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Diffusion of Oxygen Isotopes in Thermally Evolving Planetesimals and Size Ranges of Presolar Silicate Grains

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Abstract

Presolar grains are small particles found in meteorites through their isotopic compositions, which are considerably different from those of materials in the solar system. If some isotopes in presolar grains diffused out beyond their grain sizes when they were embedded in parent bodies of meteorites, their isotopic compositions could be washed out, and hence the grains could no longer be identified as presolar grains. We explore this possibility for the first time by self-consistently simulating the thermal evolution of planetesimals and the diffusion length of ^{18}O in presolar silicate grains. Our results show that presolar silicate grains smaller than $\sim 0.03\,\mu$m cannot keep their original isotopic compositions even if the host planetesimals experienced a maximum temperature as low as 600 °C. Since this temperature corresponds to that experienced by petrologic type 3 chondrites, isotopic diffusion can constrain the size of presolar silicate grains discovered in such chondrites to be larger than $\sim 0.03\,\mu$m. We also find that the diffusion length of ^{18}O reaches ~ 0.3–$2\,\mu$m in planetesimals that were heated up to 700–800°C. This indicates that, if the original size of presolar grains spans a range from $\sim 0.001\,\mu$m to $\sim 0.3\,\mu$m like that in the interstellar medium, then the isotopic records of the presolar grains may be almost completely lost in such highly thermalized parent bodies. We propose that isotopic diffusion could be a key process to control the size distribution and abundance of presolar grains in some types of chondrites.

Key words: diffusion – meteorites, meteors, meteoroids – planets and satellites: formation

1. Introduction

Primitive meteorites contain unique tiny materials called presolar grains. The fundamental property of the presolar grains is their isotopic compositions, which deviate significantly from extremely homogeneous values of materials in the solar system. In particular, presolar silicate grains are identified via the oxygen isotopic ratios of ^{17}O/^{16}O and ^{18}O/^{16}O (e.g., Clayton & Nittler 2004). Since such isotopic anomalies must have been preserved over the entire history of the solar system (which is why we can currently measure the differences in isotopic composition), investigation of presolar grains can provide us with important clues to understand the formation and evolution of the solar system.

It is considered that the presolar grains originally formed in nearby stars during post-main-sequence phases such as supernovae and/or asymptotic giant branch (AGB) stars. They were then transported to the presolar nebular, which is a preliminary stage to the formation of the Sun, and were finally incorporated into planetesimals that are the parent bodies of meteorites. Some theoretical studies suggest that dying stars could inject relatively large grains with radii of 0.1–1 μm into the interstellar medium (ISM, e.g., Nozawa et al. 2007; Yasuda & Kozasa 2012). On the other hand, while these grains were traveling to the presolar nebula, many of them might fragment into grains smaller than 0.1 μm as a result of shattering in interstellar turbulence (Hirashita & Yan 2009). It is nonetheless important to point out that such small ($<0.1\,\mu$m) presolar grains have rarely been detected in the currently available samples; the typical size distribution of presolar silicate grains is from ~ 0.1 to $\sim 1\,\mu$m (e.g., Zinner 2003; Hynes & Gyngard 2009; Nguyen et al. 2010; Leitner et al. 2012; Hoppe et al. 2015).

The abundance of presolar grains varies among different petrologic types of meteorites: in general, presolar grains are most abundant in type 3, but their abundance decreases as the type number increases from 3 to 6 (Huss 1990; Huss & Lewis 1995). These types are based on the degree of metamorphism experienced by meteorites when they were embedded in planetesimals. Since the metamorphism depends on the temperature, petrologic types are regarded as representing the peak temperature experienced by the planetesimals. For instance, it is widely accepted that the ordinary chondrites of petrologic type 3 experienced a peak temperature less than 700°C, that experienced by type 6 was higher than 800°C, and types 4 and 5 fall between them (Huss et al. 2006). Thus, this classification suggests that the abundance of presolar grains may be related to the thermal history of planetesimals.

It is implicitly presumed that metamorphism totally erases the isotopic records of presolar grains. This may be because metamorphism creates new minerals or crystalline structures by breaking the original atomic bonds and forming new ones. If metamorphism were the dominant process in washing out the isotopic compositions, more presolar grains would be discovered in unmetamorphosed (primitive) chondrites. While the least metamorphosed type 3 chondrites have the highest abundance of presolar grains among chondrites (Nguyen et al. 2007, 2010; Floss & Stadermann 2009, 2012; Nittler et al. 2013), it is still much lower than that in interplanetary dust particles (IDPs), which are regarded as the most primitive materials (Messenger et al. 2003; Floss et al. 2006; Busemann et al. 2009). Therefore, metamorphism would not explain the difference in abundance of presolar grains between type 3 chondrites and IDPs. On the other hand, even if the chemical compositions of minerals are not changed through metamorphism, the replacement of one atom by another—so-called atomic diffusion—could occur in slightly thermalized planetesimals. Therefore, it can be anticipated that atomic diffusion can also delete the original isotopic composition possessed by presolar grains in their parent bodies.

In this paper we explore this possibility for the first time by computing the diffusion length of ^{18}O in presolar silicate grains in meteorites and by comparing it with the actually measured size of presolar grains. Since the diffusion length is sensitive to temperature, we numerically simulate thermal evolution of planetesimals with different radii and formation times. We find that the diffusion process of oxygen atoms can regulate the size distribution of presolar silicate grains and that only grains larger than $\sim$0.3 μm can keep their original isotopic properties in chondrites of types 4–6. Our results also suggest that, even in type 3 chondrites that experienced a peak temperature of only 600°C, atomic diffusion can entirely erode the isotopic records of presolar silicate grains smaller than $\sim$0.03 μm. This may be viewed as a potential explanation of why the currently available samples of presolar silicate grains in type 3 chondrites have a size larger than $\sim$0.05 μm. Thus, we conclude that isotopic diffusion is one of the important processes governing the survival of presolar grains in thermally evolving planetesimals.

2. Isotopic Diffusion in Thermally Evolving Planetesimals

In order to compute the diffusion length of oxygen isotopes in presolar silicate grains, we adopt a diffusion coefficient of ^{18}O in olivine from Dohmen et al. (2002). The diffusion coefficient $D(T)$ [m^2 s^{-1}] is given as

$$D(T) = 10^{-8.34}\exp(-3.38 \times 10^5/RT), \tag{1}$$

where R is the gas constant [J K^{-1} mol^{-1}] and T is the temperature [K]. Since the diffusion coefficient is obtained for a temperature range of $1100°\mathrm{C} < T < 1500°\mathrm{C}$ (Dohmen et al. 2002), we extrapolate the results down to lower temperatures. Measurements with a transmission electron microscope show that the chemical compositions of presolar silicate grains vary on the scale of a few tens of nanometers (Nguyen et al. 2007; Busemann et al. 2009; Leitner et al. 2012). While it would be interesting to see how the diffusion coefficients depend on stoichiometry of elements in silicate, there are no data for silicate compositions with a variety of Mg/O, Fe/O, and Si/O ratios. Thus, we assume that grains have a homogeneous composition of Mg-rich forsterite as used in Dohmen et al. (2002).

As can be seen from Equation (1), the diffusion coefficient depends strongly on the temperature. Thus, we need to numerically simulate the thermal evolution of planetesimals to reliably estimate the diffusion length of ^{18}O in silicate. We assume that decay energy of the short-lived radioisotope ^{26}Al heats up materials within a planetesimal, as most thermal modeling studies have done (e.g., Miyamoto et al. 1982). A heat conduction equation,

$$\rho c\frac{\partial T}{\partial t} = \frac{1}{r^2}\frac{\partial}{\partial r}\left(r^2 K\frac{\partial T}{\partial r}\right) + A\exp(-\lambda t), \tag{2}$$

is solved numerically based on Wakita et al. (2014), where t is time measured from the formation time of planetesimals t_0, r is the distance from the center of the planetesimals, A is the radiogenic heat generation rate per unit volume, and λ is the decay constant of the radionuclides. We adopt physical parameters of thermal conductivity $K = 2$ [J s^{-1} m^{-1} K^{-1}], density $\rho = 3300$ [kg m^{-3}], and specific heat $c = 910$ [J kg^{-1} K^{-1}] (Yomogida & Matsui 1983; Opeil et al. 2010). We assume that these parameters do not depend on the temperature. The radius and formation time of planetesimals, both of which are expected to affect the maximum temperature they reach (see Figure 8 in Wakita et al. 2014), are parameterized; the formation time of planetesimals t_0 is determined based on a formation time of Ca–Al-rich inclusions (CAIs), 4567 Myr ago, when they had the initial ratio of $^{26}\mathrm{Al}/^{27}\mathrm{Al} = 5.25 \times 10^{-5}$ in the solar nebula (e.g., Connelly et al. 2012). The radiogenic heat generation rate A falls in direct proportion to the initial ratio of $^{26}\mathrm{Al}/^{27}\mathrm{Al}$, which depends on the formation time of planetesimals t_0 (see Figure 15 in Wakita & Sekiya 2011). Since the abundance of ^{26}Al decreases with time (its half-life is 0.72 Myr), a planetesimal formed at a later time has fewer heating sources than one formed earlier.

A diffusion length (L) is calculated as $dL^2 = D(T)dt$ at each time step during the thermal evolution of planetesimals. It takes about 10^3 yr to diffuse ^{18}O entirely in particles with a size of 1 μm at 1000°C, and a much longer time (10^9 yr) is needed at 600°C. These times are long enough compared with the time step for calculating the thermal evolution of planetesimals (dt), which is of the order of one year. We compute the accumulated diffusion length of ^{18}O given by $L^2 = \sum_t dL^2$. We assume that planetesimals do not experience any kind of disruption after their formation.

Figure 1 shows the temperature evolution at the center of planetesimals (dashed lines) and the accumulated diffusion length of ^{18}O (solid lines) for planetesimals with different radii (50 km and 100 km) and formation times ($t_0 = 1.9$ Myr and 2.4 Myr). The results show that the temperature evolution controls how the diffusion length develops over time: as the temperature increases, the diffusion length becomes longer. When the temperature starts to decrease, the increase in diffusion length ceases (see the left panels of Figure 1). Since we evaluate the accumulated diffusion length, it does not decrease once the temperature starts to drop. This means that the maximum temperature governs the diffusion length of ^{18}O in grains.

Our results also indicate that for larger planetesimals (see the right panels of Figure 1), the diffusion length at the center gradually increases even after the temperature reaches its maximum value. This arises because larger planetesimals can keep the maximum temperature for a longer time than smaller ones (see Figures 1(b) and (d)). It should be noted that the maximum temperature, and accordingly the diffusion length, is more sensitive to the formation times of planetesimals than their radii. As seen from Figure 1, the diffusion lengths of planetesimals formed at earlier times ($t_0 = 1.9$ Myr, top panels of Figure 1) are much longer ($\gtrsim 10^{-4}$ m) than those ($\lesssim 10^{-6}$ m) formed at later times ($t_0 = 2.4$ Myr, bottom panels of Figure 1). This is because, if their sizes are the same, the planetesimals formed at earlier epochs have more abundant ^{26}Al and reach a higher maximum temperature. Note that the above results are obtained for the central region of planetesimals. The diffusion lengths are different at different radii of the planetesimals. As mentioned above, the resultant diffusion length is a function of only the maximum temperature. Therefore, we can estimate a diffusion length at every location in a planetesimal by simply referring to the maximum temperature achieved there.

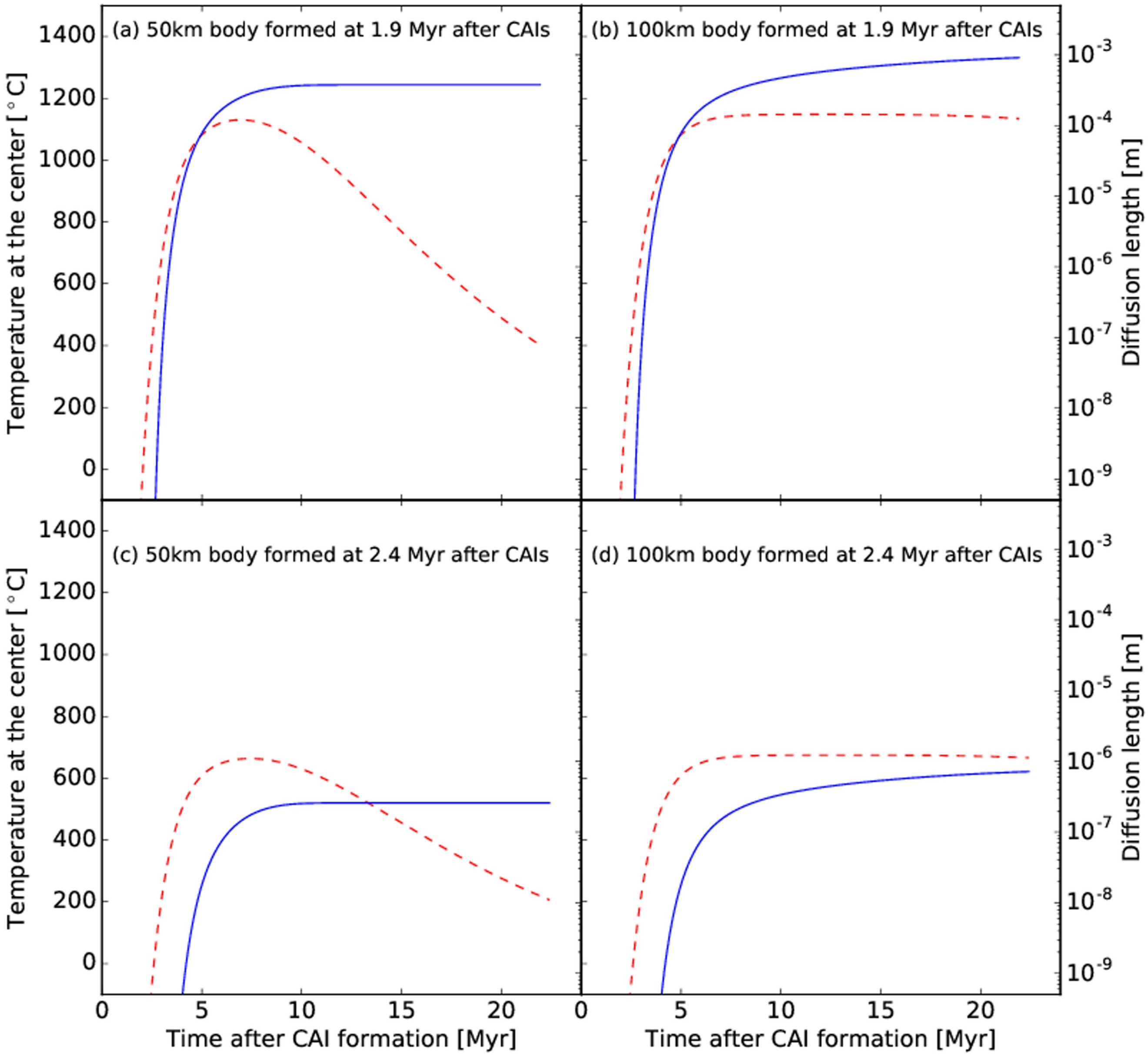

Figure 1. Evolution of temperature at the center of planetesimals (dashed lines with left axes) and the corresponding evolution of diffusion lengths of ^{18}O in olivine (solid lines with right axes) as a function of time after CAI formation. Panels give the result for planetesimals with radii of (a) 50 km and (b) 100 km formed 1.9 Myr after CAI formation, and with radii of (c) 50 km and (d) 100 km formed 2.4 Myr after CAI formation.

We also find that the diffusion length of ^{18}O can be approximately described by the following analytical formula:

$$L^2 = D(T_{\max})\Delta t_{\max}, \quad (3)$$

where $\Delta t_{\max}$ is the duration of the maximum temperature $T_{\max}$. Figure 2 represents the diffusion lengths obtained from numerical simulations and from Equation (3) with $\Delta t_{\max} = 1$ Myr as a function of maximum temperature. It should be emphasized that the dependence of the diffusion length on the maximum temperature follows a simple relation and is reasonably described as a function of $T_{\max}$ by Equation (3). The maximum temperature achieved at each location in a planetesimal decreases with increasing radius. Thus, each curve in Figure 2 can be viewed as representing the radial dependence of diffusion length, where the greatest diffusion length is achieved in the center of a planetesimal. While we adopt $\Delta t_{\max} = 1$ Myr in Equation (3), which is a typical duration for planetesimals of 50 km radius, it works well for planetesimals with a radius of 100 km. Thus, our results demonstrate that our analytical formula (Equation (3)) reproduces the numerical results very well and the maximum temperature can be used as an indicator to estimate the diffusion length of ^{18}O.

3. Discussion and Conclusions

Here we discuss the implications of our results for the size distribution and abundances of presolar grains in various petrologic types of meteorites. In Figure 2, we compare the calculated diffusion lengths with the size ranges of presolar silicate grains obtained from meteorites. For two data points on grain sizes, the vertical error bars cover the maximum and the minimum sizes, and the horizontal ones are the suggested

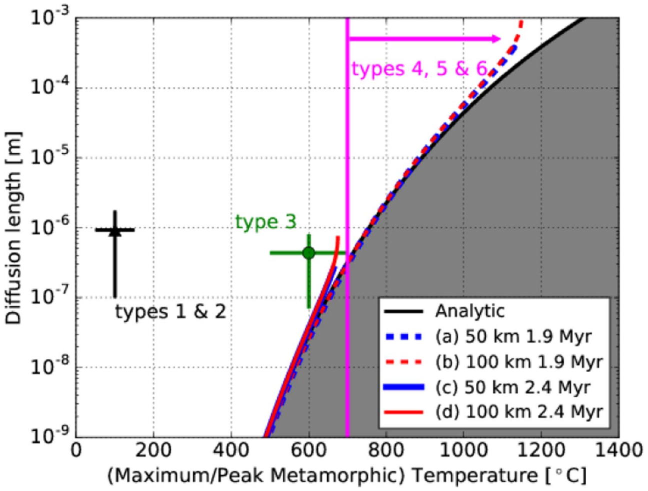

Figure 2. Diffusion length of ^{18}O as a function of maximum temperature, which can be translated to radial depth of the planetesimal. Numerical results are denoted by colored lines, and the analytical result by the black one. We adopt the same values as in Figure 1 for the radii and formation times of planetesimals, and the parameter set for each line is given in the legend of the figure. The shaded region denotes a regime where diffusion can lead to complete erosion of presolar grains of a certain size. Symbols with bars plot the ranges of size and peak temperature derived for presolar silicate grains of chondrites of type 3 (green circle) and types 1 and 2 (black triangle). The vertical magenta line with a rightward arrow indicates the lower value of the peak temperatures expected for chondrites of types 4–6.

ranges of peak metamorphic temperatures. The sizes of presolar silicate grains are 0.1–1.7 μm and 0.07–0.6 μm in petrologic chondrites of types 1 and 2 and type 3, respectively (Zinner 2003; Hynes & Gyngard 2009; Nguyen et al. 2010; Leitner et al. 2012; Hoppe et al. 2015). The metamorphic temperature of type 3 chondrites is likely to be between 500°C and 700°C (Huss et al. 2006; Krot et al. 2007), whereas that of types 1 and 2 carbonaceous chondrites might be around 100°C (Krot et al. 2015).

The maximum temperature governs the diffusion lengths. The length is $\sim$0.001 μm when the maximum temperature is 500°C. If planetesimals reach a maximum temperature of 700°C, the diffusion length is $\sim$0.3 μm. Hence, the diffusion length of ^{18}O in silicate grains in type 3 chondrites ranges from $\sim$0.001 μm to $\sim$0.3 μm, depending on the maximum temperature that they experienced. This indicates that, once we know the minimum size of presolar silicate grains, we can estimate the maximum temperature of their parent bodies. For example, chondrites that contain the smallest presolar silicate grains of 0.07 μm would not have experienced a peak temperature higher than 630°C when they were embedded in their parent bodies. Our calculations imply that, even if typical type 3 chondrites have undergone a peak temperature of 600°C, presolar grains smaller than $\sim$0.03 μm can lose their oxygen isotopic anomaly as a result of isotopic diffusion. This may be one of the reasons why we do not find such small presolar silicate grains in type 3 chondrites.

Petrologic type 3 chondrites can be subdivided into types 3.0 to 3.9 based on their characteristics of thermoluminescence and compositions of minerals (Sears et al. 1980; Scott & Jones 1990; Scott et al. 1994; Grossman & Brearley 2005; Bonal et al. 2006). It is suggested that type 3.0 and 3.1 chondrites experienced lower peak temperatures of $\sim$200–400°C than typical type 3 chondrites (Cody et al. 2008). If this would be the case, our results imply that the original abundance of presolar silicate grains would be retained in such type 3.0 and 3.1 chondrites. In fact, it is interesting to notice that a type 3.0 chondrite, ALHA 77307, is considered to be a primitive chondrite with one of the highest abundances of presolar silicate grains among type 3 chondrites (Nguyen et al. 2007). Although the subtypes of type 3 chondrites cannot be clearly distinguished by their peak metamorphic temperatures (e.g., Huss et al. 2006), it would be valuable to see how the abundance of presolar silicates changes in each subtype.

If the birthplace of presolar silicate grains is the outflowing gas from supernovae and AGB stars, their initial sizes could be dominantly in the range from 0.1 to 1 μm (e.g., Nozawa et al. 2007, 2015; Höfner 2008). As seen in Figure 2, the upper end ($\sim$1 μm) of this range roughly corresponds to the maximum size of grains that could form in supernovae and AGB stars. This implies that the largest presolar grains that might be produced in stellar sources can survive any destructive events (e.g., Hirashita et al. 2016) and they may contain intact information that was recorded at the time of their formation. On the other hand, many submicron-sized grains collide with each other and fragment into nanometer-sized grains as a result of shattering in interstellar turbulence (Hirashita et al. 2010; Asano et al. 2014). Hence, presolar grains should also include grains smaller than 0.1 μm. Nonetheless, the size ($\sim$0.1 μm) of the smallest silicate grains measured in type 3 chondrites is much larger than the minimum size ($\sim$0.001 μm) of interstellar dust. In fact, the critical value of 0.1 μm coincides with the accumulated diffusion length expected from our results. This may suggest that the smallest size of presolar grains in type 3 chondrites might be determined by the diffusion process triggered in thermally evolving planetesimals.

For type 1 and 2 chondrites, which experienced peak metamorphic temperatures of only $\sim$150°C or less, the expected diffusion lengths of ^{18}O are extremely short. Therefore, any size of presolar silicate grains can survive against the isotopic diffusion. However, the sizes of presolar grains measured in these petrologic types are confined to the range 0.1–1.7 μm, and no grains smaller than 0.1 μm have been discovered so far. In general, in type 1 and 2 chondrites, aqueous alteration may have played an important role in eliminating the original isotopic information of grains smaller than 0.1 μm. Leitner et al. (2012) suggest that the initial abundance of presolar silicate grains might be 10 times larger than current values in a type 2 chondrite, and this reduction may be due to destruction of the grains by aqueous alteration.

It should be kept in mind that there is a limitation to the size measurement of presolar grains. Most of the data on the size of presolar grains are obtained from in situ measurements with a secondary ion mass spectrometer (SIMS), whose beam size is comparable to 0.15 μm. This indicates that significant amounts of presolar grains of that size or smaller could be undetected. An optimized setting of NanoSIMS makes it possible to identify smaller ($<$0.1 μm) presolar grains (Hoppe et al. 2015). However, measurements with a spatial resolution of 0.1μm might suffer from instrumental biases in detecting smaller grains (Nguyen et al. 2007, 2010). Thus, more sophisticated techniques are desired to find presolar silicate grains smaller than 0.1 μm in chondrites of types 1–3 and to check our scenario in which isotopic diffusion (and aqueous alteration) can affect the size distribution of presolar grains surviving in planetesimals.

In chondrites of types 4, 5, and 6, which are considered to have experienced a peak metamorphic temperature above 700°C (the vertical magenta line with a rightward arrow in Figure 2), the expected diffusion length is comparable to or even greater than $\sim 0.3\,\mu$m. Therefore, if the original size distribution of presolar grains is limited to below $\sim 0.3\,\mu$m following the grain size distribution in the ISM (Mathis et al. 1977; Nozawa & Fukugita 2013), no presolar grains in these chondrites can keep their original isotopic compositions, because of isotopic diffusion. Note that both isotopic diffusion and thermal metamorphism may be able to delete the original information of presolar grains in these highly thermalized chondrites. Nonetheless, if the process of isotopic diffusion is more efficient in affecting their original isotopic composition than thermal metamorphism, then there is a chance of finding a large presolar grain of $>1\,\mu$m in chondrites of types 4–6. As demonstrated above, their minimum sizes give valuable hints to the maximum temperatures experienced by the parent bodies. Therefore, the search for presolar grains in such fully metamorphosed meteorites is highly encouraged.

Finally, it would be worth discussing the abundances of presolar grains in chondrites and IDPs. The abundances of presolar grains in IDPs are higher than those in chondrites (Messenger et al. 2003; Floss et al. 2006; Busemann et al. 2009). It is very likely that IDPs, which have never experienced any kind of metamorphism, can retain the most primitive information about the chemical composition in the solar nebula. Thus, it may be plausible to consider that the abundance of presolar grains in IDPs represents their original abundance in meteorites. There should be some processes that reduce the abundances of presolar grains in chondrites: aqueous alteration and thermal metamorphism. Although these processes can explain the abundances in chondrites of types 1 and 2 and types 4–6, respectively, they cannot be effective for type 3 chondrites. Hence, isotopic diffusion may be a primary process to cause the difference in abundance of presolar grains between primitive chondrites and IDPs.

As we examine the diffusion of ^{18}O in thermally evolving planetesimals, we can also apply the same approach to the diffusion of other atoms. There are two interesting presolar silicate grains ($\sim 0.2\,\mu$m) that were found in an ungrouped carbonaceous chondrite (type 2 or 3) and have a few nanometers of iron-rich rims (Floss & Stadermann 2012). This discovery suggests that these rims might be an outcome of a kinetic process or a diffusion process. We estimate the diffusion length of iron using Equation (3) with the diffusion coefficient of iron in olivine, which is described in terms of temperature, oxygen fugacity, and iron content (Miyamoto et al. 2002; Dohmen & Chakraborty 2007). At any temperature, the diffusion coefficient of iron would be much larger than that of ^{18}O. Thus, iron can easily diffuse into whole grains while oxygen retains its original content. When we assume an iron content of $\mathrm{Fe}/(\mathrm{Fe}+\mathrm{Mg}) = 0.5$, the diffusion length of iron would be of the order of a nanometer when the maximum temperature is about 200°C. Hence, the diffusion process could explain a thin Fe-rich rim around the presolar grains while they keep the original oxygen isotopic composition that they had when they were in such a lightly heated parent body.

In this paper, we have examined an isotopic diffusion process in thermally evolving planetesimals, which has never been investigated carefully. We find that the diffusion can be viewed as an important process to wash out the original isotopic composition of presolar silicate grains in certain meteorites. We show that isotopic diffusion can regulate the lower size limits of presolar silicate grains in various petrologic types of chondrites, while the upper limits probably originate from the formation processes of the grains in stellar envelopes. For carbonaceous chondrites (usually types 1–3), measurements of grains $<0.1\,\mu$m are needed to firmly address how important the diffusion is in altering the size distribution of presolar grains in planetesimals. The methodology developed in this paper is applicable to any other isotopes in a variety of minerals provided that their diffusion coefficients are given. The measured sizes of presolar grains, combined with the simulations of diffusion lengths of the relevant isotopes, will surely advance our understanding of presolar grains and the parent bodies of their host meteorites.

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