138La-138Ce-136Ce nuclear cosmochronometer of the supernova neutrino process

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The ¹³⁸La ($T_{1/2} = 102$ Gyr)-¹³⁸Ce-¹³⁶Ce system is proposed to be used as a nuclear cosmochronometer for measuring the time elapsed from a supernova neutrino process. This chronometer is applied to examine a sample affected by a single nucleosynthesis episode as presolar grains in primitive meteorites. A feature of this chronometer is to evaluate the initial abundance ratio of ¹³⁶Ce/¹³⁸Ce using an empirical scaling law, which was found in the solar abundances. We calculate the age of the sample as a function of isotopic ratios $^{136}Ce/^{138}Ce$ and 138 La/ 138 Ce and evaluate the age uncertainty due to theoretical and observational errors. It is concluded that this chronometer can work well for a sample with the abundance ratio of $^{138}La/^{138}Ce \geq 20$ when the ratios of 136Ce/138Ce and 138La/138Ce are measured within the uncertainty of 20%. The availability of such samples becomes clear in recent studies of the presolar grains. We also discuss the effect of the nuclear structure to the *ν* process origin of 138La.

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I. INTRODUCTION

Long-lived radioactivities are used as nuclear cosmochronometers for dating the ages of nucleosynthesis episodes, metal-poor stars $[1,2]$, and the Milky Way $[3]$. Rutherford suggested the original idea, the uranium chronometer, in 1929 [\[4\]](#page-4-0). Recent progress of astronomical observations has enabled us to detect actinoid elements of early generations of stars [\[1,2\]](#page-4-0). A fraction of the uranium in a metal-deficient star CS 31082-001 was measured and the stellar age was estimated with the 238 U chronometer for the first time in 2001 [\[1\]](#page-4-0). Only six chronometers with half-lives of 1–100 Gyr that are suitable for measuring cosmological ages were known. The other chronometers are $40K$ and $232Th$, suggested by Burbidge *et al.* [\[5\]](#page-4-0), 87Rb and 187Re, suggested by Clayton [\[6\]](#page-4-0), and 176Lu, suggested by Audouze *et al.* [\[7\]](#page-4-0). It should be noted that these chronometers measure the age of slow or rapid neutron-capture reaction nucleosynthesis (*s* or *r* process). However, there is no cosmochronometer for neutrino-induced reactions in supernova explosions (*ν* process).

The *ν* process was suggested as the origin of heavy and light elements $[8-15]$ and is of importance for studying neutrino spectra from the supernovae $[11,12]$ and for discussing neutrino oscillation [\[15\]](#page-4-0). Among many heavy elements, only two isotopes of 138La and 180Ta are considered to be synthesized primarily by the *ν* process [\[8,11\]](#page-4-0), but there is another possibility for 180Ta that is produced by the *s* process $[16]$. The solar abundance of 138 La was quantitatively explained by the *ν* process in theoretical calculations [\[11,17\]](#page-4-0). Thus, 138La is a key for understanding the *ν* process. In this article, we propose a new cosmochronometer based on the abundance ratios of the three isotopes, 138 La, 138 Ce and 136 Ce, in a single sample affected strongly by a single nucleosynthesis episode. Recent studies of presolar grains in primitive meteorites provide samples affected strongly by single supernova nucleosynthesis episodes [\[18,19\]](#page-4-0). The isotopic patterns of heavy elements such as molybdenum and barium in the silicon-carbide (SiC) presolar grains originating from the supernovae have been measured [\[20,21\]](#page-4-0). On the basis of these technological developments, the abundances of ¹³⁶*,*138Ce and 138La in a single sample as the presolar grain are expected to be determined in the future.

II. PROPOSAL OF 138La- 138Ce- 136Ce NUCLEAR COSMO CHRONOMETER

The meta-stable isotope 138 La decays to 138 Ce or 138 Ba with a half-life of 102 Gyr. The 138 La- 138 Ba (or 138 Ce) system has been used as a geochronometer to date the ages of rocks [\[22\]](#page-4-0). Audouze *et al.* pointed out that the ¹³⁸La system is not useful as the cosmochronometer because the abundance of the daughter nucleus populated by the decay of 138La may be miner relative to its initial abundance [\[7\]](#page-4-0). However, nucleosynthesis models cannot predict generally the initial abundances of individual presolar grains, because the supernovae produce both of 138La and 138Ce and their initial abundance ratios in individual grains are different. After the destroy of pre-existing 138La and 138Ce (and 136Ce) by the weak *s* process in evolutionary stages of progenitors, these nuclei are resynthesized in different layers in the supernova explosion; 138La is synthesized by the *ν* process in He- and C-rich layers [\[11\]](#page-4-0) and ¹³⁶*,*138Ce are synthesized by the γ process in O/Ne layers [\[23,24\]](#page-4-0). Meteorite compositions are produced in stellar outflows after mixture of materials originating from different layers, which may include both the Ce isotopes and 138La. The mixing ratios of the different layers in the individual meteorite compositions are different.

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FIG. 1. A partial nuclear chart and nucleosynthesis flows around 138La. The nucleus 138La is shielded by stable isobars against *β* decay and is produced by neither the *s* nor the *r* process. In the *ν* process, ¹³⁸La is mainly synthesized from ¹³⁸Ba via the charged current reaction. Two *p* nuclei, ¹³⁶Ce and ¹³⁸Ce, are mainly produced from an *s* nucleus 140 Ce via successive (γ , *n*) reactions.

Therefore we should evaluate the initial abundances in each sample by an alternative method.

Here we propose a novel empirical method to estimate the initial abundance of ¹³⁸Ce. A similar empirical method was suggested for a ¹⁴⁶Sm nuclear cosmochronometer, wherein an initial abundance of 146Sm is evaluated by the solar abundance pattern [\[25\]](#page-4-0). In our proposed method, the initial abundance of 138Ce can be calculated by an empirical scaling law for *p* nuclei, which was found in the solar abundances [\[26\]](#page-4-0). The three isotopes, 138 La, 138 Ce, and 136 Ce, are classified to the *p* nuclei. The *p* nuclei are located on the neutron-deficient side from the *β*-stability line and their isotopic fractions are small (typically 0.1–1%). There are nine pairs of the two *p* nuclei with the same atomic number. As shown in Fig. 1, the first and second *p* nuclei are lighter than the *s* nucleus by two and four neutrons, respectively. We found the empirical scalings that the abundance ratios of N_{\odot} (1st *p*)/ N_{\odot} (2nd *p*) and $N_{\odot}(s)/N_{\odot}$ (1st *p*) are almost constant over a wide range of atomic number, respectively, where N_{\odot} is the solar isotope abundance [\[26\]](#page-4-0). These scalings are a piece of evidence that the most probable origin of the *p* nuclei is the supernova *γ* process. In addition, we found a novel concept of "the universality of the *γ* process," wherein the scalings hold for nuclides produced in individual supernova *γ* processes under various astrophysical conditions [\[26\]](#page-4-0). We calculated the γ process of core-collapse supernovae under various astrophysical conditions, such as metallicity, progenitor mass, and explosion energy, and found that the scalings hold for individual nucleosyntheses independent of the astrophysical conditions assumed [\[27\]](#page-4-0). We presented a detailed mechanism why the scalings fold for various supernovae in our previous articles [\[27,28\]](#page-4-0). Our proposed chronometer is based on the universality of the scaling for the two *p* nuclei.

Here we improve the empirical scaling for the two *p* nuclei. There are nine pairs of the two *p* nuclei in the solar system, but we omit three elements of Mo, Ru, and Er in Fig. 2 because the origin of *p* nuclei of Mo and Ru is considered to be different from other *p* nuclei $[13,14,26]$ and a *p* nucleus ¹⁶⁴Er is largely contributed from the *s* process [\[29\]](#page-4-0). The N_{\odot} (1st p)/ N_{\odot} (2nd p) ratios in the other six elements increase with increasing the atomic number (see Fig. 2). We obtain a function,

$$
R_{pp}(Z) = 0.0581 \times Z - 2.18,\tag{1}
$$

using a χ^2 fitting. As shown in Fig. 2, the observed N_{\odot} (1st p ^{$)/N_{\odot}$ (2nd *p*) ratios are described well by this equation and} the standard deviation of N_{\odot} (1st *p*)/ N_{\odot} (2nd *p*) is only 9%.

If abundance ratios of $N(^{138}La)/N(^{138}Ce)$ and $N(^{136}Ce)/N(^{138}Ce)$ in a sample affected strongly by a single supernova are known, the age *T* elapsed from the supernova episode is calculated by

$$
T = -\frac{T_{1/2}(\text{^{138}La})}{\ln 2} \ln \left\{ \frac{N(\text{^{138}La})/N(\text{^{138}Ce})}{N(\text{^{138}La})/N(\text{^{138}Ce}) + \frac{1}{b}[1 - R_{pp}(Ce) \times N(\text{^{136}Ce})/N(\text{^{138}Ce})]} \right\},
$$
(2)

where *N* is the observed abundance in the sample, R_{pp} is the initial $N(1st\ p)/N(2nd\ p)$ ratio at the freeze-out of the *γ* process, and *b* is the branching ratio of the *β*[−] decay of ¹³⁸La: $[\lambda(\beta^-)/(\lambda(\beta^-) + \lambda(EC))]$. The scaling holds for nuclides produced by individual supernova nucleosyntheses and the function (1) would hold for individual presolar grains. The $R_{pp}(Ce)$ ratio is estimated to be 1.19 using the function (1). Therefore, the age can be obtained from the two observed ratios of $N(^{138}La)/N(^{138}Ce)$ and $N(^{136}Ce)/N(^{138}Ce)$ in the sample.

A part of ¹³⁸La may be contributed from the γ process, but the age obtained from this chronometer is not affected by the contamination of the γ process, because this chronometer measure the age elapsed from a supernova and both the *γ* and *ν* processes occur at the same supernova. The cosmic ray process [\[30\]](#page-4-0) also has been suggested as the origin of

FIG. 2. Observed isotope ratios of the first *p* nuclei to the second *p* nuclei with the same atomic number. The solid line is the result of *χ*² fitting. The standard deviation of the fitting is 9%.

FIG. 3. Calculated age as a function $R_{pp} \times N(^{136}Ce)/N(^{138}Ce)$. The age is the time elapsed from a nucleosynthesis episode to the present, which includes the solar system age 4.6 Gyr. The solid curves of (a), (b), (c), and (d) are the calculated ages with mixing ratios $N(^{138}La)/N(^{138}Ce) = 20, 10, 5,$ and 1, respectively. The dot-dashed line is the age of $T = 10$ Gyr. The solid, dashed, and dotted lines are the upper and lower limits arisen from uncertainty of R_{pp} or $N(^{136}Ce)/N(^{138}Ce)$ for (a), (b), and (c), respectively.

¹³⁸La. However, the presolar grains originating from the supernovae do not contain the comic ray products, because the pre-existing 138La and ¹³⁶*,*138Ce originating from the interstellar media, which include the cosmic ray products, are destroyed by the weak *s* process before the supernovae. The *p* nuclei that include the three isotopes are located on the neutron-deficient side from the *β*-stability line, and hence they cannot be synthesized by neutron capture reactions in the weak *s* process. However, the pre-existing *p* nuclei are destroyed by the neutron capture reactions. Therefore, the three isotopes in the presolar grains from the supernovae are newly synthesized by single nucleosyntheses without any pre-existing contamination such as the cosmic ray products.

Here we discuss the uncertainty of the age using the chronometer. Figure 3 shows the calculated ages as a function of R_{pp} ·N(¹³⁶Ce)/N(¹³⁸Ce) under the mixing ratio $N(^{138}La)/N(^{138}Ce) = 1, 5, 10, and 20. We also calculated$ the ages as a function of $N(^{138}La)/N(^{138}Ce)$ under the ratio $N(^{136}Ce)/N(^{138}Ce) = 1.5, 2.0,$ and 3.0 (see Fig. 4). The

FIG. 4. Calculated age as a function $N(^{138}La)/N(^{138}Ce)$. The age is the time elapsed from a nucleosynthesis episode. The solid curves of (a), (b), and (c) are the calculated ages with the ratios $N(^{138}Ce)/N(^{136}Ce) = 1.5, 2.0,$ and 3.0, respectively. The dot-dashed line is the age of $T = 10$ Gyr. The solid, dashed, and dotted lines are the upper and lower limits of the assumed uncertainty of $N(^{138}La)/N(^{138}Ce)$ for (a), (b), and (c), respectively.

uncertainty of the age is generally due to theoretical and observational errors. In the present proposed chronometer, the theoretical errors come only from the uncertainty of the initial abundance ratio of $R_{pp}(Ce)$. The uncertainty of R_{pp} (Ce) is estimated from the deviation of the observed N_{\odot} (1st p ^{$)/N_{\odot}$ (2nd *p*) ratios (see Fig. [2\)](#page-1-0). The standard deviation of} the observed ratios is 9% and we take 9% as the uncertainty of $R_{pp}(Ce)$. The observational errors are due to the errors of the $N(^{136}Ce)/N(^{138}Ce)$ and $N(^{138}La)/N(^{138}Ce)$ ratios in samples. Recently, measurement techniques with mass separation have been rapidly developed and the isotopic fractions of heavy elements such as Mo and Ba in SiC grains originating from the supernovae have been measured within the uncertainties of $7-15\%$ [$20,21$]. Thus, we consider two cases that the observational errors of both the two ratios are 10 or 20%. Finally, we calculate the complete uncertainty due to both the theoretical and observational errors, namely the errors of R_{pp} , $N(^{136}Ce)/N(^{138}Ce)$, and $N(^{138}La)/N(^{138}Ce)$. If the ratios of $N(^{136}Ce)/N(^{138}Ce)$ and $N(^{138}La)/N(^{138}Ce)$ are measured with the uncertainty of 10%, respectively, the complete uncertainty is about 20, 50, or 90% in the case of $N(^{138}La)/N(^{138}Ce)$ = 20, 10, or 5. With observational errors of 20%, the complete uncertainty turns out to be about 30% for a sample of $N(^{138}La)/N(^{138}Ce) = 20$. It should be noted that the complete uncertainty suddenly decreases with increasing the ratio of $N(^{138}La)/N(^{138}Ce)$.

Recent progress of the astronomical observation has enabled us to detect actinoid elements of metal-poor stars. The ages of these metal-poor stars have been evaluated with U and Th chronometers within uncertainty of $~\sim 50\%$; [\[1,2\]](#page-4-0). Therefore the present proposed chronometer is as useful as the cosmochronometers have been used in the astronomy.

The discussion mentioned above shows that the present proposed chronometer can work well if the presolar grains of $N(^{138}La)/N(^{138}Ce) \ge 20$ are found with the observational errors of 20%. A question we have to ask here is whether the presolar grains of $N(^{138}La)/N(^{138}Ce) \ge 20$ exist or not in nature. Because the solar abundance ratio N_{\odot} (138 La)/ N_{\odot} (138 Ce) is about 0.14, it seems to be difficult to find the samples of $N(^{138}La)/N(^{138}Ce) \ge 20$. However, recent studies of the presolar grains originating from the supernovae suggest that such samples are available. The carbon-rich grains of the SiC grains and low-density graphite grains are chemically condensed in a carbon-rich environment of C/O *>* 1. This suggests that the main component of the carbon-rich grains originating from the supernovae are produced in their C-rich layers, where 138La is synthesized by the *ν* process. In contrast, the contribution of O/Ne layers, where ¹³⁶*,*138Ce are produced by the γ process, is considered to be small. The isotopic fractions of several elements in individual presolar grains from supernovae were measured and theoretical calculations were performed to reproduce these isotopic fractions by proper mixture of materials produced in different layers [\[31,32\]](#page-4-0). The observed isotopic fractions were reproduced consistently without the contribution from the O/Ne layers [\[31\]](#page-4-0) and with the small contribution from the O/Ne layers that is smaller than that from the C-rich layers by 2–4 orders of magnitude [\[32\]](#page-4-0). We stress again that ^{136,138}Ce are dominantly synthesized in the O/Ne layers by the γ process. Therefore, we expect that

FIG. 5. Synthesis and destruction of 138La in the *ν* process. Lowspin excited states such as $J^{\pi} = 1^{+}$ are strongly populated from ¹³⁸Ba by Gamow-Teller transitions. If the 1^+ isomer exists, these excited states strongly decay to the 1^+ isomer that decays to daughter nuclei.

samples with the abundance ratio of $N(^{138}La)/N(^{138}Ce) \ge 20$ will be found. Finally, we would like to stress the complete uncertainty suddenly decreases with increasing the ratio of $N(^{138}La)/N(^{138}Ce)$. The discussion mentioned above shows that we can find samples, of which the $N(^{138}La)/N(^{138}Ce)$ ratios are much larger than 20, and thereby the complete uncertainties of the ages of the sample become much lower than 30%.

Although recent studies of the astrophysics suggest that 138La may be dominantly synthesized by the *ν* process $[11,17]$, the origin of 138 La has not been elucidated in astronomical observations or meteorite analyses. To take a hypothetical example, suppose that 138La is dominantly synthesized by the cosmic-ray process and the presolar grains are formed in an interstellar environment. The age estimated by this chronometer would be longer than that in the case where 138La is dominantly synthesized by the *ν* process because the cosmic-ray nucleosynthesis is a continuing process from the Galaxy formation to the solar system formation. Thus, the age measured in the presolar grains is of importance in understanding the origin of 138La.

III. EFFECT OF THE NUCLEAR STRUCTURE TO THE *ν* **PROCESS ORIGIN**

Here we point out that the nuclear structure of 138 La may affect the performance of the chronometer and the *ν* process origin of 138 La. A fact that the spin and parity of the ground state of an isotone ¹⁴⁰Pr ($N = 81$) is $J^{\pi} = 1^+$ suggests that a 1^+ excited state may exist at a low excitation energy in 138 La. Although a 1^+ state at 296 keV in 138 La was reported [\[33\]](#page-4-0), the lowest 1^+ state has not been established experimentally. If this 1^+ excited state exists at energy lower than 72 keV that is the energy of the first excited state, the 1^+ state may be a β unstable isomer because β decay can compete with an internal *E*4 transition. In such a case, our proposed clock cannot work well because the initial abundance cannot be estimated by using the scaling. In addition, this isomer affects the *ν* process origin of 138La because 138La synthesized by the *ν* process is destroyed via the isomer (see Fig. 5).

We would like to stress that the observed 1^+ ground state of 140Pr cannot be understood by a simple shell-model picture that would lead to the ground state with $J^{\pi} = 2^{+}$ or 3^{+} [\[34\]](#page-4-0). This indicates that there is a mechanism decreasing the energy

FIG. 6. The calculated and measured levels in 140Pr and 138La. The observed levels of ¹⁴⁰Pr and ¹³⁸La are taken from Ref. [\[36\]](#page-4-0) and Refs. [\[34,35\]](#page-4-0), respectively.

of the 1^+ state. To examine the existence of the 1^+ isomer, we calculate the nuclear structures of ¹³⁸La and ¹⁴⁰Pr in a shell-model. In the mass region around 138 La, the last proton (neutron) can occupy either $d_{5/2}$ or $g_{7/2}(d_{3/2})$ orbit around the Fermi surface. The 1^+ and 5^+ states in 138 La and 140 Pr can be understood generally by $(\pi d_{5/2}) \otimes (\nu d_{3/2})^{-1}$ and $(\pi g_{7/2}) \otimes$ $(vd_{3/2})^{-1}$ configurations, respectively, where π (*v*) is proton (neutron). Figure 6 show the calculated results. We

successfully describe the observed excited states in ¹³⁸La [\[34,](#page-4-0) 35] and ¹⁴⁰Pr [\[36\]](#page-4-0) lower than 500 keV and, in particular, the ground state spin of 140Pr. We find that the coupling of twoand four-particle excited configurations of $\pi (g_{7/2})^{-2} (d_{5/2})^3$ and $\pi (g_{7/2})^{-4} (d_{5/2})^5$ leads to the 1^+ ground state. A calculated 1⁺ state in 138La locates at 169 keV. This result suggests that the 1^+ state in 1^{138} La is not an isomer but an experimental determination is desired.

IV. CONCLUSION

In summary, we propose a novel ¹³⁸La ($T_{1/2} = 102$ Gyr)-138Ce-136Ce nuclear cosmochronometer for measuring the time elapsed from the supernova *ν* process. We introduce a novel method using the universality of the empirical scaling for p nuclei to evaluate the initial abundance of 138 Ce. The chronometer is applied to examine a sample affected by single supernova nucleosyntheses as presolar grains in primitive meteorites. Recent studies of the presolar grains provide isotopic fractions of several different elements of single samples. The chronometer can work well for a sample, of which its abundance ratio of $^{138}La^{138}Ce$ is larger than 20, when the ratios of $^{136}Ce/^{138}Ce$ and $^{138}La/^{138}Ce$ are measured within the uncertainty of 20%. The availability of such samples becomes clear in recent studies of the presolar grains. With the chronometer, we can measure both the age of the supernova associated with neutrino-driven wind and the isotopic abundance patterns of elements synthesized by the supernova. We present that the energy of the lowest 1^+ state may affect the chronometer performance and the *ν* process origin of 138La, and its measurement is desired.

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