RADIATION-INDUCED NANO-DIAMOND FORMATION AND ITS IMPLICATIONS FOR INTERSTELLAR DIAMONDS

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Introduction: Interstellar grains of presolar origin are known to exist in the matrices of primitive carbonaceous meteorites; these grains include: aluminum oxide, graphite, silicon carbide, titanium carbide, and diamond. Diamond is unique among all presolar grains because of its association with Xe-HL. Several theories have been proposed for the origin of interstellar nano-diamonds, including chemical vapor deposition (CVD) from stellar outflows [1], impact shock metamorphism driven by supernovae [2], and UV annealing of carbonaceous grains [3]. Common to all these theories is the problem regarding the incorporation of Xe-HL into the nano-diamonds. However, this is not a difficulty if a radiation-induced formation mechanism is considered. High energy particles emitted during the supernova stage of a massive star (including Xe ions) could have interacted with carbonaceous grains in stellar envelopes [4]. These energetic particles may subsequently have become entrapped within the carbonaceous material when it recrystallized as diamond.

Based on the observation of submicroscopic diamonds in uranium-bearing sedimentary rocks with high carbon contents (kerogens, lignite, coal, and kerite) [5], Kaminsky [6] speculated that carbonados were formed in coal deposits irradiated by the spontaneous fission products of U and Th inclusions. Carbonado is a polycrystalline porous-aggregate of fine-grain diamond which is only found in placer deposits and is characterized by many features associated with the earth's crust including: biogenic carbon isotopic ratio, mineral inclusions of crustal assemblages, tightly trapped atmospheric noble gases, and the lack of association with kimberlites or lamproites. These suggest an alternative to formation from static high-pressure as in the earth's mantle. An indication to the mechanism responsible for the formation of carbonado may be found in the observation of a considerable amount of fission Xe and Kr in carbonados [7]. The presence of such large amounts of fission Xe and Kr can only be possible if the carbonaceous precursors to diamond (as well as the diamond themselves) were originally part of a finely dispersed U-rich material over geological time scales, consistent with the mechanism proposed by Kaminsky.

Discussion: In previous investigations [8,9], acid residues of carburanium [10], a U-rich ($\approx$5%) coal-like amorphous carbonaceous material of Precambrian age (1.7 $\pm$ 0.2 Ga) from North Karelia, Russia was examined with x-ray diffraction for the presence of diamonds; however, inconclusive results were obtained. In the present work, these acid residues were reexamined using transmission electron microscopy (TEM). The carburanium residue was observed to contain ($\approx$ 500 nm) polycrystalline aggregates, several of which yielded selected area diffraction (SAD) patterns demonstrating the presence of diffraction maxima that index well to that of cubic-diamond (Fig. 1). From the intensity of these reflections, it can be inferred that these aggregates contain a significant abundance of nano-diamonds. Furthermore, the width of these diffraction peaks places a lower limit of 26 Å on the size of the nano-diamonds after instrumental broadening is taken into consideration. Additional diffraction peaks are also observed in Figure 1, presumably arising from other resistant phases that survived the acid treatments. Energy dispersive x-ray spectroscopy (EDXS) of the aggregates indicated that in addition to carbon, Si, O, Fe, Al, and Mg with trace amounts of Ti were also present. High-resolution (HR) TEM images of thin areas near the perimeter of these aggregates reveal euhedral multiple-twinned nanocrystals with lattice spacings and cross-fringe angles consistent with that of the [110] zone axis of cubic-diamond (Fig. 2).

Therefore, the combination of SAD and HR-TEM results presents strong evidence supporting Kaminsky's hypothesis that carbonados are formed by the irradiation of carbonaceous coal-like materials with U and Th fission fragments. An energetic ion bombardment mechanism, applied to meteoritic diamonds, readily explains their nanometer size and, more importantly, their association with Xe-HL. Therefore, ion bombardment warrants further consideration as a possible mechanism for the formation of meteoritic diamonds that may compete, in stellar envelopes [4] or *in situ* in the host meteorites [11], with either shock metamorphism and/or CVD-type processes.

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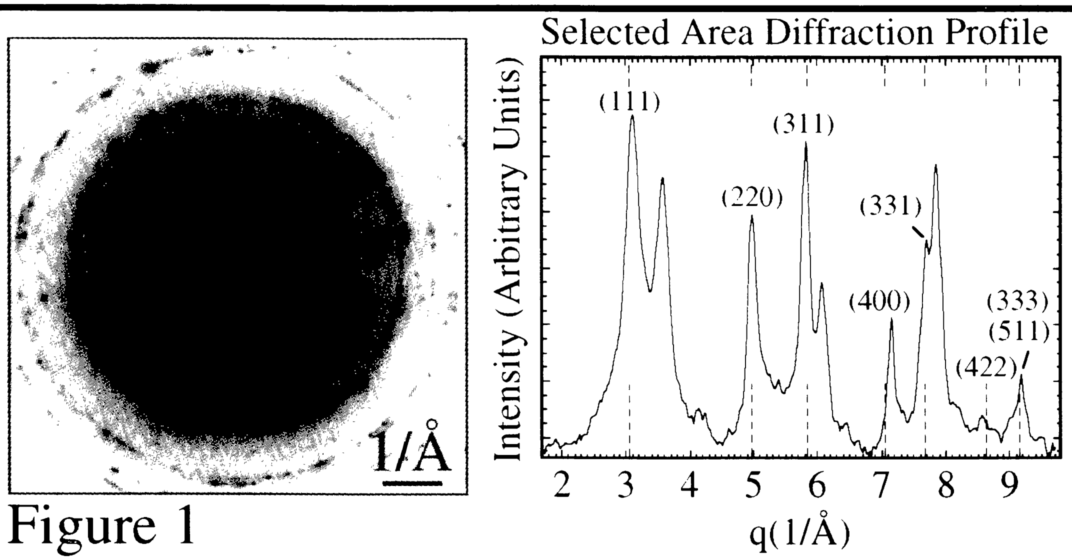

Figure 1

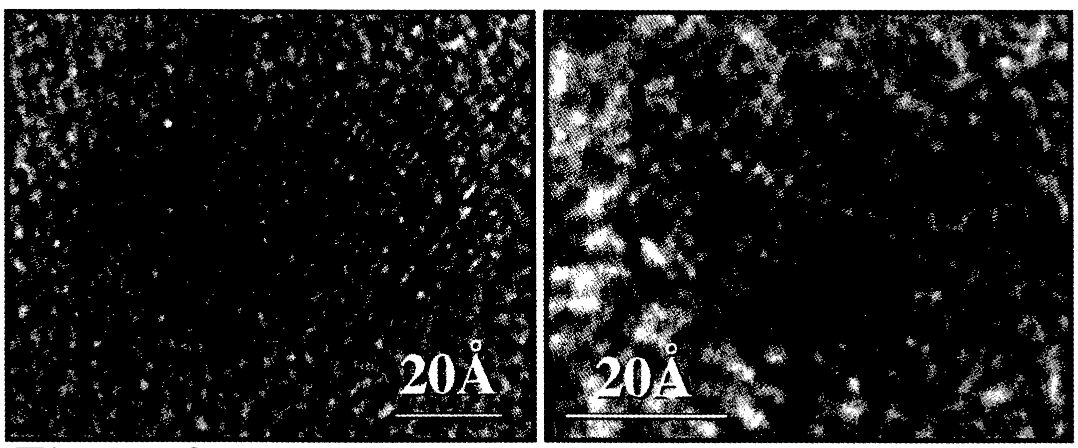

Figure 2

Figure 1: A selected area diffraction (SAD) pattern recorded from a polycrystalline aggregate observed in the carburanium residue. The profile plot measured across the SAD pattern shows that many of the major peaks can be indexed to cubic-diamond. For reference, all the diamond reflections that fall within the range displayed in the abscissa are indicated by dashed vertical lines. The background intensity was fitted to a power-law and subtracted.

Figure 2: HR-TEM images near the perimeter of the polycrystalline aggregates, which produce SAD patterns similar to that shown in Figure 1, reveal the presence of multiply-twinned nano-diamonds.

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