series of data (continuous and dashed lines refer to 1 and 2 mm diameter die series, respectively). In the graph of each compound (b–e), the power-law fitting curves of the unfilled HDPE, represented with thinner lines, are also reported (continuous and dash-dot lines refer to 1 and 2 mm diameter die series, respectively).

wide reservoir, such as the rheometer barrel, into a capillary die, it is subjected to a converging (or entry) flow in the region of the die entrance. It has been shown that, for a polymer melt filled with discontinuous glass fibres, the converging flow at the die entrance aligns the fibres, and the extent of the alignment process, which is reflected in the value of ΔP_{ent} , is governed by the aspect ratio of the fibres [11,13]. The role played by the rigid particles in the converging flow of a polymer melt filled with lamellar particles could be interpreted by analogy with the action exerted by discontinuous glass fibres – this has been proposed even in the presence of nano-platelets (silicate layers) [12,14]. Thus, the increase in ΔP_{ent} observed for GF and talc compounds, with respect to the unfilled melt, might be explained as the consequence of the alignment processes of the highly anisotropic rigid particles during converging flow in the die entrance region. Literature indicates that, depending on the system examined, the presence of rigid spherical particles, which cannot undergo alignment processes in the entry flow, can produce different effects. Glass spheres can either increase ΔP_{ent} (as observed by Hristov and Vlachopoulos [15] for filled HDPE, with narrow molecular weight distribution, and lowdensity polyethylene melts), or leave ΔP_{ent} practically unchanged (as observed by Laun [11] for a filled polyamide 6 melt), or even induce a decrease in ΔP_{ent} (as observed by Liang [16,17] for filled polypropylene and low-density polyethylene melts), with respect to the unfilled polymer melt. The effect induced by glass beads in the uGB and cGB compounds examined here is of this latter type. The mechanisms at the basis of this effect, which has important implications for polymer processing, are still not fully understood, and this interesting phenomenon deserves further analysis (the processability of compounds containing glass beads in processes that might benefit from ΔP_{ent} reduction in the converging flows, such as microinjection moulding, is under investigation [18]).

From Fig. 3, it also clearly emerges that, for both the filled and the unfilled melts, ΔP_{ent} at a given τ_w depends on the die entry geometry. In particular, ΔP_{ent} is higher with the 1 mm die entry geometry than with the 2 mm one. In converging flow that is constituted by a superposition of shear and elongational flows, the polymer melt undergoes strong stretching along the die axis direction. This extensional process contributes to ΔP_{ent} , which can be directly related to the elongational viscosity of the polymer melt (the higher the elongational viscosity, the higher is ΔP_{ent}). Several models (converging flow models) have been proposed in literature to determine elongational viscosity data from ΔP_{ent} data measured for a known die entry geometry, and the most used are those due to Cogswell [19], Binding [20] and Gibson [21]. The considerable die diameter effect on ΔP_{ent} observed here for the unfilled HDPE can be interpreted as the consequence of the dependence of the elongational viscosity on the elongational strain, ε , experienced during the flow. In isothermal uniform elongational flow at a given strain rate, the elongational viscosity increases with ε , up to, if reached, the achievement of a steady-state - the elongational behaviour of unfilled HDPE melts with macromolecular characteristics similar to the HDPE analysed here can be found in [22]. In particular, it can be ascribed to the different levels of elongational strain accumulated globally, $\varepsilon_{g,a}$, by the material in converging flow through the two different die entry geometries. $\varepsilon_{g.a.}$ can be calculated as:

$$\varepsilon_{\text{g.a.}} = 2\ln\left(\frac{D}{d}\right)$$
 (2)

where D and d are the diameter of barrel and die, respectively [23]. In the converging flow that takes place with the 1 mm die, the level of $\varepsilon_{g.a.}$ is higher than in the converging flow with the 2 mm die $(\epsilon_{g.a.} \text{ is 5.4 s.u. and 4.0 s.u. for the}$ 1 mm die and the 2 mm die converging flows, respectively). For the filled melts, this die diameter effect on ΔP_{ent} is either enlarged or reduced, with respect to the unfilled melt, depending on the type of particles dispersed in the polymer melt. More specifically, in the presence of glass beads, either large and uncoated (uGB) or small and coated (cGB), the die diameter effect appears enhanced, whereas in presence of glass fibres it appears strongly contained, with respect to the unfilled melt. These results seem to suggest that the ε dependence of the elongational viscosity of the polymer melt also governs the die diameter effect on ΔP_{ent} in the presence of rigid fillers dispersed in the melt, even if the specific distortions to the flow of the polymer in the die entry region due to the presence of rigid particles

also have a role in the filled melts. The action exerted by the spherical particles, which interact with the surrounding polymer melt in such a way to foster the flow of the material into the die, seems to be enhanced in converging flow into a die with larger diameter. By contrast, the alignment processes undergone by the glass fibres in both the two different entry flows, responsible for the high levels of ΔP_{ent} of the GF compound, tend to overshadow the $\varepsilon_{g.a.}$ dependence of ΔP_{ent} exhibited by the polymer melt. This is not observed for the talc compound, for which an appreciable die diameter effect on ΔP_{ent} is found. This is probably due to the fact that the alignment processes undergone by lamellar talc particles in the converging flow produce, globally, less effective distortion of the flow of the polymer melt compared to glass fibres, which are much larger (fibres have an average length of $\approx 500 \,\mu\text{m}$, whereas talc platelets a lateral dimension of $\approx 10 \ \mu m$) and closer to the die diameter.

3.2. Flow curves

Fig. 4a–e show the steady-state $\tau_w vs \dot{\gamma}_{app,w}$ curves for the unfilled and the filled melts, obtained from the tests performed with the 1 mm and the 2 mm diameter dies. In order to facilitate comparison between the unfilled polymer and the filled melts, the τ_w curve of the unfilled HDPE obtained with the 1 mm die is also reported in all graphs of Fig. 4 that refer to a specific compound. It clearly emerges that, irrespective of the die diameter considered, only the presence of glass fillers makes the filled melts significantly more viscous than the unfilled one in the whole range of $\dot{\gamma}_{app,w}$ explored. By contrast, talc particles embedded in the polymer melt induce very little increase in viscosity only at the lowest levels of $\dot{\gamma}_{app,w}$ whereas, above 100 s⁻¹, they make the filled fluid slightly less viscous than neat polymer. The fact that, at the levels of $\dot{\gamma}_{app,w}$ explored here, the presence of talc particles produces very little effect on the steady-state shear viscosity function of the filled melt, with respect to the unfilled one, is also reported in other literature [6,24–26].

The slight decrease in viscosity observed for the talc compound with respect to the unfilled HDPE should be directly related to the ability of the lamellar talc particles to be oriented during the flow in the capillary at high rates. This viscosity decrease might be the consequence of slip processes localized at the interface between the polymer melt and the aligned talc platelets, in the region of the die where the material undergoes higher shear rates. Indeed, the authors in a previous paper [9] showed that for the talc compound, in the viscometric flow through a circular die at a moderately high shear rate (100 s^{-1}), two regions characterized by remarkably different particle alignment levels can be distinguished: an annular region near the die wall (where the fluid experiences the highest shear rates), in which the platelets are locally aligned parallel to the die wall; a circular region around the die axis, where the low shear rates are not sufficient to align the talc particles. In support of the above discussion is the observation that the results obtained here for the talc compound with the two different die diameters (discussed below) are opposite to what is expected in the presence of slip at the capillary wall



Fig. 4. Shear stress at the die wall, τ_{w} , as a function of the apparent shear rate at the die wall, $\dot{\gamma}_{app,w}$, for (a) unfilled HDPE, (b) uGB compound, (c) cGB compound, (d) GF compound, (e) talc compound, from experiments carried out with dies having diameter of 1 mm (full symbols and continuous line) and 2 mm (open symbols and dashed line). In the graph of each compound (b–e), the data points of the unfilled HDPE (1 mm diameter die), represented with smaller circles, are also indicated.

[27], suggesting that wall slip should not give a contribution to the apparent viscosity decrease, with respect to the neat HDPE. Also, in the presence of nano-platelets (silicate layers) dispersed in a polymer melt, a decrease in viscosity with respect to the unfilled polymer has been observed [12,14,28].

The increase in viscosity induced by the glass fillers in GF, uGB and cGB compounds was expected on the basis of literature results [3] and, as underlined by Laun [11], it arises from the fact that these rigid particles cannot deform with the matrix, giving rise, at a microscopic scale, to shear rates experienced by the polymer melt higher than those imposed macroscopically, as well as to extra-flow of the polymer melt around the solid particles. Among the glass

fillers examined here, uGB particles appear the most effective at promoting an increase in viscosity with respect to the unfilled melt (the viscosity increase varies between 40%, at 17.8 s⁻¹ with the 2 mm die, and 24% at 316 s⁻¹ with the 1 mm die). Nevertheless, the difference between the values of τ_w measured for the two different systems containing glass beads (uGB and cGB compound, respectively) is small (τ_w variation is smaller than 7% with both the die diameters, leaving out of consideration the datum of the cGB compound measured at 178 s⁻¹ and with d = 1 mm). This indicates that, for these glass bead filled melts flowing through 1 or 2 mm diameter dies at high levels of $\dot{\gamma}_{app,w}$, (i.) surface treatment of glass beads (uGB are uncoated, whereas cGB coated with a coupling agent for

polyethylene), which should govern filler-polymer interactions, and (ii.) number density of spheres (there are two orders of magnitude between the number densities estimated for the two different compounds, by considering the average diameters of the spheres) play a minor role, with respect to the role played by the volume fraction of the secondary phase.

Focussing attention on the die diameter effect, by comparing the curves obtained for each material with the two different die diameters, it can be observed that: (i.) for the unfilled polymer and the uGB compound, the curves obtained with the two different die diameters overlap well within the whole range of $\dot{\gamma}_{app,w}$ explored; (ii.) for the cGB compound, the two τ_w curves overlap up to 100 s⁻¹ and then diverge; (iii.) for the GF and the talc compounds, both containing non-spherical particles, the two τ_w curves do not appear superimposed. It is worth noting that, in this latter case, the variations observed between τ_w data measured with the two different dies at a given $\dot{\gamma}_{app,w}$ are limited (the highest variation, observed for the GF compound at 17.8 s⁻¹, is of 9%).

For the GF compound, for which the die diameter effect on τ_w is more evident, the $\tau_w vs \dot{\gamma}_{app,w}$ curve obtained with the 1 mm die is above the 2 mm die curve over the whole range of $\dot{\gamma}_{app,w}$. The difference observed at a given $\dot{\gamma}_{app,w}$ might be ascribed to the occurrence of specific fibre/wall interactions, which are expected to increase the energy necessary to sustain the capillary flow to an extent that depends on the surface/volume ratio characteristic of the channel through which the filled fluid is forced to flow. In the flow of a fibre filled melt, the length of the discontinuous fibres are often comparable to the dimensions of the cross-section of the flow channels (that here is the rheometer die), and the rheological behaviour of the filled melt might be affected by interactions with the solid boundaries [3]. The higher the surface/volume ratio of the die, that is the case of the 1 mm die for the testing geometries here examined, the more enhanced should be the fibre/wall interactions. The extent of these interactions might be related also to the macro-structural characteristics of the fibre filled fluid, which can vary depending on the die diameter through which the fluid is forced to flow. Wu [29] pointed out that, in the capillary flow of a polymer melt filled with discontinuous glass fibres, order-disorder transitions in fibre orientation can occur by changing the level of $\dot{\gamma}_{app,w}$, and that the $\dot{\gamma}_{app,w}$ at which a transition takes place depends on the capillary diameter. Therefore, the possibility that τ_w values evaluated for the GF compound at a given $\dot{\gamma}_{app,w}$ with the two different die diameters refer to two different levels of fibre orientation with respect to die axis cannot be ruled out. In connection with this, it is interesting to observe also that, for the GF compound, the fibres coming from the 2 mm die were, on average, longer than those from the 1 mm die. The length of the fibres was measured on the residue of calcination (in a muffle furnace at 550 °C for 60 min) by means of an optical microscope; weight-average and number-average fibre lengths of 570 and 450 μ m, respectively, evaluated as described in [29], were obtained for the 2 mm die, and 490 and 380 µm, respectively, for the 1 mm die. The extrudates analyzed were obtained at a $\dot{\gamma}_{app,w}$ of 178 s⁻¹ with dies chosen in

such a way to ensure that similar levels of extrusion pressure were obtained with the two different die diameters (d = 1 mm and l = 20 mm, and d = 2 mm and l = 40 mm,respectively). By considering the average fibre length measured on the pellets (see Table 1), it clearly emerges that glass fibres undergo fragmentation during the capillary rheometer test, in which the filled melt experiences a converging flow followed by a simple shear capillary flow, and that the 1 mm die test is more effective than the 2 mm die at promoting such fragmentation. It is reasonable to think that both fibre/wall and fibre/fibre interactions are responsible for these fibre breaking processes.

For the talc compound, in spite of the die diameter effect on τ_w being very contained, it can be observed that the two τ_w curves cross. τ_w from the 2 mm die is higher than that from the 1 mm die for the lowest levels of $\dot{\gamma}_{app.w}$, whereas the opposite occurs at the highest levels of $\dot{\gamma}_{app,w}$ explored. By considering that (i.) most of the alignment processes undergone by high aspect ratio particles is expected to occur in the entry flow at the die entrance [11], and that (ii.) at a given $\dot{\gamma}_{app,w}$ the entry flow that takes place with the 1 mm die differs from that with the 2 mm die, both for the level of $\varepsilon_{\sigma,a}$ and the absolute time scale of the process (with the 2 mm die, the flow rate is eight times that produced with the 1 mm die), the slight die diameter effect on τ_{w} observed here might be explained as the consequence of different arrangements of talc platelets in the polymer melt during the capillary flow (different levels of local alignment and/or different relative extensions of the two regions described above). For the highest levels of $\dot{\gamma}_{app,w}$ explored, where the lower the die diameter the higher the level of τ_{w} , a possible contribution of platelet/wall interactions cannot be ruled out. As evidenced by Joseph et al. [7], particles dispersed in a polymer melt can come in contact with the die wall, enhancing the level of τ_w to an extent that depends on die diameter.

For the cGB compound, the lack of overlapping beyond 100 s^{-1} might be ascribed to the activation of processes (such as wall slip) in the 1 mm die, that is the die with the highest surface/volume ratio, that should probably be related to the occurrence of "stick-slip" instability observed with this die diameter at 316 s⁻¹.

4. Conclusions

The effects of the die diameter on the measurement by capillary rheometry of the high rate steady-state flow properties of micro-filled HDPE melts were investigated. The micro-compounds examined had the same filler content (\approx 10 vol.% in the melt state), but differed in the nature and geometry of filler (glass beads, discontinuous glass fibres and talc).

A die entry geometry effect on the localized pressure drop at the die entrance, ΔP_{ent} , was observed for both the unfilled and for each filled melt: a higher value of ΔP_{ent} is obtained with the highest barrel to die diameter ratio, D/d, at any given shear stress at the die wall. The results indicate that this effect, ascribable for the unfilled polymer to the strain dependence of the elongational viscosity, can either be enhanced, in the presence of glass beads, or largely reduced, in the presence of discontinuous fibres. With a fixed die entry geometry, the effects promoted by the presence of the rigid fillers on ΔP_{ent} , with respect to the unfilled polymer turned out to be largely dependent on the type of filler: fillers with relatively high aspect ratio (lamellar particles of talc and discontinuous fibres) induce an increase in ΔP_{ent} , whereas a decrease in ΔP_{ent} is promoted by spherical particles, irrespective of sphere size and particle surface treatment.

For the filled melts, the steady-state high rate shear viscosity function turned out to be governed by the type of fillers dispersed in the polymer melt, with the die diameter playing only a minor role. A die diameter effect was noticed only for the melts filled with talc particles and discontinuous fibres. This seems to indicate that only in the presence of particles with relatively high aspect ratio, which may undergo orientation during the flow, the test geometry used in the capillary rheometer tests affects the macroscopic steady-state shear viscosity response of the filled polymer melts.

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