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^{48}Ca HETEROGENEITY IN DIFFERENTIATED METEORITES

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ABSTRACT

Isotopic heterogeneities of ^{48}Ca have been found in numerous bulk meteorites that are correlated with ^{50}Ti and ^{54}Cr anomalies among differentiated planetary bodies, and the results suggest that a rare subset of neutron-rich Type Ia supernova (nSN Ia) was responsible for contributing these neutron-rich iron-group isotopes into the solar system (SS). The heterogeneity of these isotopes found in differentiated meteorites indicates that the isotopic compositions of the bulk SS are not uniform, and there are significant amounts of nSNe Ia dust incompletely mixed with the rest of SS materials during planetary formation. Combined with the data of now-extinct short-lived nuclide ^{60}Fe, which can be produced more efficiently from an nSN Ia than a Type II supernova ejecta, the observed planetary-scale isotopic heterogeneity probably reflects a late input of stellar dust grains with neutron-rich nuclear statistical equilibrium nuclides into the early SS.

Key words: meteorites, meteors, meteoroids – nuclear reactions, nucleosynthesis, abundances

Online-only material: machine-readable table

1. INTRODUCTION

Isotopic anomalies of ^{50}Ti and ^{54}Cr have been reported in numerous bulk meteorites recently, and the ^{50}Ti and ^{54}Cr anomalies seem to have coupled within different planetary bodies (Trinquier et al. 2007, 2009). The distinct isotopic heterogeneity is due either to a significant amount of late stellar input(s) during the accretion of the solar system (SS), or to the galactic memory effect preserved in the carrier grains. The stellar source of these neutron-rich iron-group isotopes was most likely from a rare subset (~2%) of neutron-rich version of Type Ia supernovae (nSNe Ia; Woosley 1997). However, besides nSNe Ia, the observed ^{50}Ti and ^{54}Cr enrichments can also be produced via slow neutron capture process (*s*-process) in asymptotic giant branch (AGB) stars (Hoppe et al. 1994; Woosley et al. 2002; Lugaro et al. 2004). Furthermore, both *p*-process (e.g., ^{92}Mo) and *r*-process (e.g., ^{100}Mo) Mo isotope excesses have been found in differentiated meteorites, which may imply a relative deficit of *s*-process (e.g., ^{96}Mo) contribution to those differentiated parent bodies (Dauphas et al. 2002). Such a scenario will only work if ^{50}Ti, ^{54}Cr, and ^{96}Mo all came from the same stellar source.

In addition to ^{50}Ti and ^{54}Cr, the most neutron-rich stable isotope among the neutron-rich iron-group isotopes, ^{48}Ca, is a better candidate to verify whether a late input from an nSN Ia explosion was indeed responsible for the observed isotopic anomalies, since theoretical work suggested that ^{48}Ca cannot be produced in quantity in other stellar sources (Woosley 1997). Generally speaking, ^{48}Ca is synthesized under neutron-rich condition of nuclear statistical equilibrium process (nNSE; Hartmann et al. 1985), and both core-collapsed and accretion-induced supernovae explosion are able to produce ^{48}Ca. However, instead of ejecting into the interstellar medium, most of the ^{48}Ca synthesized by core-collapsed explosion will ultimately fall back into the collapsed star shortly after the explosion (K. Nomoto 2010, private communication). Consequently, a late injection of Type II supernova ejecta will not be able to generate observable enrichment of ^{48}Ca/^{44}Ca (Woosley & Weaver 1995; Nomoto et al. 2006). Alternatively, Woosley (1997) proposed that a cold carbon–oxygen white dwarf accreted at a slower rate could grow up to the Chandrasekhar mass, and then be ignited at a higher core density ($>2 \times 10^9$ g cm^{-3}) to become an nSN Ia. An nSN Ia generates significant amounts of neutron-rich iron-group nuclides, such as ^{48}Ca, ^{50}Ti, and ^{54}Cr, but relatively fewer other nuclides (Woosley 1997); therefore, a late input of an nSN Ia explosion to the SS can explain the observed ^{50}Ti and ^{54}Cr anomalies, while showing the negligible effect to other nuclides, e.g., ^{40}Ca, ^{44}Ca, and ^{48}Ti. It is also possible to synthesize ^{48}Ca through neutron capture process; however, a more significant anomalous effect of ^{46}Ca would be observed. Based on the coupled ^{48}Ca and ^{50}Ti anomalies observed in the Ca–Al-rich inclusions (CAIs; Niederer et al. 1981; Jungck et al. 1984), the estimated ^{48}Ca anomalous effect in achondrites is only about 2 epsilon units (ε; parts per ten thousand). This means that a sub-epsilon level of analytical precision is necessary to resolve the expected ^{48}Ca anomaly. Recently, Moynier et al. (2010) detected small isotope shifts in ^{48}Ca at the 2 s.d. (~3ε) level that may suggest the lack of complete mixing persisted even all the way to the time of planetesimal formation. In this Letter, high-precision Ca isotopic data of several differentiated meteorites are reported in order to test whether ^{48}Ca is indeed coupled with ^{50}Ti and ^{54}Cr anomalies in differentiated planetary bodies, as well as their potential stellar source and the mechanism that introduced them into the early SS.

2. RESULTS

We separated approximately 1 mg of Ca by conventional cation ion exchange column chemistry (Tera et al. 1970; Russell et al. 1978) from each dissolved 1–2 g whole rock sample residue left over from the W isotopic measurement (Lee et al. 2009). The blank contribution was negligible (less than 12 ng). Roughly ~2 μg of Ca was loaded onto an outgassed Re filament for Ca isotope measurement using the Triton, a thermal ionization mass spectrometry, at the Institute of Earth Sciences (IES). A double filament technique was used to obtain a Ca$^+$ ion current around 0.2–0.5 nA and >1.5 nA for two separate static runs to measure sequentially from mass ^{40}Ca to ^{44}Ca and ^{42}Ca to ^{48}Ca for about 1000 and 2000 s, respectively.

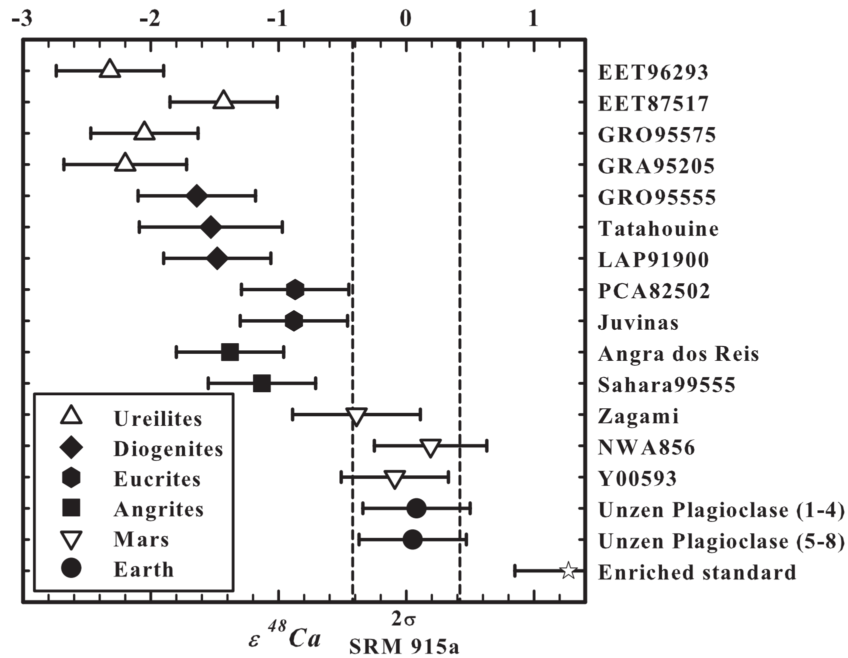

Figure 1. ε^{48}Ca of the mean for all the samples analyzed in this study. Data from Table 1, while the uncertainty quoted for each sample is the higher for either 2-fold standard deviation (2s s.d.) of the four replicate measurements or the long-term reproducibility of four replicated standard runs, which are shown by the dashed lines.

Table 1
Ca Isotopic Composition of Bulk Differentiated Meteorites, a Terrestrial Plagioclase, and a Man-made ^{48}Ca Enriched Standard*

Sample	ε^{40}Ca$^{\#}$	2σ	ε^{43}Ca$^{\#}$	2σ	ε^{46}Ca$^{\#}$	2σ	ε^{48}Ca$^{\#}$	2σ
Ureilites								
EET96293-1	−2.1	0.6	0.06	0.14	10.1	3.1	−2.75	0.19
EET96293-2	0.1	0.7	−0.06	0.17	14.7	4.6	−2.14	0.22
EET96293-3	−0.8	0.5	−0.09	0.14	−3.0	3.2	−2.08	0.20
EET96293-4	−1.3	0.8	−0.05	0.15	0.6	3.5	−2.32	0.21
Mean	−1.02	0.91	−0.03	0.06	5.61	8.20	−2.32	0.30
EET87517-1	−1.0	0.7	−0.01	0.15	3.3	3.5	−1.34	0.19

(This table is available in its entirety in a machine-readable form in the online journal. A portion is shown here for guidance regarding its form and content.)

The long-term instrumental reproducibility of each run, based on 68 repeated measurements of NIST SRM915a standard, of ^{40}Ca/^{44}Ca, ^{43}Ca/^{44}Ca, ^{46}Ca/^{44}Ca, and ^{48}Ca/^{44}Ca are 1.6, 0.31, 7.5, and 0.68ε (2σ s.d.), respectively, after normalizing to ^{42}Ca/^{44}Ca $= 0.31221$ (Russell et al. 1978). In general, four separate runs are done for each individual sample to ensure the reproducibility of the isotopic measurements, and the analytical uncertainty (2σ) can be further improved to 0.87ε, 0.13ε, 4.6ε, and 0.42ε for ^{40}Ca/^{44}Ca, ^{43}Ca/^{44}Ca, ^{46}Ca/^{44}Ca, and ^{48}Ca/^{44}Ca. An artificial standard enriched with 1.3ε of ^{48}Ca by mixture of NIST SRM915a and ^{48}Ca spike had been analyzed three times ($1.15 \pm 0.37\varepsilon$, $1.43 \pm 0.19\varepsilon$, and $1.24 \pm 0.50\varepsilon$, respectively) to verify the accuracy of our high-precision ^{48}Ca measurement.

Eight replicate measurements of a terrestrial volcanic plagioclase gave identical and consistent Ca isotopic compositions as the NIST SRM 915a standard (Table 1). All the differentiated meteorites analyzed in this study, including SNCs, angrites, eucrites, diogenites, and ureilites, have normal isotopic compositions of ^{40}Ca/^{44}Ca, ^{43}Ca/^{44}Ca, and ^{46}Ca/^{44}Ca within the analytical errors, but clearly resolvable ^{48}Ca/^{44}Ca deficits relative to the NIST SRM 915a, with the exception of SNC meteorites (Figure 1). The most deficit of ε^{48}Ca is preserved in ureilites (up to $-2.0 \pm 0.4\varepsilon$, the mean of all four ureilites measurements), and the ε^{48}Ca deficit gradually reduces in diogenites, angrites, and eucrites ($-1.55 \pm 0.10\varepsilon$, $-1.23 \pm 0.18\varepsilon$, and $-0.88 \pm 0.02\varepsilon$, respectively). Meteorites from the same group show comparable results, but a different group of achondrites yields different ^{48}Ca/^{44}Ca. Comparing the ^{48}Ca/^{44}Ca of this study with recently reported ^{50}Ti/^{48}Ti and ^{54}Cr/^{52}Cr data for the same differentiated meteorites (Trinquier et al. 2007, 2009) and the mean Cr isotopes of ureilites (Yamakawa et al. 2010; Qin et al. 2010), an apparent linear correlation of ε^{48}Ca: ε^{50}Ti: ε^{54}Cr $\approx$ 2.5:2.1:1 (Figure 2) can be resolved by using the least-squares fitting method (York 1966).

3. DISCUSSION

The results of this study demonstrate that there are well-resolved ε^{48}Ca deficits (up to 10σ effect) among different achondrites, while ε^{40}Ca, ε^{43}Ca, and ε^{46}Ca are homogeneous within the inner SS. Since the observed ^{48}Ca anomaly shows no correlation with other Ca nuclides, the neutron capture effect can be ruled out. Consequently, the most plausible explanation for the observed ^{48}Ca anomaly is the contribution from the nNSE process of an nSN Ia explosion.

The correlation among ε^{48}Ca, ε^{50}Ti, and ε^{54}Cr in differentiated meteorites (Figure 2) reflects the incomplete homogenization of the solar nebula between a component similar to the ureilite parent body (UPB), and a component with high ε^{48}Ca, ε^{50}Ti, and ε^{54}Cr, e.g., nSNe Ia, in the early stage of the solar evolution. Since the observed ε^{50}Ti and ε^{54}Cr anomalies are in the same level as the ε^{48}Ca anomaly, which most likely came from an nSN Ia, potential contributions from AGB stars for ^{50}Ti and ^{54}Cr can, therefore, be neglected. Even though the UPB has the most ε^{48}Ca deficits, there is insufficient data to constrain whether UPB truly reflects the mean composition of the SS prior to the addition of the nSNe Ia component; hence it is difficult to determine the nSNe Ia contributions quantitatively. Nevertheless, using the UPB as the upper limit of the original

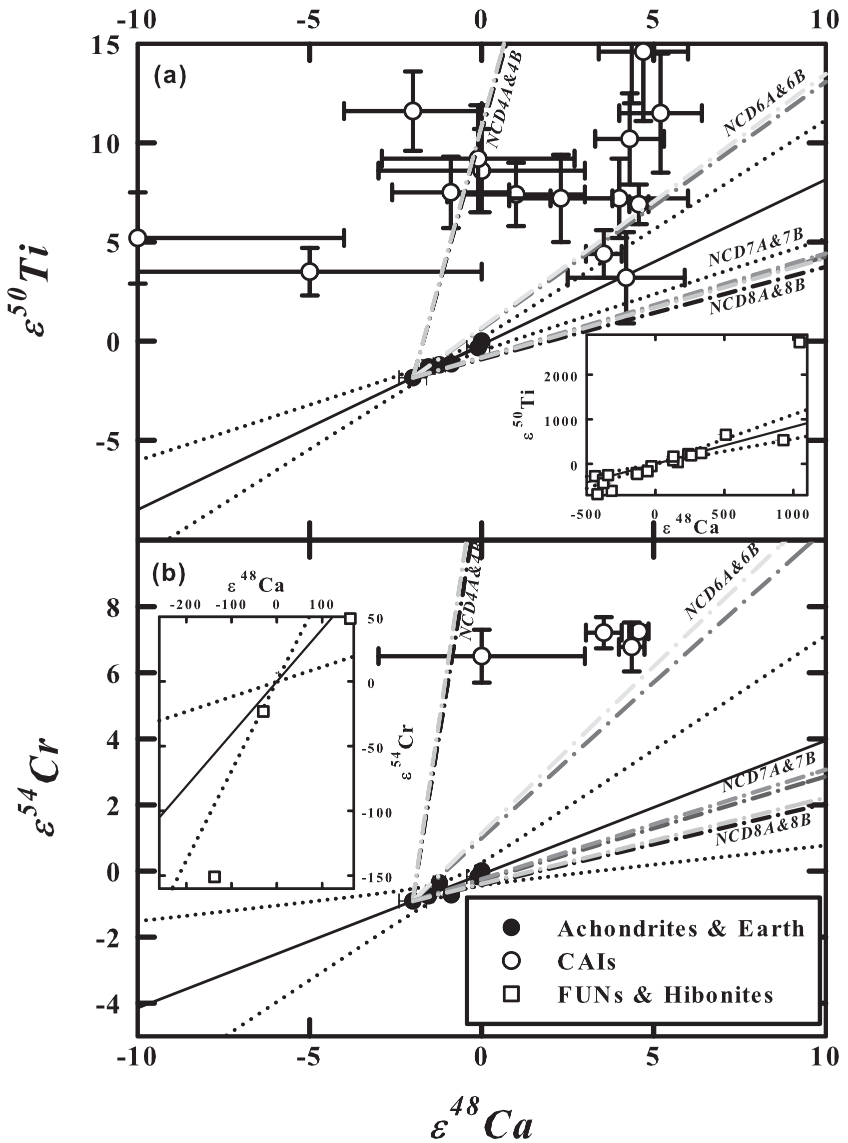

Figure 2. Plots of ε^{48}Ca vs. ε^{50}Ti (a) and ε^{54}Cr (b) anomalies of meteorites in the SS. The ε^{48}Ca data of differentiated meteorites are from this work (mean data in Table 1), while the ε^{50}Ti and ε^{54}Cr data are the mean of individual bulk meteorites (Trinquier et al. 2007, 2009; Yamakawa et al. 2010; Qin et al. 2010). Note that the ε^{48}Ca, ε^{50}Ti, and ε^{54}Cr data of each group of meteorites are not all from the same meteorites. As for the rest of CAIs, FUNs, and hibonites, the data are from the references listed in the text. All bulk-differentiated meteorites show well-resolved ε^{48}Ca, ε^{50}Ti, and ε^{54}Cr deficits relative to Earth, and a non-weighted linear correlation (solid lines) with the 95% confidence level (dot lines) can be constructed. The normal CAIs data are, however, not correlated with the linear regression line (black line) in the 95% confidence level (dot lines). Also shown are the FUN CAIs and hibonites with anomalous effects that also lie generally in the regression lines of differentiated meteorites. The dash-dotted lines are the calculated curves of an assumed initial solar composition (ureilites) mixed with different igniting density conditions (e.g., NCD6A) of nSNe Ia (Woosley 1997).

composition of the SS and the apparent correlation of ε^{48}Ca, ε^{50}Ti, and ε^{54}Cr, it is possible to place critical constraints for the nSNe Ia, e.g., core density. Using the isotopic abundance of the nSNe Ia ejecta of Woosley (1997), assuming the same efficiency of Ca, Ti, and Cr ejecta carriers captured by the SS, an nSN Ia explosion with an igniting density about 5.8–7.4 $\times$ 10^9 g cm^{-3} can account for the observed ε^{48}Ca, ε^{50}Ti, and ε^{54}Cr anomalies.

The SS is made of remnants with vastly different isotopic compositions ejected from many different stars (Anders & Zinner 1993; Zinner 2004), yet early theoretical and experimental works suggested that these materials were effectively homogenized in differentiated planetary bodies, despite that refractory grains can be preserved in primitive meteorites, e.g., CAIs and hibonites, from planetary bodies that had experienced very little thermal alteration (Podosek 1978). However, the heterogeneous ε^{48}Ca, ε^{50}Ti, and ε^{54}Cr observed in differentiated parent bodies provide stringent constraints that either the planetary formation and differentiation processes were ineffective to homogenize the SS completely, or an injection of these nNSE nuclides at a late stage of planetary formation prevented them from being mixed thoroughly in all planetary bodies. Although volatility control of carrier grains preservation can explain the ^{54}Cr anomaly (Trinquier et al. 2007), this will not work for refractory elements, such as ^{48}Ca and ^{50}Ti. The most direct and simplest scenario is a late input event of nNSE nuclides; however, stable isotopes cannot provide any time constraint.

Ordinary SNe Ia, with a central igniting density $< 2 \times 10^9$ g cm^{-3}, does not produce sufficient neutron-rich short-lived ^{60}Fe ($\sim 2.3 \times 10^{-9}$ solar mass; Nomoto et al. 1984). In contrast, an nSN Ia is capable of ejecting a significant amount of ^{60}Fe (on the order of 10^{-3} solar mass; Woosley 1997),

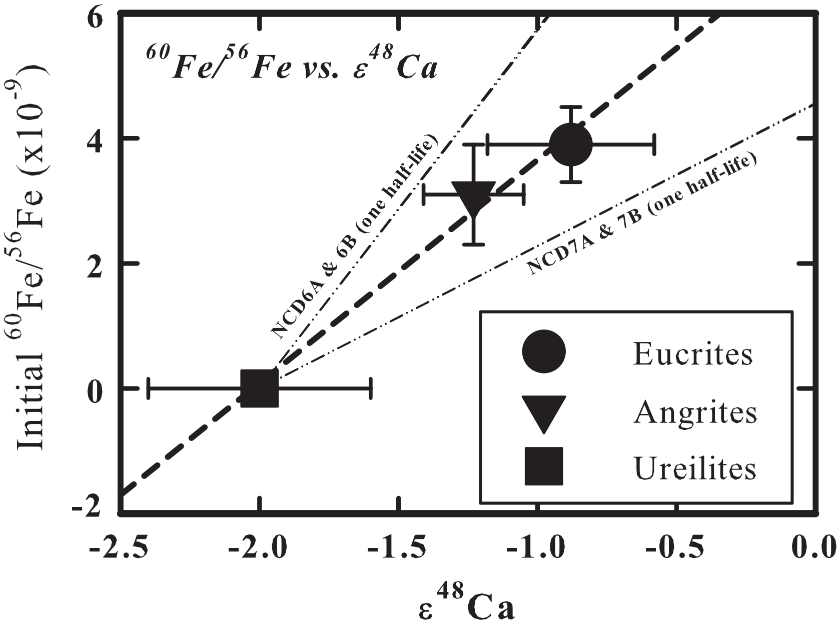

Figure 3. Plot of ε^{48}Ca anomaly vs. initial ^{60}Fe/^{56}Fe of different achondrites. ε^{48}Ca data are from the means of the same clast of achondrites in Table 1, and ^{60}Fe/^{56}Fe are from Quitté et al. (2010) and Shukolyulov & Lugmair (1993). The dashed line represents the linear correlation defined by the means of three achondrites, and the dash-dotted line is the mixing line for two nSNe Ia model calculations after one half-life of ^{60}Fe since the supernova exploded.

approximately one to three orders of magnitude more than SNe II explosions (Woosley & Weaver 1995; Woosley 1997; Iwamoto et al. 1999). Recently, live ^{60}Fe/^{56}Fe of 3.1 ± 0.8 × 10^{-9} was found in several bulk angrites measurements (Quitté et al. 2010). However, no live ^{60}Fe was identified in ureilites. Since ureilites also have the largest deficits of ^{48}Ca, ^{50}Ti, ^{54}Cr anomalies in all of the achondrites that have been studied, this is consistent if these nNSE nuclides (^{60}Fe included) all share a common origin. Comparing the initial ^{60}Fe/^{56}Fe and ε^{48}Ca results among differentiated meteorites, a linear correlation can be resolved (Figure 3), indicating that the ^{60}Fe might also be contributed from the nSNe Ia ejection, and all of the achondritic parent bodies should be formed within a short period.

Anomalous ε^{48}Ca (Lee et al. 1978, 1979; Jungck et al. 1984; Niederer & Papanastassiou, 1984; Papanastassiou & Brigham 1989), ε^{50}Ti (Niederer et al. 1981; Niemeyer & Lugmair 1981, 1984; Papanastassiou & Brigham 1989), and ε^{54}Cr (Birck & Allegre 1984; Papanastassiou 1986; Papanastassiou & Brigham 1989; Bogdanovski et al. 2002; Chen et al. 2010) have also been found in CAIs, and despite considerable variance, most of the CAIs are characterized with positive ε^{48}Ca, ε^{50}Ti, and ε^{54}Cr (Figure 2). It is well established that CAIs were among the earliest phases solidified from the accretion disk, while planetesimals and terrestrial planets formed subsequently through coagulation, runaway growth, and giant impacts. Albeit with larger uncertainties, CAIs are characterized with quite variable and also the largest excesses of nNSE nuclides; however, they seem to follow a different trend from that of the differentiated parent bodies. If the injection of these nNSE nuclides occurred just before the collapse of the solar nebula and the formation of CAIs, they would not have sufficient time to be mixed thoroughly throughout the accretion disk. With CAIs having formed early, followed by differentiated parent bodies, CAIs should have larger and more variable nNSE nuclides signatures relative to the larger and later-formed differentiated parent bodies. The different ε^{48}Ca, ε^{50}Ti, and ε^{54}Cr correlations between the CAIs and differentiated bodies can be attributed to the contribution of a late nucleosynthetic input that was responsible for the observed ε^{50}Ti and ε^{54}Cr in CAIs, or another late injection of nSNe Ia with an igniting density between 4 and 5.8 × 10^9 g cm^{-3} (Figure 2). Regardless of whether either of the above two scenarios is true, either scenario must have occurred after the injection of the initial nSNe Ia responsible for the nNSE nuclide signatures in differentiated bodies, and prior to the formation of first solid in the SS, in order to match the observed data. Given the rarity of an nSN Ia (Woosley 1997), the chances for two different nSNe Ia events to occur sequentially near the solar nebula during the early stages of the formation and differentiation of the SS are rather slim, although it is entirely possible. In addition to the observed ε^{50}Ti and ε^{54}Cr in CAIs, there are ample data supporting a late nucleosynthetic injection of various short-lived radionuclides, e.g., ^{26}Al, ^{53}Mn, ^{60}Fe, and ^{107}Pd. However, there is not yet any reliable constraint or general consensus about the timing of the input, or the actual nucleosynthetic site(s). In contrast, the refractory hibonite grains show larger ε^{48}Ca, ε^{50}Ti, and ε^{54}Cr anomalies that are roughly consistent with the correlation of differentiated meteorites observed from this study (Fahey et al. 1987; Ireland 1990; Hinton et al. 1987). It is believed that hibonites formed earlier than the CAIs, and thus the data are consistent with only one late nSNe Ia injection, and a second nucleosynthetic input occurred just prior to the formation of CAIs. If this is correct, the timing and the distance of the second nucleosynthetic event could be further constrained; however, these will be determined depending on better knowledge about the production rates and observed initial abundances of the short-lived radionuclides that are known to have originated from this event.

The observed ^{48}Ca heterogeneity clearly supports the presence of an nSN Ia contribution to the solar nebula. Using the solar abundance of Anders & Grevesse (1989), the mean ^{48}Ca/^{44}Ca of the SS can be modeled as a simple mixture of the original solar nebula (component P) and the late nSN Ia input (component N). It is not possible to constrain the original ^{48}Ca/^{44}Ca of the solar nebula with the current data; therefore, the lowest ^{48}Ca/^{44}Ca obtained from this study, the ureilites, is used as the component P, which represents a minimum estimate of the original solar composition, while the theoretical nSN Ia production rate for ^{48}Ca/^{44}Ca (Woosley 1997) is used for component N:

$$(^{48}\mathrm{Ca}/^{44}\mathrm{Ca})_{\mathrm{SS}} = (^{48}\mathrm{Ca}_{\mathrm{P}} + ^{48}\mathrm{Ca}_{\mathrm{N}} \times f_o)/(^{44}\mathrm{Ca}_{\mathrm{P}} + ^{44}\mathrm{Ca}_{\mathrm{N}} \times f_o),$$

where the f_o represents the fraction of nSNe Ia ejecta input the SS and is also known as the dilution factor (Wasserburg et al. 2006) to describe the efficiency of the SS acquiring the supernova dust from an isotopic ejecta envelope in order to evaluate the distance of a nearby stellar explosion event. Assuming that the SS is perpendicular to the explosion envelope with a diameter of about 60 AU (Neptune orbit) and that all the incoming stellar ejecta will be efficiently merged into the early SS, the derived dilution factor (1.5–6.7 $\times 10^{-9}$) is consistent with the distance of a nearby event at $\sim$ 0.9–1.9 pc from the SS, a reasonable distance to contribute SN ejecta without destroying the newly formed solar nebula (Adams 2010).

The quantitative estimation of ^{48}Ca heterogeneity observed in this study among various differentiated meteorites offers a stringent constraint about the presence of a late injection of rare nNSE isotopes to the SS. Incomplete mixing between the original solar nebula and the nSNe Ia ejecta due to the late arrival of the latter is the most direct and simple explanation for the results of this study. Alternatively, inefficient homogenization during the planetary formation process cannot be ruled out completely (e.g., slightly but not well-resolvable inconsistency between eucrites and diogenites which are thought to be from the same known asteroid, 4 Vesta), while more data and quantitative estimation are necessary in order to explore more complex scenarios.

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