

LEAD DIFFUSION AND CLOSURE TEMPERATURE IN FeS: IMPLICATIONS FOR DATING IRON METEORITES. H.C. Watson¹, Z. Zhang¹, J. Linerelli¹, S. Wang² and D.J. Cherniak³, ¹Union College, Department of Physics and Astronomy (watsonh@union.edu), ²Columbia University, Department of Environmental Engineering ³Rensselaer Polytechnic Institute, Department of Earth and Environmental Sciences

Introduction: Many iron meteorites have enough radiogenic lead in their sulfide phases to allow for absolute age dating using the Pb-Pb system (Chen and Wasserburg, 1983). Absolute ages have been measured by ²⁰⁷Pb/²⁰⁶Pb dating of troilite nodules in the IVA iron meteorites (e.g. Blichert-Toft et al. 2010). This, in conjunction with established cooling rates and the age of core segregation from ¹⁸²Hf/¹⁸²W isotopes, (Kleine et al. 2005, 2009) can provide constraints on the timing from accretion and differentiation through crystallization and final cooling. Scarce data on Pb diffusion in sulfides preclude using an accurate closure temperature for this system, limiting its utility in constraining thermal histories. Proxies such as the closure of Os in pyrrhotite (Brenan et al. 2000) have been used but may not be realistic. We conducted a series of experiments to measure Pb diffusion in pyrrhotite (Fe_{1-x}S), with direct applicability to the closure temperature of Pb in this system.

Methods: Diffusion experiments were performed in evacuated silica capsules at 1 atm and 500°C to 850°C for a period of several hours to two weeks. Pb was introduced at the surface of polished pyrrhotite (FeS) crystals either as a presynthesized FeS powder doped with ~1 at% Pb or as a thin film of either pure Pb or galena (PbS) deposited by an evaporator coater. The resulting concentration profiles were measured by Rutherford backscattering spectroscopy (RBS) at the University at Albany Ion Beam laboratory. Some experiments were also analyzed at the Union College Ion Beam Analysis Laboratory (UCIBAL) for comparison. Depth resolution with the detector used in measurements is ~10nm, and detection limits to a few tens of ppm (atomic) for Pb given typical beam currents and acquisition times used in this work.

Results: The RBS profiles of Pb following diffusion anneals were fit with a model solution to the diffusion equation to determine the diffusion coefficients (*D*). Diffusion is modeled as simple one-dimensional, concentration independent diffusion in a semi-infinite medium with a source reservoir maintained at constant concentration. Figure 1 shows a representative concentration profile (blue symbols) and the resulting fit (black curve).

Our preliminary results over the studied temperature range suggest that the closure temperature for Pb is significantly higher than that of Os, and wide scale melting (of sulfides and metals) would likely be required to reset this isotopic clock.

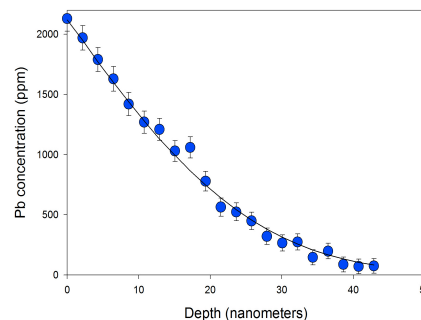


Figure 1: Representative concentration profile for a Pb diffusion experiment measured using RBS (blue symbols) and resulting fit (black curve). This experiment was run at 750°C for 24 hours.

Figure 2 shows the results of closure temperature models for a range of grain sizes and cooling rates. The closure temperatures were calculated using the model presented by Ganguly and Tirone (2001). The activation energy for Pb is approximately 87 kJ/mol, compared to 211 kJ/mol for Os. At relatively high temperatures, the diffusivity of Pb is actually significantly less than that of Os. In fact, the diffusivity is close to six orders of magnitude slower than Os at temperatures close to FeS crystallization (~1000°C), resulting in a much higher capability of retaining an isotopic signature.

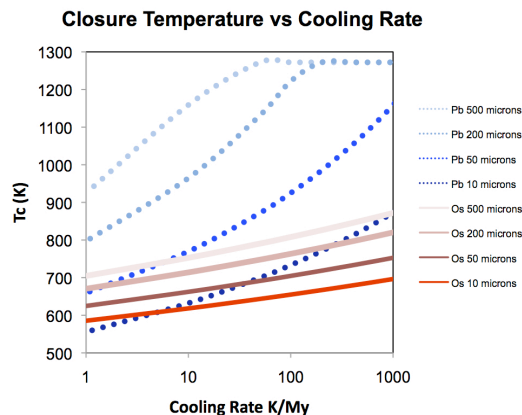


Figure 2: Preliminary calculations of closure temperature as a function of cooling rate and grain size for Pb and Os.

Conclusions: The key points that have emerged from our preliminary data are as follows 1. Pb in sulfide appears to have a relatively high closure temperature compared to Os in most cases (for grains larger than ~10 microns) 2. Larger grain sizes have a closure temperature that approximates the peak temperature (eutectic) at fast cooling rates. 3. This dependence on cooling rate is also largely dependent on grain size, with larger grains retaining the peak T_0 at slower cooling rates. So, in iron meteorites with sufficient radiogenic lead (and large enough troilite nodule) to get an accurate age measurement, this age is likely from early crystallization of the sulfide close to the peak temperature.

References: [1] Chen and Wasserburg 1983 *JGR*, 90, 1151–1154. [2] : Blichert-Toft, et al. (2010). *EPSL*, 296, 469-480 [3] Kleine et al., 2005, *GCA*, 69:24, 5805-5818. [4] Kleine et al. 2009, *GCA* 73, 5150-5188. [5] Brennan et. al., 2000, *EPSL* 180, 399-413 [6] Ganguly J., and Tirone, M., 2001 *MAPS*, 36, 167-175.