Spectroscopic studies of synthetic and natural ringwoodites, gamma-(Mg1-xFex)2SiO4

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Synthetic ringwoodite γ-(Mg_{1-x},Fe_x)$_2SiO_4$ with 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. Three synthetic samples were studied in addition with Mössbauer spectroscopy. The Mössbauer spectra consist of two doublets caused by $^{VI}Fe^{2+}$ and $^{VI}Fe^{3+}$, with IS and QS parameters close to those established elsewhere. The Fe^{3+}/Fe_{total} ratio evaluated from the spectra, ranges from 0.04 to 0.1. Optical absorption spectra of all synthetic samples are qualitatively very similar as they are directly related to the iron content differing mostly in the intensity of the observed 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 of them are attributed to splitting of electronic spin-allowed $^5T_{2g}$ ^5E_g transitions of $^{VI}Fe^{2+}$ and 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. This observation is not consistent with the idea of their intensification by exchange coupling mechanism. The IVCT band at ~16400 cm^{-1} displays strong temperature dependence decreasing with temperature and practically disappearing at about 497 K. With pressure the absorption edge shifts to lower energies while the spin-allowed bands shift to higher energy and apparently decreases in intensity. The intensity of the IVCT band decreases with pressure and vanishes at about 9 GPa. We assigned this effect to be due to pressure-induced reduction of Fe^{3+} in ringwoodite.

By analogy with synthetic samples three broad bands in spectra of natural (meteoritic) blue ringwoodite are assigned to electronic spin-allowed transitions of $^{VI}Fe^{2+}$ (~8600 cm^{-1} and ~12700 cm^{-1}) and Fe^{2+}/Fe^{3+} IVCT transition (~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)$_2SiO_4$-spinels and the band is caused by the split spin-allowed ^5E $^5T_2$ 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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