

THERMAL ALTERATION OF ORGANICS AND VOLATILES IN CARBONACEOUS CHONDRITES: INSIGHTS FROM IN-SITU HEATING EXPERIMENTS. P. Haenecour¹, J. Y. Howe², T. J. Zega^{1,3}, T. Sunaoshi⁴, M. S. Thompson⁵, S. Dogel⁶, and J. Sagar⁷. ¹Lunar and Planetary Laboratory, University of Arizona, Tucson, AZ, USA. ²Dept. of Materials Science and Engineering, and Dept. of Chemical Engineering and Applied Chemistry, University of Toronto, Ontario, Canada. ³Dept. of Materials Science and Engineering, University of Arizona, Tucson, AZ, USA. ⁴Hitachi High Technologies Co., Hitachinaka, Japan. ⁵Dept. of Earth, Atmospheric, and Planetary Sciences, Purdue University, West Lafayette, IN, USA. ⁶Hitachi High Technologies Canada Inc., Toronto, Canada. ⁷Oxford Instruments NanoAnalysis, High Wycombe, UK. (Email: pierre@lpl.arizona.edu).

Introduction. Carbonaceous chondrites contain fine-grained material consisting generally of a mixture of crystalline and amorphous silicates, oxides, sulfides, Fe-Ni metal grains, and carbonaceous matter that accreted together from the solar protoplanetary disk, *e.g.* [1]. Some of this ‘primary’ material was affected by secondary processing, including both heating and aqueous alteration, on their host asteroid, *e.g.* [1]. The response of primary materials to secondary alteration is important for understanding active processes on the surfaces of and within the parent asteroids of meteorites in the early solar system. Thermal metamorphism, in particular, played an important role in alteration, *e.g.* melting, volatile loss, elemental diffusion between grains, and driving hydrothermal processing. Here we report initial results from in-situ heating of samples of the Murchison (CM2) and Tagish Lake (C2-ung.) carbonaceous chondrites that provide insight into the effects of heating on the composition and microstructure of their volatile-rich components. While these meteorites were affected by aqueous alteration on their parent-body asteroids, and hence contain abundant carbonaceous materials and water-bearing minerals [2-4], they experienced minimal thermal alteration. Thus, our work examines the response of these materials to thermal processing.

Sample and Experimental Methods. Fine-grained material of the two meteorites were crushed and then deposited onto Norcada heating chips. Using the Hitachi Blaze heating holder, we carried out *in situ* heating of each sample in a Hitachi SU9000 scanning transmission electron microscope (STEM/SEM) in vacuum ($< 10^{-5}$ Pa) at temperatures up to 1075°C. The SU9000 is equipped with an Oxford X-Max 100TLE energy-dispersive X-ray spectroscopy (EDS) system and Hitachi electron energy-loss spectroscopy (EELS) system.

The two samples were exposed to different heating protocols. The Murchison meteorite was progressively heated at temperature intervals and held isothermally for different durations: 200 °C (16 min), 400 °C (12min), 600 °C (11 min), 800 °C (2 min), and 1000 °C (10 min). In comparison, the Tagish Lake sample was rapidly heated to 800 °C (~30 sec), then to 900 °C (~30 min),

and 1075°C (~10 min). Before the heating, we acquired images and EDS elemental maps of the samples. During the heating, we recorded video to observe potential microstructural changes. After reaching each heating step, the sample was cooled back down to 200 °C for imaging and EDS mapping. Simultaneous secondary electron (SE), STEM bright-field (BF) and dark-field (ADF) images were acquired throughout the experiment to document changes to the surface (SE) and throughout the bulk (ADF and BF).

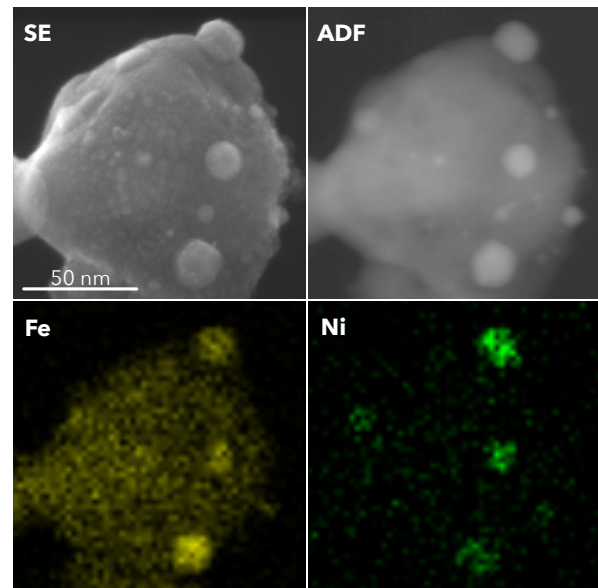


Fig. 1. STEM SE and ADF images, and false-color composite EDS elemental maps of a silicate grain from the Murchison meteorite after heating up to 1000°C.

Results and Discussion: 1) Heating of the Murchison meteorite. Initial EDS analysis of a local part of the sample before heating show that it is composed of ferromagnesian silicate, oxide, sulfides, Fe-Ni metal, and carbonaceous grains. We did not observe any noticeable change to the meteorite in terms of grain structure, surface morphology or elemental composition of the sample until it was heated up to 600 °C. After heating at 800 °C, STEM imaging and EDS measurements revealed changes in the surface morphology and formation of Fe-Ni nanoparticles on the surface of silicate grains (Fig. 1). These particles

appear to have similar elemental compositions as the space-weathering-induced nanophase iron that form on the surface of lunar soil particles [5]. Heating to 1000 °C caused the particles to sinter and form larger assemblages. Previous stepped heating experiments of the Murchison meteorite [6] showed the formation of Ni-rich magnetite particles. However, EDS mapping indicated that the Fe-Ni nanoparticles formed in our experiment do not contain much O but rather have an elemental composition consistent with Fe-Ni metal.

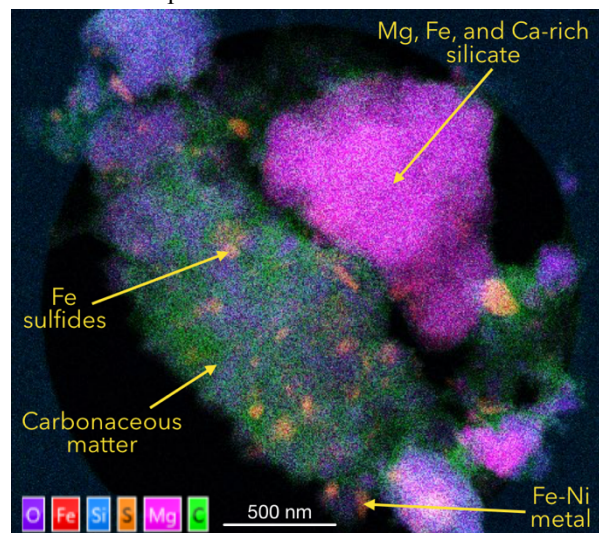


Fig. 2. False-color composite EDS elemental map of the Tagish Lake fine-grained material on the heating chip prior to heating.

2) Heating of the Tagish Lake meteorite. Initial EDS mapping before heating showed that the Tagish Lake sample is composed of a mixture of carbonaceous material, ferromagnesian silicates, Fe sulfides, and Fe-Ni metal (Fig. 2).

STEM imaging showed that flash heating to 800 °C caused significant melting and a reduction of the overall size of the grains. Increased melting and sintering occurred progressively at 900 °C and then 1075 °C. Similar to the Murchison sample, heating also caused the formation of nanoparticles. However, these nanoparticles contain minor Si and so are not pure Fe-Ni metal. This observation is consistent with identification of Si in Fe-Ni metal grains in some carbonaceous chondrites [7]. Furthermore, EDS mapping at each step shows migration of the Fe into the nanoparticles and loss of volatile elements, such as sulfur from the iron sulfide grains (Fig. 3). STEM imaging indicates that heating graphitizes the carbonaceous material, consistent with the increase of the C signal observed in the EDS maps.

Summary: Our initial results from in situ heating of Murchison and Tagish Lake fine-grained material indicate that significant changes to their microstructure and elemental compositions, such as melting and formation of Fe-Ni metal nanoparticles, occurred only after heating above 600°C. Heating up to 1075°C caused a significant loss of volatiles (e.g., S) and the graphitization of the carbonaceous matter.

Acknowledgments: This work is supported by NASA Grants NNX15AD94G (NExSS EOS program) and NNX15AJ22G, and NSF Grant 1531243. The work was carried out at the Kuiper Core Imaging and Microscopy Facility, University of Arizona.

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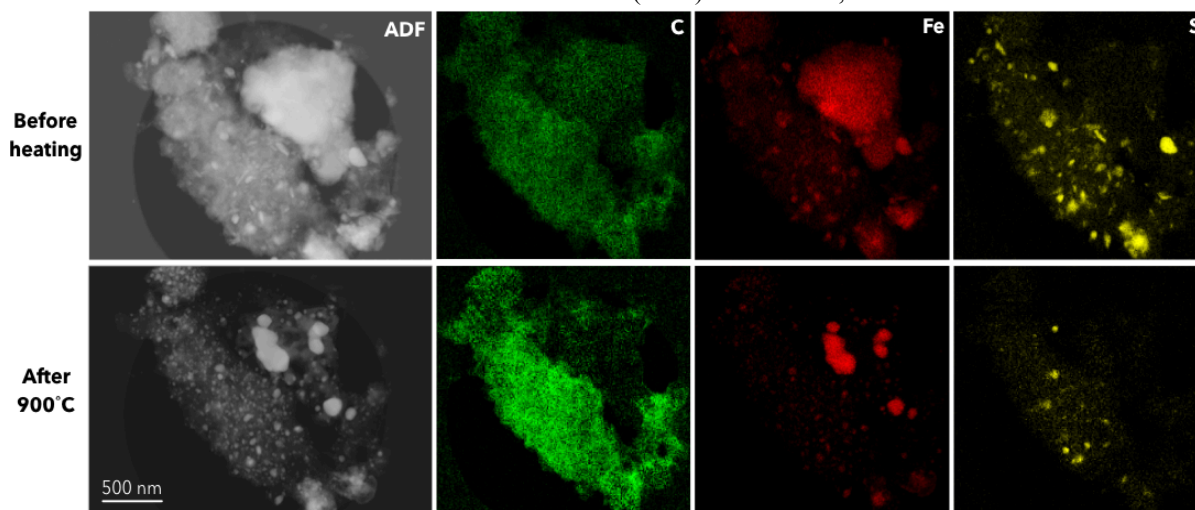


Fig. 3 STEM dark-field (ADF) images and false-color EDS maps (green = C, red = Fe, and yellow = S) of a Tagish Lake sample on the heating chip before and after heating up to 900°C.