COSMOGENIC PRODUCTION RATES IN PRESOLAR SiC GRAINS.

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Introduction: Presolar grains formed from the outflow of dying stars and were incorporated into meteorite parent bodies at the formation of the Solar System. These grains recorded snapshots of stellar nucleosynthesis. However, the time gap between their and solar system formation is still a conundrum. Two recent studies [1,2] analysed He, Ne, and Li isotopes in such grains and derived cosmic ray exposure (CRE) ages. Due to the small grain size, cosmogenic nuclides can be lost due to recoil. Here we present a physical model to calculate cosmogenic production rates and recoil loss in presolar SiC grains.

Model: Cosmogenic production rates were calculated for SiC assuming a galactic cosmic ray (GCR) spectrum with no solar modulation, an integral flux of $17.4 \, \text{cm}^{-2} \, \text{s}^{-1}$ [3], and assuming 92% protons and 8% α particles [3]. The cross section database used is the same as in [4], however, several excitation function were calculated using TALYS-1.2 [5] and INCL/ABLA-4.5 [6] and adjusted to experimental data (if available). Recoil losses for energies up to 240 MeV were constrained using the same model as [4] using recoil spectra from the TALYS-1.2 code. For higher energies we calculated recoil spectra using INCL/ABLA-4.5, converted them into the same format as the TALYS-1.2 calculations, and plugged these spectra into our model [4].

Results: Our newly calculated cosmogenic production rates [in $10^{-10} \, \text{cm}^3 \, \text{STP} \, \text{g}^{-1} \, \text{Ma}^{-1}$] are 322 ± 167 for ^{3}He, 72.8 ± 13.6 for ^{6}Li, 128 ± 24.0 for ^{7}Li, 41.0 ± 15.6 for ^{21}Ne, and 50.8 ± 18.1 for ^{22}Ne. The quoted uncertainties are purely from the cross sections and do not include uncertainties in the GCR spectrum. Retention rates for these isotopes in, e.g., a presolar grain with 5 µm radius are 6%, 17%, 26%, 62%, and 65%, respectively.

Disucssion: The calculated production rates are similar to but more state-of-the-art than previous estimates [7]. Retention of cosmogenic nuclides are rather small for ^{3}He and increase with mass of the produced nuclide. The low retention of, e.g., ^{3}He yields a rather large uncertainty in CRE ages calculated via ^{3}He. Reevaluating the data of [1] and [2] using the new model results in older CRE ages. The ages increases by an average factor of 2.7 for ^{3}He and 1.1 for ^{21}Ne. Due to the large uncertainties in the ^{3}He production and retention rates, the ^{21}Ne measurements are preferred. CRE ages for Li are about a factor of 5 higher than given in [2], mostly due to the fact that retention rates are much lower than previously estimated. Uncertainties in CRE ages determined via Li isotopes are again rather large but they are still underestimated because the uncertainties of the Li/Si ratio, which are needed for proper modeling, are not given in [2]. All reevaluated grains show ages between 20 Ma and 3.5 Ga prior to Solar System formation.

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DUST ENRICHMENT: LESS THAN MEETS THE EYE.

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Introduction: Total evaporation of a dust-enriched region is often called upon to provide a solar nebular environment of high enough f_{O2} at high temperature to account for the near-ubiquitous presence of FeO in chondrites. Unless the dust contains substantial water, however, this mechanism is of limited efficacy.

Results: Equilibrium condensation calculations were done with the VAPORS code [1] on systems enriched by 10^4x in dry chondritic (OC) dust [2], but otherwise solar in composition. A total pressure of 10^{-3} bar ensured that the condensation sequence in a cooling gas is MELTS liquid [3], olivine, FeNi metal, sp+opx. Log f_{O2} falls from IW at 2200K to IW-1.5 by 1950K (Fig. 1) due to consumption by condensing silicate melt of the free oxygen that was released by high-T vaporization of the dust [2]. Olivine Fa_6 coexists with melt containing 19 wt% FeO at 1950K. Over the next 500K of cooling, X_{Fa} rises to only 0.17 as reduction lowers the oxidized fraction of the total iron by 27% (Fig. 2), causing FeO in the liquid to plunge to 5 wt%. If the dust contains just 5 wt% H_2O, reduction is more subdued, as a smaller fraction of the free oxygen recondenses, keeping f_{O2} higher, IW-1.2 to -1.0 until T reaches 1450K, where X_{Fa}=0.29 and the melt contains 14 wt% FeO. Massive enrichment in anhydrous dust thus yields only transient oxidizing conditions, and condensates so formed will show obvious signs of reduction with falling T.

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Fig. 1

Fig. 2
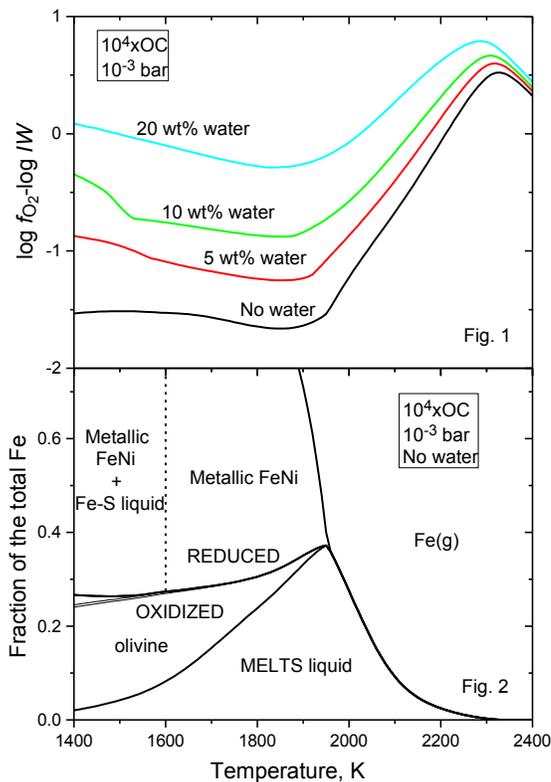
CHRONOLOGY OF AQUEOUS ACTIVITY AND SOURCES OF WATER ON THE CHONDRITE PARENT BODIES: TESTING THE GRAND TACK MODEL

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Introduction: Most chondrite parent bodies accreted water ices together with anhydrous minerals and subsequently experienced aqueous/hydrothermal alteration and fluid-assisted thermal metamorphism resulting in formation of a diverse suite of secondary minerals [1]. It is currently being debated whether hydrated asteroids formed in the main asteroid belt [2] or whether they formed further away from the Sun and were subsequently implanted into the main belt during dynamical evolution of the Solar System (SS) [3,4]. To test these models, we attempted to constrain the accretion regions of hydrated chondrite parent bodies (CPBs) using ^{53}Mn-^{53}Cr and O-isotope systematics of aqueously formed minerals, thermodynamic calculations, and physical modeling.

Results and Discussion: The ^{53}Mn-^{53}Cr chronology of datable secondary minerals by SIMS with matrix-matched standards – calcite in CM and CI [5], and fayalite in CO3, CV3, and LL3 chondrites [6] – indicates that aqueous activity on the CPBs started ~3–5 Myr after formation of CV CAIs with the canonical $^{26}Al/^{27}Al$ ratio [7], consistent with ^{26}Al being the major heat source in these bodies. The peak metamorphic temperatures reached by the CPBs [8], the ^{26}Al-^{26}Mg ages of chondrule formation [9,10], the ^{53}Mn-^{53}Cr ages of aqueous alteration [5,6], and the physical modeling suggest that the CPBs accreted ~2.0–4 Myr after CV CAIs [5,6,11]. The inferred water-to-rock mass ratios in ordinary and carbonaceous CPBs range from <0.1 to ~0.6 (could be higher in CIs), which is significantly lower than the solar value of 1.2 [12]. We suggest that most CPBs accreted close to the snow line; CIs may have accreted further away from the Sun than other chondrite groups. Because the snow line for the 2.5–4 Myr-old disk is expected to be within 2–3 AU from the Sun [13,14], we infer that CPBs sampled by meteorites accreted in the main asteroid belt, consistent with the inferred D/H ratio of asteroidal water [2] and near the terrestrial O-isotope compositions ($\Delta^{17}O$ from –2‰ to +5‰) of aqueously formed fayalite, magnetite and carbonates. Therefore, the existing meteorite observations provide no clear evidence supporting the predictions of the Grand Tack dynamical model of the SS evolution that hydrated asteroids formed between and beyond the giant planets and were subsequently implanted into the main asteroid belt during migration of the giant planets [3]. We note, however, that there may be other types of hydrated or water ice-bearing planetesimals that were implanted into the main asteroid belt, but have not been sampled by the known meteorites.

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^{26}AL-DEPLETIONS IN ANOMALOUS AND SOLAR PLAC-LIKE CAIS SUGGEST HIGH DEGREES OF PROCESSING IN THE EARLY SOLAR NEBULA

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Introduction: Due to their large nucleosynthetic anomalies [1], platy hibonite crystals (PLACs) are thought to be among the oldest solids that formed in the solar system. Most of them lack evidence for incorporation of ^{26}Al, suggesting formation prior to injection and/or homogenization of ^{26}Al in the solar system [1].

Our combined study of O, Ca and Ti isotopes in a large number of PLACs and related CAIs (hereafter PLAC-like CAIs) revealed a correlation between Δ^{17}O, i.e., the mass-independent variation in O isotopes, and the range of large-scale anomalies in ^{48}Ca and ^{50}Ti (>100‰; [2]). Such a distribution is expected if the primordial (protosolar molecular cloud) dust was depleted in ^{16}O relative to the inferred solar value [2,3]. In addition, we can now use this relationship to distinguish between solar (solar O, δ^{48}Ca and δ^{50}Ti ≈ 0‰) and anomalous PLAC-like CAIs (^{16}O-depleted relative to solar, range of δ^{48}Ca and δ^{50}Ti).

We have studied the Al-Mg systematics in the same CAIs with the WiscSIMS Cameca IMS 1280 to investigate whether both solar and anomalous PLAC-like CAIs belong to the ^{26}Al-depleted population or whether these CAIs represent multiple generations with respect to their incorporation of ^{26}Al, in which case we would expect to find relationships between the degree of nucleosynthetic heterogeneity, O isotopes and ^{26}Al incorporation.

Results and discussion: Our results confirm that most solar and anomalous PLAC-like CAIs show no evidence for having formed in the presence of live ^{26}Al (δ^{26}Mg* ≈ 0‰). Two CAIs of the anomalous population (δ^{48}Ca ≈ −20‰ and 80‰, δ^{50}Ti ≈ −20‰ and 60‰, respectively) have slightly enhanced δ^{26}Mg* values, which could indicate ^{26}Al-incorporation at a ^{26}Al/^{27}Al ratio of ~(1–1.5)×10^{-5} or that the Mg isotopic heterogeneity in the early solar system was on the order of ~15‰.

Importantly, the results show that both isotopically anomalous and solar PLAC-like CAIs are depleted in ^{26}Al and that its abundance cannot be used for a relative chronology of PLAC-like CAIs. Both solar and anomalous PLAC-like CAIs likely formed in the earliest stages of solar system history, prior to arrival of this short-lived radionuclide. In particular, the presence of ^{26}Al-free PLAC-like CAIs with solar O, Ca and Ti suggests that highly homogenized reservoir(s) had been established prior to a widespread distribution of ^{26}Al.

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ISOTOPIC COMPOSITION OF PRESOLAR SILICON CARBIDE GRAINS ANALYZED WITH CHILI.

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Introduction: The Chicago Instrument for Laser Ionization (CHILI), a resonance ionization mass spectrometry (RIMS) instrument, is designed for isotopic and chemical analysis at the ~10 nm scale with a useful yield of ~40 % [1,2]. After several years of instrument development, we report the first analyses of presolar grains performed with CHILI. Preliminary results on a subset of the grains were reported previously [3].

Samples and analytical procedures: Twenty-two presolar SiC grains from Murchison separate KJG were analyzed for Sr, Zr, and Ba isotopes. Six tunable Ti:sapphire lasers allow simultaneous resonance ionization of three elements with two-photon ionization schemes. Atoms were desorbed with a 351 nm laser beam, focused to ~1 μm using a Schwarzschild optical microscope. Standards showed isotopic precision in the few ‰ range.

Results and discussion: Ten of the 22 grains selected had sufficient Ba content for isotope analysis with CHILI, and eight of these ten grains also had sufficient Sr concentration. However, none of the grains had detectable Zr. The reason for the nondetection of Zr is unclear. Previous analyses [4–7] suggested that 30–50 % of presolar SiC grains should have detectable Zr.

Seven grains have Sr and Ba isotopic ratios similar to those previously observed in mainstream SiC grains [8,9], consistent with formation in low-mass asymptotic giant branch (AGB) stars.

Relative to ^{86}Sr and ^{136}Ba, another grain is depleted in ^{84}Sr, ^{87}Sr, ^{134}Ba, and ^{135}Ba and enriched in ^{88}Sr, ^{137}Ba, and ^{138}Ba. This is consistent with formation in a neutron burst caused by a shock wave passing through ejecta of a type II supernova. Calculations predict that ^{137}Ba and ^{138}Ba should be similarly enriched relative to ^{136}Ba after complete decay, but much of the ^{137}Ba comes from ^{137}Cs ($t_{1/2}$ = 30 a) [10]. Our results, δ^{137}Ba = (+310±100) ‰ and δ^{138}Ba = (+1670±180) ‰ (2σ errors), would indicate grain formation within a few years of a supernova explosion.

Two other grains are depleted in ^{84}Sr, ^{87}Sr, and ^{88}Sr (or enriched in ^{86}Sr) and enriched in ^{134}Ba/^{136}Ba relative to mainstream grains [8,9]. These patterns are very difficult to produce in AGB stars, but might be produced in supernova ejecta.

Conclusions and outlook: CHILI has analyzed its first natural samples and provided first relevant data from presolar grains. Although optimization of CHILI continues, CHILI is already more capable than earlier RIMS instruments. A wide variety of cosmochemical problems will be explored in the near future, including isotopic studies of presolar grains, CAIs, and samples returned to Earth by the Genesis and Stardust spacecraft.

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OXYGEN ISOTOPES AND HIGH ^{26}Mg EXCESSES IN A U-DEPLETED FINE-GRAINED ALLENDE CAI.

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Introduction: CAIs are thought to be among the first solids formed in the early Solar System (ESS). As such, they are prime samples to study when (1) investigating ESS high-temperature processes, and (2) searching for evidence of short-lived radionuclides at the time of formation of the SS. A recent systematic study of fine-grained CAIs characterized by a Group II REE pattern from Allende [1], found an extremely large ^{235}U excess (δ^{235}U >50 ‰ rel. to CRM-112a) in one sample: ME-3364 3.2. The discovery of this large ^{235}U excess provides definite evidence of the existence of live ^{247}Cm in the ESS, as previously suggested by [2]. In this study, we analyzed the oxygen isotope compositions and Al-Mg systematics of CAI ME-3364 3.2 to constrain the conditions of its formation.

Method: Petrologic and mineralogical studies were carried out by SEM equipped with EDS at UCLA. Secondary minerals such as nepheline, sodalite, Fe-rich pyroxene, and Fe-rich spinel were observed in ME-3364 3.2. Analyses for oxygen isotopes and Al-Mg systematics were performed on the IMS 1270 ion probe in UCLA. The instrumental mass fractionation for both oxygen and Mg isotopes was corrected by comparison to San Carlos olivine, pyroxene and Burma spinel. Excesses in ^{26}Mg (δ^{26}Mg*) were calculated by adopting an exponential law with a mass fractionation exponent of 0.516 obtained from the analysis of standards.

Results and Discussion: Despite being U-anomalous and full of secondary phases, the oxygen isotopic compositions of this fine-grained CAI are not too different from those of other non-FUN CAIs. The Δ^{17}O values of sodalite and nepheline range from -20‰ to -5‰, similar to values previously obtained on Efremovka fine-grained CAIs [3]. Oxygen isotopes are fractionated slightly along a mass dependent fractionation line at Δ^{17}O~ -7‰, which could be caused by imperfect corrections for instrumental mass fractionations due to the lack of proper standards for fine-grained CAIs. Large excesses in ^{26}Mg have been identified in ME-3364 3.2 over a large range of ^{27}Al/^{24}Mg values (from 42 to 667), yet the data do not define an isochron. Instead, the ^{26}Mg excess is approximately uniform across all spots analyzed. All the spots are characterized by large but variable negative δ^{25}Mg (from ~ -9‰ to -19‰).

The elevated, yet homogenous, δ^{26}Mg* and negative δ^{25}Mg in ME-3364 3.2 indicates Mg isotope exchange must have taken place in a closed system to avoid dilution with chondritic Mg, although our data cannot constrain a reliable timescale of formation of the secondary phases. Further investigation is required to explain ^{26}Mg excesses and negative δ^{25}Mg associated with U depletion in this fine-grained inclusion.

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PRESOLAR NEON-22 IN INDIVIDUAL GRAPHITIC SUPERNOVA SPHERULES FROM ORGUEIL

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Introduction: Stellar sources of presolar graphitic spherules (hereafter called grains) from the CI chondrite Orgueil have been determined [1,2]. While high-density grains can have multiple stellar sources, low-density grains (LD; <2.0 g cm^{-3}) predominantly originate in supernovae (SN) [1,2]. In 2010, we presented the first noble gas data from Orgueil grains and found that 4 out of 15 LD grains contained presolar ^{22}Ne barely above detection limit [3]. While the fraction of grains with detectable ^{22}Ne was similar to the one found for grains from Murchison [4,5], the measured gas amounts were much lower. This could be due to significant erosion by ion beam sputtering during *extensive* NanoSIMS analyses (C, N, O, Al-Mg, Si, K, Ca, Ti isotopes) prior to noble gas extraction. In order to test whether LD grains from Orgueil and Murchison represent different populations, we analyzed for Ne in 7 Orgueil LD fraction grains of similar size that had not been exposed to an ion beam.

Methods: Grains were imaged in the SEM with short electron beam dwell times to minimize heating. Gases were extracted by melting individual grains with an IR laser, and He and Ne isotopes were analyzed in a high-sensitivity compressor noble gas mass spectrometer [6] by a method developed specifically for presolar grains [7]. Subsequently, melted grains were imaged with SEM and analyzed for C and O isotopes with the NanoSIMS.

Results & Discussion: Most (5 of 7) of the grains contain orders of magnitudes more ^{22}Ne than our previously analyzed LD grains [3]. This implies that grain erosion by extensive ion beam sputtering prior to noble gas analysis leads to loss of noble gases and can mislead interpretations of gas amounts. We detected neither ^{20}Ne nor ^{21}Ne. The pure ^{22}Ne is Ne-R ((^{20}Ne/^{22}Ne)$_{measured}$ < (^{20}Ne/^{22}Ne)$_G$) from the decay of the radioactive nuclide ^{22}Na ($t_{1/2}$=2.6 a) that was extant in the grains when they formed. Na-22 forms predominantly in SN [e.g., 8] and indicates that Ne-R containing grains originate in SN. Melt residues appear to be diluted with normal material and do not reveal large C and O isotopic anomalies except in one Ne-rich grain.

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