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MÖSSBAUER SPECTROSCOPY OF ELECTRON IRRADIATION EFFECTS IN ORTHOPYROXENES

R.B. Scorzelli, E.B. Saitovitch and J. Danon

Orthopyroxenes (Fe,Mg)$_2$Si$_2$O$_6$ occur commonly as a major constituent of mafic, igneous and metamorphic rocks, being a mineral of great mineralogic and geophysical interest. The orthopyroxene crystal structure provides two non-equivalent octahedrally-coordinated positions, M$_1$ and M$_2$, and the only cations present in substantial quantities at these positions are magnesium and iron (1). The site population observed in natural and synthetic orthopyroxenes can be used as a thermal indicator on the basis of thermodynamic and kinetic arguments (2). Several factors modify the distribution equilibrium in these minerals, such as high pressures arising from shock (meteoritic impact). Radiation effects from particle bombardment (cosmic rays and natural radioactivity) can possibly disturb the equilibrium site population.

In this paper an attempt has been made to determine the influence of a 2 MeV electron irradiation on the orthopyroxene structure. We present the results of a Mössbauer study of the distribution of Fe$^{2+}$ in the irradiated orthopyroxene as a function of the radiation doses and the annealing behaviour of the system.

The studied samples are highly purified natural OPX orthopyroxene with the composition Fe$_{0.55}$Mg$_{0.47}$SiO$_3$. The experiments have been performed with powder samples both source and absorber being kept at 110K. A 2 MeV electron beam had been used for the irradiation experiments according to previous description (3).

The hyperfine spectrum of the orthopyroxene Fe$_{0.88}$Mg$_{0.12}$SiO$_3$(OPX) at 110K consists of: a inner doublet assigned to Fe$^{2+}$ ions in the M$_2$ site (IS=0.90 mm/s $\Delta E_Q=2.08$ mm/s) and the outer one assigned to Fe$^{2+}$ in the M$_1$ site (IS=0.92 mm/s $\Delta E_Q=2.85$ mm/s). The intensities of the inner two peaks are not equal, due to texture effects, the line at lower velocity being the more intense.

The Mössbauer spectrum of the irradiated OPX orthopyroxene shows an alteration compared to the spectrum of the unirradiated sample with the appearance of a third doublet (IS=0.91 mm/s, $\Delta E_Q=2.19$ mm/s), which can be attributed to a species formed at expenses of iron in the M$_2$ site (Table I).

<table>
<thead>
<tr>
<th>(\Delta E_Q=2.85) mm/s</th>
<th>(\Delta E_Q=2.08) mm/s</th>
<th>(\Delta E_Q=2.19) mm/s</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fe$^{2+}$ site</td>
<td>Fe$^{2+}$ site</td>
<td>Fe$^{2+}$ site</td>
</tr>
<tr>
<td>M$_1$ site</td>
<td>M$_2$ site</td>
<td>M$_2$ site</td>
</tr>
<tr>
<td>IS=0.90 mm/s</td>
<td>IS=0.92 mm/s</td>
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<tr>
<td>$\Delta E_Q=2.08$ mm/s</td>
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</tr>
</tbody>
</table>

By keeping the irradiated sample in liquid nitrogen the third doublet has been found to disappear exponentially with the time that has elapsed after the end of the irradiation (see Table I).

Similar results have been found in a XZZ orthopyroxene (Fe$_{0.88}$Mg$_{0.12}$SiO$_3$), which has better resolved doublets for iron in the M$_1$ and M$_2$ sites. In this case the position of the new doublet can be seen in the Mössbauer spectrum as is illustrated in fig. 1.

The effect of electron irradiation on orthopyroxenes differ completely from that of shocks (4,5). When shock is applied to orthopyroxene crystals, a permanent alteration of the cation distribution takes place, meaning that the initial crystal...
structure has been destroyed and that a new phase has been reached.

Our experiments with 2 MeV electron irradiation reveal a different behaviour, namely that there is a complete recovery even at low temperature. This means that irradiation with doses up to 5400 Mrad produces only a transient effect, at least as seen by Mössbauer spectroscopy, connected to a local lattice disturbance.

References