Relationship Between Flow Index and Apparent Viscosity of Pseudoplastic Polymers

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Abstract: *This work presents a proposal to relate the melt flow index with rheological parameters such as apparent viscosity, taking into account its dependence on the shear rate. The results obtained for polypropylene, which is pseudoplastic, proved that the proposed model is adequate, accurately fitting the experimental data.*

Keywords: *Melt flow index, Apparent viscosity, Pseudoplastic polymers, Volumetric flow rate..*

I. INTRODUCTION

When Oakes created the melt flow index (*MFI*), he was only interested in developing a parameter or an index that would provide information regarding the processability and quality of the polyethylene produced by ICI [1]. Currently, *MFI* is standardized by as ASTM and DIN and is a widely used method with the initial objective, but for all thermoplastic polymers.

Because it is a simple measure, the *MFI* is often used as the only parameter for determining the processing conditions of a thermoplastic polymer, given that the necessary rheological functions depend on expensive equipment. Some works have been published relating *MFI* indirectly with properties such as molar mass [1] and extrusion variables [2], and directly with volume flow [1,3]

The relationship between *MFI* and volumetric flow rate is direct, since *MFI*, given in g/10min, is a measure of mass flow rate, and it is easy to convert it into volumetric flow rate, Q, in cm^3/s, as follows:

$$MFI = \alpha\rho Q \qquad (1)$$

where α is the conversion factor from cm^3/s to g/10min, and ρ is the density given in g/cm^3.

Methods that attempt to relate *MFI* to the most important rheological variables fail to consider the polymer viscosity, η, as being Newtonian. This is seen in Brener's proposition [1], who, using the Hagen-Poiseuille equation for Newtonian fluids in capillaries, proposed if:

$$Q = \frac{\pi\Delta P R^4}{8\mu l} \qquad (2)$$

where μ is Newtonian viscosity, in Pa.s, $\frac{\Delta P}{l}$ is the pressure gradient, in Pa/m, R is capillary radius, in m.

Combining Eq.(1) and Eq. (2) results in

$$MFI = \left(\frac{\pi\alpha\Delta P R^4}{8l}\right)\mu^{-1} \qquad (3)$$

Eq. (3) is named Brener's model. However, few polymers, even in dilute solution, have viscosity in the melt state independent of the shear rate, $\dot{\gamma}$. Even under very low shear rates, as is the case verified inside the *MFI* equipment (MFIe), most thermoplastic polymers have their viscosity profoundly altered due to pseudoplasticity, been convenient to use the term apparent viscosity, η_a, which depends on the shear rate. For this reason, the volumetric flow rate of pseudoplastic fluids in capillaries (see Figure 1) is much higher than that predicted by the Hagen-Poiseuille equation (Eq. (2)).Thus, the *MFI* experimental data present values significantly different from those produced by Eq. (3), especially if the power-law index, *n*, present in Eq. (4) stray far from the unit value for totality of melt polymers.

Almost all polymers don't obey Newton's Law for the viscosity, $\tau = \mu\dot{\gamma}$, and follow the Ostwald Power-Law seen in Eq. (4).

$$\tau = K\dot{\gamma}^n \qquad (4)$$

where $K = \eta_a \dot{\gamma}^{1-n}$ is the consistency index, usually given in Pa.s^n and $\eta_a = \tau/\dot{\gamma}$ is the apparent viscosity in Pa.s, τ is the shear stress in Pa and $\dot{\gamma}$ is shear rate in s^{-1}.

The solution for the volumetric flow of power-law fluids in capillaries, according to [livros navarro], is shown in Eq. (5).

$$Q = \frac{n}{3n+1} \pi R^3 \left(\frac{R\Delta P}{2Kl}\right)^{1/n} \tag{5}$$

Comparing Eqs. (1) and (5), we obtain the relationship between *MFI* and Q for power-law fluids given by:

$$MFI = \alpha\rho \frac{n}{3n+1} \pi R^3 \left(\frac{R\Delta P}{2Kl}\right)^{1/n} \tag{6}$$

or

$$MFI = \alpha\rho \frac{n}{3n+1} \pi R^3 \left(\frac{R\Delta P}{2\eta_a \dot{\gamma}^{1-n} l}\right)^{1/n} \tag{7}$$

Eq. (7) may be used for any shear rate.

Although the power-law flashes in regions at and the $\dot{\gamma} \to 0$ and $\dot{\gamma} \to \infty$, therefore it does not provide precise results for the apparent viscosity in both region, η_0 and η_∞ respectively, it is possible to derive an analytical expression for the $\eta(\dot{\gamma}) = \eta_a$ dependence that satisfy the conditions of finite values for $\eta(\dot{\gamma})$ when $\dot{\gamma} \to 0$ and $\dot{\gamma} \to \infty$, assuming that in the intermediate range for $\eta(\dot{\gamma})$, that is $0 \ll \dot{\gamma} \ll \infty$, it follows Eq. (4). According to Vinogradov & Malkin [4],

$$\eta(\dot{\gamma}) = \eta_\infty + \frac{\eta_0 - \eta_\infty}{1 + K\dot{\gamma}^n} \tag{8}$$

As, for most polymer, $\eta_\infty \ll \eta_0$, Eq. (8) reduces to

$$\eta(\dot{\gamma}) = \frac{\eta_0}{1 + K\dot{\gamma}^n} \tag{9}$$

Substituting Eq. (9) in Eq. (7), we get

$$MFI = \alpha\rho \left[\frac{n}{3n+1} \pi R^3 \left(\frac{R\Delta P(1+K\dot{\gamma}^n)}{2\eta_a \dot{\gamma}^{1-n} l \eta_0}\right)^{1/n}\right] \tag{10}$$

which relates MFI to η_a and provides satisfactory results as long as the temperature, shear stress and shear rate applied to both the capillary rheometer and the MFIe are the same. For rheometers with other geometry, in Appendix I, there are other solutions relating MFI and Q not used in this work.

According to [3], the average values of such and point range in the MFIe are given by:

$$\dot{\gamma}_{MFI} = \frac{MFI}{\alpha\rho R_M^3} \tag{11}$$

$$\tau_{MFI} = \frac{R_M}{8R_C^2 l_M} F \tag{12}$$

the values provided by Eqs. (11) and (12) must be adjusted for use in the capillary rheometer.

II. EXPERIMENTAL PROCEDURE

The polymer used was polypropylene supplied by Polibrasil (Camaçari, Bahia, Brazil) as it came from the manufacturer, with a density of 0.758 g/cm^3 and a melt flow index of 3.39 g/10min, measured at 190°C. Melt flow index measurements were made on a standard MFIe with a cylinder diameter of 9.474 mm, die diameter, $d_M = 2R_M$ of 2.0955 mm and die length, l_M of 8.0 mm and l/d ratio of about 4.0. The distance traveled by the piston was 25.0 mm and the applied force, F, was 5.0 Kgf. The operating temperature was 190°C. For other shear rates and l/d ratios, a HAAKE SYSTEM 90 rheometer was used, with a single screw extruder, and the temperature in all regions of the extruder fixed at 190°C.

III. RESULTS AND DISCUSSIONS

As show in Table 1, the results obtained with Eqs. (1) and (2) showed great discrepancy with the experimental results, due to not taking into account the power-law index, n, of the sample at a temperature of 190°C. As shown in Figure 1, as $n \to 0$, moving away from the unitary value, the values of the volumetric flow rate, Q, measured in the same pressure gradient and $1.0 \ll {}^l/_d \ll 10.0$, move far away from the value calculated for a Newtonian fluid. The model proposed in this work proves to be adequate, producing compatible values for $0 \ll n \ll 1.0$.
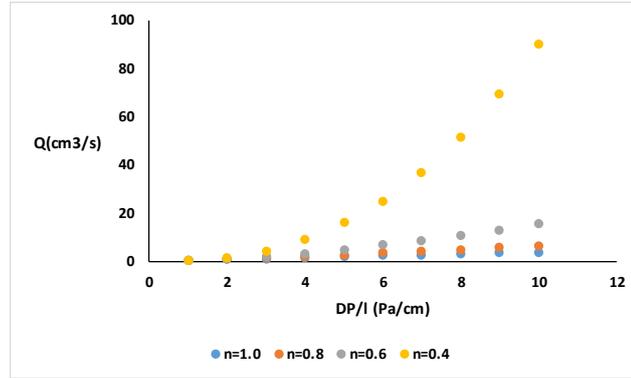
Figure 1. Influence of n on the Q x pressure drop ratio.

l/d	*MFI* Eq. (3) (g/10min)	*MFI* Eq. (10) (g/10min)	*MFI* EXP(g/10min)
4.0	0.328	3.580	3.500
6.0	0.062	0.410	0.420
8.0	0.020	0.099	0.100
10.0	0.008	0.032	0.032

Table 1. Experimental values of the MFI and those obtained by Eqs. (3) and (10).

Table 1 also presents MFI's data for *l/d* of 4.0, 6.0, 8.0 and 10.0, for a pressure gradient of 5.9 Pa/cm. The difference between the experimental values and those produced by Eq. (3) increases as the shear becomes more severe, that is, as the *l/d* ratio increases. This is because the index *n* characterizes not only the departure from Newtonian behavior but also indicates the degree of sensitivity to shear [5]. As this model does not consider the index *n*, it fails. Eq. (10), on the contrary, includes the power-law index and can be used in any shear range, that is, for any *l/d* ratio.

IV. CONCLUSION

Experimental data show that the model proposed in this work fits perfectly experimental data, making it suitable to correlate the relationship between MFI and apparent viscosity under constant temperature and pressure gradient for any power-law index, $0 < n < 1.0$, and l/d ratio.

Conflict of interest

There is no conflict to disclose.

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APPENDIX I

The following equations were compiled from Navarro, 2018 [6].

1) Slit rheometer,

$$Q = \frac{2wB^2}{m+2}\left(\frac{\Delta PB}{Kl}\right)^m \tag{A1}$$

Where $m = \frac{1}{n}$, w is the width of the slit and B is the half of slit height.

So, Eq. (1) becomes

$$MFI = \alpha\rho \frac{2wB^2}{m+2}\left(\frac{\Delta PB}{Kl}\right)^m \tag{A2}$$

2) Annular geometry rheometer with linear movement of the inner cylinder

$$Q = \frac{2\pi R^2 V}{\Gamma^{1-m}-1}\left[\frac{(1-\Gamma^{3-m})}{3-m} - \frac{1-\Gamma^2}{2}\right] \tag{A3}$$

Where R is inner radius of outer cylinder and Γ is the ratio between the outer radius of the inner cylinder and R. So,

$$MFI = \alpha\rho \frac{2\pi R^2 V}{\Gamma^{1-m}-1}\left[\frac{(1-\Gamma^{3-m})}{3-m} - \frac{1-\Gamma^2}{2}\right] \tag{A4}$$

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3) Circular duct with angle of convergence φ

$$Q = \frac{\pi R_0^2}{m+3}\left(\frac{\Delta P R_0}{2Kl}\right)^m \left[\frac{3(\varphi-1)}{m(1-\varphi^3)}\right]^m \qquad (A5)$$

Where R_0 is larger radius or input radius.

$$MFI = \alpha\rho\,\frac{\pi R_0^2}{m+3}\left(\frac{\Delta P R_0}{2Kl}\right)^m \left[\frac{3(\varphi-1)}{m(1-\varphi^3)}\right]^m \qquad (A6)$$

4) Radial flow between two parallel disks

$$Q = \frac{4\pi n}{2n+1}\left[\frac{(1-n)(P_1-P_2)}{K(r_2^{1-n}-r_1^{1-n})}B^{2n+1}\right]^{1/n} \qquad (A7)$$

Where r_1 is the input radius, r_2 is the disk radius, B is the half the separation distance between the disks.

$$MFI = \alpha\rho\,\frac{4\pi n}{2n+1}\left[\frac{(1-n)(P_1-P_2)}{K(r_2^{1-n}-r_1^{1-n})}B^{2n+1}\right]^{1/n} \qquad (A8)$$