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An atomic force microscopy study of diamond dissolution features: the effect of H2O and CO2 in the fluid on diamond morphology

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Many lines of evidence indicate that diamonds undergo significant dissolution while residing in the mantle and during the ascent in kimberlite magmas. Recent experimental studies have demonstrated that the composition and conditions of diamond-destructive fluids dictate the morphology of surface dissolution features on diamonds. Dissolution features recorded on the majority of natural diamonds can provide a direct tool to study fluids in the mantle source of diamonds and magmatic fluid in kimberlites. However, the lack of any comprehensive model explaining the nature of the various dissolution features limits examination of diamond surfaces to purely descriptive. This study for the first time uses atomic force microscopy (AFM) to obtain detailed crystallographic information for the different types of diamond resorption features and to examine their dependence of the oxidizing media. It allowed us to identify a new type of diamond surface features with much higher relief than the commonly described trigonal etch pits and to quantitatively characterize crystallographic orientation of the micro-faces, which form individual etch pits on diamonds.

The study uses diamonds resorbed in experiments in piston-cylinder apparatus at pressure of 1 GPa and temperatures 1150, 1250, and 1350oC in H2O-rich and CO2-rich fluids, and natural resorbed diamonds recovered from some Canadian kimberlites. Diamond surfaces were examined under optical and scanning electron microscope and then AFM images were collected. Dissolution features were examined on (111) face of diamond crystals using a tapping AFM mode with the scanned areas 50x50 µm to 1x1 µm. The AFM results show that the well-described diamond resorption features (trigonal etch pits and hillocks) while well seen in optical microscope and SEM, have often hardly any relief. On contrary, small "dirt-like" spots present on all experimentally dissolved and natural diamonds and always ignored in the studies of diamond dissolution features, at high magnification of AFM appear as regular "pyramid-like" features with the height (up to 400 nm) well above the depth of the trigonal etch pits. Their longer axes are oriented along [001] or [011] direction of diamond lattice depending on fluid composition and the temperature. Their occurrence on experimentally resorbed diamonds indicates the development during dissolution and not growth processes. The size of the "pyramids", the orientation, and the length to width ratio vary with the temperature and H2O:CO2 ratio in the fluid. Section analyzes of the AFM images of macro- and micro-trigonal etch pits allowed to identify crystallographic indexes of the faces forming the trigonal pits on different diamonds and their dependence from the oxidation conditions. This confirms that different volatile species in diamond-destructive fluids preferably attach to the directions of diamond crystal lattice with the configuration of carbon – carbon bonds fitting better the configuration of the molecules in the fluid (or melt). This controls the relative oxidation rates in different directions of diamond crystals and determines the type of the resorbed surface on diamonds. Furthermore, AFM results show that diamonds preserve no pristine surfaces untouched by resorption. What appear to be flat remnant of (111) faces under optical microscope and SEM, is a combination of several surfaces with an angle ∼2o. The initial results of AFM investigation of diamond resorption features demonstrate that the detailed crystallographic information can be obtained for extremely small features down to 300 – 500 nm. Similar resorption features on diamonds oxidized at different conditions differ by the indexes of crystallographic faces forming these features. The widespread micro- "pyramids", described here for the first time, are very sensitive "tracers" of the conditions in the reacting fluid and can possibly be used to constrain emplacement conditions in rising kimberlite magma.