

BIMODAL THERMAL RELEASE OF NOBLE GASES FROM METEORITIC NANODIAMONDS: ARE P3 NOBLE GASES LIKELY EVOLVED AT HIGHER TEMPERATURES? A. V. Fisenko¹, A.B. Verchovsky², L. F. Semjonova¹ and I. P. Wright². ¹Institute of Geochemistry and Analytical Chemistry, Russian Academy of Sciences, Moscow 119991 Russia; ²Planetary and Space Sciences Research Institute, Open University, Milton Keynes MK7 6AA, U. K.; e-mail: anat@chgnat.ru

Introduction. It is generally accepted that the enrichment of heavy and light Xe isotopes in Xe-HL is associated with *p*- and *r*-process nucleosynthesis in type II supernovae [1-3]. However these excess represent only a small fraction of Xe-HL in which an isotopically normal component dominates. The origin of the isotopically normal component remains unknown as does when and how the mixing with the anomalous nucleosynthetic components took place. Unsuccessful attempts to separate Xe-HL into individual components by a number of different methods have resulted in the conclusion that Xe-HL represents a single component formed before implantation into nanodiamonds [2-4]. This component is released from nanodiamonds at relatively high temperatures during stepped pyrolysis. The other, isotopically normal nanodiamond component, P3, is liberated at significantly lower temperature.

Recently, on the basis of experimental simulation of ion implantation into synthetic nanodiamonds, it was suggested that the high temperature release is also dominated by P3 noble gases i. e. HL is effectively the result of the release of a small amount of the highly anomalous supernova components along with P3 gases at the high temperature [5]. According to this suggestion the isotopically anomalous and normal (P3) components have been trapped in the same energetic sites so that during release by pyrolysis they cannot be separated from each other (and therefore appear as a single component). The suggestion is based on the similarity in the release pattern of artificially implanted noble gases into synthetic ultradispersed nanodiamonds (UDD) [6] and P3 and HL noble gases from meteoritic nanodiamonds [5]. Here we examine this suggestion using arguments which have not been considered by the authors of the original concept.

Results and discussion. From a general point of view it remains unclear how, during a single physical process of implantation, implanted ions create two different types of defects (or noble gas sites) in the diamond structure. If the sites existed in the diamonds before irradiation, then again it is unclear why synthetic and meteoritic nanodiamonds have the

same defects. Our experiments on ion implantation into the same synthetic nanodiamonds, in which we implanted monoisotopic ions, indicate that the release temperature of the implanted ions depends on total radiation dose: the higher the dose the lower the release temperature [7]. At a dose of $\sim 10^{16}$ cm⁻² the release temperature can be as low as 300°C, while at doses less than 10^{14} cm⁻² the release temperature is approaching 1400°C – the graphitization temperature of nanodiamonds. We have not observed a bimodal release of the implanted ions, although the peaks were broad. From the point of view of the observations the simplest explanation for the bimodal release of a mixture of several implanted noble gases could be that they are present in two layers of nanodiamond target in which implanted ions produce radiation damages corresponding to two different doses. The total dose, 3.2×10^{15} cm⁻², calculated for the experiment done by Koscheev et al. [5] is high enough to reduce the release temperature of the implanted noble gases to 300-500°C. In the second layer the dose was lower and hence the release temperature is high. In detail, however, when considering the grain size of the synthetic nanodiamonds and the energies of the implanted ions this becomes not so obvious. Thus, implantation of nanodiamonds with a mixture and monoisotopic noble gases show different results in terms of noble gas release pattern that have no satisfactory explanation so far. Therefore, explanation of the laboratory simulation experiments for the natural samples remains unclear.

Having very similar release pattern noble gases in synthetic and meteoritic nanodiamonds [5, 6] have very different variations in their elemental composition. If during pyrolysis of nanodiamonds from Orgueil, ³⁶Ar/¹³²Xe and ⁸⁴Kr/¹³²Xe monotonically decrease with increasing temperature, in the case of synthetic nanodiamonds the variations are quite different showing a clear maximum between the peaks of the noble gas release (Fig. 1). The observed trend in the elemental composition versus temperature for Orgueil nanodiamonds could be the result of mixing two components, P3 and HL, if each of them is dominated in one of the temperature

ranges of release: P3 with high $^{36}\text{Ar}/^{132}\text{Xe}$ and $^{84}\text{Kr}/^{132}\text{Xe}$ - at the low temperature and HL with low $^{36}\text{Ar}/^{132}\text{Xe}$ and $^{84}\text{Kr}/^{132}\text{Xe}$ - at the high temperature [8]. In other words, the systematic decrease of the ratios with increasing temperature is simply a result of lower abundance of P3 gases relative to HL. Variations in the elemental composition during pyrolysis of the implanted synthetic nanodiamonds are much more difficult to explain. At the moment it is not clear whether these variations just reflect different element distributions in grains of different sizes (as a result of different implantation ranges for the elements), or it is an effect caused by implantation into two nanodiamond target layers. Anyway, the difference in the elemental variation between natural and artificially implanted samples points to a significant difference in the implantation origin of noble gases in these two cases.

Finally, when considering the elemental ratios for the nanodiamonds from meteorites with different metamorphic histories [4], we can see systematic variations in these ratios which can be simply explained as a result of different P3/HL mixing ratios (Fig. 2). Being highly susceptible to thermal metamorphism the low-temperature P3 component is partly lost, while the more resistant, high-temperature component, HL, remains unchanged. As a result of this we see corresponding variation in the elemental and isotopic compositions (Fig. 2) which can be explained as two components mixing line. Moreover, the extrapolated $^{136}\text{Xe}/^{132}\text{Xe}$ ratio at $^{36}\text{Ar}/^{132}\text{Xe}$ and $^{84}\text{Kr}/^{132}\text{Xe}$ equal to zero gives values very close to the accepted $^{136}\text{Xe}/^{132}\text{Xe}$ in HL as was originally defined by Huss and Lewis [4].

Conclusion. Data available at the moment suggest that the high temperature component, HL, in the meteoritic nanodiamonds has not been formed during implantation of P3 and a highly anomalous supernova component. Rather we contest it results from the mixing of the supernova and isotopically normal (not necessarily equal to P3) components which took place before implantation (to form HL) and this has been implanted independently of the P3 component.

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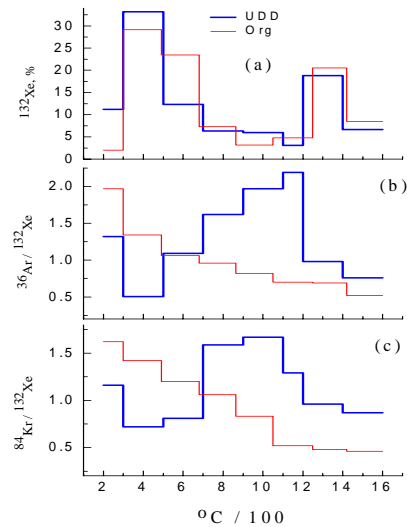


Figure 1. The release pattern of Xe (a), and variations of elemental ratios of noble gases (b, c) at pyrolysis of Orgueil CI (Org) and synthetic nanodiamonds (UDD) according to [4, 5].

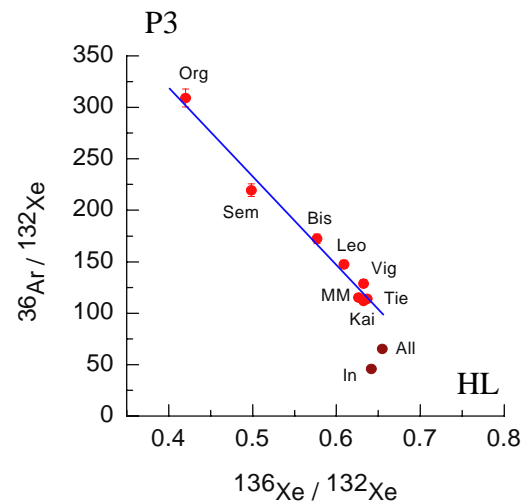


Figure 2. Variations of $^{36}\text{Ar}/^{132}\text{Xe}$ with Xe isotopic composition ($^{136}\text{Xe}/^{132}\text{Xe}$) for bulk samples of meteoritic nanodiamonds [4]. The data points make a mixing line between P3 and HL compositions. All - Allende CV3, Bis - Bishunpur LL3.1, In - Indarch EH3-4, Kai - Kainsaz CO3, Leo - Leoville CV3, MM - Mezö Madaras L3.5, Org - Orgueil CI, Sem - Semarkona LL3.0, Tie - Tieschitz H3.6, Vig - Vigarano CV3.