Lifetimes of interstellar dust from cosmic ray exposure ages of presolar silicon carbide

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We determined interstellar cosmic ray exposure ages of 40 large presolar silicon carbide grains extracted from the Murchison CM2 meteorite. Our ages, based on cosmogenic Ne-21, range from 3.9 ± 1.6 Ma to ~3 ± 2 Ga before the start of the Solar System. ~4.6 Ga ago. A majority of the grains have interstellar lifetimes of <300 Ma, which is shorter than theoretical estimates for large grains. These grains condensed in outflows of asymptotic giant branch stars <4.9 Ga ago that possibly formed during an episode of enhanced star formation ~7 Ga ago. A minority of the grains have ages >1 Ga. Longer lifetimes are expected for large grains. We determined that at least 12 of the analyzed grains were parts of aggregates in the interstellar medium: The large difference in nuclear recoil loss of cosmic ray spallation products 3He and 21Ne enabled us to estimate that the irradiated objects in the interstellar medium were up to 30 times larger than the analyzed grains. Furthermore, we estimate that the majority of the grains acquired the bulk of their cosmogenic nuclides in the interstellar medium and not by exposure to an enhanced particle flux of the early active sun.

Interstellar dust is an important component of our galaxy. It influences star formation as well as the thermal and chemical evolution of the galaxy. Although dust only presents ~1% of the mass in the interstellar medium (ISM) (1), it carries a large fraction of the elements heavier than He (2), including the elements that form terrestrial planets and are essential for life. Thus, interstellar dust is a key ingredient of stars and habitable planetary systems, making increased knowledge about its composition and lifecycle desirable. Compositional, structural, and size information of interstellar dust can be obtained through astronomical spectroscopic observations (3), but dust lifetime estimates mainly rely on sophisticated theoretical models. These models, however, focus on the more common small dust grains and are based on assumptions with large uncertainties. These uncertainties primarily pertain to the residence time of the dust in various regions of the ISM, which exhibit different rates of dust destruction through sputtering and collisions in supernova shock waves (4–9). Most of these models currently predict an average lifetime of interstellar grains on the order of 100 Ma. However, more recent models and a few models for larger grains predict much longer survival times in the ISM of up to billions of years (10–12).

Here, we present a laboratory-based approach of determining the interstellar lifetimes of individual large presolar silicon carbide (SiC) stardust grains (Fig. 1). The presolar grains analyzed in the present study were isolated by chemical methods (see Materials and Methods) from the Murchison CM2 meteorite, where they had remained unaltered since their incorporation into the meteorite parent body in the early Solar System 4.6 Ga ago. These grains are identified as presolar by their large isotopic anomalies that exclude an origin in the Solar System (13, 14). Presolar stardust grains are the oldest known solid samples available for study in the laboratory, represent the small fraction of material that formed in circumstellar environments, and survived processing in the ISM and Solar System. The presolar stardust grain abundance in our parent interstellar cloud was a few percent of all interstellar dust present in this cloud (15), with the other dust having condensed in the ISM. In the solar nebula, more dust condensed from the cooling gas and presolar stardust became an even more minor component. Most presolar grains were subsequently destroyed after accretion in their parent bodies during thermal metamorphism and aqueous alteration. Thus, their abundance in the most primitive Solar System materials that evaded destructive parent body processing is a few parts per million (ppm) to ~200 ppm (16) except for interplanetary dust presumably from comet Grigg-Skjellerup dust, which contains up to ~1% presolar materials (17). We used mass spectrometry to analyze the abundance of nuclides produced in the grains by spallation reactions with galactic cosmic rays (GCR)—which comprise mostly high-energy protons and α-particles—during their residence in the ISM. When these high-energy particles hit a grain, small fractions of the target nuclides break up.

Significance

Dating of interstellar dust directly with astronomical methods is not possible. Neither is dating based on the decay of long-lived radioactive nuclides, due to current analytical limitations and unknown initial isotopic compositions. Here we present interstellar ages of individual presolar SiC grains from a meteorite. The ages are based on Ne isotopes produced by galactic cosmic rays. Lifetimes of ~60% of our grains are <3 × 10⁶ y, while at least 8% are >10⁹ y, in line with what is expected for large grains. The former could be the end products of stars originating in an enhanced star formation episode. Presolar grains are the oldest datable solid samples available and provide invaluable insight into the presolar chronology of our galaxy.

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The resulting atomic fragments accumulate in the grain, and their concentrations are proportional to the timespan the grains were irradiated. Suitable daughter elements to study are those with a very low initial abundance in the grains such that the cosmic ray-produced (“cosmogenic”) fraction becomes detectable. This is the case for He, Ne, and Li. SiC is the best-suited interstellar phase for cosmogenic nuclide dating, due to its relatively large grain size, high retentivity of cosmogenic nuclides, and durability. Even though SiC is only a small fraction of the total amount of interstellar dust (9), due to its durability, we consider it a useful tracer.

In the most common SiC grains, the ones that originate from low- to intermediate-mass asymptotic giant branch (AGB) stars (14), the initial He and Ne isotopic compositions, incorporated from their parent stars, are well known (18-20), so the cosmogenic fraction can be readily identified. Improved knowledge of production and retention of such cosmogenic nuclides enabled us to obtain ages with improved reliability. While radiometric dating based on the U–Pb decay system can provide ages with high accuracy (21) and is often the method of choice for samples of Solar System materials, it has not yet been successfully applied to presolar grains. These grains have masses that are orders of magnitudes smaller than samples dated so far. Furthermore, presolar grains have large isotopic anomalies in essentially every element, so each grain may have a distinct initial lead isotopic composition, uranium isotope ratio, and age, robbing the U–Pb system of some of its most desirable characteristics for geochronology. Until these obstacles are overcome, exposure age dating is the preferred method for determining presolar ages of individual Stardust grains.

The first such studies were made on assemblages of thousands of SiC grains from chemical separates. Ages of $\sim 10^7$ to $10^8$ y were derived from $^{21}$Ne, but it was suggested that individual grains might show much higher presolar ages of up to 2 Ga (18, 22). However, Ott and Begemann (23) showed that much of the measured $^{21}$Ne was not cosmogenic but was implanted neon from the He shell of the parent AGB star, while, at the same time, they concluded that losses of cosmogenic $^{21}$Ne upon production due to recoil out of the grains were much larger than assumed. Ott et al. (24) deduced much lower presolar ages for bulk SiC assemblages of a few times $10^7$ y only, based on cosmogenic Xe, for which recoil losses are smaller. The first interstellar exposure ages on individual exceptionally large ($\sim 5$ to 60 $\mu$m) SiC grains were reported by Gygard et al. (25) based on Li isotopes and by Heck et al. (20) with He and Ne. The large grains contain greater amounts of cosmogenic nuclides, and, more importantly, require a smaller recoil correction (26). The studies by Heck et al. (20) and Gygard et al. (25) both reported ages of between a few megayears to about 1 Ga, but the average of the Li-based ages was considerably higher than the average noble gas age. Heck et al. (20) suggested that the many ages of $<200$ Ma may be explained by increased dust production after a galactic starburst 1 to 2 Ga prior to the birth of the sun.

In this work, we provide presolar ages based on cosmogenic Ne isotopes, significantly increasing the total number of presolar grain ages. We also present reevaluated ages from previously published data. This will enable us to further advance our understanding of the lifetimes of interstellar dust. Previous interstellar production rates of cosmogenic nuclides were based on fluxes deep within the heliosphere that were extrapolated to interstellar space (20, 27, 28). Here, we use, instead, improved interstellar production rates that were determined with the purely physical model of Trapptisch and Leya (29) that uses a state-of-the-art nuclear cross-section database and an interstellar GCR spectrum based on data collected by NASA’s Voyager 1 space probe at the edge of the heliosphere. Voyager 1 recorded the low-energy part of the GCR spectrum, something that is not possible deeper within the heliosphere. To correct for recoil losses of cosmogenic nuclides from SiC grains, we use a physical recoil model that considers the energies of GCR protons and $\alpha$ particles from the new cosmic ray spectrum (29).

Another aspect that was not considered in previous studies is the potential exposure of presolar grains to the enhanced particle flux of the early active sun. Large excesses of cosmogenic noble gases in single olivine grains in some primitive meteorites have been attributed by some workers to a high flux of energetic particles from the early sun (e.g., refs. 30–32), although others contested this conclusion (33). Recently, however, unambiguous evidence for an enhanced exposure of hibonite (an aluminum–calcium oxide)—possibly the earliest solar nebula condensate—to energetic particles from the early active sun was reported by Kööp et al. (34). This implies that some of the presolar grains we studied might have been exposed to the same enhanced solar particle flux. We are, therefore, also required to estimate the upper limit of cosmogenic nuclides concentrations produced in the early Solar System rather than in the ISM.

Results and Discussion

Presolar Grain Ages. We processed our noble gas data from 27 SiC grains and reprocessed data from published results from 22 SiC grains (20) to calculate an internally consistent set of presolar cosmic ray exposure ages for nearly 50 grains with the improved cosmogenic nuclide production rates and nuclear recoil corrections (SI Appendix, Table S1). The cosmogenic Ne component can be clearly resolved from the two other main components, nucleosynthetic Ne (Ne–G) and adsorbed atmospheric Ne based on distinct isotopic Ne compositions (SI Appendix, Fig. S6A). Ne–G is implanted into circumstellar grains from the hot post-AGB star wind emanating from the exposed He shell, and its concentrations decrease with increasing grain size (SI Appendix, Fig. S6B and refs. 19 and 20). Using C, N, and Si isotopes, all but three grains have been classified as mainstream SiC, originating in the outflows of low- to intermediate-mass (post) AGB stars (14, 35) (SI Appendix, Fig. S1 and Table S2). The three other grains are of AB type, based on their low $^{12}$C/$^{13}$C ratios (SI Appendix). All newly analyzed grains are mainstream SiC.

We determined $^3$He and $^{21}$Ne exposures ages ($T_3$ and $T_21$) of 30 and 24 grains, respectively, and obtained upper age limits for

![Fig. 1. Presolar SiC morphology. Scanning electron microscope images (secondary electrons) of representative samples of the two morphological types of presolar SiC grains studied here.](Image)
12 (3He) and 16 (21Ne) grains (SI Appendix, Table S3). For 18 grains, we have obtained both T3 and T21. Nominal recoil-corrected T3 for 16 out of these 18 grains are higher than recoil-corrected T21, whereas uncorrected ages show an opposite trend (Fig. 2). Helium is more easily lost through heating and through recoil than Ne, so both effects would result in lower nominal T3 than T21 before a recoil correction. Hence, a recoil correction will be larger for 3He than for 21Ne. SI Appendix, Fig. S2 shows that, for grains of <10 μm, nominal cosmogenic 3He recoil losses are >94% for the smallest grains analyzed here, whereas corresponding losses for 21Ne are >40%. Hence, any uncertainties in recoil corrections will result in a larger uncertainty of T3. Heating of grains to high temperatures (>900 K) would result in near-complete He loss (36). Helium loss works in the opposite direction of the trend we see in the data. This implies that, while some He loss cannot be excluded, no significant loss occurred; otherwise, much more He than Ne would have been lost, and even overcorrected T3 would be smaller than T21. The T3 are less reliable than T21, mainly because of larger uncertainties in the 3He recoil correction. The 16 recoil-corrected T3 exceeding recoil-corrected T21, consequently, indicate an overestimation of the recoil loss for 3He. The reason for this may be that these grains were actually irradiated in the ISM as parts of larger grains or as grain aggregates, or the grains were coated with large mantles of ices and organics while in the ISM. We estimate the original sizes of the irradiated objects in the ISM by varying the grain size and modeling the resulting recoil correction until the recoil-corrected T3 and T21 match (SI Appendix, Fig. S3). The estimated object diameters during irradiation are factors of ∼3× up to ∼30× higher than those of the analyzed grains. This results in ages of 45 to 85% of the original recoil-corrected ages (Fig. 3). In principle, it would be possible to test this result with cosmogenic Xe that has a much smaller recoil loss.

Unfortunately, the amounts of cosmogenic Xe produced are below current detection limits for single-grain analyses, due to the low amounts of suitable target elements for Xe production in SiC (23). Bulk analyses of SiC give mixed signals and are not useful in this regard, as these do not resolve cosmogenic gas contributions from grains with different lifetimes. Seventy-five percent of the 16 analyzed grains that were part of much larger objects have euhedral shapes, which indicates they are not fragments of larger grains and were more likely parts of aggregates. The remainder look like they are shattered fragments of larger SiC grains (SI Appendix, Table S3 and Dataset S1), but, given the large object sizes estimated during ISM irradiation, larger than any known presolar SiC grain, they were likely also part of aggregates. Aggregates of minerals, suspected by some to be presolar, in an organic material matrix were recently observed in interplanetary dust particles (37). Bernatowicz et al. (38) observed organic coatings on ~60% of pristine presolar SiC that they physically separated from their host meteorite without the use of chemical reagents. However, no aggregates or clustering of larger presolar grains have yet been observed during the in situ imaging searches of polished sections of meteorites (e.g., ref. 16). The lack of such clustering of larger grains could be due to preferred breakup of larger clusters of several dozen to hundred micrometers during accretion onto planetesimals in the early Solar System, while smaller clusters composed of smaller grains which have lower inertia, such as the ones observed by Ishii et al. (37), stayed intact. We propose that grains in the size range we analyzed formed in the outflows of (post) AGB parent stars (39) and coagulated there with organic matter to form larger aggregates. While large SiC dust grains are rare in the ISM, they are consistent with observations of circumstellar dust around AGB and post-AGB stars (40). Far-infrared excess associated with such dust may indicate the presence of up to millimeter-sized grains (41). Up to 5-mm-large dust grains were proposed to explain radio observations of dust around the Egg Nebula, a post-AGB star (42). Jura et al. also propose that the
high-density winds from post-AGB stars are the sources of the large presolar SiC grains, such as the size fraction studied here.

We also obtained Li isotope data for 19 SiC grains. Many of these grains have a Li⁷/Li⁶ ratio below the chondritic (“solar”) value of 12.06 ± 0.03 (43), indicating the contribution of a cosmogenic Li component [the end-member cosmogenic Li⁷/Li⁶ ratio is ~1.2 (29)]. However, the nominal Li ages determined from different spots on the same grains are highly variable. Li ages also correlate with the total, noncosmogenic Li concentration (SI Appendix, Fig. S4 and Table S4). These observations could be due to a combination of contamination with terrestrial or Solar System Li, matrix effects (25), or additional, unidentified Li components that would have contributed to the measured Li concentration. Because of low concentrations of cosmogenic Li and high abundance of normal Li, a reliable determination of cosmogenic Li is very difficult. Currently, Li does not allow us to obtain reliable ages, as discussed in more detail in SI Appendix.

Evidently, the Ne ages are more reliable than the Li and He ages, and we will base the following discussion mainly on ²¹Ne ages. They range from 4 ± 2 Ma (±1σ) to 3,200 ± 2,300 Ma (Figs. 2 and 3), and upper limits range from 3 to 3,300 Ma. We obtained ²¹Ne ages for two out of three AB grains; the calculated ages, 65 ± 9 Ma and 260 ± 59 Ma, fit into the age range of the mainstream grains. No age was determined for the third AB grain, due to an insufficient gas amount; the 2-μm-sized grain was the smallest one analyzed in this study.

Overall, the ²¹Ne age distribution trend (Fig. 3) is similar to what was previously reported for a smaller sample set (20), with most exposure ages below 300 Ma (60%) and 50% below 200 Ma. This is consistent with most theoretical lifetime estimates for much smaller, <1 μm interstellar dust of 100 to 300 Ma (4–9), but in contrast to the longer lifetimes expected for large grains (10–12). Assuming constant dust production rates from AGB stars and constant dust destruction rates, we would expect to encounter younger grains more frequently than older grains simply because older ones have a higher probability of encountering a destructive process. However, our age distribution does not fit any of the assumed steady-state models for different average lifetimes (SI Appendix, Fig. S5A).

Having many large grains in a relatively narrow age range seems to require an explanation other than simply a lifetime effect, which would apply to small grains. We propose that this age distribution is caused by high-speed winds from post-SNR gas and experience rare collisions with other large grains (10). Gradual erosion by collisions with smaller grains would leave cratered surfaces (10), something that has not been observed with SiC grains to date (38). Possible evidence of a microimpact crater was so far only found in a large presolar aluminum oxide grain (51). Some of the old grains could have been shielded from destructive processes in clumps. Such protective density inhomogeneities have been observed astronomically in shocked regions of the ISM (e.g., ref. 52).

The oldest grains based on both He and ²¹Ne ages are the smallest, and an inverse trend between age and grain size is apparent (Fig. 4 and SI Appendix, Fig. S6), consistent with the preliminary trend observed by Ott and Begemann (23) in Xe from bulk SiC analyses but in contrast to the prediction by Hirashita et al. (12). The trend persists in the recoil-corrected data and in the size-corrected subset but gets less prominent in the latter. Smaller grains are more abundant than larger grains in the ISM (3), resulting in a higher number of smaller grains that are old compared to larger ones. We can exclude a sampling bias, as we have not disproportionally analyzed small grains; on the contrary, only 12 of the 49 grains are <4 μm.

Gyngard et al. (53) proposed that grains with presolar ages older than the sun’s galactic year (230 Ma (54)) might have had the time to radially migrate from the inner parts of the galaxy toward the galactocentric distance of the forming Solar System. Because of the compositional gradient within our galaxy, we would expect these grains to reflect the metallicity of their parent stars. However, we do not observe a correlation between age and Si isotopic composition, which is a proxy for metallicity of stellar sources (55). Either our dataset is too small to reveal such a trend, the grains did not migrate as suggested, or there is no galactic gradient for Si isotopic composition, in contrast to O isotopic composition (56) and [Fe/H] (57). Recent astronomical observations (58) did not find a galactocentric ⁶⁷⁶³Si trend within ~200%, a range that was less than expected from galactocentric variations in other isotope ratios but similar to the one measured in presolar SiC mainstream grains.

We should highlight that, at the end of their interstellar journey, the presolar grains could have been exposed to enhanced particle...
hibonites are indeed early Solar System products and not of presolar origin.

We note that a presolar exposure age of a SiC grain is a nominal age and that the actual residence time in the ISM might have been shorter if the grains were exposed to a high energetic particle flux from other nearby stars in addition to background GCR exposure. We estimate that the chances of such a close encounter for the average interstellar SiC are low and that such exposure could have also led to destruction of the grain. Modeling of this probability is difficult due to many unknowns and beyond the scope of this work.

Conclusions

With this study, we have increased the number of presolar SiC Ne exposure ages, calculated with improved recoil corrections and cosmogenic nuclide production rates. Based on Ne isotopes, we conclude that a majority (~60%) of the large presolar SiC grains analyzed have interstellar cosmic ray exposure ages below 300 Ma before the formation of the Solar System. This is compatible with most theoretical estimates of interstellar dust lifetimes of 100 to 200 Ma. This age distribution is also consistent with the hypothesis that these grains originate from stars that initially formed during an enhanced SFR ~7 Ga ago and became dust-producing AGB stars between ~4.9 and ~4.6 Ga ago. Furthermore, a significant fraction has presolar ages above 300 Ma, with at least ~8% above 1 Ga, making them the oldest dated samples so far. These old ages require that these grains evaded destruction in supernova shockwaves, possibly in dense clumps that formed in such shockwaves. Based on a comparison of cosmogenic He and Ne, it is clear that some grains were part of larger particles or aggregates and might have had large mantles of ices and organics during cosmic ray exposure in the ISM.

The studied presolar grains might have acquired a small but, in most cases, undetectable fraction of their cosmogenic Ne during exposure of energetic particles from the early active sun. However, only particularly young grains with very low interstellar residence times might have received a significant fraction of their cosmogenic nuclides in the early Solar System, before accretion onto planetesimals. The specifics of this exposure, such as the solar particle flux and exposure, are currently unknown.

We conclude that Ne exposure age dating is currently the only viable method to date presolar grains. While the method provides ages relative to the start of the Solar System and suffers from relatively large uncertainties, it can provide unique information about the interstellar dust cycle and star-forming events in the Galaxy before the birth of the sun.

Materials and Methods

This paragraph describes the materials and methods in brief. More detailed information, data and figures of the samples, analytical methods, and models are provided in SI Appendix. Large presolar SiC from the original so-called “LS+LU” separation from the Murchison meteorite were characterized with electron microscopy and classified with nanoscale secondary ion mass spectrometry (NanoSIMS). Isotopes of Li were analyzed with NanoSIMS, and He and Ne isotopes were analyzed with noble gas mass spectrometry. We determined cosmogenic components and recoil corrections before calculating cosmic ray exposure ages with interstellar production rates. The systematic uncertainties of the ages include uncertainties in the production rates and recoil corrections. The uncertainties of the data are based on counting statistics, blank corrections, and sample mass errors. A detailed discussion of the uncertainties is given in SI Appendix. Ages are considered upper limits if their uncertainty is larger than the age.

Data Availability Statement. All data discussed in the paper is available in the SI Appendix.

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