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Positron Annihilation Study of Irradiated HOPG-type Graphite

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Positron annihilation angular correlation measurements (ACAR : angular correlation of annihilation radiation) were made for HOPG (highly oriented pyrolytic graphite) and PGCCL (HOPG-type graphite, Le Carbone-Lorraine) irradiated by various particles, such as electrons, neutrons and deuterium particles. The ACAR measurements made under the condition of P_z // c-axis for unirradiated specimens showed a clear minimum at $\vartheta = 0°$, but $P_z \perp$ c-axis measurement did not show this minimum. This minimum is considered to be due to the annihilation of positrons with π electrons the momentum distribution of which is extended to the direction parallel to c-axis. On the other hand, by neutron irradiation this minimum disappeared, which suggests that positrons are trapped at radiation-induced defects, probably vacancy sites, and the probability of the annihilation with π electrons was decreased to the large extent. But, in the case of hydrogen irradiation the minimum was not affected so much, which suggests hydrogen atoms do not disturb the π electrons, probably because hydrogen atoms are trapped at boundary regions, such as those between adjacent crystallites or region of stacking disorder.

KEYWORDS : positron annihilation, angular correlation, HOPG, neutron irradiation, electron irradiation, hydrogen irradiation

1. Introduction

Recently, graphite has become one of the important fusion reactor materials which must face to plasma, because of some reasons, such as low atomic number (so-called low Z) , good high temperature characteristics and so on[1)] . Of course graphite has some negative properties, e.g., absorption and desorption of hydrogen atoms (plasma particles) , which definitely affects the plasma confinement. To obtain the better control of the whole system of plasma and wall, the detailed investigation of the behaviour of hydrogen atoms in graphite must be elucidated. Therefore, the information on the atomistic structure in graphite which provides trapping sites for hydrogen atoms is required. The positron annihilation technique has become a powerful tool to study the defects in solids[2,3]. In this paper, results of the positron annihilation angular correlation measurements (ACAR : angular correlation of annihilation radiation) for HOPG (highly oriented pyrolitic graphite) - type graphite will be presented.

2. Experimental

Positron annihilation angular correlation (ACAR) measurements were performed by using Cu^{64} (~ 1 Ci) positron source at room temperature. The electron irradiation was carried out by using the high energy electron LINAC in KURRI at 77K. Neutron irradiation was performed in JMTR in JAERI-Oarai. The deuterium irradiation was carried out under the condition of 8 keV D_2^+, R.T., 3.46 x 10^{18} D_2^+/cm^2. The reason for using deuterium beam instead of hydrogen beam is to avoid the hydrogen contamination from the environment. Also, the annealing in H_2 gas was performed at 700 ℃ for 5 hrs to supply hydrogen atoms into HOPG.

3. Results and discussion

Figs. 1 and 2 show the ACAR curves of the unirradiated HOPG (UCC, ZYA) specimen obtained at room temperature under the condition of P_z // c-axis and $P_z \perp$ c-axis, respectively. In the former there is a clear minimum at $\vartheta = 0°$, but no such minimum in the latter. This is considered to be due to positron annihilation with π electrons, wave functions of which are extended along the c-axis, i.e., parallel to the normal direction of the basal atomic planes in the case of P_z // c-axis, but in the case of $P_z \perp$ c-axis the minimum did not appear at $\vartheta = 0°$,because the momentum distribution has a maximum at $\vartheta = 0°$.

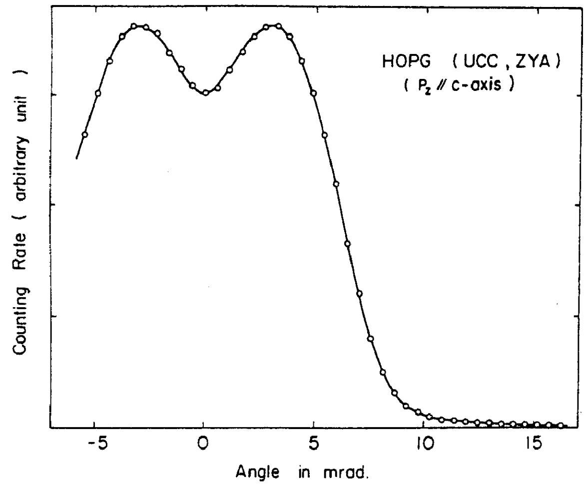

Fig. 1 ACAR curve of HOPG (UCC, ZYA) obtained under the condition of P_z // c-axis.

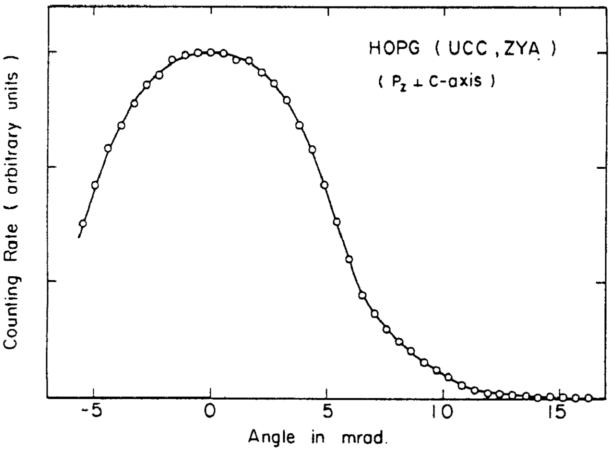

Fig. 2 ACAR curve of HOPG (UCC, ZYA) obtained under the condition of $P_z \perp$ c-axis.

Fig. 3 shows the ACAR curve of the HOPG (UCC, ZYA) irradiated with neutrons in JMTR to the fluence of 1 x 10^{19} n/cm^2 at 150℃, obtained under the condition of P_z // c-axis. The clear minimum at $\vartheta = 0°$ seen in Fig. 1 almost disappears here, which suggests that positrons are trapped and annihilated at radiation-induced defects, mainly at vacancy sites where π electrons are missing[5].

Figs. 4 and 5 show the ACAR curves of the HOPG (UCC, ZYA) annealed in high-purity H_2 gas at 700 ℃ for 5 hrs, and irradiated by 8 keV D_2^+ at R.T. to the fluence of 3.46 x 10^{18} D_2^+/cm^2, respectively. In both cases the minimum at

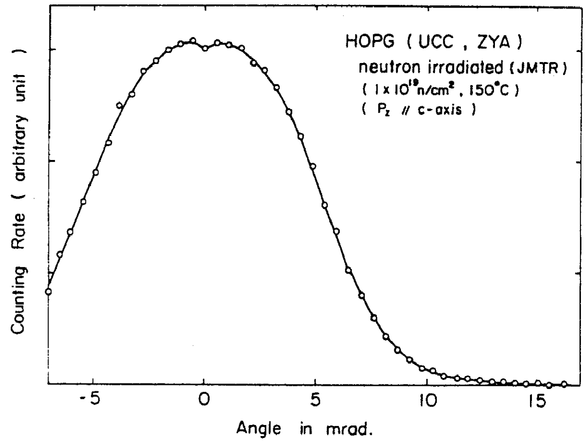

Fig. 3 ACAR curve of the HOPG (UCC, ZYA) irradiated with neutrons in JMTR to the fluence of 1 x 10^{19} n/cm^2 at 150℃, obtained under the condition of P_z // c-axis.

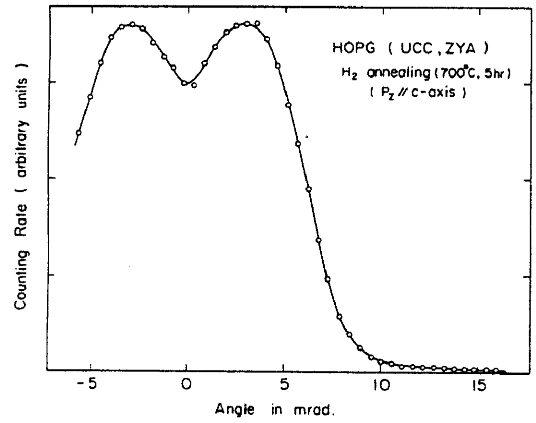

Fig. 4 ACAR curve of the HOPG (UCC, ZYA) annealed in high-purity H_2 gas at 700 ℃ for 5 hrs, obtained under the condition of P_z // c-axis.

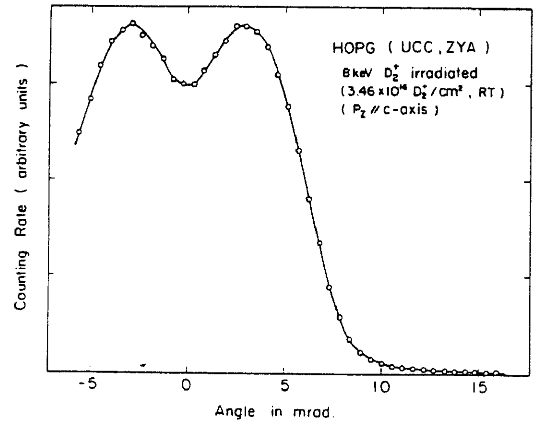

Fig. 5 ACAR curve of the HOPG (UCC, ZYA) irradiated by 8 keV D_2^+ at R.T. to the fluence of 3.46 x 10^{18} D_2^+/cm^2, obtained under P_z // c-axis.

$\vartheta = 0°$ clearly exists as well as the unirradiated one. If hydrogen atoms exist in the matrix of HOPG, probably forming some chemical bondings, they must affect the π electrons and consequently ACAR curve must be changed. But no such change is seen, which suggests that hydrogen atoms are not in the matrix, but in the boundary regions, such as those between adjacent crystallites or region of stacking disorder. In the case of deuterium irradiation the existing range of deuterium from the surface of HOPG is ~ 100nm, then deuterium atoms existing only in the surface region are not able to affect the whole ACAR curve.

Fig. 6 shows ACAR curve of PGCCL (HOPG-type graphite, Le Carbone-Lorraine) , obtained under the condition of P_z // c-axis, which is almost the same as that in Fig. 1, but the dip at $\vartheta = 0°$ is slightly shallow. This corresponds to the fact that positron annihilation lifetime for PGCCL is longer (225.9psec) than that for HOPG (210.5psec) obtained in the previous work[3,4] . Namely, the crystal perfectness of PGCCL is lower than that of HOPG. The electron irradiation introduces vacancies into PGCCL, and the ACAR curve was changed as shown in Fig. 7, where the dip at $\vartheta = 0°$ becomes very shallow, because of the same reason as that in neutron irradiation, that is, positrons are trapped at vacancy sites where π electrons are missing.

Figs. 8 and 9 show ACAR curves of PGCCL irradiated by high energy carbon beam and hydrogen beam, obtained under the condition of P_z // c-axis, respectively. ACAR curves are not changed so much. In both cases, ranges are long enough (carbon -- 61μm, hydrogen -- 1.38mm) , but damage level is not so high enough to trap positrons (less than 0.001dpa in flat region) .

To understand the change of ACAR curve more in detail the calculation of the deuterium state in matrix and in a vacancy is definitely necessary and this is a near future work.

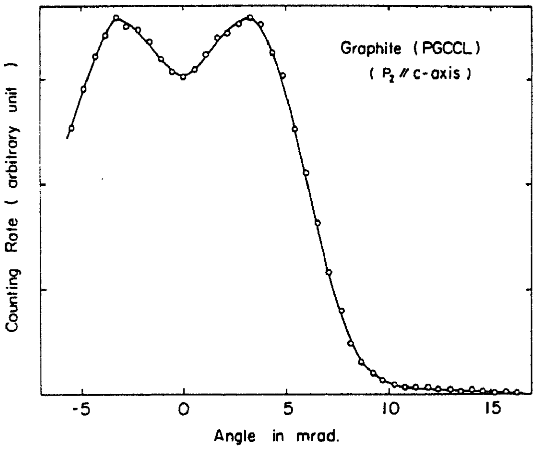

Fig. 6 ACAR curve of PGCCL (HOPG-type graphite, Le Carbone-Lorraine) obtained under the condition of P_z // c-axis

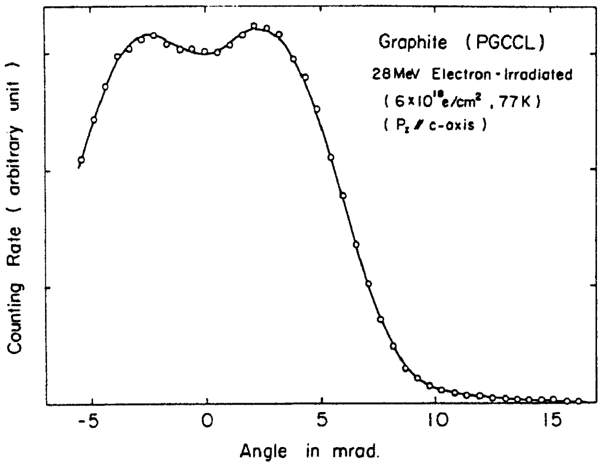

Fig. 7 ACAR curve of PGCCL (HOPG-type graphite, Le Carbone-Lorraine) irradiated by 28MeV electrons at 77K to the fluence of $6\text{x}10^{18}$ e/cm^2, obtained under the condition of P_z // c-axis

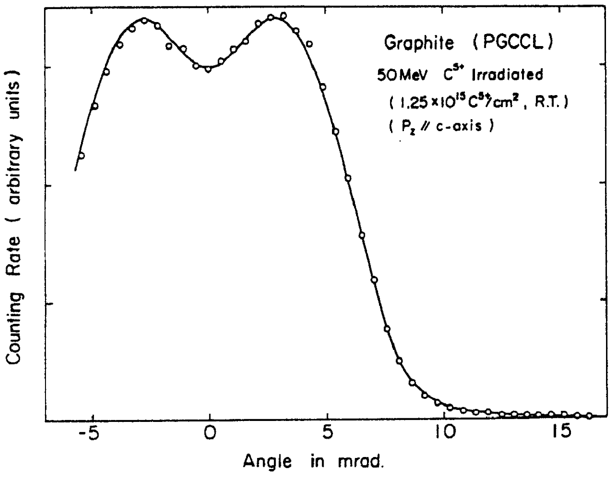

Fig. 8 ACAR curve of PGCCL (HOPG-type graphite, Le Carbone-Lorraine) irradiated by 55MeV carbon beam at R. T. to the fluence of $1.25\text{x}10^{15}$ C^{5+}/cm^2, obtained under the condition of P_z // c-axis

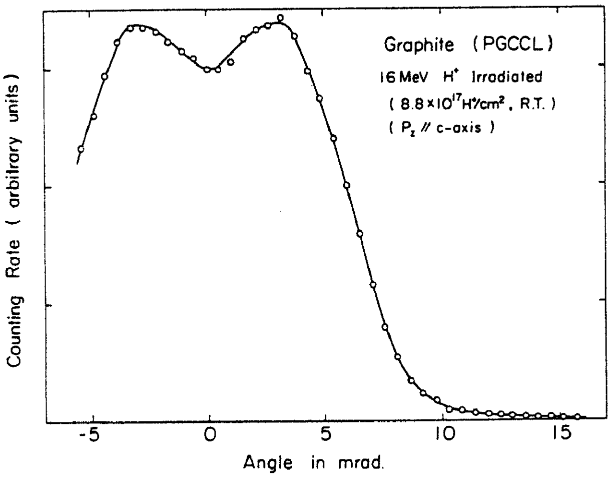

Fig. 9 ACAR curve of PGCCL (HOPG-type graphite, Le Carbone-Lorraine) irradiated by 16MeV hydrogen beam at R. T. to the fluence of 8.8 $\text{x}10^{17}$ H^+/cm^2, obtained under the condition of P_z // c-axis

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