HELIUM BUBBLES IN CYCLOTRON-BOMBARDED METALS AND ALLOYS STUDIED BY POSITRON ANNIHILATION SPECTROSCOPY

G. Amarendra, B. Viswanathan and K.P. Gopinathan+

Indira Gandhi Centre for Atomic Research, Kalpakkam 603 102, India

ABSTRACT

Helium has been injected using α-particles from a cyclotron to a dose of 100 appm in Cu, Al, Ni and Ni-Ti alloys. Positron annihilation studies have been carried out on the irradiated materials to identify the nucleation and growth process for helium bubbles. From an analysis of experimental positron lifetimes, the characteristic bubble parameters have been deduced.

1. INTRODUCTION

Ever since the recognition of material property changes caused by high dose neutron irradiation, simulation of neutron damage by charge particle bombardment at cyclotron has been a promising approach[1]. Light ions of energy 5-40 MeV have ranges as well as damage rates suitable for the study of bulk defect properties in materials. In this paper, the use of positron annihilation spectroscopy for a detailed understanding of the properties of helium bubbles in cyclotron bombarded metals and alloys is discussed on the basis of examples from our recent studies. The extremely low solubility of helium in metals results in precipitation into bubbles which cause degradation of the material properties. Studies on helium-defect interaction, helium diffusion and nucleation and growth of bubbles are therefore of importance both from a fundamental as well as a technological point of view.[2]

Positron annihilation spectroscopy (PAS) is an established powerful technique for a qualitative and quantitative understanding of vacancy-type defects in metals and alloys[3]. Vacancies due to the missing atom cores, act as attractive centres to trap positrons. This results in positron localisation into bound states at vacancies or their clusters . When a vacancy cluster contains helium atoms,there is a sharp increase in electron density at the centre brought about by the core electrons of helium. Consequently, the positron samples a region of relatively higher electron density leading to a reduction of its lifetime,as compared to that in a helium free vacancy cluster. Furthermore, the presence of helium atom cores in the cluster alters measurably the electron momentum distribution around the positron trap. Recent work[4] has shown that positron lifetime is markedly sensitive to the size as well as helium-to-vacancy ratio for small clusters, while for large bubbles there is a strong correlation between the lifetime and helium atom density (He pressure).

2. EXPERIMENTAL

2.1 Helium Irradiation

The material under study was implanted with 40 MeV α-particles at the Variable Energy Cyclotron, Calcutta. Uniform distribution of helium over a depth of 200 μm from the sample surface was achieved by continuously degrading the alpha beam energy from 40 MeV to 1 MeV. The irradiation arrangement coupled to the beam line, along with the target assembly is shown in fig.1. The present arrangement enabled strong overlap of the helium profiles with the profile of the positron probe (fig.1c), thereby increasing the strength of annihilation signals from helium sites. During alpha bombardments, helium dose, irradiation temperature and beam profiles were continuously monitored.

2.2 Positron Experiments

Positron lifetime spectra were measured with a conventional fast-slow coincidence spectrometer with a time resoltuion of 300 ps (FWHM). In the study on Ni and Ni-Ti alloys, a higher resolution spectrometer with FWHM = 200 ps was used. The measured lifetime spectra have been analysed for the various components using

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+Presently at Hahn-Meitner Institut,Berlin(West)

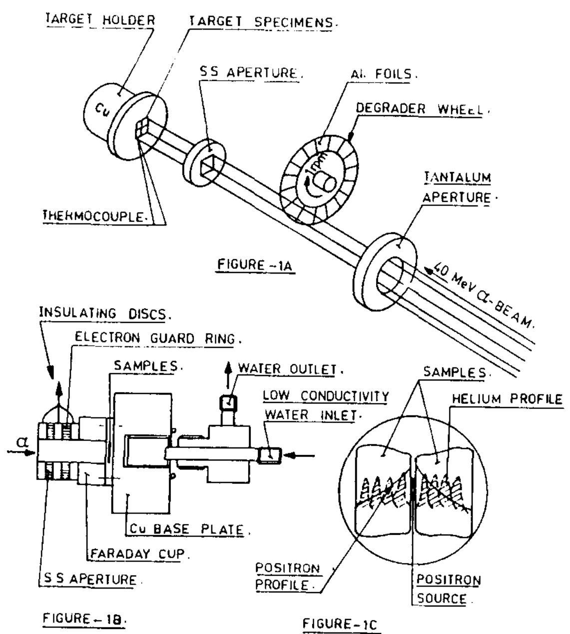

Fig.1 (A) Helium irradiation arrangement, (B) Target assembly, (c) Overlap of the implanted He profiles with the profile of the positron probe.

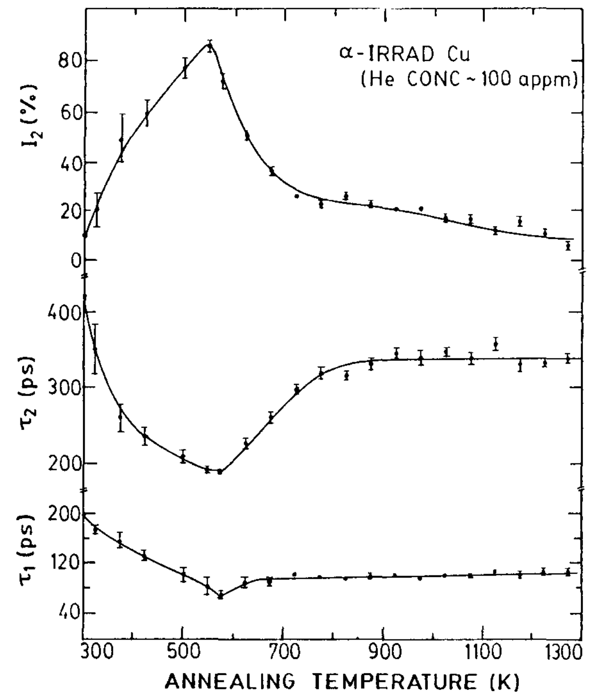

Fig.3 Resolved lifetime parameters vs. annealing temperature for α-irradiated Cu.

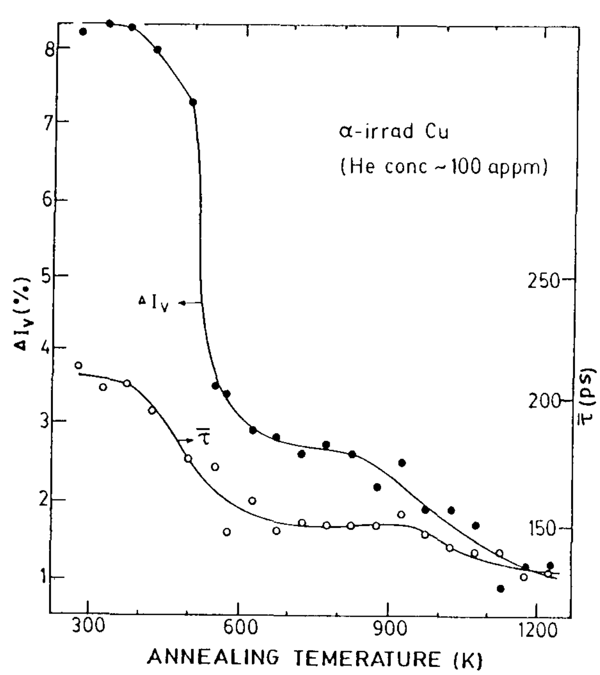

Fig.2 Doppler lineshape parameter and mean lifetime $\bar{\tau}$ vs. annealing temperature for α-irradiated Cu.

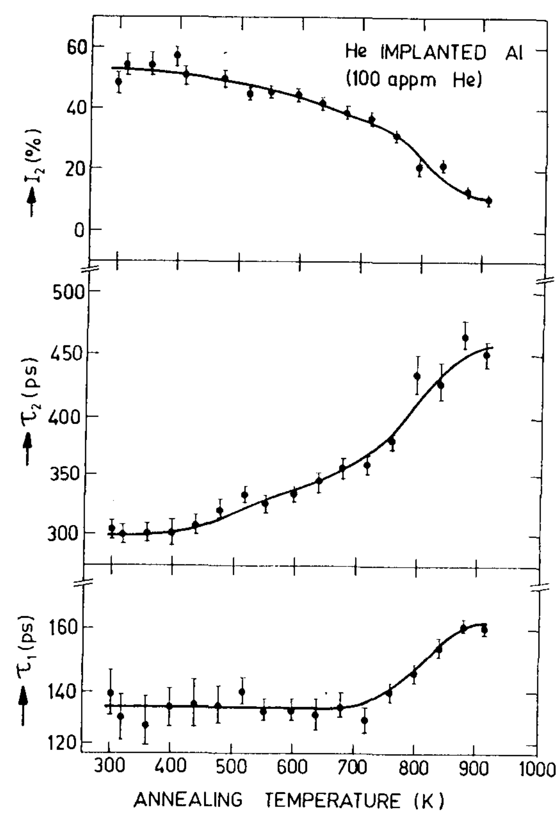

Fig.4 Resolved lifetime parameters vs. annealing temperature for α-irradiated Al.

the programs RESOLUTION and POSITRONFIT. Doppler broadening lineshape measurements were made with a high pure Ge detector with an energy resolution of 1.1 keV at 514 keV γ-ray of ^{85}Sr. A lineshape parameter I_V, which is the integrated counts over a small central segment about the 511 keV peak of the normalised Doppler spectrum was used for defect characterization.

3. RESULTS AND DISCUSSION

3.1 Helium Irradiated Cu

Figure 2 shows the variation of the Doppler lineshape I_V as well as mean lifetime $\bar{\tau}$ as a function of annealing temperature in α-irradiated Cu (He dose 100 appm). Both I_V and $\bar{\tau}$ decrease sharply in the interval 400-600 K, exhibiting a shoulder around 600 K. The latter feature observed in the present study has not been seen either in the case of electron irradiated Cu or n-irradiated Cu. Hence the stage seen around 600 K can be associated with the behaviour of helium. Beyond 800 K, I_V and $\bar{\tau}$ decrease again. The lifetime spectra have been analysed in terms of two components as shown in fig.3. In the as-irradiated state, the unconstrained fit gives τ_1 = 180±10 ps with 92% intensity and τ_2 = 420±30 ps with 8% intensity. The shorter lifetime τ_1 of dominant intensity is explained as due to the combined effects of positron trapping at small He-V complexes and irradiation induced dislocation loops. The larger lifetime τ_2 is understood as due to positron trapping at helium-free voids formed by the agglomeration of mobile vacancies. Between 300 and 570 K, τ_1 decreases towards the bulk value, τ_2 decreases to a minimum and I_2 increases to a maximum. This behaviour of τ_2 and I_2 may be understood in terms of the formation of bubble nucleus, controlled by a thermally activated process of helium migration to vacancy traps. The kinetics of bubble nucleation has also been studied [5] by isothermal annealing measurements on Cu (100 appm He) at different temperatures in the range of 500-600 K. An analysis of the isothermal curves gives an estimate of activation energy as 1.25 eV. This value which is higher than that for interstitial He migration might support the mechanism of trap-hindered migration of He to the nucleating site. Beyond 600 K, the sharp increase of τ_2 towards a saturation value accompanied by a concommittant decrease of I_2 is indicative of post-nucleation growth of He clusters enroute to the formation of stable bubbles. An analysis of the experimental lifetimes on the basis of the positron surface state model [4] of the bubble,gives helium pressure in the bubble as P_τ = 550 MPa at T_A = 1200 K. This value for P_τ compares satisfactorily with the independent estimates of equilibrium pressures for the bubble.

Helium was also produced through the reaction $^{10}B(n,\alpha)^7Li$ in n-irradiated Cu-B alloys [6] with an irradiation time of nearly two months in the reactor. This is to be compared with the direct alpha irradiation time of only 8 hours to produce an equivalent dose of helium in the material. This shows the merit of charge particle simulation of neutron effects using a cyclotron. A comparison of lifetime results in n-irradiated Cu-B and α-irradiated Cu shows that helium bubble growth processes are similar for the two types of irradiation.

3.2 Helium Irradiated Al

Figure 4 shows the variation of the resolved lifetime parameters in α-irradiated Al with 100 appm He. Since the onset of vacancy and helium mobilities in Al is at lower temperature, the nucleation stage of bubbles is not seen in fig.4. However, the behaviour of τ_2 and I_2 is indicative of the post-nucleation growth. Using the relation [4] between helium atom density n_{He} and lifetime τ_2, the deduced n_{He} decreases from 77 nm^{-3} at 500 K to 21 nm^{-3} at 900 K. The bubble concentration decreases from 1.4×10^{22} m^{-3} to 2.8×10^{20} m^{-3} in the same interval. Helium retention in bubbles is found stable in Al even upto the melting point. The present results are in agreement with those in Al irradiated with 600-800 MeV protons [7].

3.3 Helium Irradiated Ni and Ni-Ti Alloys

PAS studies of helium implanted Ni and Ni-Ti alloys containing Ti in the concentration range of 0.5 to 5 at% have been made to understand the effect of Ti alloying on helium bubble growth. The He dose in all samples studied was 100 appm. The lifetime behaviour observed in pure Ni is qualitatively similar to that discussed earlier in copper. Bubble nucleation has been identified by the characteristic reduction of τ_2 and increase of I_2 around 750 K. On the other hand, in all Ni-Ti alloys, only a single lifetime of ~ 150 ps is observed in the as-irradiated state, indicating complete positron trapping at Ti-associated defects. Beyond 700 K, two lifetimes have been resolved in the alloys. The lifetime corresponding to stable bubbles during growth is found to decrease progressively as Ti-concentration is increased and these values in turn are smaller than that for pure Ni. The variation of bubble parameters, namely, radius, concentration and total bubble volume, as deduced from an analysis [8] of the experimental lifetimes are shown in fig.5. A decrease of bubble radius r_B and an increase of bubble density C_B are seen as a function of increasing Ti-concentration. Bubble swelling V_B is found to decrease sharply till 1 at% Ti and levels off at higher concentration. These results in fig.5 may be understood as follows: An oversized substitutional Ti impurity with local electron density different from the host Ni, acts as an efficient trapping site for helium. Hence alloying Ni with Ti increases the possible

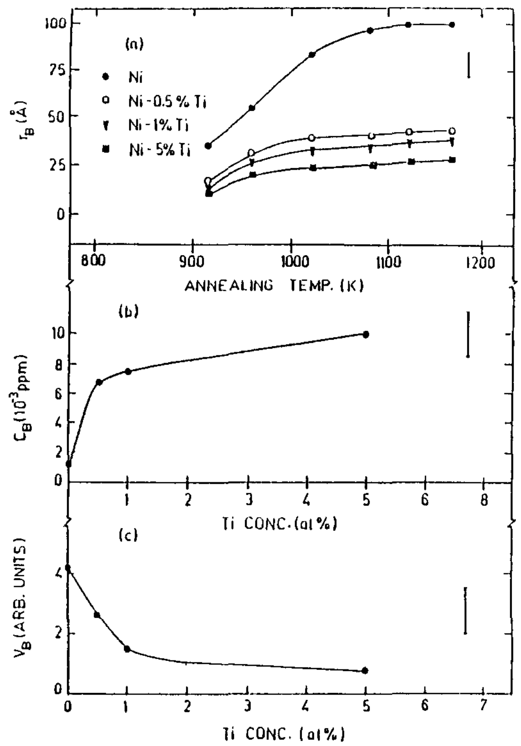

Fig.5 Results of the analysis of experimental positron lifetime in Ni and Ni-Ti alloys (a) Bubble radius r_B vs. annealing temperature, (b) Bubble density C_B vs. Ti-concentration, (c) Total bubble volume V_B vs. Ti-concentration.

nucleation centres for bubble formation with increased helium occupancy.

In summary and conclusion, positron annihilation studies on helium in Cu, Al, Ni and Ni-Ti alloys have shown that detailed understanding of nucleation and growth of bubbles can obtained after ion bombardment at the cyclotron.

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