

MEGASIMS UPDATE: OXYGEN TRANSMISSION, DESTRUCTION OF OH MOLECULAR IONS, AND STABILITY OF THREE-ISOTOPE MEASUREMENTS. P. H. Mao¹, T. Kunihiro¹, K. D. McKeegan¹, C. D. Coath^{1,2}, G. Jarzebinski¹, and D. Burnett³ ¹Dept. of Earth & Space Sciences, UCLA, Los Angeles, CA. 90095-1567 USA (peterm@ess.ucla.edu); ²Dept. of Earth Sciences, Univ. of Bristol, Bristol, BS8 1RJ, UK; ³Div. Geol. & Planetary Sci., Caltech, USA.

Introduction: The GENESIS Discovery Mission [1] seeks to measure the average elemental and isotopic composition of the solar system to a precision and accuracy sufficient to address important questions in planetary science. In September of 2004, the GENESIS sample return capsule (SRC) returned to Earth with its payload of solar wind samples, captured in ultra-pure target materials. Due to an assembly error, the parachute on the sample return capsule never opened, causing the capsule to impact Earth at terminal velocity, ~300 km/h. Most of the target materials were broken and all of the science materials suffered some degree of surface contamination. Fortunately, however, both of the SiC concentrator targets, one of which we will request for oxygen and nitrogen analysis, returned fully intact [2].

The UCLA MegaSIMS consists of a secondary ion mass spectrometer (SIMS) front end coupled to an accelerator mass spectrometer (AMS) through a band-pass mass filter (recombinator). This instrument has been designed specifically to tackle the unique analytical challenges posed by the Genesis samples: dilute elemental concentrations, limited sample material, and close proximity of likely surface contamination to the implanted solar wind ions. The design criteria and overall instrument description were reported in McKeegan et al., 2004 [3].

Soon after completing the beamline in late 2004, we were able to inject a recombined three-isotope beam (Si) into the accelerator mass spectrometer, verifying proper operation of all major beamline components and determining the mass resolution of the system [4]. Over the past year, major milestones include completion of the sample introduction airlock, installation of a low energy Ar gun for in-situ sample surface cleaning, and the installation of electron multiplier detectors at the high energy mass focal plane. The airlock was a necessary modification to the Cameca ims-6f in order to accommodate the relatively large Genesis concentrator samples. Its completion allowed us to introduce a magnetite sample, from which we obtain a strong oxygen beam.

Oxygen transmission: With an oxygen beam available, we can now determine optimal operating conditions for analyzing the Genesis concentrator sample. The three major results we are presently pursuing are (1) charge-state distributions and transmission through the accelerator as a function of accelerator terminal voltage and stripper pressure, (2)

molecular destruction as a function of stripper pressure at optimal terminal voltages, and (3) determination of the instrument stability over time scales from minutes to days.

Charge-state distributions were measured at Ar-stripper densities ranging from 1 to 3 $\mu\text{g}/\text{cm}^2$ and terminal voltages from 400 to 1200 kV. For this measurement, the magnets and lenses of the recombinator were set up to recombine oxygen with ^{17}O on the beamline axis but the slits were set up to only pass ^{16}O . The beam current was measured by Faraday cups in front of the accelerator and after the high-energy electrostatic analyzer (ESA). Based on 250-550 kV AMS results of Synal et al. [5], we expected to find maximum throughput with the $q=+1$ charge-state at a terminal voltage near 400 kV. Our results for $q=(+1, +2, +3)$, shown in Figure 1, demonstrate that O^+ has maximal transmission at ~550 kV, but we also found that the O^{2+} gave an even higher transmission value at 1200 kV than the $q=+1$ state did at 550 kV. Transmission through the accelerator of O^{2+} at $V_{\text{terminal}} = 1200$ exceeds 45%. This is a very encouraging result because angular straggling, due to beam-stripper collisions, improves with terminal voltage as $1/V_{\text{terminal}}$ and molecular destruction ought to be more efficient at higher charge states. The O^{3+} transmission maximum is not accessible with our accelerator system.

Molecular destruction: As with ^{14}C AMS carbon-dating, the main purpose of the accelerator in our system is to eliminate molecular ions from the beam by collisional dissociation. In early tests with the instrument, we observed that at very low stripper densities ($\sim 0.1 \mu\text{g}/\text{cm}^2$) $^{16}\text{OH}^+$ could be detected at the mass focal plane. Injecting all three isotopes of oxygen into the accelerator, we found that the ratio of the 18 amu/16 amu signal was within a few percent of SMOW, but that the 17 amu/16 amu ratio was about a factor of five too high. This indicates that the molecular ion signal ($^{16}\text{OH}^+$) was about four times stronger than the $^{17}\text{O}^+$ signal. When the stripper density was raised to $\sim 1 \mu\text{g}/\text{cm}^2$, the molecular ion signal was significantly suppressed and the 17 amu/16 amu ratio decreased to within a few percent of SMOW. Clearly we need to repeat these measurements to precisely quantify the molecular destruction as a function of stripper density. As an indicator to what we expect, Synal et al. [5] demonstrated a 3 order of magnitude reduction in the $^{13}\text{CH}^+$ signal corresponding to an increase in stripper

density from 0.1 to 0.8 $\mu\text{g}/\text{cm}^2$.

After we have determined our optimal accelerator settings for terminal voltage and stripper density, we will obtain the repeatability and stability of oxygen isotope ratio measurements with the instrument. At this time, although we have not had any significant problems with these measurements, we do not have sufficiently controlled results to report.

Precise measurements of the accelerator's molecular destruction efficiency and the stability of three-isotope oxygen composition of magnetite will be reported at the March meeting.

References: [1] Burnett D.S. et al., (2003) *Spa. Sci. Rev.* 105: 509-534. [2] Wiens R. C. et al., (2004) *EOS Trans. Am. Geophys. U.* 85: 497-498. [3] McKeegan et al., (2004) LPSC 35 : #2000. [4] Mao et al., (2005) LPSC 36 : #2259. [5] Synal et al., (2000) *Phys Rev B*, **161-163** (2000) 29-36.

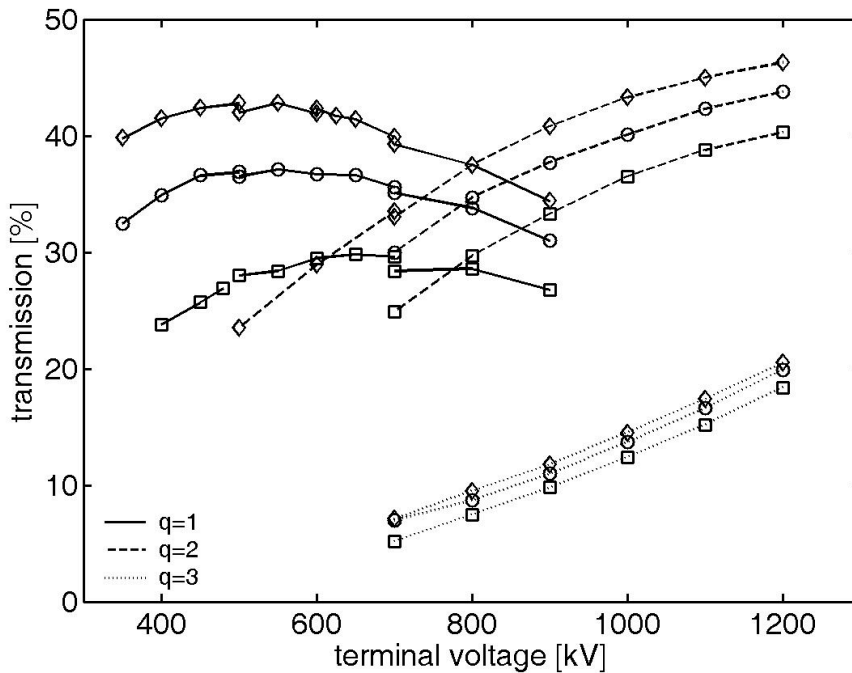


Fig. 1: Charge-state distributions in ^{16}O transmission at Ar-stripper densities ranging from 1 to 3 $\mu\text{g}/\text{cm}^2$. Data symbols: diamond, circle, and square correspond to 1, 2, and 3 $\mu\text{g}/\text{cm}^2$, respectively.