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^{138}La ANOMALY IN THE EARLY SOLAR SYSTEM

Jason Jiun-San Shen and Typhoon Lee
Institute of Earth Sciences, Academia Sinica, Box 1-55, Nankang, Taipei 11529, Taiwan;
mountain@earth.sinica.edu.tw, typhoon@asiaa.sinica.edu.tw
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ABSTRACT

For every 1100 lanthanum atoms in the solar system, only one is ^{138}La. Relative to this low abundance, even a tiny additional ^{138}La made by irradiating its more abundant neighboring nuclides with energetic protosolar flare particles would cause a large, hence detectable, percentage increase in ^{138}La. Such early solar irradiation can produce many now-extinct short-lived radio nuclides (e.g., ^{41}Ca, ^{53}Mn, and ^{26}Al) and is the only way to make the newly discovered ^{10}Be and the possibly detected ^{7}Be, because stars destroy rather than produce Be. The alternative hypothesis to produce extinct nuclides is the injection of freshly synthesized radioactivity from a nearby asymptotic giant branch star or supernovae during solar system formation. Hoping to clarify the origin of extinct nuclides, we have been searching for ^{138}La excess and its possible correlation with extinct nuclides. Here we report the detection of up to 0.6% (~7.5 σ) ^{138}La excesses in five Allende calcium-aluminum–rich inclusions. Surprisingly, they do not correlate with ^{26}Al, thus offering no support for making ^{26}Al by early irradiation. Instead, ^{138}La excess correlates with ^{50}Ti excess. Current nucleosynthesis models produce ^{50}Ti in a rare subset of Type I supernovae whose core underwent significant gravitational collapse before carbon deflagration. Our observed correlation thus suggests that ^{138}La also came from these rare sources, perhaps in the mantle of the white dwarf, by reactions induced by the neutrino burst emitted during core neutronization. After the explosion, ^{138}La was incorporated into Ti-rich dusts that later became the building material of our solar system.

Subject headings: nuclear reactions, nucleosynthesis, abundances — Sun: flares

1. INTRODUCTION

^{138}La is a rare (^{138}La/^{139}La $= 0.0009079 \pm 7$) radio nuclide that decays into ^{138}Ce and ^{138}Ba with a half-life (106 Gyr) so long that over the age of the solar system, only 1.5% of it decayed. It is one of the very few naturally occurring "odd (proton)–odd (neutron)" nuclides whose origin has long been a mystery (Goriely et al. 2001). It is shielded from both β^+ and β^- decays and is not on the slow neutron-capture path. So it cannot be produced by *r*-, *s*-, or conventional (i.e., proton capture) *p*-process nucleosynthesis that successfully accounts for almost all nuclides beyond the iron group. Because of its unpaired nucleons, ^{138}La is less well bound than its neighboring nuclides, thus easy to destroy but hard to produce inside stars. That is why its abundance is thousands of times less than the neighboring *r*- or *s*-nuclides and 10 times less than the rare *p*-nuclides. So far, four processes have been proposed for the production of ^{138}La: (1) irradiation of interstellar material by a high-abundance component of galactic cosmic rays (Hainebach, Schramm, & Blake 1970) whose energy is too low to reach the earth, (2) nuclear reaction caused by energetic photons from stellar thermonuclear fusion before they are thermalized (Harrison 1976), (3) the new *p*-process that relies on high-temperature (~10^9 K) thermal photons to dissociate high *Z* nuclei (may be enhanced by a prior *s*-process) into low *Z* nuclei (Woosley & Howard 1978), and (4) neutrino-induced spallation reactions (Woosley et al. 1990). Most astrophysicists now believe that the first two are not viable because their yields are too small to even account for ^{138}La's low abundance. The most up-to-date effort to model the photodissociation production showed that the yield for ^{138}La would probably be still too low when the rest of the *p*-process nuclides were made in solar proportions (Goriely et al. 2001). However, under special circumstances, with ad hoc mixing it may still have an outside chance to make this process viable (Cameron 2003; Fujimoto et al. 2003). The neutrino reactions, especially captures by ^{138}Ba, are thought to be the most promising (Goriely et al. 2001; Woosley et al. 1990) but in need of better determination of relevant cross sections. The site of the photodissociation is believed to be the O/Ne zone inside massive stars. The same zone may also be the site for neutrino spallation when the core neutronizes.

2. METHOD

Detecting La isotopic anomalies is technically challenging, because to measure ^{138}La/^{139}La ~ 1/1100 to 1 per mil precision requires reducing the background and interferences to much less than 1 part per mil (ppm). Also, La has only two natural isotopes, hence no extra isotope ratio to be normalized for correcting the large and variable mass fractionation during thermal ionization mass spectrometry. We previously developed a novel way for La isotope analysis using LaO ions instead of the La element. It enables not only a high ionization efficiency of greater than 3% but also fractionation correction by normalizing to the oxygen isotope ratios (Shen, Lee, & Chang 1992). Using this method, we have previously found evidence of small La anomalies (around 1.2 per mil $= 3\ \sigma$), whole rock samples of the Allende and Murchison carbonaceous chondrites but not in ordinary chondrites (Shen, Lee, & Chang 1994). Since then we have further refined our technique by reducing the lab contamination blank to 10 pg and measuring all LaO isotopes except the main peak at ^{139}La^{16}O on secondary electron multipliers in pulse counting mode instead of Faraday cups. Our present method allows calcium-aluminum–rich inclusion (CAI) weighing, typically 7 mg and containing about 85 ng La, to be analyzed with 2 σ precision of 1.6 per mil initially but later improved to 0.8 per mil.

CAIs weighing about 7 mg each were dissolved in concentrated HNO_3 and HF and then converted to chloride form. Rare earth elements (REEs) were first separated from all other elements in an ion exchange column with TRU-Spec Resin (sup-

TABLE 1
Isotopic Anomalies in ^{138}La, ^{50}Ti, ^{46}Ca, and ^{48}Ca and Initial ^{26}Al and ^{10}Be for CAIs

CAI Allende	$\delta(^{138}La/^{139}La)$* (per mil)	$(^{26}Al/^{27}Al)_0$ ($\times 10^{-5}$)	$\delta(^{50}Ti/^{48}Ti)$ (per mil)	$(^{10}Be/^{9}Be)_0$ ($\times 10^{-4}$)	$\delta(^{46}Ca/^{44}Ca)$ (per mil)	$\delta(^{48}Ca/^{44}Ca)$ (per mil)
Sample 1	6.0 ± 1.6	…	…	…	…	…
Sample 2	−0.8 ± 1.6	…	…	…	…	…
	0.4 ± 1.6	…	…	…	…	…
Sample 3	3.1 ± 1.6	…	…	…	…	…
	3.2 ± 1.6	…	…	…	…	…
USMN 3898	4.4 ± 1.6	4.5 ± 0.7 (1)	1.16 ± 0.20 (5)	…	2 ± 3 (7)	−0.2 ± 0.4 (7)
3529-Z	3.0 ± 0.8	4.0 ± 0.1 (2)	0.96 ± 0.26 (6)	7.6 ± 2.6 (2)	…	…
EGG-3	2.6 ± 1.6	4.9 ± 0.5 (3)	0.69 ± 0.10 (5)	…	…	0.3 ± 0.2 (7)
	2.3 ± 0.8	…	…	…	…	…
EGG-6	−0.1 ± 1.6	4.4 ± 0.6 (4)	0.44 ± 0.12 (5)	…	…	…

Notes.—*δ: per mil deviations relative to the terrestrial normal (Ames La metal). The errors are 2 σ. The numbers in parentheses indicate that the data are from (1) Armstrong, Hutcheon, & Wasserburg 1984; (2) McKeegan et al. 2001; (3) Podosek et al. 1991; (4) G. J. Wasserburg & G. R. Huss 2002, private communication; (5) Niederer et al. 1985; (6) Niemeyer & Lugmair 1981; and (7) Niederer & Papanastassiou 1984.

plied by ElChroM). Then La was separated from other REEs by the 2-MLA chromatography technique. Using reagents purified by subboiling distillation, we reduced our total procedure blank from 2500 to 10 pg. The chemical yield of the entire procedure was better than 80%. Before loading onto the filaments, the extracted La was exposed to intense UV light for ~20 minutes to oxidize and vaporize the hydrocarbons to improve the ionization efficiency of La.

La was analyzed as LaO by using MAT-262Q TIMS as detailed before (Shen et al. 1992, 1994). Briefly, signals at 154, 155, and 157 amu were measured so that $^{138}La/^{139}La$ was obtained from 154/155 ($^{138}La^{16}O/^{139}La^{16}O$), $^{18}O/^{16}O$ fractionation normalization from 157/155 ($^{139}La^{18}O/^{139}La^{16}O$), and consistency plus linearity checks for $^{138}La/^{139}La$ from the mixed ratio 157/154. For the smaller samples analyzed here, it is crucial to diligently guard against artifacts caused by interferences and backgrounds. So we always carefully scanned the mass spectrum between 145.5 and 164.5 amu using multipliers in the pulse counting mode both before and after data acquisition. For meteoritic samples, small signals of ^{153}BaF (10 counts s^{-1} for $^{134}Ba^{19}F$) and ^{158}NdO (2200 counts s^{-1} for $^{142}Nd^{16}O$) were found when $^{138}La^{16}O$ was about 60,000 counts s^{-1}. The isotopic pattern for the mass region between 158 and 164 amu fits the isotopic pattern of NdO perfectly. Therefore, the upper limit for $^{142}Ce^{16}O$ at 158 amu can be inferred with some certainty that in turn implies an insignificant upper limit at $^{138}Ce^{16}O$ interference for $^{138}La^{16}O$. We believe that these NdO signals originated not from poor chemical separation but from the TIMS itself, since it has been our workhorse for a large number of Nd analyses of sizable geological samples. A possible interference that cannot be monitored was $^{141}Pr^{16}O$. However, the residue from incomplete chemical separation should have more Ce than Pr because Ce is closer to La in atomic number. So we can also infer an upper limit for PrO by assuming that PrO < CeO, and this limit implies a negligible PrO interference at 157 amu (Shen et al. 1994). BaF contributions at 154 and 157 amu were estimated to be about 0.5 and 2.3 per mil, respectively, based on monitoring $^{134}Ba^{19}F$ at 153 amu. The consistency between the $^{138}La/^{139}La$ inferred from 157/154 and 154/155 means that the correction is reasonable. Background contribution of the tails of the large ^{155}LaO peak was significant on the small ^{154}LaO peak; however, throughout our experiments the contribution of 155 tailing at 154 amu was always less than 1 ppm of the ^{155}LaO signal. We have carefully established that the background due to the 155 amu tail at 154 amu was roughly 3 times that at 153.5 amu that was monitored to correct the tail effect at 154 amu. A typical background correction at 154 amu was ~0.5 per mil. Overall correction at mass 154 amu was typically 1 per mil, which should be good to at least 20%, thus implying an uncertainty of 0.2 per mil. Therefore, we believe that our ^{138}La anomaly is not an artifact owing to interference or background.

We have carried out extensive tests in order to establish the size of uncertainty of our results. The early error estimate of 1.6 per mil was initially based on the 2 σ reproducibility of the normal ratios over about a year. Eight analyses of the solution prepared from high-purity La metal plus two runs of the USGS standard granite G2 were carried out. This estimate is broadly consistent with the isotopic differences measured for the four pairs of separately prepared repeat samples from three CAIs (1.2, 0.1, and 0.3 per mil, respectively) and G2 (0.5 per mil). The grand mean of the four differences was about 0.5 per mil. Two artificially enriched standards were prepared by adding highly enriched ^{138}La to normal La to generate standards with expected offsets of 4.4 and 7.7 per mil based on gravimetry. Our mass spec results for them were 5.6 and 7.9 per mil, respectively, implying that we can detect isotope shifts at the 4–8 per mil level with an average error of about 0.7 per mil. These data demonstrated that the 2 σ precision of our ^{138}La measurements using the method described here was about 1.6 per mil except the two CAIs studied last (i.e., 3529 Z and EGG-3), for which our reproducibility was improved to 0.8 per mil based on eight repeats of the La metal standard over several weeks.

3. RESULTS AND DISCUSSION

We have so far studied seven CAIs (Table 1). The first three were small (~10 mg) millimeter-sized Ca-rich but otherwise uncharacterized Allende inclusions from our own collection. They were analyzed mainly to validate our analytical method. We then turned to four large well-studied CAIs with much published data to look for ^{138}La correlation. USNM-3898 is a type-A CAI containing mostly melilite, spinel, and hibonite, whereas 3529-Z, EGG-6, and EGG-3 are type-B CAIs consisting primarily of melilite, Ti-rich clinopyroxene, and anorthite, as well as spinels. Five out of the seven have well-resolved (>5 σ) positive anomalies (Table 1). No negative anomalies were found so far. The largest ^{138}La excess among them was 0.6% (about 7.5 σ), and the average is 0.27%. Strictly speaking, the positive anomaly in $^{138}La/^{139}La$ can be caused by either an ^{139}La deficit or an ^{138}La excess. However, the latter is favored because (1) there are good reasons to expect ^{138}La anomalies from either early solar flare irradiation or the fluctuation of a small number of rare dusts originated from unusual stars, (2) it would be difficult to have anomalies in a major nuclide such as ^{139}La that is produced by both *r*- and *s*-processes without anomalies in other major nuclides made in the same processes (e.g., ^{137}Ba, ^{138}Ba, ^{142}Nd, ^{145}Nd, etc., which are not detected; McCulloch & Wasserburg 1978), and (3) the number of atoms that has to be added to cause a few per mil excess in ^{138}La is much smaller than that needed to be destroyed to cause a few per mil deficit in ^{139}La.

In Figure 1 and Table 1, we summarize the data on ^{138}La, ^{26}Al, and ^{50}Ti plus a few $^{46,\,48}$Ca, ^{10}Be. All four CAIs have well-behaved ^{26}Al isochrones, indicating the initial presence of ^{26}Al at the canonical abundance level of $^{26}\mathrm{Al}/^{27}\mathrm{Al} \sim 4.5 \times 10^{-5}$ (Armstrong, Hutcheon, & Wasserburg 1984; Podosek et al. 1991; McKeegan et al. 2001; G. J. Wasserburg & G. R. Huss 2002, private communication) when they last solidified in the early solar system. Neither their Mg nor Al data show any sign of later disturbances or possibly nonuniform initial ^{26}Al/^{27}Al, which is common among other CAIs (Podosek et al. 1991). These four CAIs have ^{138}La excesses that vary from -0.1 to 4.4 per mil, thus obviously not correlating with the nearly constant initial ^{26}Al.

Let us consider first the most naïve model in which both ^{26}Al and ^{138}La* (* = excess made by irradiation) were made by irradiation as a single component then added to the normal solar system matter. In such cases, constant ^{26}Al/^{27}Al would be associated with constant ^{138}La excess, and a variable ^{138}La excess should correlate with ^{26}Al/^{27}Al variation. Our data thus rule out such models because CAIs have constant ^{26}Al/^{27}Al but variable ^{138}La excesses. On the other hand, it is instructive to recall that we can detect only the difference in the ^{138}La* produced by early solar irradiation, whereas the absolute initial abundance of ^{26}Al can be inferred even when everything was homogenized. Therefore, more sophisticated models can probably be devised to account for constant ^{26}Al but variable ^{138}La*. Moreover, the most recent study of ^{10}Be indicated that it does not correlate with ^{26}Al in CAIs (Marhas, Goswami, & Davis 2002). So ^{26}Al may not be produced by irradiation at all but did come from an asymptotic giant branch star or supernovae. Therefore, it will be interesting to search for ^{10}Be in the remaining three CAIs if their boron isotopes were not disturbed by later alteration.

Clearly resolvable ^{50}Ti anomalies from 0.44 to 1.16 per mil for the four large CAIs have been reported before (Niederer, Papanastassiou, & Wasserburg 1985; Niemeyer & Lugmair 1981). There seems to be a positive correlation between the ^{138}La and the ^{50}Ti excesses in Figure 1. This observation has several far-reaching implications on the origin of ^{138}La and the mixing of presolar components into the normal solar system material.

^{50}Ti is the most neutron-rich isotope of Ti. Similar neutron-rich isotopes in the iron peak region including ^{48}Ca, ^{54}Cr, ^{58}Fe, and ^{66}Zn were known to show correlated anomalies in CAIs (cf. Völkening & Papanastassiou 1990 and references therein). These nuclides are the main products of the neutron-rich version of the nuclear statistical equilibrium (NSE). This process occurs near the central core of stars where temperature is high enough (greater than or equal to a few times 10^9 K) so that the abundances are determined almost entirely by thermodynamic equilibrium that depends not on reaction rates but on temperature and nuclear binding energy for the nuclides. However, it is called "nuclear statistical" instead of thermal because

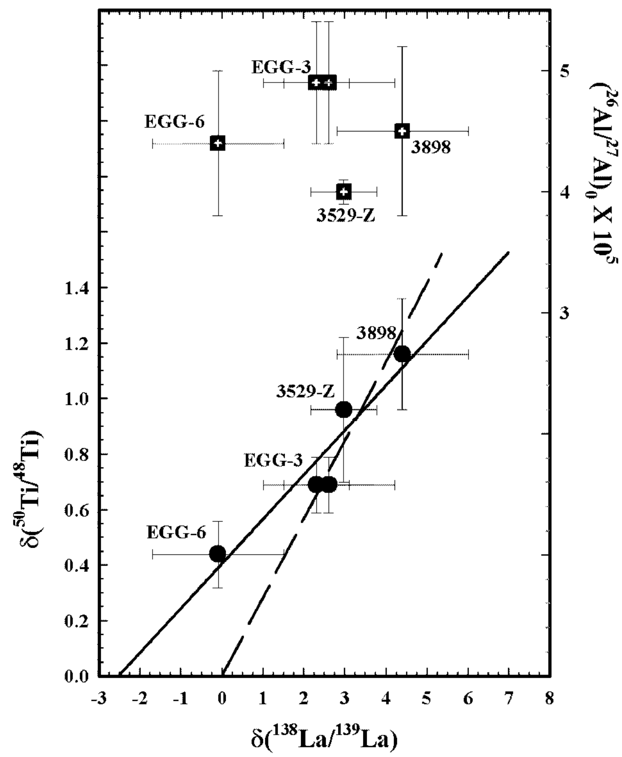

FIG. 1.—Plot of ^{50}Ti anomaly (*bottom panel, left axis*) and initial ^{26}Al/^{27}Al (*top panel, right axis*) vs. ^{138}La anomaly for the four large and well-characterized Allende CAIs. They all show ^{26}Al/^{27}Al close to the canonical ratio of 5×10^{-5} but very different δ^{138}La from -0.1 to 4.4 per mil (*filled square*). Therefore, the ^{138}La variations were not consistent with the expectation from models in which ^{26}Al and ^{138}La were coproduced by flare irradiation. However, there seems to be a correlation between ^{50}Ti anomalies and ^{138}La anomalies (*filled circles*). The observed spread of $\pm 4\varepsilon$ in ^{50}Ti/^{48}Ti agrees with the expectation of $\pm 3\varepsilon$ because of the fluctuation of rare grains of ^{50}Ti and ^{138}La from a rare subset of SN Ia. Note that the broken line is forced to go through the origin. If we relax that constraint, then the least-squares fit (*continuous line*) would not go through the origin, suggesting that there may be another uncorrelated source for ^{50}Ti.

weak interaction is too slow to equilibrate, thus requiring one additional parameter (such as the neutron-to-proton ratio) to characterize it. The neutron-rich version of NSE operates when the increasing density causes stronger electrostatic repulsion between protons so that more of them capture electrons to become neutrons and emit neutrinos. So, the natural consequence of the neutronization process is to have an associated burst of neutrinos. The currently favored site for NSE is a rare subset of Type Ia supernovae (SNe Ia; Woosley 1997). The most accepted current model for SNe Ia is the sudden nuclear burning when a carbon-oxygen white dwarf ignites under degeneracy because the mass transfer from its companion star drives its mass beyond the Chandrasekhar limit. So the white dwarf collapses toward a neutron star, and the carbon and oxygen fuse violently all the way to iron. For a rare subset of SNe Ia, the ignition starts slowly and the thermonuclear fusion front moves at subsonic speed, then the white dwarf has time to collapse to a high-density and high-temperature state before the star explodes. Such a SN Ia would make NSE products including ^{50}Ti and disperse them, since the collapse of the core toward high density would cause neutronization, which in turn should emit neutrinos. It seems logical to infer that the observed ^{50}Ti-^{138}La correlation implies that ^{138}La should have also come from these special SNe Ia and was produced by neutrino-induced reaction on ^{138}Ba and light REEs. Moreover, such a source should be very rare, perhaps only 1/50 of the SN Ia rate, because otherwise it will produce too much iron with a strange composition to the Galaxy.

It is likely that the SN Ia only partially fused to iron, leaving the outer one-third still mainly C and O. Therefore, one of the best carrier grains for Ti would be perovskite (CaTiO3), assuming O/C > 1 and the NSE material can be mixed into the O-rich zone when condensation finally occurred in the SN Ia ejected. If we assume that each such grain weighs 1 pg (10^{-12}) and all Ti is ^{50}Ti, then each dust should have $\sim 3 \times 10^{-13}$ g ^{50}Ti. A moderately sized CAI may weigh 10 mg and contain 1.3% TiO_2. The total ^{50}Ti atoms then weigh around 4×10^{-6} g, so that a CAI contains on the order of 10 million $CaTiO_3$ dust grains from the rare special type of SNe Ia. If the distribution of grains follows Poisson statistics, then we would expect that ^{50}Ti abundance may vary by $1/\sqrt{10^7} = \pm 3 \times 10^{-4}$ typically. This compares favorably with the spread of $\pm 4 \times 10^{-4}$ anomaly for the CAIs studied. CAIs usually are enriched in REEs at a level 20 times the chondritic abundance. So, they contain 12 ppm La equilibrated to $\sim 10^{-10}$ g of ^{138}La, since $CaTiO_3$ is usually rich in REEs, perhaps containing 500 ppm La (Wark & Boynton 2001). So in the 1 pg $CaTiO_3$ grain, there is 5×10^{-16} g ^{139}La. Therefore, the ^{138}La/^{139}La for the carrier grain would be 50, which means that ^{138}La is enriched by a factor of 22 in the source. The enrichment factor of neutrino-induced ^{138}La is estimated to be 5–36, depending on neutrino flux for SNe II (Goriely et al. 2001). For a reduced neutrino flux by a factor of 3 in SNe Ia, the enrichment can be up to a factor of 12, which is in reasonable agreement with the factor of 22 estimated here. These estimates show that with very reasonable values for the parameters, we have a plausible scenario in explaining the spread of ^{138}La anomalies as the fluctuation rare small grains from unusual stars. However, the correlation line between δ^{138}La and δ^{50}Ti should go through the origin if they both came exclusively from the same source. The marginally significant offset of 0.04% δ^{50}Ti at 0 δ^{138}La may mean that parts of ^{50}Ti came from a different source.

4. CONCLUSION

We have succeeded in measuring the ^{138}La/^{139}La ratio of individual CAIs with a 2 σ precision less than 1 per mil. Five out of seven CAIs studied so far show ^{138}La excesses up to 6 per mil (about 7.5 σ). For four CAIs with known ^{26}Al, the ^{138}La excesses do not correlate with ^{26}Al. The lack of correlation between ^{26}Al and this sensitive refractory recorder of spallation by energetic particles offers no support for producing ^{26}Al inside the early solar system by flare irradiation. However, it will be important to check for ^{138}La-^{10}Be correlation, because Be (McKeegan, Chaussidon, & Robert 2000; Sugiura, Shuzou, & Ulyanov 2001; Marhas, Goswami, & Davis 2002; Chaussidon, Robert, & McKeegan 2002; Srinivasan 2002) can come only from energetic particle irradiation, while at least part of ^{26}Al was from outside the solar system and ^{26}Al does not seem to correlate with ^{10}Be (Marhas et al. 2002). Surprisingly, the ^{138}La excesses correlate with ^{50}Ti excesses, suggesting that these two rare nuclides were made by the same source and carried to the solar system by the same presolar dust grains. Since ^{50}Ti is thought to have been made by neutron-rich nuclear statistical equilibrium in a rare subset of SNe Ia (Woosley 1997), our newly discovered correlation then implies that ^{138}La was also made there. So our result may solve the long-standing mystery of the nucleosynthetic origin of the rare odd-odd nuclides such as ^{138}La. The most likely production mechanism is reaction induced by the burst of neutrinos from the neutronization of the core of the collapsing white dwarf before carbon deflagration blows it apart. An obvious candidate carrier grain is perovskite ($CaTiO_3$), which is known to be highly enriched in rare earth elements including La. The fluctuation of a small number of such rare ^{48}Ca^{50}Ti $^{16}O_3$ grains would explain the range of the anomalies and the correlation among ^{48}Ca, ^{50}Ti, and ^{138}La anomalies. This hypothesis thus provides important insight toward the mixing of distinct nucleosynthetic components into the solar system normal composition.

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