

Defects in cubic diamonds from the placers in the northeastern Siberian platform: results of IR microspectrometry

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Abstract

Defects in yellowish-green, yellow, and orange diamonds of cubic habit from placers of the northeastern Siberian Platform were studied by IR spectroscopy. In addition to the main A, C, and, probably, B defects, the diamonds contain X and Y centers and amber defects of different types and show absorption bands at 1240, 1270, and 1290–1295 cm^{-1} , peaks in the region 1350–1380 cm^{-1} , and bands between 3100–3300 cm^{-1} . Diamonds of different colors contain different associations of structural defects, though they belong to the same variety II according to the Orlov classification. According to the integral spectra of the diamond crystals, the content of structural nitrogen impurity is low, 60–265 ppm. However, spatially resolved spectroscopic examination of diamond plates has revealed highly nonuniform distribution of defects in all diamond crystals. The general regularity for the studied diamonds is a decrease in the total nitrogen content and in the relative fraction of the major A defect from core to periphery of a crystal. The content of structural nitrogen impurity in the core reaches 900 ppm, which is higher than the average N content in widespread octahedral diamond crystals. The presence of C, Y, and X defects in the majority of the samples indicates short postgrowth annealing of these diamonds. The genetic significance of the obtained data on structural defects is discussed.

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Introduction

Diamonds with cubic crystals, forming a continuous color series from yellowish-green to yellow and orange and assigned to variety II according to the Orlov (1984) classification, are found in abundant placers in the northeastern Siberian Platform. The content of variety II diamonds reaches 12.7% (locally, even 50%) in the placers but does not exceed 1% in the Siberian kimberlites. The sources of these placers have not been identified yet, despite huge prospecting efforts (Grakhanov et al., 2007). The intense investigations of these placer diamonds are caused by the necessity to locate their sources (Logvinova et al., 2011; Shatsky et al., 2014; Zedgenizov et al., 2011). Diamonds of variety II are assumed to be related to the unknown type of diamondiferous rocks, in particular,

to eclogite massifs or metamorphic complexes (e.g., Zudina et al., 2013).

Structural defects in cubic placer diamonds of variety II were studied by different spectroscopic methods (Bokii et al., 1986; Mineeva et al., 2013; Nadolinny et al., 2012; Orlov, 1984; Samoilovich et al., 1972; Zudina et al., 2013). A set of specific defects was revealed, and correlation between the photoluminescence centers H3, S1, 525.1, 564.9, 574.5, 610.5, 615.4, 635.1, and 636.8 nm and the diamond color was established (Zudina et al., 2013).

The IR spectra of cubic diamonds of variety II assign them to the subtype Ia+Ib. A specific feature of many natural Ia+Ib diamonds is an intricate IR spectrum in the one-phonon region (Bokii et al., 1986; Collins and Mohammed, 1982; Hainschwang et al., 2012; Massi, 2006; Woods and Collins, 1983). For many diamonds the experimental spectrum cannot be satisfactorily described as a superposition of the bands of the A and C defects. This is due to the absorption of other optical centers, such as E, F (Clark and Davey, 1984), and Y

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(Hainschwang et al., 2012). The color of these diamonds is only partly related to the C defects (Hainschwang et al., 2013). Note that these spectral peculiarities are not observed in cubic diamonds of variety III rich in fluid and mineral inclusions (Zedgenizov et al., 2011).

In this work, we studied in detail structural defects in cubic gem diamonds of variety II from the placers of the north-eastern Siberian Platform. To elucidate the conditions of their formation and postcrystallization changes, we examined both bulk IR spectra of whole crystals and IR spectra of different zones of cut-out plates, which gave an insight into the distribution of structural defects in the crystals.

Samples and methods

We analyzed a collection of variety II diamonds from the Anabar River placers, which was earlier studied by EPR spectroscopy (Mineeva et al., 2013) and photoluminescence (Zudina et al., 2013). Sixteen faceted 0.25–2.06 carat crystals and six thin plates cut out of the 0.92–1.38 carat stones were examined. According to GIA atlas (King, 2006), the visual color characteristics of the crystals covered continuous color series of cubic diamonds from these placers, from yellowish-green to yellow and orange, of different hues and saturations. One of the faceted crystals was of red-orange color extremely rare among natural diamonds.

The IR spectra of the faceted crystals were recorded on a Perkin-Elmer 2000 Fourier spectrometer with a diffuse scattering attachment. The IR spectra of the thin plates were recorded on a Spectrum One (Perkin-Elmer) FTIR spectrometer equipped with an AutoImage FTIR microscope (aperture 100 μm). Measurements along the profiles across the optical growth zoning were made to study the spatial distribution of nitrogen defects; 2D maps of defects distribution were compiled for several samples. The spectra were recorded at a moderate resolution of 4 cm^{-1} because of the time limitations of the mapping. This has led to a slight reduction in the intensity of narrow peaks, e.g., the C defect peak at 1344 cm^{-1} .

After the baseline subtraction and standard normalization to the lattice absorption, the concentrations of the major nitrogen defects (A, C, and X) were calculated by decomposition of the one-phonon region of the IR spectra into individual components, using known absorption coefficients for each defect (Kiflawi et al., 1994). However, quantitative calculations of nitrogen content in natural diamonds with a significant fraction of Y defects are difficult because of the spectrum distortion (Hainschwang et al., 2012, 2013) (Fig. 1). For such crystals it is difficult to employ the absorption at 1130 cm^{-1} for estimation of the C defect content because of the shift of the absorption peak to ~ 1140 – 1145 cm^{-1} . The ratio $I(1344 \text{ cm}^{-1})/I(1130 \text{ cm}^{-1})$, where I is the absorption intensity at the corresponding wavelength, also decreases. Note that this behavior is observed even in the spectra with a resolution of 1 cm^{-1} and higher. The narrow peak at 1344 cm^{-1} is attributed to the vibration of carbon atoms around nitrogen, and the band

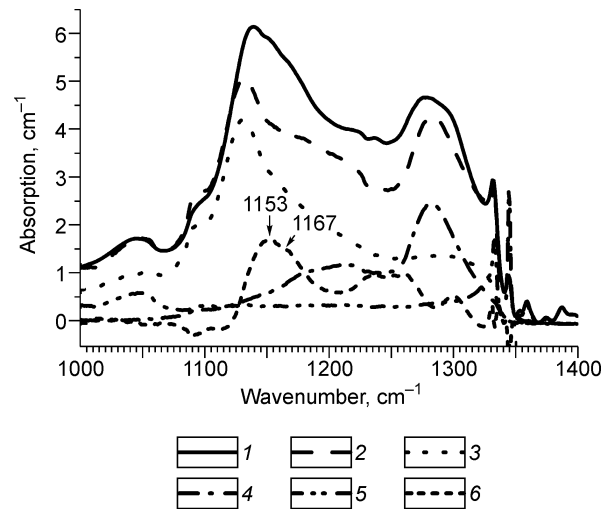


Fig. 1. Decomposition of the one-phonon region of IR spectrum of crystals with Y defects. 1, experiment; 2, (A + C + X) defects; 3, C defect; 4, A defect; 5, X defect; 6, residue. Arrows mark the position of the peak of the Y defect. There is a noticeable difference between the experimental data and the sum of components near the peak at 1344 cm^{-1} . For diamonds with Y defects, the absorption band of C defect differs from the standard one (Hainschwang et al., 2012).

at 1130 cm^{-1} involves vibrations of both nitrogen and carbon (Briddon and Jones, 1993). The change in the ratio of peak intensities is probably due to differences in the local environment of the C defects between such crystals and standard samples. Because of this uncertainty, the upper bound of nitrogen content is imposed by absorption at 1130 cm^{-1} , and the lower bound is given by absorption at 1344 cm^{-1} .

Results

Integral spectra of the bulk diamond crystals

The integral IR spectra of faceted crystals classify the studied diamonds as types Ia, Ia+Ib, and Ib+Ia. The content of nitrogen is low (60–265 ppm), which is common for variety II diamonds (Bokii et al., 1986). Most of the yellowish-green and greenish-yellow diamonds contain only A centers, up to 260 ppm (Fig. 2a). The yellow-orange diamonds have both A and C defects in comparable amounts (Fig. 2b, c). In the orange crystals C defects with concentrations up to 125 ppm dominate (Fig. 2d). As follows from the IR spectroscopic data, the relative formation temperatures of the yellow-green variety II crystals were apparently higher than those of the red-orange crystals.

Besides the well-known nitrogen defects, several other spectral peculiarities have been revealed in the studied cubic diamonds. After the decomposition of the IR spectra of most of the studied crystals into major components (A, C, and X) and their subtraction from the experimental spectrum, residual absorption is observed (Fig. 1). Such a residual spectrum with the main peak in the region 1145–1167 cm^{-1} has been recently identified in natural Ib diamonds and termed the Y defect

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