TDPAC Measurements of He-decolated Vacancies in 111In-implanted Cu and Stainless Steels

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The presence of He in a metal has a strong influence on its mechanical properties. Studies of helium embrittlement are important in connection with developing suitable materials for controlled thermonuclear reactors (CTR). Helium atoms will be generated as (n,α) transmutation products within the first wall and blanket structures of CTR and are insoluble in most materials; therefore, it has a tendency to diffuse and precipitate within materials. There are few techniques to study the behaviour of He diffusion and precipitation in a microscopic way. Recently, by the Groningen group, however, striking effects have been found in the TDPAC spectra of sources of ^{111}In implanted in Ni and Cu post-implanted with He. It has been concluded that He-decolated vacancies are formed with almost 100 % capturing efficiency at the ^{111}In impurities.[1,2)]

In this report, we show that TDPAC, through the formation of ^{111}In-He complexes, can be a new technique in the study of He behaviour in metals, and some experimental results are given as an example.

Sources were prepared on polycrystalline metal foils of Marz grade by implantation with 2.8 d ^{111}In activity with a dose of about 1×10^{12} ions at an implantation energy of 42 keV at room temperature. This source-preparation procedure was reported previously.[3)] In the present experiment the method has been refined and the ^{111}In-implantation efficiency reached 3-5 %. In every case, the source sample was prepared with and without a pre-implantation He-dose of 5×10^{16} to 1×10^{17} ions/cm^2. The He implantation energies were 5 to 40 keV.

In the TDPAC measurements we used a standard fast-slow coincidence spectrometer with three $1\frac{1}{2}$"$\phi\times1\frac{1}{2}$" NaI(Tl) detectors to detect the 171-245 keV γ-γ cascade in ^{111}Cd (daughter of ^{111}In). The resolving time was 1.8 ns (FWHM). Two coincidence spectra, with counter angles of 90° and 180°, were combined to obtain the perturbation function

$$A_2G_2(t) = R(t) = 2\frac{N(t,180°)-N(t,90°)}{N(t,180°)+2N(t,90°)} . \qquad (1)$$

For a randomly directed axial electric field gradient, the perturbation function for nuclear spin I = 5/2 has the time dependence[4)]

$$G_2(t) = \sum_{n=0}^{n=3} \exp(-\tfrac{1}{2}(n\sigma t)^2)s_{2n}\cos(n\omega_0 t) , \qquad (2)$$

where $\omega_0 = 2\pi/t_0 = (3\pi/10)\nu_Q$ and $\nu_Q = eQV_{zz}/\hbar$. Here t_0 is a period of the modulation of the R(t) curve and Q is the quadrupole moment of the $5/2^+$ state of ^{111}Cd, V_{zz} is the z-component of the electric field gradient at the nuclear site, σ is a frequency distribution around ω_0 (Gaussian type) and s_{2n} is the attenuation coefficients tabulated in ref. (4).

Figure 1 is a typical TDPAC spectrum for Cu pre-implanted with He at 5 keV measured at room temperature after 1 h of annealing at 873 K. The solid line in Fig. 1 is the curve of eq. (2) determined with a least-squares method. This spectrum can be considered as a signal of the He-decolated vacancy which consists of He atoms, vacancies and a ^{111}In atom, because the frequency of ν_Q = 217 MHz obtained from the present experiment agrees with the value of 229 MHz previously reported for He-decolated vacancies of Cu.[1)]

Figure 2 is the TDPAC spectra measured with polycrystalline stainless steel foils (Good Fellow Metal Co. AISI#316) at room temperature after steps of 1 h of isochronal annealing at 300, 873 and 973 K with and without He-pre-implantation. The energy and dose of He pre-implantation were 10 keV and 5×10^{16} ions/cm^2, respectively. The modulation of the R(t) curve did not appear until the thermal annealing at 873 K. At 973 K this signal became dominant. In comparison with the spectra observed without He-pre-implantation, in which only a low-frequency (13 MHz) signal was seen even after the high-temperature annealing, and with the He reemission data of SUS#316, we interpret this modulation as the eqQ signals with a fast damping which was induced by the formation of He-decolated vacancies.

Figure 3 shows a typical TDPAC spectrum for #316 stainless steel measured after 1 h of annealing at 973 K with He pre-implantation dose of 1×10^{17} ions/cm^2. Fitting the R(t) spectrum to eq. (2) with a least-squares method, we obtained the following results; the central frequency ν_Q = 247 MHz and the width of the Gaussian type distribution $\delta = \sigma/\omega_0$ = 13 %. Moreover, we made similar experiments with other alloys, and obtained for SUS#304 (Good Fellow Metal Co. AISI#304) ν_Q = 245 MHz and δ = 28 %, and for Inconel (Ni72/Cr16/Fe8) ν_Q = 247 MHz and δ = 8 %.

The present observations of the ^{111}In-He complexes by TDPAC for a variety of stainless steels are very significant, because these materials are candidate structural materials for CTR and their He behaviour can be studied by this method microscopically. Through all the measurements, the central frequency of modulation has similar values. This fact confirms that the He-decolated vacancies in Cu and stainless steels have a common geometrical structure of vacancy complex. In addition to such an information, the activation energy E_A for He migration in SUS#316 can be determined by applying a simple diffusion equation which is the function of annealing time, annealing temperature and activation energy E_A.[6)]

Figure 4 shows the changes of the eqQ signals corresponding to isochronal annealing (from 973 to 1073 K for 10 min) and to isothermal annealing (at 973 K from 10 to 60 min). The analysis of these data is now in progress.

References

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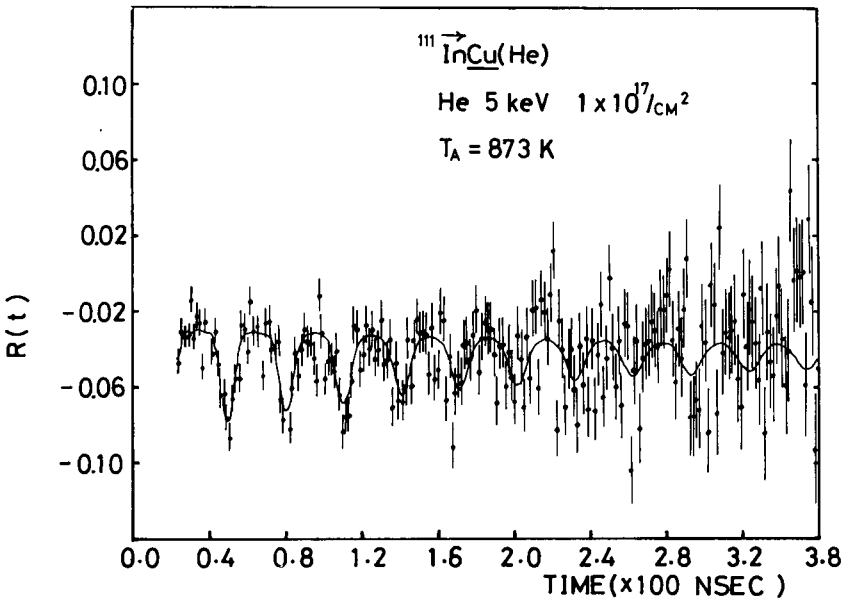

Fig. 1. A TDPAC spectrum of Cu pre-implanted with He ions at 5 keV measured at room temperature after 1 h of annealing at 873 K

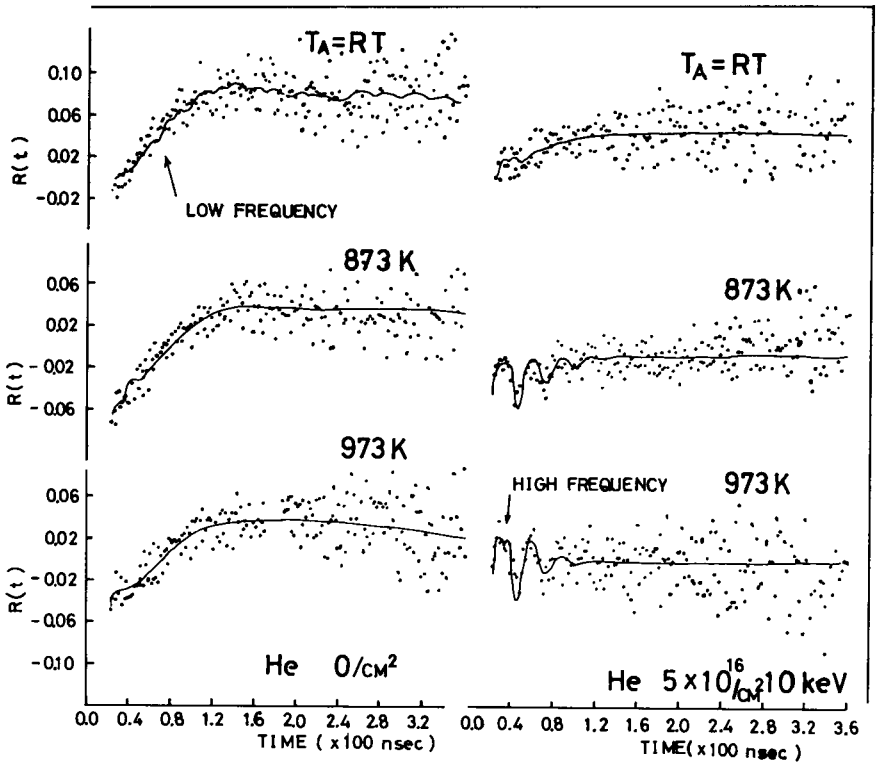

Fig. 2. TDPAC spectra measured for polycrystalline SUS#316 foils at room temperature after 1 h steps of isochronal annealing at 300, 873 and 973 K with and without He-pre-implantation. The energy and dose of He pre-implantation were 10 keV and 5×10^{16} ions/cm^2, respectively.

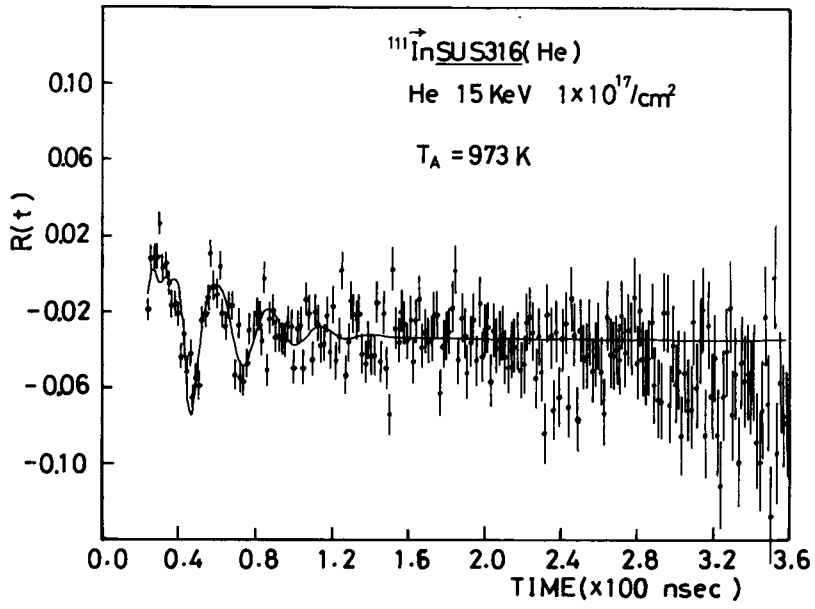

Fig. 3. A TDPAC spectrum for SUS#316 measured after 1 h of annealing at 973 K. The energy and dose of He pre-implantation were 15 keV and 1×10^{17} ions/cm^2, respectively.

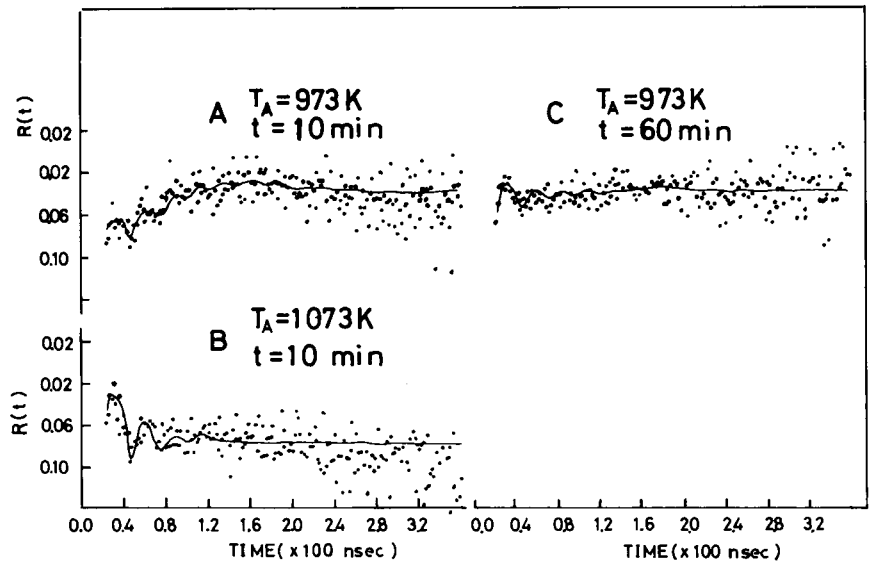

Fig. 4. TDPAC spectra for SUS#316 corresponding to an isochronal annealing from 973 K to 1073 K for 10 min (A→B), and to an isothermal annealing from 10 min to 60 min at 973 K (A→C). He pre-implantation energy and dose were 40 keV and 1×10^{17} ions/cm^2, respectively.