NITROGEN AGGREGATION AND LINEAR GROWTH RATE IN SYNTHETIC DIAMOND SINGLE CRYSTALS

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When examining single crystals of synthetic diamond grown in the metal-carbon system, it was found that the content of A centres (two nearest-neighbour substitutional nitrogen atoms) with respect to C centres (single substitutional nitrogen atoms) increases appreciably in octahedral growth sectors formed at a higher average linear growth rate [1]. Present work is concerned with a more detailed study of the dependence of nitrogen aggregation rate (C=>A) on linear growth rate, which is derived from particularities of distribution of nitrogen defects in synthetic diamonds.

The examined synthetic diamond was grown using BARS high pressure apparatus in the Fe-Ni-C system at P=6.0 GPa and T=1470-1495^{o}C. Additionally the temperature gradient method was combined with the induced zoning technique. In this case the temperature was varied during the growth process, amplitude of oscillation being about 3 degrees (relative to nominal temperature) and oscillation period being 22.3(3) minutes. Finally the single crystal of a cube-octahedral habit exhibited pronounced growth zoning, which was readily seen when a 0.4mm plate was cut from this crystal along (110) and viewed under a microscope. The zoning observed is proposed to be a result of micro-fluctuation of some impurity concentrations (for example, nitrogen, nickel) in diamond structure. Presence of zoning allowed us to determine the time of any point formation by direct counting the number of growth zones (of known duration) between the point and seed of the crystal. The absolute size of zones, which were formed for a mentioned period of time, provided us with the information about the linear growth rate at a certain point.

In order to study the local distribution of C and A defects we employed IR-Fourier micro-spectroscopy (Bruker IFS 113v, A-590 microscope, 50μm round aperture). Nitrogen contents in the form of A and C centres were derived by decomposing the spectrum in one-phonon region into corresponding components with subsequent calculating of the concentrations using absorption coefficients reported in [2,3]. Previously evaluated N^{+} (1332cm-1) component was determined only in near-seed area in concentration no higher than 2 ppm and so was left out of account.

Some linear sections in octahedral growth sectors of the diamond plate were chosen for detailed inspection. Having data on contents nitrogen defects and the duration of annealing, which is determined with the help of zoning, the rate constants of transformation of C centers into A centers were derived for all points of analysis. The calculations were performed using a second order equation $dC/dt = -KC^2$, where C is the concentration of dispersed nitrogen, K is the rate constant, t is the annealing time. This equation describes kinetics of simple homogeneous irreversible reactions of diffusion type and is usually used to describe nitrogen aggregation in diamond (it should be noted, however, that possible deviations from this model were reported and discussed in [4-6]). Then derived the rate constants were correlated with linear growth rate values obtained by measuring zoning at points of analysis. The relationship between nitrogen C=>A aggregation rate and linear growth rate in octahedral growth sectors of the sample inspected has the following main peculiarities:

The aggregation rate constant can changes widely within the separate octahedral growth sectors of the same single crystal (from $3.0*10^{-7}$ to $7.5*10^{-10}$ $ppm^{-1}*sec^{-1}$, e.g., LnK= -15.0 to -20.9);

The aggregation rate constant varies non-linearly depending on the linear growth rate in octahedral growth sectors. It increases sharply if the growth rate increases from 20 to 40 μm/h and is almost invariable at V_{lin}>40 μm/h.

Taking into account that "the concentration of nickel is related to the kinetics of diamond growth rather than to the growth temperature itself" [7] we propose that determined relation of aggregation rate and linear growth rate is connected with the process of nickel incorporation in diamond lattice. It is indirectly supported by the presence of intensive yellow-green photoluminescence and N^{+} - component in the near-seed area, that indicate the presence Ni-containing centers in the region where growth and aggregation rates were highest. Also a sharp increase in the aggregation rate constant as linear growth rate increase from 20 to 40 μm/h is seems to attribute to a progressive increasing of nickel concentration in the diamond lattice.

In conclusion it should be noted that the wide range of nitrogen aggregation rate observed in octahedral growth sectors of diamond single crystal could account for existing quantitative discrepancy between experimental data on kinetics of this process in synthetic diamonds [4, 8-11].

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