第 31 卷 增刊 I 高 能 物 理 与 核 物 理 Vol. 31, Supp. I
2007 年 7 月 HIGH ENERGY PHYSICS AND NUCLEAR PHYSICS Jul., 2007

Solid Material Development with the Argonne National Laboratory ECR Ion Sources*

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Abstract Development work with solid materials at the Argonne National Laboratory ECR ion sources has been focused on two areas – introduction of materials with low vapour pressures, and increasing the beam intensities of heavy beams (i.e. – lead and uranium). An induction oven, with a demonstrated operating temperature extending to 2000°C, has been utilized to produce a Ti-50 beam with an intensity of 5.5eμA (12+). In addition, a refinement of the sputter technique has been employed which has resulted in a 42% improvement in lead beam intensities. Details of the induction oven as well as the refined sputter technique will be presented.

Key words ECR ion source, sputter, induction oven

1 Introduction

In order to provide a wider range of beam species to the user community, as well as increased beam intensities for low cross section measurements, development work with solid materials at the Argonne National Laboratory ECR ion sources has focused on two areas – introduction of materials which have low vapour pressures (highly refractory materials), and increasing the beam intensities of heavy beams (i.e. – lead and uranium). In many cases, the low vapour pressure materials being used are isotopically enriched, with their preferred form being the metal rather than the oxide. The sputter technique has been the dominant method of material introduction[1]. It is robust, easy to use, and works with almost all solid materials. A difficulty was encountered however in the production of a Ti-50 beam. The isotopic material, when converted to metallic form, was brittle and difficult to press and its performance was rather poor. Therefore, an induction oven was developed to introduce the Ti-50 metal into the plasma via evaporation.

The sputter technique itself has an inherent limiting factor in that it requires a relatively high gas pressure in the plasma chamber for adequate sputter yield. The high pressure limits the production of the high charge states as well as intense beams of the mid charge states. To mitigate this, a technique has been developed to lower the overall pressure in the plasma chamber by introducing the working gas at the tip of the sputter sample. This has the effect of introducing the gas in the region it is most needed for a good sputter yield as opposed to diffusing throughout the entire plasma chamber.

2 Refinement of the sputter technique

The sputter technique was originally developed at ANL in 1994 and has been used extensively since that time. The method has several advantages.

1) Almost any material can be sputtered. The most demanding application has been in accelerator mass spectroscopy (AMS)[2] work where the success-

Received 20 April 2007
* Supported by the U.S. Department of Energy, Office of Nuclear Physics, contract W-31-109-ENG-38
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ful use of small sample sizes (5.0mg) and various material forms (oxides, unrefined ores, mixes of materials) has demonstrated the robustness of the technique.

2) Large beam currents can be obtained when the configuration has been optimized – 100eμA of Pb^{23+} and 39eμA of U^{26+}.

3) It is also useful for the higher charge states – 6.4eμA of Pb^{37+} and 0.25eμA of U^{46+}.

2.1 Sputter technique limitations

The technique's limiting factor, especially in the production of the high charge states, is the relatively high operating pressure required to obtain an adequate sputter yield. Typical operating pressure for the ECR2 ion source[3] is between 6.0×10^{-8} and 2.0×10^{-7} Torr measured at the plasma chamber. Such good pressure is obtained due to the open hexapole structure and the pumping radial slots afford (6.3×85mm). When operating with the sputter probe however, the pressure required for large beam currents (100eμA of Pb^{23+}) increases to 3.0×10^{-7} due to the higher sputter yield required, and this higher pressure negatively impacts overall source performance. When optimizing for high charge state production, the operating pressure is typically 1.0×10^{-7} Torr, but then the sputter yield decreases due to an insufficient amount of gas in the region of the sputter probe and large beam currents (>10eμA of Pb^{37+}) can not be achieved.

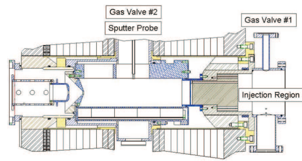

Fig. 1. Sputter probe configuration on ECR2. The gas from Valve #1 diffuses into the plasma chamber via the injection region. The gas from the Valve #2 flows into the plasma chamber directly from the sputter probe.

The central problem is that the support gas is normally introduced into the plasma chamber via the injection side of the source using gas valve #1, as shown in Fig. 1. The gas diffuses into the chamber and provides the plasma working gas, but it also affects the sputter process. And the gas pressure required for the sputter process is higher than that required by the plasma for optimum source performance. It was necessary to find a way to decouple the overall plasma chamber pressure and the sputter yield.

2.2 Introduction of working gas

In order to reduce the overall plasma chamber pressure and hence provide good plasma conditions for the intense production of high charge states, a method has been developed for introducing the sputter gas directly at the sputter probe tip using another gas valve. This has the effect of decoupling the support gas (Valve #1) and sputter gas (Valve #2) levels and providing an increased local pressure at the probe tip where it is needed for an adequate sputter yield. The gas flow is controlled via a metering valve which operates independent of the plasma support gas valve. The gas flows through a stainless steel tube which holds the sputter sample and is introduced into the plasma chamber through a 0.5mm diameter hole in the sputter sample, shown in Fig. 2.

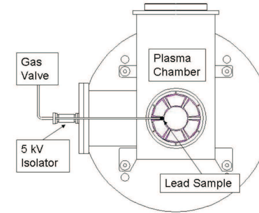

Fig. 2. Sputter probe configuration on ECR2. The 5kV isolator allows the sample to be biased. The sample has a 0.5mm hole through which the working gas flows.

For the tests the source was run with oxygen as the support gas. Oxygen was also used as the working gas introduced at the sputter probe tip. Time did not allow varying the sputter working gas in order to increase the sputter yield, but these tests are planned for the future.

2.2.1 Results

The source was run in two-frequency heating mode (14 + 10.85GHz) with the total RF power level kept constant at 500W. The extraction voltage was 14kV, and the solenoid coil currents were not varied between runs. A natural lead sample was used with the sputter voltage kept constant at −0.28kV (0.1mA drain).

The results from the two operating modes are shown in Table 1. With the gas flowing in only through the injection region (Valve #1), the total source drain current was 1.07mA with a vacuum of 1.0×10^{-7}Torr. With the gas flowing in only through the sputter sample (Valve #2), the total drain current decreased to 0.83mA, reflecting the decrease in the amount of gas within the plasma chamber, and the vacuum increased to 1.4×10^{-7}Torr in the plasma tank. This measurement is outside of the plasma chamber and at this level may not accurately reflect what is occurring within the plasma chamber itself.

Table 1. Beam intensities for ^{Nat}Pb.

charge state	intensity/eμA using valve #1	intensity/eμA using valve #2
31+	6.75	9.67
32+	6.00	8.36
33+	4.88	6.88
34+	3.95	5.59
35+	3.00	4.22
36+	2.08	3.04

As can be seen in the table, the beam currents for the highly charged ions (HCI) of lead increased on average by 42%. This demonstrates that the technique of lowering the base pressure within the plasma chamber by introducing the working gas at the sputter sample location improves the HCI production. More tests are scheduled to determine the effect on the mid-charge state production.

3 Induction oven for Ti-50 production

The introduction of highly refractory materials into the ECR plasma can be accomplished in several ways – sputter, MIVOC, laser ablation, direct insertion, and oven. All of the techniques have their advantages and disadvantages. In the case of the sputter technique which is used extensively at ANL, the difficulty arises when the sample material is isotopically enriched. The material does not always behave as the naturally occurring isotope does, as was the case with Ti-50. When the enriched material was reduced from the oxide to the metal, the resulting material was dark in color and brittle. Beam production with this sample was only 1.0eμA and unstable. This contrasts with beam production with natural titanium metal where 8.0eμA was stably produced for more than 24hours.

An attempt was made to use our standard resistively heated ovens[4], but they could not sustain the required temperature for more than 36 hours before material interactions destroyed the oven. Hence an inductively heated oven was developed for the production of a Ti-50 beam.

3.1 Induction oven design and heat shielding

The induction oven is powered by a commercial power supply available from Ameritherm Inc[5]. It is a HotShot 1.0kW model which operates in the frequency range of 150—400kHz. The induction coil is a 3.2mm diameter copper tube consisting of six turns and cooled with deionized water. The outer diameter of the coil is 24.5mm and the inner is 19mm.

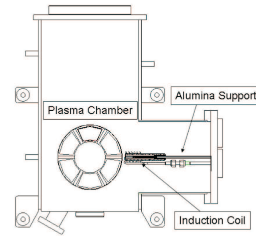

Fig. 3. Induction oven configuration on ECR2. The crucible assembly is supported by an alumina tube. The material vapor enters the plasma chamber via the radial slot.

Several crucible configurations were tested for use with the titanium. The first configuration had only a graphite crucible which acted as the susceptor. The disadvantage of this configuration was that as the temperature of the graphite increased, it became a poor susceptor to the RF. Hence at temperatures >1800℃, the power/temperature curve started to flatten. A new configuration was chosen which still had the graphite crucible but around this was mounted a tantalum tube. The tantalum, whose properties improve with temperature, then acted as the susceptor which radiatively heated the graphite crucible.

An additional consideration had to be given to the choice of crucible material due to material interactions at elevated temperatures. A tungsten crucible was first tested with poor results. The sample material melted at 1175℃, well below the melting point of

titanium. In addition, the sample material attacked the tungsten crucible and compromised the crucible integrity. Conversations with the GSI ECR group[6] revealed that the tungsten apparently reacted with a nickel contaminant in the titanium. Since the enriched material to be used had a 60ppm contaminant of nickel, the use of a tungsten crucible was not possible.

A graphite crucible could be in contact with titanium, even with the nickel contaminant, but it absorbed the material when the titanium melted at 1675℃. Since it was not desired to absorb the enriched material into the graphite crucible, a technique first developed for target making was utilized[7]. The crucible was first heated off-line with a load of natural titanium. Upon melting, the titanium was absorbed into the wall of the crucible. The process was repeated until no more material was absorbed into the graphite wall. The enriched material was then loaded and the entire assembly was placed into the ECR source.

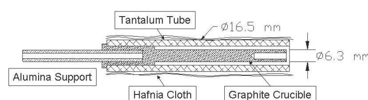

Fig. 4. Induction oven crucible configuration. The crucible assembly is supported by an alumina tube. The tantalum tube acts as the RF susceptor and radiatively heats the graphite crucible which holds the material charge. The hafnium oxide cloth protects the RF coil from direct heat.

Different heat shielding configurations were tested using alumina and graphite felts, a zirconium oxide foam, and combinations of pyrolytic boron nitride and alumina tubes. The final heat shield configuration simply consists of hafnium oxide cloth which is wrapped around the tantalum tube. It was found that this provided the same thermal protection as the other methods and had a much lower outgassing rate and a lower bulk.

In addition to the crucible heat shielding, a water cooled copper heat shield was placed around the entire induction coil assembly. This greatly reduced the amount of heat going to the chamber walls and the associated outgassing. It also served to protect the hexapole from the direct heat of the induction oven.

3.2 Oven operation and Ti-50 production

The oven has been tested to 2000℃ with an input power of 1kW (the maximum power available). Long term off-line tests had the oven at 1830℃ for three days with no performance degradation or material interactions. During the Ti-50 experiment, the oven ran for seven days at ∼1600℃.

A beam of $^{50}Ti^{12+}$ (the peak of the charge state distribution) was produced for seven days with an intensity of 5.5eμA at the source and 2.5eμA reaching target. The consumption rate was 0.70mg/hr with some of this material plating out onto the heat shield and the radial port.

4 Conclusion

Due to the ever increasing demand for beams produced from highly refractory or exotic elements, the development work at ANL has focused on the introduction of these materials into the ECR plasma. An RF induction oven has been successfully used for the production of a 5.5eμA beam of $^{50}Ti^{12+}$ for a period of seven days with no degradation in the oven materials.

The sputter technique, which has been in use at ANL for more than a decade, continues to be refined. The addition of a gas feed at the sputter sample has reduced the amount of support gas required for stable beam production. Due to this reduction in source operating pressure, higher intensities of the HCI are available.

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