A new detection method in studying penetration depth of low-energy heavy ions in botanic samples

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Abstract A new detection method has been applied to study the penetration depth of low-energy heavy ions in botanic samples. Highly oriented pyrolytic graphite (HOPG) pieces were placed behind the target samples with certain thickness to receive energetic penetrated ions during the irradiation. After irradiation, statistic number density of protrusion-like damage induced by energetic penetrated ions can be obtained through scanning tunneling microscope (STM) observation on the surfaces of HOPG. The results of test indicate that the detection limit can be as low as 1.0×10^9 protrusions/cm^2. With the method, the penetration depth of at least $60\,\mu$m can be detected in kidney bean slices irradiated by N^+ ions with dose of $0.3\text{-}3\times10^{17}$ions/cm^2.

Keywords Low energy ion irradiation, Transmission, Botanic slice, STM observation, HOPG (piece)

CLC numbers O571.33,TN305

1 INTRODUCTION

Low-energy heavy ion implantation has found its new application in crop breeding in recent years. It has been reported that the implantation of 20~300 keV N^+ ions into dry plant seeds such as rice and wheat in the dose range of 10^{15}-10^{17} ions/cm^2 produced significant inheritable mutation effects[1]. The mutation effects are guessed to be induced by the direct interaction between energetic implanted ions and the DNA molecules in the embryo of seeds. However, based on the LSS theory[2], the range of such low-energy ions in condensed materials is no more than $1\,\mu$m, far less than the distance from the embryo to the surface of the seeds ($100\,\mu$m at least).

It is difficult to clarify the problem because the measurement requires high sensitivity. In the crop breeding by high-energy ion irradiation, ions can penetrate through the embryo of seeds and produce penetration to DNA molecules in the embryo, the dose is no more than 1×10^8 ions/cm^2 [3]. Similarly, if the mutation effects were induced by direct interaction in the crop breeding by low-energy heavy ion implantation, the dose of

Supported by the National Natural Science Foundation of China under Grant No.19675004 and (No.19890300)

Manuscript received date: 2001-03-26

energetic ions which can produce damage on the DNA molecules in embryo would also be low. In the former experiment, only high sensitive method such as particle induced X-ray emission (PIXE)[4] and secondary ion mass spectrometry (SIMS)[5] can detect implanted ions in the 100 μm deep region below the surface of the plant seeds. While in the detection by the scanning electronic microscopy with EDAX analysis, the concentration depth of 90~200 keV implanted V^+ ions is no deeper than 30μm below the surface in wheat and peanut seeds [6]. Another difficulty is the requirement of the accuracy in the high sensitive measurement. Because the botanic samples are very tender, large measurement errors may be induced by any treatment after ion implantation, such as the sputtering in SIMS detection[5] and the cutting in PIXE detection[4]. Besides the possible measurement errors, another problem in detecting the concentration depth profile of implanted ions also needs to be taken into consideration. Diffuse will become severe by the accumulation of heat in the irradiation, and will induce the migration of implanted ions from the surface to the deep region of seeds. Since the diffuse ions or neutral atoms do not have enough energy to break chemical bonds in DNA molecules, they can not cause mutation effects.

To deal with the above difficulties, the transmission measurement is used in our experiment. In the transmission measurement, a certain thick slices of botanic samples, such as seed coats, are used to be bombarded by ion beam, the energetic ions penetrated through the samples are detected by high sensitive detectors. These ions have enough energy to break chemical bonds in DNA molecules and are able to produce mutation effects in the region deeper than the thickness of the samples. In our former transmission measurement experiment with a semiconductor detector, kidney bean slices of 120μm thick were irradiated with MeV F^+ ions and the penetrated energetic ions were detected *in situ*[7]. The ion transmission ratio kept at a very low level. It was at most 10^{-6} even after 1000 s irradiation. But to tens of keV incident heavy ions, semiconductor detectors become incapable, because the low-energy penetrated ions fall in the high noise region and even lower than the detection limits of these detectors (usually 18 keV). Normal solid track detectors, such as a highly sensitive detector CR-39, are also ineffective because of the short projectile range of low-energy heavy ion. In this article, we report a new method to detect the penetration depth of low-energy heavy ions in botanic samples with transmission measurement.

2 METHOD

2.1 Principle of detection

According to the experimental set-up of the method illustrated in Fig.1, ions will pass through the ion windows of target cap and bombard the slice samples. They will lose their energy in collisions with the atoms in botanic slice samples. Some of the projectiles may penetrate through the slice samples. The recoils in collisions will also probably have enough energy to escape from the slice samples. All these primary and secondary penetrated particles will impact on the surface of the HOPG (detecting pieces) that are placed just behind each botanic slice sample, producing damage on the surface of the HOPG piece. After irradiation, STM are used to observe the damage.

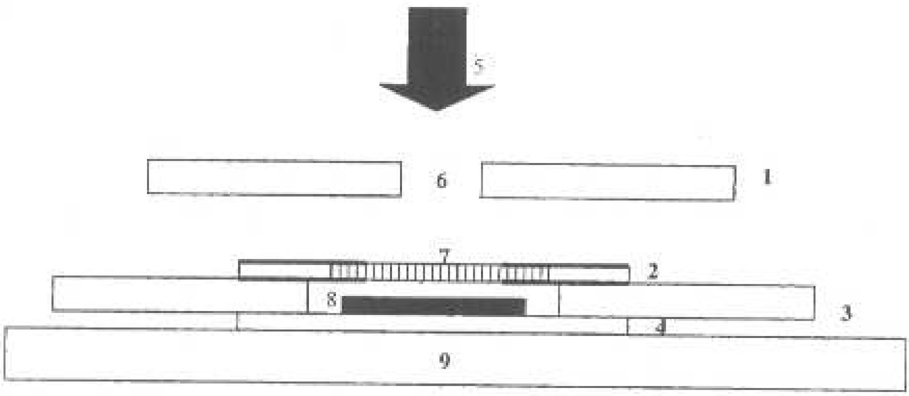

Fig.1 Schematic diagram of the transmission measurement
All the target cap, spacer, board, target sample holder and HOPG piece substrate are made of Al
1.Target cap, 2.Target sample holder, 3. Target spacer, 4.HOPG piece substrate, 5.Ion beam, 6. Beam window, 7.Target sample, 8.HOPG piece with flat surface, 9. Targe board

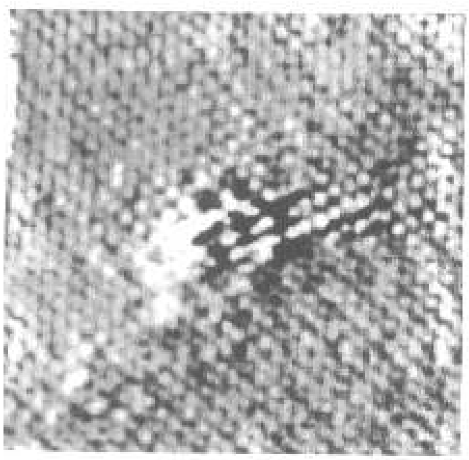

Fig.2 Typical topography of protrusion-like damage on the surface of HOPG detecting pieces placed behind slice samples ($10\times10\,\mathrm{nm}^2$)

The atomic resolution of STM makes it highly sensitive to observe isolated damages induced by single ion bombardments on the surface of HOPG. When the fluence of incidence ions is low in the irradiation, individual protrusion-like damage can be observed on the surface of HOPG with STM. As illustrated in Fig.2, the typical topography of damage is a hillock with the diameter of 1~6 nm and the height of 0.1~0.3 nm separated by flat terraces on which the normal atomic order is reserved. If the number density of damage exceeds 10^{13} protrusions/cm^2, the overlapping of damages causes the loss of the atomic resolution on the surface of the HOPG and the HOPG becomes amorphous[8]. It means the detection range can be no more than 10^{13} protrusions/cm^2.

2.2 Detection efficiency and the scale of dose

The mechanism of the damage formation on the surface of the HOPG is thought to be the creation of displacement of botanic target atom by the ion impact, followed by the change of topography of HOPG or the increase of electronic density. It has been reported that one and only one protrusion can be produced by each incident ion that has nuclear stopping power in the range of 1.5~30 eV/nm at the impacting point on the surface of HOPG[9]. This hypothesis is justified in experiments such as the damage production probability of each Ga^+ ion with the energy above 250 eV is about 1, and the probability decreases as the energy decreases[10]. The simulation of TRIM program[11] indicates that the nuclear stopping power of N^+ ions with the energy between 180 eV and 25 keV is just in the range mentioned above. Therefore, the detection efficiency of low-energy heavy ions such as N^+ ions is near 1 through the STM observation of HOPG, and the dose of ions bombarding HOPG is almost the same as the average number density of protrusion in the detection.

2.3 Detection accuracy and background

The detection accuracy of the method is determined by the background number density of protrusions. Careful STM observation on the surface of HOPG pieces which are placed in the ambient without direct irradiation verifies that similar structure like the protrusion can not be observed except those very close to the directly irradiated regions. The possible background is thought to come only from the scattering ions in the irradiation. This can be supported by two points: The scattering coefficient of ions with energy of several keV is as high as 0.5[12], and the scattering ions have enough energy to produce protrusion-like damage on the nearby surface of HOPG pieces if they impact some steps away on the HOPG surface. Experiments for tests were designed to study the influence of scattering ions in irradiation.

In order to study the existence of scattering ions in the target, some HOPG pieces were attached on the back of target cap or on the target board far away from the ion windows. The target was irradiated by 40 keV N^+ ions at the dose of 3×10^{16} ions/cm^2. After irradiation, the STM observation showed that these HOPG pieces attached on the target cap and board both have the number density of protrusions near 3.0×10^{10}/cm^2. The result demonstrates that there are ions scattering between the target cap and target board. They can also impact on the HOPG surface and produce protrusions.

In the other test experiment, the target samples were aluminum foils. They were chosen to be 30μm thick so that all the incident low-energy heavy ions would stop in them. Two kinds of HOPG set up were used. For one kind, each HOPG was enveloped. The spacer was a washer attached firmly between the target sample holder and HOPG's substrate, each HOPG piece was put in the hole of the washer, just behind the aluminum foils put below the ion window in the irradiation. For the other kind, HOPG were not enveloped. The spacer was two pieces of shims attached on the two edges of HOPG's substrate. Targets holder was attached on the shims. The ion windows above the samples were covered by a 1mm thick aluminum pieces, so the target samples would not be irradiated by incident ions. After irradiation by 40 keV N^+ ions at the dose of 1.0×10^{17} ions/cm^2, HOPG surfaces were observed with STM. It was found that the number density of protrusions on the enveloped HOPG pieces is no more than 1.0×10^9/cm^2, indicating that no protrusions can be found on enveloped HOPG surfaces when ions can not penetrate through target samples. For HOPG surface that were not enveloped, the number density of protrusions was found larger than 1.0×10^{11}/cm^2. The result demonstrated that some energetic ions bombarded HOPG surface in the irradiation though the target samples over them were not bombarded by ion beam. The protrusions on them can not be produced by the energetic ions penetrated through the samples, but the scattering ions in the irradiation.

Based on the above results, we conclude that it is necessary to make each HOPG to be enveloped to avoid the influence of scattering ions in order to keep low background. If the HOPG pieces are enveloped, the scattering ions can be successfully prevented from impacting on the HOPG surface. The number density of protrusions induced by penetrated ions can be as low as 1.0×10^9/cm^2 in the detection. The protrusions with the number density of more than 1.0×10^9/cm^2 are sure to be induced by the penetrated ions through target samples. In the following experiment, each HOPG piece was enveloped.

3 EXPERIMENT

The new transmit measurement were applied to detect the possible penetration depth of low-energy ions in ethylene terephthalate (PET) films and botanic samples.

The botanic samples were kidney bean seed slices with thickness of 30, 60 and 120 μm, prepared with traditional biological techniques. The average density of the slices samples is about 1.18 g/cm^3. The thickness of PET ($C_{10}H_8O_4$) films is 8 and 36 μm and the density of them is 1.397 g/cm^3.

N^+ ion beam with the energy of 40 keV produced by ion implanter was used in the irradiation experiment. The current density of the ion beam was about 8μA/cm^2 and the ion irradiation dose was 3×10^{16} ions/cm^2 and 3×10^{17} ions/cm^2. The vacuum in the irradiation chamber was about 10^{-3} Pa. Under the same ion beam condition, several samples were irradiated to meet the statistical requirement.

The topography of all the HOPG surface without further treatment were observed by an STM after irradiation. Usually, the tunneling current was 2 nA, and the bias voltage was −300 mV. STM tips were made by etching tungsten wires with NaOH solution. To get the statistical number density of protrusion-like damage on HOPG surface, at least three regions in each HOPG piece were selected randomly. In each region, 6×6 matrices of 100nm× 100 nm images, which was chosen as the unit area for protrusion counting, were obtained. Image of 100nm×100nm is acquired at a pixel density of 4 pixels/nm^2 to allow nanometer-scale features to be detected and to avoid missing protrusion in counting. In each sample at least one hundred unit images were collected.

4 RESULTS AND DISCUSSION

On all the HOPG behind PET films, the number density of protrusion is no more than the background in the detection. Since the dose of the ions to irradiate the PET films reaches 1.0×10^{17}/cm^2, the probability for the 40 keV N^+ ions to penetrate 8 and 36 μm thick PET films is no more than 10^{-8}. The results fit the simulation of TRIM program. This indicates that the method is feasible to detect the penetrated ions through a certain thickness target and the sensitivity of the detection can be very high.

For botanic samples, protrusions have been observed on the HOPG samples and the number densities of protrusions decrease with increasing thickness of botanic samples. As shown in Table.1, on the HOPG samples behind 30 μm and 60 μm thick slices, the number densities are 1.0~ 8.0×10^{11}/cm^2 and 0.9~ 3.0×10^{11}/cm^2 respectively. Fig.3 shows the typical image in the observation. Since both are much larger than 1.0×10^9/cm^2, the protrusions are sure to be produced by the energetic ions penetrated through the 30 μm and 60 μm thick slices. The ratios of protrusion (the ratio of the number density of protrusion to irradiation dose) are as low as $10^{-6}\sim10^{-5}$ and $10^{-7}\sim10^{-6}$, respectively for 30μm and 60 μm thick slices. It indicates that the probability of ions to penetrate such depth is very little. For 120 μm thick slices, the densities are less than 1.0×10^9/cm^2. It is still unclear whether there are energetic ions penetrated through kidney been slices with thickness larger than 120 μm.

Table 1 The number density of protrusions on HOPG samples

Target samples	Irradiation dose/cm^2	Number density of protrusion/cm^2	Ratio of protrusion
30 μm slices	1×10^{17}	7.8×10^{11}	7.8×10^{-6}
30 μm slices	3×10^{16}	9.3×10^{10}	3.1×10^{-6}
60 μm slices	1×10^{17}	1.3×10^{10}	1.3×10^{-7}
60 μm slices	3×10^{16}	6.0×10^{9}	2.0×10^{-7}
120 μm slices	3×10^{17}	$< 1\times10^{9}$	$< 3.0\times10^{-9}$

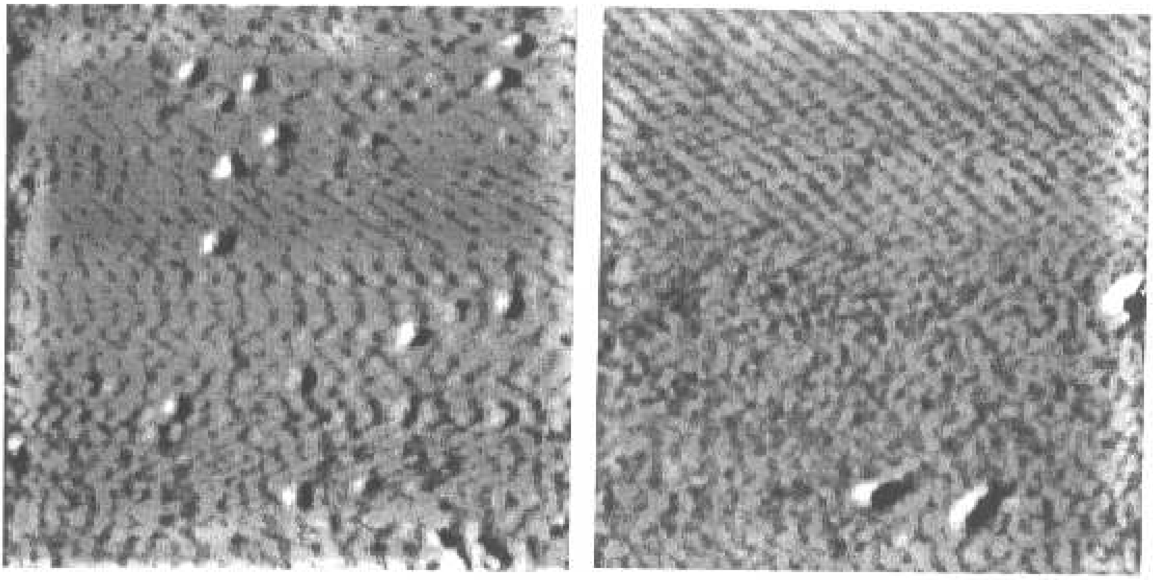

Fig.3 The typical image ($100\times100\,nm^2$) of HOPG samples irradiated with 40 keV N^+ ions
Left. 30 μm slice irradiated with the dose of 1×10^{17} ions/cm^2
Right. 60 μm slice irradiated with the dose of 1×10^{17} ions/cm^2

The results confirm that a very small amount of 40keV N^+ ions can penetrate through kindney bean slices with the thickness of at least 60 μm and produce damage on HOPG. The depth is smaller than the maximal depth (about 100 μm) of implanted ion's concentration detected by PIXE and SIMS. More experiments are still in need to clarify whether the difference is due to the diffuse effect or other reasons such as the difference of sample compositions or detection sensitivity. Anyway, all the experiments show that the penetration depth of low-energy ions in botanic samples is much larger than their theoretical range in condensed materials. To explain the phenomenon, it has been suggested that the inhomogenous mass distribution of samples and the ion irradiation damage on the samples must be taken into consideration[7]. At some very thin regions, even micro-holes may exist in the botanic samples before or after irradiation. They can be the open paths for low-energy ions to penetrate through thick botanical samples with large depth. If there are some micro-holes with area larger than 1 μm^2 in the target samples, there will be some amorphous micro-regions on their HOPG detecting pieces due to the overlap of damage. However, such amorphous micro-region on the HOPG surface has not been observed with STM in our experiment. Maybe more images are needed to fit the statistical requirement to get the overall distribution of damage on HOPG surfaces, since the total scanning area was still a very small part of the irradiation area on HOPG detecting pieces. More experiments with other methods will be carried out to clarify the existence of micro-holes in the botanic samples.

5 CONCLUSIONS

In summary, with STM observation, flat HOPG piece can be used as a sensitive counter for low-energy heavy ions. In some energy range, each ion impact can produce one and only one protrusion-like damage. The protrusions can only be produced by impact of energetic ions. The characteristic of HOPG surface make it suitable to detect penetration depth of low-energy heavy ions in botanic samples with transmission measurement. The background can be as low as 1.0×10^9protrusions/cm^2, when each HOPG piece is enveloped so that no scattering ions can bombard it in the irradiation. With the high sensitivity of the method, penetration depth of at least 60 μm has been detected in kidney bean slices.

Acknowledgements

We would like to express our thanks to professor ZHAO Shou-Guang for the helpful discussion and professors Sheng Fang and Yi Li for their technical assistance.

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