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Spectroscopic studies of synthetic and natural ringwoodite, γ-(Mg$_{1-x}$,Fe$_x$)$_2$SiO$_4$

M. Taran (1), M. Koch-Müller (2), R. Wirth (2), I. Abs-Wurmbach (3), D. Rhede (2), and A. Greshake (4)

(1) Institute of Geochemistry, Mineralogy and Ore Formation, National Academy of Science of Ukraine, Optical spectroscopy and luminescence of minerals, Kiev, Ukraine (m_taran@hotmail.com, 00380444241270), (2) Deutsches GeoForschungsZentrum, Telegrafenberg, 14473 Potsdam, Germany (mkoch@gfz-potsdam.de / 0049 331 288 1402) , (3) Institut für Angewandte Geowissenschaften, TU Berlin, Ernst-Reuter-Platz 1, 10587 Berlin, Germany (irmgardabs-wurmbach@alice-dsl.net / 00493031472218), (4) Humboldt-Universität zu Berlin, Museum für Naturkunde, 10099 Berlin, Germany (ansgar.greshake@museum.hu-berlin.de / 00493020938565)

Synthetic ringwoodite γ-(Mg$_{1-x}$,Fe$_x$)$_2$SiO$_4$ of x = 0.4 to 1.0 compositions and variously colored micro-grains of natural ringwoodite in shock metamorphism veins of thin sections of two S6-type chondrites were studied by means of microprobe analysis, TEM and optical spectroscopy. In addition, three synthetic samples were studied by Mössbauer spectroscopy. The spectra consist of two doublets caused by ^{VI}Fe^{2+} and ^{VI}Fe^{3+}, with IS and QS parameters $\sim$ 1.06 and $\sim$ 2.76 mm/sec and $\sim$ 0.25 and $\sim$ 0.38 mm/sec, respectively, close to those established elsewhere. The Fe^{3+}/Fe$_{total}$ ratio ranges from 0.04 to 0.1. Optical absorption spectra of all synthetic samples qualitatively are very similar being directly related to the iron content and differing mostly in the intensity of the absorption features. They consist of a strong high-energy absorption edge and a series of bands of different width and intensity. The three strongest and broadest absorptions are attributed to split electronic spin-allowed ^{5}T$_{2g}\rightarrow^5$E$_g$ transition of ^{VI}Fe^{2+} and to intervalence charge-transfer (IVCT) transition between ferrous and ferric ions in adjacent octahedral sites of the structure. The spin-allowed absorption bands at ca. 8000 and 11500 cm^{-1} weakly depend on temperature thus poorly matching the idea of their intensification by Fe^{2+}-Fe^{3+} exchange coupling mechanism (Keppler and Smyth 2005). The IVCT band at $\sim$ 16400 cm^{-1} displays strong temperature dependence decreasing with temperature and practically disappearing between 397 K and 497 K. Pressure induces a distinct lower-energy shift of the absorption edge. The spin-allowed bands shift to higher energies and apparently decrease in intensity. The IVCT band weakens and finally vanishes at about 9 GPa. We assigned this effect to pressure-induced reduction of Fe^{3+}. By analogy with synthetic samples three broad bands in spectra of natural (meteoritic) blue ringwoodite are assigned to electronic spin-allowed transitions of Fe^{3+} ($\sim$ 8600 cm^{-1} and $\sim$ 12700 cm^{-1}) and Fe^{2+}/Fe^{3+} IVCT transition ($\sim$ 18100 cm^{-1}), respectively. Spectra of colorless ringwoodite consist of a single broad band at ca. 12000 cm^{-1}. It is assumed that such ringwoodite grains are inverse (Fe, Mg)$_2$SiO$_4$-spinels and the band is caused by the split spin-allowed ^{5}E $\rightarrow^5$T$_2$ transition of ^{IV}Fe^{2+}. Ringwoodite of intermediate color variations between dark-blue and colorless are assumed to be partly inversed ringwoodite. No glassy material between the grain boundaries in the natural colored ringwoodite aggregates was found in our samples and disprove the cause of the coloration to be due to light scattering.

References

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