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Cation ordering in synthetic low-calcium actinolite

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ABSTRACT

A series of low-Ca (1.8–1.65 atoms Ca per formula unit) actinolites were synthesized in the system CaO-FeO-MgO-SiO_2-H_2O with bulk compositions of 0, 10, 20, and 30 mol% ferro-actinolite when projected onto the tremolite–ferro-actinolite join for the purpose of determining the cation site occupancies as a function of bulk composition. Syntheses were done by multiple treatments of the oxide-metal mixtures in internally heated gas vessels at 600–800 °C, 6 kbar, f_{O_2} near the Co-CoO buffer, which is sufficiently low to prevent the appearance of Fe^{3+} in this study, and for total durations up to 774 hours. Bulk compositions of the synthesis products were assessed by electron microprobe analysis (EMPA), analytical transmission electron microscopy (AEM), and by using unit-cell dimensions (CD method), whereas the site occupancies were assessed using powder X-ray diffraction (XRD) Rietveld refinements, Fourier-transform infrared (FTIR) spectroscopy, and Mössbauer spectroscopy. Selected-area electron-diffraction (SAED) patterns and high-resolution transmission electron microscopy (HRTEM) images were obtained to study the structure and defects of the samples. A maximum of about 2% non-amphibole chain-multiplicity faults (CMFs) were observed in the sample with 10 mol% ferro-actinolite component, indicating the strong reduction in CMFs that occurs with only a small addition of ferro-actinolite to tremolitic amphiboles. This study, as with earlier studies, found no significant preferential partitioning of Fe and Mg relative to the bulk Fe content of the sample at the M1 and M3 sites, weak partitioning of Mg relative to Fe into the M2 site, and strong partitioning of Fe relative to Mg at the M4 site. No ferric iron was detected. The partitioning of Fe and Mg at the M4 site was modeled by a simple ideal-activity, two-site exchange reaction using the actinolites from this study and those of previous studies with bulk Fe/(Fe + Mg) ≤ 0.3 and with low Al, Mn, and Na contents using 1 bar and 298K values of $\Delta G° = 62$ kJ and $\Delta S° = 54$ J/K. The FTIR data were analyzed for the presence of short-range order of Fe and Mg at the M1 and M3 sites but none was found.