

TRANSFORMATION OF GRAPHITE TO DIAMOND IN SHOCK EXPERIMENTS: A RAMAN STUDY.

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Introduction: Diamonds were found in many impact craters, e.g., [1]-[4]. A synthesis of diamond by shock wave experiments was first reported in 1961 [5], and is now widely commercialized, using artificial aggregates of porous carbon, quenchant particles and explosives. To our knowledge, a successful synthesis of diamonds from natural rocks in shock experiments has not been reported until now.

Methods: Shock recovery experiments were performed on cylindrical samples (15 mm diameter, 0.5 mm thickness) of a graphite gneiss of Kropfmühl, Germany. The gneiss consists of ~47 % graphite, ~35 % plagioclase, ~10 % quartz, ~3 % pyrite, and ~5 % biotite. We used a multiple reverberation technique with a high explosive device to achieve shock levels of 35, 45, 69, and 79 GPa in the samples, respectively. The technique is described in some detail, e.g., in [6]. A thin section of the 45 GPa sample was grinded with SiC and polished with Al₂O₃, the other samples were prepared conventionally. The Raman spectra of the polishing paste was carefully analysed to identify possible contaminations. Samples were analysed using optical microscopy, scanning electron microscopy (SEM) (without coating), and micro-Raman spectrometry (MRS). MRS was carried out with the notch filter-based system Dilor LabRam (HeNe laser, 632.8 nm, 100x microscope lens). The first-order Raman band in diamond is at 1332 cm⁻¹, and represents the main C-C bond vibration in diamond. Graphite can be identified by the dominance of the 1581 cm⁻¹ band. In the following the 1332 cm⁻¹ band is denoted as I₁, and the 1581 cm⁻¹ band as I₂. To characterize the phase transformation from graphite to diamond, we recorded (a) the I₁/I₂ ratio, (b) the full width at half peak intensity (FWHM), (c) the ratio of peak intensity to integrated background, and (d) the peak position. All measurements were carried out after removal of the background and Gauss-Lorentz fitting of peaks.

Results: Optical and SEM analyses show that most of the graphite grains persist during all shock stages, but they become heavily kinked and deformed with increasing pressure (Fig. 1). Quartz and plagioclase is transformed to diaplectic glass in all experiments. Biotite starts to melt at 35 GPa along graphite and maskelynite interfaces. A vesicular habit of the melt (Fig. 1) indicates that OH was released as vapour. EDX element mapping of the biotite melt shows high amounts of carbon incorporated into the melt. Very fine-grained

aggregates of carbon are present adjacent or within the biotite melt (Fig. 1). These aggregates become more voluminous with increasing pressure and primarily consist of disordered graphite, amorphous carbon and, to a small degree, of dispersed diamonds. Diamonds are typically very small (< 1 μm), and do not form graphite pseudomorphs.

Although the scattering is large, we found with increasing shock pressure, the following Raman characteristics for graphite (Fig. 3): (1) The Raman band I₁ displays a shift in position toward lower wavenumbers, (2) FWHM increases, (3) the peak intensity normalised to the background decreases, and (4) I₁/I₂ increases. The first peak becomes more and more prominent with increasing shock pressure (Fig. 2). Locally, very sharp I₁ peaks (Fig. 2 f), which represent diamonds, evolve in a strongly disordered carbon matrix. They are characterized by a I₁/I₂ ratio >10, a high background intensity (fluorescence), a FWHM < 20 cm⁻¹. Taking the broadness of the Raman bands as a measure of the degree of ordering, a continuous spectra range exist between graphite (Fig. 2a), disordered to amorphous graphite (Fig. 2b,c), and diamonds (Fig. 2d-f).

Discussion: Raman spectra of graphite are useful shock barometers, although the scattering of spectra characteristics increases with increasing pressure (Fig. 3). Diamond formation is restricted to the setting described above and cannot be explained via a martensitic transformation of graphite to diamond. It is the biotitic melt and its released OH content that potentially catalyses the reaction or provides suitable conditions for a persistence of diamond during pressure release. Diffusion of carbon is accelerated by the presence of OH. MRS showed that a broad range of intermediate carbon structures between graphite and diamond coexist with diamond. We favour a thermally activated formation mechanism of diamond [7]. Hot spots arose within the melt by vaporisation of OH. A sufficiently rapid quenching before the pressure decayed is probably due to the growth of the bubbles.

References: [1] Masaitis, V. L. et al. (1995) *MAPS*, 30, 541. [2] Koeberl, C. et al. (1997) *Geology*, 25, 967-970. [3] Schmitt, R. T. (1999) *MAPS*, 34, A102. [4] El Goresy, A. (2001) *Amer Mineral.*, 86, 611-621. [5] DeCarli, P. S. and Jamieson, J. C. (1961) *Science*, 133, 1821-1822. [6] Stöffler, D. and Langenhorst, F. (1994) *Meteoritics*, 29, 155-181. [7] DeCarli, P. S. (1995) *Mat. Res. Soc. Symp. Proc.*, 383, 21-31.

Figure 2

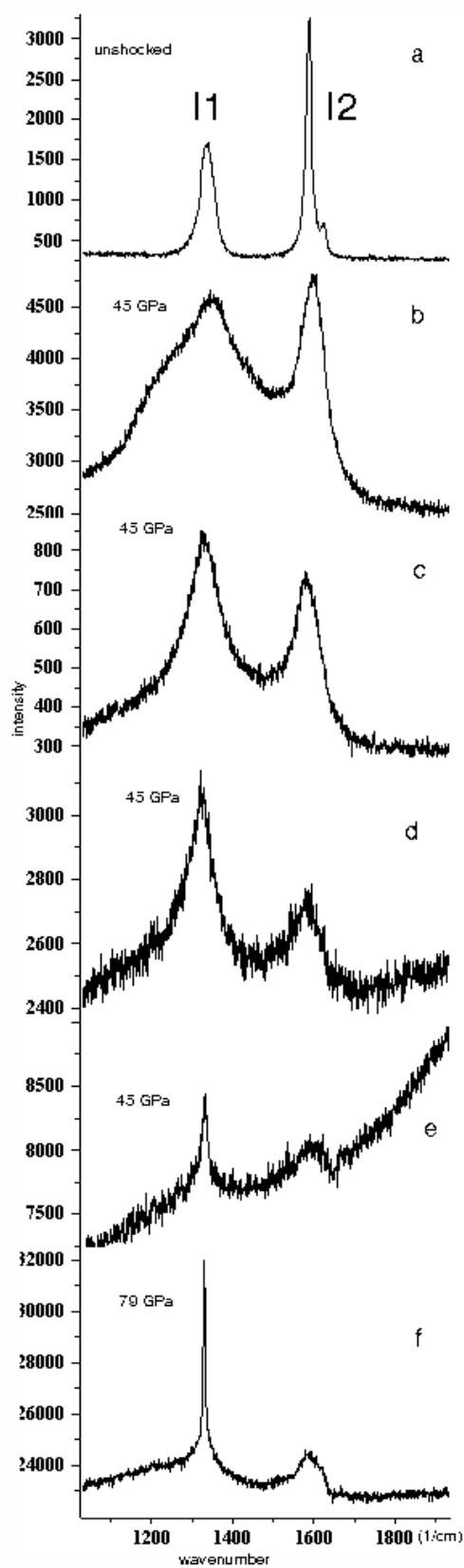


Figure 1 (a) 69 GPa, (b) 79 GPa

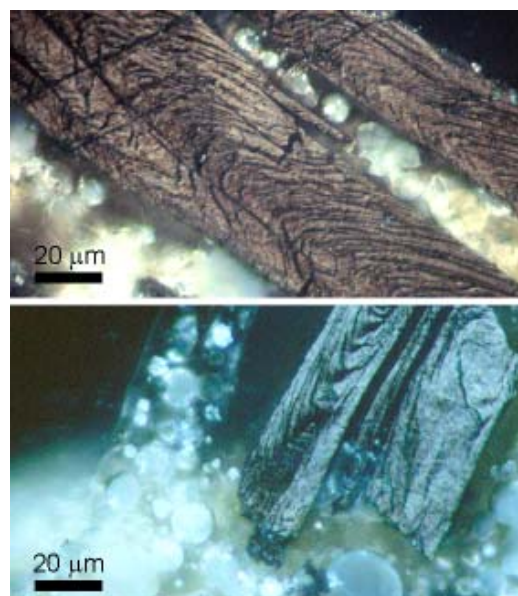


Figure 3

