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Experimental study of Cr incorporation in pargasite

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ABSTRACT

Pargasite compositions corresponding to the nominal structural formula $NaCa_2(Mg_4Al_{1-x}Cr_x)(Si_6Al_2)O_{22}(OH)_2$, with $0 \leq x \leq 1$, were investigated at 900 °C and 3 and 20 kbar P_{H_2O}. The aims of this work were to determine the extent of the solid solution, i.e., the maximum x value, as a function of experimental conditions, and to characterize the M^{3+} (Al^{3+} and/or Cr^{3+}) distribution over the M sites. A first series of experiments at 900 °C and 3 kbar showed the presence of large amounts of Cr^{6+} as sodium chromate (Na_2CrO_4) and dichromate ($Na_2Cr_2O_7$) in the final fluid phase, which promotes the extraction of Cr from the bulk solid (up to 45 mol%). The oxidizing agent was the oxygen from water, liberated by diffusion of hydrogen from water through the Pt tube wall. To prevent this oxidation, a new double-chamber tube was designed, one chamber containing powdered metallic Cr to trap the excess of oxygen and the second chamber initially containing the starting gel and water. Using this tube, (chromium)-pargasites were produced with a maximum Cr content around 0.43 apfu from EMP analyses (based on 23 O atoms), close to the maximum Cr solubility observed in naturally occurring pargasites. In FTIR spectra of Cr-free pargasite, two intense OH-stretching bands are observed at 3714 and 3681 cm^{-1}, corresponding to OH groups adjacent to Mg_3 and to Mg_2Al, respectively, and pointing toward a filled A-site. A third band is observed at 3656 cm^{-1}, which can be assigned to OH groups pointing toward vacant A-sites. Along the Al-Cr pargasite join, significant modifications are observed in the OH-stretching region: a new band appears at 3660 cm^{-1}, which can be assigned to OH groups adjacent to Mg_2Cr, and pointing toward a filled A-site. Its relative intensity increases regularly with the Cr content, showing that Cr^{3+} enters M site(s) adjacent to an OH group. Combining EMP and FTIR data, we conclude that up to 0.27 Cr per formula unit occupies the M3 site. However, these are more octahedral trivalent cations than expected, and these are balanced by lower tetrahedral Al and lower occupancies of the A and M4 sites. This charge arrangement, with important deviations from ideal stoichiometry, is apparently the only stable one under the conditions applied. Results were confirmed at 900 °C and 20 kbar P_{H_2O}, indicating that increasing pressure does not affect Cr solubility in pargasite.