Annealing of a Pre-Assembled Tungsten Positron Moderator by Direct Electron Bombardment

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We have developed a positron moderator annealing system with a handmade electron gun which allows the pre-assembled moderator temperature to be monitored directly during heating. The moderator, composed of strips of tungsten arranged in a regular lattice, was heated by bombarding electrons emitted from a hot tungsten filament. Moderator annealing was successfully achieved up to a temperature of 2600 °C in the central region of the moderator with a thermionic power of about 1200 W. Positron annihilation lifetime measurements of as-received and annealed tungsten foils were performed using a pulsed slow positron beam. As a result, it was confirmed that residual defects contained in the as-received tungsten foil were eliminated by conducting the annealing process.

1. Introduction

Positron annihilation spectroscopy with a slow positron beam is a powerful technique to detect vacancy-type defects or free-volumes near the surface of a sample. In a slow positron beam system, [22]Na radioisotopes, electron linear accelerators (LINAC), or nuclear reactors are utilized as the positron source. However, the initially obtained positrons have a broad energy distribution (~MeV). Therefore, in order to form a slow positron beam, it is necessary to moderate the positrons. Materials (such as tungsten) that have a negative work function for positron emission are used as a positron moderator [1–3]. Moderators assembled from thin strips of tungsten arranged in a lattice configuration are often used in LINAC- or reactor-based slow positron beam systems since: 1) the moderator assembly has large surface area, 2) the assembly can cover the whole area of the positron converter [4,5]. Such lattice-like structures are used as positron moderators in the AIST LINAC-based slow positron beam system [6–9].

Among positrons implanted into moderator materials, only those which diffuse back to near the surface are reemitted into vacuum [10]. Increasing the positron diffusion length is an important factor to improve the moderation efficiency. On the other hand, vacancy-type defects act as positron-trapping centers, and cause the positron diffusion length to be reduced. Removal of surface contaminations is also an important factor to improve moderator efficiency [10–12]. Thus, an elimination of residual defects and surface contaminations by means of annealing treatment is essential for moderator materials [3, 4, 11–17]. Since tungsten has a high melting temperature ($T_M \sim 3700$ K), an annealing temperature of around 2300 °C (2573 K), which corresponds to $0.7\,T_M$, is desired [18] in order to remove residual defects. Additionally, in order to remove surface contaminations such as oxides, an annealing temperature of around 1500 °C is needed [11].

Although the annealing treatment can be performed easily, annealed tungsten strips are difficult to be assembled in a lattice-like configuration as the strips are brittle. In addition, plastic-deformation-

induced defects will be introduced even if the lattice can be assembled after annealing. Therefore, it is desirable to anneal pre-assembled lattice-like tungsten moderators. However, annealing the pre-assembled lattice-like structures up to such a high temperature by Joule heating is difficult. In order to overcome the problem, we have adopted electron bombardment for the moderator annealing system. Wada *et al.* [4] have also applied an electron bombardment method to the lattice-like moderator at the KEK slow positron beamline, where the moderator components were encased in a box made from tungsten, and the box was irradiated with a high current electron beam obtained from an electron beam welder. However, they measured only the surface temperature of the box. Measuring the temperature of the moderator directly with a pyrometer is difficult in their method, although the surface temperature of the box can be easily measured. It is considered that the surface temperature of the box differs from the actual temperature of the moderator assembly which is encased in the box. Even if a bare lattice-like moderator is installed in the electron beam welder without encasing in the box to monitor the temperature directly, the moderator will not be sufficiently heated since the colliding area of the electron beam is too small. In this study, we developed a moderator annealing system with a handmade electron gun. In order to achieve a direct monitoring of the moderator temperature and maximize the collision area of the electron beam, a bare lattice-like moderator was installed in the annealing system at an angle of $\theta = 45°$ to the electron beam direction.

2.   System configuration and heating characteristics

Figure 1 shows a schematic diagram and photographs of the constructed moderator annealing system and the lattice-like tungsten moderator. The device is assembled with 100-μm-thick tantalum foils by spot welding, and composed of double layers for a thermal barrier. A slit is cut for monitoring the temperature with a pyrometer. The device contains a tungsten mesh filament ($\phi30$ μm, 100 wires·inch^{-1}) with a size of 20 mm × 10 mm as an electron source. A power-supply with output voltage/current of 10 V/10 A is used for filament heating. Electrons emitted from the filament are accelerated and are incident on the moderator which is floated to a high-voltage (+5 kV). The moderator is put on a tilted tungsten plate shown in Fig. 1(c) in order to collide electrons directly. With such an arrangement, it becomes possible to monitor the temperature of the whole area of the moderator by a pyrometer
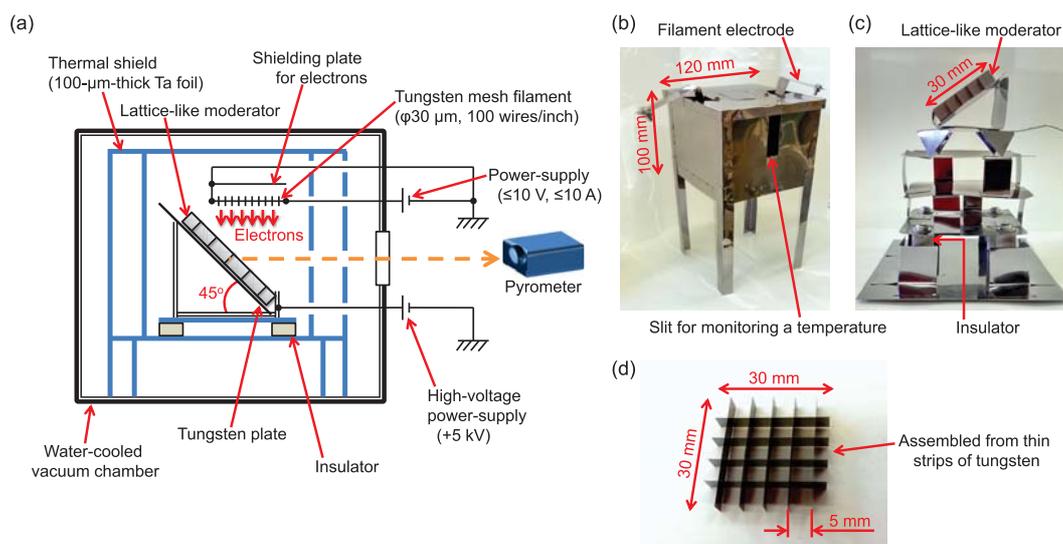

Fig. 1.   (a) Schematic diagram of the constructed annealing system. (b) External view of the annealing device. (c) Internal assembly of the annealing device. (d) Lattice-like pre-assembled tungsten moderator.

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during annealing. This annealing device was installed in a water-cooled vacuum chamber. The lattice-like moderator is prepared from thin strips of tungsten with a size of 30 mm × 5 mm × 0.05 mm as shown in Fig. 1(d). Each strip has some slits for assembling in the lattice-like configuration.

For monitoring the moderator temperature, a Chino Model IR-AH3SU portable radiation thermometer was used, which has a measurement diameter of $d = L/250$ mm (L: measurement distance). In our device configuration, the measurement area of temperature on the tilted ($\theta = 45°$) tungsten ribbons was about 2.8 mm × 2 mm ellipsoidal size, because the measurement distance between the lattice-like moderator and the pyrometer was around 500 mm. To measure the temperature of an object the thermometer observes visible light with a wavelength of 0.65 μm. Thermal radiation from actual objects is less than that from a perfect black body. Thus, in a radiation thermometry, it is necessary to correct measured temperatures by using the emissivity of the measured object. The corrected true temperature T is given by $1/T = 1/T_\lambda + (\lambda/c_2) \ln \tau_\lambda \varepsilon_\lambda$, where T_λ is called the brightness temperature and is the measured temperature assuming the measuring object is a perfect black body, λ (m) is wavelength of the observation light, c_2 (m·K) is Planck's second radiation constant, τ_λ is the spectral transmissivity of vacuum window, and ε_λ is spectral emissivity of the measured object, respectively [19]. Planck's second radiation constant c_2 (m·K) is also given by $c_2 = ch/k = 0.0144$ m·K, where c (m·s^{-1}) is the speed of light in vacuum, h (m^2·kg·s^{-1}) is the Planck constant, and k (m^2·kg·s^{-2}·K^{-1}) is the Boltzmann constant, respectively. Now, the wavelength of the observed light λ is 6.5×10^{-7} m, the spectral transmissivity of the borosilicate glass window τ_λ and the spectral emissivity of tungsten ε_λ are assumed to be 0.9 and 0.43, respectively [20, 21]. True temperatures were obtained by correcting the measured brightness temperatures using above values.

Heating characteristics of the developed annealing system were investigated. The relationship between the temperature of the central region of the moderator and the injected thermionic power is shown in Fig. 2. Using the above-mentioned method, we have successfully achieved an annealing temperature of 2600 °C in the central region of the moderator with a thermionic power of about 1200 W (5 kV, 240 mA). In the temperature range above 2000 °C, temperature differences of around 200°C were observed between the center and edges of the lattice-like moderator. Therefore, in order to anneal the whole area of the moderator with the required temperature of more than $0.7 T_M$ (2300 °C), the central region of the moderator was heated to 2600 °C. This simple tilting setup ($\theta = 45°$) of
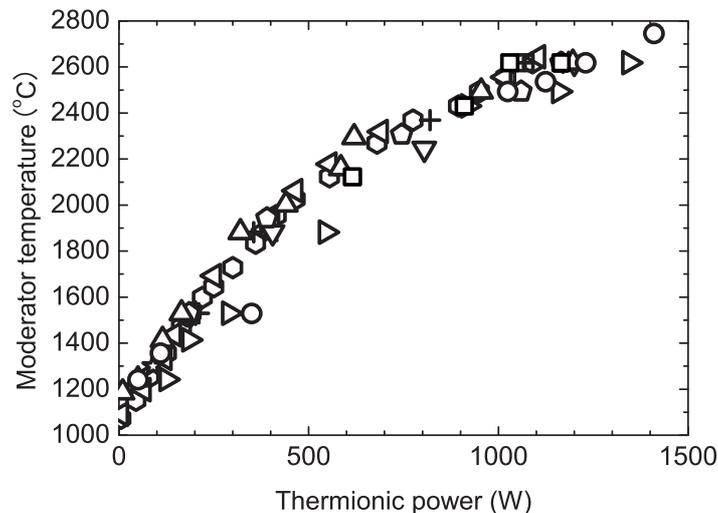
Fig. 2. Temperature of central region of the moderator as a function of irradiated thermionic power. Different annealing series are denoted by different symbols.

the moderator against the electron beam direction enables us to monitor the lattice-like moderator temperature directly even at 2600 °C. Since the electron beam current was stable for several minutes during annealing, time fluctuation of the moderator temperature was not observed.

3.　Annealing procedures and characterization methods

The annealing procedure was performed as described below by referring to previous studies [11, 16]. Temperatures recorded in this study were values obtained from the central region of the lattice-like moderator. The edges were also estimated to be heated to the desired temperature of 2300 °C by heating the central region to 2600 °C.

(1)　Evacuate the chamber to 5×10^{-6} Pa.

(2)　Apply a high voltage (+5 kV) to the moderator.

(3)　Anneal the moderator to 2600 °C for 3 minutes.

(4)　Stop heating.

(5)　Introduce oxygen gas to the chamber. (1.3×10^{-4} Pa)

(6)　Anneal to 1500 °C for 1 minute.

(7)　Evacuate the introduced oxygen gas.

(8)　Anneal to 1900 °C for a few seconds.

(9)　Repeat Steps 5–8 for 3 cycles.

(10)　Anneal to 2600 °C for 3 minutes, and allow to cool in vacuum with a cooling rate of roughly $10 \, °C \cdot s^{-1}$–$20 \, °C \cdot s^{-1}$.

(11)　Break the vacuum, rotate the moderator by 90° and return to Step 1.

Carbon is known as one of the major impurities in tungsten, and carbon atoms have been reported to segregate on the surface at high temperature [11, 22, 23]. Additionally, the surface-segregated carbon layer is also known to be removable by annealing in oxygen [12, 24]. The tungsten moderator (99.95 % purity) used at AIST contains carbon impurities with an average concentration of 15 ppm. Thus, in Step 3, the moderator is subjected to a high-temperature annealing as a process which segregates carbon atoms contained in a bulk of the tungsten to the surface. The diffusion coefficient D of carbon atoms in a tungsten is given by $D = D_0 \exp(-Q/kT)$, where D_0 ($m^2 \cdot s^{-1}$) is pre-exponential factor and Q (eV) is the activation energy for diffusion. In the temperature range 2073 K–3073 K, D_0 and Q are reported to be 9.22×10^{-7} $m^2 \cdot s^{-1}$ and 1.75 eV, respectively [25]. In addition, the diffusion length L of carbon atoms is given by $L = \sqrt{Dt}$, where t (s) is the diffusion time. Since the diffusion rate is estimated to be 19 $\mu m \cdot s^{-1}$ at 2300 °C, almost all carbon impurities in the 50-μm-thick tungsten foil are considered to be segregated to the surface during a few seconds. The surface-segregated carbon layer is removed as CO gas by annealing in oxygen atmosphere (Step 6). Step 6 forms an oxide on the tungsten surface, however, the oxide layer is also reported to be removable by annealing over 1500 °C [11, 24]. Steps 5–8 are repeated for 3 cycles. Phosphorus is also known as a major non-metal impurity in tungsten. The tungsten foil used in this study contains phosphorus with an average concentration of 20 ppm. Phosphorus has been reported to be segregated at grain boundaries in polycrystalline tungsten [26]. Therefore, the tungsten surface is also estimated to be coated with phosphorus after heating. However, similar to the carbon removal, the surface segregated phosphorus will be removed as a gas during oxygen annealing. Although tungsten contains other impurities such as Mo or Fe, the existence of such metal impurities in the bulk will not affect positron reemission properties at the surface significantly. Finally, to remove residual defects, the tungsten moderator is subjected to annealing to 2600 °C for 3 minutes, and then cooled to room temperature gradually (Step 10). After cooling, the moderator is rotated by 90° and the procedure is repeated from Step 1 in order to anneal the tungsten strips in the perpendicular orientation.

The effect of the annealing process on the lattice-like tungsten moderator was characterized by measuring positron annihilation lifetimes. Positron lifetimes of tungsten foils before and after annealing were measured using the AIST LINAC-based brightness enhanced pulsed positron microbeam with a time resolution of about 250 ps (full width at half maximum) [6–8]. A central region was cut out as 5 mm × 5 mm foil from the pre-assembled tungsten moderator and was subjected to positron annihilation lifetime measurements. The lifetime spectra are decomposed into a sum of exponential terms: $L(t) = \sum(I_i/\tau_i)\exp(-t/\tau_i)$, where τ_i and I_i are the ith lifetime component and its intensity, respectively. The total intensity of all components is unity ($\sum I_i = 100$ %).

4. Measurement results of positron lifetimes and discussion

Figure 3 shows positron annihilation lifetime spectra for as-received and annealed foils obtained at a positron energy of $E = 5$ keV and 25 keV. The mean positron implantation depth at $E = 5$ keV and 25 keV is 27 nm and 358 nm, respectively [27–29]. Comparing the spectra before and after annealing, the short-lived lifetime components become further shortened by annealing. On the other hand, long-lived components are also observed in each lifetime spectrum. Therefore, two-component analysis was conducted, and the fitting result is shown in Fig. 4.

At $E = 5$ keV, the long-lived intensity I_2 for the annealed foil is increased compared to that for the as-received foil without changing the long-lived lifetimes. This indicates that the number of positrons which diffuse back to the surface is increased by annealing. Thus, it suggests that the observed long-lived positron lifetimes τ_2 of about 350 ps correspond to a surface-state lifetime.

At $E = 25$ keV, the intensities I_2 are decreased compared to $E = 5$ keV. This means that the number of positrons which diffuse back to the surface is decreased since the positron implantation depth is increased. Calculated positron lifetimes of tungsten bulk and monovacancy have been reported to be 108 ps and 200 ps, respectively [30]. The observed short-lived lifetime τ_1 (184 ps) in the as-received foil is somewhat shorter than the reported monovacancy lifetime. The positron lifetime of monovacancies associated with dislocations is reported to be shorter than that of isolated monovacancies [31, 32]. Thus, the observed τ_1 for the as-received foil is considered to correspond to dislocation-related vacancy defects. After annealing, the observed τ_1 is reduced to 103 ps and is in good agreement with the value of bulk lifetime reported in Ref. [30]. Two positron lifetimes, corresponding to annihilation in the defect-free bulk and at the surface-state, can be derived by the fitting for the annealed foil. Therefore, we conclude that vacancy-type defects in the tungsten foil are successfully eliminated by conducting the above-mentioned annealing process.
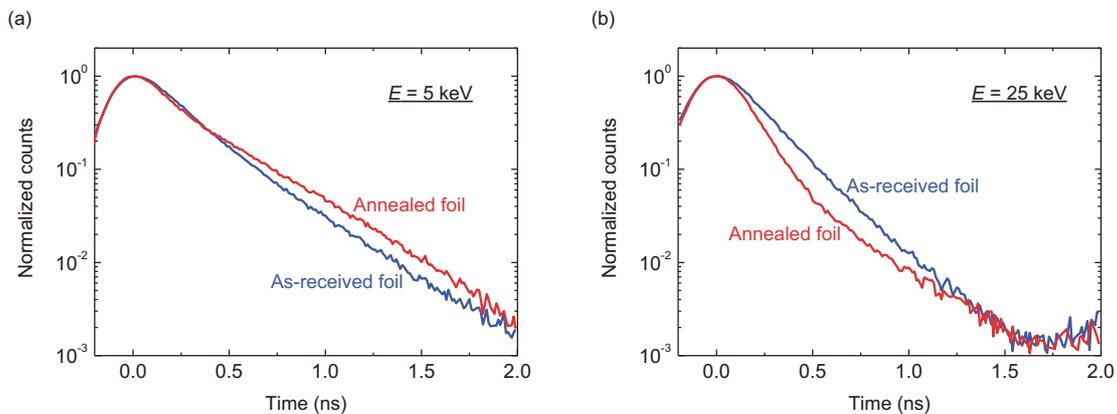

Fig. 3. Positron annihilation lifetime spectra for as-received and annealed foils obtained at (a) $E = 5$ keV and (b) $E = 25$ keV.
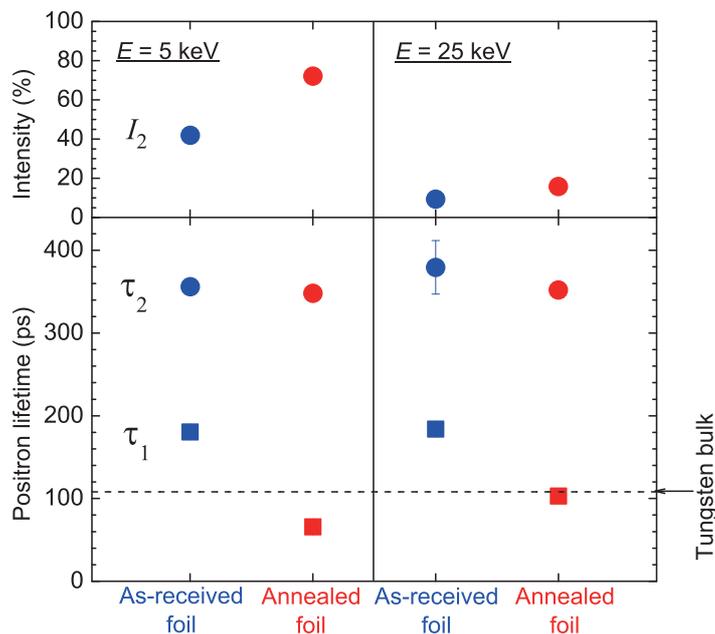
Fig. 4. Positron lifetimes (τ_1 and τ_2) and intensities (I_2) obtained for the tungsten foils before and after annealing at E = 5 keV and 25 keV. Calculated bulk lifetime reported by Troev *et al.* [30] is depicted by a horizontal dashed line.

5. Summary

In this study, we have developed a lattice-like moderator annealing system with a handmade electron gun. The temperature of the bare moderator assembly was directly monitored by using a pyrometer, and the central region of the tungsten moderator was successfully annealed up to 2600 °C with a thermionic power of about 1200 W. The whole area of the moderator was estimated to have been heated to more than 2300 °C which is the required temperature for eliminating residual defects in tungsten. Positron annihilation lifetime measurements were performed on the tungsten foils before and after annealing. As a result, residual defects contained in the as-received tungsten foil were eliminated by annealing. Therefore, it is concluded that the constructed device meets the required specifications as an annealing system for positron moderators.

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