Flash lamp annealing of tungsten surfaces marks a new way to optimized slow positron yields

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Abstract. Tungsten in the form of a mono-crystalline foil with an optimum thickness of about 2 µm is often used as a positron moderator in mono-energetic positron beams with 22Na positron sources. The efficiency of such a moderator strongly depends on its prior heat treatment, i.e. an annealing procedure with considerable difficulty at temperatures of about 2000 °C under vacuum conditions. Flash lamp annealing (FLA) has been tested as new method to quickly anneal W foils in order to produce easy manageable, low-cost moderators with a high efficiency. With FLA, just the surface of a W foil is heated above the melting point (3422°C) within 1 to 3 ms, i.e. without melting the whole foil volume. In this way, a surface cleaning is reached connected with a considerable increase in the positron diffusion length. Conventional polycrystalline W foils of 9 µm ± 25% thickness, heat treated by FLA, were characterized and tested as positron moderators. First promising tests result in a moderator efficiency of ~3*10⁻⁴ and clearly demonstrate that FLA is also applicable to tungsten meshes.

1. Introduction

The worldwide scarcity of commercially available 22Na sources and the resulting dramatic rise in its price demand new thinking about an optimal application of 22Na as a β⁺ source in mono-energetic positron beams for depth-dependent Doppler broadening measurements [1]. The positron moderator is the crucial factor for the creation of enough mono-energetic positrons for their use in such investigations. There are two main requirements for the moderator properties: a stable, high moderation efficiency and manageability at low costs. Tungsten generally meets both requirements well. Nevertheless, the commonly used 2 µm single crystalline tungsten foils are especially susceptible to radiation damage and require frequent defect annealing, which is associated with considerable difficulties. Fig. 1 shows the decrease in the count rate of such a foil in comparison to the decrease calculated with the decay law. The foil was mounted in front of a 2.4 GBq source. It becomes clear that moderator annealing is essential for an effective operation of mono-energetic positron beams. A new method was sought to replace the laborious annealing of the thin foils mostly done by means of electron heating under high vacuum conditions by a more simple method. Flash lamp annealing (FLA) seems to be viable for this purpose and experimental results regarding the first application of flash-lamp-annealed tungsten foils are presented.
2. Efficiency of tungsten foils with different thicknesses
FLA of tungsten requires opaque foils in order to avoid local overheating, which causes the formation of holes in the foils. Opaque polycrystalline foils are commercially available with a minimum thickness of 9 µm ± 25%. However, thicker moderator foils lead to decreasing moderator efficiency. For this reason, a short calculation of the efficiency of a 2 µm foil in comparison to the thicker foil of 9 µm was made. The calculations were performed with the following assumptions: The positrons from the source capsule penetrate a 5 µm thick Ti window and enter the moderator foil, which is directly mounted behind the window. The energy distribution of the positrons only marginally changes after passing the Ti window. The fraction of the positrons $F(z)$ which is stopped in a certain depth $z$ within the moderator can be calculated by use of the energy distribution of the positrons $N(E)$ and assuming a Makhovian profile $P(E,z)$ for the depth distribution according to Eq. (1).

$$F(z) = \int_{0keV}^{545keV} N(E)P(E,z)dE$$  \hspace{1cm} (1)

Fig. 2 shows the dependence on the penetration depth for positrons stopped in a tungsten foil. It becomes clear that the optimum thickness of a tungsten moderator foil is about 2 µm. A thickness of 9 µm correlates with the mean positron implantation depth, with only 59% of stopped positrons below the surface compared to the thinner foil. The expected moderation efficiencies for both foils are shown in Fig.3. Thereby the moderator efficiency is defined as ratio of moderated positrons divided by fast positrons entering the moderator. Assuming that the bulk diffusion length $L_{+}^{bulk} = 135$ nm [2] the
The expected maxima of the efficiencies should be $4.5 \times 10^{-3}$ for 2 \(\mu\)m and $2.7 \times 10^{-3}$ for 9 \(\mu\)m thick foils. In Ref.[3], it was reported a moderator efficiency of a 6 \(\mu\)m thick tungsten foil of $2.6 \times 10^{-4}$ after annealing at 2100 \(^\circ\)C in vacuum. Surface properties, grain boundaries, impurities and other influences, which were not included in the rough calculations, are responsible for the considerable discrepancy between calculation and reality.

3. Flash lamp annealing of tungsten foils

FLA [4] was used for cleaning the surface of the foils and for defect annealing of the poly-crystalline foils. It offers a chance for the optimization of the moderator properties. This method enables the heating of the surface of the W foil above the melting point of 3422 \(^\circ\)C in 1 to 3 ms without melting the whole volume. The very short heat input into the surface is realized by xenon-filled gas discharging lamps, which were charged by a capacitor bank with a capacitance of 4.8 mF and a maximum charging voltage of about 4 kV. The whole capacitor bank is discharged via induction coils for the adjustment of the duration of the flash. Twelve Xe flash lamps generate the short light pulse with a spectral distribution between 400 and 700 nm, peaking around 500 nm. For the FLA of the 9 \(\mu\)m W foils, a light pulse of 1.25 ms has been chosen with an energy density of 32 J/cm\(^2\) resulting from an electrical power of 31 MW. This heat treatment was carried out stepwise in an Ar flow at normal pressure, starting with an energy density of 8 J/cm\(^2\). The complete cleaning of the surface could be observed at an energy density of 12.5 J/cm\(^2\), which corresponds to a surface temperature of about 1600 \(^\circ\)C. While the surface cleaning could be obtained below 2000 \(^\circ\)C, temperatures above the melting point had to be applied in order to obtain longer positron diffusion lengths due to grain growth and defect annealing (Fig.4a and b and Fig.5).

4. Characterisation and test of the moderator foils and meshes

The mono-energetic positron beam “SPONSOR” at Helmholtz-Zentrum Dresden-Rossendorf [5] has been operating with a 2.4 GBq \(^{22}\)Na source and a 2 \(\mu\)m single crystalline W moderator for about nine years. Finally, in 2011 the source had decayed to 308 MBq and the measured count rate below the 511keV line decreased from 1800 c/s at the beginning to 22 c/s. Therefore, the moderator was replaced by a flash lamp annealed 9 \(\mu\)m polycrystalline W foil and a nearly 20 times higher count rate of 400 c/s could be immediately reached. Taking into account the distance between Ge detector and sample, efficiency and size of the Ge detector, the number of positrons entering the moderator and other experimental influences, the efficiency of the moderator could be estimated to be about $3 \times 10^{-4}$, which is marked as a star in Fig.3. Fig.5 shows the measured S parameter versus the incident positron energy E for the as-received W foil and the moderators that were used. The positron diffusion lengths L\(_+\) were fitted using the VEPFIT software [6]. The fits result in very short L\(_+\) \~ 1 nm for the as-received foil, in L\(_+\) = 48 (8) nm for the 2 \(\mu\)m foil and 94 (13) and 99 (16) nm for FLA 1 and 2 foils, respectively. Because of 25% deviation in thickness of the poly-crystalline foils a complete surface melting could not be obtained and the diffusion length in the FLA foils is shorter than the expected one [2]. From the
comparison of the diffusion lengths of the 2 µm foil and the poly-crystalline foil it becomes clear that the shorter diffusion length only contributes to half of the degradation of the efficiency, which is more than an order of magnitude in total. Structural changes at the surface primarily influence the moderator properties. In this context, it is worth mentioning that all W foils showed a decrease in the efficiency of about 20% under vacuum conditions of 5x10^-7 Torr after one week. The primary efficiency could be restored by exposure of the foils to air for a few hours.

W meshes can be etched with a homogenous volume degradation, which leads to a uniform diameter of the W wires. For this reason, heating of the entire surface to the melting point is possible without damage by local overheating, as seen in Fig. 6. Taking into account an efficiency like that of the poly-crystalline foils and three times higher surface area of the meshes (diameter of the wire 10-15 µm), a total efficiency of 9x10^-4 should be achievable using 10-12 meshes.

**Figure 5.** S(E) of W moderators.  
**Figure 6.** W meshes treated by FLA.

5. Conclusions
FLA is a suitable annealing procedure for W foils of a thickness of 9 µm. By heating the surface to near the melting point, it could not only be cleaned, but a longer positron diffusion length could be reached, which would be impossible with a thermal treatment of around 2000 °C. The obtained moderator efficiency of 3x10^-4 agrees well with the published value [3], though it falls short of the optimum, and further efforts for its improvement are needed. Nevertheless, the short annealing time and stable properties of the foils even when exposed to air legitimate this method. The application of FLA to W moderator meshes is promising. The intention is to realize this as a next step.

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