Hydrothermal synthesis of amphiboles along the tremolite-pargasite join and in the ternary system tremolite-pargasite-cummingtonite

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ABSTRACT

We report here an investigation of amphibole synthesis within the ternary system Ca$_2$Mg$_5$Si$_8$O$_{22}$(OH)$_2$-NaCa$_2$Mg$_4$Al$_3$Si$_6$O$_{22}$(OH)$_2$-Mg$_7$Si$_8$O$_{22}$(OH)$_2$ (= TR-PG-MC) to define better the limits of solid solution for amphiboles formed along and near the join tremolite-pargasite. Hydrothermal syntheses were conducted in the range of 750–1000 °C and 1–6 kbar. Syntheses at 10 mol% compositional intervals along the TR-PG join produced incomplete yields of amphibole, and the resultant amphiboles were found to be associated with varying amounts of accessory clinopyroxene, plagioclase, and gehlenite. Syntheses of end-member pargasite with progressively greater amounts of the MC component produced a very good amphibole yield for a pargasite composition containing 2.5 mol% MC. The persistence of clinopyroxene or gehlenite and the improved yield of pargasitic amphibole with minor addition of the MC component indicate a shift toward MC enrichment for amphiboles made from bulk compositions directly on the TR-PG join. Syntheses within the TR-PG-MC ternary system suggest that the Ca-rich limit of solid solution is a slightly curved field in the TR-PG-MC field. Amphiboles made along this join showed a very systematic change in the (151) and (3–31) peak spacing with mol% PG, providing a simple technique for deducing the composition of amphiboles made near the tremolite-pargasite join. Electron microprobe analysis confirms that the amphiboles for the most part conform to the “pargasite” substitution. The one exception being amphiboles formed near 80–90 mol% PG compositions, which proved both difficult to synthesize (~75 wt% amphibole yield) and displayed a small, but significant, excess of Na in their structure. High yields of amphibole could be formed from this same bulk composition with the use of a dilute NaOH solution instead of pure water, suggesting that there was insufficient Na in the starting mixture to allow complete amphibole formation. Structural refinements using X-ray diffraction (XRD) Rietveld refinement confirmed the high Na content for this sample and also revealed a non-linear trend in the unit-cell volume with composition, with a maximum at the 80 mol% PG bulk composition. The difficulty in synthesizing amphiboles near this bulk composition and its unusually high volume suggest that immiscibility in natural hornblendes may initiate with increasing pressure for pargasite-rich bulk compositions.

INTRODUCTION

The join tremolite [Ca$_2$Mg$_5$Si$_8$O$_{22}$(OH)$_2$ = TR]-pargasite [NaCa$_2$Mg$_4$Al$_3$Si$_6$O$_{22}$(OH)$_2$ = PG] provides a useful first approximation to the (admittedly complex) chemical variation that occurs between actinolite and hornblende, as shown by the amphibole chemistry plots from many meta-mafic rock localities (e.g., Kamineni 1986; Schumacher 1991; Gillis et al. 1993; Lee and Cho 1995). Both end members of this join have been the focus of research in recent years. Tremolite is one of the chemically simplest end-members of the calcic amphiboles and yet a mineral of this ideal composition is not commonly found in nature and has proven difficult, if not impossible, to synthesize (see recent reviews by Maresch et al. 1994; Hawthorne 1995; Yang and Evans 1996; Zimmermann et al. 1996). Cation distribution in synthetic pargasite has been studied recently by Welch et al. (1994) using NMR and IR techniques and by Oberti et al. (1995) using single-crystal XRD analysis. Pargasite has not been so problematic as tremolite to synthesize (e.g., Raudsepp et al. 1991), but it also may display some deviation from its ideal composition as discussed below.

There have been two previous studies devoted to the (Fe-free) tremolite-pargasite join, which were both by Oba (1980, 1990). These studies have focused on establishing the temperature-composition phase equilibria along this join at various pressures. In addition to confirming the higher thermal stability of pargasite relative to tremolite, Oba (1980) reported the presence of an extensive miscibility gap existing along this join at 1 kbar but not at 5 kbar. A second investigation of this join at 10 kbar (Oba 1990) again showed complete miscibility, even at a temperature (800 °C) that was well inside the miscibility gap at 1 kbar. If these results represent true equilibrium, supported by one very long duration (4280 h) compositional re-equilibration experiment at 800 °C and 1 kbar, then one would anticipate immiscibility in hornblendes to be present at pressures below 5 kbar but not above. This pressure

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