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Polymer 47 (2006) 7455–7459
polymer
www.elsevier.com/locate/polymer

Empirical determination of equation of state for zero internal pressure in rare gas solids and semi-crystalline polymers

S. Saeki*, F. Wang, Y. Tanaka

Department of Materials Science and Engineering, Fukui University, Fukui 910-8507, Japan

Received 28 March 2006; received in revised form 3 August 2006; accepted 7 August 2006
Available online 1 September 2006

Abstract

An equation of state for zero internal pressure in rare gas solids and semi-crystalline polymers has been determined based on the empirical functions of thermal pressure coefficient γ_V with respect to volume at constant pressure. The experimental data of *PVT* over wide range of temperature and pressure published by Anderson and Swenson and Syassen and Holzapfel for rare gas solids and Olabisi and Simha and Zoller for semi-crystalline polymers are used to evaluate γ_V. The function of γ_V with respect to volume determined at constant pressure is given by $\gamma_V = A\{(V-V_0)/V\}^{\varepsilon}(V_0/V)^{1+c}\exp\{-c(V-V_0)/V\}$ where V_0 is the volume at 0 K, A, ε and c are constants. The function of internal pressure $P_{\mathrm{i}} = \gamma_V T - P$ with respect to temperature at constant pressure is determined by converting the function of $\gamma_V(V)$ to a function of temperature $\gamma_V(T)$. An empirical equation of state for zero internal pressure determined by pressure P^*, volume V^* and temperature T^* at which $P_{\mathrm{i}} = 0$ is expressed by $P^*V^*/\mathrm{R}T^* = C^* - D^*V^*$ for rare gas and semi-crystalline polymer where C^* and D^* are constants. The practical meaning of the equation of state for $P_{\mathrm{i}} = 0$ in the semi-crystalline polymers has been discussed.

Keywords: Internal pressure; Thermal pressure coefficient; Semi-crystalline polymer

1. Introduction

The main purposes of the experimental and theoretical studies on the equation of state have been the accurate determination of *PVT* over wide range of pressure and temperature and their theoretical interpretation. The theoretical equation of states for semi-crystalline polymers have been derived by Pastine [1,2], Simha and Jain [3], Midha and Nanda [4], which give theoretical prediction of *PVT* data quantitatively. The extensive experimental studies of the *PVT* for the semi-crystalline polymers have been done by Olabisi and Simha [5] and Zoller [6] over the wide range of temperature and pressure. On the other hand the experimental and theoretical studies of *PVT* for rare gas solids have been done by Anderson and Swenson [7], Syassen and Holzapfel [8] and Bernardes and Swenson [9]. Based on these experimental *PVT* data it becomes possible to determine a function of isothermal compressibility and thermal expansion coefficient with respect to pressure and temperature, respectively. In previous works [10–15] we have derived a simple equation that enables an accurate determination of isothermal compressibility and thermal expansion coefficient. In this work we have determined an empirical function of thermal pressure coefficient with respect to volume at constant pressure over wide range of pressure from which the function of internal pressure $P_{\mathrm{i}} = \gamma_V T - P$ with respect to temperature can be determined. The experimental data of P_{i} against temperature at various constant pressures provide enough way to determine a characteristic pressure P^*, volume V^* and temperature T^* at which the P_{i} becomes zero. The main purpose of this work is to determine the equation of state for zero internal pressure in rare gas solids and semi-crystalline polymers by the experimental data of P^*, V^* and T^*.

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* Corresponding author. Tel.: +81 776 27 8622; fax: +81 776 27 8767.
E-mail address: saeki@matse.fukui-u.ac.jp (S. Saeki).

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doi:10.1016/j.polymer.2006.08.018

2. Empirical determination of thermal pressure coefficient and internal pressure in rare gas and semi-crystalline polymers from *PVT* data

Two steps are involved in the determination of thermal pressure coefficient. First, the isothermal compressibility is determined from the $P-V$ data at constant temperature. Second, the thermal expansion coefficient is evaluated from $V-T$ data at constant pressure. The two empirical equations derived in the previous work [12,15] are used in this work. The first equation is for the function of volume $V(P,T)$ with respect to pressure at constant temperature and is given by

$$V(P_0,T)/V(P,T) = [\{P+P_x(T)\}/\{P_0+P_x(T)\}]^{m_x} \quad T = \text{constant} \tag{1}$$

where the volume $V(P_0,T)$ is V at ($P_0 = 0$ bar,T), $P_x(T)$ is a function of temperature and m_x is a constant. The second equation is a function of temperature with respect to volume $T(V)$ at constant pressure and is given by

$$\ln(T/T_1) = B\{(V-V_1)/V\}^{n_x} \quad P = \text{constant} \tag{2}$$

where $T_1 = 1$ K, V_1 is the volume at $T_1 = 1$ K. The values of B and n_x are constant. The compressibility β_T is calculated from Eq. (1) as

$$\beta_T = m_x/\{P+P_x(T)\} \tag{3}$$

The thermal expansion coefficient α_P obtained from Eq. (2) is expressed by

$$\alpha_P = (V-V_1)/(n_x V_1 T \ln T) \tag{4}$$

From the thermal pressure coefficient γ_V defined by $\gamma_V = \alpha_P/\beta_T$ the internal pressure P_i is obtained as $P_i = \gamma_V T - P$. The typical comparisons between the experimental data and values calculated by Eqs. (1) and (2) are shown in Table 1 where the difference between the specific volumes in linear polyethylene [5] for the $P-V$ isotherm is 0.007% in average over the pressure range up to 2000 bar, while the difference for the $T-V$ isobar is 0.064% in average over the temperature range of 19.5–129 °C in semi-crystalline state of polyethylene.

We have selected the equations for the determination of β_T and α_P towards an accurate determination of γ_V. For example, Eq. (1) is used for the determination of β_T, while Eq. (2) is used for α_P. The α_P may be determined by Eq. (1) with less accuracy because m_x is not constant. The values of m_x and $P_x(T)$ used to determine β_T for rare gas solids are listed in Table 2, while those for semi-crystalline polymers are taken from literature [13].

It is somewhat unfortunate that Eqs. (3) and (4) for α_P and β_T cannot give an accurate function for $\gamma_V = \alpha_P/\beta_T$ because m_x varies on temperature slightly and irregularly and the function $P_x(T)$ with respect to temperatures is not smooth enough to obtain a simple function of temperature. We have tried to determine a simple empirical function of γ_V with respect to volume at constant pressure. The mathematical procedure for the determination of $\gamma_V = (\partial S/\partial V)_T$ from entropy S has been described elsewhere [14,15]. One of the useful functions of γ_V found by the mathematical procedure is given by

$$\gamma_V = A\{(V-V_0)/V\}^{\varepsilon}(V_0/V)^{1+c}\exp\{-c(V-V_0)/V\} \tag{5}$$

where V_0 is a volume at $T = 0$ K. We assumed in the calculation that V_0 is equal to V_1 at $T_1 = 1$ K in Eq. (4). In the mathematical procedure [14,15] we used an assumption that $\gamma_V(V)$ is a function of the variable $(V-V_0)/V$ and is expressed by its power law, which enables a transformation of $\gamma_V(V)$ to $\gamma_V(T)$ easily as is described below. The variable $(V-V_0)/V$ is a measure of free volume fraction in the solid state and plays an important role in the function of thermal pressure coefficient with respect to volume. It is found that Eq. (5) allows good prediction of $\gamma_V(V)$ at constant pressure. The constants A, ε and c in Eq. (5) determined by fitting the experimental data of γ_V are listed in Table 3 for rare gas solids and Table 4 for semi-crystalline polymers where values of volume V_0 at 0 K for various pressure are also listed. Typical data of $\gamma_V(V)$ for polypropylene are shown in Fig. 1 where the line is calculated by Eq. (5).

Table 1

Comparison between the experimental data (V_{exp}) of *PVT* for polyethylene [5] and values (V_{cal}) calculated by Eq. (1) with $m_x = 0.0909$, $P_x = 3470$ bar, $V(P_0 = 1\ \text{bar}, T = 329\ \text{K}) = 1.0341$ (cc/g) and Eq. (2) with $B = 6.741$, $V(P = 1200\ \text{bar}, T = 1\ \text{K}) = 0.9876$ (cc/g), $n_x = 0.0376$

Volume (cc/g) at various pressure at 55.8 °C				Volume (cc/g) at various temperature at 1200 bar			
P (bar)	V_{exp} (cc/g)	V_{cal} (cc/g)	$V_{exp}-V_{cal}$	Temp (°C)	V_{exp} (cc/g)	V_{cal} (cc/g)	$V_{exp}-V_{cal}$
1	1.0341	1.0341	–	19.5	0.9981	0.9981	–
100	1.0315	1.0315	–	32.3	1.0017	1.0004	0.0013
200	1.0289	1.0289	–	47.6	1.0048	1.0037	0.0011
300	1.0264	1.0264	–	55.8	1.0064	1.0057	0.0007
400	1.0240	1.0239	0.0010	66.1	1.0091	1.0085	0.0006
600	1.0192	1.0192	–	76.4	1.0117	1.0116	0.0001
800	1.0148	1.0148	–	84.8	1.0139	1.0144	−0.0005
1000	1.0105	1.0106	−0.0001	94.5	1.0174	1.0180	−0.0006
1200	1.0064	1.0066	−0.0002	104.5	1.0221	1.0220	0.0001
1400	1.0028	1.0028	–	115.5	1.0290	1.0269	0.0021
1600	0.9993	0.9991	0.0002	128.9	1.0337	1.0336	0.0001
1800	0.9957	0.9956	0.0001				
2000	0.9922	0.9922	–				

Table 2
The constants in Eq. (1) for rare gas solids determined at constant temperatures

	T (K)	$V_{0,T}$ (cc/g)	m_x	$P_x(T)$ (bar)
Ar [7]	0	0.5646	0.1513	4459
	4	0.5646	0.1559	4643
	20	0.5668	0.1548	4442
	40	0.5766	0.1520	3749
	60	0.5914	0.1456	2858
	77	0.6079	0.1421	2222
Kr [7]	0	0.3233	0.1474	5001
	4	0.3234	0.1510	5178
	20	0.3249	0.1463	4698
	40	0.3294	0.1450	4142
	60	0.3350	0.1454	3700
	77	0.3405	0.1414	3151
	90	0.3456	0.1395	2740
	100	0.3502	0.1392	2480
	110	0.3550	0.1394	2261
Xe [7]	0	0.2645	0.1420	5191
	4	0.2646	0.1464	5387
	20	0.2653	0.1470	5298
	40	0.2676	0.1454	4881
	60	0.2708	0.1451	4464
	77	0.2740	0.1405	3900
	80	0.2743	0.1411	3897
	100	0.2782	0.1417	3556
	120	0.2819	0.1426	3290
	140	0.2868	0.1390	2751
	159	0.2932	0.1311	2062

As far as we know there is no data available for low temperature range near 0 K for polypropylene, as is shown in Fig. 1. However, in the determination of the empirical function of $\gamma_V(V)$ we have used hypothetical point obtained from a thermodynamic condition that $\gamma_V(V_0) = 0$ at V_0.

3. Results

The determination of empirical function of $\gamma_V(T)$ has been carried out by using Eq. (5). The function of $\gamma_V(V)$ in Eq. (5) is converted into $\gamma_V(T)$ by using a relation $\ln(T/T_1) = B\{(V - V_0)/V\}^{n_x}$ in Eq. (2). The empirical function of $\gamma_V(T)$ is expressed by

$$\gamma_V = A\left(\frac{\ln\left(\frac{T}{T_1}\right)}{B}\right)^{\varepsilon/n_x}\left[1-\left(\frac{\ln\left(\frac{T}{T_1}\right)}{B}\right)^{1/n_x}\right]^{1+c} \times \exp\left[-c\left(\frac{\ln\left(\frac{T}{T_1}\right)}{B}\right)^{1/n_x}\right] \tag{6}$$

Eq. (6) allows the determination of $P_\mathrm{i} = \gamma_V T - P$ as a function of temperature at constant pressure. Therefore a temperature T^* at which $P_\mathrm{i} = 0$ can be determined at $P = P^*$. The typical experimental values of $P_\mathrm{i} = \gamma_V T - P$ for solid xenon are plotted against temperature at constant pressure in Fig. 2 where a good agreement between the experimental data and the lines calculated by Eq. (6) are obtained over wide range of temperatures. The value of V^* for $P_\mathrm{i} = 0$ is also determined by T^*, P^* and Eq. (2). All constants in Eq. (6) and values of P^*, V^* and T^* evaluated are listed in Tables 3 and 4.

We have found that a relation between P^*, V^* and T^* is expressed by an empirical equation $V^{*-1} = a_0 + b_0(P^*/T^*)$. From this equation we have derived an equation that

$$P^*V^*/\mathrm{R}T^* = C^* - D^*V^* \tag{7}$$

This is an empirical equation for PVT surface with zero internal pressure. Typical data of P^*, V^*, T^* for polyethylene in three dimensions are shown in Fig. 3. The constants C^* and D^* are listed in Table 5. The thermal pressure coefficient for the equation of state with $P_\mathrm{i} = 0$ is given using Eq. (7) by

$$\gamma_V^* = \frac{\mathrm{R}(C^* - D^*V^*)}{V^*} \tag{8}$$

Table 3
The constants in Eqs. (2) and (5) for rare gas solids determined at constant pressure and the values of P^*, V^* and T^* and its ratio $P^*V^*/\mathrm{R}T^*$ in Eq. (7)

Rare gas solid	P (bar)	A (bar/K)	c	ε	B	n_x	$V_1 = V_0$ (cc/g)	P^* (bar)	V^* (cc/mol)	T^* (K)	$P^*V^*/\mathrm{R}T^*$
Ar [7]	1	118.93	3.593	0.3784	6.169	0.1327	0.5646				
	500	130.63	4.455	0.3697	6.433	0.1376	0.5554	500	22.35	25.73	5.22
	1000	116.15	4.596	0.3480	6.647	0.1404	0.5474	1000	22.44	42.36	6.31
	1500	107.83	4.699	0.3353	6.823	0.1421	0.5402	1500	22.22	61.80	6.48
Kr [7]	1	155.65	5.008	0.4240	6.711	0.1473	0.3233				
	500	124.59	5.008	0.3703	6.943	0.1512	0.3188	500	26.91	26.99	5.99
	1000	110.04	5.077	0.3418	7.136	0.1535	0.3147	1000	26.81	44.52	7.22
	1500	97.23	5.021	0.3140	7.299	0.1550	0.3110	1500	26.79	64.76	7.46
	2000	88.37	4.960	0.2932	7.446	0.1560	0.3077	2000	26.85	90.44	7.14
Xe [7]	1	86.76	4.020	0.3400	7.328	0.1542	0.2645				
	500	65.13	4.518	0.2706	7.537	0.1570	0.2611	500	34.52	31.60	6.57
	1000	63.89	4.429	0.2673	7.614	0.1550	0.2580	1000	34.43	55.36	7.48
	1500	58.72	4.883	0.2465	7.892	0.1608	0.2552	1500	34.45	83.46	7.45
	2000	56.15	4.849	0.2369	8.008	0.1610	0.2526	2000	34.55	117.63	7.07
	2500	57.41	5.289	0.2381	8.164	0.1621	0.2501	2500	34.87	169.95	6.17
	3000	56.82	5.479	0.2348	8.285	0.1636	0.2479	3000			
	4000	56.72	5.857	0.2366	8.526	0.1652	0.2440				

Table 4
The constants in Eqs. (2) and (5) for semi-crystalline polymer determined at constant pressure and the values of P^*, V^* and T^* and its ratio P^*V^*/RT^* in Eq. (7)

Polymer solid	M_0 (g/mol)	P (bar)	A (bar/K)	c	ε	B	n_x	$V_1 = V_0$ (cc/g)	P^* (bar)	V^* (cc/g)	T^* (K)	P^*V^*/RT^*
PP [18]	42.08	1	224.3	7.209	0.7618	6.932	0.0405	1.0793				
		500	241.9	8.216	0.7586	7.027	0.0580	1.0591	500	1.0629	158.6	1.70
		1000	90.7	5.443	0.5238	7.095	0.0561	1.0530	1000	1.0580	191.8	2.79
		1500	178.9	7.313	0.6706	7.079	0.0508	1.0484	1500	1.0533	219.3	3.65
		2000	159.1	5.304	0.6329	6.932	0.0405	1.0456	2000	1.0490	244.1	4.35
PE [5]	28.05	1	87.6	3.187	0.4207	6.637	0.0383	1.0036				
		400	472.8	11.275	0.7706	6.715	0.0404	0.9965	400	0.9976	163.7	0.82
		800	283.0	8.188	0.6928	6.841	0.0453	0.9883	800	0.9908	184.9	1.45
		1200	101.5	2.597	0.4816	6.741	0.0376	0.9876	1200	0.9900	216.7	1.85
HMWPE [5]	28.05	1	251.0	4.296	0.7411	6.512	0.0379	1.0495	0			
		400	493.9	5.826	0.8880	6.553	0.0384	1.0398	400	1.0409	149.0	0.94
		800	255.9	3.677	0.7308	6.622	0.0409	1.0291	800	1.0329	193.6	1.44
		1200	141.9	1.908	0.5822	6.694	0.0436	1.0197	1200	1.0244	198.9	2.09
P4MP1 [6][a]	84.15	1	73.1	4.377	0.5851	7.068	0.0674	1.1570	0			
		200	42.1	3.951	0.4011	7.346	0.0841	1.1372	200	1.1388	68.0	3.32
		400	336.6	7.824	0.9347	7.698	0.1076	1.1130	400	1.1238	106.8	4.18
		600	123.2	6.369	0.6448	7.811	0.1105	1.1086	600	1.1197	109.0	6.12
NY66 [18]	226.31	1		5.359	1.0758	6.968	0.0546	0.8542				
		500	133.6	2.824	0.6556	7.025	0.0553	0.8448	500	0.8475	165.9	6.95
		1000	155.6	2.742	0.6661	7.019	0.0521	0.8398	1000	0.8445	211.8	10.85
		1500	71.3	0.747	0.4360	7.021	0.0502	0.8340	1500	0.8381	216.2	15.83
POM [19][a]	30.03	1	541.9	7.702	0.8190	6.801	0.0454	0.6929				
		400	315.1	6.206	0.6844	6.921	0.0508	0.6874	400	0.6882	134.5	0.73
		800	1.446×10^3	10.373	1.0335	6.965	0.0515	0.6841	800	0.6868	187.3	1.04
		1200	14.31×10^3	15.559	1.6319	7.261	0.0680	0.6742	1200	0.6812	204.4	1.41
		1600	10.71×10^3	16.437	1.4898	7.078	0.0540	0.6789	1600	0.6848	238.6	1.63
		2000	190.1×10^3	24.444	2.1673	7.115	0.0549	0.6763	2000	0.6848	267.8	1.81

M_0: M_w of monomer unit, PP: polypropylene, PE: polyethylene, HMWPE: high molecular weight polyethylene, P4MP1: poly-4-methyl-pentene-1, NY66: Nylon66, POM: polyoxymethylene.

[a] The units of pressure for P, A and P^* are kg/cm^2.

where γ_V^* is defined by $(\partial P^*/\partial T^*)_{V^*}$ It is obvious from Eqs. (7) and (8) that the internal pressure P_i^* is zero at every points on the $P^*V^*T^*$ surface.

4. Discussions

According to the third law of thermodynamics the entropy of the material approaches to a constant value S_0 in the limit of 0 K, which does not depend on volume and pressure. Therefore $(\partial S/\partial P)_T \to 0$ at $T \to 0$, which gives $(\partial V/\partial T)_P = V\alpha_P = 0$ due to the Maxwell relation and then $\alpha_P \to 0$ and $\gamma_V \to 0$ at $T \to 0$ [20]. This is a criterion in deriving the empirical function of γ_V in Eq. (5) where γ_V is zero at $V \to V_0$ through $T \to 0$. An existence of empirical equation of state for $P^*V^*T^*$ with $P_i = 0$ is confirmed by the two thermodynamic facts that $P_i = \gamma_V T - P < 0$ at $T \to 0$ under high

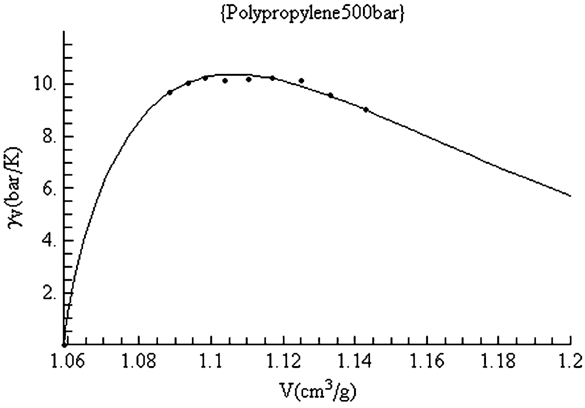

Fig. 1. γ_V vs. V plot for polypropylene at 500 bar. The experimental data are taken from Ref. [18].

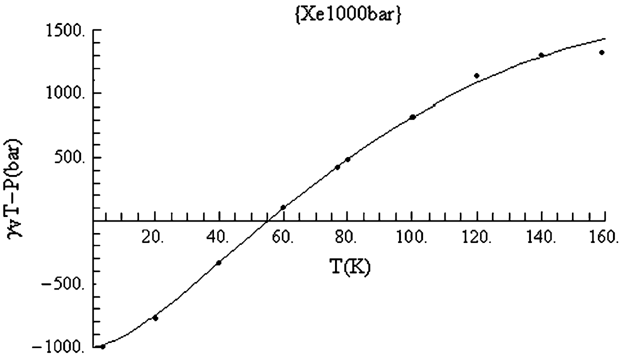

Fig. 2. $\gamma_V T - P$ vs. T plot for Xenon at 1000 bar. The experimental data are taken from Ref. [7].

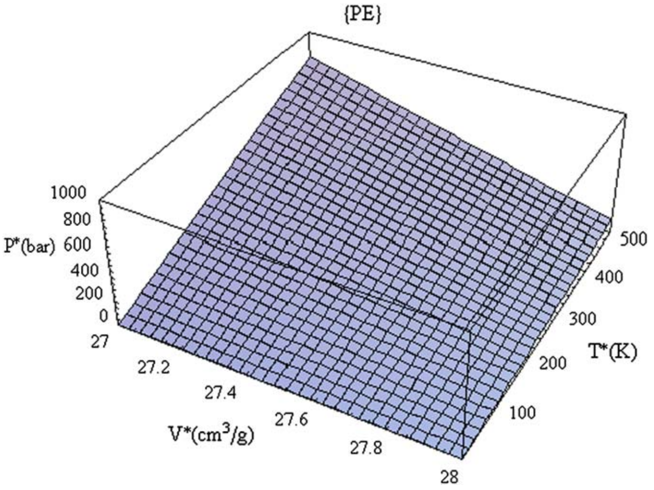

Fig. 3. $P^*V^*T^*$ surface for the equation of state with $P_i = 0$ in polyethylene calculated by Eq. (7) and values of C^* and D^* in Table 5.

pressure $P > 1$ since γ_V is zero at $T \to 0$ and the experimental result that $P_i > 0$ at higher temperature such as $T_1 > 0$. Therefore $P_i^* = \gamma_V^* T^* - P^* = 0$ exists between $0 < T^* < T_1$ (see Fig. 2).

It is interesting to discuss the practical meaning of the equation of state for zero internal pressure for solids. The three dimension $P^*V^*T^*$ plot is shown in Fig. 3 where the region above the $P^*V^*T^*$ surface correspond to high pressure region where $\gamma^*T^* = P^* < P$ or $P_i < 0$, while the region below the surface shows $P_i > 0$ and the surface shows $\gamma^*T^* = P^*$ or $P_i = 0$. The internal pressure P_i is related to the internal energy U by

$$P_i = (\partial U/\partial V)_T \tag{9}$$

No internal energy change occurs by changing the volume at constant temperature on the $P^*V^*T^*$ surface due to the cancellation between the external pressure P and thermal pressure $\gamma_V T$. The surface of $P^*V^*T^*$ with $(\partial U/\partial V)_T = 0$ may be meaningful in a processing such that a rare component is introduced into the inside of solid material without energy change.

Pastine [1,2] has derived the equation of state for polymer solid where a pressure consists of two factors that 0 K isotherm and thermal pressure contribution above 0 K and derived the equation of state for polyethylene of arbitrary crystallinity. In this work we have not discussed the effect of crystallinity on the equation of state. The thermal pressure coefficient evaluated in this work and constants obtained are limited for the semi-crystalline polymer from which the data were obtained.

It is interesting to discuss about the theoretical works of γ_V with respect to volume or temperature at constant pressure. The equation of state for polymer solids derived by Hartmann and Haque [16] is given in reduced form by $\overline{P}\,\overline{V} = \overline{T}^{3/2} - \ln \overline{V}$ which can predict a maximum of γ_V with respect to volume at constant pressure. Cho et al. [17] also predicted a maximum in the thermal pressure coefficient with respect to temperature in the semi-crystalline polymer. These results obtained by Hartmann and Haque and Cho et al. [17] are consistent with the experimental data of the γ_V vs. V curve with a maximum at constant pressure obtained in this work.

Table 5
The constant in the Eq. (7) for rare gas solids and semi-crystalline polymers

Solid	C^*	D^* (mol/cc)
Xe	109.3	2.960
Kr	308.4	11.230
Ar	240.6	10.540
POM	188.6	9.098
HMWPE	72.9	2.464
PE	122.4	4.344
PP	209.2	4.644
NY66	801.9	4.145

5. Conclusions

In this work an empirical equation for the γ_V vs. V curve with a maximum at constant pressure has been determined based on the PVT data available and empirical equations Eq. (1) for $V(P)$ and Eq. (2) for $V(T)$ in this work. An equation of state for zero internal pressure has been given by $P^*V^*/\mathrm{R}T^* = C^* - D^*V^*$ where the characteristic pressure P^*, volume V^* and temperature T^* are the values at which $\gamma_V T - P = 0$, respectively, and C^* and D^* are constant.

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