

MINERALOGY AND *IN SITU* MICRO-RAMAN STUDIES OF CARBON PHASES IN UREILITE JAH

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Introduction: Ureilites are primitive olivine achondritic meteorites enriched in carbon which is presented by graphite, diamond, lonsdaleite [1]. The origin of high density carbon polymorphs in ureilites is debatable. Ureilite diamonds are generally thought to be produced from graphite by impact shock pressure [2]. An alternate hypothesis is the chemical vapor deposition (CVD) mechanism of diamond origin, developed by [3]. Based on our preliminary results reported here, we suggested that ureilite diamonds that simultaneously demonstrates characteristic features of shock and CVD origin could be the products of carbon multiprocessing that includes both of these mechanisms.

Analytical procedure: We studied thick polished section of the Jiddat al Horasis 054 (JaH 054), received from the Collection of Meteorites RAS. We used optical microscopy for petrographic description and Raman spectroscopy to analyze carbon material in JaH 054 *in situ* using Raman spectrometer Dilor XY-500 with the $\lambda = 488$ nm of an ionized-argon laser focused to an area of about $2\mu\text{m}$ diameter.

Results: Optical study of a thick polished section of JAH 054 in reflected light shows that carbon mainly fills the interstitial spaces between the olivine grains. Carbon occurs as elongated “blades” up to 1.5 mm length, comprised of graphite and diamond aggregates of different textures: 1) fine-grained graphite aggregates; 2) granular graphite-diamond aggregates (5-20 μm diamonds contained in graphite matrix); 3) layered ones – pile of ~ 1 μm of thick layers of diamond and graphite; 4) massive ones – relatively large (10-20 μm) aggregates of diamond grains surrounded by thin graphite rims. Fine-grained graphite aggregates occasionally demonstrate traces of tiny layered texture, oriented across the long dimension of carbonaceous blades. Granular graphite-diamond aggregates usually have thin linear graphite bridges oriented across the long dimension of the blades. The bridges could be result of impact event induced shear deformations. Geometry of massive diamond aggregates corresponds to that of host carbon blades indicating *in situ* genesis of diamond from graphite. Rare observed well polished areas of massive diamond aggregates demonstrate that they have optic anisotropy and thin (0.2-0.5 μm) layered texture, usually co-oriented with layers of surrounding graphite, that also could be an evidence of *in situ* transformation of graphite to diamond.

Average content of acid-insoluble carbon-rich material in JAH 054 is 3.25 wt%. The X-ray powder diffraction study of the insoluble material indicated that it comprised of graphite and diamond.

The carbon aggregate of granular structure (Fig. 1) was studied by Raman spectroscopy on the area of 100×100 μm . The received spectra show well defined peaks of diamond and graphite (Fig. 2). It should be noted that the FWHM (full width at half maximum) of the Raman line at 1332 cm^{-1} from the above diamond is about 8 cm^{-1} , using the slit width of 1.5 cm^{-1} spectral resolution. As well known, a crystal size affects only the Raman line itself. For instance, the nano-diamonds produced by different methods, 2-10 nm in size have Raman line at 1322 cm^{-1} , i.e. it decreases depend on the size. Position of characteristic line of a diamond at 1332 cm^{-1} indicates that the crystal size is from 100 nm to 200 nm, as usual Raman line of mono-crystal diamond. The above diamond peak with maximum at 1332 cm^{-1} is quite narrow, suggesting the small abundance of tensions of the lattice and well ordering of measured diamond crystals.

Raman spectra of graphitic material on the studied area exhibit one intrinsic band (so called G-band, 1588 cm^{-1}) [4] originating from the lattice vibration of graphite and in addition defect-induced band (so called D-band, 1355 cm^{-1}). The ratios of intensities of these two bands can be used for evaluation of the crystallite size and structural ordering of graphite materials. In our case, I_G/I_D is about 1.6 and it corresponds to semi-ordered graphite crystals with average size ~ 10 nm.

Discussion: According to [5], JAH 054 is the medium shock level ureilite. Variations in texture and modal composition of the carbon blades could indicate that solid-state phase transitions were out of equilibrium. The graphite aggregates seems to be affected by mechanic deformations of different intensity, from shearing of layered aggregated to crushing that leads to formation of fine-grained graphite blades. The deformations could take place before diamond formation and the texture of graphite could control a growth of the diamonds.

Occurrence of well-ordered mono-crystals of diamond, about 100-200 nm in size, within fine-grained graphite media of granular-textured carbon blade suggests an existence of appropriate conditions of diamond growth (carbon concentration, pressure, tem-

perature) during relatively long time. The narrowness of the JAH 054 nano-diamond peak mostly corresponds to natural endogenous diamonds rather than impact diamonds that have the wide peaks.

Sato [6] reported that the FWHM of the Raman line near 1330 cm^{-1} depends on the method of the diamonds synthesis. The FWHM for natural diamonds and for synthetic diamonds under static high pressure is less than 3 cm^{-1} . The FWHM for synthetic diamond formed under shock-induced high pressure is larger than 20 cm^{-1} . The FWHM for vapor-growth diamonds usually ranges from $6\text{ to }10\text{ cm}^{-1}$ and peak position of the Raman line is $1332.5 \pm 3\text{ cm}^{-1}$.

So, both the FWHM and peak position of the Raman line, obtained from diamonds within granular carbon blade of the JAH 054 are in the ranges of those of vapor-growth diamonds were previously measured. Earlier study of JaH 054 [7] showed that FWHM values of JaH 054 diamonds were similar to FWHM of CVD diamonds and differ significantly from shock-produced diamonds, what agrees well with the results obtained by [8].

These results suggest that observed nano-diamonds probably do not have impact genesis and could be formed by CVD mechanism. The massive diamond aggregates in JAH 054 probably were formed in situ from graphite under the shock pressure. However, the CVD-generated nano-diamonds, scattered in the source graphite, could be a nucleation centers for impact growth of the larger diamonds.

An origin of identified semi-ordered nano-crystalline graphite is not clear. It could be an intervening product of amorphous carbon ordering, that lead to formation of graphite sheets, or, instead, a result of disordering of well-crystallized graphite by shock deformations.

Conclusions: Our study showed that carbonaceous material in the JAH 054 ureilite could be affected by several shock events of different energy. Studied granular-textured carbonaceous blade probably contains nano-crystals of diamond, probably formed by the CVD mechanism. It could take place in interior of the JAH 054 parent body in a flow of methane, mobilized by internal or impact heating. These CVD-formed nano-diamonds could be deposited together with other endogenous carbon phases, and finally converted into graphite. Some of nano-diamonds scattered through graphite probably could be crystallization centers for a later shock - induced growth of the more coarse-grained massive diamond aggregates. Farther study of the JaH 054 carbon minerals *in situ* and in insoluble residue is planned to be done in future for understanding of a sequence of processes that lead to formation of carbon allotropes in the ureilite.

Acknowledgements: The work was partly supported by RFBR grants 12-05-00160-a and 12-05-01161-a. We are grateful to A. M. Bychkov, our former colleague, for help with X-ray analysis.

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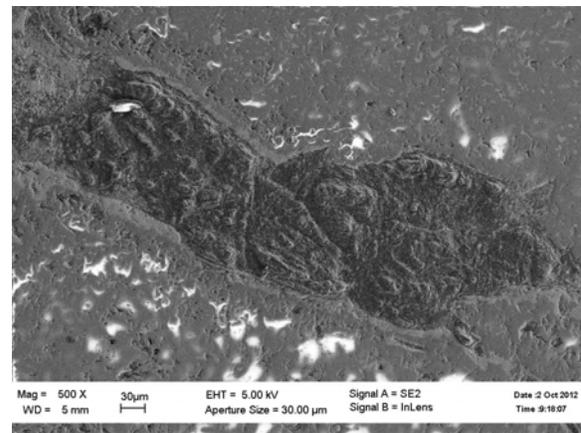


Fig. 1. The secondary electron image of carbon blade of granular texture in the JaH 054 ureilite.

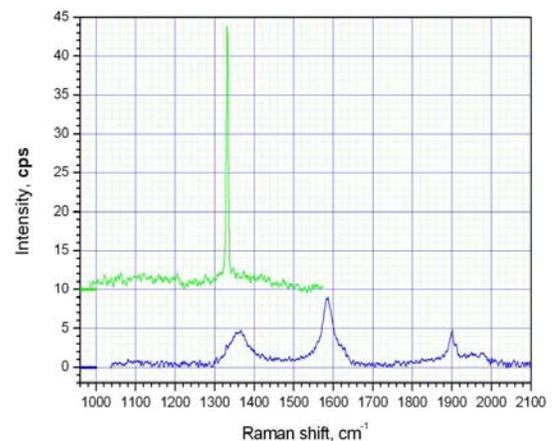


Fig. 2. Raman spectra of diamond (green) and graphite (blue) in the JAH 054.