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Equations of state of Plagioclase Feldspars

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Abstract The volume variation with pressure of seven intermediate plagioclase feldspars has been determined by high-pressure single-crystal X-ray diffraction. The bulk moduli of plagioclases for a 3rd-order Birch-Murnaghan EoS can be described by the following pair of equations:

$$K_{T0} = 54.1(3) + 0.39(1)X_{An}X_{An}<50$$

$$K_{T0} = 59.5(3.1) + 0.23(4)X_{An}X_{An} > 50$$

with $K'_0 = 5.8$ for plagioclase with $X_{An} < 20$ and $K'_0 = 3.2$ for $X_{An} > 35$. These parameters can also be used in a Murnaghan EoS to describe the volume variation of plagioclase feldspars up to pressures of 3 GPa. For a Murnaghan EoS with $K'_0 = 4$, the values of the bulk moduli can be described by a single equation, $K_{T0} = 57.7(6) + 0.24(1)X_{An}$, with a small loss in the accuracy of the predicted volumes up to pressures of 3 GPa.

Introduction

Plagioclase are among the most common minerals in crustal rocks, yet an examination of both the published literature and the thermodynamic data gathered in databases reveals that there is little or no recent information available on either their room-pressure bulk moduli or their equations of state (EoS). Experimental measurements of the compressibility of plagioclases do, however, go back at least to Adams and Williamson (1923). These and subsequent determinations (Adams and Gibson 1929; Bridgman 1948; Yoder Jr. and Weir 1951; Vaidya et al. 1973) were carried out by a variety of press techniques, wherein the change in length of a cylinder of the material of interest was measured as a function of pressure. These experiments were fraught with difficulties arising from the small changes in the length of the specimen under pressure and the need to maintain hydrostatic conditions, to remove all void space from the sample and to correct for the elasticity and the friction in the apparatus. Nonetheless, many of these measurements are sufficiently accurate to reveal that feldspars are significantly stiffer than other framework silicates such as quartz, but softer than olivines and pyroxenes, and that the bulk moduli of plagioclase feldspars increase by about 40% from albite to anorthite. These general results were confirmed by the measurement of the individual components of the elastic constant tensors of five plagioclase samples by Rhyzova (1964). However, these ultrasonic measurements demanded large single-crystal samples and, as a result, several of the measurements were made on twinned specimens while the data were reduced on the assumption of monoclinic rather than the true triclinic symmetry. It is not clear what influence these assumptions might have on the values of the resulting bulk moduli.

Even if these previous experimental data were accurate, they only provide values for the bulk modulus (K_{T0}) at room pressure while both $> K_{T0} = -V_0(\partial P/\partial V)_{P=0}$ and its pressure derivative $K'_0 = (\partial K_T/\partial P)_{P=0}$ are required to describe the volume evolution of phases with pressure. Therefore, the full determination of the EoS of crystals requires either precise volume data collected at high pressure by X-ray diffraction methods, or direct measurement of the elastic tensor at high pressures. The triclinic symmetry of feldspars places the latter beyond current experimental capabilities, at least for routine measurements. Angel

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et al. (1988) reported some single-crystal high-pressure diffraction data for end-member feldspars that again confirmed that anorthite was significantly stiffer than albite. However, the precision of the data collected by Angel et al. (1988) was insufficient for both K_{T0} and K'_0 to be determined. In the intervening years the experimental techniques for high-pressure diffraction have improved significantly (Angel et al. 2000) and in this paper new P-V data of higher precision are reported for seven different intermediate plagioclase feldspars. As for all other sub-solidus properties of feldspars, these data reveal that the high-pressure behavior of plagioclase feldspars is complex. A full description of this behavior is not, however, the purpose of this contribution. Instead the aim here is to derive parameters for the EoS of plagioclase feldspars that are sufficiently reliable for use in thermodynamic calculations at modest temperatures and pressures.

Samples and experiments

Single crystals of natural anorthite-rich plagioclases were selected from mineral separates prepared for previous studies by Dr. M.A. Carpenter of the University of Cambridge. Samples designated "Hawk b", "Lake Co", "101377a" and "87975a" are described in full by Carpenter et al. (1985). All of these samples were heat-treated at 800 °C for ~12 hours in the preparation for the previous calorimetric measurements by Carpenter et al. (1985). Two further crystals were selected from crushed portions of rocks of specimens 67791 and 67783 of the Harker collection, University of Cambridge. All of these plagioclase samples have low orthoclase contents (a maximum of 3 mol% in Hawk-b) and were chosen because they are probably representative of the highest state of Al/Si tetrahedral order possible at each composition. Full single-crystal structure analyses of samples 87975a, 101377a and Lake Co were reported by Angel et al. (1990). Sample101377a/1 was heat-treated at 1300 °C to induce partial Al/Si disorder (Carpenter et al. 1985), which was then determined by subsequent single-crystal structure determination (Angel et al. 1990). The compositions of the individual crystals used in the high-pressure measurements were not determined, but the compositional range of each sample as determined by electron microprobe analyses is reported in Table 1.

Each crystal was loaded in turn into a BGI diamond-anvil pressure cell with a 4:1 mixture of methanol and ethanol as the hydrostatic pressure medium and a second crystal of quartz or fluorite for use as an internal pressure standard. Unit-cell parameters of each crystal at each pressure were obtained by vector least-squares fit (Ralph and Finger 1982) to the diffractometer setting angles of between 20 and 30 strong reflections determined by the SINGLE software (Angel et al. 2000) on a Huber four-circle diffractometer (Angel et al. 1997). Pressures were determined from the measured unit-cell volumes of the standard crystals via their known equations of state (Angel 1993; Angel et al. 1997). Pressures and unit-cell parameters are listed in Table 2.

These data were supplemented by the dataset for low albite published by Downs et al. (1994). The parameters of the equations of state for each sample were determined by a least-squares fit of the pressure-volume data with v5.2 of the EoSFit software (Angel 2000). Weights derived from the experimental uncertainties in both pressure and volume were assigned to each data point in every fit. Birch-Murnaghan EoS (Birch 1947) based upon the Eulerian definition of finite strain were used to fit the data and to provide the basis for discussion, but the parameters for the Murnaghan EoS (Murnaghan 1937) are provided at the end for those thermodynamic databases that use this formulism.

Table 1 Structural state and compositional data for plagioclase used in this study

	Sample	*Source*	*Structural state*	*Compositional range*	*Mean composition*
An20	Hawk b	Harvard University mineral collection 97608	$C\bar{1}$ with some weak and diffuse *e* reflections	$An_{20-21}Ab_{76-77}Or_3$	$An_{20}Ab_{77}Or_3$
An37[a]	67791	Harker Collection, University of Cambridge	$C\bar{1}$ with *e* reflections	$An_{35-58}Ab_{60-62}Or_{2-3}$	$An_{37}Ab_{61}Or_2$
An46[a]	67783	Harker Collection, University of Cambridge	$C\bar{1}$ with *e* and *f* reflections.	[b]$An_{45-47}Ab_{51-53}Or_2$	$An_{46}Ab_{52}Or_2$
An68	Lake Co.	USNM collection no. 115900	$I\bar{1}$ *b* reflections slightly elongated	$An_{66-70}Ab_{29-33}Or_{0-1}$	$An_{68}Ab_{31}Or_1$
An78o	101377a	Harker Collection, University of Cambridge	$I\bar{1}$ sharp *b* reflections, diffuse *c* reflections	$An_{76\text{-}78}Ab_{22-24}Or_0$	$An_{78}Ab_{22}Or_0$
An78d	101377a/1	101377a heat-treated by Carpenter et al (1985)	$I\bar{1}$ sharp, weak *b* reflections, very diffuse *c* reflections	$An_{76-78}Ab_{22-24}Or_0$	$An_{78}Ab_{22}Or_0$
An89	87975a	Harker Collection, University of Cambridge	$I\bar{1}$ sharp *b* reflections	$An_{88-90}Ab_{10-12}Or_0$	$An_{89}Ab_{11}Or_0$

Data from Carpenter et al. (1985) except [a]Carpenter (personal communication) and [b]O'Brien (personal communication).

Table 2 Pressures and unit-cell parameters of the sample crystals

P GPa	V Å^3	esd(P) GPa	esd(V) Å^3
An20			
0.000	667.893	0.000	0.076
0.832	659.186	0.004	0.047
1.364	653.925	0.005	0.061
1.939	648.564	0.005	0.053
2.843	640.524	0.005	0.039
3.488	635.014	0.005	0.053
4.737	624.891	0.006	0.037
5.510	618.875	0.007	0.050
6.448	611.789	0.007	0.059
7.104	606.932	0.007	0.036
7.746	602.068	0.008	0.053
8.234	598.699	0.007	0.049
An38			
0.000	667.893	0.000	0.076
0.832	659.186	0.004	0.047
1.364	653.925	0.005	0.061
1.939	648.564	0.005	0.053
2.843	640.524	0.005	0.039
3.488	635.014	0.005	0.053
4.737	624.891	0.006	0.037
5.510	618.875	0.007	0.050
6.448	611.789	0.007	0.059
7.104	606.932	0.007	0.036
7.746	602.068	0.008	0.053
8.234	598.699	0.007	0.049
An46			
0.000	668.463	0.000	0.044
0.708	662.078	0.005	0.082
1.297	656.915	0.006	0.054
2.014	650.774	0.006	0.049
2.808	644.589	0.005	0.109
3.424	639.483	0.007	0.059
4.059	634.523	0.007	0.115
4.638	630.195	0.005	0.089
5.276	625.467	0.009	0.069
6.290	618.240	0.010	0.060
7.359	611.071	0.012	0.104
An68			
0	1339.017	0.000	0.064
1.240	1317.696	0.009	0.072
2.111	1303.213	0.035	0.124
2.905	1291.452	0.011	0.074
3.430	1283.456	0.009	0.076
3.897	1276.494	0.009	0.076
4.425	1269.012	0.013	0.054
4.727	1264.816	0.013	0.088
5.277	1257.312	0.013	0.058
5.630	1252.383	0.012	0.119
An78 ordered			
0	1339.213	0	0.062
2.149	1303.680	0.008	0.068
2.592	1297.165	0.009	0.052
3.214	1287.831	0.009	0.056
3.429	1284.867	0.008	0.080
4.062	1275.736	0.010	0.055
4.489	1269.459	0.012	0.105
4.929	1263.471	0.012	0.116
An78 disordered			
0	1339.968	0.000	0.082
0.939	1324.349	0.006	0.100
1.620	1313.109	0.011	0.093
2.149	1304.477	0.008	0.127
2.592	1297.913	0.009	0.087
3.214	1288.772	0.009	0.085
3.429	1285.685	0.008	0.119
4.062	1276.700	0.010	0.124
4.489	1270.600	0.012	0.098
4.929	1264.776	0.012	0.086
5.748	1253.350	0.009	0.099
An89			
0	1339.014	0.000	0.098
0.473	1331.229	0.005	0.057
1.071	1321.727	0.006	0.054
1.637	1312.883	0.005	0.064
2.254	1303.502	0.006	0.062
2.785	1295.805	0.006	0.065
3.411	1286.811	0.009	0.054
3.904	1279.488	0.007	0.077
4.104	1276.493	0.007	0.065
4.560	1270.227	0.008	0.051
4.971	1264.357	0.009	0.067
5.238	1260.803	0.007	0.048
5.649	1255.450	0.009	0.085
6.594	1242.858	0.010	0.075
7.133	1235.746	0.009	0.080

Results

The volumes of all of the samples studied evolve smoothly with increasing pressure (Fig. 1). However, this apparently simple picture actually masks much more complex and subtle trends that are more apparent if the volume-pressure data is transformed into an *f-F* or *normalised stress—finite strain* plot for the purposes of diagnostics (e.g. Angel 2000). For a Birch-Murnaghan EoS the finite strain is $f_E = \left[(V_0/V)^{2/3} - 1\right]/2$, a *normalised stress* is defined as $F_E = P/3f_E(1+2f_E)^{5/2}$, and the EoS can be re-written as a polynomial in the strain (e.g. Stacey et al. 1981):

$$F_E = K_0 + \frac{3K_0}{2}\left(K'_0 - 4\right)f_E + \frac{3K_0}{2}\left(K_0K'' + (K'-4)(K'-3) + \frac{35}{9}\right)f_E^2 + \ldots \quad (1)$$

If the *P-V* data are transformed into f_E and F_E and plotted with f_E as the abscissa, a direct indication of the compressional behavior is obtained. If the data points all lie on a horizontal line of constant F_E then $K' = 4$, and the data can be fitted with a 2nd-order truncation of the Birch-Murnaghan EoS. If the data lie on an inclined straight line, the slope is equal to $3K_0(K'-4)/2$ and the data will be adequately described by a 3rd-order truncation of the EoS. If the value of K'' differs significantly from the value implied by the 3rd-order truncation, then the coefficient of f^2 in Eq. 1 is not zero and the data fall on a parabolic curve in the *F-f* plot. In all cases, the intercept on the *F* axis is the value of K_{T0}.

The *f-F* plot (Fig. 2) of the plagioclase data clearly indicates that the pressure derivatives of the bulk moduli differ significantly across the plagioclase join. For albite-

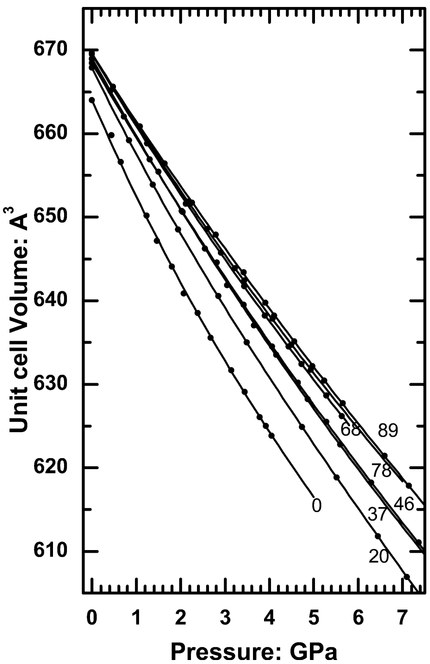

Fig. 1 Variation of the volume of plagioclase feldspars with pressure. *Symbols* are measured data as listed in Table 2, *numbers* are the sample compositions in X_{An}. The *lines* are fitted Birch-Murnaghan EoS with the parameters listed in Table 3. Data for the An78d sample are omitted, as they are indistinguishable on this scale from the data for An78o. Data for plagioclases with $X_{An} > 50$ have been plotted as one-half of the true $I\bar{1}$ unit-cell volume so as to appear on the same plot. Data for low albite are taken from Downs et al. (1994)

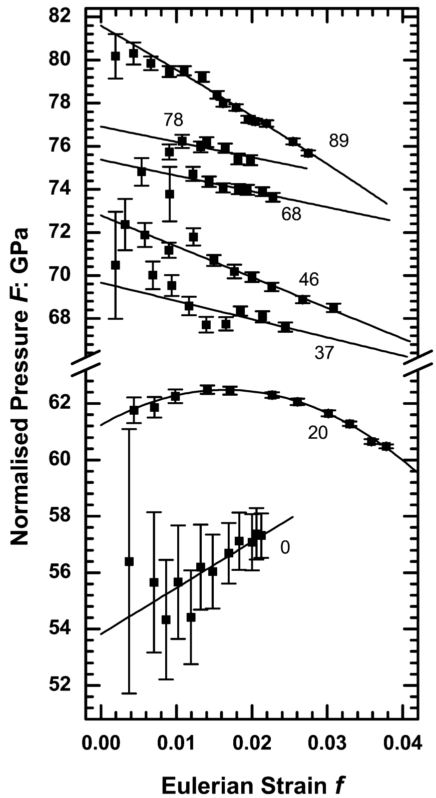

Fig. 2 The volume-pressure data of the plagioclase feldspars displayed as a plot of the normalized pressure F against the Eulerian strain f. *Numbers* are the sample compositions in X_{An}. Data for the An78d sample are omitted, as they are indistinguishable on this scale from the data for An78o. Data for low albite are taken from Downs et al. (1994). The *lines* are fitted Birch-Murnaghan EoS with the parameters listed in Table 3

rich samples (albite itself and An20) the positive slopes indicate that the room-pressure values of K'_0 are significantly greater than 4. By contrast, all of the samples with An contents of 38% or higher exhibit negative slopes in the f-F plot and hence values of K'_0 less than 4, mostly around a value of 3.2. In this sense, the An20 sample appears to be intermediate in its behavior, in that the low-pressure value of K'_0 is certainly greater than 4. However the plot shows that K''_0 is negative, so that K'_0 decreases with increasing pressure until it is 4 at around 3.5 GPa and less than 4 at higher pressures. The data for An20 can therefore only be fit satisfactorily by a 4th-order EoS whereas the remainder are fit with a 3rd-order EoS. Lastly, the f-F plot of the data for the An89 sample shows an abrupt change in slope at a pressure of 3.6 GPa as a result of the smeared transition from $P\bar{1}$ symmetry to $I\bar{1}$ symmetry (Angel et al. 1989). Parameters for the Birch-Murnaghan EoS of all the samples are reported in Table 3.

Leaving aside these other complications, the room pressure bulk moduli of the plagioclase feldspars increase with increasing anorthite content (Fig. 3) in agreement with the ultrasonic data of Rhyzova (1964). However, the values of K_S derived from the ultrasonic data are between 2 and 5 GPa lower than the values obtained from the new P-V datasets. This may be a result of the use of twinned crystals and the assumption of monoclinic symmetry in the ultrasonic measurements (Rhyzova 1964). The new data suggest that, in addition, there appears to be a break in the dependence of the bulk modulus with composition around An50 that coincides with the phase transition from the $C\bar{1}$ structures of the albite-rich compositions to the $I\bar{1}$ structures

Table 3 Parameters for Birch-Murnaghan EoS

Comp	N_{data}	P_{max} *GPa*	V_0 *Å³*	K_0 *GPa*	K'	K'' GPa^{-1}	χ^2_w	ΔP_{max} *GPa*
An0	13*	4.05	664.04(9)	53.8(9)	6.0(6)	[-0.186]	0.48	0.054
An20	12	8.23	667.88(7)	61.2(5)	5.6(4)	-0.94(14)	0.49	0.017
An37	10	5.59	669.01(7)	69.7(7)	3.2(3)	[-.054]	2.21	0.037
An46	11	7.36	668.46(4)	72.8(4)	2.7(2)	[-.059]	0.82	0.030
An68	10	5.63	1339.02(7)	75.4(5)	3.4(2)	[-.049]	0.29	0.026
An78o	8	4.93	1339.21(7)	76.9(6)	3.4(3)	[-.047]	1.29	0.015
An78d	11	5.75	1339.98(10)	77.5(5)	3.2(2)	[-.048]	1.47	0.023
An89	7	3.41	1339.02(9)	80.4(7)	3.2(4)	[-.046]	0.18	0.004

Note: Numbers in parentheses represent esd's in the last digit. Numbers in square brackets are the implied values of the parameters. ΔP_{max} is the largest absolute misfit to a P-V data point.*Data from Downs et al. (1994). The data point at 0.4 GPa was omitted because it clearly lies off the trend defined by all of the other data points.

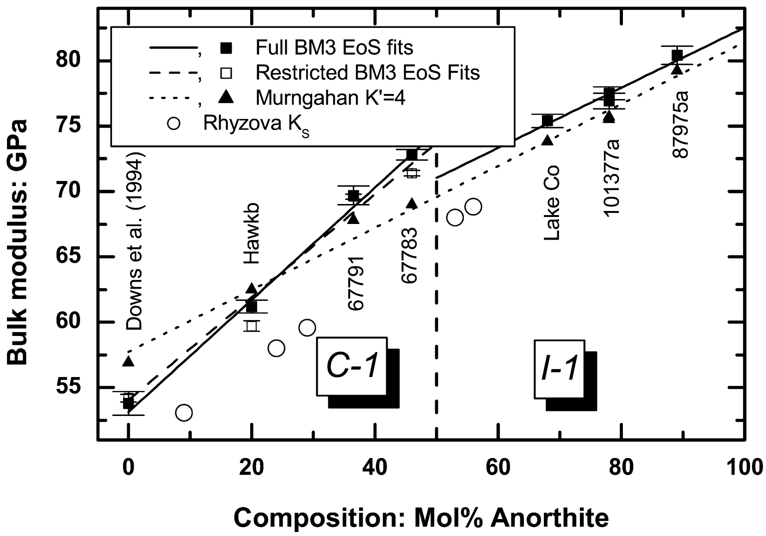

Fig. 3 The variation of the bulk moduli of plagioclase feldspars with composition. The *filled squares* with *error bars* are the isothermal bulk moduli of the fitted Birch-Murnaghan EoS with the parameters listed in Table 3, *open squares* are from the constrained Birch-Murnaghan EoS (Table 4) and the *triangles* are the bulk moduli for the Murnaghan EoS with $K_0' = 4$ (Table 5). The values of K_{S0} from Rhyzova (1964) are plotted as open *circles*

of the anorthite-rich plagioclases. This transition is also accompanied by a change in the behavior of the lattice parameters of the ordered (so-called "low") plagioclases with composition (Bambauer et al. 1967) and reflects the change in the pattern of ordering of the Al and Si within the tetrahedral framework.

There is no reason to suppose that the bulk modulus should vary linearly with composition, even over part of the plagioclase join. In fact, the demonstrated existence of the "plateau" effect in alkali feldspars (Hayward and Salje 1996; Hayward et al. 1998) would suggest that the relationship should be non-linear, especially near the end-members and near phase transitions. Nonetheless, the bulk moduli can be approximated by two linear segments (Fig. 3) with equations:

$$K_{T0} = 53.1(7) + 0.43(2)X_{An}X_{An}<50$$

$$K_{T0} = 59.5(3.1) + 0.23(4)X_{An}X_{An} > 50$$

Extrapolation of the second segment to An100 would suggest that the bulk modulus of $I\bar{1}$ anorthite would be around 82.5 GPa, well within the range of 78–86 GPa reported for $P\bar{1}$ anorthites by Hackwell and Angel (1992).

The other important observation is that the bulk moduli of the natural and disordered samples of An78 composition are indistinguishable (Table 3). So, for petrologically relevant ranges of Al/Si disorder it appears that ordering has no significant effect on the bulk elastic properties of $I\bar{1}$ plagioclases. Since disordering in plagioclase is accompanied by a small increase in the room-pressure unit-cell volume (Bambauer et al. 1967; Carpenter et al. 1985) this means that the disordered material will remain less dense than the ordered material at all pressures. Therefore, the driving force for ordering will increase as the $P\Delta V$ term increases with increasing pressure. The same conclusion was drawn from a structural study of albite by Downs et al. (1994).

Discussion

For the purposes of the incorporation of these data into thermodynamic databases aimed at predicting the thermodynamic properties of minerals at pressures and temperatures of relevance to rocks, some reasonable simplifications can be made. First, we note that most of the anomalous behavior in the EoS of plagioclases that has been described above occurs at pressures in excess of 3 GPa, well beyond the pressure range at which plagioclase is thermodynamically stable. There are two caveats to this. Presumably, for compositions between An20 and An37 there may be complex EoS behavior at lower pressures. The boundary of the $P\bar{1}$ to $I\bar{1}$ phase transition in anorthite-rich feldspars has also yet to be mapped out in *P*-*T*-*X* space although the work of Angel et al. (1989) and Hackwell and Angel (1995) suggest that it is restricted to very low temperatures and is therefore not a factor in determining the elastic properties of anorthites even at the lowest metamorphic grades. Secondly, the values of K_0' obtained from the data fall into two groups, around 5.8 for the albite-rich samples, and 3.2 for the remainder. Parameters for 3rd-order Birch-Murnaghan EoS were therefore obtained by re-fitting the data with these constraints and are reported in Table 4. The bulk moduli of the feldspars with $X_{An} > 50$ do not differ significantly from the values obtained in the unconstrained fits reported in Table 3 and the trend with composition is indistinguishable from that reported above. By contrast, the constrained fits result in considerably more scatter in the values of the bulk moduli of the albite-rich plagioclases (Fig. 3), with the result that it is not clear whether the trend of these data with $K_{T0} = 54.1(3) + 0.39(1)X_{An}$ represents a significant difference from that reported above.

Up to a pressure of 3 GPa the plagioclase structures undergo about 5% compression. This is sufficiently small for the Murnaghan EoS to provide an adequate

Table 4 Parameters for 3rd-order Birch-Murnaghan EoS with K'_0 fixed

Comp	N_{data}	P_{max}	V_0	K_0	K'	χ^2_w	ΔP_{max}
		GPa	*Å³*	*GPa*			*GPa*
An0	13	4.05	664.03(9)	54.2(3)	5.8	0.45	0.058
An20	6	3.49	668.04(10)	59.7(4)	5.8	3.48	0.019
An37	10	5.59	669.02(6)	69.6(2)	3.2	1.93	0.036
An46	11	7.36	668.55(7)	71.4(2)	3.2	3.2	0.048
An68	10	5.63	1339.00(7)	75.7(2)	3.2	0.33	0.031
An78o	8	4.93	1339.20(7)	77.3(2)	3.2	1.17	0.018
An78d	11	5.75	1339.98(9)	77.5(2)	3.2	1.32	0.024
An89	7	3.41	1339.02(7)	80.5(2)	3.2	0.15	0.005

Table 5 Parameters for Murnaghan EoS with $K'_0 = 4$

Comp	V_0 *Å³*	K_0 *GPa*	χ^2_w	ΔP_{max} *GPa*
An0	663.93(10)	56.9(3)	1.46	0.087
An20	667.83(5)	62.5(2)	0.59	0.007
An37	669.08(9)	67.8(3)	4.4	0.047
An46	668.7(2)	69.0(5)	19.7	0.127
An68	1339.08(9)	73.8(2)	2.16	0.036
An77o	1339.2(1)	75.7(2)	2.56	0.034
An77d	1340.2(2)	75.5(3)	4.6	0.053
An89	1339.11(7)	79.2(2)	0.90	0.011

Data ranges same as for Table 4.

representation of the *P-V* relations in this pressure range, with the same values of bulk modulus and K'_0 as reported in Table 4. However, the deviation of the volumes predicted by the Murnaghan form from that predicted by the Birch-Murnaghan formulism at higher pressures means that the Murnaghan formulism *must not* be used to predict the volumes of plagioclase feldspars at pressures above 3 GPa. Similarly, care must be taken in extrapolating the Birch-Murnaghan EoS to pressures in excess of the reported data as there is no reason to suppose that further complexity in behavior, beyond that already observed in the current datasets, does not occur at higher pressures.

Several thermodynamic databases incorporate the Murnaghan EoS with K'_0 set to 4 for all phases (e.g. Holland and Powell 1998). Therefore Table 5 lists values of bulk moduli obtained by fitting the P-V data with $K'_0 = 4$. The increased values of both the misfits (ΔP_{max}) and the χ^2_w over the fits listed in Tables 3 and 4 clearly indicate that the restriction of K'_0 to 4 is not consistent with the *P-V* data and therefore results in a bias in the values of K_{T0}. For albite-rich plagioclase whose true value of K'_0 is ~5.8, the value of K_{T0} is increased when K'_0 is reduced to 4, while the opposite is true for the remainder of the plagioclase compositions with true values of $K'_0 \sim 3.2$This shift in values does have the advantage that the bulk moduli can be described by a single equation:$K_{T0} = 57.7(6) + 0.24(1)X_{An}$ In the worst case, the use of these moduli with $K'_0 = 4$ results in about a 0.1% error in the volume predicted for intermediate plagioclases at pressures of 3 GPa. The discrepancy increases at higher pressures but, as noted above, this is not of practical concern.

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