Composition of synthetic tremolite-tschermakite solid solutions in amphibole + anorthite- and amphibole + zoisite-bearing assemblages

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ABSTRACT

The composition of synthetic amphiboles was investigated experimentally along the tremolite-tschermakite join in the system CaO-MgO-Al2O3-SiO2-H2O-Br2. Compositions of these amphiboles were studied within the phase assemblages: amphibole-anorthite-quartz-diopside (I), amphibole-anorthite-quartz-talc (II), amphibole-anorthite-quartz-enstatite (III), amphibole-anorthite-talc-clinochlore (IV), amphibole-zoisite-talc-quartz (V), and amphibole-zoisite-talc-clinochlore (VI). Assemblages were synthesized from oxide-hydroxide mixtures in the presence of a CaBr2-bearing solution between 600–800 °C and 200–2000 MPa. Solid phases were investigated using SEM, HRTEM, EMP, and XRD techniques. EMP data show that the amphiboles produced are solid solutions of the ternary system tremolite-tschermakite-cummingtonite. Enstatite, diopside, talc, and clinochlore showed small deviations from their respective end-member compositions due to incorporation of some Al.

The thermodynamic properties of the tschermakite end-member and the mixing properties along the tremolite-tschermakite join were extracted from corresponding exchange reactions of the un-reversed synthesized phase assemblages I–VI. Various ideal mixing models were tested for Al-Mg and Al-Si substitution at octahedral M2 and M3 sites and at tetrahedral T1-sites. Best fits were obtained for a two-site coupled model, resulting in $\Delta H_m^o = -12528.3 \pm 11.7$ kJ/mol and $S_m^o = 556.5 \pm 12.0$ J/(mol·K) for the tschermakite end-member. Similar calculations were carried out for magnesiohornblende, and values of $\Delta H_m^o = -12418.7 \pm 5.9$ kJ/mol and $S_m^o = 562.8 \pm 6.1$ J/(mol·K) were extracted. Calculated phase relations and amphibole compositions agree well with experimental data if the derived thermodynamic data of tschermakite and a two-site mixing model for Al incorporation in amphibole solid solutions are applied.

INTRODUCTION

In greenschist- to granulite-facies rocks, Al-bearing calcic amphiboles are common. They occur either in metabasites (e.g., Robinson et al. 1982), metamorphosed marls (e.g., Baker and Matthews 1994) or in calc-alkaline granitoids (e.g., Hammarstrom and Zen 1986; Hollister et al. 1987). In calc-alkaline granitoids, the Al content of amphibole commonly increases with depth. This has been used to derive an empirical “Al in hornblende” geobarometer (Hammarstrom and Zen 1986; Hollister et al. 1987; Johnson and Rutherford 1989; Thomas and Ernst 1990; Schmidt 1992). The Al content of amphibole, however, is not only a function of pressure and temperature but also depends on bulk composition as given by a particular mineral assemblage. This complicates using the Al content in amphibole as a simple petrogenetic indicator. Léger and Ferry (1991), for example, demonstrated that calcic amphiboles at the same metamorphic grade have highly variable Al contents of 0.4 to 3.3 Al\textsuperscript{4+} apfu due to the presence of different Al-bearing minerals.

Because of its suitable ionic size (IV)Al\textsuperscript{4+} = 0.39 Å, (VI)Al\textsuperscript{3+} = 0.54 Å; Shannon 1976) Al\textsuperscript{4+} enters various tetrahedral and octahedral sites in the amphibole structure. The octahedral M2 site and the tetrahedral T1 site are the most probable locations (Hawthorne 1983; Makino and Tomita 1989), but the M3 site (Oberti et al. 1995a; Welch and Knight 1999) and the T2 sites (Oberti et al. 1995b; Welch et al. 1998) have also been suggested for Al incorporation in pargasite. Jenkins et al. (1997) considered the incorporation of Al\textsuperscript{4+} at all octahedral (M1–M3) and tetrahedral sites (T1–T2). In synthetic amphiboles along the tremolite-tschermakite join, incorporation of octahedral Al\textsuperscript{4+} at M2 and M3 sites and of tetrahedral Al\textsuperscript{4+} at T1 sites has been discussed in recent studies (Hawthorne et al. 2000; Najorka and Gottschalk in preparation).

Using Al-free tremolite (Ca\textsubscript{2}Mg\textsubscript{5}(Si\textsubscript{8}O\textsubscript{22}(OH)\textsubscript{2})) as a reference, Al is incorporated as a coupled substitution represented by the tschermaks AlAl(SiMg)\textsuperscript{1-} exchange vector. The thermodynamic properties of Al-bearing amphibole, i.e., its Gibbs free energy, depend on the intracrystalline Al-distribution at the different tetrahedral and octahedral sites. It is clear that the Al concentrations in calcic amphiboles could be used more effectively in geothermobarometry if the thermodynamic properties of the amphibole end-members, their mixing properties and particularly the distribution of Al\textsuperscript{4+} between different crystallographic sites were better known.

There have been many experimental studies on the tschermaks substitution in amphibole, its respective phase equi-