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Strontium and barium isotopes in presolar silicon carbide grains measured with CHILI—two types of X grains

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Abstract

We used CHILI, the Chicago Instrument for Laser Ionization, a new resonance ionization mass spectrometer developed for isotopic analysis of small samples, to analyze strontium, zirconium, and barium isotopes in 22 presolar silicon carbide grains. Twenty of the grains showed detectable strontium and barium, but none of the grains had enough zirconium to be detected with CHILI. Nine grains were excluded from further consideration since they showed very little signals (<1000 counts) for strontium as well as for barium. Among the 11 remaining grains, we found three X grains. The discovery of three supernova grains among only 22 grains was fortuitous, because only ~1% of presolar silicon carbide grains are type X, but was confirmed by silicon isotopic measurements of grain residues with NanoSIMS. While one of the X grains showed strontium and barium isotope patterns expected for supernova grains, the two other supernova grains have $^{87}Sr/^{86}Sr < 0.5$, values never observed in any natural sample before. From their silicon isotope ratios, the latter two grains can be classified as X2 grains, while the former grain belongs to the more common X1 group. The differences of these grains in strontium and barium isotopic composition constrain their individual formation conditions in Type II supernovae.

Keywords: Presolar grains; Silicon carbide; Supernovae; Nucleosynthesis; Resonance ionization mass spectrometry (RIMS); Strontium isotopes; Barium isotopes

1. INTRODUCTION

Primitive meteorites and interplanetary dust particles contain small quantities of isotopically anomalous refractory dust grains that are older than our Solar System and commonly called "presolar grains" (Davis, 2011; Zinner, 2014). They condensed in the winds of evolved stars and in the ejecta of stellar explosions, i.e., they represent a sam-

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ple of stardust that can be analyzed in the laboratory. Presolar minerals identified to date include, e.g., diamond, silicon carbide (SiC), graphite, silicon nitride (Si_3N_4), oxides, and various types of silicates. Silicon carbide is the best characterized presolar mineral. Based on the isotopic compositions of carbon, nitrogen, and silicon, it is divided into distinct populations, namely, mainstream (the majority of SiC grains) and the minor subtypes AB, C, X, Y, Z, and nova grains. Mainstream, Y, and Z grains are from low-mass asymptotic giant branch (AGB) stars with about-solar (mainstream grains) and subsolar (Y and Z grains) metallicities. C and X grains are believed to come from Type II supernovae. The origin of AB grains is still a matter of debate; among the proposed stellar sources are J-type carbon stars (Lambert et al., 1986), born-again AGB stars (Asplund et al., 1999), and Type II supernovae (Liu et al., 2016).

CHILI, the Chicago Instrument for Laser Ionization is a new resonance ionization mass spectrometry (RIMS) instrument developed for elemental and isotopic analysis of small samples, such as dust returned to Earth by spacecraft and presolar grains from meteorites, at high spatial resolution and high sensitivity (Stephan et al., 2016). CHILI is especially suited for the analysis of trace element isotopic compositions in presolar grains. We used CHILI to measure the isotopic compositions of strontium, zirconium, and barium in 22 presolar SiC grains. These elements are particularly important for understanding *s*-process nucleosynthesis in AGB stars (Lugaro et al., 2003) because of their sensitivity to branching between neutron capture and β-decay. These elements are also particularly important since they have isotopes with magic neutron numbers, which makes them sensitive to the total neutron exposure. Strontium, zirconium, and barium have been measured before in presolar SiC with RIMS (Nicolussi et al., 1997, 1998; Savina et al., 2003a; Barzyk et al., 2007; Liu et al., 2014, 2015) using the CHARISMA instrument (Ma et al., 1995; Savina et al., 2003b) at Argonne National Laboratory.

Among the 22 SiC grains analyzed in the present study, three grains turned out to be X grains, as was confirmed by subsequent silicon isotopic analyses performed with the CAMECA NanoSIMS 50 at Washington University in St. Louis. Only a few X grains had been analyzed by RIMS for strontium, zirconium, and barium isotopes before, and those measurements suffered from relatively large uncertainties (Pellin et al., 1999, 2000, 2006; Davis et al., 2002). X grains are typically attributed to an origin in Type II supernovae (Amari et al., 1992; Davis, 2011; Zinner, 2014). According to a study by Hoppe et al. (1994), in which 720 SiC grains ranging in size from 1 to 10 μm, including 181 KJG grains, were analyzed, only ~1% of presolar SiC grains from the KJG size fraction from the Murchison meteorite, which was analyzed in the present study, should be X grains. Smaller size fractions contain up to 2% X grains (Hoppe et al., 2010). Assuming an X grain abundance of 1–2% among presolar SiC grains, the probability for finding three X grains out of 22 grains analyzed was calculated, using the binomial distribution, to be $(1.3 - 8.4) \times 10^{-3}$.

The new data obtained with CHILI and discussed here provide new insights into the nucleosynthetic processes preceding grain formation. We compare our results to models of supernova nucleosynthesis with a full nuclear network (Rauscher et al., 2002). Such models have been successfully used before to explain X grain properties, at least qualitatively, by involving extensive multizone mixing (Yoshida and Hashimoto, 2004; Yoshida, 2007), but have failed to quantitatively explain all observed X grain isotope systems so far (Lin et al., 2010; Hoppe et al., 2010, 2012; Davis, 2011; Zinner, 2014). Pignatari et al. (2013, 2015) recently presented new supernova models, in which a carbon- and silicon-rich zone at the bottom of the helium-burning shell could be the site for SiC grain formation without involving selective, large-scale mixing. In these one-dimensional models, the maximum temperature at the bottom of the helium-burning shell has a large impact on the isotopic signatures of such grains. Multidimensional models (Nomoto et al., 2013; Müller, 2016) taking into account the asymmetry of core-collapse supernovae (Grefenstette et al., 2014) should provide a more realistic picture in order to explain the wide range of isotopic compositions observed in supernova grains (Hoppe et al., 2018). The data presented here add new constraints to existing and to future models of supernova nucleosynthesis.

2. EXPERIMENTAL

2.1. Samples

Presolar SiC grains that had been extracted from the Murchison CM2 meteorite more than 20 years ago were analyzed in this study. The grains are from the KJG sample, which refers to the 1.5–3 μm size fraction of the KJ SiC separate (Amari et al., 1994). In contrast to recent work on Murchison SiC grains (Levine et al., 2009; Liu et al., 2014, 2015; Trappitsch et al., 2018), the samples in this study were not additionally treated with concentrated acids to remove contamination from parent-body or terrestrial material. The grains were mounted on a high-purity gold foil by depositing them from a suspension and pressing them into the gold with a sapphire window. Prior to RIMS analysis, energy-dispersive X-ray spectroscopy (EDS) images of the mount were acquired by scanning electron microscopy (SEM) to locate the SiC grains. From these images, 22 silicon-rich grains were randomly selected for analysis with CHILI.

2.2. RIMS measurements

A detailed description of the CHILI instrument and the analytical procedures has been given by Stephan et al. (2016). Here, we only provide a short summary of the analytical procedures, especially those specific to these measurements. The SiC grains were located using CHILI's integrated scanning electron microscope. The samples were ablated by a 351 nm wavelength laser beam from a frequency-tripled Nd:YLF desorption laser, focused to ~1 μm with a Schwarzschild-type optical microscope, and rastered over an area of about 10 × 10 μm^2. The beam