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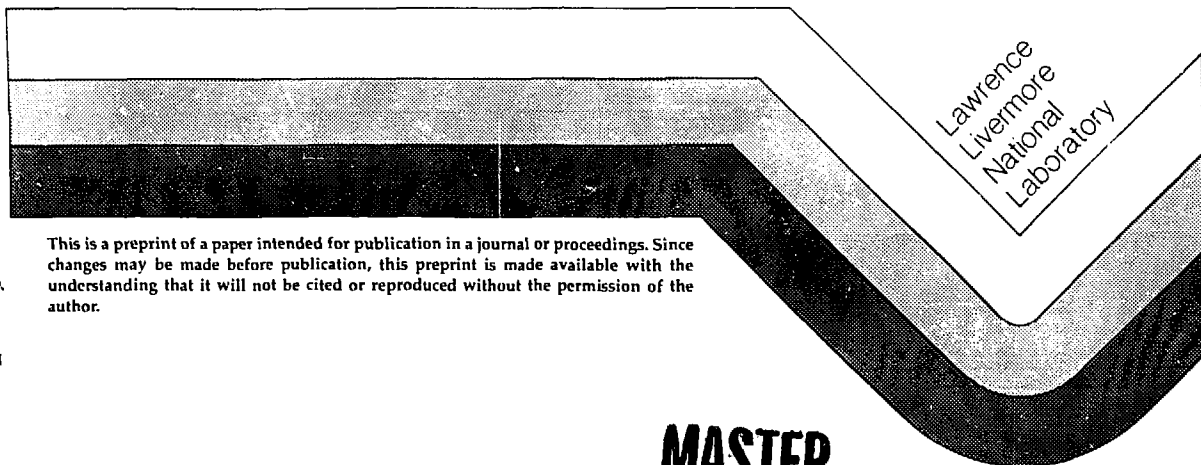
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ABSTRACT

A technique for preparing selenium films onto 50.8 microns thick beryllium foils is described. The selenium was deposited in vacuum from a resistance heated evaporation source. Profilometry measurements of the coatings indicate deposit thicknesses of 5.5, 12.9, 37.5, 49.8 and 74.5 microns. The control of deposition rate and of coating thickness was facilitated using a commercially available closed-loop programmable thin film controller. The x-ray transmission of the coated substrates was measured using a tritiated zirconium source. The transmissivities of the film/substrate combination are presented for the range of energies from 4 to 20 keV.

## I. INTRODUCTION

The interest and demand for both free-standing or coated foils for use in nuclear physics experiments have encouraged the development of numerous preparation techniques. A variety of methods have been used to deposit pure or alloyed films of selenium: Jansen (1) developed a novel apparatus fashioned after the flash evaporation technique which allows the positive control of the deposition rate of low melting point (As/Sb/Se) (alloys; Johnson and Schlie (2) deposited thin films of zinc, selenium, and non-stoichiometric ZnSe by photodissociation with UV energies; and even electrochemical deposition methods(3) have been applied. Physical vapor deposition methods such as reactive sputtering (4) and evaporation from electron beam (5) and resistance heated (6-8) sources have also been used with good results.

Because of its inherent simplicity, the resistance heated source was selected to produce selenium coatings on beryllium substrates. Early test runs had indicated that, although the quality of the coatings was adequate, the integration of closed-loop deposition controller would be advantageous. Selenium has a relatively high vapor pressure (0.1 Torr @ 277 C)<sup>9</sup>, is rather toxic. It was desirable to control the amount of material introduced to the vacuum system. To minimize the coating of chamber walls and to reduce the possible contamination of the vacuum system, the evaporation source and substrates were enclosed within a water-cooled containment fixture.

## II. EQUIPMENT AND PROCEDURE

The evaporator which was used to deposit selenium coatings consisted of a LN<sub>2</sub> trapped oil diffusion pumped vacuum system (Figure 1). The walls of the stainless steel chamber were protected from the deposition of selenium by the use of a water-cooled enclosure. The evaporation source, the substrates, a shutter, and the deposition monitor are located within the enclosure.

A resistance heated tantalum source was used to evaporate the selenium. The source was of an open-boat design and had sufficient capacity to allow the deposition of 50 micron thick films when fully charged with 100 grams of high purity selenium shot.<sup>(10)</sup> The source temperature was monitored with an intrinsic chromel-alumel thermocouple, which was spot-welded to the bottom of the boat. A programmable deposition controller <sup>(11)</sup> was used to control the source power supply in a closed-loop control of preheat time and temperature, deposition rate and deposited thickness.

The substrates, in this case beryllium sheet material approximately 3 cm diameter and 50.8 microns thick, were attached to an aluminum holder. The holder was in turn clamped to the water-cooled lid of the containment enclosure. The source to substrate distance has about 37 cm and the substrates were arranged within a 15 cm circle. Although the temperature of the substrates was not monitored during deposition the estimated maximum temperature is on the order of 40<sup>0</sup> C. No substrate motion was used during the deposition. However, because many of the

coatings were too thick to be deposited in a single coating run, the coating uniformity was improved by manually rotating the substrates while the system was at atmosphere and the evaporation source was being reloaded. The amount of rotation depended upon the number of coating runs which were required (i.e. 180 degrees for 2 runs, 120 degrees for 3 runs, etc.)

A shutter was used to protect the substrates and the quartz crystal monitor from contamination during the initial heating and degassing of the source material. The shutter was actuated by the deposition controller; opening after a timed preheat and closing after reaching the desired deposit thickness. The measured temperature of the selenium source during a typical coating run is shown in Figure 2.

The coating thickness deposited during each of the runs was determined using stylus profilometry.<sup>(12)</sup> Polished glass slides were included in every coating run and were used as witness pieces. The thickness of the coating deposited on the glass slide was measurable and was representative of the coatings deposited on the beryllium substrates.

Two other techniques were used to verify the total thickness of the selenium coatings: gravimetric and computation of x-ray density. The first technique, the measurement of the weight gain of a given substrate due to the deposition of a coating over a known and calculable area, depends upon the assumption of bulk density in order to estimate the coating thickness. The second technique also requires the assumption of certain bulk properties such as density and x-ray cross section. The

latter technique is a by-product of the x-ray measurements and calibrations which were performed on each of the coated foils.

### III. RESULTS AND DISCUSSION

The use of the controlled deposition rate technique proved to be a major improvement over previously used methods. Prior to integrating the closed-loop control system, the deposition method depended upon the complete evaporation of pre-weighed charges of evaporant. In addition to the lack of real time monitoring of coating thickness, the major drawback of this method was the inability to properly degas the evaporant prior to the start of the deposition cycle. In applying the closed-loop control system, we no longer had to use pre-weighed charges, the melting and degassing could proceed as required and the coating process could be terminated upon reaching a desired (and measurable) thickness. The nominal deposition rate used for these coatings was on the order of 200 to 210 Å/second to a maximum thickness of about 35 microns during any one run.

The use of the water-cooled enclosure as a means of reducing the coating of the vacuum chamber walls was also successful. During nearly 20 coating runs, many of which required the deposition of relatively thick selenium coatings, the chamber walls remained clean and the vacuum pumping system was not affected by the deposited material. Pump down times and base pressures were substantially the same regardless of the thickness of the coatings which were deposited on the inner walls of the enclosure.

The manual rotation of the substrates between coating runs and the favorable source-to-substrate distance apparently combined to good advantage. The coating uniformity, measured by a nonquantitative beta-backscattering method, (13) was on the order of +5 percent of the nominal coating thickness.

Comparison of the results of stylus profilometry and gravimetric measurements indicated that the density of the coatings was about 93 percent of the bulk density of vitreous selenium. However, the computation of the coating density by means of the x-ray transmission and photo-interaction cross sections indicated that the coating densities were more on the order of 90% of bulk. While it is not entirely clear at the present time just which measurement is most correct, the vacuum deposition of thin films can often result in coatings which are of less than bulk density. While we are interested in resolving the disparity in measurements, in our present application of these coatings the most important data is the transmission of x-ray energies through the selenium-beryllium coating-substrate combination.

The x-ray transmission of the coated substrates was measured over the range of 3 keV to about 20 keV using a tritiated zirconium source. (14 and 15) Since the beryllium substrates are essentially transparent to x-rays over this energy range, the transmission values are that of the selenium coatings. Each of the coated foils was calibrated thusly, providing the experimenter with the particular characteristics of every coating. The percentage x-ray transmission as a function of incident energy for five different thicknesses of selenium coatings is shown in Figure 3.



#### IV. SUMMARY

The integration of a simple, "Tried and True" method of deposition with a modern, closed-loop thin film control system has been illustrated. The use of a water-cooled enclosure during the deposition of a high vapor pressure material such as selenium has proved effective in minimizing deleterious effects upon the vacuum coating system. The use of the enclosure also reduced maintenance and system cleanup requirements. The same scheme might be expanded to include other applications. The thick selenium films produced under these conditions were quite uniform and had densities approaching those of bulk material. The x-ray transmission of selenium coated beryllium foils was measured over the range of energies from 3 keV to 15 keV.

#### V. ACKNOWLEDGEMENTS

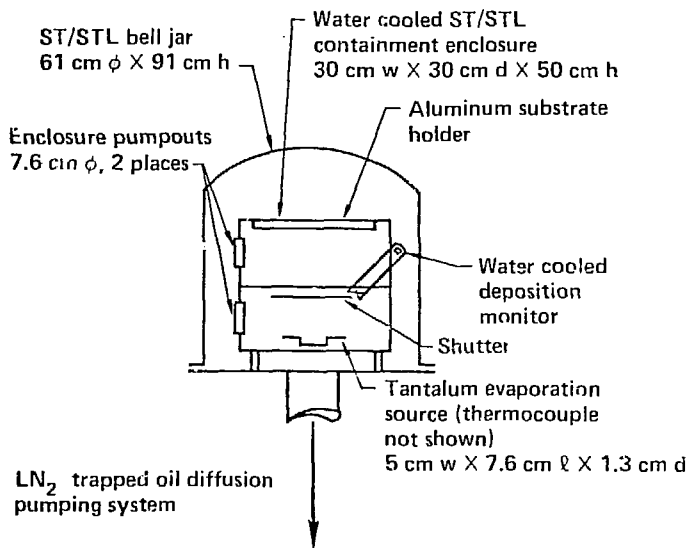
This work was performed under the auspices of the U. S. Department of Energy by Lawrence Livermore National Laboratory under Contract Number W-7405-ENG-48. We would like to extend our thanks to Geana Hattery and Bill Junk for information concerning the use and test results of the coated foils, to Jack Button and Jerry Gaines of the x-ray Calibration and Standards Laboratory for the measurements and graphs of the foil transmissivity and to physicists Tom Harper, Ted Strand and Jim Woaser for their assistance in the interpretation of the measurements as well as discussions concerning the applications of such foils in nuclear research.

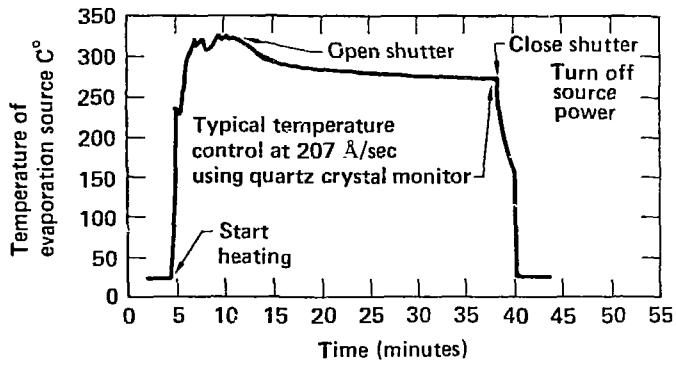
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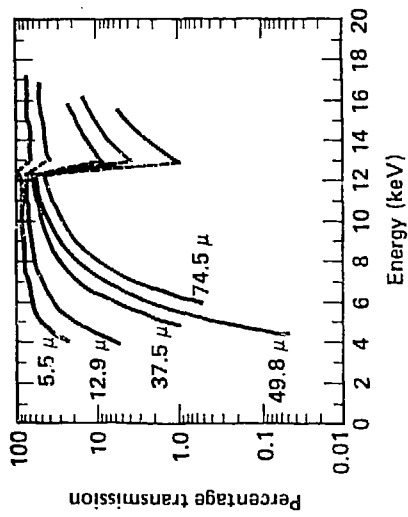
FIGURE 1 A schematic diagram of the evaporation system (and the containment enclosure) which was used to deposit selenium coatings.

FIGURE 2 A graph of the temperature of the evaporation source as a function of time during a typical closed-loop coating cycle at an average deposition rate of 200-210  $\text{A}^0/\text{second}$ .

FIGURE 3 A graph of the measured x-ray transmission of selenium coated beryllium foils as a function of incident energy. The substrates were approximately 50.8 microns thick and the thickness of the selenium coatings is as indicated.







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14. For further information concerning LLNL's x-ray measurement capabilities, contact:

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