FORMATION OF SIDERITE IN LUNAR REGOLITH PARTICLES DURING IRRADIATION FOR INSTRUMENTAL NEUTRON ACTIVATION ANALYSIS. R A. Zeigler*, L. A. Haskin, B. L. Jolliff, A. Wang, and R. L. Korotev, Dept. of Earth and Planetary Science, Washington University, 1 Brookings Dr., Campus Box 1169, St. Louis MO, 63130; *zeigler@levee.wustl.edu.

Introduction: Last year in these proceedings we reported on the occurrence of siderite in a lunar regolith sample [1]. We postulated that the siderite was deposited on the surface of the Moon through the interaction of a fluid produced by the vaporization of a comet or carbonaceous chondrite with the lunar regolith [2]. Because this was an extraordinary finding and interpretation, and because siderite has not previously been identified in a lunar sample, we designed several experiments to verify that we did not somehow produce the carbonate as part of our analytical procedures. These experiments involving additional lunar samples, iron meteorites, and Fe-wire have shown, to our dismay, that the siderite was produced not on the Moon, but during sealing of samples and in the reactor during neutron irradiation for instrumental neutron activation analysis.

Methods: Samples sent to the reactor for irradiation are sealed in ultra-pure silica tubes with a methane and oxygen torch. Samples are irradiated for 6 to 48 hours. During irradiation, they are immersed in the water of the reactor pool, constraining the temperature to <100°C. Raman spectra of samples were taken using a Kaiser Hololab5000 Raman spectrometer (excitation laser wavelength is 532 nm, a spot size of ~6µm). Back-scattered and secondary electron images (BSE and SEM) and quantitative siderite analyses were obtained with a JEOL 733 electron microprobe, at 15 KeV accelerating voltage, 10 nA beam current, and a spot size of 1 µm.

Results: The original particle, 65903,16-7, has siderite and associated goethite occurring as depositional phases on the exterior of the particle, in vugs within the particle, and filling veins down to sub-micron size levels. It shows no evidence that the siderite or goethite was reaction products of any phase within the sample. Alteration products (siderite or otherwise) associated with the Fe,Ni metal or schreibersite (Fe_3P) grains are absent in this sample.

Raman spectroscopic analysis revealed siderite in 8 of 34 additional irradiated Apollo 16 2-4 mm particles (Ni-rich impact melt breccias). Electron microprobe analyses (EMPA) show that, in contrast to the original particle, the morphology of siderite in these particles suggests an origin as alteration products of Fe-rich phases. Siderite grains were typically found as rims on, or partially replacing troilite, schreibersite, ilmenite, and Fe,Ni metal (Fig. 1). The siderite in these grains is compositionally indistinguishable from that in the original particle (Fig. 2). A similar Raman analysis of 41 unirradiated Apollo 16 1-2 mm particles (from rake sample 65902), including one Fe,Ni metal grain, found no trace of siderite.

Pieces of the North Chile iron meteorite [3] irradiated for 6, 12, and 36 hours were analyzed with the Raman spectrometer. No trace of siderite was detected on the 4 pieces that were irradiated for 6 hours, but siderite was widespread on the pieces that were irradiated for 12 and 36 hours. Back-scattered electron images of the 12-hour piece show the rounded surface morphologies of the siderite (Fig. 3) and EMPA results support the identification as siderite (Fig. 2).

Several pieces of Fe-wire were sealed in tubes using the torch in our usual manner and others were sealed under normal laboratory atmosphere, excluding combustion products from the torch. These samples were irradiated for 36 hours. A mixture of siderite and Fe-oxide

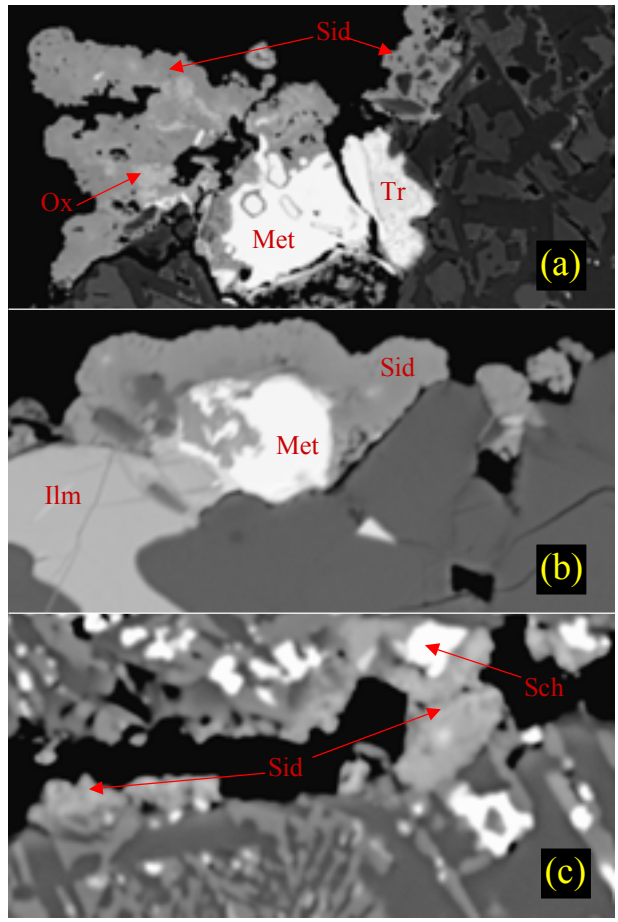

Figure 1: BSE images of siderite (Sid). Fields of view are 125, 160, and 40 µm. **(a)** In 60053,2-10 mixed with Fe-oxide (Ox) replacing Fe,Ni metal (Met) and troilite (Tr). **(b)** In 65903,17-10 replacing Fe,Ni metal and ilmenite (Ilm). **(c)** In 62243,10-12 replacing schreibersite (Sch) in a vein.

(hematite) was identified in the Fe-wire sealed in the usual way. Wire that was sealed under laboratory atmosphere had Fe-oxide alteration but no siderite.

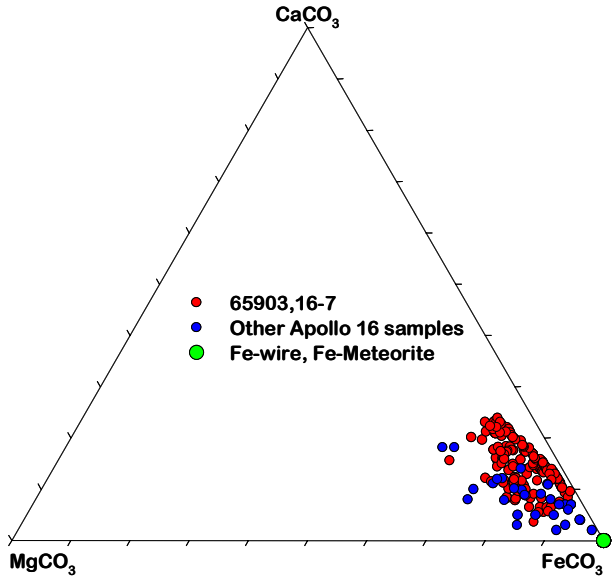

Figure 2: Ternary diagram for the Fe-Ca-Mg carbonate system. The carbonate compositions for the original particle (65903,16-7 in red) overlap with the carbonate compositions in the other 8 samples containing carbonate.

Discussion: Based on the depositional morphology, and the lack of evidence for alteration anywhere in the sample, we interpreted the original occurrence of siderite as lunar. We argued that the samples had not been subjected to any conditions that would produce siderite in the laboratory. That judgement was erroneous; sealing the samples for reactor irradiation did provide an atmosphere that led to formation of siderite and goethite. Although most siderite formation appears to require the warmth of the reactor pool, in at least one case, small amounts of siderite were observed on an Fe-wire that was sealed but not irradiated. The observation that Fe-rich phases have been altered to siderite in our recent experiments and the failure to find siderite in some 40 unirradiated lunar samples is strong evidence that the siderite is terrestrial after all.

The lack of siderite on the Fe-wire sealed in with the Earth's atmosphere is hardly surprising. Assuming complete combustion, the gases from the torch would be H_2O and CO_2 in a 2:1 ratio. These gases have a CO_2 concentration some 4 orders of magnitude higher than that of Earth's atmosphere, which we had previously assumed for the tubes. The lack of siderite observed on the pieces of North Chile irradiated for 6 hours and occurrence on the piece irradiated for 12 hours may indicate a time constraint on the formation of siderite, or it may reflect sealing without the usual amount of contamination of combustion products from the torch.

So, we have shown that siderite is produced in the reactor when sealed with a methane/oxygen torch and perhaps during the act of sealing. As there is no evidence for siderite in unirradiated lunar samples, we conclude that despite apparent textural evidence to the contrary in sample 65903,16-7, the siderite we observed in lunar regolith particles was produced here on Earth.

References: [1] Zeigler R. A. et al. (2001) *LPS XXXII,* 1242. [2] Zeigler R. A. et al. (2001) *LPS XXXII,* 1243. [3] Wasson G. H. et al. (1989) *GCA, 53,* 735-744.

Acknowledgements: This work was supported by NASA grant NAG5-10485 through Larry Haskin. We would also like to thank K. Kuebler for help with the Raman analyses.

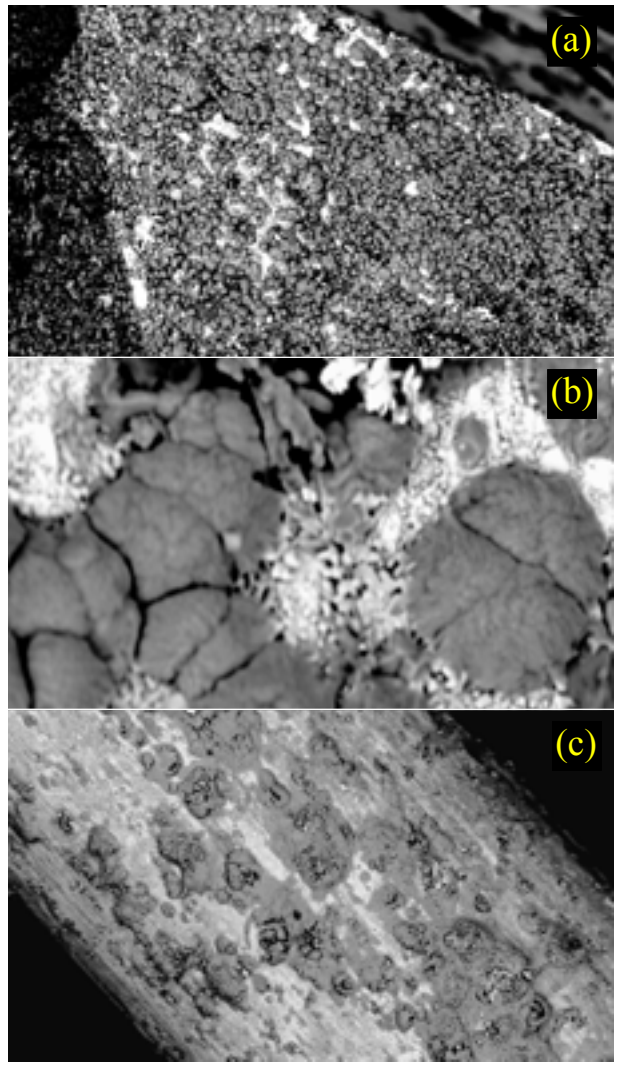

Figure 3: BSE images. Fields of View are 700, 65, and 700 µm respectively. **(a)** The surface of the North Chile meteorite (12 hour irradiation). Nearly the entire surface is covered with siderite alteration. **(b)** Close up view showing the rounded, fractured morphology of the siderite. The mottled areas are mixtures of siderite and metal. **(c)** The surface of the Fe-wire sealed under normal conditions. All of the medium grey raised areas are a mixture of siderite and hematite.