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Analysis of P-V-T Relationship & High Derivative Thermo elastic Properties of Al2O3

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ABSTRACT

Al2O3 is an important ceramic material and geophysical mineral. It has a high value of bulk modulus (Ko=252 GPa) and a lower value of K'0= 3.99. A compression of 40 percent in volume for this material requires a pressure in the rang 300GPa-350GPa. The pressure-volume-temperature relationship and variations of thermoelastic properties with compression and temperature have been studied in the present work using the formulations based on the Holzafel AP2 equation of state.

Keywords: Al2O3, Holzafel AP2 EOS, Bulk modulus, Pressure derivatives of bulk modulus, Grüneisen parameter

I. INTRODUCTION

Al_2O_3 perovskite, is an important geophysical mineral considered to be a major constituent of the Earth lower mantle[1,2]. Al_2O_3 is the fourth most prevalent component. The most prevalent mineral in the Earth's mantle, in the pyrolite composition under lower mantle conditions. Furthermore, the addition of Al_2O_3 can considerably alter the crystal chemistry of bridgmanite and, as a result, its physical properties, such as thermoelastic properties[3]. In the geochemical comportement of aluminium, it is unlikely that a large amount of such element could enter the core of the Earth, though at any depth of Earth's mantle and crust it constitutes a major constituent of many minerals [4]. It is stable for a wide range of pressures and temperatures [5–8]. Equations of state (EOS) describe the pressure-volume-temperature (P-V-T) relationship for a material at high pressures and high temperatures, including material properties such as bulk modulus (K) and its pressure derivatives and the Grüneisen parameters and $q=(d\ln\gamma/d\ln V)_T$. Physically meaningful EOS must satisfy boundary conditions at atmospheric pressure (P = 0) and also in the limit of infinite pressure, i.e. at extreme compression ($V\rightarrow 0$). We have fit Al_2O_3 perovskite in the Holzapfel AP2 EOS [9,10] based on the Thomas-Fermi model. EOS satisfy the infinite pressure conditions universally for investigated materials [11–15]. The results for P, K and K' for Al_2O_3 obtained from the Holzapfel AP2 EOS are used to establish a relationship between K' and P/K. The expressions for the volume dependences of and q obtained by Shanker and Singh [16] from

the free-volume theory [17] have been used in the present calculations section 2.

II. METHOD OF ANALYSIS

The Holzapfel AP2 EOS can be written as [10,18]

$$P = 3K_0 x^{-5}(1-x)[1+c_2 x(1-x)]\exp[c_0(1-x)] \quad (1)$$

where x = $(V/V_0)^{1/3}$, and

$$c_0 = -\ln\left(\frac{3K_0}{P_{FG0}}\right) \quad (2)$$

$$P_{FG0} = a_{FG}\left(\frac{Z}{V_0}\right)^{5/3} \quad (3)$$

$$\gamma = \frac{\alpha K_T V}{C_V} \quad (4)$$

with $a_{FG} = 0.02337$ GPa nm^5. Z is the total number of electrons in the volume V_0. In the case of Al_2O_3 we have Z=50 per molecule. This is to be multiplied by the Avogadro number when V_0 is given in the units of cm^3/mole. The constant c_2 in Eq.(4) [19] is related to K_0', the value of $K' = dK/dP$ at P = 0, as follows

$$c_2 = \frac{3}{2}(K_0' - 3) - c_0 \quad (5)$$

We use Eq.(1) for determining P-V relationships along different isotherms for Al_2O_3. The required input data on V_0, K_0 and K_0' at different temperatures have been taken from the literature[19,20,21,22] and given in Table 1. The volume ratio V/V_0 in the Table represents V(T,P)/V(T,0) along different isotherms at selected temperatures T. The amount of pressure required to produce the same change in V/V_0 decreases continuously with the increase in temperature. This is related to the fact that the bulk modulus becomes less, and the material more compressible at higher temperatures.

In addition to P-V isotherms we can also calculate the high derivative properties along different isotherms using values of input parameters appropriately corresponding to each temperature. This method has successfully been used by earlier workers[20,21,23]. The expressions for the bulk modulus K and its pressure derivatives $K' = dK/dP$ and $K'' = d^2K/dP^2$ are obtained using the following relationships

$$K = -V\left(\frac{dP}{dV}\right) = -\frac{x}{3}\left(\frac{dP}{dx}\right) \quad (6)$$

$$K' = -\frac{x}{3}\left(\frac{dK}{dx}\right) \quad (7)$$

$$KK'' = \frac{x^2}{9K}\left(\frac{d^2K}{dx^2}\right) - K'\left(K' + \frac{1}{3}\right) \quad (8)$$

where

$$\frac{dK}{dx} = -\frac{x}{3}\left(\frac{d^2P}{dx^2}\right) - \frac{1}{3}\left(\frac{dP}{dx}\right) \quad (9)$$

and

$$\frac{d^2K}{dx^2} = -\frac{x}{3}\left(\frac{d^3P}{dx^3}\right) - \frac{2}{3}\left(\frac{d^2P}{dx^2}\right) \quad (10)$$

The pressure P as a function of x is given by Eq.(1), the Holzapfel AP2 EOS. The high derivative thermoelastic properties include the Grüneisen parameter γ (Eq.4) and its volume derivative q. The modified free volume theory yields the following expression[16]

$$\gamma = \frac{K'}{2} - \frac{1}{6} - \varepsilon \quad (11)$$

where

$$\varepsilon = \frac{f(K - K'P)}{(3K - 2fP)} \quad (12)$$

The parameter f takes different values for different formulations of the Grüneisen parameter γ. Thus f=0 for Slater's formula[24], f=1 for the Dugdale-MacDonald formula[25], and f=2 for the Vashchenko-Zubarev formula[26]. Value of f for a given material can also be determined by taking $\gamma = \gamma_0$ at P=0. For Al_2O_3 $\gamma = \gamma_0 = 1.32$ [19], we have

$$\gamma_0 = \frac{K_0'}{2} - \frac{1}{6} - \frac{f}{3} \quad (13)$$

giving f=2.47 for $K_0' = 3.99$. Eq.(13) is obtained from Eqs.(11) and (12) at P=0, $\gamma = \gamma_0$, and $K' = K_0'$. The

second Grüneisen parameter q is obtained by differentiating Eq.(11)

$$q = \frac{V}{\gamma}\left(\frac{d\gamma}{dV}\right)_T = \frac{1}{\gamma}\left[\frac{-KK''}{2} + K\frac{d\varepsilon}{dP}\right] \quad (14)$$

where

$$K\frac{d\varepsilon}{dP} = -\frac{\left[fKK''P + \varepsilon K(3K' - 2f)\right]}{(3K - 2fP)} \quad (15)$$

We have used equations (26-30) for determining γ and q at different compressions and temperatures.

III. RESULTS AND DISCUSSIONS

The results for P, K, K' and KK'' obtained with the help of Eqs. (2, 3 and 5-10) for Al_2O_3 along different isotherms are given in Tables 2-6. There is good agreement between the results obtained in the present study and the pressure, volume, bulk modulus data for Al_2O_3 reported in the literature[19,20,27,28]. For studying high derivative properties, we have found that the results given in Tables 2-5 for P, K and K' plotted in Figure 2 as $1/K'$ versus P/K satisfy the following relationship[29]

$$\frac{1}{K'} = A + B\left(\frac{P}{K}\right) + C\left(\frac{P}{K}\right)^2 \quad (16)$$

where A= $1/K'_0$, B= $-K_0K_0''/K_0'^2$, and C= $(K_\infty'/K_0'^2)[K_0K_0'' + K_0'(K_0' - K_\infty')]$. The parameters A, B, and C are found to be temperature-dependent (Table 6). The validity of Eq.(16) has been discussed recently by Shanker et al [29,30] and Kushwah and Bharadwaj[31]. The second pressure derivative of bulk modulus is obtained from Eq.(16) as follows

$$KK'' = -K'^2\left(B + 2C\frac{P}{K}\right)\left(1 - \frac{P}{K}K'\right) \quad (17)$$

Values of A, B and C given in Table 6 when substituted in Eqs.(16) and (17) yield good agreement with the values of K' and KK'' given in Tables 4 and 5 determined in the present study using the Holzapfel AP2 EOS. Thus equations (16) and (17) are compatible with the results based on the Holzapfel AP2 EOS.

The Grüneisen parameter γ and its volume derivative q along different isotherms have been determined with the help of equations (11) and (14) using the values of P, K, K' and KK'' given in Tables 2-6. First we have determined ε and K(d ε /dP) using equations (12) and (15) at different compressions and temperatures. The results are given in Tables 7 and 8. Variations in the values of γ as well as q are quite significant (Figures 2 and 3). γ and q both increase with the increase in temperature, and decrease with the increase in pressure. The results for γ and q obtained in the present study are based on the free volume formulation derived from the fundamental relationship between thermal pressure and thermal energy[3] using the pressure derivatives of bulk modulus determined from the Holzapfel AP2 EOS. This EOS has been found compatible with the results based on the ab initio molecular dynamics[10,18]. Values of as well as q (Figures 2 and 3) increase substantially with the increase in temperature. This finding is consistent with the critical analysis of equations of state for Al_2O_3 presented by Jeanloz et al [32]. He predicted that the strongest temperature dependence is characteristic of at constant pressure, and this is arising from the intrinsic anharmoncity of the solid.

Along different isotherms γ decreases with the increasing pressure, i.e. decreasing volume. Earlier workers have used the following relationship[19]

$$\frac{\gamma}{\gamma_0} = \left(\frac{V}{V_0}\right)^q \quad (18)$$

Equation (18) is valid only when q is assumed to remain constant. However, it has been found that q does not remain constant, and changes significantly along different isotherms with the change in pressure or volume (Figure 4). It may be more appropriate to write [33]

$$\frac{q}{q_0} = \left(\frac{V}{V_0}\right)^\lambda \quad (19)$$

where is the Jeanloz parameter and known as third order Grüneisen parameters. To examine the validity of Eq.(19), the plots between $\ln(q/q_0)$ and $\ln(V/V_0)$ are presented in Figure 5. The nature of plots reveals that in Eq. (19) does not remain constant with the increase in compression. The slope of the plots in Figure 5 decreases with the increase in pressure along different isotherms leading to the conclusion that decreases with the increase in pressure. The simple relationships such as equations (18) and (19) are therefore inadequate for representing the volume dependences of and q. A model for the volume dependence of consistent with the generalized free-volume theory has been developed by Burakovsky and Preston[34]

$$\gamma = \gamma_\infty + a_1\left(\frac{V}{V_0}\right)^n + a_2\left(\frac{V}{V_0}\right)^{1/3} \qquad (20)$$

where γ_∞ is the value of at extreme compression $V\rightarrow 0$. Burakovsky and Preston[18] have taken $\gamma_\infty = 1/2$, based on the Thomas-Fermi theory. a_1, a_2 and n are positive constants for a given material. n is taken greater than 1 in equation (20) for [34,35]. Thus the two volume-dependent terms in equation (20) represent concave up and concave down variations respectively. Equation (20) on differentiation with respect to V yields [36]

$$q = \frac{V}{\gamma}\left(\frac{d\gamma}{dV}\right)_T = \frac{1}{\gamma}\left[a_1 n\left(\frac{V}{V_0}\right)^n + \frac{a_2}{3}\left(\frac{V}{V_0}\right)^{1/3}\right] \qquad (21)$$

and

$$\lambda = \frac{V}{q}\left(\frac{dq}{dV}\right)_T = \frac{\left[a_1 n^2\left(\frac{V}{V_0}\right)^n + \frac{a_2}{9}\left(\frac{V}{V_0}\right)^{1/3}\right]}{\left[a_1 n\left(\frac{V}{V_0}\right)^n + \frac{a_2}{3}\left(\frac{V}{V_0}\right)^{1/3}\right]} - q \qquad (22)$$

It is thus evident from equations (21) and (22) that q and both depend on volume. Equations (18) and (19), which are based on the assumptions that q and are constants, are not adequate. The results obtained in the present study (Figures 2-4) support this finding.

It should also be mentioned that in the limit of extreme compression ($V\rightarrow 0$, $P\rightarrow\infty$), becomes $_\infty$ (a finite positive value), but $q\rightarrow q_\infty\rightarrow 0$ (Eq.21). The third order Grűneisen parameter tends to $_\infty$ as $V\rightarrow 0$ (Eq.22), and becomes $_\infty = 1/3$. The study of variation of with pressure is very important for understanding the higher derivatives of thermodynamic properties of solids.

IV. CONCLUSIONS

The thermoelastic properties of Al_2O_3 were investigated using the Holzapfel AP2 EOS over a wide range of pressures and temperatures. The EOS produced results consistent with the results of ab-initio molecular dynamics [18,37]. Using the EOS results in the free-volume theory, the thermoelastic behaviour has been explored in terms of the Grüneisen parameter and its volume derivative q, the second Grüneisen parameter. The Grüneisen parameter and its volume derivative q are found to alter dramatically with changes in volume owing to pressure and temperature differences. Because Al_2O_3 is an important high-temperature-high-pressure calibrator for determining experimental data [38,39], the results are useful.

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Table-1: Values of input parameters for Al_2O_3 used in Holzapfel AP2 EOS [19,20,21,22,37,38]

T (K)	V_o=V(T,0) (cm^3/mol)	K_o (GPa)	K'$_o$	P_{FG0} (GPa)	C_0	C_2
300	25.606	252	3.99	3070	1.401	0.084
500	25.709	247	4.05	3049	1.415	0.160
1000	26.038	231	4.21	2985	1.460	0.355
1500	26.415	214	4.37	2915	1.513	0.542
2000	26.818	196	4.56	2842	1.575	0.765
2500	27.307	178	4.76	2758	1.642	0.998
3000	27.798	160	4.95	2677	1.719	1.206

Table-2: Pressure-volume relationships for Al_2O_3 along different isotherms at selected temperatures T.

	300K		500K		1000K		1500K		2000K		2500K		3000K	
V(T,P)/V(T,0)	V(T,P) (cm^3/mol)	P (GPa)	V(T,P) (cm^3/mol)	P (GPa)	V(T,P) (cm^3/mol)	P (GPa)	V(T,P) (cm^3/mol)	P (GPa)	V(T,P) (cm^3/mol)	P (GPa)	V(T,P) (cm^3/mol)	P (GPa)	V(T,P) (cm^3/mol)	P(GPa)
1	25.606	0	25.709	0	26.038	0.0	26.415	0	26.818	0	27.307	0	27.798	0
0.95	24.326	14.3	24.424	14.1	24.736	13.2	25.094	12.3	25.477	11.3	25.942	10.3	26.408	9.31
0.90	23.045	32.7	23.138	32.2	23.434	30.3	23.774	28.3	24.136	26.2	24.576	24.0	25.018	21.8
0.85	21.765	56.5	21.853	55.6	22.132	52.7	22.453	49.4	22.795	45.9	23.211	42.3	23.628	38.5
0.80	20.485	87.4	20.567	86.2	20.830	81.9	21.132	77.1	21.454	71.9	21.846	66.6	22.238	61.0
0.75	19.205	128	19.282	126	19.529	120	19.811	114	20.114	107	20.480	99.2	20.849	91.3
0.70	17.924	181	17.996	179	18.227	172	18.491	163	18.773	153	19.115	143	19.459	132
0.65	16.644	252	16.711	250	16.925	241	17.170	229	17.432	217	17.750	204	18.069	189
0.60	15.364	349	15.425	346	15.623	334	15.849	320	16.091	304	16.384	287	16.679	267

Table-3: Values of bulk modulus K(GPa) for Al_2O_3 at different compressions and temperatures

V(T,P)	Bulk modulus K(GPa)						
V(T,0)	300K	500K	1000K	1500K	2000K	2500K	3000K
1	252	247	231	214	196	178	160
0.95	308	302	285	266	246	225	204
0.90	375	370	351	330	307	284	259
0.85	459	453	433	409	383	357	328
0.80	563	557	535	508	479	449	415
0.75	693	687	663	633	601	566	527
0.70	859	853	827	794	757	717	670
0.65	1072	1067	1039	1001	960	913	858
0.60	1352	1348	1317	1275	1258	1174	1108

Table-4: Values of pressure derivative of bulk modulus (K') for Al_2O_3at different compressions and temperatures

V(T,P)	Pressure derivative of bulk modulus K'						
V(T,0)	300K	500K	1000K	1500K	2000K	2500K	3000K
1	3.99	4.05	4.21	4.37	4.56	4.76	4.95
0.95	3.78	3.83	3.97	4.10	4.26	4.42	4.57
0.90	3.60	3.64	3.76	3.87	4.00	4.14	4.26
0.85	3.44	3.48	3.57	3.67	3.78	3.90	4.00
0.80	3.30	3.33	3.41	3.50	3.59	3.69	3.78
0.75	3.17	3.20	3.27	3.34	3.42	3.51	3.59
0.70	3.05	3.08	3.14	3.20	3.27	3.35	3.41
0.65	2.95	2.97	3.02	3.08	3.14	3.20	3.26
0.60	2.85	2.87	2.91	2.96	3.01	3.07	3.12

Table-5: Values of second pressure derivatives of bulk modulus for Al_2O_3 at different compressions and temperatures

V(T,P)	Second pressure derivatives of bulk modulus KK"						
V(T,0)	300K	500K	1000K	1500K	2000K	2500K	3000K
1	-4.51	-4.71	-5.29	-5.89	-6.66	-7.51	-8.36
0.95	-3.70	-3.86	-4.28	-4.72	-5.27	-5.87	-6.46
0.90	-3.07	-3.19	-3.51	-3.84	-4.24	-4.68	-5.09
0.85	-2.57	-2.66	-2.91	-3.16	-3.46	-3.78	-4.09
0.80	-2.16	-2.23	-2.43	-2.63	-2.86	-3.10	-3.32
0.75	-1.83	-1.89	-2.04	-2.20	-2.38	-2.56	-2.73
0.70	-1.56	-1.60	-1.73	-1.85	-1.99	-2.14	-2.27
0.65	-1.33	-1.37	-1.47	-1.57	-1.68	-1.79	-1.90
0.60	-1.14	-1.17	-1.25	-1.33	-1.42	-1.51	-1.60

Table-6: Values of the parameters for Al_2O_3 in the reciprocal K-primed equation

T (K)	A	B	C
300K	0.251	0.283	0.499
500K	0.247	0.287	0.502
1000K	0.238	0.298	0.509
1500K	0.229	0.308	0.517
2000K	0.219	0.320	0.524
2500K	0.210	0.332	0.530
3000K	0.202	0.341	0.537

Table-7: Values of the parameter ε for Al_2O_3 in the free volume theory

V(T,P)	ε						
V(T,0)	300K	500K	1000K	1500K	2000K	2500K	3000K
1	0.508	0.508	0.508	0.508	0.508	0.508	0.508
0.95	0.439	0.439	0.435	0.432	0.429	0.425	0.422
0.90	0.383	0.381	0.376	0.372	0.367	0.361	0.357
0.85	0.335	0.333	0.328	0.323	0.317	0.311	0.306
0.80	0.295	0.293	0.287	0.282	0.277	0.271	0.266
0.75	0.260	0.258	0.253	0.249	0.243	0.238	0.234
0.70	0.231	0.229	0.225	0.22	0.216	0.211	0.207
0.65	0.205	0.204	0.199	0.196	0.192	0.189	0.185
0.60	0.183	0.182	0.179	0.176	0.173	0.169	0.167

Table-8: Values of K(dε/dP) for Al_2O_3 in the free volume theory

V(T,P)	Kdε/dP						
V(T,0)	300K	500K	1000K	1500K	2000K	2500K	3000K
1	-1.51	-1.54	-1.62	-1.71	-1.80	-1.90	-2.00
0.95	-1.18	-1.20	-1.24	-1.28	-1.33	-1.37	-1.42
0.90	-0.94	-0.94	-0.96	-0.98	-1.00	-1.01	-1.03
0.85	-0.74	-0.75	-0.75	-0.76	-0.76	-0.76	-0.76
0.80	-0.60	-0.59	-0.59	-0.59	-0.58	-0.58	-0.57
0.75	-0.48	-0.48	-0.47	-0.46	-0.45	-0.44	-0.44
0.70	-0.39	-0.38	-0.37	-0.36	-0.36	-0.35	-0.34
0.65	-0.31	-0.31	-0.30	-0.29	-0.28	-0.27	-0.26
0.60	-0.25	-0.25	-0.24	-0.23	-0.22	-0.21	-0.21

Figure 1: Plots between V and P for Al2O3 along different isotherms

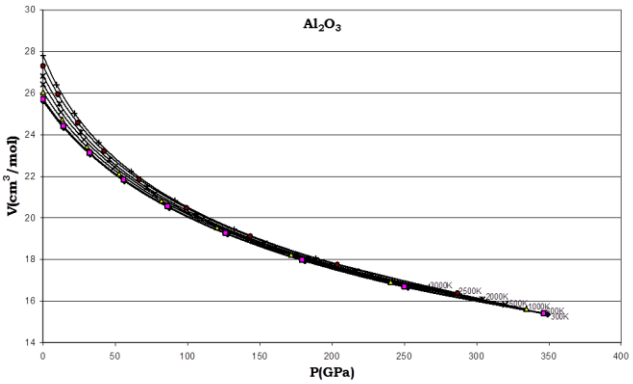

Figure 2: Plots between 1/ and P/K for Al2O3 along different isotherms

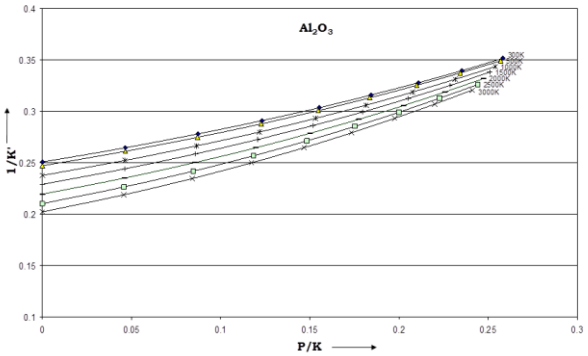

Figure 3: Volume dependence of for Al2O3 along different isotherms

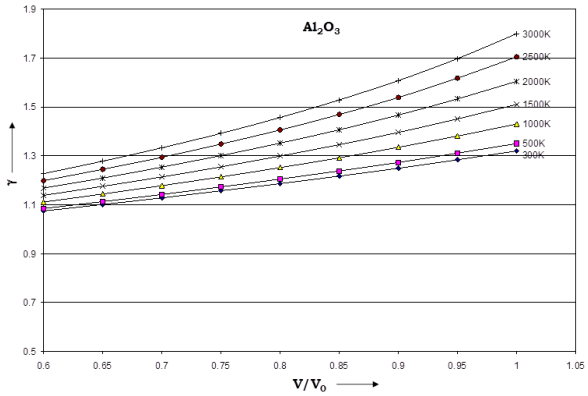

Figure 4: Volume dependence of q for Al2O3 along different isotherms

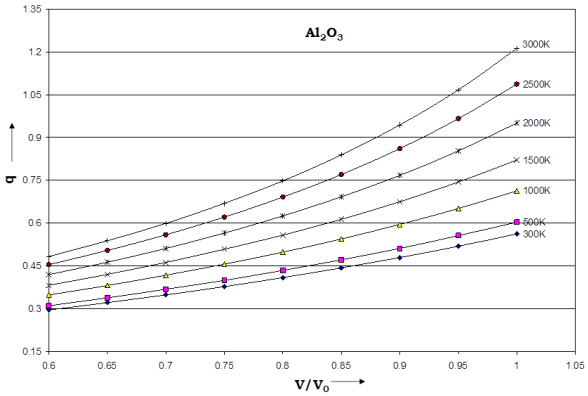

Figure 5: Plots between ln(q/q0) and ln(V/V0) for Al2O3 along different isotherms