

Nanodiamonds in meteorites: properties and astrophysical context

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Properties

ABSTRACT

Purpose: This contribution provides an overview on properties and origin of nanodiamonds in primitive meteorites. Nanodiamond are a type of stardust, i.e. “pre-solar” grains that formed in the outflows or ejecta of stars.

Design/methodology/approach: We summarize previously obtained results and include our results dealing with recoil loss from nanoparticles during radioactive decay of trace elements within them.

Findings: Nanodiamonds in primitive meteorites have a mean size of ~2.6 nm and an abundance reaching up to ~0.15 % by weight. They contain trace noble gases, notably xenon, with an unusual isotopic composition. The latter is reminiscent of the p- and r-processes of nucleosynthesis that are thought to occur during supernova explosions. Our new results show that recoil loss during β decay of implanted ^{22}Na does not exceed what is expected from energy distribution and range-energy relations in matter. While a CVD origin for the diamonds appears likely (but is not assured), the noble gases were probably introduced by ion implantation.

Research limitations/implications: The isotopic pattern of Xe contained in nanodiamonds indicates some unconventional types of element synthesis in stars or modification by secondary processes. Recoil loss from nanometer-sized grains during decay of unstable precursor nuclides has been suggested as an explanation, but our experiments do not support this idea.

Originality/value: Other processes must be invoked for explanation of the isotopically unusual xenon trapped in meteoritic nanodiamonds. Ion implantation experiments suggest of “trapped” cosmic ray ^3He for deriving an age for the diamonds.

Keywords: Presolar grains; Nanodiamond; Noble gases; Nucleosynthesis; Nuclear recoil

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1. Discovery of stardust diamonds

One of the important and more spectacular developments in the study of meteorites has been the realization about two decades ago of the presence in the more primitive ones of presolar materials ([1, 2; more recent 3]). These grains of “stardust” predate the formation of the Solar System; they formed from

material in the strong stellar winds of evolved stars or from material ejected during the explosion of supernovae. Suspected to be present based on variations in the isotopic composition of noble gases [4, 5], the first ones were detected in 1987 [6, 7] by following the lead of the noble gases. Luckily, it turned out that the important presolar noble gas carriers – diamond, silicon carbide and the later found graphite [8] – were resistant to acids and could be concentrated in rather pure form by chemical / physical separation methods as described in, e.g., [9]. This allows

detailed analysis of a large number of grains without interference from adjacent minerals. Newly developed NanoSIMS instrumentation (e.g., [10]) has allowed in the meantime the detection *in situ* of presolar minerals and also led to the discovery of other types, in particular silicates that do not survive the chemical treatments. Table 1 gives a summary of the current status of pre-solar grains identified in meteorites. Here I present an overview of properties of meteoritic nanodiamonds (with up to ~1500 ppm the nominally most abundant presolar grain type) and describe isotopic characteristics of noble gases and other properties relating to their stellar origin and history.

Table 1.

Major types of identified presolar grains in meteorites and some of their properties

mineral type	abundance [wt ppm] ⁺	size [μm]	isotopic signatures*	stellar source(s) [#]
diamond	~ 1500	~0.0026	noble gases	supernovae(?)
SiC	~ 30	~0.1-10	C, N, Ne, (Mg) s-process elements	AGB stars (supernovae)
graphite	~ 10	~0.5-10	C, N, Ne, Si, (Mg, K, Ca)	supernovae, AGB stars
refractory oxides	~ 50	~0.1-5	O, (Mg)	RGB/AGB stars (supernovae)
silicates	~ 200	~0.1-1	O, Si	RGB/AGB stars (supernovae)
Si ₃ N ₄	~ 0.002	~1	C, N, Si, (Mg)	supernovae

⁺Abundances vary with meteorite type. Listed values are maximum reported abundances.

*Elements with characteristic isotopic signatures. Elements in parentheses show enhanced abundances of isotopes from decay of extinct radionuclides: ²⁶Mg from ²⁶Al (half-life 7×10⁵ a), ⁴¹K from ⁴¹Ca (1×10⁵ a) and ⁴⁴Ca from ⁴⁴Ti (60 a). S-process elements are elements heavier than Fe (e.g., Sr, Zr, Mo, Ba, Nd) with significant contributions from the slow neutron capture process.

[#]Listed are the dominant stellar sources (minor sources in parentheses). RGB/AGB stars = Red Giants on the Red Giant Branch and Asymptotic Giant branch, respectively, in the Hertzsprung-Russell diagram.

2. Physical properties of meteoritic nanodiamonds

A major distinction between the meteoritic nanodiamonds and the other presolar materials found in meteorites so far is their size. The size of presolar silicon carbide and silicates, for example, is typically in the range of tenths of and reaches up to several micrometers, which allows them to be analyzed individually for major and more abundant trace elements, using the NanoSIMS (e.g., [11, 12]). In contrast, the average size of the diamonds is on the order of 2-3 nm (Fig. 1), as established by extensive high-resolution TEM work [13] and confirmed by the technique of MALDI-TOF-SIMS [14, 15]. The size distribution is log-normal, consistent with expectations for growth processes and indicating

minimal secondary processing. An unpleasant side effect of the small size (on average some 1000 carbon atoms) is that it is not possible to perform chemical / isotopic studies of single diamond crystals. Notably, the “average” carbon isotopic composition of the diamonds is not indicative of a stardust origin, so only for those that carry isotopically unusual noble gases such as xenon – which led to their identification in the first place – a pre-solar origin can be considered as ascertained. This fraction is exceedingly small, since there is roughly only one Xe atom per a million diamond grains, but it is a lower limit only, of course.

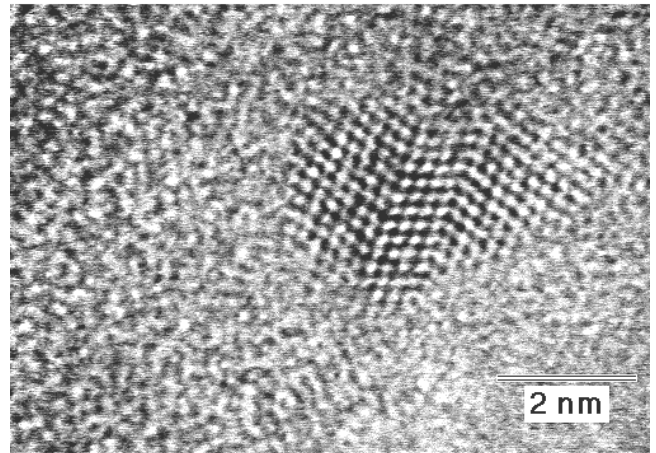


Fig. 1. High-resolution TEM image of a pre-solar nanodiamond grain from the Murchison meteorite. Courtesy F. Banhart

Diamond is a typical high-pressure phase and one of the processes suggested for its origin is shock transformation from graphitic material. As described in [16], the process is physically possible by the action of supernova shock waves traveling the interstellar medium, but some quantitative problems appear in the context. Other possibilities are Chemical Vapor Deposition (CVD), or maybe even homogeneous nucleation from the gas phase [17]. Raman spectroscopy has been tried as a tool to possibly distinguish between high- and low-pressure origin [18, 19], but the results are inconclusive. The observed peak broadening and downward shift by 6 cm⁻¹ of the characteristic 1332 cm⁻¹ band may be due to shock [20-22], but also may be related to small grain size or phonon / quantum confinement effects [23-25]. More telling may be the results from the work of [13] who performed an extensive TEM study and comparison with analogous observations on CVD and detonation diamonds. According to their results, the ratio of twins/single crystals (1.25), the ratio of linear to non-linear multiple-twins (0.87) and the absence of dislocations are significantly more in line with a CVD origin than a shock origin. The match is, however, not perfect. As an example, the lonsdaleite (2-H) polymorph that is usually associated with shock origin is present in the detonation diamonds (as expected) and has not clearly been observed in the CVD sample (not surprising). But it is present also in the meteoritic nanodiamonds investigated by [13]. Thus, the possibility needs to be entertained that there are various sources of the nanodiamonds [13], similarly as inferred from the carbon isotopes as discussed above.

3. The stellar / interstellar connection

3.1. Xenon isotopes

The most telling isotopic feature observed in the analysis of “bulk samples” of meteoritic nanodiamonds is in the noble gas xenon. As noted above, the major element, carbon, has an unspectacular isotopic composition, consistent with an origin of the majority of diamonds in a solar system setting. Similarly, the major trace element nitrogen has a $^{14}\text{N}/^{15}\text{N}$ isotopic ratio of ~400 [26], which is distinct from the terrestrial value (272 in air), but is identical within uncertainties with nitrogen measured in the atmosphere of Jupiter [27] and possibly also in the solar wind, as inferred from measurement on one set of samples returned by the Genesis spacecraft [28].

The Xe isotopic composition is characterized by overabundances of the heaviest (neutron-rich) and the lightest (proton-rich) isotopes relative to those of intermediate mass [6, 29] – hence named HL-xenon, as shown in Fig. 2. The nuclear origin of these isotopes are the rapid neutron capture process (r-process) and the so-called p-process (more likely (γ, n) reaction rather than proton capture), both of which are considered to take place in massive stars ending their life as supernovae type II.

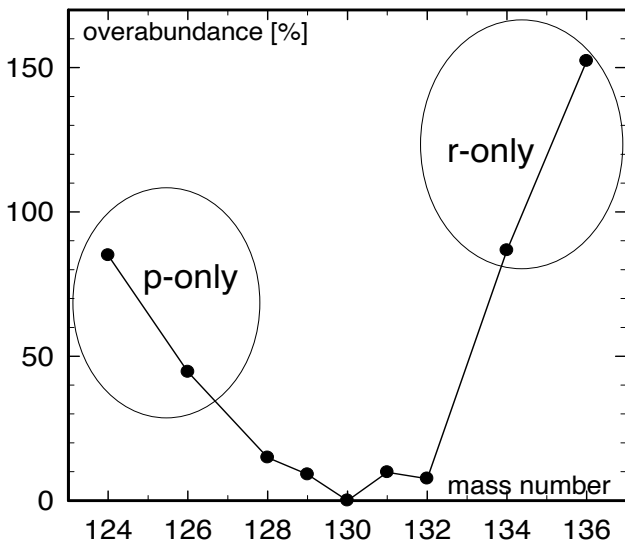


Fig. 2. Isotopic pattern of HL-Xenon released from meteoritic nanodiamonds at high temperature. Shown are deviations in % of ^{130}Xe -normalized ratios from those in the solar wind xenon

In both cases one hardly expects any production of the middle isotope ^{130}Xe , and a composition for the “pure HL-Xenon” can be obtained by extrapolating mixing relations to $^{130}\text{Xe} \equiv 0$. The composition of the thus derived heavy part (“H-Xenon”) is reminiscent of the r-process, but in detail quite distinct from that of r-process xenon derived from solar system abundances (Fig. 3; [30]).

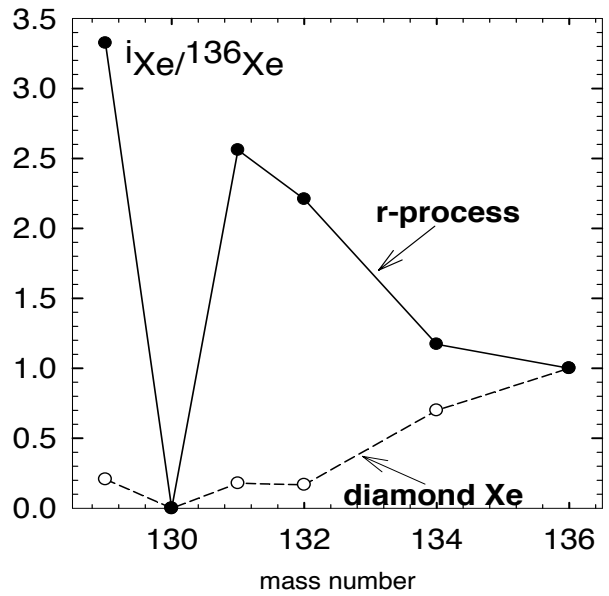


Fig. 3. Isotopic pattern of Xe-H compared with r-process xenon

3.2. Rapid separation of r-process products (?)

Comparison of predictions of neutron burst [32] and “r-process plus early separation” [30] models with composition of Xe-H as derived from analysis of presolar nanodiamonds is shown in Fig. 4.

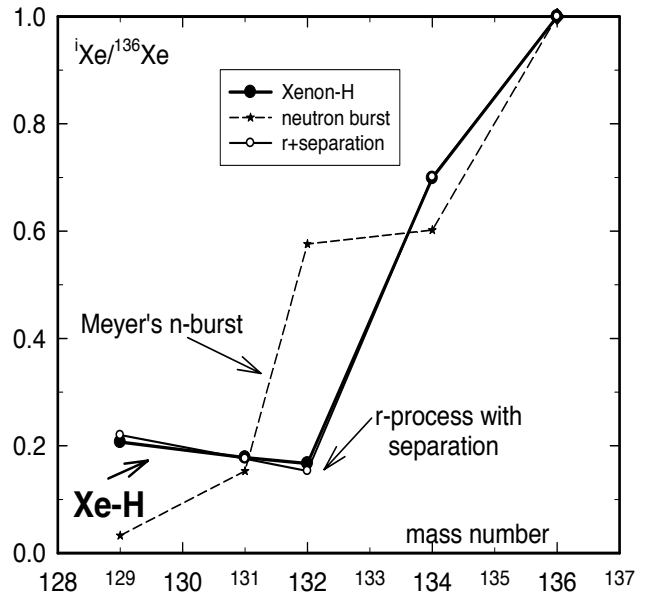


Fig. 4. Comparison of predictions of neutron burst [32] and “r-process plus early separation” [30] models with composition of Xe-H as derived from analysis of presolar nanodiamonds

3.3. Nuclear recoil in β -decay

It is not straightforward, however, to identify a process leading to the “early separation” that realistically can operate under the conditions characteristic of the immediate neighborhood of a supernova some 2 hours after the explosion. Three possible scenarios have been considered in the original work of [30]: separation according to volatility, recoil loss from nanometer-sized grains of β -decay daughter nuclei and separation according to charge/mass ratio in the strong magnetic field of the nascent neutron star (the final state of the exploding supernova). Of these, the volatility-based separation has been immediately ruled out by [30], while the importance of charge/mass separation is difficult to ascertain. In contrast, the nuclear recoil effect is amenable to experimental test. Compared to α -decays where a much more massive particle is ejected and hence significantly larger recoil energy is transferred to the residual nucleus, recoil in β -decays is usually considered negligible. But then, usually one is not dealing with decay happening in nanometer-sized grains. A test that is also relevant in context of the unexpected absence from the diamonds of ^{26}Al and ^{44}Ti (half lives of 0.7 million years and 60 years, respectively) that now would show up as relative overabundances of the decay products ^{26}Mg and ^{44}Ca [35] was performed by [36, 37]. In this work we implanted radioactive ^{22}Na (half life 2.6 years) into nanodiamonds and determined the abundance of the decay product ^{22}Ne . The results indicate that nuclear recoil would not have resulted in an early separation. A new idea has recently been put forward by [38] who suggest early condensation of less volatile Xe precursors in grains that are later reheated and destroyed, but with the spatial separation of Xe from precursors remaining.

3.4. Noble gas implantation and release

Implantation of ions has long been considered the most promising mechanism for introducing the noble gases into the nanodiamonds [39] and this has been confirmed in experiments using terrestrial detonation nanodiamonds as analogs [40, 41, 34]. Interestingly, the noble gas abundance as a function of detailed nanodiamond grain size can be used as a measure for the energy of the implanted noble gas ions [41]. A further clue to the history and properties of the meteoritic nanodiamonds is the bimodal temperature release pattern, with isotopically unremarkable Xe (Xe-P3) released in the first and Xenon-HL in the second peak (Fig. 5). Most likely the noble gases are trapped in diamonds by defects of two types with different binding energies [42], and two populations of gas-bearing nanodiamonds with different histories may exist [40]. A third noble gas component (P6) is released at even higher temperature, but its contribution to the noble gas budget is minor [29]. Also, since it is only seen mixed with Xe-HL, it is not clear whether P6 is isotopically unusual (and hence presolar) or – like the low-temperature P3 component – possibly of “local” origin.

To study structural changes related to gas release, we have also performed a series of experiments in which nanodiamonds were heated to various temperatures (600 °C, 800°C and 1000°C where most of the P3 component has been released – Fig. 6), and their appearance and structure was checked by TEM [37, 43]. No clear-cut correlation was found, with some onion-like structures observed after heating to 600 °C already, and some diamonds surviving heating to 1000 °C. More work along these lines will be needed.

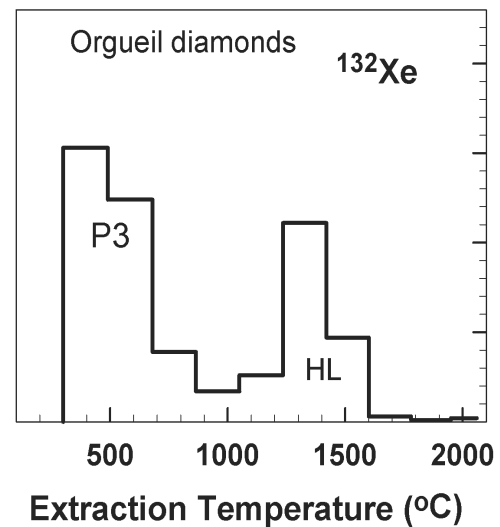


Fig. 5. Thermal release of Xe from nanodiamonds of the Orgueil meteorite (data from [29])

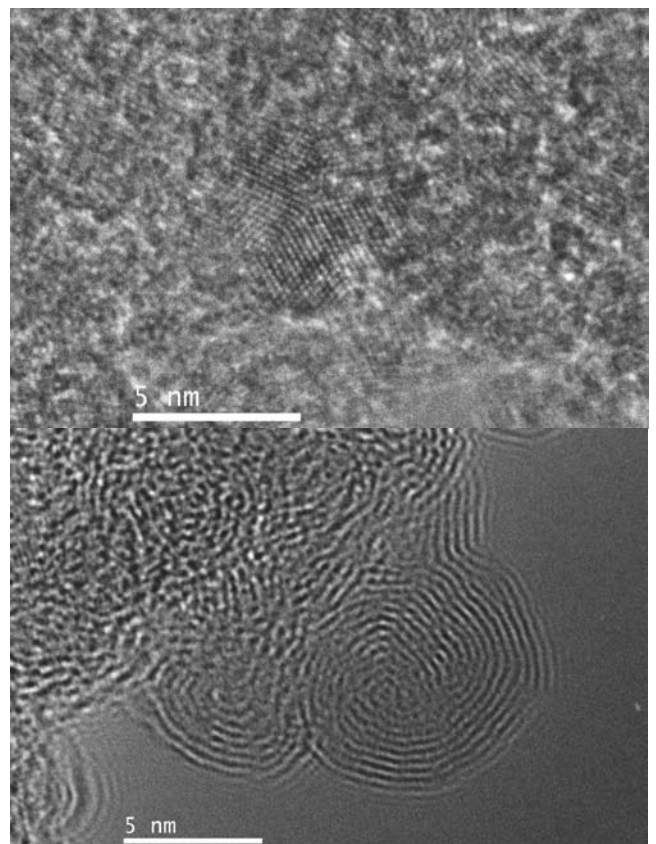


Fig. 6. TEM investigation of Allende nanodiamonds heated to 800 °C (from [37]). The area in the upper image shows nanodiamond, while the lower image is dominated by a region where diamonds have been transformed to onions

3.5. An age for presolar nanodiamond (?)

Determining an age for presolar materials is not a trivial task. Classical radioactive age dating using long-lived radionuclides is ruled out because of the nucleosynthetic anomalies that result in imperfect knowledge of the isotopic composition of mother and daughter elements before decay. For silicon carbide grains it has been possible to determine (with some uncertainty) the time they were exposed to cosmic rays during travel from their stellar sources towards the (forming) Solar System [44, 45], but the same approach does not work for the diamonds because of their chemical composition and small size. Cosmogenic ^3He has a recoil range of tens of micrometers [46], for example; however, ^3He contained in the cosmic rays themselves – where it is unusually abundant – may become trapped after having been slowed down, from which a presolar cosmic ray trapping (rather than exposure) age of some tens of million years has been estimated [47].

4. Conclusions

Nanodiamond contained in primitive meteorites with an abundance of up to 1500 ppm has been the first of the presolar phases in meteorites that has been identified, but possibly only some fraction of it is truly of presolar origin. An origin by a CVD process seems most likely but is not assured. The nanodiamonds contain noble gases with exotic isotopic composition that tie them to element synthesis in supernovae and that were probably introduced by ion implantation. Because of their small size it is not possible to perform chemical / isotopic analysis of single grains, which is in contrast to the case of the other identified presolar grains. This makes nanodiamond the most enigmatic and difficult to study type of “stardust” and even does not allow determining if all or only a small fraction of the diamonds is of presolar origin. For the same reason determining a presolar age is difficult. A possible option is a presolar cosmic ray trapping time based on ^3He . Many open questions remain that need to be studied by refined methods in future work.

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