IMPROVED PRECISION FOR THE *CHEMISTRY AND DATING EXPERIMENT* USING FS-LASER ABLATION.

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Introduction: We have developed a portable Rb-Sr and Pb-Pb dating instrument called the Chemistry and Dating EXperiment (CDEX) to address billion year uncertainties in inner solar system chronology [1-3]. CDEX uses laser ablation resonance ionization mass spectrometry (LARIMS) to produce isochrons, test concordance, and map geochemistry of samples. We have demonstrated Pb-Pb dates with an average precision of ~±50 Ma years, and Rb-Sr dates with an average precision of ~±180 Ma. We hypothesized that the difference in precision between Rb-Sr and Pb-Pb dates originates with time dependent elemental fractionation caused by low intensity (~60 GW/cm^2) 213-nm nanosecond laser ablation. Thus, elemental fractionation limits the Rb-Sr precision, but not the Pb-Pb precision. In order to test this hypothesis, we have replaced our ablation laser with a femtosecond laser with intensities up to ~400 PW/cm^2. Using this fs-laser, we have made new measurements of the Zagami meteorite that show the CDEX measurement precision has improved by ~5X, resulting in a Rb-Sr date of 191±56 Ma, $(^{87}Sr/^{86}Sr)_i$ of 0.725, and MSWD of 1.17 with 415 spot measurements. These results are consistent with previous laboratory measurements [4], and suggest that increasing ablation intensity can improve Rb-Sr precision to the instrumental limits consistent with our intra element Pb-Pb measurements.

Relevance: The chronology of the inner solar system is based on models relating the crater densities of planetary surfaces to calibrated radiometric dates of well-provenanced lunar samples that primarily constrain the era between 3.5 and 4.2 Ga, as well as the very recent past. These results have been extrapolated to Mars, and throughout the solar system. However, recent work comparing the numerous lunar chronology models in the literature [e.g., 5, 6], illustrates differences between the models of up to one billion years for the period between ~2.8 to 3.3 Ga [7]. Improving CDEX measurements opens the door to rapid, simple, and precise chronology measurements on rocky bodies throughout the inner solar system. CDEX dates with a precision of ±50 Ma are more than sufficient to address the billion year uncertainties in solar system history from 1-3 Ga. Furthermore, because CDEX can produce dates in as little as 5 hours, it can be used to produce measurements on multiple samples thought to be from comagmatic lithologies, potentially improving precision sufficiently to constrain the age of large impact basins.

Laser Ablation: The laser ablation literature is replete with demonstrations of how higher intensity (>1 GW/cm^2 [e.g., 8]), shorter wavelength (<266 nm [9, 10]), and shorter pulse duration improve ion and particulate production [11-13]. Based on these observations, high intensity, nanosecond pulse length, UV ablation lasers with TRL of 6+ are being developed for spaceflight. Recent technological developments enable the potential for shorter pulses in flight systems [14, 15]. Our new ablation laser is capable of producing 266, 400, and 800-nm wavelengths, pulse durations of 40-1000+ femtoseconds, and power of up to 3.5W. After careful studies of measurement reproducibility [16], we have identified optimal measurement conditions at 150-fs and 250 TW/cm^2. Current measurements have only used the 800-nm capability, with shorter wavelength measurements to be completed in the near future.

Conclusion: In addition to new measurements of Zagami, we are currently making new measurements of Sudbury impact lapilli, and expanding our understanding lunar Pb reservoirs to improve our ability to derive lunar Pb-Pb dates. We are also developing methods to improve isotopic measurements while reducing instrument size, complexity, and cost, using new approaches like using photon momentum for isotopic separation.

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