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MORPHOLOGIES OF FISSION FRAGMENT IMPACTS IN DIAMOND AND SILICA

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Abstract

The morphologies of fission-fragment impact craters in diamond and silica were investigated by atomic force microscopy. The impacts produced micron-sized craters that were especially obvious in diamond; irradiations in air may have allowed the cratering in carbon to be oxidatively enhanced. The ejecta deposit preferentially at ordered sites and have the appearance of hillocks of a few tenths microns in size. On quartz, the hillocks have a parallel-perpendicular, x-y pattern; on diamond, the hillocks form one dimensional, parallel rows. In contrast, the hillocks on amorphous silica fiber show a random pattern.

The likely explanation is that back sputtering from impact sites produces plasma haloes from which condensation occurs on surface faults having the geometric arrangement of the observed hillocks. The fission fragments, with mean energies of 79.4 and 103.8 MeV, were emitted from a californium source, and collimated through air to impact the targets at near normal angles.

Introduction

As an extension of our multiyear efforts to understand and measure radiation - induced, damage tracks in solids (1,2), we have added atomic force microscopy to study surface damage on a nanometer scale. In the current work, we chose to irradiate diamond and silica in air with the fission fragments from californium (^{252}Cf) having the binary distribution of energies, 79.4 and 103.8 MeV. We focused our attention on the morphologies of the impact sites and of the deposited ejecta.

Experimental Procedures

The atomic force microscope (ATM) was made by TM Microscopes-Veeco Metrology Group, model Autoprobe CP Research. The ATM probe was a silicon cantilever with a silicon conical tip of height 5.7 microns and 10 nanometer tip radius, and applied to the target with a force of 10 nN. More details can be found in our recent companion article(3).

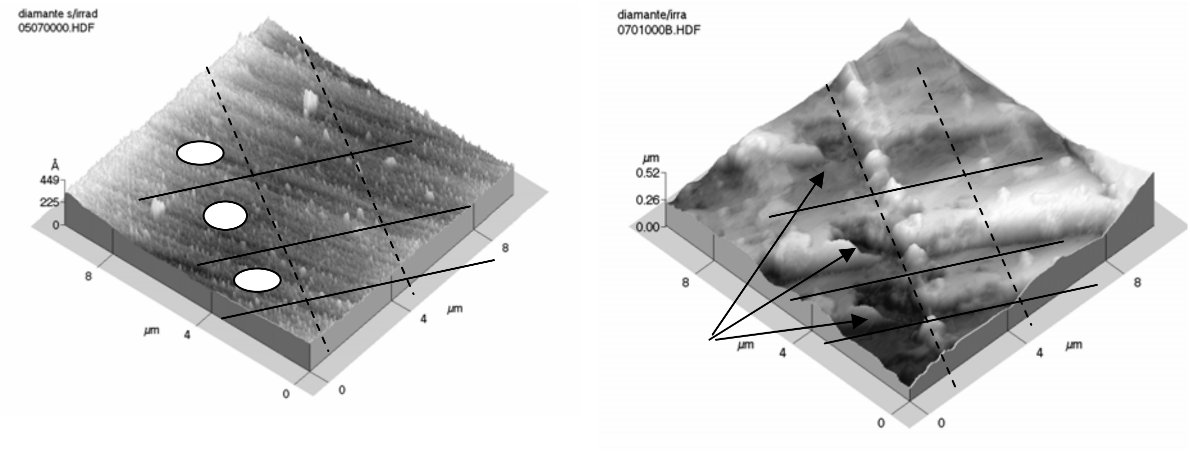
Three target materials were investigated: commercial silica optical fiber (Nokia fiber), natural quartz crystals cut to small size, and small natural diamonds of about 1 mm.

The radiation source was ^{252}Cf with an activity of 98 fission fragments (ff) per min/cm^2. Irradiations were made in air through an aluminum foil, 10 microns thick, and though which collimating holes had been made by pricks with a fine needle. Thus the ffs were collimated approximately normal to the target surface.

The calculated rate of surface impacts was 1 ff per week per area of 10,000 microns^2. Irradiations were made for up to 20 days and produced about 5 impacts per zone observed in the AFM.

Results and Discussion

ATM micrographs of diamond surface, before and after irradiation, are shown in Fig.1a, and 1b; three impact craters are arrowed in 1b with the analogous positions on smooth, undamaged surface being shown in 1a. The grid helps one to recognize two parallel rows of hillocks, the more distinct row being the one closest to the line of impact craters. The scale shows that the irregularly shaped craters have dimensions of up to a few microns, while the hillocks appear to be a few tenths microns in size, up to a maximum of 0.5 microns.



a) b)

Fig. 1.- Diamond surface images:
a) original surface; and b) after exposure to fission fragments.

A more detailed image of an impact crater - Fig. 2 - reveals a quite irregular, three-dimensional shape and with a depth of nearly 0.5 microns in the center of the crater.

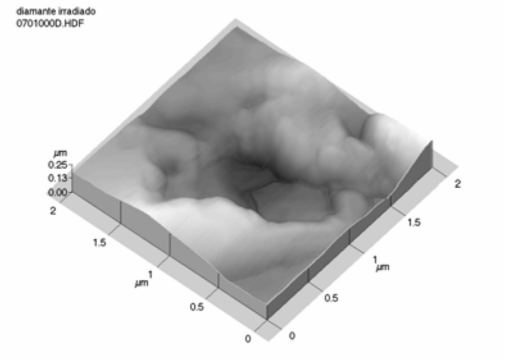
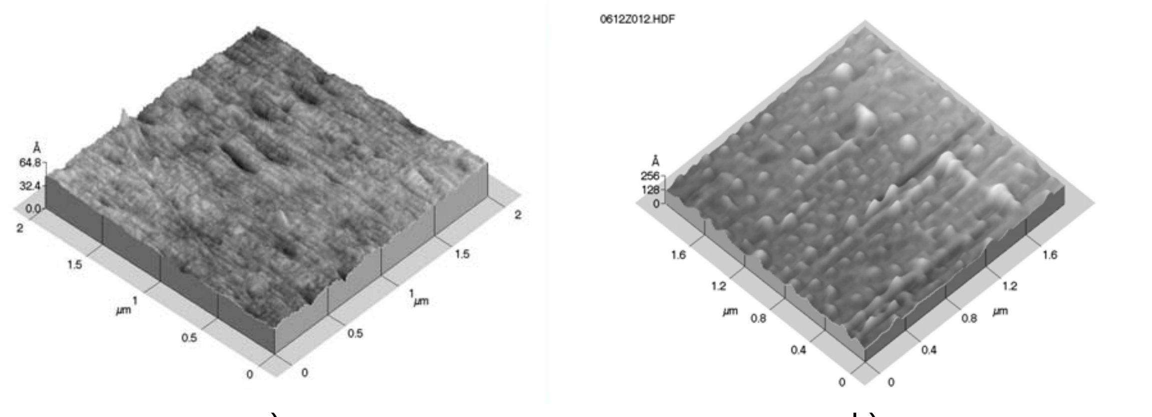


Fig. 2.- Detail of three-dimensional impact crater in diamond surface.

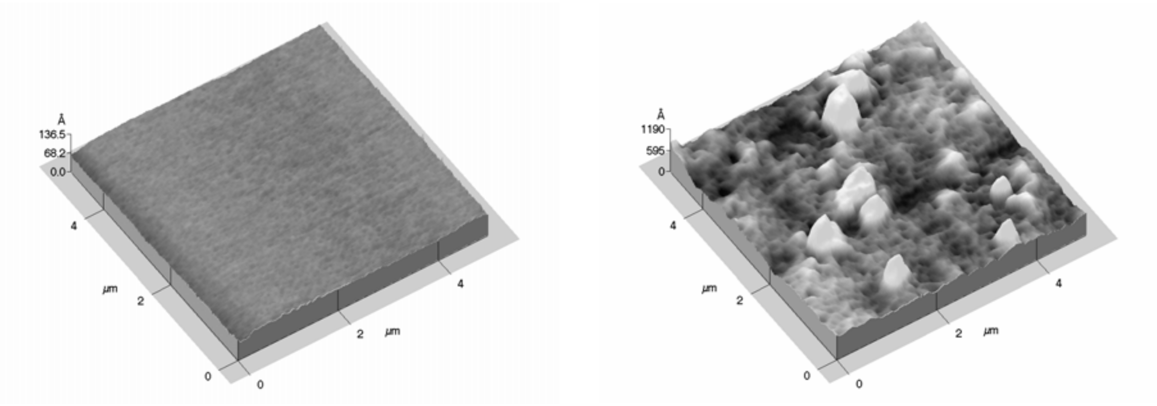
Views of unirradiated and irradiated quartz are shown in Fig. 3a and 3b, respectively. Impacts result in a well- developed array of rounded hillocks, in the size range 0.05 to 0.2 microns, and in a distinct x-y array (Fig. 3b). There is little evidence of any impact cratering, in contrast to the situation with diamond. The unirradiated quartz surface, (Fig. 3a), shows a smooth topology with features of on the scale of 10 Å.



a) b)

Fig. 3.- Quartz surface: a) unirradiated, and b) irradiated.

Fig.4 has the analogous AFM micrographs of amorphous silica fiber. The original fiber surface (Fig. 4a) is smooth on a scale above 15 Å. After irradiation with ff's, a random pattern of hillocks is clearly seen (Fig. 4b), with heights of up to 0.07 microns. This finding contrasts with the ordered patterns of condensed ejecta that are produced with the crystalline samples. We are undecided on whether or not the darker regions seen in Fig. 4b are indicative of cratering.



a) b)

Fig. 4.- Optical fiber surface: a) unirradiated, and b) irradiated.

Let us now consider the physical processes involved in ff impacts on the targets, and their relevance to the observations we have seen. The fundamental sputtering process is one of converting the forward momentum of the projectile into reverse momentum of the target components - the ion explosion-spike model. The projectile ions, having energies of the order 100 MeV, produce a rich halo of ejecta ions that, after neutralization, condense back on the target surface. In the case of the crystalline surfaces, deposition can be seen to occur preferentially on geometrically ordered sites. It seems reasonable to assume that these sites are protruding surface faults, such as steps, jogs and intersecting stacking faults. The amorphous silica fiber would, in contrast, be expected to exhibit a random pattern of deposition sites, and it does.

In diamond there is obvious cratering at impact sites. The irradiations were conducted under atmospheric conditions so that some oxidation of carbon ions would have taken place in the plasma haloes, as well as at the impacts sites themselves. Perhaps this condition is promoting oxidatively enhanced cratering that we see in the diamond but not for the silica. Future studies of ff impacts on diamond should be done in a non-oxidizing atmosphere, such as nitrogen, to determine the role of oxygen in cratering on diamond.

The crystalline form of the condensed carbon ejecta will be graphitic, not diamond. This expectation was confirmed using microscale, X-ray diffraction examination of the hillocks on irradiated diamond. Not surprisingly, the material in, and in the immediate vicinity of the craters was also graphitic.

As well as craters and hillocks, the ff's produce damage tracks penetrating into the bulk of the targets. These damage tracks below the surface are largely invisible to the AFM. A subsequent etching will reveal these damage tracks as etch pits, as was found recently in silica by Espinosa et al(4). The range of the ff's in diamond was calculated using SRIM-2003.26; most ff's penetrate to a depth of about 8 microns, with energy deposition of 4.7 MeV per cm.

To investigate further the damage in the subsurface, etching of ff tracks in diamond should, in the future, be attempted in an oxidizing medium. Possibilities are oxidants such as hydrogen peroxide solution, or heating in air at temperatures that are mildly combustive

The higher resolution micrograph (Fig. 3) of a crater in diamond shows a flat-bottomed crater. The flattening is perhaps caused by disrupted atoms relaxing back into a minimum surface area configuration. Craters of a similar nature have been observed by AFM in other organic crystals such as benzoyl glycene (5).

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