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The initial ${}^{41}Ca/{}^{40}Ca$ ratios in two type A Ca–Al-rich inclusions: Implications for the origin of short-lived ${}^{41}Ca$

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Abstract

This paper reports new ⁴¹Ca⁻⁴¹K isotopic data for two Type A CAIs, NWA 3118 #1Nb (Compact Type A) and Vigarano 3138 F8 (Fluffy Type A), from reduced CV3 chondrites. The NWA CAI is found to have carried live 41 Ca at the level of $(4.6 \pm 1.9) \times 10^{-9}$, consistent with the proposed Solar System initial 41 Ca $/{}^{40}$ Ca = 4.2 × 10⁻⁹ by Liu et al. (2012a). On the other hand, the Vigarano CAI does not have resolvable radiogenic ⁴¹K excesses that can be attributed to the decay of ⁴¹Ca. Combined with the ²⁶AI data that have been reported for these two CAIs, we infer that the ⁴ not homogeneous when ²⁶Al was widespread at the canonical level of ²⁶Al/²⁷Al = 5.2 × 10⁻⁵. Such a ⁴¹Ca heterogeneity can be understood under two astrophysical contexts: in situ charged particle irradiation by the protoSun in the solar nebula that had inherited some baseline ¹⁰Be abundance from the molecular cloud, and Solar System formation in a molecular cloud enriched in ²⁶Al and ⁴¹Ca contaminated by massive star winds. That said, more high quality ⁴¹Ca data are still needed to better understand the origin of this radionuclide.

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1. INTRODUCTION

The origin of shortest-lived radionuclides $(t_{1/2} \le 10 \text{ Myr})$ provides important constraints on the birth environment of the Sun, and is also a crucial piece of information if one intends to use these radioactivities for early Solar System chronology. From theoretical viewpoints, these extinct radioisotopes could come into the forming Solar System as a stellar product, either from a dying star (supernova, asymptotic giant branch star, or Wolf–Rayet star; see e.g., [Huss et al., 2009; Wasserburg et al., 2006;](#page--1-0) [Arnould et al., 2006](#page--1-0)) or from the background molecular cloud [Young \(2014\)](#page--1-0). Some radionuclides could also be a result of spallation-induced nuclear reactions between energetic charged particles (protons and alphas) and ambient gas/solid near the proto-Sun (e.g., [Gounelle et al., 2006](#page--1-0)).

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Radionuclides synthesized in the same process should in principle exhibit a concordant decay behavior.

The former existence of ${}^{41}Ca$, which decays to ${}^{41}K$ with a half-life of 0.1 Myr, can be in theory accounted for by either stellar nucleosynthesis or in situ irradiation. If ${}^{41}Ca$ was derived from outside the solar nebula, not only would this radionuclide be potentially useful for chronology, but its initial abundance could set an upper limit for the timespan between the collapse of the parental molecular cloud and Solar System formation. On the contrary, spallation near the proto-Sun would have resulted in a ${}^{41}Ca$ heterogeneity in the solar nebula, rendering chronological interpretations difficult. However, it has not been possible to favor one source over the other because the limited amount of data available for 41Ca has hindered our understanding of its initial abundance and distribution, and therefore its relationship with other short-lived radionuclides, in the early Solar System.

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The prior presence of ${}^{41}Ca$ is inferred through the detection of radiogenic ⁴¹K excesses (\equiv ⁴¹K*) in the oldest dateable Solar System materials, namely Ca–Al-rich Inclusions (CAIs). However, such measurements require analytically challenging mass spectrometry (see below; also see [Hutcheon et al., 1984; Srinivasan et al., 1996; Ito et al.,](#page--1-0) [2006; Liu et al., 2012a](#page--1-0) for more details). Earlier work on the 41Ca abundances in CAIs was performed on smallgeometry secondary ion mass spectrometers (SIMS, such as CAMECA ims-3f and 4f), and these workers discovered large ${}^{41}\text{K}^*$ correlating with the ${}^{40}\text{Ca}/{}^{39}\text{K}$ ratios of the phases in CV3 (Allende and Efremovka) Type B CAIs ([Hutcheon et al., 1984; Srinivasan et al., 1996; Sahijpal](#page--1-0) [et al., 2000](#page--1-0)) and of CM2 (Murchison) hibonite grains $(CaAl₁₂O₁₉)$ ([Sahijpal and Goswami, 1998; Sahijpal et al.,](#page--1-0) 2000). The results indicated that $4\degree$ Ca existed in the solar nebula with an initial abundance of ${}^{41}Ca/{}^{40}Ca \sim$ 1.4×10^{-8} . It was also found that short-lived ²⁶Al $(t_{1/2}=0.7$ Myr) was correlated with ⁴¹Ca in terms of presence or absence: when a CAI sample contains ⁴¹Ca/⁴⁰Ca \sim 1.4 \times 10⁻⁸, it is normally characterized by ⁴¹Ca/⁴⁰Ca ~ 1.4 × 10⁻⁸, it is normally characterized by 26 Al/²⁷Al = 5 × 10⁻⁵. On the other hand, samples (e.g., CM2-chondrite platy hibonite crystals) lacking $41K^*$ that could be due to the decay of 41 Ca are free of 26 Al ([Sahijpal and Goswami, 1998; Sahijpal et al., 2000](#page--1-0)). Given that ²⁶Al/²⁷Al = 5.2 \times 10⁻⁵ having characterized the early solar nebula (or at least the CAI forming region) could have been a result of external seeding of ²⁶Al followed by hydrodynamic mixing (e.g. [Boss, 2007, 2011\)](#page--1-0), the correlation between ${}^{41}Ca$ and ${}^{26}Al$ implies that ${}^{41}Ca$ was also derived from an external source and was cogenetic with 26 Al ([Sahijpal and Goswami, 1998](#page--1-0)).

Recent reanalysis with a large-geometry SIMS (CAMECA ims-1280HR2) of the CAI samples (Efremovka E44 and E65), in which ⁴¹Ca/⁴⁰Ca = 1.4×10^{-8} was pri-
marily inferred (Sriniyasan et al., 1996), revealed that their $\rm{^{41}Ca/^{40}Ca}$ ratios were 7–10 times lower than the previously determined value [\(Liu et al., 2012a](#page--1-0)). After correcting for the resetting time calculated from the sub-canonical 26 Al/²⁷Al ratios reported for these two inclusions ([Young et al.,](#page--1-0) [2005; Srinivasan and Chaussidon, 2013\)](#page--1-0), [Liu et al.](#page--1-0) [\(2012a\)](#page--1-0) found a converging 41 Ca/⁴⁰Ca ratio $\sim 4 \times 10^{-9}$, perfectly consistent with the ${}^{41}Ca/{}^{40}Ca$ value by [Ito et al.](#page--1-0) [\(2006\)](#page--1-0) in the Allende EGG3 CAI that is also characterized by canonical ²⁶Al/²⁷Al = (5.29 \pm 0.39) \times 10⁻⁵ [\(Wasserburg](#page--1-0) [et al., 2012\)](#page--1-0). Based on this apparent synchronicity between the two radionuclides, [Liu et al. \(2012a\)](#page--1-0) proposed that ⁴¹Ca/⁴⁰Ca \sim 4 \times 10⁻⁹ should be the best representative initial abundance in the early Solar System and that ²⁶Al and 41 Ca should have come into the Solar System together as stellar products.
It should be pointed out that the synchronicity between

 26 Al and 41 Ca was inferred based on the data reported for three CAIs, which are all Type B inclusions. Petrological examinations showed that Type Bs have complicated thermal histories, e.g., multiple-stage (partial) remelting and (partial) recrystallization. Sometimes such processing could make the ²⁶Al data difficult to decipher (e.g. [MacPherson](#page--1-0) [et al., 2012](#page--1-0)). Therefore, Type Bs are not good targets for

constraining the Solar System $4\degree$ Ca/ $4\degree$ Ca initial value. Instead, samples that appear to have escaped remelting since their formation, such as Fluffy Type A (FTA) CAIs, should be used. In hopes of further testing the concordant decay behavior of 26\AA l and 41Ca , samples that have subcanonical ²⁶Al/²⁷Al ratios corresponding to \sim (1–2) \times 10⁵ years compared to 5.2×10^{-5} would be needed. In this paper, we report the ${}^{41}Ca-{}^{41}K$ isotopic results of two Type A CAIs from reduced CV3 chondrites, NWA 3118 and Vigarano, and discuss possible origins of short-lived 41Ca.

2. EXPERIMENTAL

2.1. Samples

The samples used in this study are the NWA 3118 #1Nb (Compact Type A, ''CTA") and Vigarano 3138 F8 (FTA) CAIs. The NWA 3118 and Vigarano chondrites are reduced CV3; therefore, CAIs within them should have experienced less secondary processing than those in oxidized CV3 chondrites, such as Allende. Detailed petrological and mineralogical descriptions of the two CAIs can be found in [MacPherson et al. \(2012, 2013\)](#page--1-0). In general, the samples are petrologically free of secondary alteration and coarse-grained, making them suitable for the K isotope measurements. Recent high precision Mg isotope analyses of the two CAIs revealed well-defined 26Al isochrons with very little scatter, with NWA 3118 #1Nb and Vigarano 3138 F8 being characterized by 26 Al/²⁷Al = (4.64 \pm 0.09) \times 10⁻⁵ ($\tilde{\chi}^2$ = 1.5) and ²⁶Al/²⁷Al = (5.29 \pm 0.28) \times 10^{-5} ($\tilde{\gamma}^2 = 1.9$), respectively ([MacPherson et al., 2012;](#page--1-0) [MacPherson et al., 2013](#page--1-0)). The canonical 26 Al/²⁷Al ratio, along with the petrological texture of Vigarano 3138 F8, suggests that this CAI has never experienced postformation remelting and isotopic resetting (at least for Mg). The very tight isochron derived for the CTA indicates that no isotopic disturbance took place after the last melting event. The difference in the 26 Al/²⁷Al ratio between FTA and CTA can be translated to a \sim 100,000 year timespan, which is the half-life of ${}^{41}Ca$. Therefore, if the synchronicity between 26 Al and 41 Ca holds, one should expect that Vigarano 3138 F8 contains twice as much $^{41}Ca^{40}Ca$ as does NWA 3118 #1Nb.

2.2. Ion microprobe techniques

Measurements of K isotopes were carried out on the new CAMECA 1290 ion microprobe at UCLA by following the analytical protocol developed by [Liu et al.](#page--1-0) [\(2012a\)](#page--1-0). A polished sample was sputtered with a 10 nA, 23 keV ${}^{16}O^-$ primary beam ($\phi \sim 20$ µm). 25 min presputtering was applied to the sample prior to each spot analysis to minimize the surface contaminations. The field aperture with a width of 2500 μ m was used to block out scattered K ions from the edge of the analysis crater. The mass resolution ($M/\Delta M$) was set at 8000, which is sufficient to fully separate ${}^{41}\text{K}^+$ from ${}^{40}\text{CaH}^+$ and other minor interferences from the peaks of interest, but is incapable of resolving $(^{40}Ca^{42}Ca)$ ⁺⁺ from ⁴¹K⁺ (M/ $\Delta M = 34,000$ is required). Therefore, the contribution of $(^{40}Ca^{42}Ca)^{++}$ at $m/e = 41$

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