

## PREPARATION OF ISOTOPIC RUTHENIUM TARGETS USING A FOCUSED ION BEAM SPUTTERING SOURCE

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### Abstract

The preparation of ruthenium targets to be used in heavy-ion experiments has always presented challenges due to its physical properties and the expense of the separated isotope. The straightforward approach of focused ion beam sputtering provides an efficient deposition technique, which allows for conservation of the isotopic starting material. A description of the target preparation will be presented along with results for several ruthenium isotopes.

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## 1. Introduction and Motivation

For several recent heavy-ion experiments using the Fragment Mass Analyzer (FMA) [1] at the Argonne Tandem Linear Accelerator System (ATLAS), targets of  $^{96,102,104}\text{Ru}$  having a thickness of  $\sim 500 \mu\text{g/cm}^2$  were required. These targets were prepared by the method of focused ion beam sputtering from a saddle-field ion source using noble gases. Backings of carbon, aluminum and gold with a variety of thicknesses were used.

Ruthenium is a notoriously difficult target to fabricate, having a high melting point ( $2334^\circ\text{C}$ ) and therefore, low vapor pressure. From the literature, preparation of ruthenium targets by the method of electrodeposition has been presented by Morgan in 1977 [2] and later by Cecchi [3]. A solution of  $\text{RuCl}_3$  in 0.1 N hydrochloric acid was placed in the electrochemical cell and plated, using a Pt anode, onto a copper backing foil acting as the cathode. The current density used was  $10 \text{ mA/cm}^2$ . Target thickness of  $0.5\text{--}2 \text{ mg/cm}^2$  on sturdy backings can be produced using this technique. This is a very efficient means for producing these targets, an important consideration when dealing with expensive isotopic material. For our target requirements we would need  $0.5 \text{ mg/cm}^2$  deposit on a thin backing, allowing for the emission of evaporation residues. Investigations into using this procedure for preparing ruthenium targets are currently in progress.

Thin targets of  $5\text{--}100 \mu\text{g/cm}^2$  by the method of physical vapor deposition (PVD) have been achieved by Maier-Komor [4] using electron bombardment. By employing an electron beam source of the Pierce principle, ruthenium depositions were carried out on  $3\text{--}5 \mu\text{g/cm}^2$  carbon backings. Although in our laboratory we have a similar apparatus, it is not clear whether the desired thickness needed for these experiments can be achieved using this technique.

Ruthenium deposition by sputtering has been shown by Maier [5] to be a viable means to obtain the desired targets. Focused ion beam sputtering using compact saddle-field ion sources has been available as a means of target production in our laboratory for some time [6]. It was decided therefore, to use this approach for the present situation.

## 2. Focused Saddle-Field Ion Source

For the depositions carried out in the present work, a Model FAB11N Fine Beam Ion Source (Ion Tech Ltd., Teddington, Middlesex, England) was employed. Through the introduction of a noble gas and an accelerating potential, this compact, water-cooled source produces a saddle-field ionization of the gas employed with a specially shaped central anode. The resulting beam emerges symmetrically along the source axis with the forward directed beam impinging on the target material to be sputtered and the beam current collected out the rear of the source on a Faraday cup arrangement. The source was operated at an accelerating voltage of 8 kV and an average beam current of approximately

40  $\mu$ A provided by a Model B50 constant voltage power supply (Ion Tech Ltd., Teddington, Middlesex, England).

### *2.1 Experimental Setup and Sample Preparation*

The experimental arrangement is identical to that used previously in our laboratory [7]. The source is mounted in an aluminum block on sliding rails at an angle of 30° [8] with respect to the plane of the sputtering target and adjustable in distance from the material to be sputtered. The distance from the front of the ion source to the sputter material was 5 cm. The substrate, or backing foil was mounted approximately 2 cm above the sputter target. At this distance, efficiencies approaching 10% may be achieved for a single target.

The depositions were carried out in a Model VE-775 vacuum evaporator (Veeco Instruments, Inc., Plainview, Long Island, NY) employing a 1500 l/s cryopump station. Under operating conditions, the noble gas was introduced to a partial pressure of approximately  $2.7 \times 10^{-3}$  Pa. The entire sputtering system is installed on a removable vacuum feed-thru ring, which may be easily incorporated into the vacuum evaporator. A photograph of the sputtering arrangement is shown in Figure 1.

The ruthenium samples were obtained as highly enriched separated isotope from Oak Ridge National Laboratory (ORNL). In the case of  $^{104}\text{Ru}$ , the sample was prepared as a compacted pellet from the powder, using approximately 100 mg and formed with a .452 mm diameter die (Parr Instrument Co., Moline, IL). The pellet is positioned on a glassy carbon support as suggested by Maier [9] and held in place using double-sided tape. The observed material consumption using this method was small and allowed for the pellet to be reused several times.

For the  $^{96}\text{Ru}$  isotope, a small electron beam melted sample of approximately 10 mg was used. Similarly, a 17 mg sample was used in the case of  $^{102}\text{Ru}$ . The glassy carbon support was modified by the machining of a small dimple in the center of the plate to hold the spherical sample bead. Previous work with this source by our laboratory [10] revealed a projected beam spot size of approximately 2 mm x 4 mm, which would clearly impinge on the carbon support as well as the sample. Therefore, at one point during the target depositions, the  $^{96}\text{Ru}$  bead was removed and sputtering done on the empty dimple to determine the amount of carbon sputtered. Using a small glass witness disk, a deposition rate of  $<0.2 \mu\text{g}/\text{hr}$  was observed for the carbon.

It should be pointed out that for the spherical beads, sputtering only occurred from the roughly hemispherical surface illuminated by the beam. In addition, the directionality of the sputtered particles is drastically altered. An estimate of the collection efficiency under these conditions versus that obtained from a planar surface was not attempted. The reduction in deposition rate due to these considerations required many long hours of sputtering time to obtain any appreciable target thickness.

## 2.2 Calculated Sputter Yields

The sputter yield in atoms/ion was determined for the various sputtering beam/isotope combinations used to prepare the targets. The program SRIM version 2000.39 [11] was used to perform the calculations, which were carried out on a personal computer. The sputter yields were calculated using 8 kV ion beam incident at 60° with respect to the normal. The sputter target thickness varied for each of the isotopes. The thickness for the <sup>104</sup>Ru pressed pellet was 1.5 mm while for the <sup>96</sup>Ru and <sup>102</sup>Ru melted beads, the approximate thicknesses used were the sample diameters of 1.2 mm and 1.4 mm respectively.

Comparisons are shown for sputtering <sup>104</sup>Ru using both argon and xenon incident ions. Investigations were also carried out for sputtering from a pressed pellet. From density considerations, an estimated reduction in yield was expected to occur for sputtering from a pressed pellet versus the solid [12]. Table 1 gives the results of the SRIM calculations for sputter yields and also calculated deposition rates [9] using 20  $\mu$ A beam current.

## 3. Ruthenium Target Preparation

For the various experimental requirements, targets of several ruthenium isotopes were prepared on carbon, aluminum and gold back foils. The targets were mounted on standard ATLAS aluminum target frames as well as those used for GAMMASPHERE. The size of the aperture in each case was 12.5 mm diameter.

### 3.1 <sup>104</sup>Ru on Carbon, Aluminum and Gold Backings

Target preparation began using argon sputtering on a pressed pellet of <sup>nat</sup>Ru powder with the material collected on a 40  $\mu$ g/cm<sup>2</sup> carbon coated slide in addition to a carbon backing foil mounted on a standard frame. This attempt was not successful as the target ruptured and the deposited coating began to prematurely release from the glass substrate. Next, a 100  $\mu$ g/cm<sup>2</sup> foil and slide were substituted with similar results. Success was finally obtained after employing ~200  $\mu$ g/cm<sup>2</sup> carbon coated slide and mounted backing foil. For the natural material, the sputtering rate was approximately 7  $\mu$ g/cm<sup>2</sup>/hr. Next, using a pressed pellet of <sup>104</sup>Ru and a 180  $\mu$ g/cm<sup>2</sup> C backing, two backed targets of 85 and 100  $\mu$ g/cm<sup>2</sup> <sup>104</sup>Ru were obtained. The measured sputtering rate for the isotope was 4-5  $\mu$ g/cm<sup>2</sup>/hr. Thicker deposits were not attempted.

In an effort to increase target deposition, sputtering using xenon ions was carried out on a 30  $\mu$ g/cm<sup>2</sup> mounted Al backing foil. The aluminum backing foils were too thin and ruptured after a short time. Thicker (100  $\mu$ g/cm<sup>2</sup>) aluminum backings were then used for subsequent depositions. The observed sputtering rate was now approximately 10  $\mu$ g/cm<sup>2</sup>/hr. Target preparation onto gold backings of 300 and 600  $\mu$ g/cm<sup>2</sup> were also successful. Teepol (Shell Chemicals, Geismar, Ascension Parish, LA) was used as the parting agent. Several targets were made, mostly from depositions onto gold coated slides.

### 3.2 $^{96}\text{Ru}$ on Gold Backings

The ruthenium isotope was sputtered onto gold-coated slides using Xe as the sputtering ion at an accelerating potential of 8 kV. In this case, due to the small amount of  $^{96}\text{Ru}$  isotope, many hours were necessary to accumulate a target of several  $\mu\text{g}/\text{cm}^2$ . Initial depositions suggested a rate of approximately  $2 \mu\text{g}/\text{cm}^2/\text{hr}$ . Optimal source performance, especially positioning of the Xe beam on the small bead of isotope, was crucial in maintaining even this rate. Thick deposits (i.e.  $> 100 \mu\text{g}/\text{cm}^2$ ) on mounted  $300 \mu\text{g}/\text{cm}^2$  gold were not successful and ended with rupture of the backing foil, presumably due to built up stress. The gold backing foil was increased to  $500 \mu\text{g}/\text{cm}^2$  and a  $400 \mu\text{g}/\text{cm}^2$   $^{96}\text{Ru}$  target was successfully prepared but later broke upon standing after several days.

Next, in addition to the mounted  $500 \mu\text{g}/\text{cm}^2$  backing foil, a  $400 \mu\text{g}/\text{cm}^2$  gold coated slide was placed above the sputtering target to collect the ruthenium. The resulting foils on  $400 \mu\text{g}/\text{cm}^2$  Au backings were found to be highly stressed when released during floating. No attempt was made to try and anneal the films. Nevertheless, several targets of  $400\text{-}500 \mu\text{g}/\text{cm}^2$   $^{96}\text{Ru}$  were successfully prepared from this deposition.

### 3.3 $^{102}\text{Ru}$ on Gold Backings

For this target, the sputtering was done from a melted bead of  $^{102}\text{Ru}$  using a xenon beam and collected on  $300 \mu\text{g}/\text{cm}^2$  gold-coated slides. The deposition rate observed from this small sample was only  $1\text{-}2 \mu\text{g}/\text{cm}^2/\text{hr}$ . Nevertheless, several targets were successfully produced for use in a FMA/GAMMASPHERE experiment.

## 4. Results and Conclusion

In conclusion, targets made from several of the ruthenium isotopes have been prepared by our laboratory using the method of focused ion beam sputtering. Table 2 lists the various targets prepared using this technique. The compact size of the saddle-field source allows for close deposition geometry, thus conserving precious isotopic starting material. Targets of  $^{96,102,104}\text{Ru}$  have been produced by this method for use in experiments with thickness  $100\text{-}500 \mu\text{g}/\text{cm}^2$  on carbon, aluminum and gold backing foils.

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Table 1. Sputter Yields Calculated Using SRIM

Sputter Target	Thickness (mm)	Noble Gas Used	Sputter Yield (atoms/ion)	Calculated Rate ( $\mu\text{g}/\text{cm}^2/\text{hr}$ )
$^{104}\text{Ru}$ (powder)	1.5	Ar	3.09	21
$^{104}\text{Ru}$ (solid)	1.5	Ar	7.25	51
$^{104}\text{Ru}$ (powder)	1.5	Xe	5.15	36
$^{96}\text{Ru}$ (solid)	1.2	Xe	11.70	75
$^{102}\text{Ru}$ (solid)	1.4	Xe	11.10	76

Table 2. Isotopic Ruthenium Targets Prepared by Focused Ion Beam Sputtering

Target	Target Thickness ( $\mu\text{g}/\text{cm}^2$ )	Backing Foil	Backing Foil Thickness ( $\mu\text{g}/\text{cm}^2$ )
$^{96}\text{Ru}$	110	Au	300
$^{96}\text{Ru}$	468	Au	401
$^{96}\text{Ru}$	513	Au	401
$^{102}\text{Ru}$	242	Au	300
$^{102}\text{Ru}$	330	Au	300
$^{104}\text{Ru}$	85	C	168
$^{104}\text{Ru}$	100	C	177
$^{104}\text{Ru}$	73	Al	100
$^{104}\text{Ru}$	106	Au	293
$^{104}\text{Ru}$	186	Au	293
$^{104}\text{Ru}$	349	Au	293
$^{104}\text{Ru}$	132	Au	300
$^{104}\text{Ru}$	384	Au	311
$^{104}\text{Ru}$	144	Au	600

## Figures

Figure 1. Photograph of the sputtering setup within the evaporator.

