#121

Phys Chem Minerals (1988) 15:313–318

Papers from the GEOPHYSICAL LABORATORY
Carnegie Institution of Washington NO. 2060

PHYSICS AND CHEMISTRY OF MINERALS

Comparative Compressibility of End-Member Feldspars

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Abstract. The compressibilities of the three end-member feldspars have been determined between 1 bar and 50 kbar by single crystal X-ray diffraction techniques, using a Merrill-Bassett type diamond anvil cell with three crystals loaded simultaneously. Low albite (ordered aluminium-silicon distribution) and high sanidine (disordered Al–Si) show similar behaviour on compression, with bulk moduli (linear fit to volume-pressure data) of 0.70 and 0.67 Mbar respectively. The most compressible cell axis of all three feldspars studied is *a*, indicating that the major change in the feldspar framework with pressure is a shortening of the overall length of the "crankshaft chains" by reduction of T–O–T angles.

Anorthite shows anomalous behaviour in that we have observed a previously unreported reversible phase transition at a pressure between 25.5 and 29.5 kbar. This transition is marked by large discontinuities in the unit cell angles and a small decrease of 0.2 percent in the cell volume with increasing pressure. The high-pressure phase is less compressible than the low-pressure phase, the bulk moduli being 0.94 and 1.06 Mbar respectively. There was no evidence of a monoclinic to triclinic inversion in sanidine that was expected to occur between 20 and 30 kbar on the basis of previous work on intermediate alkali feldspars.

Introduction

Feldspar minerals are a major component of igneous and metamorphic rocks, and comprise approximately 60 percent of the Earth's crust. Of the three principal rock-forming end-members, anorthite and albite are restricted to the crust and uppermost mantle (pressures less than 20 kbar), whereas sanidine is stable to at least 40 kbar and may be an important host for potassium in the mantle. A comprehensive understanding of the response of the feldspar crystal structure to temperature and pressure is therefore critical to an understanding of the geochemistry of igneous and metamorphic processes of the crust and upper mantle, as well as to an understanding of the geophysical properties of the crust.

In recent years a more thorough understanding of the complex behaviour of feldspars at elevated temperatures and low or atmospheric pressures has been developed through the integration of crystallographic, calorimetric, and spectroscopic techniques. However, apart from experimental phase equilibria studies there has been little work done on feldspars at elevated pressures. Compressibility studies of low albite (Hazen and Prewitt 1977) and of intermediate alkali feldspars (Hazen 1976) have demonstrated that the effects of pressure upon the feldspar structure are, in general, the opposite to those induced by an increase in temperature. For example, Hazen (1976) found that the monoclinic to triclinic displacive transition associated with the collapse, with decreasing temperature, of the tetrahedral framework around the Na site in albite could be induced in orthoclase-rich feldspars by an increase in pressure.

In this study we have measured the compressibility of the three end-member feldspars by single crystal X-ray diffraction techniques. The results presented here provide new compressibility data for the thermodynamic characterisation of feldspars at elevated pressures, as well as a basis for further high pressure structural studies of the feldspar minerals.

Experimental

The feldspar specimens used in this study were chosen because they represent essentially end-member compositions, and have previously been well-characterised by a variety of techniques. The albite is from Amelia Court House, Virginia, and has a composition of $Ab_{98}Or_1An_1$ (Carpenter et al. 1985). Neutron and X-ray diffraction studies by Harlow and Brown (1980) and Smith et al. (1986), solution calorimetric measurements by Carpenter et al. (1985), and NMR studies by Kirkpatrick et al. (1985) indicate that Amelia albite has an almost completely ordered aluminium/silicon distribution in a triclinic $C\bar{1}$ structure. The anorthite is from the Val Pasmeda locality in Austria. Crystals are reported as having variable composition between 99.5 percent and 100% anorthite component (Adlhart et al. 1980, Redfern and Salje 1987). A single crystal structure determination was carried out on a crystal from this specimen by Wainwright and Starkey (1971). More recently, samples from this locality have been studied by calorimetry (Carpenter et al. 1985) and Si^{29} NMR spectroscopy (Kirkpatrick et al. 1987), while lattice constants have been determined at elevated temperature (Redfern and Salje 1987). These all indicate that Val Pasmeda anorthite is completely ordered at room temperature and pressure with a $c = 14$Å $P\bar{1}$ cell. The K-feldspar used in the study was an $Or_{98}Ab_2$ high sanidine found in a grospydite nodule from a kimberlite pipe (Smyth and Hatton 1977). Its cell parameters and a recent single-crystal structure determination (Scambos et al. 1987) indicate that it is highly disordered with $t_1 = 0.266$ and $t_2 = 0.234$ on the basis of mean T–O bond lengths.

Table 1. Cell parameters of feldspars at pressure

P (Kbar)	a: (Å)	b: (Å)	c: (Å)	alpha: °	beta: °	gamma: °	Volume: (Å^3)
Albite							
0.0	8.141 (1)	12.783 (1)	7.161 (1)	94.23 (1)	116.61 (1)	87.69 (1)	664.39 (12)
6.5	8.100 (1)	12.761 (1)	7.146 (1)	94.27 (1)	116.72 (1)	87.71 (1)	657.94 (9)
14.0	8.049 (2)	12.727 (1)	7.129 (2)	94.24 (1)	116.91 (1)	87.78 (1)	649.44 (19)
19.0	8.025 (1)	12.718 (2)	7.117 (1)	94.23 (1)	116.87 (1)	87.83 (1)	646.21 (15)
25.5	7.968 (1)	12.684 (1)	7.100 (1)	94.17 (2)	116.98 (1)	87.93 (1)	637.75 (14)
29.5	7.952 (2)	12.676 (2)	7.095 (2)	94.23 (2)	116.97 (2)	87.87 (2)	635.60 (19)
35.5	7.922 (2)	12.663 (3)	7.081 (1)	94.18 (3)	117.08 (1)	87.99 (2)	630.79 (21)
41.5	7.864 (1)	12.627 (2)	7.058 (1)	94.19 (2)	117.09 (2)	87.99 (2)	622.31 (16)
49.5	7.833 (2)	12.614 (2)	7.049 (2)	94.09 (1)	117.07 (1)	88.08 (1)	618.57 (21)
Anorthite							
0.0	8.171 (3)	12.870 (2)	7.085 (2)	93.12 (2)	115.89 (2)	91.28 (2)	668.38 (28)
6.5	8.152 (1)	12.849 (1)	7.072 (1)	93.03 (2)	115.87 (1)	91.39 (1)	664.68 (13)
14.0	8.120 (2)	12.815 (2)	7.048 (2)	92.88 (2)	115.82 (2)	91.56 (2)	658.28 (33)
19.0	8.111 (2)	12.805 (2)	7.037 (6)	92.87 (3)	115.80 (3)	91.61 (2)	656.25 (58)
25.5	8.080 (3)	12.770 (1)	7.015 (2)	92.70 (2)	115.74 (2)	91.81 (2)	650.16 (25)
29.5	8.051 (2)	12.755 (2)	6.992 (2)	92.27 (3)	115.27 (2)	92.58 (2)	647.30 (23)
35.5	8.029 (2)	12.733 (1)	6.971 (2)	92.14 (2)	115.23 (2)	92.71 (1)	642.66 (22)
41.5	8.004 (3)	12.711 (2)	6.957 (2)	92.02 (2)	115.20 (1)	92.81 (1)	638.42 (32)
49.5	7.987 (2)	12.694 (2)	6.941 (2)	91.97 (2)	115.16 (2)	92.91 (2)	634.90 (25)
Sanidine							
0.0	8.604 (2)	13.032 (1)	7.180 (3)	90.00	115.98 (2)	90.00	723.66 (42)
6.5	8.557 (1)	13.017 (1)	7.169 (2)	90.00	116.11 (2)	90.00	717.10 (31)
14.0	8.489 (1)	12.993 (1)	7.141 (1)	90.00	116.19 (1)	90.00	706.80 (22)
19.0	8.470 (1)	12.985 (1)	7.135 (1)	90.00	116.22 (1)	90.00	704.01 (18)
25.5	8.405 (1)	12.967 (1)	7.109 (2)	90.00	116.30 (1)	90.00	694.59 (25)
29.5	8.387 (3)	12.963 (3)	7.107 (7)	90.00	116.35 (5)	90.00	691.81 (94)
35.5	8.337 (1)	12.948 (1)	7.081 (1)	90.00	116.38 (1)	90.00	684.80 (19)
41.5	8.278 (1)	12.934 (1)	7.059 (2)	90.00	116.42 (2)	90.00	676.78 (28)
49.5	8.246 (2)	12.927 (2)	7.042 (3)	90.00	116.47 (2)	90.00	672.00 (41)

Notes: Estimated standard deviations given in parentheses. The c cell parameter of the average structure of anorthite is reported; the true cell has an axis of $2c$

It therefore has monoclinic symmetry, and space group $C2/m$.

Suitable twin-free crystals of all three samples were selected on the basis of omega and psi scans of single crystals mounted in a conventional manner on single-crystal X-ray diffractometers. Even the best high sanidine crystals had noticeably broader diffraction peak profiles than those from albite or anorthite. This may be due to the high state of Al–Si disorder which would give rise to strains on a unit-cell scale. A TEM examination of sanidines from this sample showed the complete absence of any evidence of even the earliest stages of ordering, consistent with rapid cooling from high temperature (Smyth and Hatton 1977).

All three crystals used in the high pressure study were loaded together in a Merrill-Bassett diamond-anvil cell of the type described by Hazen and Finger (1982). The precautions involved in orienting three crystals of similar lattice constants within one diamond cell have recently been described by McCormick et al. (1988). The pressure medium used was a 4:1 methanol: ethanol mixture which is hydrostatic to over 100 kbar (Piermarini et al. 1973). Pressure calibration, which was performed before and after every set of cell determinations, was by the standard laser-induced ruby fluorescence technique, using the R_1 line from small ruby chips included in the cell. The diamond cell was mounted on an automated Picker four-circle diffractometer equiped with a Mo tube ($\lambda_{\bar{\alpha}}=0.7107$ Å). Cell parameters were determined from the centering of between 10 and 13 reflections from each crystal ($9.5°<2\theta<16.5°$) at eight positions on the diffractometer. This technique is designed to eliminate any errors in peak positions arising from zero or crystal centering errors (Hamilton 1974, King and Finger 1979), and results in more reliable cell parameters. The cell parameters of sanidine were obtained by a constrained fit to the centering data, after it was determined that the unconstrained values of alpha and gamma unit cell angles did not deviate significantly from 90°.

Unit cell parameters of all three crystals were determined at eight pressures from room pressure to 49.5 kbar, and are reported in Table 1. The cells were also redetermined at nominal room pressure within the diamond cell after the high pressure experiments. The results were identical, within the estimated errors, to those measured at the start of the experiment, and are therefore not reported in Table 1. The room pressure unit cell parameters are also in good agreement with those reported for samples from the same localities by Harlow and Brown (1980) (albite), Carpenter et al. (1985) (albite and anorthite), and by Scambos et al. (1987) for sanidine.

Discussion

Although the concept of structurally analagous variables (Hazen 1977) is of limited applicability to feldspars (Brown

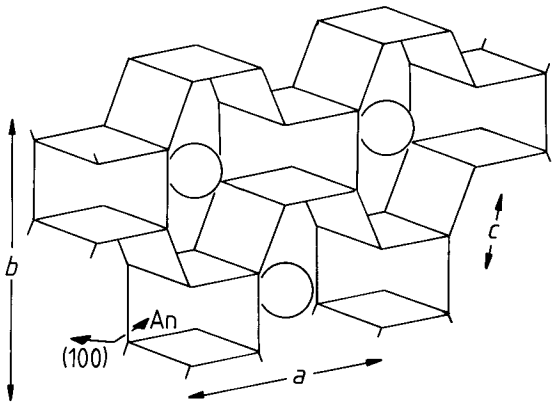

Fig. 1. A diagrammatic representation of the feldspar framework after Megaw (1974). The (100) plane normal, and the direction of maximum compressibility in anorthite (An) are indicated

et al. 1984), the type of structural changes that occur with changes in pressure are expected to be similar to those observed with changes in temperature. Megaw (1974) demonstrated that the major change in the feldspar structure in response to temperature is the deformation of the tetrahedral framework mainly by changes in the T–O–T bond angles, as confirmed by numerous structural studies (Foit and Peacor 1973, Ohashi and Finger 1974, 1975, Prewitt et al. 1976, Winter et al. 1977, 1979, Smith et al. 1986, Swanson 1986). The internal dimensions of the tetrahedra themselves change very little with temperature. This deformation of the framework is coupled with the behaviour of the large cation site of the structure (Figure 1). When this cation is relatively large (Ba^{2+} in celsian, or K^+ in sanidine) the framework around the cation site is "held" open and the structure is metrically monoclinic. The same effect is achieved when a feldspar with a smaller cation (e.g. Na^+ in albite) is heated, and a displacive transition occurs from a metrically triclinic to a monoclinic framework. This simple view of the feldspar structure is complicated by the potential for aluminium-silicon ordering within the tetrahedral sites of the framework. Ordering not only reduces the topochemical symmetry, but the strains induced in the framework by this ordering have been shown to couple with the strains associated with framework collapse around the cation sites (Salje 1985, Salje et al. 1985, Salje 1987, Redfern & Salje 1987). This coupling changes the temperature (and presumably the pressure) at which structural collapse of the framework around the cation sites takes place. With these considerations in mind, we will now discuss the results of the present high pressure study.

Comparative Compressibilities

The variation of the unit cell parameters of all three feldspars with pressure are shown in Figure 2. The most striking feature of the results is the large discontinuity in all three cell angles of anorthite between 25.5 and 29.5 kbar. This is indicative of a previously unknown phase transition in end-member $P\bar{1}$ anorthite which will be discussed below. Apart from the discontinuities associated with this phase transition the cell parameters and cell volumes of all three feldspars vary linearly with pressure over the pressure range studied, within the uncertainties associated with the determination of pressure.

The patterns of linear compressibilities of the three axes (Table 2) of sanidine and albite are very similar, with the *a* axis the most compressible, the *c* axis intermediate, and the *b* axis the least compressible. Calculations with the

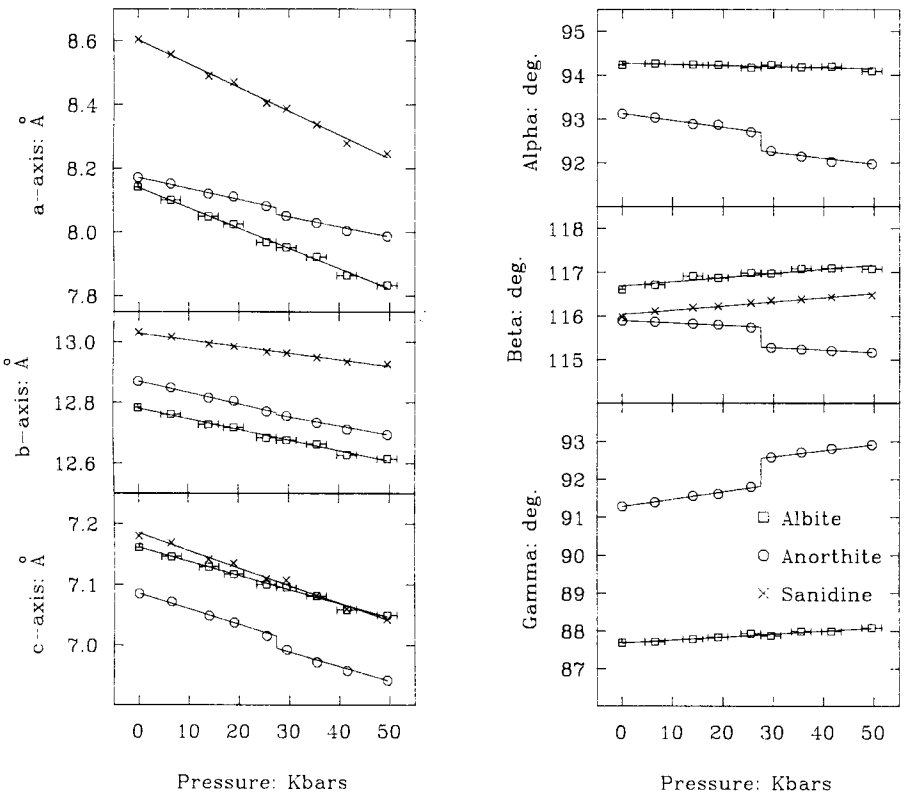

Fig. 2. The variation of feldspar unit-cell parameters with pressure. The *c* cell parameter graphed for anorthite is one half of the true *c* repeat. Error bars on albite data points represent pressure uncertainties of +/− 2 kbar. Esd's of cell parameters are less than the size of the symbols

Table 2. Linear Compressibilities ($Mbar^{-1}$)

	Albite	Sanidine	Anorthite	
			0–25 kbar	30–50 kbar
a	0.78	0.87	0.43	0.40
b	0.28	0.17	0.30	0.24
c	0.33	0.40	0.39	0.36
V	1.43	1.49	1.06	0.94

Table 3. Major axes of strain ellipsoids

	Unit strain	Angle with		
		+A	+B	+C
Albite 0–49.5 kbar	$-0.85\ Mbar^{-1}$	24	83	94
	−0.32	110	110	17
	−0.25	102	21	74
Albite 26–488 C	$21.2\ K^{-1}$	42	67	81
(Stewart & von Limbach	6.6	126	43	61
1967)	−0.4	109	124	30
Sanidine 0–49.5 kbar	$-0.92\ Mbar^{-1}$	23	90	93
	−0.37	113	90	3
	−0.16	90	0	90
Sanidine 20–500 C	$16.9\ K^{-1}$	20	90	96
(Henderson 1979)	−0.7	110	90	6
	−1.8	90	0	90
Anorthite 0–25.5 kbar	$-0.58\ Mbar^{-1}$	140	125	53
	−0.33	68	96	48
	−0.17	121	35	64
Anorthite 25–200 C	$22.4\ K^{-1}$	39	106	78
(Redfern and Salje 1987)	4.8	110	53	40
	−0.4	58	41	127

Note: unit thermal strains have been multiplied by 10^6

STRAIN program of Ohashi (In Hazen and Finger 1982, Appendix II) show that the direction of maximum compression of both of these structures is close to the (100) plane normal (Figure 1 and Table 3). The compression along this direction accounts for around 65 percent of the volume compressibility of these two structures. The thermal expansion of both albite (Stewart and von Limbach 1967) and high sanidine (Henderson 1979) is also highly anisotropic with the linear expansion of the major axis in each accounting for between 75 percent and 85 percent of the bulk thermal expansivities. This suggests that the short M–OA2 bond, which is almost parallel to this major axis, plays a passive role in the structure. The obverse argument (Henderson 1979) would lead to the expectation of this direction being the *least*, not the *most*, compressible. The metrical changes observed with increasing pressure therefore probably reflect structural adjustments driven by a change in the T–O–T bond angles.

However, while sanidine and albite behave almost identically, anorthite is far more isotropic in compression, with the most compressible direction rotated some 55° from the (100) plane normal (Figure 1). Anorthite is also stiffer than either albite or sanidine (Figure 3), suggesting that the large cation site plays a major role in controlling the overall amount of compression allowed in a given structure. As

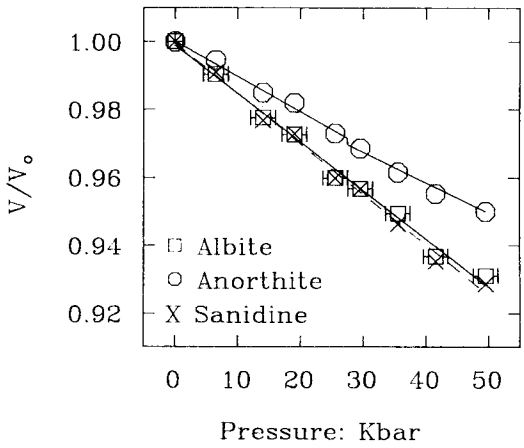

Fig. 3. Feldspar unit-cell volume variation with pressure, normalised to room pressure volume (V_o)

albite and sanidine have singly charged cations, and anorthite a doubly charged cation, this may be understood as a coulombic effect. Hazen and Finger (1982) showed that in such cases the bulk modulus of the structure increases in proportion to the cation charge as observed here.

Anorthite

Whereas the thermal expansion and compressibility behaviour of feldspars arise from continuous changes in the geometry of the tetrahedral framework, displacive phase transitions result from discontinous changes in framework geometry around the large cation sites. Although the exact temperature or pressure at which these transitions take place can be modified by the state of Al–Si order within the framework, such transitions are primarily associated with changes in the nature of the occupancy of the large cation site.

The volume change associated with the newly discovered phase transition observed between 25.5 and 29.5 kbar in anorthite is estimated to be 1.3 $Å^3$ on the basis of a $c = 7Å$ cell, a relative change of 0.2 percent. The spontaneous strain associated with the transition from the low pressure phase to the high pressure phase is constant (within experimental uncertainties) between 29.5 and 41.5 kbar. The diagonalised strain tensor components, or principal strains, are $+55 \times 10^{-4}$, $+25 \times 10^{-4}$, and -90×10^{-4}. This is in contrast to the $I\bar{1}$–$P\bar{1}$ transition at 240° C (Redfern and Salje 1987), in which the $P\bar{1}$ cell of anorthite is in a state of plane strain with respect to the $I\bar{1}$ cell.

As *b*, *c*, and *d* type reflections are still present in the diffraction pattern of the structure at pressures above the transition, it is quite clear that the transition does not result in an increase in symmetry. An examination of the available structural data on anorthite (Wainwright and Starkey 1971) and anorthite-rich plagioclase (Kempster et al. 1963, Berking 1976, Smyth 1986) also provides a possible explanation for the phase transition. At room temperature the $P\bar{1}$ structure has four distinct Ca sites. Two of them, Ca(ooo) and Ca(zio), have significantly larger temperature factors than the others, indicative of residual disorder. Wenk and Kroll (1984) and Smyth (1986) successfully refined these as split sites. The phase transition observed at 27.5 kbar may therefore be driven by the ordering of Ca on these two sites, which would be expected to reduce the symmetry of the

structure from $P\bar{1}$ to $P1$. A single crystal structural study in a diamond cell is currently in progress to investigate the structural changes associated with this transition.

Sanidine

In contrast, no phase transition was detected in sanidine. Previous work by Hazen (1976) on intermediate alkali feldspars had demonstrated that framework collapse resulting in a reduction in symmetry from $C2/m$ to $C\bar{1}$ could be induced by increasing pressure. Extrapolation of the measured inversion pressures of Or_{67} (12 kbar), Or_{82} (18 Kbar) and that inferred for Or_{35} (0 Kbar; Hovis 1980, Kroll and Ribbe 1983) would suggest that the sanidine used in this study (Or_{98}) should have inverted to a triclinic phase between 20 and 30 kbar.

There are several possible explanations for this apparent discrepancy. Firstly, our results suggest that the calibration used by Hazen (1976), which was based on work by Adams and Williamson (1923), under-estimates pressures by about 20 percent. This would of course contribute to an underestimation of the transformation pressures, but this alone cannot account for the discrepancy. Other contributory factors may be related to the use of a potentially non-hydrostatic medium (refractive index oil) by Hazen (1976). Such a medium can support deviatoric stress which could contribute to a drastic reduction in the transition pressure of a transformation of this sort. If so then the results reported here would indicate that the "stress-free" transition pressure is in excess of 50 kbar, and that the presence of deviatoric stress may reduce the transition pressure by more than 30 kbar. Another possibility is that the transformation did take place, but we were unable to detect it, because the twin domains arising out of the $C2/m$ to $C\bar{1}$ symmetry reduction were sufficiently small for the crystal to retain the appearence of monoclinic symmetry. A similar phenomenon is observed during the earliest stages of ordering in adularia (McConnell 1965) in which the local symmetry is $C\bar{1}$, but the crystal as a whole appears to retain monoclinic symmetry. A non-hydrostatic medium, on the other hand, may promote the formation of larger, and therefore detectable twins. Further studies of a suite of alkali feldspars are currently underway to resolve these questions.

Conclusions

This study has provided compressibility data for the three end-member feldspars albite, anorthite, and sanidine. Low albite and high sanidine show almost identical behaviour on compression which indicates that the state of aluminium-silicon order within the feldspar framework does not have a major effect on the compressibility of this structure type. When these new data are combined with thermal expansion data (Stewart and von Limbach 1967, Henderson 1979), some indication of the bulk behaviour of the alkali feldspars within the crust and upper mantle can be predicted. Albite has ratio of $\frac{\alpha}{\beta}=16.4$ bar. K^{-1}, and sanidine has $\frac{\alpha}{\beta}=9.6$ bar. K^{-1}. These are significantly less than typical values of between 18 and 20 bar. K^{-1} for most minerals and compares with the average initial geothermal gradient which is also about 20 bar. K^{-1}. Thus, in contrast with most other mineral phases, alkali feldspars become denser with depth in the crust. However, this analysis assumes that the derivatives $\partial\alpha/\partial P$ and $\partial\beta/\partial T$ are zero. If, as Brown et al. (1984) suggest, the high sanidine framework is almost fully expanded at room temperature and pressure, this assumption would break down, and the thermal expansion coefficient of sanidine, α, would be expected to increase with pressure. This would result in a $\frac{\alpha}{\beta}$ ratio, and thus behaviour at depth within the earth, much closer to that of other minerals.

The compression of anorthite differs from the other feldspars studied in two respects. Firstly it is less compressible than either albite or sanidine, a result attributable to the greater charge of the interstitial cation. Secondly a new phase transition in anorthite has been discovered, which may be related to further ordering of the calcium on two of the four Ca sites within the structure. Although the pressure at which this transformation occurs is outside the stability field of anorthite within the crust, the behaviour of the low pressure $P\bar{1}$ phase remains difficult to predict, as the thermal expansion behaviour at temperatures between 25° C and 240° C is dominated by the strain preceeding the $P\bar{1}$–$I\bar{1}$ transformation (Redfern and Salje 1987). Further work is therefore required to investigate how this spontaneous strain varies as a function of temperature at pressure, as well as to delineate the composition dependence of the newly-discovered high-pressure transition.

Acknowledgements. We would like to thank Dr. Michael Carpenter and Dr. Stuart Agrell, both of the Earth Sciences department of the University of Cambridge, for kindly donating the anorthite and albite specimens used in this study. Discussions with Dr. Michael Carpenter, Dr. Ekard Salje, and Simon Redfern of the University of Cambridge, and an extensive review by Dr. William Brown of CNRS Nancy, are gratefully acknowledged. This study was supported by grants from the National Science Foundation, EAR86-18602 to CTP, EAR87-09226 to JRS, by a NATO Overseas Research Fellowship to RJA, and by the Carnegie Institution of Washington.

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Received September 21, 1987