CATHODOLUMINESCENCE OF MURCHISON FORSTERITE: TEMPERATURE DEPENDENCE AND STRUCTURAL IMPLICATIONS.

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Forsterite in extraterrestrial material shows compositional differences among different meteorite groups [1]. These compositional and other differences are reflected in cathodoluminescence (CL) spectra. While most CL spectra have been obtained at room temperature, low temperature spectra may reveal additional CL details related to composition and structure. Below we explore the modifications of the CL at low temperature using forsterite from Murchison with spectra obtained in situ to preserve petrographic integrity. We describe the effects of accelerating voltage and incident current on resulting spectra and describe the spectral modifications at temperatures as low as 140K which include sharper peaks, changes in relative intensity, and peak splitting. These effects must be related to the composition and structural details of forsterite and indirectly to growth conditions.

While room temperature CL spectra suggested a difference between C2 and C3 forsterite based on a peak shift of the red or Cr^{+3} component of the spectra [2], low temperature (T) studies [3] have allowed recognition of two contributions to this red peak. The relative magnitudes of these two contributions differ between Allende (C3) and Murchison (C2) forsterite thus offering an explanation for the peak shift in the combined peak at room temperature [2]. The origin of the two CL emissions near 720nm has been attributed to Cr in two different structural sites, probably M1 and M2 [3], with the different low T response reflecting Cr in the higher symmetry M1 site giving sharp emissions while Cr in lower symmetry M2 giving broad CL emission at low T. Using parallel studies of synthetic Cr^{+3} forsterites [4], the CL behavior is due to Cr^{+3}.

While CL is usually considered qualitative, the apparent correlation of CL emissions with individual sites, the different response of the CL at low T from these sites, and the observed differences in apparent Cr^{+3} occupancy in these sites for forsterite known to differ in origin [1] suggests that low T CL may provide useful information for constraining formation/alteration conditions of these grains. Murchison forsterite [5] was used to study the CL emission as a function of sample temperature. The experiment uses a microprobe cold stage cooled by liquid nitrogen gas. Because samples were examined in situ on normal thin sections, however, temperatures of only 140K as measured by a copper-constantan thermocouple attached to the section surface were attained. The acquisition system is described in [2].

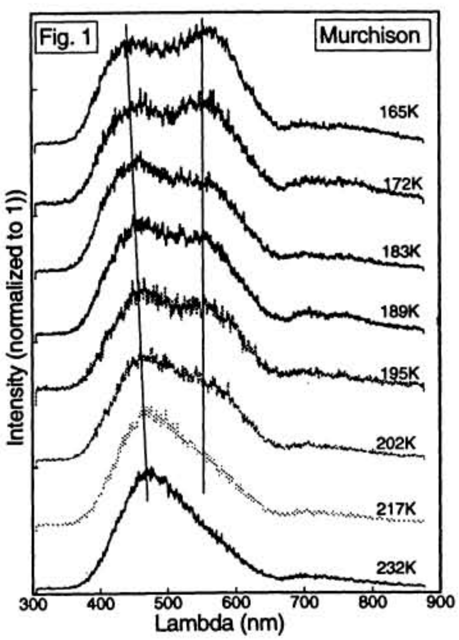
Experimental: All measurements were made in the homogeneous central area of a Murchison forsterite grain [1]. Different accelerating voltages and incident currents were used to evaluate their effects on CL. Previous work had shown the presence of CL emission in the red due mainly to Cr and in the blue due to unknown effects. At constant voltage, an increase in incident current resulted in an increase in the ratio of blue (480nm) to red (700nm) intensity. Both peaks are asymmetric. A similar effect was observed by decreasing the voltage at constant current. Thus, either the red or blue emissions could be emphasized by selecting appropriate current and voltage.

CL response as a function of temperature: Figure 1 shows the blue CL emission as a function of temperature at 15 kV and 50 nA. The asymmetric peak observed at room temperature clearly becomes a doublet at 140K. In addition there is a shift toward shorter wavelength and an increase in the intensity of the 560nm peak relative to the shorter wavelength peak. Figure 2 shows a similar sequence of spectra with conditions emphasizing the peaks in the red (25kV, 0.5nA). With decreasing temperature, the main peak near 700nm becomes sharper and at the lowest temperature, a sharp emission appears near 680nm. A weak, broad peak is present in all spectra near 625nm.

Discussion: The separation of two peaks in the blue at low temperature has not been previously described. A blue peak is common to many silicates and the separation shown in Fig. 1 may either represent a splitting of this peak or a general sharpening of two broad peaks to reveal a peak resulting from a minor element. Parallel studies on pure forsterite may answer this question. The peaks in the red can be assigned to Mn (625nm), and Cr (700nm); the appearance of a sharp peak at the lowest temperature is similar to what is easily seen in [3] but at much lower temperature, higher kV, and low current, all of which promote more intense peaks in the red.

The Cr^{+3} CL emission is a function of the temperature and experimental conditions, including accelerating voltage and incident current. The importance of these data with respect to olivine origin is that the two chemical types of forsterite represented by Allende and Murchison, respectively, apparently show a different occupancy of Cr^{+3} in the two octahedral sites. This distribution can be caused by a number of factors of which formation temperature and/or annealing time are possibly the most important. Equally important however, is the realization that only a small fraction of the Cr may be trivalent and the distribution of total Cr may be quite different. CL has the potential given careful calibration to both provide indications of the amount of Cr^{+3} relative to total Cr and the distribution between sites with a possible calibration of a controlling factor, probably temperature.

References: [1] Geochim. Cosmochim. Acta **50**,1379-1396; [2] ACS Symposium Series, **415**, 150-164; [3] Benstock, Buseck and Steele to be submitted, Am. Mineral. ; [4] Institute of Electrical and Electron. Engineer. J. of Quan. Electron., **27**, 114-120; [5] LPS **XXIV**, 1345-1346.
Acknowledgements: Support obtained through NASA NAGW 3416 (IMS) and the Brasilian funding agency, CNPq (MAZV).



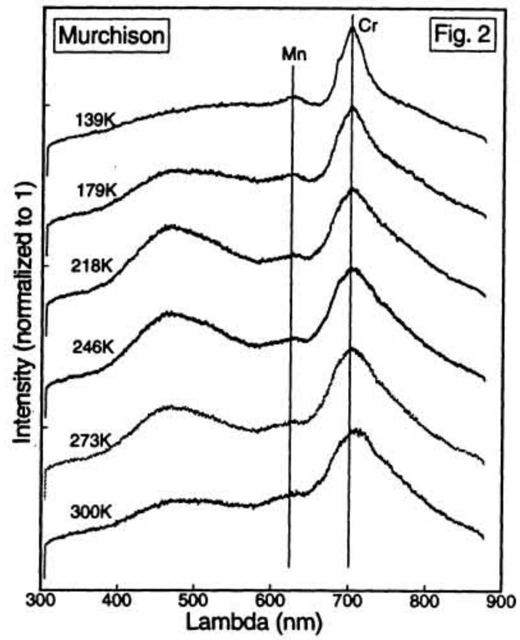


Figure 1. Sequence of CL spectra of Murchison forsterite emphasizing the blue spectral region. Each curve represents a different temperature as labeled. As the sample is cooled the asymmetric blue peak forms a clear doublet with the lower wavelength peak shifting to lower wavelength.

Figure 2. Sequence of CL spectra of Murchison forsterite emphasizing the red spectral region. Each curve represents a different temperature from room temperature to 139K. With cooling the main red peak a 700nm becomes sharper and at the coldest temperature shows sharp peaks as seen in [3]. The peak due to Mn shows no change in intensity or position. A second broad peak near 800nm is seen only as a asymmetric side on the main peak.