OXYGEN ISOTOPES IN LUNAR METAL GRAINS – A NATURAL GENESIS EXPERIMENT.
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Introduction: The oxygen isotopic compositions of materials from the solar nebula are highly diverse and uniquely represent a myriad of potential effects from diverse nucleosynthetic inputs, high-temperature fractionations through to low temperature alteration, with non-mass-dependent and mass dependent isotope fractionations.

The main variation in solar system materials is in the abundance of $^{16}$O. In standard three-isotope plots, $^{16}$O as the most abundant isotope is used as the denominator and so variations in $^{16}$O abundance lie on a 1:1 correlation line with enrichment in $^{16}$O towards the origin (Figure 1). Most solid materials (as bulk meteorites, terrestrial, lunar) of the solar system lie within less than 1 % or so of each other in terms of normalized $^{16}$O abundance. The earliest known objects in the solar system, refractory inclusions and related objects have large enrichments of $^{16}$O up to 7% relative to terrestrial. Chondrules can show $^{16}$O enrichments up to a few percent.

Figure 1. Potential solar O isotopic reservoirs: (1) Close to terrestrial and most meteorite bodies, (2) $^{16}$O enriched close to refractory inclusions, (3) $^{16}$O-depleted and potentially elevated above the terrestrial mass fractionation line (dependent on gas/dust enrichment).

Various considerations of the solar O-isotope composition have been made and these vary markedly, over the full range of solar-system-related O-isotopic measurements.

Most models place the Sun with a composition close to the solid materials [1,2]. The premise has been that early solar system materials were all enriched in $^{16}$O and then reacted with solar gas to produce the bulk compositions now seen. Only the most refractory materials preserve the original solid composition. Other models are dependent on the gas to dust ratio of the solar nebula and lower levels of $^{16}$O in the Sun would be expected for higher dust/gas enrichments, up to $^{16}$O of +54 % for a 200% enrichment [2]. Recently, it has been postulated that the refractory inclusions themselves represent the solar value, and that the meteorites represent a modified composition due to nebula processing [3]. In this case, all solid material has been substantially modified in the solar nebula.

In effect, we do not know the isotopic composition of solar oxygen within a range of nearly 10 %. This is of course a major priority of the Genesis mission in that a sample of solar wind is to be collected and returned to Earth for analysis. In preparation for this return, we have begun an experiment with lunar materials in an effort to at least constrain possible endmembers of the solar composition.

Solar Wind: The implantation of solar wind into the lunar surface is a well-known phenomena and a known resource for determining solar isotopic compositions, notably for the noble gases but also for light elements such as N and possibly C [4, 5]. Implantation of oxygen into silicate materials would offer an enhancement of the solar composition in the surface micron or so, but the level of intrinsic lunar oxygen would make this a very difficult measurement in practice. However, FeNi metal grains are present in lunar soils and these may act as solar wind collectors with little intrinsic oxygen. As such, this potentially represents a natural Genesis experiment but with a much longer exposure interval. There are many issues concerning contamination, diffusion, assimilation, and redistribution of volatiles in the lunar regolith environment, but the lunar samples are good proxies for Genesis materials and at least they will give an indication of the problems to be encountered.

Metal Grains: A 0.5 g aliquot of lunar soil (10084) was sieved to pass 75 μm and then magnetic grains were obtained by passing a hand magnet over the sample. The magnetic separation was then hand picked for opaque metallic grains, but these grains were dominated by ilmenite. A bulk sample of the hand magnetics was scattered onto double-sided tape, set into epoxy and polished to reveal surfaces. In this way metal grains could be identified through their higher reflectivity compared to ilmenite. Three
suitable grains were found for analysis ranging in size from ca. 40 to 80 µm. These three grains were removed from epoxy, and mounted into a gold substrate and were designated [ ], the largest grain which is a spherule, [ ], a metallic shard, and [ ], a small spherule about 40 µm in diameter.

**Analytical Techniques:** The analyses were carried out on SHRIMP II in multiple collection mode. Mass resolution for $^{16}$O and $^{18}$O were maintained at 2000 (M/ΔM 10% peak) and 5500 for $^{17}$O. Isotopic ratios were referenced to a magnetite from a southeast Australian granite as well as the lunar ilmenites. A 1 nA Cs+ beam was used to sputter both the oxide and the metal grains and all samples were analyzed with a faraday cup for $^{16}$O and CDEM for $^{17}$O and $^{18}$O. Magnetite and ilmenite analyses were interspersed with analyses of two of the metal grains. The count rates from the oxides were kept relatively low to minimize gain changes from the CDEM (order 40,000 c/s of $^{18}$O from ilmenite). Count rates from the metal grains were lower by an order of magnitude compared to the oxide grains. Internal precisions ranged from 1-2 % for the ilmenite measurements, and around 5 % for the metal analyses. Oxygen counts from the gold substrate were negligible indicating oxygen signals are associated with the grains.

**Results:** The first grain analyzed ( [ ]) displayed relatively constant $^{16}$O count rates and a near-normal composition and probably reflects contamination of this metal grain from an adhering silicate particle.

Grain [ ] was flattened slightly during pressing into the gold foil. Analysis of the middle of the grain revealed a growing then decaying O– signal consistent with the expectation of implantation of solar wind in the surface of the grain. Analyses to the edge of the flattened area showed slower signal decay. The O isotopic composition of this grain is systematically heavier than the standards. The decaying signal of the first analysis moved back to near normal suggesting a minor component of O in the metal. Four areas on the grain were analyzed. All show similar trajectories. The extreme analyses are enriched in both $^{17}$O and $^{18}$O by up to 80 ‰.

Various experimental tests were carried out to verify the behavior of grain [ ]. Inappropriate ion optic tuning could cause isotopic shifts but these were largely related to mass-dependent fractionation. Reducing the count rates on an ilmenite caused no major isotopic shift indicating the isotopic composition is not related to a base-line effect of the electrometer. Furthermore, the composition of particle [ ] is near normal, mass fractionated by less than 10 % from the neighboring ilmenite analyses. As such, the composition of [ ] cannot be related simply to a geometric effect.

**Oxygen Isotopes in Solar Wind:** The lunar metal grain [ ] is characterized by a decaying profile that is consistent with implantation from solar wind (± solar energetic particles). As such, the composition of the implanted oxygen could be taken as solar. The composition of the oxygen isotopic component is unknown in any meteoritic reservoir. The enrichment of both $^{17}$O and $^{18}$O is consistent with a $^{16}$O-depleted reservoir, the complement of the $^{16}$O-enriched component in refractory inclusions. As such, the data presented here are not consistent with a solar oxygen isotope composition near refractory inclusion compositions [3], or compositions close to terrestrial planets and planetesimals. The solar oxygen isotopic composition would be that envisaged for a high dust/gas enrichment [2]. However, the isotopes of K and S in lunar soils are known to be heavy due to loss of light isotopes from the lunar regolith caused by solar wind induced sputtering or impact vaporization [6]. Perhaps a similar mechanism could be invoked to explain the heavy nature of surface O in lunar metal grains. It is premature to infer the solar composition based on this implanted component alone. It is possible that some analytical artifact is affecting the analyses although we have gone to some length to document these. Analysis of the isotopic composition of oxygen in lunar metal grains is a good proxy for the implanted wafers of the Genesis experiment. The lunar metal grains have much higher concentrations of solar wind due to their long exposure and so the analytical protocols are more straightforward. On the other hand, the history of the metal grains may be more complicated than originally envisaged and their oxygen isotopic composition may not directly reflect implanted solar wind alone. Further analyses of these materials will provide an interesting comparison for Genesis results.