

AN ^{57}Fe MÖSSBAUER STUDY OF THE OLIVINE SOLID SOLUTION SERIES: IMPLICATIONS FOR METEORITE CLASSIFICATION AND DECONVOLUTION OF UNEQUILIBRATED CHONDRITE SPECTRA. O. N. Menzies¹, P. A. Bland¹ and F. J. Berry². ¹Planetary and Space Sciences Research Institute, The Open University, Milton Keynes MK7 6AA, UK, ²Department of Chemistry, The Open University, Milton Keynes MK7 6AA, UK.

Introduction: Mössbauer spectroscopy has been used to characterise and quantify the mineralogy of chondritic meteorites by detecting Fe bound in a crystal lattice [1]. The technique investigates the oxidation state and electronic configuration of Fe atoms in minerals [2]. Mössbauer spectra for meteorites frequently show overlapping or poorly separated absorption peaks for olivine and pyroxene, which are difficult to separate during the fitting process [3]. This is particularly true for studies of unequilibrated chondrites where their absorptions are often treated as a single peak and their mineral abundances considered together [4]. Some discrepancy has also been observed between pyroxene abundances as measured by Mössbauer spectroscopy and X-ray diffraction in carbonaceous chondrites [5]. Pyroxene appears to have been overestimated in samples analysed by the Mössbauer technique, possibly due to overlapping of peaks associated with olivine. The large spread of olivine compositions in unequilibrated chondrites suggests that some of the more fayalitic compositions may have been mis-identified as pyroxene. It was proposed that if one of these components could be isolated, the fitting procedure for meteorites could be improved and more accurate mineral abundances obtained. As well as improving interpretation of meteorite Mössbauer spectra, such a method could also point the way for a rapid method of characterising silicate chemistry and identifying the degree of equilibration in chondrites.

Olivine has a simple chemistry and displays a complete solid solution series from forsterite to fayalite. This study aims to use ^{57}Fe Mössbauer spectroscopy to analyse a number of mono-mineralic olivine samples within this range and determine how the Mössbauer parameters vary with changing proportions of Fe (recorded as olivine fayalite mol%). Bancroft et al [2] carried out a similar procedure on four samples of olivine of varying Fa mol%, but with the purpose of determining cation distribution in olivine.

Methodology: Six synthetic olivine samples ranging from pure fayalite, Fe_2SiO_4 (Fa₁₀₀), to forsteritic olivine, $(\text{Mg}_{80}\text{Fe}_{20})_2\text{SiO}_4$ (Fa₂₀), were obtained for analysis. Approximately 100mg of powder was placed in a lead sample holder. The spectra were recorded at 298K with an ^{57}Fe Mössbauer spectrometer using a $^{57}\text{Co}/\text{Rh}$ source. Spectra were fitted

with a constrained non-linear least squares fitting programme of Lorentzian functions. Three parameters reflecting the energy state and chemical environment of electrons at the nucleus are observed by Mössbauer analysis, two of which, chemical isomer shift and quadrupole splitting, are important for investigating olivine. The former is used to tell the oxidation state of Fe while quadrupole splitting gives information about the symmetry of the structure [1].

Results: The Mössbauer spectrum recorded for olivine shows a well-defined doublet (two-line absorption) of strong intensity as seen in Figure 1. The quadrupole splitting values for each of the synthetic olivine standards were plotted against the Fa mol% (Figure 2).

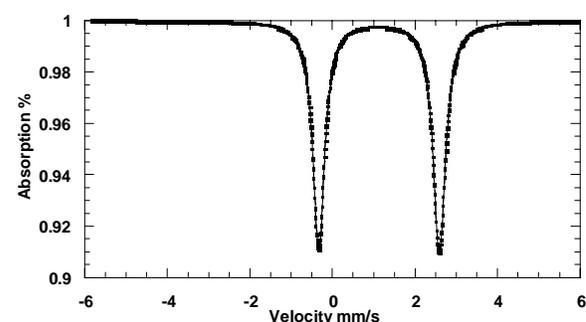


Figure 1. ^{57}Fe Mössbauer spectrum for mono-mineralic olivine ($\text{Mg}_{60}\text{Fe}_{40}$)₂SiO₄ showing a well-defined doublet.

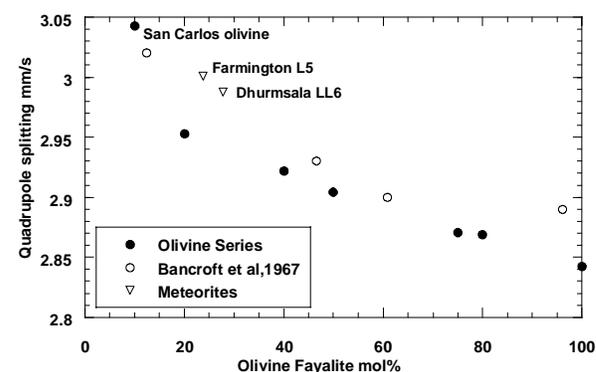


Figure 2. Quadrupole splitting values for a range of olivine standards, including olivine from the San Carlos meteorite and earlier data from Bancroft et al 1967. Also plotted are two unequilibrated ordinary chondrites.

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Quadrupole splitting values are highest for pure forsterite and decrease linearly as Fe^{2+} substitutes for Mg^{2+} in the olivine structure. Quadrupole splitting can be used as a measure of the degree of asymmetry of a compound. The negative slope of the graph can be explained by the fact that the more asymmetric form of olivine, forsterite, has a high quadrupole splitting value and as Fe^{2+} progressively substitutes for Mg^{2+} , the structure becomes more symmetrical and the value of this parameter decreases.

The relationship is confirmed by plotting Bancroft's data points onto the same graph (Figure 2).

Chemical isomer shift values have been plotted against the Fa mol% and also yield a linear correlation although not as marked as that for quadrupole splitting.

Discussion: A standard olivine sample of known composition (Fa_{10}), from the meteorite San Carlos, was analysed and found to plot slightly above the line in Figure 2. Similarly, Bancroft's sample of $\text{Fa}_{12.5}$ plots in this location. As pure forsterite contains no iron it could not be sampled by this technique to determine whether there is a departure from the linear relationship observed between quadrupole splitting and olivine Fa mol% in the synthetic samples at the forsteritic end.

To explore the application to meteorites, two samples of equilibrated ordinary chondrites were analysed, in this case, with known olivine Fa mol% values [6],[7]. The samples were Farmington (L5) and Dhurmsala (LL6). The Mössbauer spectrum of Dhurmsala with clearly defined olivine peaks is shown in Figure 3. The olivine in these meteorites plot on the trend defined by the standard olivines suggesting that Mössbauer spectroscopy could be used in baseline meteorite classification.

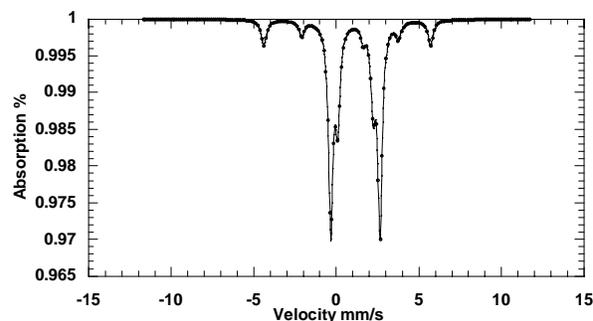


Figure 3. ^{57}Fe Mössbauer spectrum for Dhurmsala. The two highest intensity peaks are those associated with olivine. (Note the velocity difference between this and previous Figure 1.)

Conclusion: The study shows that Mössbauer spectroscopy can be related to the mineralogy of olivine. The quadrupole splitting parameter shows a clear trend with changing olivine Fa mol%. The olivine of equilibrated meteorites fits this trend and with more analyses it may be possible to determine the degree of equilibration within the ordinary chondrites. This data will thus be important in deconvoluting spectra from unequilibrated meteorites.

References: [1] Berry, F. J. (1983). *Physics Bulletin* **34**, 517-519; [2] Bancroft, G. M., Maddock, A. G. and Burns, R. G. (1967). *G.C.A.* **31**, 2219-2246; [3] Bland, P. A., Sexton, A. S., Jull, A. J. T., Bevan, A. W. R., Berry, F. J., Thornley, D. M., Astin, T. R., Britt, D. T. and Pillinger, C. T. (1998). *G.C.A.* **62**, 3169-3184; [4] Hoffman, E., Seifu, D. and Oliver, F. W. (2000). *Met. And Planet. Sci.* **35**, 431-434; [5] Bland, P. A. *Pers. comm.* [6] Keil, K. and Fredriksson, K. (1964). *J. Geophys. Res.* **69**, p3487. [7] Fredriksson et al., in Ahrens, L.H. (1968), *Pergamon*. P487.