Chemical variations in sodium gedrite from Labrador

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Abstract

Sodium gedrite occurs in a xenolith of ferro-aluminous gneiss enclosed by granitic rocks of the Nain complex, Labrador. The sodium gedrite is zoned, but the most Na-rich compositions approximate the formula $Na_1R_2^{2+}(R_4^{2+}R_1^{3+})Si_6Al_2O_{22}(OH, F)_2$. These are among the more Fe-rich gedrites ever reported ($X_{Mg} = 0.34–0.52$) and they are far more fluorine-rich than any orthoamphibole ever reported ($X_F = 0.243–0.493$). The gedrites have a range of compositions from $Na(A) = 0.48$, $Al(IV) = 1.44$ to $Na(A) = 0.95$, $Al(IV) = 2.10$. The most common composition is $Na(A) = 0.80$ and $Al(IV) = 2.00$. The magnitude of the "edenite" coupled substitution relative to the "tschermakite" coupled substitution is much greater in these gedrites than in most other gedrites. The factors that control the $A/Al(IV)$ ratios, or edenite/tschermakite ratios, in gedrites appear to be crystal-chemical/crystallographic in origin. High X_{Mg} favors the tschermakite exchange at the expense of the edenite exchange, perhaps by causing a slight distortion of the A site. High X_F favors the edenite exchange at the expense of the tschermakite exchange, perhaps because the hydrogen in the hydroxyl group slightly repels Na from entering the A site or because F forces Fe^{2+} into the M2 site.

Introduction

Ever since the early work by Rabbitt (1948) and Seki and Yamasaki (1957), orthoamphibole compositional variation and the nature of the anthophyllite–gedrite solid solution series have received considerable study (Lal and Moorhouse, 1969; Robinson and Jaffe, 1969; Robinson et al., 1971, 1982; Papike and Ross, 1970; Stout, 1971, 1972; Kamineni, 1975; James et al., 1978; Spear, 1980; Otten, 1984). After reviewing their data as well as the data available in the literature, Robinson et al. (1971) suggested that end-member gedrite possessed a half-filled A site (principally Na) and two Al atoms in tetrahedral coordination, yielding the formula:

$$Na_{0.5}R_2^{2+}(R_{3.5}^{2+}R_{1.5}^{3+})Si_6Al_2O_{22}(OH)_2$$
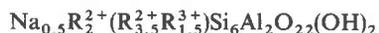
This conclusion has been generally accepted or supported in other work (cf. Papike and Ross, 1970; Cameron and Papike, 1979; Spear, 1980).

In this paper, I describe the optical properties, mineral chemistry, and crystal chemistry of the first reported occurrence of sodium gedrite[1] (Berg and Wiebe, 1978). The most Na-rich compositions have greater than 90% of the A sites filled and approximate the formula:

$$Na_1R_2^{2+}(R_4^{2+}R_1^{3+})Si_6Al_2O_{22}(OH)_2$$
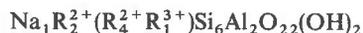
This sodium gedrite occurs in a xenolith of ferro-aluminous gneiss engulfed in a granitic body of the 1.4-b.y.- old anorthositic Nain complex in Labrador. The geology and petrology of this ferro-aluminous xenolith which is located near the abandoned Eskimo village of Zoar are described by Berg and Wiebe (1985).

Petrologic setting

The xenolith of ferro-aluminous gneiss occurs in a granitic body which was intruded along the contact between a large leuconorite pluton and older ferro-aluminous gneisses before the residual liquids of the leuconorite had completely solidified (Wiebe, 1978, 1980). This indicates that the xenolith probably endured a two-stage contact metamorphic history: the first stage being high temperature and dry, and the second stage being lower temperature and volatile-rich (Berg and Wiebe, 1985). The xenolith is a cordierite–plagioclase–biotite–spinel–ilmenite–pyrrhotite gneiss with ovoids, 1–3 cm in diameter, consisting of a mixture of fine-grained and granular cordierite, olivine, spinel, orthopyroxene, plagioclase, ilmenite, and pyrrhotite enclosed by poikilitic grains of garnet and the sodium gedrite. Biotite occurs both as poikilitic grains and as inclusions in garnet and sodium gedrite. The textures indicate a two-stage contact metamorphic history in which the granular minerals that formed during the first stage reacted to produce the poikilitic minerals during the second stage, the simplified reaction being:

cordierite + olivine + spinel + plagioclase + orthopyroxene

→ garnet + sodium gedrite

Distribution-coefficient data for mineral pairs from four

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[1] Amphibole nomenclature is based on Leake (1978) where sodium gedrite has Na ≥ 0.75.

different ovoids suggest that all of the minerals in the simplified reaction, except possibly orthopyroxene, ultimately reached equilibrium (Berg and Wiebe, 1985). The good agreement between the phase relationships in the ovoids and the experimental study of the almandine-pyrope-water system (Hsu and Burnham, 1969) led Berg and Wiebe (1985) to conclude that the ovoids originally were garnet porphyroblasts which were decomposed during the first-stage contact metamorphism. Berg and Wiebe (1985) infer that pressure was about 2 ± 1 kbar, fO_2 was between MW and FMQ, and temperature was about 800°C for the first stage and 615°C for the second stage of contact metamorphism.

Textural relations and optical properties

The sodium gedrite occurs as large poikilitic masses (~ 1.5 mm in diameter) composed of many smaller poikilitic crystals (~ 0.2 mm in diameter). The crystals in each mass are within only a few degrees of being in optical continuity throughout the mass. The crystals are riddled with small inclusions, principally of olivine and spinel, but locally of cordierite and hypersthene.

The sodium gedrite is pleochroic with X = pale tan, Y = olive-gray, and Z = bluish gray. It has a positive sign, and the estimated $2V = 65°$. The refractive indices determined by the dispersion method are $\alpha = 1.6768(\pm 0.0008)$, $\beta = 1.6808(\pm 0.0004)$, $\gamma = 1.6948(\pm 0.0009)$, and the birefringence is 0.0180. Most of the variation in the refractive index determinations is due to moderate compositional variation. The optical properties were determined on sodium gedrite from one of the more iron-rich ovoids (ovoid 1 of Berg and Wiebe, 1985).

Mineral chemistry

More than 175 electron microprobe analyses of the sodium gedrite have been obtained. Analytical conditions were similar to those described by Berg and Wiebe (1985). Table 1 lists 10 representative analyses along with structural formulae determined on the basis of 23 oxygens. In calculating the structural formulae, tetrahedral Si and Al were constrained to equal eight, and, where necessary, Na was assigned to M4 in order to bring the cation total for the M1, M2, M3, and M4 sites to seven. If the cation total for the M1, M2, M3, and M4 site was in excess of seven, none

Table 1. Representative electron microprobe analyses of sodium gedrite

	1	2	3	4	5	6	7	8	9	10
SiO_2	40.95	41.45	40.21	39.45	39.96	38.38	38.57	37.05	38.85	37.74
Al_2O_3	14.28	15.13	17.00	17.96	16.79	18.13	18.00	19.07	16.98	16.26
TiO_2	0.06	0.26	0.08	0.13	0.05	0.18	0.09	0.09	0.25	0.23
Cr_2O_3	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	0.04	0.04
FeO*	27.07	28.77	22.20	25.05	24.85	28.75	27.00	26.80	28.67	28.76
MnO	--	0.29	0.10	0.18	0.59	0.32	0.25	0.22	0.65	0.77
MgO	10.60	10.03	13.57	12.26	12.81	9.45	10.12	9.82	8.67	9.60
CaO	0.32	0.26	0.41	0.46	0.45	0.25	0.35	0.37	0.25	0.28
Na_2O	1.96	2.25	2.34	2.50	2.62	2.67	2.73	2.81	3.32	3.13
F	1.62	1.76	1.77	1.53	1.73	1.34	1.39	1.39	n.d.	n.d.
Total	96.18	99.46	96.93	98.88	99.12	98.91	97.91	97.03	97.68	96.81
Si	6.361	6.281	6.078	5.921	6.004	5.871	5.912	5.743	5.992	5.904
Al(IV)	1.639	1.719	1.922	2.079	1.996	2.219	2.088	2.257	2.008	2.096
Al(VI)	0.974	0.981	1.103	1.098	0.975	1.139	1.165	1.229	1.081	0.905
Ti	0.004	0.026	0.009	0.013	0.004	0.018	0.009	0.009	0.029	0.027
Cr	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	0.005	0.005
Fe*	3.516	3.643	2.804	3.141	3.121	3.676	3.459	3.472	3.700	3.765
Mn	--	0.035	0.009	0.022	0.075	0.040	0.031	0.027	0.084	0.102
Mg	2.453	2.264	3.057	2.740	2.865	2.154	2.313	2.271	1.995	2.240
Ca	0.050	0.040	0.066	0.070	0.070	0.040	0.054	0.059	0.042	0.046
Na	0.588	0.658	0.683	0.727	0.758	0.790	0.810	0.842	0.993	0.951
Total Cations	15.585	15.647	15.731	15.811	15.868	15.857	15.841	15.909	15.929	16.041
F	0.793	0.840	0.843	0.727	0.820	0.642	0.675	0.678	n.d.	n.d.
Na(A)	0.585	0.647	0.683	0.727	0.758	0.790	0.810	0.842	0.929	0.951
Na(A)/Al(IV)	0.357	0.376	0.355	0.350	0.380	0.371	0.388	0.373	0.463	0.454
X_{Mg}	0.411	0.383	0.522	0.466	0.479	0.369	0.401	0.395	0.350	0.373
X_F	0.397	0.420	0.422	0.364	0.410	0.321	0.338	0.339	n.d.	n.d.

*No ferric-iron calculation has been attempted.
Cl was analyzed for but not detected in samples for which F was analyzed.
K_2O is below the detection limits in all cases. X_{Mg} = Mg/(Mg + Fe). X_F = mol. fraction of F in O3 sites.

of these cations (Al[VI], Ti, Mg, Fe, Mn, Ca) were assigned to the A site. Instead, it was assumed that some of the total Fe, all of which is listed as Fe^{2+} in Table 1, was actually Fe^{3+}. Seventy-five percent of all the gedrite analyses in this study had excess cations tentatively assigned to the M1, M2, M3 and M4 sites. The mean cation total for these sites was 7.035. Forcing the cation total for these sites to equal 7.000 requires that the average gedrite contain about 0.140 Fe^{3+} in the structural formula (~ 1.2 wt.% Fe_2O_3). This would be a minimum value for Fe^{3+}, and conversion of additional Fe^{2+} to Fe^{3+} would require the movement of Na into M4. Because of the uncertainties in estimating Fe^{3+}, the analyses are presented in Table 1 with all Fe as Fe^{2+}. Also, sodium assigned to the A site must be considered a maximum value, because the conversion of large amounts of Fe^{2+} to Fe^{3+} would ultimately require reassigning Na from the A site to M4, as described above. By analogy with wet chemical analyses of gedrites summarized in Robinson et al. (1982) and because of the intermediate $f(O_2)$ estimated for the crystallization of this sodium gedrite, the actual amounts of Fe^{3+} and Na(M4) are thought to be trivial.

The most Na-rich analysis contains 0.993 Na(total); however the analysis with the most Na(A) contains 0.951 Na(A) (Table 1). Except for Na-rich gedrites recently described by Schumacher (1980) and Otten (1984), previously reported gedrite analyses do not exceed 0.658 Na(A) (Spear, 1980). Schumacher (1980) reported orthoamphibole compositions with Na(A) contents comparable to those reported here; however those high Na contents occur only in orthoamphiboles whose Al(IV) far exceeds the typical limit of 2.0 for normal gedrite (Leake, 1978). Otten (1984) reported a single gedrite composition with an Na(A) content of 0.883.

Only 25 analyses of gedrites contain results for F and Cl, and some of these are listed in Table 1. Although Cl is not detectable in most cases (maximum is 0.02 wt.%), these sodium gedrites contain far more F than any other orthoamphibole ever reported. The gedrite with the highest F content reported here has 2.03 wt.% F and 0.985 F in the structural formula ($X_F = F/(F + OH) = 0.493$), whereas the highest F content reported previously from a gedrite is 0.064 (0.13 wt.%; Seki and Yamasaki, 1957), and from any orthoamphibole is 0.237 (0.52 wt.%; Sundius, 1933). The average sodium gedrite from Zoar has 0.724 F in the structural formula (about 1.5 wt.% F and $X_F = 0.362$). Because of the low count rates, the F results are subject to greater uncertainty than the other elements. Thus it is not surprising that there are no obvious correlations between F and other elements in the limited number of analyses and limited compositional range of the Zoar gedrites. Although grain compositional variation was not mapped, it was observed to be patchy and probably like that shown in Figure 91 of Robinson et al. (1982).

Variations in gedrite crystal chemistry

Because of the unusually high Na(A) content of these gedrites and the emerging evidence for a more extensive range of Na(A) content in gedrite, the data on the Zoar gedrites, as well as gedrites from the literature, have been examined for insights into the factors affecting Na(A)-site occupancy in gedrite.

Coupled-substitution relationships

A plot of maximum A-site occupancy vs. tetrahedral Al is presented in Figure 1. Also identified in this diagram is the "ideal gedrite" end-member composition of Robinson et al. (1971). As with most amphiboles, these data show a moderately strong correlation between A-site occupancy and Al(IV). This is due in part to the edenite coupled substitution as described by Robinson et al. (1971):

$$Na^A + Al^{IV} = \square^A + Si^{IV}$$
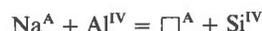
Al(IV) also enters the structure by the tschermakite (Ts) coupled substitution:

$$Al^{VI} + Al^{IV} = Mg^{VI} + Si^{IV}$$
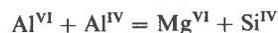
A strong correlation between A-site occupancy and Al(IV) indicates a constant ratio between the two substitutions. Based on the data available at the time, Robinson et al. (1971) suggested that in typical orthoamphiboles the two substitutions occur in a nearly fixed Ed:Ts ratio of 1:3, edenite (ED): tschermakite (Ts), leading to the "ideal gedrite" composition with a half-filled A-site where Al(IV) = 2.0 (Fig. 1). The Zoar data, however, suggest an Ed:Ts ratio much closer to 1:1.5. These data indicate that there is no ideal gedrite end member and that the relative proportions of the two coupled substitutions, Ed and Ts, can vary considerably.

To illustrate further these conclusions, the compositional fields of gedrites from seven other geographical areas are presented in Figure 2, and these clearly show that there is a wide range of Ed:Ts ratios (~ 0.1–1) or A/Al(IV) (~ 0.5–0.1). Although gedrites from each area show a correlation of A with Al(IV), each area has a different slope, A/Al(IV). Therefore, A-site occupancy does not appear to be as important as A/Al(IV) in discriminating between suites. The Zoar gedrites represent the highest proportion of edenite substitution to tschermakite substitution, whereas those from southern Norway represent the lowest. Thus it is evident from Figure 2 that for any given gedrite crystal or suite of gedrites, there must be a variable, or set
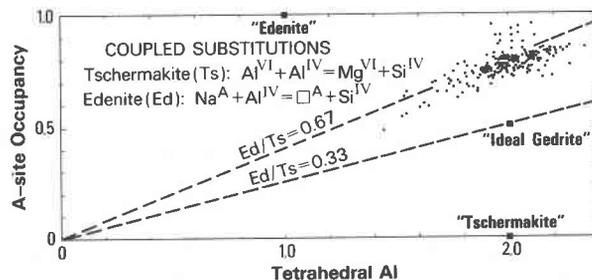
Fig. 1. Maximum A-site occupancy vs. tetrahedral Al for gedrites from near Zoar, Labrador.
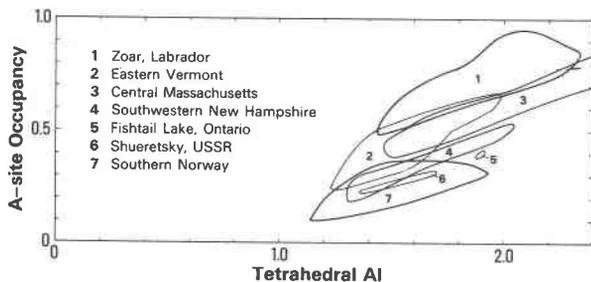
Fig. 2. A-site occupancy vs. tetrahedral Al for gedrites from seven different regions. Sources of data: (1) this paper; (2) Spear, 1980; (3) J. S. Schumacher in Robinson et al., 1982; (4) Robinson and Jaffe, 1969; (5) Lal and Moorhouse, 1969; (6) Deer et al., 1963; (7) Beeson, 1978.

of variables, that systematically control the proportion of edenite substitution to tschermakite substitution.

Effect of X_{Mg}

Spear (1980) has shown that the gedrites from eastern Vermont display an inverse correlation between A-site occupancy and X_{Mg}. A similar, but very weak, correlation is evident in the gedrite data from Labrador. Because of the above arguments against using A-site occupancy as a discriminant, A/Al(IV) vs. X_{Mg} is plotted in Figure 3 for gedrites represented in Figure 2, as well as other gedrites selected from the literature. Despite the wide range of environments and mineral assemblages from which these gedrites derive, there is a moderate correlation. The data from Kamineni (1975) show unusual stoichiometries for ge-
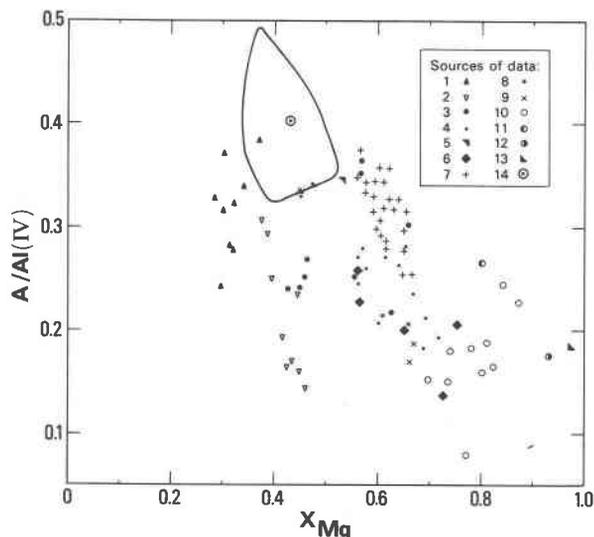
Fig. 3. A/Al(IV) vs. X_{Mg} for gedrites from different regions. Line encloses field of Zoar gedrites. Sources of other data: (1) Abraham and Schreyer, 1973; (2) Kamineni, 1975; (3) Stout, 1972; (4) Spear, 1980; (5) James et al., 1978; (6) Robinson and Jaffe, 1969; (7) Schumacher, pers. comm., 1983; (8) Lal and Moorhouse, 1969; (9) Deer, Howie, and Zussman, 1963; (10) Beeson, 1978; (11) Papike and Ross, 1970; (12) Rabbitt, 1948; (13) Zotov and Siderenko, 1967; (14) Otten, 1984.

drites, e.g., the M sites sum to ~6.5, which may reflect analytical problems. If these data are deleted the correlation is slightly improved.

Robinson et al. (1982) suggested that such a correlation could result from a slight collapse of the M1-3 sites due to increasing Mg contents. As Mg replaces Fe^{2+} in the M1 and M3 sites, these sites would collapse somewhat, which might cause a concomitant shrinking of the M2 site. This would of course favor the smaller Al over Mg in the M2 site, or in other words, the tschermakite substitution would be favored. Conversely, replacement of Mg by Fe^{2+} in the M1-3 sites would result in an expansion of these sites. Such an expansion might change the shape of the A site. In Mg-rich orthoamphiboles the A site has a rather tightly coordinated configuration which is essentially six-coordinated (Papike and Ross, 1970). This change in shape might increase the coordination of the A site resulting in easier accommodation of Na, and thus favoring the edenitic substitution.

Other factors that might affect the ratio of the two coupled substitutions include the mineral assemblage, pressure, and temperature. In evaluating all of the gedrites presented in Figures 2 and 3, there is no strong correlation between assemblage and A/Al(IV). However, because there is a slight correlation between assemblage and X_{Mg}, there is a very crude secondary correlation between assemblage and A/Al(IV). The lack of reliable pressure–temperature data for a number of gedrites with approximately the same X_{Mg} makes it virtually impossible to evaluate the effects of P and T. An indication of how unimportant pressure, temperature, and mineral assemblage appear to be relative to other factors can be seen from a comparison of gedrites from Zoar with those from the contact aureole of the Brocken Massif in the Harz Mountains, Germany (Abraham and Schreyer, 1973). The Brocken Massif gedrites have a paragenesis virtually identical to those in the Zoar sample, and they probably crystallized at a similar temperature and pressure (Berg and Wiebe, 1985). Yet the Brocken Massif gedrites have significantly lower A/Al(IV) ratios than the Zoar gedrites (Fig. 3).

Effect of fluorine

It is obvious from Figure 3 that X_{Mg} cannot be used to explain the differences in A/Al(IV) between the Zoar and Brocken Massif gedrites. The major difference between these two suites of gedrites appears to be the fluorine content. Although F was not analyzed in the Brocken Massif gedrites, a low F content can be inferred from the X_{Mg}'s of the volatile-bearing minerals. Berg and Wiebe (1985) have shown that high F content can shift the compositions of volatile-bearing minerals, in this case gedrite and especially biotite, to higher X_{Mg}'s due to the Fe^{2+}–F avoidance (Rosenberg and Foit, 1977). Within analytical error the volatile-free minerals in the Brocken Massif sample are identical in composition to those in the Zoar sample, but the Brocken Massif gedrites are somewhat more Fe-rich and the biotite is much more Fe-rich than their counterparts from Zoar (Berg and Wiebe, 1985). Hence, the F

content of the Brocken Massif gedrite, although unspecified, is inferred to be low. The high F content of the Zoar gedrites apparently shifts them to higher A/Al(IV) values relative to the Brocken Massif gedrites and to the general trend of all gedrites (Fig. 3).

Unfortunately, very few reliable gedrite analyses with fluorine determinations exist. The only modern analyses with fluorine determinations, where contamination does not appear to be a problem, are those reported by Robinson and Jaffe (1969). These F contents are all too low to permit the definition of any trend, and one is left with only the shift of the Zoar gedrites to high A/Al(IV) relative to the Brocken Massif gedrites, as the evidence for the fluorine effect.

Because structural studies of F-bearing orthoamphiboles do not exist, consideration of fluorine effects must be drawn from relevant work on clinoamphiboles. Fluorine substitutes for the hydroxyl anion in the O3 site, which is shared by two M1 and one M3 cations. In OH-bearing amphiboles the proton in the hydroxyl group is repelled away from the octahedral strip toward the A site (Appleman et al., 1966; Hawthorne, 1981). In clinoamphibole the hydrogen repels the A-site cation off the special position (Appleman et al., 1966; Docka et al., 1980). The deviation of the cation off the A-site special position is limited by the coordinating oxygens bonded to the T sites (J. A. Docka, pers. comm., 1983). Because the coordination number of the A site in orthoamphibole is much smaller than in clinoamphibole, it is possible that the hydrogen might actually inhibit the entrance of a cation such as Na into the A site. The substitution of F for OH would eliminate this effect, and would thus serve to favor or enhance the edenite substitution.

Another fluorine effect involves relations restricted to the octahedral strip. Fe^{2+} normally orders into M1 and M3 in preference to M2 of hydroxyl-bearing amphiboles (Hawthorne, 1981; Cameron and Papike, 1979). However, if F substitutes for OH in the O3 site, Fe^{2+} apparently will order into the M2 site in preference to M1 and M3 (Hawthorne, 1983). This happens because of the Fe^{2+}–F avoidance and the fact that M2 has no O3 ligands and M1 and M3 each have two. The net result is that some Mg will be displaced by Fe^{2+} in the M2 site. Based on the evidence that the tschermakite substitution involves Mg in the M2 site (Spear, 1980), that substitution will likely be retarded. Thus the substitution of F for OH in gedrite is likely to both favor the edenite substitution and inhibit the tschermakite substitution resulting in a significant increase in the A/Al(IV) relative to F-free gedrite. A better understanding of the controls of gedrite crystal chemistry must await the determination of fluorine contents in a broad spectrum of gedrites. Further structural and chemical studies elucidating the detailed nature of the tschermakite substitution are also needed.

Acknowledgments

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