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The origin of porosity in carbonado diamond

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The C–C bonds in diamond are among the strongest in nature, and one might expect that diamonds would serve as an effective medium for waste storage. Our analysis of carbonados presents evidence to the contrary. Carbonados are black, polycrystalline diamonds with grain sizes that range from less than 1 to several hundred micrometers. Radiometric dating of carbonado and its inclusions yield formation dates of 2.6 to 3.8 Ga. The genesis of carbonado remains highly controversial. Proposed crystallization pathways include precipitation within mantle-hosted kimberlite pipes, transformation of organic carbon during a meteorite impact, and radiogenic conversion of amorphous carbon to diamond.

Profuse pores that range up to several mm in diameter are especially difficult to reconcile with the high pressures required to stabilize the diamond structure. It is unlikely that the cavities in carbonado originally contained mm-sized mantle minerals that have since dissolved, as no high-pressure phases have been documented in carbonado.

Based on previous spectroscopic studies and our new transmission electron microscopy (TEM) analysis, here we propose that the porosity in carbonado resulted from the dissolution of amorphous carbon produced by the spallation of ²³⁸U hosted by included phosphates. We cite the following lines of evidence for this hypothesis: 1) The most abundant mineral that lines the pores in carbonado is florencite, a rareearth aluminophosphate with high levels of radiogenic Pb; 2) Ozima and colleagues have documented large amounts of Xe and Ne from ²³⁸U fission implanted within carbonado Carbonado diamond: 3) specimens are strongly cathodoluminescent with the exception of dark rims that envelope the pores; 4) Raman spectroscopy by C. Magee indicates that these non-cathodoluminescent rims contain amorphous C; 5) We have extracted sections from these dark rims by focused ion beam milling and compared them with neighboring cathodoluminescent regions by TEM. Electron diffraction analysis of the rim sections reveals µm-sized areas of amorphization with high porosity at the µm and nm scales. These regions of localized damage due to MeV alphascattering appear to have been sites of preferential dissolution, creating macroscale porosity.

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Methylmercury uptake by natural zeolites

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The reaction of aqueous solutions of methylmercury (CH₃HgCl) were tested experimentally with 11 different zeolites under controlled physical and chemical conditions. Organic mercury compounds are highly toxic and persistent in the environment. The well characterized zeolites were selected mainly on the basis of their channel sizes (large), and included three clinoptilolites from Mongolia, the Mud Hills clinoptilolite (California), a clinoptilolite from South Africa, mordenite (Nevada), phillipsite (Nevada), analcime (Arizona), stilbite (Skye, Scotland), Na-chabazite (Arizona) and laumontite (Skye, Scotland). Mercury solutes were equilibrated with homoionic (Na) forms of the zeolites, all pre-labelled with a radiotracer, ²²Na (isotope dilution analysis). The ²²Na released into solution due to ion exchange with CH₃HgCl was analysed by liquid scintillation counting. Selectivity factors were assessed by determining batch distribution coefficient and sorption percentage of the exchanger as a function of contact time, mecury concentration and batch factor (solution volume to exchanger mass ratio).

Mongolian clinoptilolite-2 and the South African clinoptilolite were selected for further assessment of methylmercury uptake in the presence of competing cations and anions (K⁺, Ca²⁺, Mg²⁺, NO₃⁻, Cl⁻, SO₄²⁻, and PO₄³⁻). Generally, decreased sorption was found with SO₄²⁻ and increased sorption with PO₄³⁻. No clear pattern was found with increasing concentrations of Na, but sorption tended to decrease in the presence of 0.0001M Mg²⁺ solutions relative to 0.1M.

Acid modification of both the Mongolian and South African clinoptilolites resulted in increased sorption of methylmercury (to 100%) for all three acids; nitric, sulphuric and phosphoric.

In conclusion, analcime and Na-chabazite are the most promising for potential environmental clean up of organic mercury. Mordenite was the least effective, but all zeolites studied displayed evidence of considerable reaction with organomercury solutes. Acid modification enhances the uptake of methylmercury to 100%, consistent with similar studies using other heavy metal solutes.