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Letter

The elastic tensor of monoclinic alkali feldspars

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Abstract

The full elastic tensors of two K-rich monoclinic alkali feldspars, $Or_{83}Ab_{15}$ sanidine and $Or_{93}Ab_7$ orthoclase, have been determined by using the Impulse Stimulated Light Scattering technique to measure surface acoustic wave velocities. The new data confirm that alkali feldspars exhibit extreme elastic anisotropy, so the bounds of their isotropic average properties span a wide range. The measured adiabatic moduli are, for $Or_{83}Ab_{15}$ and $Or_{93}Ab_7$, respectively, K_{Reuss} = 54.7(7), 54.5(5) GPa; K_{Voigt} = 62.9(1.1), 64.4(0.6) GPa; G_{Reuss} = 24.1(1), 24.5(1) GPa; and G_{Voigt} = 36.1(5), 36.1(7) GPa. The small differences in moduli between the samples suggests that variations in composition and in state of Al, Si order only have minor effects on the average elastic properties of K-rich feldspars. The new measurements confirm that the earliest determinations of elastic wave velocities of alkali feldspars, widely used to calculate wave velocities in rocks, resulted in velocities systematically and significantly too slow by 10% or more.

Keywords: Alkali feldspar, elastic tensor, impulse stimulated light scattering

Introduction

To be able to understand and interpret the seismic signal from the Earth in terms of phase stabilities and to obtain information about fabric, texture, and mineralogy from seismic wave speeds, full knowledge of the anisotropic elastic properties of minerals is required. From these, the wave velocities can be determined. For example *p*-wave velocities are equal to $(c'/\rho)^{1/2}$ where c' is the compressional modulus in the wave propagation direction and ρ the density. Full elastic tensors are also required to interpret diffusion in feldspars (e.g., Schäffer et al. 2014), the morphology of microstructures such as perthite exsolution in feldspars (e.g., Williame and Brown 1974) or the orientation and properties of twin walls (e.g., Salje 2015). Elastic properties are also estimated to contribute to about 50% of the free energy change of displacive structural phase transitions in feldspars (Carpenter and Salje 1994, 1998). An invariant of the full elastic tensor is the bulk modulus, required to define the volume variation with pressure and thus the thermodynamic stability of minerals.

Feldspars constitute the most volumetrically important constituent of the Earth's crust, and alkali feldspars are important in deep subduction and high-pressure metamorphism. Yet, the most widely used elastic data for K-rich alkali feldspars continues to be that of Ryzhova and Aleksandrov (1965), obtained by measurements of ultrasonic wave velocities in pseudo-single-crystals of perthites, an intergrowth of albite and K-feldspar, at room conditions. Not only are these velocity data therefore not representative of a single-phase K-rich feldspar, but in situ high-pressure wave velocity measurements on similar feldspars showed that the room-pressure measurements of Ryzhova and Aleksandrov (1965) yielded p-wave velocities that are systematically slow by between 10 and 30% (Simmons 1964; Christensen 1966). This discrepancy was attributed to the crystals containing cleavage partings and other defects, which are open at room pressure and only closed under several kilobars of external pressure. This was confirmed by the determination of the full elastic tensor of a gem-quality crystal of monoclinic $Or_{89}Ab_{11}$ sandine by ultrasonic resonance measurements (Haussühl 1993), which yielded significantly stiffer values of the individual moduli than those of Ryzhova and Aleksandrov (1965), corresponding to higher wave velocities. Furthermore, the compliances s_{ij} of Haussühl (1993) yield a value of the Reuss bulk modulus K_R = 55.7 GPa in reasonable agreement with values of 52(1) and 57(1) GPa determined from two K-rich sanidines by single-crystal diffraction (Angel 1994), and significantly higher than the range of K_R = 39 to 51 GPa from Ryzhova and Aleksandrov (1965). The single-crystal elastic moduli of albite reported by Ryzhova and Aleksandrov (1965) were also shown to be too soft by Brown et al. (2006), who also demonstrated that their data acquisition scheme was actually insufficient to determine all 13 independent elastic tensor components of monoclinic crystals.

Thus, the only previously published data for the full elastic tensor of K-rich feldspars that is not known to be problematic is the determination by Haussühl (1993). We have, therefore, undertaken a determination of the full elastic tensors of two additional well-characterized monoclinic K-rich feldspars with differing states of Al, Si order and differing compositions. Together with the results from Haussühl (1993) they provide a first indication of the possible effects of composition and state of order on the elastic properties of K-rich feldspars and, in combination with the elastic tensor of

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albite (Brown et al. 2006), an indication of the total variability of elastic properties across the entire alkali feldspar join.

Samples and methods

The two current samples (H002 and H003) were provided by J. Schlüter from the collection of the University of Hamburg, Germany. Sample H002 is a natural sanidine of approximate composition $Or_{83}Ab_{15}$ with 1.8 mol% celsian and 0.5% Sr-feldspar components, H003 is a natural orthoclase of composition $Or_{93}Ab_7$ without additional components (Angel et al. 2013). Single-crystal structure refinements (Angel et al. 2013) indicate that H002 sanidine has a more disordered Al, Si distribution than H003 orthoclase, as calculated by the method of Kroll and Ribbe (1983) from the mean tetrahedral bond lengths. In H002, the Al occupancy of the T1 site is calculated as 0.31, whereas it is 0.38 for the orthoclase. The diffraction pattern of H003 also includes the strong diffuse scattering typical of orthoclases and indicates that the local short-range order of Al and Si is higher than indicated by the average from structure refinements (e.g., Pleger 1996; Sanchez-Munoz et al. 1998). Both samples are metrically monoclinic. Densities were determined from the measured compositions and the unit-cell volumes determined by X-ray diffraction (Angel et al. 2013).

Surface elastic wave velocities were measured on several different sections of both samples by impulse stimulated light scattering (ISLS) (Abramson et al. 1999). Crystals were oriented on a four-circle X-ray diffractometer and glued to a glass slide while still attached to the goniometer to maintain the orientation. The crystals were then potted in epoxy and ground using 0.25 μm diamond powder for finishing. An aluminum film ~40 nm thick was applied to the surface to allow the coupling of the incident laser energy to the crystal surface and thus the generation of the surface waves. The aluminum film alters surface wave velocities by about 0.1%; this systematic effect is included in the data analysis (Brown et al. 2006). Nine crystals were prepared for H002 and eight crystals for H003 and the surface wave velocities were measured on each crystal slice in all directions from 0° to 180° (see Supplementary Material[1]) using steps of 10° (n.b. the data from 180° to 360° is identical to the one from 0° to 180°) within the surface (Brown et al. 2006). The elastic moduli reported in Table 1 were determined from the measured surface wave velocities through non-linear parameter optimization, using both "Levenberg-Marquardt" and "Nelder-Mead simplex" methods (Brown in preparation). Because the diagonal (e.g., c_{11}) and off-diagonal (e.g., c_{12}) moduli enter into the calculation of surface wave velocities as differences (Brown et al. 2006), the optimization was further constrained by use of the linear compressibilities $\beta_i = s_{i1} + s_{i2} + s_{i3}$ determined by fitting the unit-cell parameter variation of the same samples at high pressures were measured at Virginia Tech (N.L. Ross, personal communication) by single-crystal X-ray diffraction. The difference between isothermal elasticity (as determined under hydrostatic compression) and adiabatic elasticity (surface wave velocities) is controlled by the factor $(1+\alpha\gamma T)$ where α is thermal expansivity and γ is the Grüneisen parameter. In the case of feldspars near room temperature, the factor $(1+\alpha\gamma T)$ is, within experimental uncertainty, equal to 1 (Tribaudino et al. 2011). Thus, the difference between adiabatic and isothermal moduli and compliances is assumed negligible. As for the previous measurement of albite elasticity (Brown et al. 2006) the elastic moduli are described with respect to a Cartesian axial system whose alignment with respect to the non-orthogonal monoclinic crystal axes is **Y**//**b***, **Z**//**c**, and **X**//**Y** × **Z**. For the monoclinic samples described in this paper this means that **X**//**a***, **Y**//**b**, and **Z**//**c**.

Results

Anisotropic behavior

In Table 1 individual moduli (c_{ij}) and components of compressibility, β_i (i = 1–5) for the alkali feldspars are listed in order of increasing potassium from the pure sodium end-member albite on the left to $Or_{93}Ab_7$ on the right. The uniquely triclinic moduli of albite are not listed. In the case of albite, H002 and H003, values for β_i were independently constrained by the X-ray high-pressure measurements. In the case of $Or_{89}Ab_{11}$, they are derived from the

[1] Deposit item AM-16-55583, Supplemental Material. Deposit items are free to all readers and found on the MSA web site, via the specific issue's Table of Contents (go to http://www.minsocam.org/MSA/AmMin/TOC/).

Table 1. Elastic properties of alkali feldspars

	Brown et al. (2006)		H002		Haussühl (1993)		H003	
	Or_0Ab_{100}	Albite	$Or_{83}Ab_{15}$	Sanidine	$Or_{89}Ab_{11}$	Sanidine	$Or_{93}Ab_7$	Orthoclase
X-ray density (g/cm^3)	2.623		2.567		2.56		2.555	
(ij)	**C_{ij} (GPa)[a]**	**2σ**	**C_{ij} (GPa)**	**2σ**	**C_{ij} (GPa)[b]**	**2σ**	**C_{ij} (GPa)**	**2σ**
11	69.9	0.6	69.3	0.6	68.6	0.8	67.8	0.6
12	34.0	0.7	41.6	1.6	43.6	1.6	40.4	1.0
13	30.8	0.5	24.0	0.6	26.0	1.4	25.0	1.0
15	–2.4	0.1	0.3	0.1	–0.7	1.4	–1.1	0.2
22	183.5	2.7	176.2	5.3	176.8	1.0	181.2	3.5
23	5.5	2.2	14.3	3.2	21.0	2.0	20.6	3.9
25	–7.7	0.7	–9.4	0.6	–12.6	2.0	–12.9	0.6
33	179.5	2.3	160.8	2.3	159.9	1.2	158.4	4.0
35	7.1	0.6	7.1	0.5	6.9	1.4	10.6	0.7
44	24.9	0.1	19.2	0.1	19.3	0.4	21.1	0.1
46	–7.2	0.1	–11.5	0.1	–10.8	0.6	–11.6	0.2
55	26.8	0.2	19.4	0.1	18.0	0.8	19.4	0.1
66	33.5	0.2	33.4	0.2	33.5	0.6	33.1	0.2
			Compressibilities[c] β (TPa^{-1})					
1	11.1		11.4	0.05	11.5		11.8	0.06
2	3.4		2.6	0.12	2.4		2.4	0.03
3	3.6		4.3	0.02	4.0		4.1	0.05
5	1.0		–0.5	0.03	0.5		0.0	0.02
			Isotropic properties (GPa)					
K_{Reuss}	55.0		54.7	0.7	55.7		54.5	0.5
K_{Voigt}	63.7		62.9	1.1	65.2		64.4	0.6
K_{VRH}	59.4		58.8		60.4		59.5	
G_{Reuss}	29.8		24.1	0.1	23.6		24.5	0.1
G_{Voigt}	41.2		36.1	0.5	35.1		36.1	0.7
G_{VRH}	35.5		30.1		29.4		30.3	
			Velocity (km/s)					
v_s (VRH average)	3.7		3.4		3.4		3.4	
v_p (VRH average)	6.4		6.2		6.2		6.3	

[a] Albite is triclinic and has 21 independent moduli c_{ij}. Only those allowed to be non-zero under monoclinic symmetry are listed here for comparison with the monoclinic feldspars.

[b] The elastic moduli reported in Haussühl (1993), have been rotated by 26°—see text. Uncertainties listed are in the original coordinate system with the assumption that 1σ values rather than 2σ values were reported. This assumption is justified through comparison with uncertainties reported in other papers giving moduli for monoclinic crystals using the same technique (e.g., Isaak et al. 2006).

[c] The compressibilities are defined in terms of the elastic compliance matrix as $\beta_j = (s_{1i} + s_{2i} + s_{3i})$.

reported elastic moduli. The resulting isotropic moduli and estimated compressional and transverse wave velocities in a random aggregate of the minerals are given at the bottom of the table.

The elastic moduli for $Or_{89}Ab_{11}$ reported in Haussühl (1993) indicate that the *a*-axis is the most compressible direction and that the stiffest direction is rotated by 26° toward *a** from the *c*-axis. In addition, the tensor values for thermal expansivity in that study indicate that the *a*-axis has the highest thermal expansivity. Such results are in conflict with all recent studies showing that extrema in elasticity and thermal expansivity for feldspars are closely aligned with *a** and *c*. The elastic moduli listed in Table 1 for Haussühl (1993) have been rotated under the assumption that they were based on a coordinate system with X parallel to the *a*-axis [the coordinate system used in the pioneering work by Ryzhova and Aleksandrov (1965)]. That the rotated values are in agreement with the present study provides support for the supposition that the coordinate system used by Haussühl (1993) was that of Ryzhova and Aleksandrov (1965). The alternative assumption that the coordinate system was correctly described, leads to two equally implausible conclusions: that all recent analyses are wrong or that properties of entirely different materials are being investigated.

The elastic moduli show only modest variations across the compositional range and the three potassium-containing feldspars are remarkably similar (see Table 1). While the elasticity of albite approximates uniaxial symmetry with $c_{22} \sim c_{33}$ and $c_{12} \sim c_{13}$, the elasticity of the potassium-rich samples is triaxial with $c_{22} > c_{33}$ and $c_{12} > c_{13}$.

In Figure 1 predicted quasi-longitudinal and quasi-transverse velocities for albite and $Or_{93}Ab_7$ are shown in three planes associated with the coordinate system. Velocities for the $Or_{83}Ab_{15}$ and $Or_{89}Ab_{11}$ sanidine samples appear identical at the scale of these plots. The high degree of anisotropy previously reported for albite is maintained across the potassium-bearing samples. The degree of anisotropy exhibited by these feldspars is similar to that found in layered structures such as micas (e.g., McNeil and Grimsditch 1993) or portlandite (Speziale et al. 2008). In all samples, the longitudinal velocities are near 8 km/s along both the Y-axis (parallel to crystal *b*-axis) and the Z-axis (parallel to the *c*-axis). The lowest longitudinal velocities (near 5.5 km/s) appear parallel to the X-direction (*a** direction). Quasi-transverse velocities range from slightly more than 2 km/s in the X-Y plane to between 5 and 6 km/s in the Y-Z plane. In the Y-Z plane, the quasi-longitudinal and quasi-transverse modes become nearly degenerate between the *c*-axis and the *b*-axis. The quasi-transverse velocities show little directional dependence in the X-Y plane with the slowest velocities just above 2 km/s. Only subtle differences in the patterns of anisotropy are apparent between the albite and orthoclase samples.

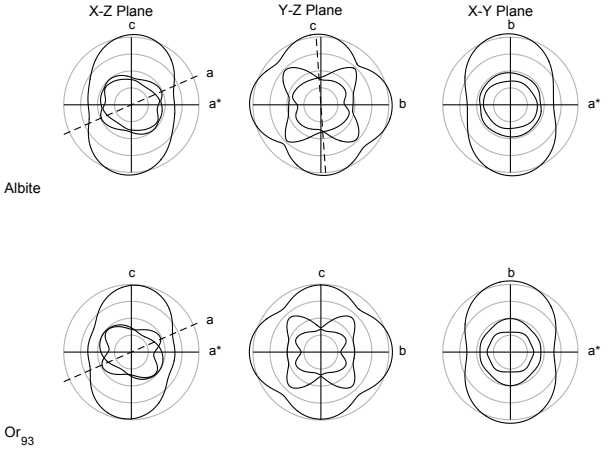

Figure 1. Velocities for albite and $Or_{93}Ab_7$ shown in three different planes. Light gray circles are velocities of 2, 4, 6, and 8 km/s. Dark curves are velocities of quasi-longitudinal and quasi-transverse elastic waves. The orientations of crystallographic axes are shown.

Isotropic average properties

For crystals of less than cubic symmetry the two bounding values of the bulk modulus, the Reuss and Voigt bulk moduli, correspond to the stiffness of the single crystal under, respectively, uniform hydrostatic stress and uniform strain. By definition, the Reuss bulk modulus is identical to the bulk modulus determined in hydrostatic compression measurements. Similar definitions corresponding to uniform shear stress and uniform shear strain conditions give Reuss and Voigt bounds on the shear moduli (e.g., Newnham 2005). These moduli correspond to the widest possible limits for the elastic properties of an aggregate (Avellaneda and Milton 1989; Avellaneda et al. 1996). The tightest constraints that can be determined without a detailed description of the microstructure of the material are provided by the Hashin-Striktmann bounds (e.g., Brown 2015). Because the Hill average of the Voigt and Reuss moduli (K_{VRH}, G_{VRH}) typically falls within the Hashin-Strikmann bounds, this average provides a convenient estimation for properties of an aggregate rock without preferred orientation and with grains locked together. If the grain boundaries are free to "slide" or relax, for example due to the presence of grain boundary fluids or melt, the average moduli will be reduced toward the Reuss bounds.

Our two new determinations of the elastic tensors of K-rich alkali feldspars, together with that of sanidine (Haussühl 1993), show that the variation in their average elastic properties is small, of the order of 1% or less in bulk moduli and 3% in shear moduli (Table 1). The VRH average wave velocities from all three samples are v_p = 6.25(5) km/s and v_s = 3.4(1) km/s, significantly faster than the velocities derived from the elastic tensors reported by Ryzhova and Aleksandrov (1965), which correspond to ranges of v_p = 5.6–5.9 km/s and v_s = 3.0–3.3 km/s. Furthermore, the bulk moduli of these three K-rich feldspars are very similar to end-member albite but the K-rich feldspars are significantly softer in shear by 5–6 GPa. This is presumably a consequence of the monoclinic-triclinic phase transition. As a consequence, the average elastic wave velocities of albite are higher, by ~0.15 km/s for v_p and ~0.3 km/s for v_s (Fig. 2).

Implications

The anisotropy of the expansion of alkali feldspars as a result of temperature increase (i.e., thermal expansion) or substitution of larger cations such as K^+ for Na^+ has been explained as arising from a particular pattern of cooperative tilting of relatively rigid tetrahedra within the structure. While many possible combinations of tetrahedral tilts are possible, the single cooperative pattern of tilts that operates is that which maximizes short O-O distances within the structure (Angel et al. 2012, 2013). These tilts make the (100) plane normal the direction of

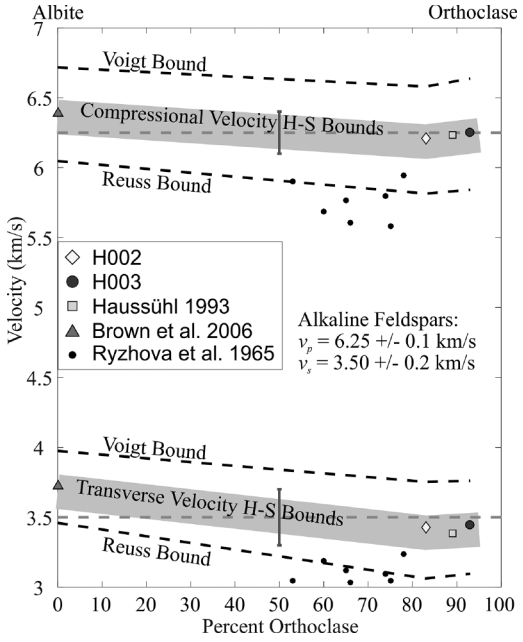

Figure 2. Bounds and averages of the compressional and transverse wave velocities for alkali feldspars. The Voigt and Reuss bounds are shown by black dashed lines, and the Hashin-Shtrikman bounds as a gray band. The gray dashed line within the Hashin-Shtrikman bounds shows the mean of the Hill averages for alkali feldspars. For comparison with the new data H002 and H003 the data from Haussühl (1993) is given as well as the velocities from Ryzhova and Aleksandrov (1965).

greatest expansion and contraction. The fact that we now observe a remarkably similar anisotropy in the elastic properties of alkali feldspars, suggests that at a crystal-chemical level the same mechanisms of tetrahedral tilting are the dominant response of the structure to applied stress, at least in the low-stress regime of linear elasticity represented by the elastic tensors reported here. Obviously, at elevated pressures the structures will become stiffer in the non-linear elastic regime and one can expect additional mechanisms such as tetrahedral deformation to become more significant.

The new data, in combination with that of Haussühl (1993), confirms the conclusion of Simmons (1964) and Christensen (1966) that the original determinations by Ryzhova and Aleksandrov (1965) were affected by open spaces in the samples, which led to their calculated average properties being too soft and thus seismic wave velocities that are significantly and systematically too low. The remaining available data now show that the average elastic properties and wave velocities across the alkali-feldspar join (Fig. 2) vary little with either composition or state of order. There is certainly less than the 15% difference in bulk modulus between sanidine and orthoclase suggested by Hacker (2003), and significantly less than the 30% difference in bulk moduli across the plagioclase feldspar join from albite to anorthite (Angel 2004). While these conclusions concerning average properties of alkali feldspars are robust, further work is required to confirm the details of variation of individual elastic moduli and compliances with composition and state of order.

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