IRRADIATION EFFECTS IN FORSTERITE AND THE NATURE OF INTERSTELLAR GRAINS: A COORDINATED INFRARED SPECTROSCOPY AND ELECTRON MICROSCOPY STUDY.

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Introduction. Crystalline and amorphous silicates condense in the outflows of low mass evolved stars and massive red supergiant stars and are injected into the interstellar medium (ISM) where they are rendered almost completely amorphous by a multitude of destructive processes (e.g. shock, grain-grain collisions, and irradiation). Irradiation effects in particular may have played an important role in the genesis and modification of primitive grains in cometary dust [1, 2], but unraveling those effects requires controlled experiments under appropriate conditions and with an emphasis on materials relevant to the ISM. Here we report our infrared (IR) microspectroscopy and transmission electron microscope (TEM) measurements on forsterite that was amorphized through irradiation by high energy heavy ions.

Methods and Samples. The irradiation experiments were carried out at the Intermediate Voltage Electron Microscope Tandem Irradiation facility (IVEM) at Argonne National Lab, which permits simultaneous irradiation experiments with real-time observation in the TEM. Additional details of the instrument design and experimental details are given in [3]. Crushed grains of San Carlos olivine (Fo_{90}) were dispersed on a Cu TEM grid with a holey carbon film support film and irradiated with 1 MeV Kr ions at room temperature to a maximum dose of $1X10^{16}$ ions/cm^2. The in situ TEM measurements were obtained using a modified Hitachi H9000 high resolution TEM. Additional measurements were made using the JEOL 2500SE 200kV field emission STEM at JSC both before and after the IR spectroscopy analyses. Infrared spectra were obtained using Fourier Transform Infrared (FTIR) microscope spectrometers at Brookhaven National Lab (beamline U10A, far-IR microscope) and at JSC to measure transmission spectra over the mid- to far-IR range (2.5-100 μm). We used a modified SpectraTech FTIR microscope with a Si bolometer detector for the far-IR measurements. The mid-IR spectra were obtained with the Nicolet Continuum FTIR at JSC equipped with HgCdTe (MCT) detectors.

Results and Discussion. TEM: At a dose of $1.7X10^{15}$ ions/cm^2, the olivine sample started to develop diffuse diffraction spacings in electron diffraction patterns consistent with the onset of amorphous olivine [3]. The amorphization of the sample (defined as the total loss of olivine reflections in the diffraction patterns) was complete at a dose of $3X10^{15}$ ions/cm^2 (Fig. 1), consistent with other results [4, 5]. Darkfield imaging combined with high resolution images showed no evidence for crystalline domains larger than ~1-2 nm. Prolonged irradiation with the electron beam resulted in rapid devitrification of the grains with the formation of numerous crystalline subgrains.

IR Spectroscopy: Figure 2 shows the "10 μm" Si-O stretching feature for both the irradiated and unirradiated olivine. Despite the lack of crystallographic order based on the electron diffraction results, the IR spectra from the irradiated grains still retain weak features from "crystalline" olivine superimposed on a strong amorphous olivine feature (Fig. 2). The crystalline features are even more pronounced in the far-IR (Fig. 3) especially the pronounced 34 μm feature.

The presence of residual crystalline order by IR and the evident lack of crystalline order by TEM is an intriguing paradox. The crystal structure of olivine contains isolated SiO_4 tetrahedra, and we hypothesize that while ion irradiation destroys crystallographic order in the grains, the local bonding environment for Si-O in the "amorphous" material must still partly resemble that in the original crystalline grains in order to give rise to the observed "crystalline" absorption features in the IR spectra of the irradiated grains. We suspect that the degree of SiO_4 polymerization has an effect on the amorphization behavior of crystalline silicates and plan additional coordinated IR and TEM experiments on ion irradiated enstatite to test this model.

Conclusions. Although forsterite is a commonly detected crystalline silicate in astronomical IR spectra of comets [6], young stars, AGB and massive stars [7], it is not detected above the percent level in lines of sight throught the diffuse ISM [8]. We conclude from our experiments that interstellar amorphous silicate-grains must be structurally disordered at nearly the atomic scale in order to erase the memory of the precursor forsterite component.

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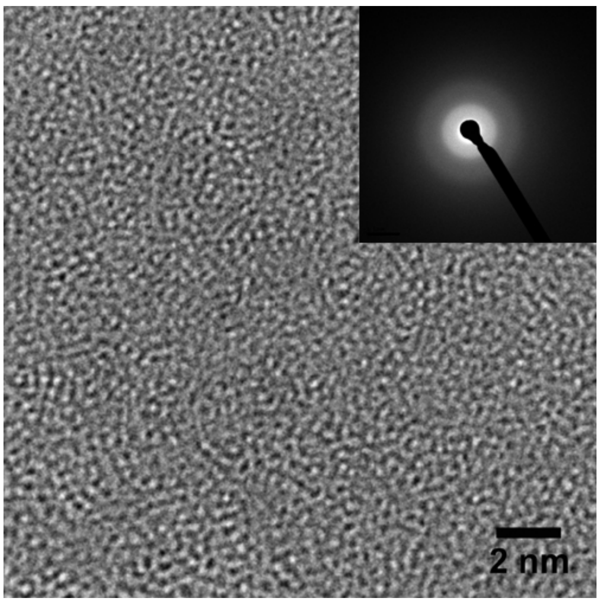

Figure 1. A high-resolution brightfield TEM image of the irradiated olivine sample and the corresponding selected-area electron diffraction pattern showing only diffuse scattering consistent with an amorphous material.

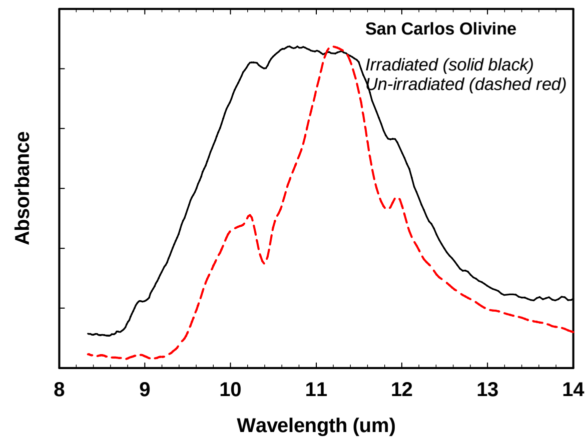

Figure 2. Mid-IR spectra for the Si-O stretch in the starting material (San Carlos olivine, red-dashed curve) and the same material after ion irradiation with 1 MeV Kr to a dose of $3x10^{15}$ ions/cm^2.(solid black curve). The irradiated sample is dominated by amorphous olvine although weak features from crystalline olivine are still eveident.

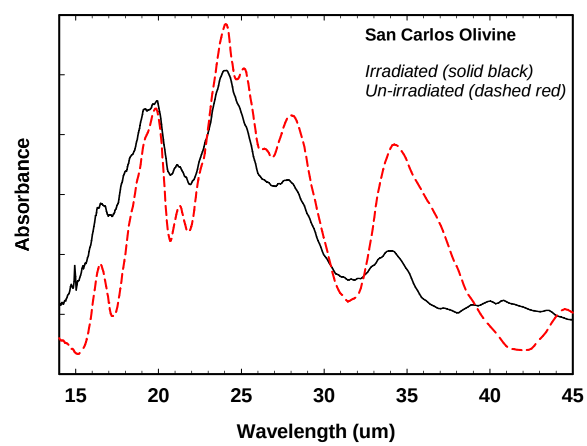

Figure 3. Far-IR spectra for the Si-O bending modes in the starting material (San Carlos olivine, red-dashed curve) and the same material after ion irradiation with 1 MeV Kr to a dose of $3x10^{15}$ ions/cm^2.(solid black curve). The residual crystalline features are well-resolved above the bulk of amorphous material.