Natural and enriched tungsten as target for heavy ion experiments

Bettina Lommel *, *Annett* Hübner, *Birgit* Kindler, *Anita* Reckziegel, *Maxim* Saifulin, *Jutta* Steiner, and *Vera* Yakusheva
GSI Helmholtzzentrum für Schwerionenforschung, Planckstr. 1, 64291 Darmstadt, Germany

Abstract. Tungsten is applied as target material for experiments in the field of nuclear spectroscopy. Typically, targets of a thickness range of 100 µg/cm² up to 1000 µg/cm² that can safely stand the irradiation by the intense heavy-ion beam over a substantially long period are requested. Natural tungsten is commercially available as thick foils and as sputtering targets. Enriched tungsten is typically available as metal powder. We report on the production of natural and enriched tungsten targets on carbon backing. We compare the different production processes, taking into account the material yield.

1 Introduction

For experiments aimed to understand the structure of matter, tungsten is one of the interesting target materials for the synthesis of the exotic nuclei in the heavy-ion induced reactions. For experiments at GSI, Helmholtz Centre for Heavy Ion Research in Darmstadt, Germany, tungsten targets, preferably highly isotopically enriched, were needed for nuclear spectroscopy experiments, e.g., at the gas-filled recoil separator TASCA and at the velocity filter SHIP [1, 2, 3]. Targets with surfaces of 1000 - 3000 mm² and with a thickness of ~ 600 µg/cm² were typically requested. In the area where the beam hits the target a homogeneity of ± 10 % was required, which applies to at least two thirds of the total target surface.

2 Prior information

2.1 Tungsten and tungsten oxide

Tungsten is a transition metal; it is rare on earth. In nature, tungsten is not available as the pure material but always in its compounds. Tungsten has the highest melting point of all elements with 3422 °C.
In natural tungsten, there are three stable isotopes: ^{182}W (natural abundance 26.5 %), ^{184}W (natural abundance 30.64 %) and ^{186}W (natural abundance 28.43 %). Additionally there are two natural isotopes, which are alpha emitters with a very long half-life: ^{180}W (natural abundance 0.12 %, half-life 1.8×10^{18} a) and ^{183}W (natural abundance 14.31 %, half-life 6.7×10^{20} a) [4]

Under sufficient oxygen, WO_3 is formed, which is the stable oxide with a melting temperature of 1473 °C. WO_3 is yellow and turns blue from $WO_{2.98}$ on. [5, 6]

2.2 Published deposition processes

In the past, targets out of natural tungsten have been produced by electron bombardment [7], extracted ion beam sputtering [8], electron beam deposition [9] and hot rolling [10].

Targets out of enriched tungsten have been manufactured by powder sedimentation [11] and electron beam deposition [12, 13].

2.3 Material availability

Natural tungsten is commercially available in various forms: as foil, as metallic sputter target, as pressed and sintered metal oxide sputter target, as metal powder and as metal oxide powder. Therefore, starting material in suitable form is available for all standard methods to produce self-supporting thin films or thin films on backings. All tungsten isotopes (mentioned earlier) can be purchased as metal powder for a maximum price of a few €/mg. The grain size of the metallic tungsten powder is defined by the manufacturing process and cannot be selected when purchasing powder. Therefore, one cannot specify a desired grain size of the powder, that would suit the target production process best. The form of the material available on the market (or in the laboratory) defines the possible target production processes.

2.4 Possible target production processes

In the target laboratory at GSI, we have different deposition techniques routinely available that in principle can be applied for deposition of tungsten and / or tungsten oxide, such as are thermal deposition,

* Corresponding author: b.lommel@gsi.de

electron beam deposition, DC magnetron sputtering and RF magnetron sputtering.

Thermal deposition is a straightforward method with a rather high yield and a high flexibility regarding form of the suitable starting material. Homogeneity and yield can be easily controlled by varying the distance from crucible to substrate. However, for thermal deposition, the most important prerequisite is the availability of a suitable crucible material. Therefore, for tungsten metal this method is no option, since it has the highest melting point of all metals and consequently no crucible material is available. For tungsten oxide with its much lower melting temperature, thermal deposition is a promising option.

Electron-beam deposition allows the evaporation of high melting materials from a water-cooled crucible [9]. Because of the high impact of the electron beam and the high heat radiation especially for high-melting substances, the distance between hearth and substrate in general has to be very large leading to a very poor yield. Therefore, electron-beam deposition is no alternative for enriched material where yield is an issue.

Magnetron sputtering inherently works independent from the melting point of the material. Here, the minimum distance from source to substrate depends on the diameter of the sputtering source. For the production of films with a good homogeneity, the yield is in general lower than with thermal evaporation. Depending on the electric conductivity of the sputtering material, either DC sputtering or RF sputtering has to be applied. For both methods, the availability of the starting material in form of a sputtering target with a suitable diameter and thickness is prerequisite.

Based on these considerations, we have identified the thermal evaporation and magnetron sputtering as the most promising methods with the best yields, namely magnetron sputtering, if it is possible to convert the available material into a suitable sputtering target, and thermal evaporation, if it is possible to convert the available material into oxide.

3 DC magnetron sputtering

Fig. 1. View of the applied Edwards AUTO 306 ® Sputter coater with the 1” DC magnetron in front.

3.1 DC magnetron sputtering with commercial sputter targets

Natural metallic tungsten was sputtered on carbon backing of ~ 100 µg/cm² on different standard frames: frames with round opening with a diameter of 10 mm or 20 mm, or banana-shaped targets with an area of up to 3000 mm² to be applied on a target wheel.

A metallic tungsten sputter target with a diameter of 25,4 mm and a thickness of 3 mm was mounted in a distance of 45 mm to the evaporation wheel with the carbon backings in the Edwards AUTO 306 Sputter coater, as shown in figure 1. With a power 100 – 150 W up to 30 minutes in 5 minute steps with at least 20 minutes break in-between tungsten targets with a thickness up to 600 µg/cm² for the small round targets and with a thickness of up to 300 µg/cm² for the banana-shaped targets could be produced.

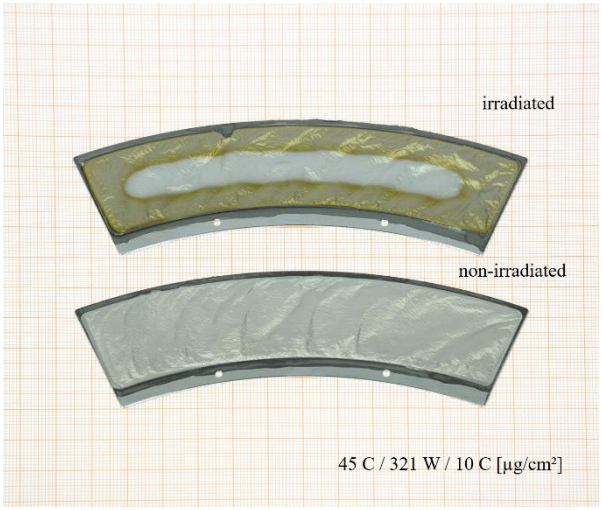

Fig. 2. Tungsten targets out of natural tungsten on a millimetre paper. Carbon backing of 45 µg/cm², natural tungsten of 321 µg/cm² and a carbon covering layer of 10 µg/cm² glued with carbon glue on a AlMg3 frame. The irradiated target is situated on top, the non-irradiated target is situated below.

Figure 2 shows metallic tungsten targets with a thickness of 321 µg/cm² on 45 µg/cm² carbon with a 10 µg/cm² carbon covering layer. The carbon is glued with carbon glue on a frame out of $AlMg_3$, 8 carbon foils on frames are situated on an evaporation wheel which runs above the DC magnetron, so the tungsten is deposited through the wheel openings. In figure 2, a freshly produced target and an irradiated target are shown in the bottom and in the top, respectively. In the latter, a track formed during the bombardment by the heavy-ion beam is well visible.

The sputtering process of tungsten puts major stress on the carbon backing. An increased appearance of holes and large cracks at the edges can be observed.

3.2 DC magnetron sputtering with sputter targets from pressed tungsten powder

To apply DC magnetron sputtering with enriched material, sputter targets had to be prepared from the enriched powder commercially available. To develop a suitable procedure sputtering targets pressed from natural tungsten powder were produced.

With a commercial hydraulic press from Paul Weber ® and a suitable pressing mould sputter targets with a diameter of 25 mm and a thickness of 0.7 mm were obtained. 2500 mg metallic tungsten powder was needed for this size of the tablet. To enable the pressing of such hard metal powder, we optionally added paraffin and glycerine. Targets with paraffin added started splashing and spraying already at 90 W so that only thin targets up to 130 µg/cm² could be deposited. Best results were obtained with adding glycerine, pre-heating the tablet with the electron-beam gun and additionally heating the tablet thermally under vacuum.

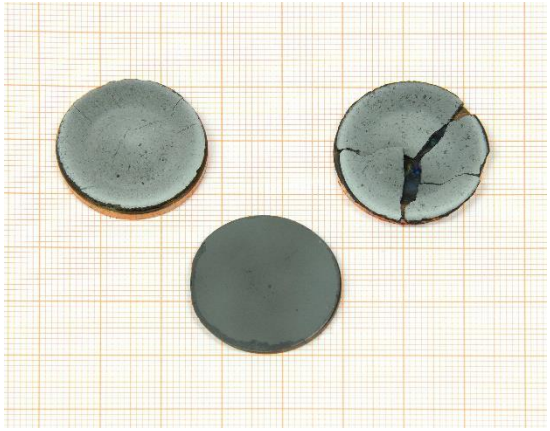

Fig. 3. Pressed tungsten sputter targets out of natural tungsten with a diameter of 25 mm on millimetre paper. The pressed target is situated below, the already applied targets with minor and major cracks after the deposition are situated above.

With these sputter targets thickness of up to 300 µg/cm² could be obtained. Nevertheless, when the produced tablets were applied as sputter targets, they were very brittle and the racetrack is formed quickly, as can be seen in figure 3, so finally the yield is unsatisfactory.

4 Targets from enriched tungsten oxide

Alternatively, we tested the conversion from tungsten to tungsten oxide to go for the thermal deposition of tungsten oxide as target material.

4.1 Conversion

Metallic tungsten was annealed in air up to 500 – 650 °C in steps until complete oxidation was reached which can be seen by the colour of the final oxide. We had to take into consideration that the final temperature depended on the grain size, since the grain sizes for the natural material and for the enriched material we had available were different

The material was heated in steps for a duration up to 2 hours. For the conversion of W into WO_3, ^{182}W into $^{182}WO_3$, ^{184}W into $^{184}WO_3$, ^{186}W into $^{186}WO_3$ a yield of 100 % could be reached.

4.2 Thermal deposition of tungsten oxide

Tungsten oxide with a thickness of up to 700 µg/cm² was deposited thermally on carbon backing with a thickness of 50 µg/cm² on standard frames with a covering layer of 5 – 10 µg/cm² carbon. The material was deposited from the tantalum crucible; the backing was heated from the backside with a quartz lamp to ~ 250 °C. The thickness was measured online with a quartz monitor and after the deposition process with a balance. The setup is shown in figure 4.

Fig. 4. View of the applied thermal deposition setup, Edwards AUTO 306 ®. The quartz lamp is situated above the evaporation wheel, the tantalum crucible is clamped between the massive copper electrodes below the evaporation wheel.

Targets with a thickness of up to 700 µg/cm² referring to the W could be obtained by thermal evaporation of tungsten oxide for ^{182}W, ^{184}W and ^{186}W.

Figure 5 shows targets out of $^{182}WO_3$ with a thickness of 421 µg/cm² referring to the ^{182}W on 48 µg/cm² carbon with a 5 µg/cm² carbon covering layer. The target at the bottom and in the middle are not irradiated; while the target at the bottom shows a complete covering of the carbon layer glued on a AlMg3 frame, the target in the middle was evaporated through a smaller window where only the area which will be covered by the beam in the experiment is deposited with tungsten oxide. This area is needed to measure and define the needed homogeneity. The target on top was irradiated in the beam; the track which was left from the ion beam is visible.

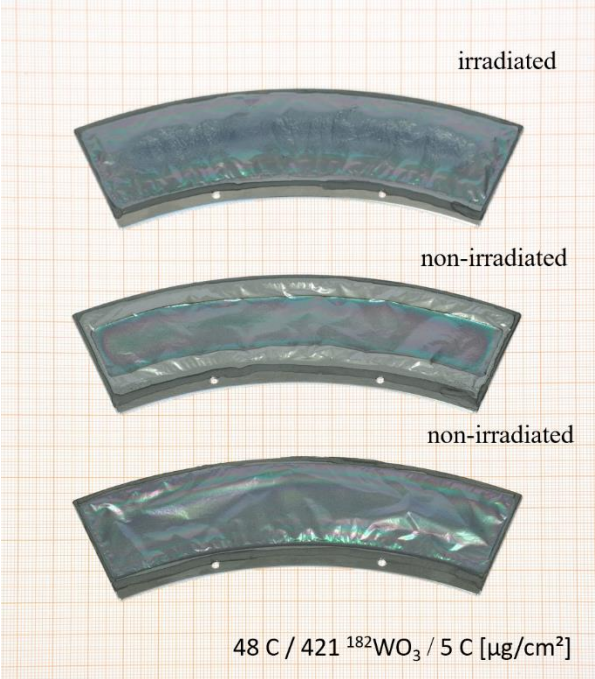

Fig. 5. Targets out of $^{182}WO_3$ with a thickness of 421 µg/cm² on 48 µg/cm² carbon and a carbon covering of 5 µg/cm². On the bottom tungsten fully deposited over the entire surface, in the middle deposition through a smaller deposition window in which the homogeneity of ± 5 % is achieved. On top the irradiated target with the track where the heavy ion hit the target can be seen.

5 Outlook

Tungsten targets with a thickness of 100 µg/cm² up to 700 µg/cm² depending on the target size could be obtained. Enriched tungsten oxide up to 700 µg/cm² for all sizes, reproducible in small series were produced.

As the next step, the metal tungsten targets produced by DC magnetron sputtering, tungsten oxide targets produced by DC magnetron sputtering and tungsten oxide targets produced by thermal deposition will be irradiated by the heavy-ion beam for the comparison of their performances. This is foreseen in the upcoming 2025 beamtime at TASCA, GSI.

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