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Polyolefins Journal, Vol. 13, No. 1 (2026)
DOI: 10.22063/POJ.2026.35836.1384

ORIGINAL PAPER

Linking pressure-dependent viscosity of polyolefin melts to compressibility and free volume

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Received: 3 January 2026, Revised: 11 February 2026, Accepted: 19 February 2026

ABSTRACT

The effect of hydrostatic pressure on the viscosity of polymer melts plays a critical role in injection molding processes, particularly under high-pressure conditions typical of thin-walled and micro-scale components. Despite its importance, pressure-dependent viscosity data are often incomplete or unavailable in commercial simulation software, especially for semi-crystalline polymers such as polyolefins. In this study, the pressure dependence of viscosity for polypropylene (PP) and high-density polyethylene (HDPE) was investigated by analyzing rheological and volumetric parameters retrieved from the Moldflow material database. The coefficient describing the pressure effect on viscosity was evaluated using a thermodynamic approach based on polymer compressibility, thermal expansion, and the temperature dependence of zero-shear viscosity, and was compared with the corresponding parameter obtained from experimental rheological measurements embedded in the Cross–WLF viscosity model. A clear linear correlation was found between the estimated and experimentally derived pressure coefficients, with the former systematically exceeding the latter by a nearly constant proportionality factor. The proposed approach enables a simple determination of material parameters for injection molding simulations and contributes to improved prediction accuracy under high-pressure processing conditions. **Polyolefins J (2026) 13: 39-44**

Keywords: Polypropylene; High density polyethylene; pressure effect on viscosity; compressibility.

INTRODUCTION

Injection molding is one of the most widely used polymer processing technologies due to its high productivity, dimensional accuracy, and suitability for complex geometries. The reliability of injection molding simulations has therefore become a key factor in reducing development costs and minimizing trial-and-error during mold design [1]. At the core of these simulations lies an accurate description of the rheological behavior of polymer melts, which is commonly modeled as a function of shear rate and temperature. However, under typical injection molding conditions - especially in thin-walled or micro-scale components - the melt is also subjected to hydrostatic pressures that can exceed several tens or even hundreds of MPa, making pressure an additional governing variable that cannot be neglected [2,3].

While the effects of shear rate and temperature on polymer melt viscosity are well documented, the influence of pressure has historically received far less attention, mainly due to experimental challenges

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associated with high-pressure rheometry [4]. As emphasized in comprehensive reviews, the scarcity and inconsistency of pressure-dependent viscosity data still represent a major limitation in polymer rheology and processing science. Nevertheless, pressure-induced viscosity increases are known to be particularly relevant for polymer processing operations such as extrusion, foaming, and injection molding, where pressure gradients directly affect flow resistance, cavity filling, and pressure transmission.

In the context of injection molding simulations, the pressure dependence of viscosity is most often introduced through phenomenological extensions of temperature-dependent models, such as the Cross-WLF formulation [5]. In this framework, pressure effects are accounted for by a shift of the reference temperature (or glass transition temperature) with pressure, typically described by a single parameter (commonly denoted as D_3) [3]. Despite its conceptual simplicity and physical interpretation in terms of free volume reduction, this parameter is frequently missing or set to zero in commercial CAE material databases, also because of the lack of standardized experimental procedures for its determination [6-8].

Studies on injection molding simulations have demonstrated that neglecting pressure-dependent viscosity can lead to significant errors in the prediction of cavity pressure evolution, flow front advancement, and filling balance, particularly for micro-injection molding of thin-walled parts or for highly viscous polymers, where high injection pressures are unavoidable [9]. These discrepancies directly impact the accuracy of simulation-based decisions regarding machine selection, mold design, and process optimization. Also, all the processing dependent properties of the materials are impossible to determine without accurate predictions [10-12]. For this reason, pressure-dependent viscosity data have increasingly been recognized as a critical input for high-fidelity injection molding simulations, especially when dealing with high surface-to-volume ratios or extreme processing conditions [13].

Among polymer families, polyolefins play a central role in injection molding due to their widespread industrial use, favorable cost-performance balance, and broad processing window. However, their semi-crystalline nature [14] and relatively low-pressure sensitivity compared to amorphous polymers [4,15-17] have contributed to an underestimation of pressure effects in both experimental characterization and simulation practice. As highlighted in the literature, even for polyolefins, pressure-dependent viscosity can significantly influence flow behavior under realistic molding conditions [18].

In this study, volumetric and rheological data from the Moldflow database were analyzed to investigate the pressure dependence of polymer melt viscosity. Moldflow is indeed one of the very few softwares which reports values for the effect of pressure on viscosity. The coefficient describing the effect of pressure on viscosity was estimated based on calculations of compressibility, thermal expansion, and the temperature dependence of viscosity. This estimated coefficient was then compared with the corresponding value reported in the Moldflow database, which is derived from experimental rheological measurements. The comparison revealed a clear quantitative relationship between the two values. These findings provide a practical approach for a more straightforward determination of the parameter governing the pressure effect on viscosity, thereby improving the accuracy of injection molding simulations.

Determination of the effect of pressure on viscosity

It is well known from the early studies by Doolittle [19], that the viscosity of polymers depends on the free volume. It is therefore quite immediate to correlate the effect of pressure on viscosity to polymer compressibility. Indeed, a mathematical relationship can be found among the parameters describing the effect of pressure and temperature on viscosity and specific volume.

In particular, if the viscosity does not change with temperature and pressure at constant volume,

$$\beta_\eta = \left(\frac{\partial ln(\eta)}{\partial P}\right)_T = \left(\frac{\partial ln(\eta)}{\partial V}\right)_T \left(\frac{\partial V}{\partial P}\right)_T = -\left(\frac{\partial ln(\eta)}{\partial V}\right)_T V \beta_V \quad (1)$$

$$\alpha_\eta = -\left(\frac{\partial ln(\eta)}{\partial T}\right)_P = -\left(\frac{\partial ln(\eta)}{\partial V}\right)_P \left(\frac{\partial V}{\partial T}\right)_P = -\left(\frac{\partial ln(\eta)}{\partial V}\right)_P V \alpha_V \quad (2)$$

where

$$\beta_V = -\frac{1}{V}\left(\frac{\partial V}{\partial P}\right)_T \quad (3)$$

is the compressibility and

$$\alpha_V = \frac{1}{V}\left(\frac{\partial V}{\partial T}\right)_P \quad (4)$$

is the volume thermal expansion.
By dividing Eq. 1 and Eq. 2 one simply obtains

$$\beta_\eta \approx \beta_V \frac{\alpha_\eta}{\alpha_V} \quad (5)$$

as reported in the book by Van Krevelen (p. 544) [20]. The effect of pressure and temperature on the zero-shear rate viscosity of polymers is considered in the so-called WLF equation, which is based on free-volume considerations and is widely used in the software for injection molding simulation:

$$\eta_T = D_1 \exp\left[-\frac{A_1\left(T - D_2 - D_3 P\right)}{A_2 + \left(T - D_2\right)}\right] \tag{6}$$

The parameters A_1, A_2, D_1, D_2 and D_3 are experimentally determined.

From Eq. 6, the parameters β_η and α_η are easily calculated as

$$\beta_\eta = \frac{A_1 D_3}{A_2 + T - D_2} \tag{7}$$

$$\alpha_\eta = \frac{A_1\left(A_2 + D_3 P\right)}{\left(A_2 + T - D_2\right)^2} \tag{8}$$

Both parameters decrease on increasing temperature, α_η increases also on increasing pressure.

The specific volume of the molten polymer can be described by the so-called Tait equation, which is also a standard equation for injection molding simulation software:

$$V_m\left(T,P\right) = \left(b_{1m} + b_{2m}\left(T - b_5\right)\right)\left(1 - 0.0894 ln\left(1 + \frac{P}{b_{3m} e^{-b_{4m}\left(T - b_5\right)}}\right)\right)$$

for $T \geq b_5 + b_6 P$

$$\tag{9}$$

The parameters b_{1m}, b_{2m}, b_{3m}, b_{4m}, b_{5m} and b_{6m} are experimentally determined. The parameter b_5 can be considered as the melting temperature at zero pressure. From Eq. 9, the parameters β_V and α_V at $T=b_5$ and $P=0$, namely at reference conditions, are easily calculated as:

$$\beta_V = \frac{0.0894}{b_{3m}} \tag{10}$$

$$\alpha_V = \frac{b_{2m}}{b_{1m}} \tag{11}$$

RESURLTS & DISCUSSION

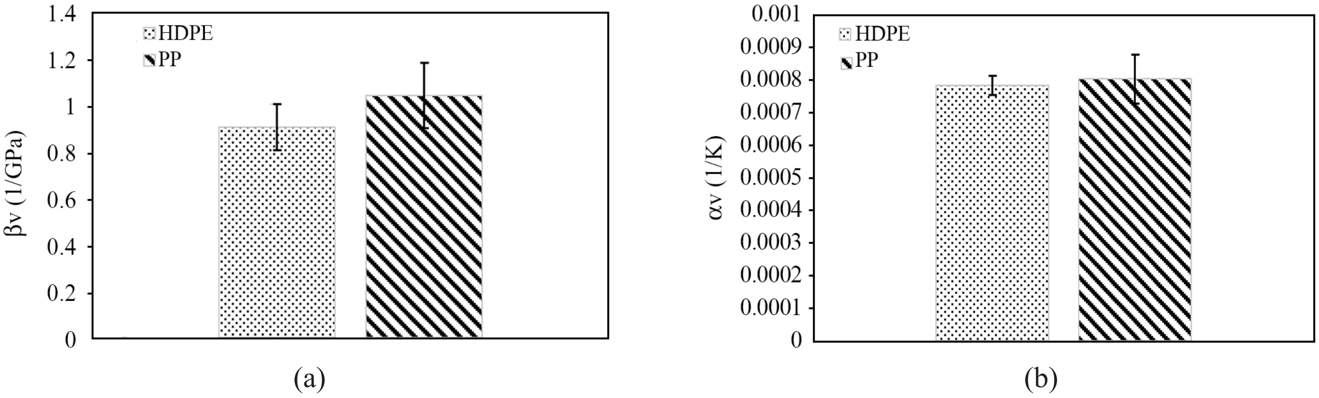
The parameters of the WLF and Tait equations were retrieved from the Moldflow database for several HDPE and PP grades. The selected grades correspond to those explicitly characterized for the pressure dependence of viscosity, namely those for which the parameter D_3 in Equation (6) was non-zero [21].

In Figure 1, the mean values of the parameters β_V and α_V are reported for both families of materials, HDPE and PP – 4 data points for HDPE, 9 for PP. The error bars refer to the standard deviations. The results show that all the materials present a quite similar dependence of the specific volume on temperature and pressure.

A similar report is given in Figure 2 concerning the parameters β_V and α_V. The results reported in Figure 2a reveal that the effect of pressure on viscosity, namely the parameter β_V, is typically lower for HDPE grades – average value 13/GPa - than for PP grades – average value 22/GPa, consistently with what reported in the literature [6,22]. The standard deviation, however, is considerably high. Concerning the parameter α_V,

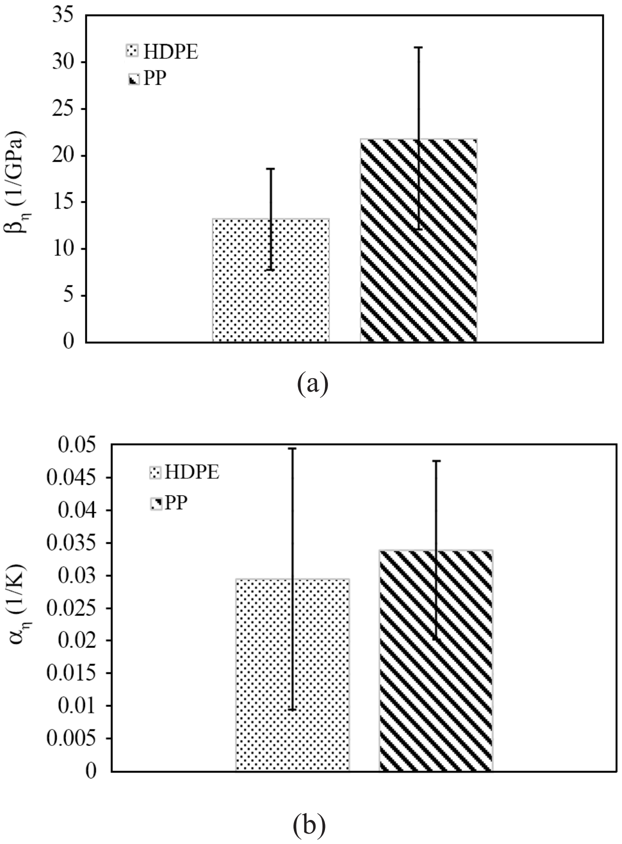
Table 1. Parameters describing the effect of temperature and pressure on specific volume and on zero-shear viscosity, as calculated from the constants reported in Moldflow database.

Polymer	**Family**	β_V **[1/GPa]**	α_V **[1/K]**	α_η **[1/K]**	$\beta_V \frac{\alpha_\eta}{\alpha_V}$ **[1/GPa]**	β_η **[1/GPa]**
RTP 0700 ABR Nat	HDPE	0.97	8.05E-04	1.48E-02	17.7	6.7
INEOS Eltex TUB 121	HDPE	0.93	7.52E-04	5.87E-02	72.5	20.0
RTP 0799 X 155233 S-46057	HDPE	0.75	7.60E-04	2.05E-02	20.2	12.9
Husky Mixed resin	HDPE	0.97	8.05E-04	2.45E-02	29.3	13.2
Sumitomo Noblen AZ864E4 - JP	PP	1.15	7.42E-04	2.97E-02	46.0	19.6
Lupol GP1007F LG	PP	1.00	8.77E-04	6.23E-02	71.2	41.2
Quingdao NH-2027H	PP	0.78	7.80E-04	3.09E-02	30.8	12.3
Aptiv M2097_002_Sup4	PP	1.00	8.27E-04	4.07E-02	49.1	21.4
Pro-fax 8523 LyondellBasell	PP	1.11	7.87E-04	3.39E-02	47.7	22.4
RTP PERMASTAT 0100 Nat	PP	0.88	9.60E-04	9.29E-03	8.5	7.0
Ineos H2E-00	PP	1.16	7.46E-04	3.31E-02	51.6	26.2
Braskem CG350HN	PP	1.06	7.63E-04	3.60E-02	50.1	28.0
Sumitomo Noblen AW564-JP	PP	1.19	7.26E-04	3.03E-02	49.8	18.0



(a) (b)

Figure 1. Mean of the values reported in Table 1 for the materials belonging to the HDPE and PP families, respectively. The error bars refer to the standard deviation. (a) compressibility; (b) thermal expansion coefficient.



(a)

(b)

Figure 2. Mean of the values reported in Table 1 for the materials belonging to the HDPE and PP families, respectively. The error bars refer to the standard deviation. a) effect of pressure on the zero-shear viscosity; b) effect of temperature on the zero-shear viscosity.

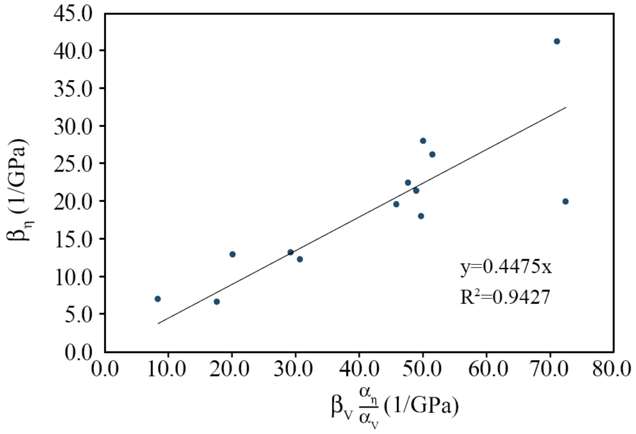


Figure 3. Relationship between the parameter β_η describing the effect of pressure on viscosity, and the same parameter estimated by Eq. (5).

reported in Figure 2b, it can be stated that the average values are similar, with a very high standard deviation.

The parameter estimated by using Eq. (5), namely $\beta_V \frac{\alpha_\eta}{\alpha_V}$, is regularly larger than β_η. The relationship between the two parameters is reported in Figure 3. It can be noticed that the two parameters are well correlated, with a linear dependence and a coefficient of proportionality equal to 0.45. Once this relationship is known, the parameter D_3 in Eq. (6) can be easily estimated by the following equation:

$$D_3 = 0.45\beta_V \frac{\alpha_\eta}{\alpha_V} \frac{A_2 + b_5 - D_2}{A_1} \tag{12}$$

which just requires the knowledge of standard WLF parameters and the description of the volumetric properties of the polymer melt.

CONCLUSIONS

In this work, the effect of hydrostatic pressure on the viscosity of polyolefin melts was investigated by combining rheological and volumetric information available in the Moldflow material database. Starting from well-established concepts linking viscosity to free volume, a quantitative relationship between the pressure sensitivity of viscosity and the thermodynamic properties of the melt - namely compressibility and thermal expansion - was derived and applied to a set of commercial HDPE and PP grades.

The analysis revealed a clear quantitative correlation between the pressure coefficient of viscosity obtained from the WLF model and the corresponding coefficient estimated from volumetric considerations. Although the latter systematically overestimates the experimentally derived values, the two parameters were shown to be linearly related, with a proportionality factor close to 0.45 for the investigated materials. This result confirms, in a quantitative manner, the physical interpretation originally proposed by Van Krevelen, according to which pressure and temperature influence polymer viscosity predominantly through their effect on free volume. The results also confirm that polypropylene exhibits, on average, a higher pressure sensitivity of viscosity than high-density polyethylene, in agreement with previous experimental findings reported in the literature.

From a practical perspective, the proposed approach provides a simple and practical method for estimating the pressure-dependent viscosity parameter in cases where direct high-pressure rheological measurements are unavailable. Since compressibility, thermal expansion, and standard WLF parameters are routinely included in commercial CAE databases, the method can be readily implemented to improve the completeness and consistency of material data used in injection molding simulations.

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