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VOLUME AND SURFACE DISTRIBUTION OF RADIATION DEFECT IN NATURAL DIAMONDS

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In the following article, we have studied the variations of natural irradiation of diamond crystals. The natural diamonds in some cases show homogeneous green colour, caused by irradiation of the entire crystals volume. Radiation damage effects, produced by the low-radiation doses, are detected by the luminescence of the GR1 system. The highradiation doses cause bluish hue, turning into a greenish colour, while the maximum level of volume irradiation produces the black crystals.

The crystals with homogeneous volumetric black colour distribution were also studied. The major source of radiation in such cases may represent a local stream containing water rich in ²²²Rn and its decay products. There is a review of the geological environment in which diamonds could be irradiated due to the decay of the ²²²Rn containing in water. *Key words:* diamond, irradiation, vacancy, FTIR

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Introduction. Diamonds from placers often record traces of irradiation in the form of irradiation spots [27]. The near-surface location, diameters of up to $60 \,\mu\text{m}$ and green or brown colours characterize these spots. Crystals with a completely coloured surface acquire a greenish or yellow-ish colour. A much more rare type of radiation traces is the homogeneous volumetric colouration of the crystals.

Numerous studies show that irradiation spots form at the points of contact between a diamond and radioactive minerals. Crystals with irradiation spots or green colouration are mainly found in placers [1, 21, 28], where heavy radioactive minerals accumulate along with the diamond. Diamonds from primary deposits less frequently have irradiation spots, and they are mainly contained in the weathering crust [1, 14, 19]. Crystals from primary deposits with green surface colouring [30] are encountered more often than those with green body colour [4]. Depending on the irradiation dose, diamond can acquire blue, light-green, and green to black colour [32].

Relevance. Black colour appears in diamonds after their *in vitro* irradiation with neutrons, protons, electrons, α -particles, and ions [32]. Only a few studies of artificially γ -irradiated diamonds have been performed [12] because diamonds absorption cross section is small [11], thus, a large exposure is required to achieve notable colouration. The visible spectral range absorption in irradiated diamond occurs on such crystal structure defects as neutral V⁰ vacancy and nitrogen-vacancy centres H3 and H4 [32]. The maximum transmission in the visible absorption spectra is 400-500 nm, which is perceived as a blue or bluish-green colour. Many other structural defects arise during irradiation, which is due to the displacement of carbon atoms from their equilibrium position. These defects cause a structureless (grey) absorption and produce narrow bands in the visible range absorption and FTIR spectra [15, 31].

Ionizing radiation of different types penetrates into the diamond at different depths. Ions, protons, α -particles, and electrons with energies up to 500 keV penetrate the crystal by 10-100 μ m and cause only surface colouring. Electrons with the energy of more than 1 MeV and neutrons penetrate inside the crystal and ensure a volume distribution of radiation defects. The «umbrella effect» is sometimes observed in irradiating large diamonds, due to the absence of radiation colour on the culet. The umbrella effect helped to determine an artificial colouring of a «Deepdene» 104-carat diamond [8]. The identification and study of crystals with a volumetric radiation-induced colour make it possible to improve the natural and artificial black colour diamonds separation methods. These studies also provide new data on the geology and features of the diamonds placer deposits formation.

Samples. This article provides the study of the nature of the colouration by the case of six black rough diamonds crystals from the Macaubas River, Minas Gerais, Brazil (Fig.1) and one black crystal from placer of the Bolshoy Shchugor River in the Middle Urals (VSEGEI collection).

Alluvial deposits with increased contents of radioactive minerals are widespread in Minas Gerais, and various geological objects with this characteristic are located in this region. Three Brazilian crystals are dissolved octahedra of 2-3 mm in size, whereas two crystals are rounded dodecahedra of 2-3 mm in size, flattened along the 4-fold axis, and one is a slightly rounded dodecahedron, 1.5 mm in size. The Urals sample U1 was 4.6 mm in size. It was previously flattened by grinding to a thickness of 1.2-1.6 mm.

All crystals have adamantine lustre, smooth surfaces and anthracite-black colour. Under the very bright transmitted light, only the smallest crystal, 10/22, weakly transmitted light with green colour; the remaining crystals are opaque.

Analytical techniques. During the first stage, we studied the crystals before the cutting process of plates (0.4-0.5 mm in thickness) from the Brazilian samples. The plates from the octahedra were cut using a laser along the [001] plane and then polished. The plates from the dodecahedron were made by grinding in the plane of flattening. The various plate production techniques were accompanied by various heating effect: the laser cut did not heat plates to temperatures above 400 °C, while the plates made by grinding were heated above 700 °C. After the above sample preparation, these plates were studied using a variety of methods.

Absorption spectra in the visible and infrared ranges (FTIR) were recorded using a VERTEX-70 spectrometer and a Hyperion 1000 microscope (Bruker). FTIR spectra were measured with a resolution of 1 cm⁻¹ and an aperture size of 100 \cdot 100 μ m. Spectra were recorded from the core, outer and rim parts of the plates to reveal possible internal inhomogeneities.

Nitrogen concentrations were calculated in the form of A and B1 aggregates; the absorption coefficients of the platelets band B2, α_{B2} , and those of the 3107 cm⁻¹ band, α_{3107} , were determined. Nitrogen concentrations were calculated based on known calibrations [9, 10]. The absorption spectra in the visible optical range were measured at a resolution of 8 cm⁻¹ at room temperature and 77 K using the Lincam THMS600 thermostat.



Fig.1. Photographs of some crystals under study and plates cut from them (bottom row). Samples 11/22, 11/24 are crystals from Brazil with a brown and green colour, the plates of which were made by grinding (with heating up to 600 °C) or cut by a laser (with heating up to 400 °C), respectively. The U1 crystal was sampled in alluvial placers of the Bolshoy Shchugor River in the Middle Urals

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Results. The crystals were opaque and transmission began from 720 nm and was minimal in the region of 750-1100 nm. At room temperature, only a shoulder at 740 nm and a system with a zero phonon line at 1076 nm was observed in the spectra. The IR absorption spectra show the absorption systems A, B1, B2, 3107 cm⁻¹. The FTIR spectra of all of the Brazilian crystals contained the band with a maximum of 1530 cm⁻¹ using an absorption coefficient, α_{1530} , of 0.6-0.7 cm⁻¹ (Fig.2, *a*).

The plates produced by the laser cut have very intense colours, and those made by grinding were brown (see Fig.1).The colouration of the plates is homogenous, and specific features in the colour of near-surface regions were not revealed. The plates do not contain any colour-inducing mineral inclusions. Minor variations in colour saturation were observed only in plate 11/25. All crystals have internal inhomogeneities in the nitrogen concentrations of the A and B1 aggregates and the absorption bands B2, 3107 cm⁻¹. Inhomogeneities in the distribution of the defects do not affect the variations in colour or intensity. The ranges of the nitrogen concentrations and absorption coefficients of the main systems are given in the te



Sample (plate colour)	A, ppm	B1, ppm	N _{tot} , ppm	$N_{B1},\%$	α_{B2}, cm^{-l}	α_{3107} , cm ⁻¹	$\alpha_{740}, \mathrm{cm}^{-1}$ (T = 300 K)	A_{740}, cm^{-2} (T = 77 K)
10/21 (brown)	770-1100	200-360	1460-970	19-25	2.2-6.5	0.1-5.3	5.2	800
11/22 (brown)	660-1020	<60	660-1080	<5	0-1.2	1.8-8.2	6	875
10/22 (brown, green before cutting)	160	200	360	55	9	0.9	4.5	1010
11/24 (green)	350-430	250-415	600-850	42-50	9.5-16.6	0.16-0.23	9.2	870
11/25 (green)	240-790	200-1350	850-1900	20-75	4-38	0.3-4	8	820
11/21 (green)	30-150	0-220	40-370	0-70	0-12.2	0-0.5	10.8	810
U1 (opaque)	140	50	190	26	0.6	0.8	-	-

Results of the plates study by absorption spectroscopy



Fig.2. Absorption spectra of: a – black diamond crystal from Ural

and plate 11/12, IR region; b - plates 11/22 and 11/24, visible re-

The 1530 cm⁻¹ band disappeared in the FTIR spectra of the plates from Brazil samples. Instead of this band, appeared bands with maxima at 1450 cm⁻¹ (H1a) and 1570 cm⁻¹. The absorption coefficient of the H1a band in different Brazilian plates ranges from 0.6-6.4 cm⁻¹, and the absorption coefficient of the 1570 cm⁻¹ band, α_{1570} , in all plates is approximately 0.8 cm⁻¹. The corresponding values for the sample U1 were 5.5 and 2.0 cm⁻¹.

Discussion. All studied crystals were initially opaque. This opacity is due to the very saturated homogenous green colour of the crystals. The green colour is a combination of the absorption bands GR1 and structureless absorption. As a result of irradiation, vacancies and interstitial atoms are generated in equal concentrations. The sign of the vacancies is the GR1 system, while the interstitial atoms are revealed by 1530 cm⁻¹ band. However, the mobility of interstitials is much higher; therefore, their concentration depends on the irradiation temperature and, apparently, its intensity. Until now, there have been no criteria to estimate the intensity of irradiation; however, there are criteria to determine the temperature above which crystals have not been heated after annealing.

The experiments on the annealing of diamond crystals with neutron irradiation [31] show the decrease in the amplitude of the 1530 cm⁻¹ band beginning at 200 °C; in another study [24] the decrease in the amplitude was observed already from 150 °C. Consequently, the studied crystals were not heated above 150 °C after natural irradiation. In the grinding process, the plates were heated to above 600 °C. These annealing parameters led to the structureless absorption decrease in the region of the phonon sideband GR1 and the change in colour from green to brown.

Let us analyze the possible radiation types that could cause a black colour. The ionizing radiation in natural systems is represented by α , β -particles, and γ -quanta. The neutron radiation is often used in laboratory experiments on the radiation defects [28, 31], but its intensity in natural systems is much lower. A unique system with neutron radiation is the famous natural nuclear reactor formed within the Oklo uranium ore deposits. During the operation of this reactor, the temperature in this natural reactor raised up to 500 °C [18], which is higher than the temperature limit established for the investigated crystals.

A study on the dependence of the increase in diamond volume on the neutron flux has shown the presence of interesting saturation effects [3]. As a result of neutron irradiation at 270 °C, crystal volume increases by up to 2-2.5 % (with a neutron flux of $2 \cdot 10^{20}$ cm⁻²), after which volume expansion stops [3]. With the irradiation temperature decrease, the threshold of this volume increase reaches 40 % [29]. The annealing of such diamonds allows the restoration of the crystalline structure if the volume increase is less than 12 %. The saturation effect, assessed at a temperature of 270 °C [3], is apparently caused by the dynamic restoration of the structure. The structure can be restored due to the extremely high mobility of interstitial carbon atoms, even at room temperature [14]. Similar processes of structural restoration can presumably occur as the result of the natural low-intensity irradiation. The stimulation effect of the crystal structure defects annealing by γ -radiation is also known [25], which can play a significant role in long-term processes. These factors can explain the similarities in the absorbances of the 1530 cm⁻¹ and 1570 cm⁻¹ bands and the GR1 system for all of the Brazilian crystals. Irradiating crystals with reactor neutrons with fluxes of above 10^{18} cm⁻² not only leads to the appearance of the 1530 cm⁻¹, 1570 cm⁻¹, and GR1 bands but also leads to the growth of structureless absorption, especially in the single-phonon region up to 1350 cm⁻¹ and above 2500 cm⁻¹ [20, 24]. This structureless absorption begins to decrease during annealing above 200 °C; this can thus be considered to be an indicator of high-flux artificial irradiation.

Alpha-particles penetrate into diamonds up to a depth of 10-30 μ m, where they form surface irradiation spots [7, 27]. The high radiation dose makes these spots visible in the surface texture and has very contrast boundaries, which is apparently the result of the high internal stresses, caused by an increase in the volume of the irradiated zone (Fig.3, *a*).





Fig.3. Images of crystals with spots of coloration: a – the surface of the crystal with brown spots in transmitted light, a contour is visible due to internal stresses; b – CL image (by A.V.Antonov) of diamond plates with traces of surface irradiation; the white arrow shows the luminescent zone of irradiation by a point source (from the radioactive mineral up to a few µm in size); both bright and dark areas (one of them is marked by a black arrow) are visible on the surface; this areas indicates increased dose of irradiation quenching luminescence

The irradiated regions without the specific colour can brightly luminesce under electron or photoexcitation. With the irradiation dose increase, the luminescence is completely quenched, and the irradiated regions acquire colour, becoming real spots of pigmentation. In Fig.3, b, the central part of the irradiated zone in which the luminescence is quenched is indicated by a black arrow. A clear boundary of the irradiated volume arises after penetration of the α -particles or electrons with a fixed energy into the crystal [27]. The white arrow also identifies the irradiated region with a clearly visible bright ring marking the boundary of the α -particles penetration from the radioactive mineral. For α -particles with the energy of 8.8 MeV, penetration depth is 28 μ m [27]. Radiation defects are uniformly distributed in a volume irradiated by a parallel beam of α-particles. The concentration of radiation defects in spots of pigmentation can be 20 times or higher than in the described black crystals. Thus, the diamond optical density in the coloured spots in the phonon sideband of the GR1 band can reach 0.3 [16, 27], which at an irradiation depth of 0.002 cm corresponds to an absorption coefficient of 150 cm⁻¹. In the black crystals under study, the absorption coefficient in the phonon sideband of the GR1 band reaches only 10 cm^{-1} . There are completely opaque brown and green spots with even higher values of the absorption coefficient [16]. In studied crystals, such near-surface inhomogeneities are either absent or weak (crystal 11/21).

The next kind of radiation, β -rays, is generated in many radioactive systems. In accordance with the Kanaya-Okayama formula [29], the penetration depths of electrons into diamonds at energies of 20 keV, 0.7 MeV, and 1 MeV are 2.5 µm, 1 mm and 2 mm, respectively. These values indicate that electrons with energies of less than 0.7 MeV could not induce a uniform concentration of defects in crystal volumes of larger than 2 mm. In the decay series of the isotopes of ²³⁸U and ²³²Th, the electron energy can reach 5 MeV (²¹⁰Pb, ²⁰⁸Pb), which corresponds to the penetration of electrons into the diamond to a depth of 30 mm. The maximal electron energies of some isotopes exceed 1 MeV (i.e., ²¹⁰Bi, ²¹⁴Bi, ²¹⁴Pb, ²²⁸Ac, ²³⁴Pa, ²¹⁴Po). A characteristic feature of β -decay is the continuous spectrum of electron energy, in which the average value of electron energy is one-third of the maximum value. Due to this continuous energy spectrum, the zone of natural electron irradiation in diamond has no clear boundaries. The concentration of defects is expected to decrease from the surface to the centre of the crystals due to the decay of short-lived isotopes with maximum β -decay energy of 1 MeV or less (²³⁴Th, ²²⁸Ra, and ²¹²Pb). Based on these considerations, β -irradiation, even that produced by the direct contact of a radioactive medium with a diamond,

could not cause homogenous colouration in a given volume. The crystals under discussion did not come in contact with radioactive minerals but were in a medium with a high radiation background.

The last possible form of radiation, which can lead to the volume induction of crystal structure defects, is γ -radiation, which always accompanies α - and β -decay. In laboratories, γ -radiation is very rarely used to induce radiation defects in diamond [16] because its absorption cross section is small and long exposure is necessary to achieve a significant concentration of defects. A diamond is considered to be «transparent» for γ -ray radiation, but its low-absorption cross section is not equivalent to the low defect generation rate. Calculations have shown that the efficiency of the generation of vacancies by γ -ray radiation is comparable with the efficiency of electrons of the same energies [11]. Thus, despite the low-absorption cross section, γ -radiation is a powerful generator of defects in the crystalline structure of diamond that is often not taken into account. Thus, in the study of plates cut from diamond crystals, we repeatedly detected the GR1 luminescence system in the photoluminescence spectra of the crystal core [2].

Based on the above data, the estimation of the neutron irradiation dose from the absorption coefficient of the 1530 cm⁻¹, GR1, and 1076 cm⁻¹ bands is possible only for experiments that are equivalent in terms of their temperature and flux intensity. In addition, there is a stress zone around each formed defect, within which the energy of atom displacement is reduced and the constants of the interaction of the matrix with radiation can vary significantly. Because of these factors, the rate of radiation defects formation increases with increasing flux [26]. Assuming that the concentration of induced defects depends only on neutron flux, the radiation dose can be estimated as follows: in the previous study [24], after undergoing irradiation with reactor neutrons with a flux of 10^{19} cm⁻² at 50 °C, the absorption coefficient of 1570 cm⁻¹ band was $a_{1570} = 5.5$ cm⁻¹. In the investigated crystals, a_{1570} is approximately 0.9 cm⁻¹, which corresponds to a hypothetical neutron flux of $1.1.6 \cdot 10^{18}$ cm⁻² [24].

It is now possible to attempt to evaluate the flux of γ -rays that is necessary to achieve the observed vacancies concentration. As shown in [11], γ -quanta with the energy of 1 MeV produce 0.03 vacancies per centimetre. Accordingly, to achieve the observed vacancies concentration of 10^{18} cm⁻³, a γ -ray flux of $3.3 \cdot 10^{19}$ cm⁻² is required. After irradiation by γ -rays with energies of 1.17 and 1.33 MeV (⁶⁰Co source) with a flux of $4 \cdot 10^{18}$ cm⁻², the absorption coefficient of the zero-phonon line of the GR1 system was 0.18 cm⁻¹ [12], which is by 8.5 times less than that observed here. To obtain the observed GR1 absorption coefficient, a flux of $4 \cdot 10^{18} \cdot 8.5 = 3.4 \cdot 10^{19}$ cm⁻² is required, which is consistent with the abovementioned calculated value.

It is thus possible to estimate the concentration of 238 U in the environment that is necessary to induce the observed number of vacancies in diamond as a consequence of γ -rays. Assuming that the radioactive, diamond-containing medium comprised a silicate-carbonate matrix, its absorption characteristics are equivalent to those of concrete. We restrict the considered volume to a zone defined by the 100-fold attenuation of quantum flux with an energy of 1 MeV (the attenuation of photon energy loss is not taken into account), which is 50 cm in porous concrete with a density of $\rho = 1.8 \text{ g/cm}^3$ [17].

The complete decay of the ²³⁸U series emits more than 50 γ -quanta, but the energy of the majority of these is less than 0.25 MeV [22]. Using explicitly overstated parameters: let the decay of a ²³⁸U atom emit 20 quanta with the energy of 1 MeV. The generation of the observed vacancies number requires flux that is created by a decay series of $1.65 \cdot 10^{18} \ ^{238}$ U atoms. In the model of an infinitely radiating medium, the number of decays of 238 U in 1 cm³ that is necessary to provide these quanta should be $1/50 \cdot 1.65 \cdot 10^{18} \ \text{cm}^{-3} = 3.3 \cdot 10^{16} \ \text{cm}^{-3}$. Over a geological duration of 1 million years, the required concentration, calculated according to the radioactive decay law, is C [²³⁸U] = $2.2 \cdot 10^{20} \ \text{cm}^{-3}$. Given our assumptions, this value is equal to approximately 2.5 wt %.



Such a high content would inevitably cause strong surface irradiation with α -particles, the signs of which were not observed here.

It is well known that the main γ -emitters in the uranium decay series are the volatile ²²²Rn and its products, which form short-lived isotopes. It is probable that the irradiation of the investigated diamonds occurred in a local stream containing water saturated with ²²²Rn and the products of its decay. The highest activity of ²²²Rn in water can reach 10⁸ Bq/m³ [6]. With indicated activity in the model of an infinite medium, the duration of diamond irradiation to reach a neutral vacancy concentration of 10¹⁸ cm⁻³ is 10⁹ years. This length of irradiation for diamonds in placer is not geologically reasonable. To explain the observed colour, it is thus necessary to assume that in fairly rare geological situations, radon activity can reach a value of 10¹¹-10¹² Bq/m³ (10¹¹ at/cm³). In the case of irradiation caused by the products of ²²²Rn decay, the contribution of colouration by α -particles is less significant than by γ -quanta. Each decay of ²²²Rn and its products forms four α -particles (5.3-7.7 MeV). The number of α -particles crossing a diamond surface from an outer 30-micron-thick layer to achieve the specified vacancy concentration is 4·10¹⁴ cm⁻². As was shown by Nasdala et al. [27], the patterns of the 10¹⁴ cm⁻² (8.8 MeV) neutron flux cannot be recognised in photographs. In comparison with irradiation spots, black diamonds are «colourless»: the absorption coefficients of their GR1 system is 15 times lower.

The samples from Brazil have the same concentration of primary radiation defects, and hence either the same dose of irradiation or the saturation effect, similar to that observed with neutron irradiation. The uniqueness of crystals with such a black colour makes us doubt that six samples could have the same dose of irradiation under natural conditions, and therefore we should allow the possibility of artificial irradiation of these crystals.

Geological conditions that can ensure long-term γ -irradiation of diamond crystals should satisfy the following requirements:

1) since diamonds with volumetric black colour are found in placers, there should be specific lithogenic conditions, favorable for their formation;

2) a large required exposure involves Precambrian age of primary placers and their continued safety before erosion and diamonds transportation to recent placers;

3) conditions favorable for the accumulation of radioactive minerals should be realized in diamond placers;

4) there should be an active water exchange, which determines the transfer of radon from the radiation source of to diamonds.

These conditions are possible within the uranium deposits of the craton basement [5]. Uranium and gold-uranium deposits in ancient conglomerates and unconformity-related deposits are most closely aligned to these conditions. In these deposits, uranium mineralization is confined to horizons of Precambrian coarse clastic rocks that are favourable for the accumulation of alluvial diamonds. Diamonds with a green coloured surface are well known in the early Proterozoic uranium-bearing conglomerates of the Witwatersrand deposit. Both types of deposits are structurally related to superposed Proterozoic basins with significant depths of depression, which determines the possibility of active water exchange during water migration from the sides (benches) of the basins to its central most depressed area.

The typical environment of the unconformity-related deposits is favourable for the specified formation conditions of diamonds volumetric black colour in the presence of this mineral in the basal horizon of coarse clastic rocks. Such conditions do not require the uranium accumulation in high concentrations typical for industrial deposits. In these cases, an increased uranium concentration in the unconformity area and a long-term migration of waters containing dissolved radon are sufficient. The considerable duration and polychronism of the given uranium deposits formation [5]

also work towards the achievement of the γ -irradiation dose required for the diamonds volumetric black colouration.

The craton uranium deposits of other types could also be the radioactive source. For example, the rocks of crystalline basement bedding the Brazilian diamond placers are characterized by the presence of iron-uranium deposits. These rocks could also serve as a radon source.

Thus, spots of colouration are a kind of radiation exposure traces independent of crystals volumetric colour. These two colouring types can coexist within one crystal, but the geological conditions of each colouration type are completely different. The stability of radiation defects in diamond makes it possible to use it as a natural dosimeter by analogy with kaolinite, quartz, feld-spars. The green volumetric colour of the famous diamonds «Orlov», «Green Dresden», «The Ocean Dream» [8], apparently has a radiation nature too.

Conclusions. The study shows that, in addition to widespread surface pigmentation spots, the crystals can have a volumetric colouration of the radiation nature. In rare cases, volumetric irradiation can lead to a black colour induced by long exposures of γ -radiation. Here, the high concentrations of uranium-bearing minerals can be the radiation source. In these settings, crystals should have near-surface layers with a stronger colouration gained due to α -irradiation. The source of radiation in such cases can be explained by a local source of 222 Rn, whose decay products are effective γ -emitters but do not lead to intense contact patterns of α -irradiation. Black diamonds with radiation colour are rare in nature. Moreover, these crystals are not extracted by luminescent separators and also can be incorrectly diagnosed during hand picking. Similar stones with less pronounced green body colours occur not only in the Urals and Brazil, but also in Yakutia, Africa, and Borneo. Constraining the nature of the geological environment in which diamonds receive their colour requires further research.

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