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PRELIMINARY EVALUATION OF A MODIFIED MUELLER-HORTIG GEOMETRY NEGATIVE ION SOURCE USING A NEGATIVE ION SOURCE TEST FACILITY

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Summary

A negative sputter-type ion source of the Mueller-Hortig geometry is described which utilizes a positive surface ionization source. Among the ion beams which have been produced are: Al⁻0.2 μA; AlO⁻3 μA; Au⁻6 μA; C⁻25 μA; C₂⁻20 μA; Cl⁻100 μA; Cu⁻0.5 μA; CuO⁻0.8 μA; F⁻40 μA; I⁻26 μA; O⁻30 μA; Pt⁻3 μA; S⁻44 μA; TaO₂⁻2 μA. The yields of Cl⁻, F⁻, O⁻, and S⁻ produced from gaseous or high vapor pressure compounds are observed to be sensitively dependent on the metallic surface from which they are generated - indicating an intermediate surface chemical effect. While the source produces very intense beams of the halogens, sulfur, and oxygen and relatively intense beams of the metallic oxides, there appears to be a need for additional surface cesium to ensure maximum negative ion yields from many of the elemental materials.

Introduction

Since the discovery by Krohn,¹ in 1962, that yields of negative ion are greatly enhanced by sputtering in the presence of alkali metals, several negative ion sources, utilizing the principal, have been developed. The source of Mueller and Hortig was the first of this type,² followed by the very versatile version of Middleton and Adams.³ In recent months, several other sources have been developed - some of which are described in these proceedings.⁴⁻⁷

The original ion source of Mueller and Hortig clearly demonstrated the wide range of ions and high intensity capabilities of a source which incorporates sputtering and simultaneous cesium surface activation. However, the design, while possessing the same potential, lacked the flexibility, versatility, and long lifetime demonstrated by the later Middleton-Adams³ source. Among the aspects which affected the overall performance of the source were:

- 1) The length of time for continuously uninterrupted operation was limited principally by the cathode lifetime in the plasma discharge source (~ 40-80 hrs).
- 2) The number of negative ion species which could be produced during an uninterrupted operational period was limited by the fact that only one sample could be bombarded during this period. This sample was also quite large and relatively expensive to fabricate.
- 3) Because of the oblique angle of incidence of the positive ion beam with respect to the sample surface, the bombarded area is elliptical in shape and quite large. (A circularly symmetric beam, incident at 10° with respect to the sample surface, would project an image perpendicular to the bombarded surface with major axis 5.67 times that of the minor axis.)

Aside from the features of the source which appeared to need improvement, the source geometry has

certain desirable features - some of which are listed below:

- 1) Negative ions are generated over a planar area - permitting the design of an ion extraction system with good optical properties.
- 2) Under the assumption that sputtered particles leave the sample surface in a cosine distribution about the normal to the surface,⁸ one can argue qualitatively that the emittance of the ion source might be lower than that of the conventional conical geometry because of the direction in which sputtered particles leave the sample surfaces relative to the direction of negative ion extraction. (As a counter argument, the Mueller-Hortig geometry may accept a wider range of particle energies than the conventional Middleton-Adams³ geometry which acts somewhat as an energy selector, according to computer simulation studies made at ORNL.⁹)

With the indicated considerations in mind, an ion source of the Mueller-Hortig geometry was constructed and evaluated. The source, which will be described later, was evaluated using a negative ion source test facility. For this reason, the test facility will be described first.

The Negative Ion Source Test Facility

A test facility has been recently constructed which will provide a basis for future negative ion source research and development and as a facility for studying ion source related atomic-physics phenomena at ORNL. Some of its important aspects are described below.

The facility, shown in Fig. 1, consists of four principal components; an ion source modular assembly, a cylindrical lens assembly, a 90° analyzing magnet, and an ion collection chamber.

Any one of a variety of ion sources may be mounted in the modular ion source assembly (1). Extracted ions are focused through a post acceleration tube (2) onto an aperture (3) which serves as the object for the mass analyzer (6) by means of internal lensing. The aperture is adjustable in the horizontal and vertical directions thus permitting variation of the resolution of the device and measurement of the ion current distribution in each of the indicated directions. The beam then enters a cylindrical lens which compresses the beam in the vertical direction so as to maximize

*Experimental investigations show that sputtered particle exhibit, more or less, a cosine distribution about the sample normal.^{7,8} In actuality the distribution becomes more and more asymmetrical with respect to the normal as the direction of the incident particle becomes more and more oblique. This is particularly true for projectiles with masses greater than that of the target material.

*Operated by Union Carbide Corp. for the U.S. ERDA.

Fig. 1 - The ORNL Negative Ion Source Test Facility.

the transmission through the 1.59 cm gap of the magnet chamber (7). A Faraday cup (13), which is located in the cylindrical lens chamber (4), can be inserted into and withdrawn from the ion beam, and is used to determine the transmission efficiency of the system. The beam is then momentum analyzed with a 90° magnet (6), having an effective bending radius of 76 cm, and is monitored in a conventional Faraday cup, located in the collection chamber (10).

The facility has a maximum energy capability of 150 keV for singly charged particles and a resolution of > 200 -depending on the magnification or aperture setting. The full acceptance in the limiting direction of the magnet chamber is $\sim 14\pi$ cm-mrad.

Description of the Source

The ion source, shown schematically in Fig. 2, utilizes a surface ionization source to produce a 1-8 mA beam of C_s^+ ions. The ions are accelerated to energies between 20 and 30 keV and focused onto the sample surface by means of an einzel lens which will be described later. The incorporation of the surface ionization source extends the continuous operational lifetime to > 500 hours. Details of the surface ionization source will not be presented here because of its similarity to the one used in the conventional Middleton-Adams source.³

The einzel lens, used to focus the ion beam onto the sample surface, is designed to compress the beam more strongly in the horizontal than in the vertical direction in order to partially compensate for the oblique angle of incident (10°) and produce a more circular beam image as viewed along the negative ion extraction axis. (Wear patterns on the negative ion extraction electrode system indicate that the beam

shape is almost circular having a horizontal width of ~ 1.2 times the vertical height.)

Fig. 2 - The ORNL Sputter Source

The sample wheel houses up to 18, 3.2 cm diameter samples which are located circularly around the wheel. An external indexing mechanism permits selection of a particular sample for bombardment. The plane of the sample can also be moved in and out in order to compensate for sample wear, misalignment, or lens steering of the positive ion beam. The preliminary results from the source were obtained without coolant on the sample wheel which may be important for maximum retention of surface cesium; i.e., the equilibrium temperature of the sample under bombardment may be high enough to diffuse and evaporate cesium from the sample surface which could decrease the negative ion yields. In the future, coolant will be provided so that its effects can be evaluated.

The Negative Ion Extraction System

A gridded electrode, biased 1-1.5 kV positive with respect to the sample surface, is used to initially extract the negative ions. The electrode has a spherical radius of 3.8 cm with a circular aperture of 2.5 cm diameter. The ions are further accelerated by an anode (accelerating electrode) at housing potential. The negative ions converge to a focus at ~ 10 cm beyond the gridded electrode.

The special einzel lens and negative ion extraction system are preliminary designs which were determined by use of the Electron Trajectory Program written by W. B. Herrmannsfeldt.¹⁰ Figure 3 shows the trajectories of positive and negative ion beams in the source as calculated with the indicated computer program. The negative ions are assumed to be generated uniformly over the surface irradiated by the positive ion beam and leave the surface in a cosine distribution about the sample normal with a most probable energy of 2 eV. (This energy was arbitrarily chosen and is perhaps a factor of 2.5 too low based on experimental results.)^{11,12} Such computer simulation studies are extremely useful in predicting the emittances of ion sources whenever the intrinsic velocity distribution of the species can be realistically incorporated. Aberrations due to the geometry of the ion generation surface and space charge effects can be included as well.

Fig. 3 - Computer Simulation of the Positive and Negative Ion Extraction Systems.

Source Operational Parameters

The source, thus far, has exhibited excellent operational characteristics - requiring very little attention after thermal equilibrium has been established. The ionizer is heated by means of an annular cross section cylindrical heater which surrounds the ionizing end of the cesium transport tube and operates at about 1000-1100°C. The cesium oven is independently heated and is typically operated at ~ 230°C.

Typical curves of negative ion current as a function of cesium oven temperature are shown in Fig. 4. We note that the current reaches a maximum at a particular voltage - followed by a plateau region. The yield increases as a function of increasing extraction potential, principally, because the positive ion current obeys the $V^{3/2}$ law under space charge limited conditions.

Fig. 4 - Relative Negative Yields as a Function of Cesium Oven Temperature.

The dependence of the negative ion current on positive ion lens voltage is shown in Fig. 5. As anticipated, the current is rather strongly dependent on whether the positive ion beam is well focused on the sample surface or not. The maximum in the curve shifts towards higher lens voltages as the positive ion current is increased - illustrating the effect of space charge forces on the positive ion beam.

Fig. 5 - Dependence of Negative Ion Yield on Positive Ion Lens Voltage.

Emittance Estimations

From the previously indicated computer simulation studies, a full emittance of 10π cm-mrad was estimated for the source. These computations provided the starting parameters (beam size and angular divergence) subsequently used in first order transport theory calculations of the beam envelope through the test facility. These calculations indicate, even under optimum conditions, that the system will only transmit, without clipping, ~ 4-5 π cm-mrad of the beam in the limited acceptance direction of the magnet.

Figure 6 shows the horizontal and vertical current distributions of the transmitted beam as measured at the object point of the analyzer for two positive ion lens voltages. We note that the horizontal current distribution is displaced as the lens voltage is increased, indicating lens steering. The effect can be compensated by moving the plane of the sample appropriately. We also note that the vertical current distribution, which represents the transmitted beam through the limited acceptance direction, does not appear to be significantly clipped by the magnet chamber.

Beam transmission measurements, made for carbon, indicate that at least 90% of the beam is transmitted through the system. These measurements, coupled with the computer simulation studies and first order transport theory calculations seem to indicate that the source may have a full emittance of $< 10\pi$ cm-mrad for the vertical plane. It must be pointed out that due to the asymmetry of the source, the two planes may have different emittances.

Fig. 6 - Transmitted Negative Ion Beam Profiles Measured at the Object Point of the Mass

Negative Ion Yields

A partial list of the negative ion species which have been produced in the source to date is shown in Table I. As can be readily seen, the source is a good producer of molecular negative ions such as AlO^- , C_2^- , CuO^- , FeO^- , PbO_2^- , TaO_2^- , etc., and an excellent producer of atomic negative ions from high vapor pressure compounds which contain the halogens, sulfur or oxygen. However, the yields from solid elemental materials appear to be somewhat lower than the original Middleton-Adams source with the possible exceptions of Al^- , C^- , and Cu^- .

The source is equipped with a gas feed system from which high vapor pressure materials can be fed into the source at a controlled rate by means of a standard mechanical leak valve. The sample surface, onto which the material is fed, may be selected by means of the previously indicated indexing mechanism. The effect of the substrate on the ion generation rates of Cl^- , F^- , I^- , O^- , and S^- is shown in Table II. The yield of a particular negative ion is seen to be dependent on the chemical properties of the substrate - indicating that an intermediate chemical effect plays a role in the rate of negative ion generation for these materials.

Table II. Influence of the positive ion bombardment surface on the rate of negative ion generation produced from high vapor pressure materials.

Ion	Ion Current (μA)	Ion	Ion Current (μA)
Al^-	0.2	I^-	26
AlO^-	3	Ni^-	~ 0.2
Ag^-	.250	NiO^-	~ 0.3
$^*\text{Ag}^-$	1.2	Pb^-	.03
Au^-	6	PbO_2^-	0.48
C^-	25	Pt^-	3.0
C_2^-	20	S^-	44
Cl^-	100	Ta^-	.03
Cu^-	0.5	TaO_2^-	2.0
CuO^-	0.8	TaO_3^-	.6
F^-	40	Ti^-	.04
Fe^-	0.1	TiO^-	0.200
FeO^-	0.8	TiO_2^-	.07

*Sintered Ag powder saturated with C_2 .

Discussion

If a comparison of the negative ion yields from C^- , Cu^- , Ag^- , and Pb^- (elements possessing approximately the same electron affinities (~ 1.2 eV)) and their relative saturation values for materials such as cesium (see Ref. 13), we note an increase in negative ion yield with saturation value. This may imply that the lower yield materials are cesium limited and that additional surface cesium must be added in order to realize the maximum negative ion currents from low saturation value materials.

The work of Abdullayeva, Ayukhanov and Shamsiyev¹⁴ clearly demonstrated that negative ion yields can be greatly enhanced by simultaneously evaporating an optimum thickness of an alkali metal (~ 1 monolayer) during the sputtering process. Since sputtered particles are generated in the first 3 or 4 atomic layers of the material, the evaporation rate of the additional alkali metal must be precisely controlled, otherwise the alkali metal will be sputtered predominantly. Although the simplicity of the source operation and stability may

Table I
A partial list of negative ions generated in the ORNL sputter source

Ion Species	Feed Material	Ion Generation Surface									
		Al	Ag	Au	C	Cu	Fe	Ni	Pb	Ta	Ti
		Ion Current (μA)									
Cl^-	CCl_4	100	57	24	27	40	40	30	11	42	40
F^-	SF_6	25	6	5	8.5	15	17	12	9	35	40
I^-	CH_3I	26	7	4.8	16	13	13	15	10	4	23
O^-	O_2	30	1.5	3.5	7	7	20	15	3	10	20
S^-	CS_2	44	4.7	2.3	20	16	19	13	2.1	18	35

Influence of the Positive Ion Impact Surface on the Rate of Negative Ion Generation.

be compromised somewhat, plans are being made to incorporate a thermal evaporation oven in the present source in an effort to increase the negative ion yields from low producers.

The addition of coolant to the sample holder is also planned in the near future. In the present source, the samples were not directly or indirectly cooled during bombardment which may have lead to sample temperatures sufficiently high to thermally diffuse and evaporate surface cesium, decreasing the amount of cesium available for electron transfer to the sputtered particles or for affecting the work function of the composite material. The use of coolant on the sample holder in the Middleton-Adams source is considered to be very important¹⁵ and the addition of coolant is expected to affect the negative ion yields from the present source similarly.

Conclusion

The source, just described, is a very simple and versatile producer of many negative ions as evidenced by the yield data and has a potential lifetime of > 500 hours. Although the source has been tested over a very limited time period, it has performed very stably and requires very little operator interaction after equilibrium has been established. With the incorporation of coolant and possibly, provisions for evaporating additional alkali metal on the generation surface, the source is expected to produce even more intense beams of negative ions.

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