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Neutron Irradiation Effects of Iron Alloys and Ceramics*

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Synopsis

Positron annihilation angular correlation measurements have been performed for the neutron irradiated various metals and ceramics in order to obtain the information of the microvoids and positronium formation in them. Positronium (Ps) formation was observed in Nb containing a small amount of oxygen and Fe-15%Cr-16%Ni-0.006%B^{10}. In practical steels such as JPCA and JFMS no Ps formation was observed. High temperature deformation might induce microvoids into metals, but the positron annihilation angular correlation measurements could not confirm this. In non-metallic materials neutron irradiated no Ps formation has so far been observed.

I. Introduction

It is useful to investigate the microstructure changes of the various materials used under the irradiation environments by using positron annihilation techniques. This technique is useful to study vacancy type defects in materials. It is known that high temperature irradiation induces voids into materials. It is usually very difficult to study the nucleation stage of these voids, namely, microvoids stage. On the other hand, positron annihilation technique is very sensitive to the existence of these small size vacancy clusters. It is also an object of research how positrons behave in materials, such as in voids. It has recently been clarified that positrons form positronium (Ps), hydrogen like state with an electron in voids in materials. It must be studied more under what conditions this Ps can be formed in voids.

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II. Metals and Alloys

It is known that positrons are trapped in vacancy sites and microvoid sites in metals and positron lifetime usually increases and FWHM of the angular correlation curve decreases. For example, in Fe lifetime increases from 108 psec in matrix to 175 psec in vacancy sites and the FWHM decreases from 12.5 mrad in matrix to about 10 mrad in vacancy sites. In the case of microvoids this change becomes larger. These situation is, however, completely changed if Ps forms in voids. Unfortunately, in pure Fe no Ps formation has so far been observed.

II-1. Niobium

In niobium which contains about 400 ppm oxygen atoms and was neutron irradiated in JMTR a prominent positronium formation, i.e., a very narrow angular correlation curve was observed (1). As shown in Fig. 1 FWHM extremely decreased down to 2.5 mrad. Positron lifetime about 500 psec was observed, and this can be understood as the mean value obtained by frequent spin exchange of positronium, namely, Ps state changes from para-Ps (antiparallel spin, singlet, 125 psec) to ortho-Ps (parallel spin, triplet, 140 nsec) and vice versa at the collision to the void wall.

II-2. Iron-Alloys

Among iron-alloys positronum formation was observed in Fe-Cr-Ni alloy system. In ternary alloy Fe-Cr-Ni Ps formation is not so prominent, but in Fe-15%Cr-16%Ni-0.006%B^{10} alloy irradiated by JMTR a very prominent Ps formation was observed as shown in Fig. 2, especially after annealing. This can be considered to be due to the diffusion and segregation of Li atoms on the void surface, where Li atoms are created by the nuclear reaction $B^{10}(n,\alpha)Li^{7}$ (2). For Ps formation it is favorite that work function for an electron at the surface of metals is reduced by the segregation of other elements. It is known that alkali metals have this effect. It is reported that in proton irradiated Al positronium is observed in voids coated by Na which was formed by nuclear reaction (3).

II-3. Aluminium

Contrary to the expect no Ps formation was observed in Al-1%Li alloy neutron irradiated by JMTR as shown in Fig. 3. It is generally recognized that void formation is not so remarkable in aluminium, and this may be one of the reasons of less Ps formation of this metal and alloys. Even if microvoids exist in the matrix and positronium are formed in them, positron might be annihilated with other electrons than paired one, i.e., pick-off process, then resulting in disappearance of narrow angular correlation curve.

II-4. Vacancy migration temperature

Information on the vacancy migration temperature has been obtained from the positron annihilation lifetime measurements of low temperature irradiated metals and alloys. Neutron irradiation induces Frenkel pairs to materials in heterogeneous fashion, i.e., in so-called cascades formation, and this is not favorite to determine the vacancy migration temperature. Then, for this purpose the electron irradiation was performed, where defects are uniformly generated in the matrix. It was clarified that in pure iron vacancies start to migrate and form microvoids at about 220 K. This temperature is not so affected by alloying of Cr, if both Fe and Cr are fully purified in hydrogen gas. This is because size factor difference between Fe and Cr is rather small, and then trapping efficiency of Cr for vacancies is not so strong.

II-5. Vacancies generated by deformation

It is generally recognized that deformation introduces vacancies into the matrix by cutting process of dislocations. It is recently reported that in compound semiconductors, such as InP, GaAs high temperature deformation introduces microvoids which are detected by positron annihilation lifetime measurement. One of the present authors (Kuramoto) previously reported the result of lifetime measurement for the deformed GaAs and concluded that positrons were trapped at dislocation sites (4). This is quite contrary to the recent interpretation. In order to study if the same behavior is observed or not in metals, practical steels such as JPCA and JFMS were deformed at 500 C and angular correlation measurements were performed for these alloys. However, as shown in Fig. 4 no void formation (no Ps formation) was observed. Attempts will be continued in different deformation temperature and amount of deformation, but discuss on the different feature of microvoid stability and so on between semiconductors and metals may also be needed.

III. Non-metals

As well as metals and alloys investigations on non-metallic materials have been performed by using positron annihilation techniques.

III-1. Graphites

In Fig. 5 is shown the positron annihilation angular correlatioin curve of the highly oriented pyrographite (HOPG) obtained under the condition c-axis // P_z. Two side peaks come from -electrons in the basal plane. This is confirmed from the fact that side peaks are not observed in the curve obtained under the condition c-axis P_z. It is also recognized that neutron or electron irradiation to this material makes side peaks disappear, because positrons are trapped at vacancy sites and annihilate there. It is reasonable that in isotropic graphites no side peaks were observed. The FWHM has a maximum value in HOPG, and deceases in isotropic garphites and glassy carbon in this order (13 mrad - 9 mrad). On the contrary, positron lifetime increases in this order (210 psec - 420 psec). Results on the various graphites are shown in table 1. Positron annihilation lifetime in neutron irradiated isotropic graphites is about 280 psec. The positron annihilation lifetime in the electron irrdiated HOPG is about 245 psec. This difference seems to be due to the cascade formation in neutron irradiation, where some amount of small vacancy clusters are spontaneously formed, which gives longer lifetime component. Positron annihilation lifetime in the isotropic graphites are 320 psec - 370 psec, but it is not easy to determine from which site this value comes (5). Recently, a new feature has come out from the lifetime measurement of HOPG which was heat treated at rather low temperature 2800 C. In this case the lifetime was about 300 psec, which is much longer than usual HOPG. It is clear that this longer lifetime comes from the interfaces between small crystallites in the grains. It is then reasonable to interpret the lifetime obtained for the isotropic graphites as a mixed value of that at interfaces and that at grain boundaries (or pores). It can explain that isotropic graphites with higher graphitization show larger positron annihilation lifetime.

III-2. Oxides

Many measurements have so far been performed for various oxides. Sintered Al_2O_3 showed positron lifetime of 125 psec, but ordinary single crystal of this material showed 155 psec. Single crystals of higher grade (higher purity), however, showed shorter values of lifetime which comes down to that in sintered ones. On the other hand, sintered crystals irradiated by high energy oxygen ions gave the value of 155 psec. These results suggest that in single crystals of normal grade oxygen vacancies already exist. Since it is reported that at F-center sites in ionic crystals positrons form Ps, the angular correlation measurements were performed. But, no narrow component, i.e., no Ps formation was observed (6). Annealing in oxygen atmosphere is needed to change the trapping of positrons at oxygen vacancies.

No narrow component in the angular correlation curve was observed in ZnO single crystals. For this material the positron lifetime at matrix has not been established. But, recently, quite large single crystal which is not colored showed 185 psec, and this might be the value at matrix, because 'uncolored' means almost no excess Zn and almost no oxygen vacancies. For sintered materials about 210 psec has been obtained, and this might be the value at grain boundaries or pores. Single crystals of yellow color showed lifetime 185 psec - 195 psec. When this sample is annealed in vacuum to higher temperatures this value decreased to 160 psec. But, this sample has a high tendency of sublimation, so some structural change must have occurred in the matrix.

III-3. High temperature superconductor

Some measurements have been made also for high temperature superconductor $YBa_2Cu_3O_{7-x}$. Positron lifetime of this material is 175 psec, but it increased to 195 psec after it remained in air for several months. This is considered to be due to some structural change which was caused by invaded humidity from air, e.g., formation of $Ba(OH)_2$. High energy He ion irradiation induced longer lifetime component 235 psec, which was considered to be due to complex of Cu vacancy and O vacancy. No positronium formation was observed in the angular correlation measurement (7).

III-4. Carbides

Positron annihilation measurement was performed for TiC_{1-x}, (Ti,Mo)C crystals. Since these materials have plenty of structural vacancies due to non-stoichiometry, it is difficult to judge whether the lifetime 150 psec obtained is at matrix or vacancy site. Neutron irradiation did not change the lifetime value, which means this lifetime might correspond to that at vacancy sites. Annealing experiment will be necessary. No positronium formation was observed in the angular correlation curve.

IV. Subjects in Future

No positronium formation has so far been observed in stainless steels, but in samples irradiated to higher dose, such as in JOYO 10 dpa Ps formation can be expected. This will soon be made after radiation decreases to a certain level. There will be also possibility of Ps formation in voids formed by high temperature deformation. This experiment will be continued for various metals and alloys. As for non-metallic materials positron annihilation experiment must be continued to study lattice defects.

Acknowledgement

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Table 1. Results of positron annihilation measurements for various graphites

	FWHM (mrad)	Lifetime (psec)
HOPG (PGCCL)(P_z//c)	13.2	225.9
HOPG (UCC)	10.9	210.5
HOPG (Nippon Carbon)	10.7	
B-doped (10%, //) (Toyo Tanso)	10.5	271.4
B-doped (10%, ⊥) (Toyo Tanso)	10.5	289.5
UHP (Tokai Carbon)	10.3	340.5
IG430U (Toyo Tanso)	10.2	343.1
IG110U (Toyo Tanso) (1100°C,1hr degassed)	10.2	364.5
IG110U (Toyo Tanso, n-irr.)	9.8	263.2
E252G (Toyo Tanso)	9.9	326.3
Glassy Carbon (GC-L ,Tokai Carbon)	9.6	
Glassy Carbon (GC-30,Tokai Carbon)	9.3	434.3
Glassy Carbon (SG-2G, Showa Denko)	9.5	408.9

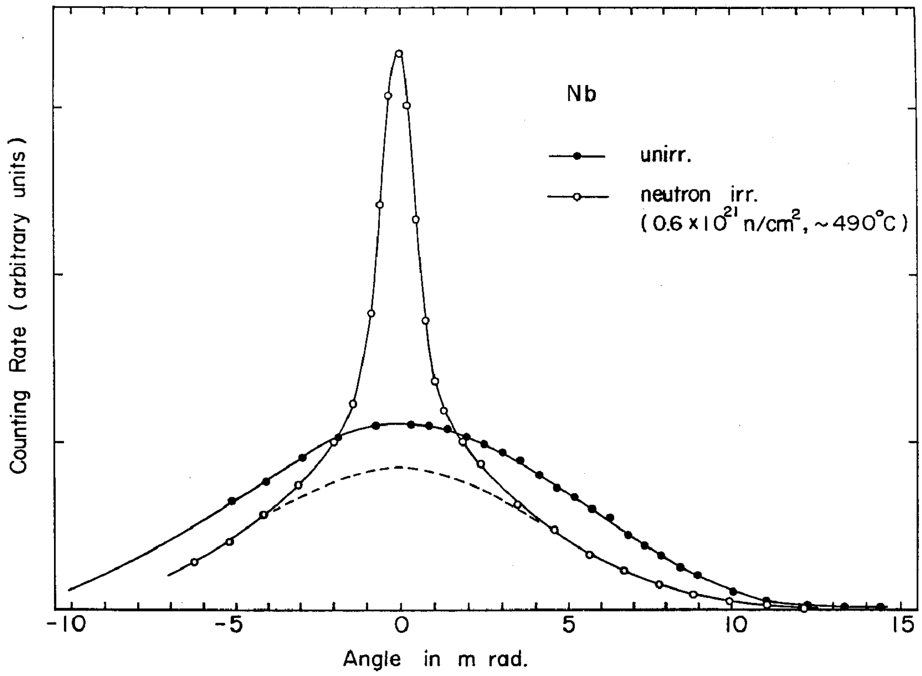

Fig. 1 Positron annihilation angular correlation curve of the neutron irradiated Nb containing small amount of oxygen atoms.

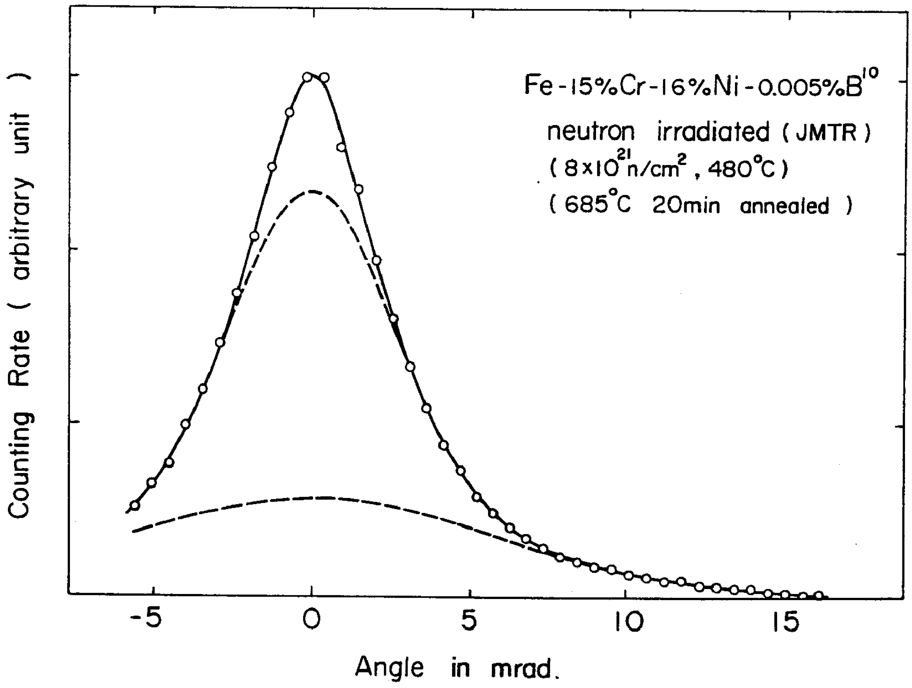

Fig.2 Positron annihilation angular correlation curve of the neutron irradiated Fe-15%Cr-16%Ni-0.005%B^{10}

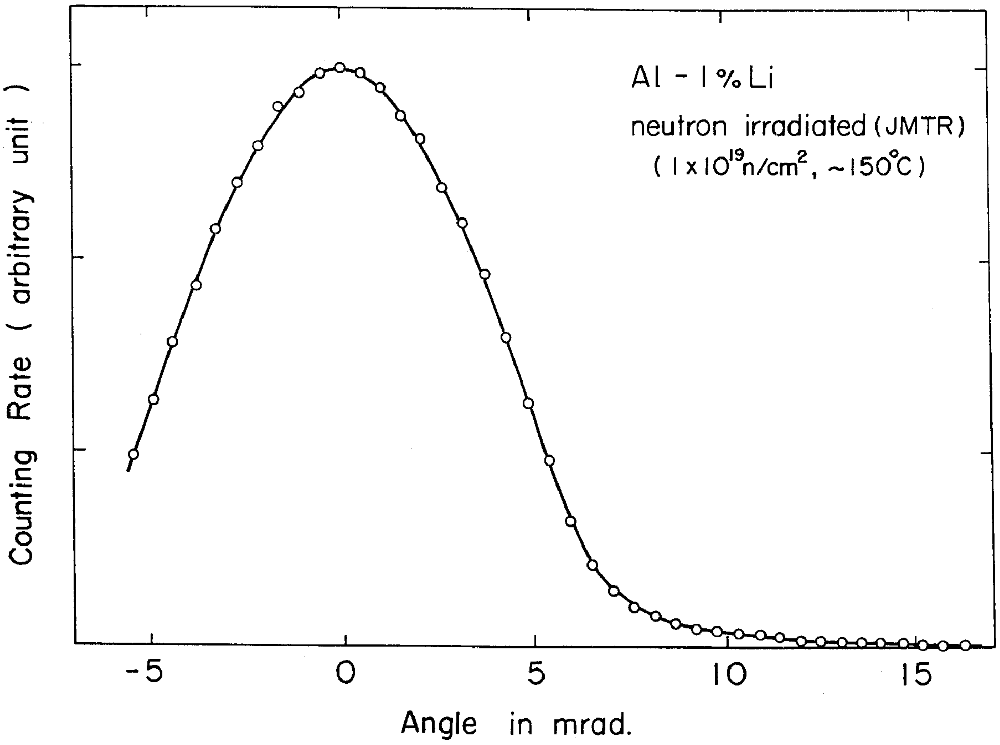

Fig. 3 Positron annihilation angular correlation curve of neutron irradiated Al-1%li.

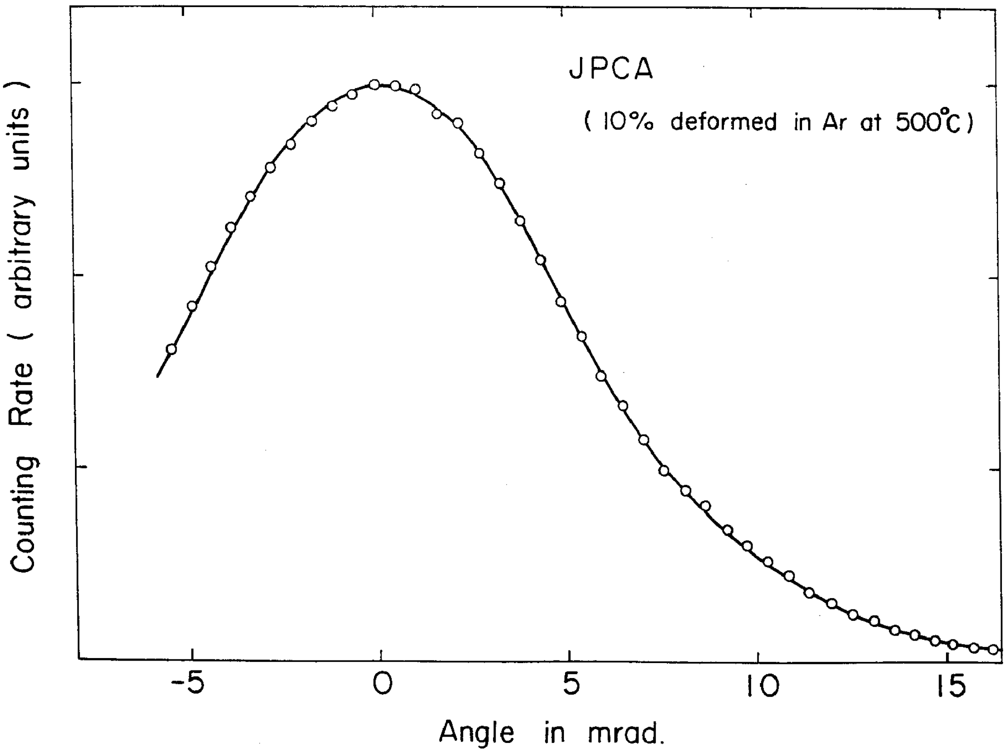

Fig. 4 Positron annihilation angular correlation curve of JPCA deformed at 500 ℃.

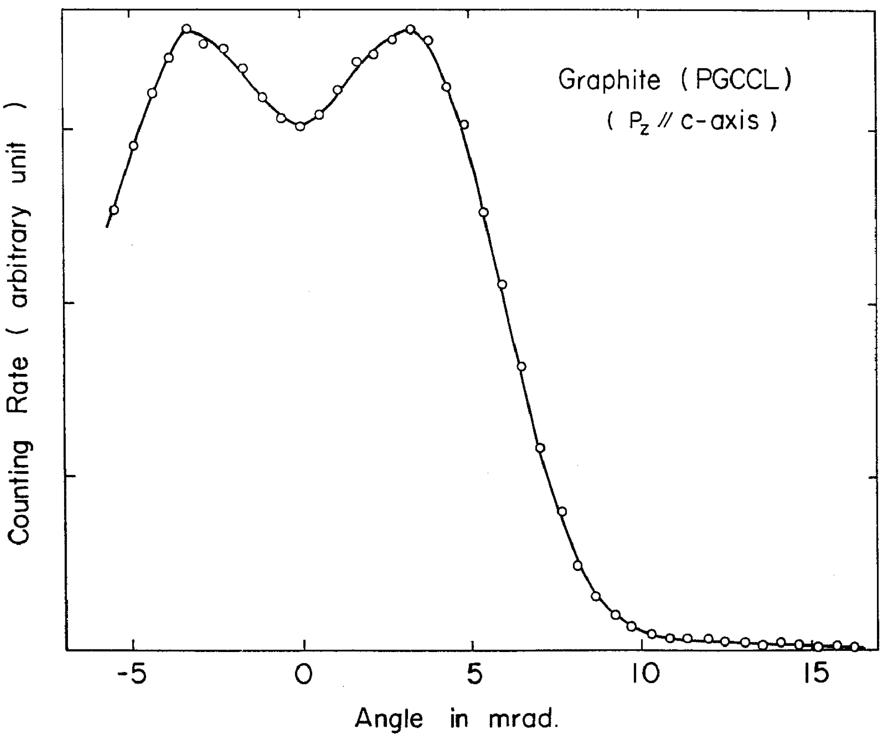

Fig. 5 Positron annihilation angular correlation curve of HOPG (c-axis // P_z).