American Mineralogist, Volume 87, pages 132–141, 2002

Mineralogical and Mössbauer spectroscopic study of a diopside occurring in the marbles of Andranondambo, southern Madagascar

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ABSTRACT

Dark pyroxene crystals found in a calcite vein cutting across marbles and skarns at Andranondambo, southern Madagascar, were investigated with various techniques in order to characterize and identify the mineral phase. Methods applied include powder X-ray diffraction, thin-section microscopy, electron-microprobe analysis, densimetry, and thermogravimetry coupled to mass spectrometry (TGA-MS), FTIR, and Mössbauer spectroscopy. The chemical composition of the mineral phase was determined as $\{Ca_{0.98},Na_{0.03}\}[Mg_{0.68},Fe^{2+}_{0.07}Fe^{3+}_{0.03},Al_{0.16},Ti_{0.04}](Si_{1.77},Al_{0.23})O_6$. The refined unit-cell parameters are: $a = 9.719(2)$ Å, $b = 8.838(2)$ Å, $c = 5.287(1)$ Å, $\beta = 106.05(2)°$, and $V = 436.33(6)$ Å^3. The refractive indices are: $\alpha = 1.690 \pm 0.002$, $\beta = 1.700 \pm 0.002$, and $\gamma = 1.712 \pm 0.002$. The density is 3.370 (55) g/cm^3. The mean refractive index calculated from the chemical composition yields a Mandarino compatibility index of $K = -0.0167$. From the various analytical data, we were able to conclude that the mineral corresponds to Fe- and Ti-bearing aluminian diopside. TGA-MS data suggest the presence of calcite solid inclusions and minor H_2O and CO_2 fluid inclusions. Chemical analyses of the calcite host matrix revealed clear rare earth element enrichment. Mössbauer spectra obtained at varying temperatures show the presence of Fe^{2+} and Fe^{3+}. Both exhibit broad quadrupole-splitting distributions, indicating widely fluctuating chemical and structural environments for the probe Fe nuclei. The broad temperature range (80–470 K) over which the spectra were collected resulted in a precise determination of the Fe^{2+}/Fe_{tot} ratio as 0.68, which is consistent with the chemical composition as determined from the microprobe analyses. For comparison, the Mössbauer spectra of two specimens of the type locality of aluminian diopside (the mineral formerly named "fassaite") were analyzed as well and found to be consistent with those obtained for the Madagascar crystals. The relation between mineral paragenesis and local geological features is briefly discussed.