U-Th-Pb ISOTOPIC COMPOSITIONS IN STARDUST SiC GRAINS FROM THE MURCHISON METEORITE.

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Introduction: Much has been learned from stardust grains about galactic chemical evolution, stellar nucleosynthesis, physical properties of stellar atmospheres, and physical and chemical conditions in the solar nebula and meteoritic parent bodies (e.g., [1]). However, many questions still remain to be answered. One of the major unresolved issues concerning stardust grains is their age. The determination of absolute ages of stardust grains is of paramount importance in astrophysics because it would enable researchers to put firm constraints on the dynamics of grain processing in the interstellar medium, and on the temporal evolution of stellar systems and their nucleosynthetic products.

While constraints on the time of formation of stardust grains in stellar outflows before arrival in the solar nebula would be very useful, obtaining chronological data on stardust grains is not straightforward. This is because for many of the long-lived chronometers (for example the ^{87}Rb–^{87}Sr decay scheme) the parent isotope is not expected to condense at the high temperatures at which stardust grains form in stellar outflows. Furthermore, the abundance of a particular radionuclide can be affected by a number of variables, such as the elemental composition of the stellar outflows, the thermochemical behaviour of the radionuclide under varying chemical and physical conditions of the stellar atmosphere, and the mechanism of incorporation into stardust grains. Another difficulty arises from the fact that concentrations of potential chronometers (parent and daughter nuclides) in stardust grains are near the detection limit for most analytical techniques. Their small sizes also make any isotopic determination an arduous task. Curiously, the same feature that helps in characterizing stardust grains, namely their anomalous isotopic signatures, hampers the application of conventional radiometric dating as the initial abundance of neither the parent nor the daughter nuclide can be estimated with certainty.

Despite these limitations, the U-Th-Pb system may still provide valuable insights into the evolutionary timescale of stardust grains and their parent stars. Because of its low condensation temperature, Pb is unlikely to be incorporated into refractory stardust phases at the time of their formation. Of the four stable isotopes of lead (^{204,206,207,208}Pb), only ^{204}Pb is nonradiogenic. The other Pb isotopes are the final decay products of three complex decay chains from uranium (^{235,238}U) and thorium (^{232}Th). Once U and Th are incorporated into SiC grains, their isotopic ratios will (in theory) only be modified by their decay, which will result, consequently, in the build-up of their daughter Pb isotopes. The possibility of U-Th-Pb chronometry on stardust grains depends, however, on the concentration of the isotopes of these elements, our ability to make precise and accurate measurements of their isotopic compositions, and knowledge of their production ratios. But above all, the assumption of no incorporation of Pb at the time of the stardust condensation must be verified. Of all types of stardust grains, SiC grains provide the best opportunity for this to be accomplished. This is because stardust SiC grains have typically high trace-element concentrations and, although most SiC grains are sub-micron in size, some are larger than 1 μm [2,3], making them suitable for multiple-element isotopic analyses in single grains.

Samples: U-Th-Pb isotopic measurements were performed on twenty four single SiC grains from the LS+LU fraction extracted from the Murchison CM2 carbonaceous chondrite. The grains range in size from 5 to 58 μm [3]. In addition to the individual grains, we also analysed a SiC-enriched bulk sample (KJB fraction, [3]). The single grains studied here show Si-, C-, and N-isotopic compositions in the range displayed by the so-called “mainstream grains”, and therefore are believed to have condensed in the outflows of low mass (~ 1.5 to 3 $M_\odot$) carbon-rich AGB stars with close-to-solar metallicity [4]. The KJB fraction also shows C-, N-, and Si-isotopic signatures consistent with an AGB origin for most of the grains [5].

U, Th, and Pb isotopic measurements: Uranium-238, ^{232}Th, and ^{204,206,207,208}Pb isotopes were measured with the ANU SHRIMP-RG ion microprobe in single collection mode, using an O_2^- primary beam of ~ 5 nA focused to sputter an area of ~ 20 μm in diameter. The analytical procedure was broadly similar to that commonly employed on zircons [6,7]. The beam was initially rastered over each grain for ~ 60 s before data acquisition to minimize surface contamination from an area larger than the analytical spot. Secondary ions were extracted at 10 kV and measured by single collector analysis on a ETP^{TM} multiplier in peak-jumping mode. A source slit of 350 μm and a 300 μm collector slit were used to obtain a mass resolution of 7000 (m/Δm, 1% peak height). The NIST-610 silicate glass and a synthetic SiC ceramic doped with heavy elements [8] were used to monitor instrumental mass fractionation. Mass interferences and background were monitored by periodically analysing a "pure" synthetic SiC single crystal and the Au foil, onto which both the KJB and LS+LU fractions had been deposited.

The ^{238}U/^{232}Th isotopic compositions of single stardust SiC grains were also measured with the SHRIMP-II ion microprobe operated in multi-collection mode. SHRIMP-II measurements were obtained with an O_2^- primary ion beam of ~ 5 nA focused to a ~ 20 μm diameter spot. The secondary ^{232}Th^{16}O$^+$ and ^{238}U^{16}O$^+$ ions were extracted at 10 kV and measured simultaneously on a Sjuts^{TM} (KBL 408) continuous dynode electron multiplier and on an ETP^{TM} discrete dynode electron multiplier, respectively. Mass resolution was 5500 (m/Δm, 1% peak height). All measurements were corrected for background contribution. Differential gain between the multipliers was calibrated through analysis of standards by switching the ^{232}Th^{16}O$^+$ peak between the detectors. The standards were routinely measured throughout the analytical session. As standards we used the NIST-610 silicate glass and the synthetic SiC ceramic doped with heavy elements [8].

Results: Lead abundances are very low, typically less than ~ 4 ppm. Most stardust SiC grains show ^{206}Pb/^{204}Pb, ^{207}Pb/^{204}Pb, and ^{208}Pb/^{204}Pb ratios lower than the most primitive Pb isotopic compositions known in the Solar System (primordial lead – Canyon Diablo Troilite, [9]). The ^{206}Pb/^{204}Pb ratios range from 5.1 to 12.5, the ^{207}Pb/^{204}Pb ratios from 3.6 to 11.0, and the ^{208}Pb/^{204}Pb ratios from 9.4 to 23.1. Thorium and U were detected in stardust SiC grains with inferred concentrations between ~ 0.03 and 5 ppm and between ~ 0.01 and 0.82 ppm, respectively. The measured ^{238}U/^{232}Th ratios in 22 out of 24 grains are close to or lower than 0.263, the present-time ^{238}U/^{232}Th ratio of the bulk Solar System [10]. The ^{238}U/^{232}Th ratios measured with SHRIMP-RG and II agree within uncertainties.

Discussion: The Pb isotopic compositions determined in this study for stardust SiC grains are clearly anomalous in comparison with those found in solar system materials, being enriched in the *s*-process only ^{204}Pb. This is not unexpected, since previous analyses of heavy elements in mainstream SiC grains have shown that these grains are strongly enriched in *s*-process nucleosynthetic products (e.g., [1]). As pointed out previously (e.g., [11]), the *s*-process signature observed in most mainstream grains appears to be the result of a mix between two components, one similar to but not exactly the same as the solar isotopic compositions (the so-called N-component), and the other with isotopic ratios close to those predicted for pure *s*-process (the so-called G-component). Our data indicate that an additional component might also play a role in determining the Pb isotopic signatures of stardust SiC grains. This is the radiogenic component (R-component), as ^{206}Pb, ^{207}Pb, and ^{208}Pb are the final decay products of ^{238}U, ^{235}U and ^{232}Th, respectively.

Given the large Pb isotopic anomalies observed in the stardust SiC grains and the unknown degree of contamination with common Pb (here defined as all Pb other than that derived from the *s*-process in the envelope of AGB stars, and from in-situ accumulation of radiogenic Pb), we cannot be certain about isotopic ratios of the initial *s*-process Pb incorporated into the grains, which makes any attempt to calculate a date for each individual grain far too uncertain.

The deconvolution of the Pb isotopic signatures into specific components is complex, as several assumptions are required in order to determine the composition of each end-member (N-component, G-component, and R-component), as well as to quantify the degree of mixing of such components. Another complication is that the Pb isotopic compositions of individual SiC grains show large variations, which might arise from different end-member compositions, and/or variations in the proportions of each Pb component.

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