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Dielectric Constants of Topaz, Orthoclase and Scapolite and the Oxide Additivity Rule

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Abstract. The dielectric constants and dissipation factors of topaz, scapolite and orthoclase were determined at 1 MHz using a two-terminal method and empirically determined edge corrections. The results are:

topaz $\kappa'_a = 6.61$ $\tan\delta = 0.0005$
$\kappa'_b = 6.82$ $\tan\delta = 0.0007$
$\kappa'_c = 6.81$ $\tan\delta = 0.0007$
orthoclase $\kappa'_a = 4.69$ $\tan\delta = 0.0007$
$\kappa'_b = 5.79$ $\tan\delta = 0.0007$
$\kappa'_c = 5.63$ $\tan\delta = 0.0011$
$\kappa'_{11} = 4.72$
$\kappa'_{22} = 5.79$
$\kappa'_{33} = 5.76$
scapolite $\kappa'_a = 6.74$ $\tan\delta = 0.0004$
$\kappa_c = 8.51$ $\tan\delta = 0.0004$

The deviation (Δ) between measured dielectric polarizabilities as determined from the Clausius-Mosotti equation and those calculated from the sum of ion polarizabilities according to $\alpha_D(\text{mineral}) = \Sigma\alpha_D(\text{ions})$ for topaz is 2.5%. The large deviations of orthoclase and scapolite from the oxide additivity rule with $\Delta = +7.2$ and $+17.6\%$, respectively, are attributed to "rattling" K ions in orthoclase and "rattling" (Na, K, Ca) ions and disordered $O^=$ and Cl^- ions in scapolite.

Introduction

The concept of additivity of molecular polarizabilities implies that the molecular polarizability of a complex substance can be broken up into the molecular polarizabilities of simpler substances according to the relation:

$$\alpha_D(M_2M'X_4) = 2\,\alpha_D(MX) + \alpha_D(M'X_2) \tag{1}$$

where the dielectric polarizability, α_D, is related to the measured dielectric constant, κ', by the Clausius-Mosotti equation:

$$\alpha_D = 1/b\,[(V_m)(\kappa'-1)/(\kappa'+2)] \tag{2}$$

V_m is the molar volume in Å^3, b is assumed to be $4\pi/3$, and κ', the real part of the complex dielectric constant, is measured in the range 1 kHz to 10 MHz (Roberts 1950, 1951). Alternatively, the ion additivity rule states that molecular polarizabilities of complex substances can also be broken up into *ion* polarizabilities according to the relation:

$$\alpha_D(M_2MX_4) = 2\,\alpha_D(M^{2+}) + \alpha_D(M^{4+}) + 4\,\alpha_D(X^=) \tag{3}$$

Ion polarizabilities are useful when compound polarizabilities are not available.

The oxide and ion additivity rules have been observed to be followed to ±0.5–1.5% in a class of "well-behaved" oxides which includes beryllates, borates, aluminates, gallates, silicates, germanates, phosphates and vanadates (Shannon 1991; Shannon and Subramanian 1989; Shannon et al. 1989, 1990, 1991a, b; 1992a, b; Subramanian and Shannon 1989). Some compounds show larger deviations (>5%) because of ionic or electronic conductivity, the presence of H_2O or CO_2 (Shannon et al. 1989; Shannon et al. 1992f; Ganguli et al. 1992), or "rattling" cations (Shannon and Rossman 1992; Shannon et al. 1992a, b, c, d, e).

The purpose of this paper is to accurately determine the 1 MHz dielectric constants of topaz, orthoclase and scapolite, and to evaluate the validity of the oxide or ion additivity rules in these materials.

Experimental

The sources of these gem-quality crystals ere: orthoclase – colorless crystal, from Madagascar; scapolite – straw-colored crystal from Morogoro, Tanzania, and topaz – a colorless crystal of unknown origin.

The samples were oriented for cutting by polarized light and by back-reflection Laue photographs. X-ray diffraction patterns were obtained on a Guinier-type focussing camera using CuKα_1 radiation and a Si SRM 640 internal standard. Cell dimensions were obtained by least-squares refinement.

Electron microprobe analyses were made using a JEOL 733 electron microprobe. Data reduction methods are described by Armstrong (1982, 1988). Although no systematic effort was made to obtain information on possible chemical zonation, analyses of 5 different points did not indicate significant variation in the composition of major constituents except in the case of scapolite, where Al ranges from 3.76 to 3.81 formula proportions.

Typical analyses indicated the following compositions: $Al_{1.99}S_{1.01}O_4F_{1.89}(OH)_{0.11}$ for topaz, $K_{0.92}Na_{0.08}Al_{0.97}Fe_{0.02}Si_{3.0}O_8$ for orthoclase, and $(Na_{2.67}K_{0.28}Ca_{1.06}Sr_{0.01})[Al_{3.79}Si_{8.19}Fe_{0.02}]\{Cl_{0.76}(CO_3)_{0.19}(SO_4)_{0.08}\}O_{24}$ for scapolite.

Rectangular-shaped samples were cut from the bulk crystals using a low-speed diamond wheel saw. Slabs were cut perpendicular to the major axes of the topaz and scapolite crystals to obtain κ'_{11}, κ'_{22}, and κ'_{33}. Slabs of orthoclase were cut perpendicular to the reciprocal axes a^*, b^*, c^* and $[1, 0, \bar{1}]$. Sputtered gold electrodes were applied over the entire parallel surfaces of the sample using a Denton Vacuum Desk II sputtering unit. Sample preparation is described in detail in Subramanian et al. (1989). Dielectric constant measurements were performed over the frequency range 30 kHz–3 MHz with a parallel-plate capacitance technique using a Hewlett-Packard 4275A LCR bridge and fixture 16043B (Test Tweezers) according to the procedure described by Subramanian et al. (1989). Edge corrections were made using the expression:

$$C_e = (0.019 \ln P/t - 0.043)P \qquad (4)$$

where t is the sample thickness and P is the perimeter in cm. The dielectric constants determined for the 4 slabs of orthoclase were: E parallel to a^* (4.69 ± 0.01); E parallel to b^* (5.79 ± 0.02); E parallel to c^* (5.63 ± 0.02); E parallel to $[1, 0, \bar{1}]$ (5.53 ± 0.05). These values were used to obtain κ'_{11}, κ'_{22}, and κ'_{33} using the procedure described by Takubo (1941). The overall accuracy of the dielectric constant measurements using the above techniques is estimated to be 1.0–1.5%. Dielectric loss errors are estimated to be 5–20% at levels of $\tan \delta = 0.002$ and 50–100% at levels of 0.0004–0.0005.

Table 1. Electron microprobe analyses, unit cell dimensions, and molar volumes of topaz, orthoclase, and scapolite

	topaz	orthoclase	scapolite
Na_2O		0.91	9.39
K_2O		15.68	1.49
Al_2O_3	54.56	17.89	21.93
SiO_2	31.82	65.21	55.89
CaO		0.01	6.75
SrO		0.01	0.08
BaO		0.11	
TiO_2	0.01	0.01	0.01
FeO	0.01	0.44	
Fe_2O_3			0.30
Ga_2O_3	0.03		
NiO		0.03	
ZnO	0.02	0.05	
F	20.39		
Cl			3.04
SO_3			0.73
CO_2			0.93
F=O	−8.586		
Total	98.25	100.35	99.84
a, Å=	4.6495(2)	8.5741(4)	12.0693(5)
b, Å=	8.7967(1)	12.9916(8)	12.0693(5)
c, Å=	8.3929(2)	7.1979(5)	7.5816(6)
β,°=		116.041(4)	
V_m, Å^3=	85.84	180.10	552.19

Table 2. Summary of single-crystal dielectric constants

κ'_{a^*}, $\tan \delta$	κ'_{b^*}, $\tan \delta$	κ'_{c^*}, $\tan \delta$	$\langle \kappa' \rangle$	Frequency	Reference
topaz					
6.608 ± 0.02 0.0005	6.816 ± 0.03 0.0007	6.810 ± 0.04 0.0007	6.74	1 MHz	This work
6.53	6.82	6.78	6.71	10 MHz	Westphal, Sils (1972)
6.30	6.70	6.65	6.55	RF	Keller (1966)
6.27	6.43	6.31	6.34	RF	Keller (1966)
6.3	6.7	6.7	6.57	1000 GHz	Liebisch, Rubens (1919)
6.25	6.54	6.44	6.41	0	Fellinger (1919)
6.30	6.70	6.65	6.55	400 MHz	Schmidt (1902)
6.58	6.95	6.62	6.72	7 MHz	Wappler (1964)
6.28	6.44	6.31	6.34	500 kHz	Takubo et al. (1953)
6.29	6.42	6.34	6.35	500 kHz	Takubo et al. (1953)
6.80, $Al_2SiO_4F_2$, undef. orientation [184.6], d = 3.500 g/cc				1 MHz	Olhoeft (1981)
6.40, $\tan \delta = 0.020$, undef. orientation from Stoneham, ME, d = 3.386 g/cc				1 MHz	Olhoeft (1981)
5.00, $Al_2SiO_4(F,OH)_2$, undef. orientation, d = 3.174 g/cc				1 MHz	Olhoeft (1981)
orthoclase					
4.688 ± 0.01 0.0007	5.792 ± 0.02 0.0007	5.630 ± 0.02 0.0011		1 MHz	This work
$\kappa'_{11} = 4.72$	$\kappa'_{22} = 5.792$	$\kappa'_{33} = 5.76$	5.42	1 MHz	This work
4.50	5.80	5.55	5.28		Keller (1966)
5.1, $\tan \delta = 0.0195$, undef. orientation [13.5], Kuggles Mine, NH; d = 2.55 g/cc				1 MHz	Olhoeft (1981)
5.7, $\tan \delta = 0.0253$, undef. orientation [82.6] Custer, CO; d = 2.587 g/cc				1 MHz	Olhoeft (1981)
4.56 (powder), $\tan \delta = 0.00006$				300 MHz	Church et al. (1988)
scapolite					
6.736 ± 0.01 0.0004		8.506 ± 0.06 0.0004	7.326	1 MHz	This work

Results

Table 1 summarizes the electron microprobe analyses, unit cell dimensions and molar volumes of the topaz, orthoclase and scapolite studied here. Table 2 summarizes dielectric constants and dielectric loss (tan δ) values at 1 MHz. The dielectric constants of orthoclase showed deviations of 0.3–0.5% over the range of frequencies 30 kHz to 3 MHz for $\bar{E}$ parallel to a^* and b^* and 0.8–1.0% for $\bar{E}$ parallel to c^*. Topaz and scapolite showed deviations of 0.3–0.6% over the same frequency range.

In Table 2 we compare our data with previously reported dielectric data for topaz and orthoclase. Dielectric data exist for a great many topaz crystals. The order of κ'_a, κ'_b, and κ'_c reported in the literature is not uniform, probably because of variation in choice of orthorhombic axes by previous authors. If we arbitrarily order $\kappa'_a < \kappa'_c < \kappa'_b$ in Table 2 as found for our topaz, then variations of κ'_a from 6.3 to 6.6, κ'_b from 6.4 to 7.0, and κ'_c from 6.3 to 6.8 are evident. Such variation is not surprising considering the variability in hydroxyl content (Deer et al. 1962). For example, hydroxylated topaz would be expected to have a higher κ' than the pure fluoride variety. Some of the variation can also be attributed to poor experimental technique, poor sample quality and decrease of κ' at high frequencies. The low values of Takubo et al. (1953), are probably caused by faulty technique since his values for quartz are ~8% low. Increasing his values by 8% increases the mean values from 6.35 to 6.90, more in line with our measurements. The low values of Schmidt (1902) and Liebisch and Rubens (1919) may be caused by reduced κ's at GHz frequencies. Values obtained by Olhoeft (1981) on 2 samples of undetermined orientation fall in these ranges but his value of 5.0 obtained on a hydroxylated topaz is unusually low. The low density of this sample is outside the range of 3.49–3.57 g/cc reported by Deer et al. (1962) and suggests that this crystal may not have been topaz.

The values of $\kappa'_a = 4.50$, $\kappa'_b = 5.80$ and $\kappa'_c = 5.55$ for orthoclase reported by Keller (1966) are reasonably close to the values reported by us. The values of 5.1 and 5.7 reported by Olhoeft for samples of orthoclase of unspecified orientation from New Hampshire and Colorado fall in the range of values observed for our sample from Madagascar. The relatively high values of tan $\delta \sim 0.02$ suggests poor sample quality. The value of $\kappa' = 4.56$ (at 300 MHz) reported by Church et al. (1988) for a powder sample of orthoclase appears anomalously low but may result from dispersion.

Discussion

Table 3 lists mean dielectric constants and molar volumes of the oxides which are used to test the oxide additivity rule. Arithmetic, rather than geometric, mean values are used. Geometric mean values are smaller by 0.0–0.2% for crystals with little or no anisotropy, whereas they can be smaller by 2.2% for crystals such as zoisite (Shannon et al. 1992c) with significant anisotropy.

Table 4 compares the total molecular dielectric polarizabilities determined from the measured dielectric constants of the minerals studied here along with the values calculated from the oxide or ion additivity rules. The discrepancy, Δ, between the observed dielectric polarizability of topaz and that calculated from the sum of the ion polarizability according to the ion additivity rule (3), is -2.5%, comparable to the typical 1–3% variation observed previously for a series of aluminates, gallates, silicates, germanates, phosphates, and vanadates (Shannon 1991; Shannon and Subramanian 1989; Shannon et al. 1989, 1990, 1991a, b, 1992a, b; Subramanian and Shannon 1989). However, both orthoclase and scapolite show deviations of $+7.2\%$ and 17.6%, respectively. Large deviations from the oxide additivity rule in other minerals, e.g. anorthite and α-spodumene (Shannon et al. 1992b), beryllonite (Shannon et al. 1992a) epidote and zoisite (Shannon et al. 1992d), pyrope garnet (Shannon and Rossman 1992) and a series of melilites (Shannon et al. 1992e) have been attributed to the presence of "rattling" cations (Orgel 1958) which are characterized by lower than ideal apparent bond valences (V′) of one or more of the cations (O'Keeffe 1989). This effect

Table 3. Dielectric constants and molar polarizabilities of simple oxides and chlorides used to calculate dielectric additivities

Compound	Orientation	κ'	$\langle\kappa'\rangle$	V_m, Å^3	α_D, Å^3	Reference
Na_2O					5.59	Shannon (1991)
K_2O					10.91	Estimated
CaO		11.95	11.95	27.83	5.22	Subramanian et al. (1989)
FeO					4.3	Shannon et al. (1991a)
SrO		14.5	14.5		6.48	Bartels et al. (1979)
BaO		31.1	31.1	42.48	9.22	Shannon et al. (1992e)
Al_2O_3	$E \parallel a$	9.395	10.126	42.45	7.627	Fontanella et al. (1974)
	$E \parallel c$	11.589				
Fe_2O_3					10.51	Shannon et al. (1991b)
SiO_2	$E \parallel a$	4.520	4.559	37.66	4.878	Fontanella et al. (1974)
	$E \parallel c$	4.637				
NaCl		5.90	5.90	44.85	6.61	Lowndes, Martin (1969)
$CaCO_3$	$E \parallel a$	8.7	8.53	61.02	10.45	Westphal, Sils (1972)
	$E \parallel c$	8.2				
Na_2SO_4					13.6	Estimated from ion polarizabilities

Table 4. Comparison of observed and predicted single crystal dielectric polarizabilities

Mineral	Composition	κ'	Pred[a] α_T	Measured α_T (exp)[b]	Δ, %
topaz	$Al_2SiO_4F_{1.89}(OH)_{0.11}$	6.74	13.80	13.46	− 2.5
orthoclase	$K_{0.92}Na_{0.08}Al_{0.97}Fe_{0.02}Si_{3.0}O_8$	5.42	23.77	25.61	+ 7.2
scapolite	$(Na_{2.67}K_{0.28}Ca_{1.06}Sr_{0.01})[Al_{3.79}Si_{8.19}Fe_{0.02}]$ $\{Cl_{0.76}(CO_3)_{0.19}(SO_4)_{0.08}\}O_{24}$	7.33	73.63	89.42	+17.6

[a] From the oxide additivity rule with the exception of topaz where: $\alpha_T(Al_2SiO_4F_2) = 2\alpha_D(Al^{3+}) + \alpha_D(Si^{4+}) + 4\alpha_D(O^{=}) + 1.89\alpha_D(F^-) + 0.11\alpha_D(OH^-)$

[b] Clausius-Mosotti equation (2)

was described by Dunitz and Orgel (1906) as a progressive "loosening of the central cation at the center of its surrounding octahedron to off-center displacements characteristic of ferroelectric and antiferroelectric substances" as the size of the central cation in an octahedron decreases. The polyhedron can adjust its configuration to adapt to the small cation by movement of the cation and/or anions resulting in polyhedral distortion. Evidence for rattling cations can be seen from decreased apparent bond valences and/or increased thermal motion in the Na(1) site of beryllonite (Shannon et al. 1992a), the Li site in α-spodumene (Shannon et al. 1992b) and Ca sites in anorthite, epidote and zoisite (Shannon et al. 1992d). A similar situation occurs in orthoclase. Using $V' = \Sigma s_i = \Sigma \exp(R_0 - R)/0.37$, the parameters of Brown and Altermatt (1985) with $R_0 = 0.92\ R_0(K) + 0.08\ R_0(Na)$, and bond distances in an orthoclase of similar composition (Colville and Ribbe 1968), the apparent valence of (K, Na) in orthoclase, $V'_{K,Na} = 0.82$ vu, is significantly lower than its ideal value of 1.0 vu. We suggest that in orthoclase the presence of "rattling" (K, Na) ions, having larger than normal polarizablities, is responsible for its deviation from the oxide additivity rule.

The deviation from additivity exhibited by scapolite is unusually large. As in orthoclase and other minerals, this large deviation might be caused by "rattling" (Na, K, Ca) ions. The structure of an Ontario scapolite of similar composition, $(Na_{2.88}K_{0.21}Ca_{0.83})[Al_{3.66}Si_{8.34}]\{Cl_{0.72}(CO_3)_{0.22}\}O_{24.27}$, to the one studied here was determined by Lin and Burley (1973). We assume that the structural features of the Ontario scapolite are similar to the Morogoro scapolite. Evaluation of bond valences in the same manner as above using the bond valence parameters of Brown and Altermatt (1985) and Brese and O'Keeffe (1991), the apparent bond valence of (Na, K, Ca) is 1.01 vu. This value which is considerably smaller than the ideal value of 1.26 vu, suggests "rattling" (Na, K, Ca) ions. The polarizability discrepancy may also arise from the usually large motion of the $CO_3^{=}$ and Cl^- ions, as evidenced by the probable positional disorder of Cl^- and carbonate oxygen ions and resultant large isotropic temperature factors. It is likely that both "rattling" cations and anion disorder play a role in the usually large polarizability of scapolite.

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