STRONTIUM ISOTOPIC CONSTRAINTS ON EARLY SOLAR SYSTEM CHRONOLOGY. R. Parai[1], S. B. Jacobsen[1] and S. Huang[1], [1]Harvard University, Department of Earth and Planetary Sciences, 20 Oxford Street, Cambridge, MA 02138, U.S.A. (parai@fas.harvard.edu).

Introduction: Precise determination of the timing and duration of early Solar System events, such as condensation from the solar nebula and subsequent planetary differentiation, are important to our understanding of the formation and evolution of the early Solar System. The long-lived ^{87}Rb-^{87}Sr system provides a useful chronometer for early Solar System events. Rb is moderately volatile, while Sr is refractory; furthermore, Rb is highly incompatible during partial melting, while Sr is moderately incompatible. Therefore, Sr isotopic ratios record information about condensation events and volatile depletion, as well as silicate differentiation in the early Solar System. High-precision analysis of Rb-Sr systematics in Solar System materials such as calcium-aluminum inclusions (CAIs), eucrites, angrites and lunar rocks can provide insight into processes that shaped the early Solar System.

Early Solar System Rb-Sr Chronology: Variation in initial ^{87}Sr/^{86}Sr among planetary materials has been interpreted to reflect differences in the timing of separation of planetary bodies from the solar nebula [e.g., 1-5]. However, Moynier et al. [6] recently argued for Sr isotopic heterogeneity throughout the Solar System based on variations in ε^{84}Sr measured in planetary materials (where ε^{84}Sr is the ^{84}Sr/^{86}Sr deviation in parts per 10,000 relative to a standard). If Sr isotopes are distributed heterogeneously among planetary bodies, then ^{87}Sr/^{86}Sr variations may partially reflect isotopic anomalies rather than radiogenic ingrowth due to ^{87}Rb decay. Non-radiogenic variation in ^{87}Sr/^{86}Sr could either reflect a direct isotopic anomaly (an excess or deficiency in ^{87}Sr relative to a standard) or an indirect effect, in which a sample with anomalous ^{88}Sr/^{86}Sr yields a spurious ^{87}Sr/^{86}Sr value after correction for instrumental mass fractionation to a "normal" ^{88}Sr/^{86}Sr value. For example, Moynier et al. [7] report ^{88}Sr/^{86}Sr variations up to 1-2 per mil among planetary materials, which would translate to a 0.5-1 per mil artifact in ^{87}Sr/^{86}Sr if samples are internally normalized to a "normal" ^{88}Sr/^{86}Sr.

Corrections for Instrumental Mass-Dependent Sr Isotope Fractionation: The earliest studies that established differences in initial ^{87}Sr/^{86}Sr among meteorites, CAIs and lunar rocks used a linear or power law [1-4] to correct for instrumental mass-dependent fractionation during analysis by thermal ionization mass spectrometry (TIMS). Russell et al. [8] demonstrated that for Ca isotopes, TIMS data deviated significantly from a linear fractionation law, and that an exponential fractionation law was most successful in fitting measured data. Subsequent studies have thus used an exponential law to correct for instrumental mass fractionation during TIMS analysis for other elements, such as Sr. However, a test of fractionation laws similar to the Russell et al. [8] study has never been extended to other elements, except for Ba [9]. Sr and Ca do not necessarily follow the same fractionation law during TIMS analysis, and correction for instrumental fractionation using an improper fractionation law may generate spurious ε^{84}Sr anomalies.

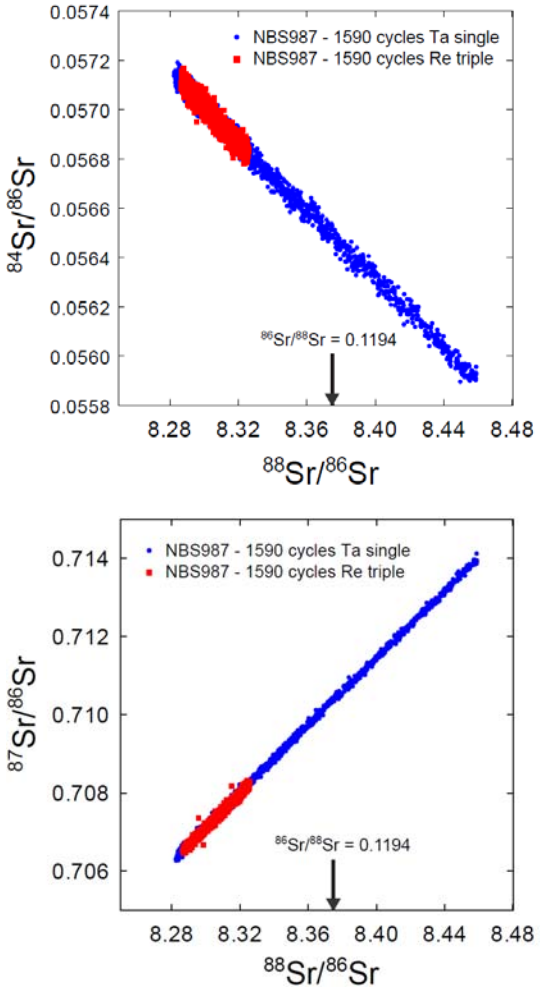

Fig. 1 Typical Sr TIMS data measured on the GV IsoprobeT at Harvard. Data for a Re triple filament run are shown as red squares, and data for a Ta single filament run are shown as blue dots. ^{87}Sr/^{86}Sr is corrected for ^{87}Rb interference.

Results: We present Sr data for NBS-SRM987 standards measured on a GV IsoprobeT at Harvard. 500 ng of Sr were loaded with 2N H_3PO_4 onto a Ta single filament, or onto one side filament of a Re triple filament assembly. Mass 84, 85, 86, 87 and 88 peaks were monitored using a single sequence method under static mode. Each cycle takes 10s, and a block consists of 10 cycles. Baselines were measured for 20s at half masses before each block. ^{87}Rb interference on ^{87}Sr was corrected using ^{87}Rb/^{85}Rb = 0.385617. This correction was typically <15 ppm for Ta single filaments and <150 ppm for Re triple filaments. For all measurements, a steady 5V ^{88}Sr signal was maintained until all Sr burnt out. A typical filament provided ~1500 cycles, corresponding to ~8 hours of instrument time. Typical raw cycle data from a single run are shown for a Ta single filament and a Re triple filament assembly in Fig. 1. The total extent of fractionation during measurement is much smaller for a Re triple filament run compared to a Ta single filament run, consistent with the result of [10] for Ca isotopes. Triple filament raw data have a very narrow ^{88}Sr/^{86}Sr range (8.28-8.32) and do not extend to the canonical "normal" ^{88}Sr/^{86}Sr value corresponding to ^{86}Sr/^{88}Sr = 0.1194.

Based on two compilations of block data from 5 Re triple filament runs and 8 Ta single filament runs, respectively, we empirically characterize instrumental mass fractionation during analysis. Linear regression in log-log space using the Williamson [11] best-fit algorithm yields slopes corresponding to the exponent b_x in a mass fractionation law of the form:

$$\left(\frac{\left({}^{x}\mathrm{Sr}/{}^{86}\mathrm{Sr}\right)_{\mathrm{sample}}}{\left({}^{x}\mathrm{Sr}/{}^{86}\mathrm{Sr}\right)_{\mathrm{normal}}}\right)=\left(\frac{\left({}^{88}\mathrm{Sr}/{}^{86}\mathrm{Sr}\right)_{\mathrm{sample}}}{\left({}^{88}\mathrm{Sr}/{}^{86}\mathrm{Sr}\right)_{\mathrm{normal}}}\right)^{b_x}$$

where x = 84, 87. We compare the empirically-determined exponents to those of various mass fractionation laws in Fig. 2. We find that the empirical values lie off the trend of expected values: the empirical values for b_{87} are close to the equilibrium value in both single and triple filaments, but the empirical b_{84} values lie between Rayleigh and exponential values.

Discussion: Based on the typical range of instrumental mass fractionation we observe in Ta single and Re triple filament runs, we may constrain the magnitude of a spurious ε^{84}Sr anomaly that could arise from the application of an improper fractionation law to correct measured values. For example, if Sr run on a Ta single filament fractionates during measurement with the series of ^{88}Sr/^{86}Sr values illustrated in Fig. 1 following a Rayleigh law, correction using the exponential law generates a spurious 0.5 ε-unit anomaly in the mean corrected ε^{84}Sr. For Sr run on a Re triple filament assembly, the same calculation for a typical

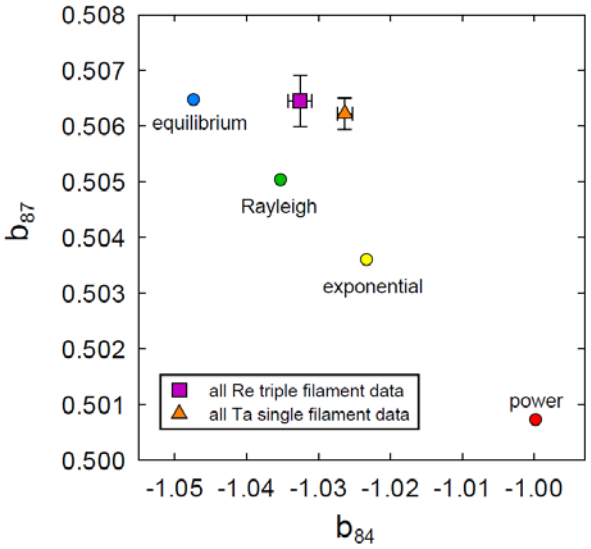

Fig. 2 Empirically-determined mass fractionation slopes from measurements of NBS SRM-987. b_{84} is the slope in log(^{84}Sr/^{86}Sr) vs. log(^{88}Sr/^{86}Sr) space, and b_{87} is the slope in log(^{87}Sr/^{86}Sr) vs. log(^{88}Sr/^{86}Sr) space. Calculated slopes reflecting power, exponential, Rayleigh and equilibrium fractionation laws are also shown.

^{88}Sr/^{86}Sr series yields a spurious 1 ε-unit anomaly in mean corrected ε^{84}Sr, due to the greater mean extent of fractionation relative to the "normal" value of ^{88}Sr/^{86}Sr. With the exception of CAIs, ε^{84}Sr anomalies reported by Moynier et al. [6] are less than 1 ε-unit in magnitude. Therefore, it is important to accurately characterize mass fractionation that occurs during measurement for a given TIMS filament assembly in order to determine ε^{84}Sr anomalies. Furthermore, application of a Sr double spike technique would allow us to fully characterize any Sr isotopic heterogeneity by constraining natural variation in ^{88}Sr/^{86}Sr.

In summary, precise determination of instrumental mass fractionation laws and development of a Sr double spike technique will provide the best constraints on the age of formation of planetary objects, such as the Moon, based on initial ^{87}Sr/^{86}Sr differences.

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