Development of a sample chamber with humidity control for an atmospheric positron probe microanalyzer

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Abstract. In order to perform positron lifetime measurements on thin films under atmospheric conditions, a slow positron microbeam was extracted into air using silicon nitride thin films (30 nm and 200 nm) as a vacuum window. Even the thinner window (30 nm) was found to reliably withstand a differential pressure of 1 atm under various stress tests. By placing the sample in an enclosed chamber through which gas with a fixed, controllable relative humidity (RH) was continuously passed, the RH dependence of the ortho-positronium lifetime for bulk fused silica was examined.

1. Introduction

The positron annihilation technique, based on keV-energy positron beams, is a powerful tool for investigating atomic defect and sub-nanometer scale pores of thin films. In practical measurements slow positrons are generated in a high vacuum, therefore, it is necessary to place samples in a vacuum chamber. Recently, extraction of slow positrons into air has been achieved with the help of a pulsed positron microbeam, produced for the positron probe microanalyzer (PPMA) in AIST[1-3]. This method is promising to be apposite for exploring nanoscopic holes of thin films under practical conditions. In this paper, we report details of reliability tests of the vacuum window and the humidity controller, specially designed for PPMA, and preliminary results of variable-energy positron lifetime measurements for bulk fused silica, performed at various humidity conditions using the developed system.

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2. Atmospheric PPMA

A schematic view of the atmospheric PPMA$^{[4,5]}$ in AIST is shown in Fig. 1. An intense positron beam produced by an electron linear accelerator (LINAC) is focused by a magnetic coil on a remoderator. The brightness enhanced positrons are transferred from the remoderator and are extracted to air through a vacuum window. The window is a silicon nitride (SiN) thin film on a silicon substrate, providing a tiny opening typically less than 1 mm$^2$ for extraction of positrons to air through the SiN film. Then the positron microbeam is focused by an objective lens down to a final beam size of 200 µm$^1$ at the vacuum window. The vacuum window is placed on the central hole of the terminal metal flange of PPMA, and is fixed by epoxy glue$^5$. The SiN film is a fragile part in the vacuum system, thus accidental breakage could cause serious damage to the vacuum system due to the rapid leak of air. In consideration of various working conditions, for instance, high humidity and/or temperature, the SiN film should not break for at least 10 h to achieve the total number of counts during positron lifetime measurements under high relative humidity (RH) at 30 °C. It is therefore necessary to ensure the reliability of the SiN windows.

3. Strength tests for SiN vacuum window

Materials such as Be, Ti and SiN are often used as vacuum windows. In our apparatus, the areal density of the window should be as small as possible to optimize the efficiency of extracting low energy positrons into air. Based on this condition SiN was chosen for the window. SiN films with thicknesses of 30 nm and 200 nm (density ~ 3 g/cm$^3$) were purchased from Silson Company. For the strength tests we built a simple vacuum system, consisting of a scroll pump, a leak valve and a vacuum gauge that are connected by a flexible metal vacuum tube with each other (Fig. 2). The SiN vacuum window was fixed at the end of this system. Three different strength tests were performed; (a) water adsorption, (b) heat treatment, and (c) sample contact, and each test was performed at least 10 times for each SiN film. During the tests the degree of vacuum was kept constant around 5.3 Pa. The details and a summary of the strength tests of SiN windows are shown in Fig. 3 and Table 1, respectively.

![Fig. 1. Schematic illustration of the atmospheric PPMA$^{[1]}$.](image)

![Fig. 2. The vacuum system for strength tests.](image)

| Properties                        | 30-nm thick | 200-nm thick |
|-----------------------------------|-------------|--------------|
| Lateral size                      | 0.6 mm×0.6 mm | 1.5 mm×1.5 mm |
| Vacuum degree (Pa)                | 5.3         | 5.3          |
| Displacement of the window center | 35 µm ~40 µm | 65 µm ~70 µm |
| Water resistance (immerse in water for 10 h) | Succeed | Succeed |
| Temperature resistance (90 °C~110 °C for 20 min) | Succeed | Succeed |
| Sample directly contact           | Failed      | Succeed      |
| Sample mounted with Kapton spacers | Succeed | Succeed      |
Fig. 3. Strength tests for the SiN vacuum window. (a) The SiN window was immersed in distilled water completely and then left to dry by evaporation at room temperature. The SiN window worked well under the effect of the surface tension of a water-drop; (b) The vacuum window and the stage were covered by 4 layers of aluminium foil. A thermal sensor was placed near the SiN window to monitor the temperature. The window was heated by a heat gun to an enough high temperature from the direction shown by arrow. The SiN window also worked normally; (c) A piece of a soft rubber film was put onto the thin vacuum window directly. In this test, the 30-nm SiN window was broken.

The 30-nm-thick SiN window failed the direct sample contact test. For this reason, 35-µm Kapton foils were placed between the sample and the vacuum window as a spacer when the 30-nm-thick SiN was used for the positron measurements. The kapton spacers keep the SiN window away from sample and protect the window preventing direct sample contact. After these three strength tests, both the 30-nm and 200-nm-thick SiN films were confirmed applicable to the vacuum window of the atmospheric PPMA.

4. Sample chamber with humidity controller

A sample chamber equipped with a humidity controller (Makuhari Rikagaku Garasu, Inc.), specially designed for the atmospheric PPMA, was installed on the air side of the SiN window (Fig. 4). A BaF$_2$ scintillator was placed outside the sample chamber to detect the annihilation γ-rays of the positrons. A magnetic coil was used as an objective lens to regulate the positron beam diameter$^{[2]}$. The beam diameter at the sample was adjusted to be smaller than the lateral size of the SiN window. The slow positrons will reach the sample surface after crossing the SiN film and air. The transmission efficiency of positrons in SiN, the positron beam properties emerging from the SiN thin window and depth profile of positrons in a sample are discussed elsewhere$^{[6]}$. The humidity controller and sample chamber were electrically floated from the ground level to supply the accelerating bias for adjusting the positron injection energy into the sample. A metal lid was used to seal the sample chamber, and a nitrogen gas with different levels of humidity were passed continuously through the chamber, allowing a fixed and controllable relative humidity and keeping the pressure constant at 1 atm. Two probes were used to monitor the humidity and temperature inside the chamber. By controlling the flow rates of a dry nitrogen gas and a nitrogen gas saturated with water vapor, RH inside the chamber can be varied from 0 to over 90%. The response and stability of RH in sample chamber is shown in Fig. 5. RH in the chamber stabilizes within 2 min with a standard deviation not more than 1.2%, when...
changing from one value to another value. Note that, based on Fick’s law of diffusion, the time of water molecules at 30 °C to pass through the channel between the SiN window and the sample is estimated as ~10 s, which is much shorter than the time to get the stability of the humidity condition in the chamber (~2 min).

5. Performance of the atmospheric PPMA
In order to check if the ortho-positronium (o-Ps) lifetime is correctly measured even if the relative humidity varies, bulk fused silica with an o-Ps lifetime of ~1.6 ns\(^{[5]}\) was selected as a standard sample. The RH dependence of the o-Ps lifetime for bulk fused silica is shown in Fig. 6. The 30-nm-thick SiN film was selected as the vacuum window for the measurements. The positron incident energy \(E\) was 4 keV and the beam diameter was 200 \(\mu\)m. The total count in each spectrum was 1\(\times\)10\(^6\). The thickness of the fused silica sample was 0.6 mm with the lateral size of 1.5 cm \(\times\) 1.5 cm. At high RH, the thickness of adsorbed water layer on the surface of fused silica is reported to be less than 1 nm\(^{[8]}\), so that the effect of the water layer on the lifetime measurements at \(E = 4\) keV is negligible. As in Fig. 6, the measured o-Ps lifetimes for the fused silica (~1.6 ns) show no dependence on RH (0, 43% RH and 85% RH). This result indicates that the apparatus worked well during the lifetime measurements.

6. Conclusion
A sample chamber equipped with a humidity controller for the atmospheric PPMA has been developed. The SiN thin films (thickness 30 nm and 200 nm, lateral size 0.6 mm \(\times\) 0.6 mm and 1.5 mm \(\times\) 1.5 mm, respectively) were confirmed suitable for the vacuum window of the atmospheric PPMA. The humidity controller was constructed to provide variable and stable relative humidity conditions in the sample chamber. The RH dependence of the o-Ps lifetimes for the fused silica was measured to confirm the normal operation of the atmospheric PPMA. This work was supported by NEDO. The SiN shape measurement was supported by the AIST Nano-Processing Facility.

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