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Abstract

A double-windowed vacuum-tight experimental chamber was developed, and calibrated on the spectroscopic ellipsometer over the energy range from .7 to 4.5 eV using a silicon wafer with approximately 25 nm oxide thickness to remove the multiple-window effects from measurements. The ellipsometric measurements were done such that incident and exit beam were at 65 degree from surface normal. The plutonium sample (3 mm diameter, .1 mm thick) was electro-polished and mounted into the sample chamber in a glove box having a nitrogen atmosphere with less than 100ppm moisture and oxygen content. The index of refraction n and the extinction coefficient k decrease from 3.7 to 1 and 5.5 to 1.1 respectively as the photon energy increases from .7 to 4.3 eV.

Key words

Plutonium, uranium, optical constants, n, k, ellipsometry

Introduction

The optical constants of plutonium metal are of scientific and practical interest, but not readily available. They are difficult to measure, because only small quantities can be handled since the material is highly radioactive. Moreover, safety regulations require that plutonium be doubly contained at all times.

Experimental

Design of the experimental chamber.

The double-windowed experimental chamber has to fulfill two requirements: Safety regulations necessitate that all openings have two independent seals, and reproducibility of ellipsometry demands that the entering and exiting photon beams are always normal to the glass surfaces, and pass them always at the same spot. The second requirement can only be fulfilled if the chamber can be fitted onto the ellipsometer's working platform into exactly the same position and orientation. Figure 1 shows an exploded view of the chamber that fulfills both requirements and was used in our experiments. Its body is machined from a solid aluminum block. It has 3 openings for windows, and one to introduce the sample. The first set of windows in each opening is sealed to the body with an epoxy known to have good vacuum properties; the second set of windows is sealed with O-rings. The windows are arranged such that the photon beam can enter or exit either normal to the target, or at an angle of 65° with respect to the target's normal. The angle of 65° was chosen to assure that the majority of the photon beam would interact with a surface of 3 mm diameter. Great care is taken to assure that all windows are seated flat on the machined surfaces and are subjected to minimal stress. The sample holder is

sealed to the bloc using an O-ring. It is machined such that it fits tightly into the bloc and that the target surface is located exactly at the intersection of the centerlines of the three windows. At the back of the sample holder is a circular hole. The second flange following the sample holder is again sealed to the bloc with an O-ring, and has alignment pins on both sides, and a recess on the backside. The alignment pin on the front side fits tightly into the hole at the sample holder, and the alignment pin on the backside fits tightly into a centering hole on the ellipsometer's work platform. The platform has two additional holes connected to a vacuum pump and concentric with that centering hole; they evacuate the recess on the backside and hold the experimental chamber securely but allow rotation of the chamber so that the flat faces of the aluminum block can be aligned parallel to the ellipsometer's work platform. All windows consisted of UV-grade fused silica.

Calibration of the chamber to eliminate window effects.

During an experiment at 65° angle of incidence the vector representing the photon beam is affected not only by the sample itself, but also by passage through 4 windows. To determine the window effects one measures over the photon energy range of interest the ellipsometric response of a small silicon wafer sample having a surface oxide layer with a thickness of ~25 nm mounted first in air and then in the experimental chamber. From these different responses a transfer function is derived that is then applied to correct the response from unknown samples. The ellipsometer and the software used for all calculations were supplied by J.A. Woolam Co. Inc. Figure 2 shows the values of Δ and Ψ measured in air fitted to a model consisting of bulk silicon, a 1 nm thick Si/SiO_2

interface layer, and a silicon dioxide layer of adjustable thickness. The best fit gives a SiO_2 thickness of 25.5 nm. Figure 3 shows the values of Δ and Ψ after the window correction was applied obtained on the same sample mounted in the chamber. These values were again fitted to the same model. The best fit gives a SiO_2 thickness of 25.8 nm. Figure 2 and 3 demonstrate that the window correction function eliminates the window effects with sufficient accuracy.

Plutonium sample characterization and preparation

The material used was gallium stabilized delta phase plutonium. The sample started out as a disk of 3 mm diameter with a thickness of less than 1 mm. The sample was mounted on a lapping fixture with low temperature melting wax. The first side was lapped until a thickness ≈ 200µm was reached using 30, 12, 3 µm abrasive paper, and finished with 1µm Al_2O_3 lapping paper. The sample was un-mounted, turned over and again lapped with the same succession of lapping films to a 1 µm finish until the thickness of the disc-shaped sample was 101µm +/- 1µm thick. The sample was annealed on a hot plate for 10 minutes @ 150 C in order to revert any alpha phase that may have formed from the mechanical polishing. The sample was then electro-polished for 5 seconds, removing ≈2µm of metal leaving a surface that appeared highly polished to the naked eye. The sample roughness is estimated to be less then 5µm and the parallelism to 10µm over 3 mm. The electro-polishing solution is 10% Nitric acid, 45% Ethanol, 45% Butoxyethanol, used at -12 degrees C, and the polishing voltage is 135V. Under these conditions the typical removal rate is ≈ 0.3µm per second. All work done in a N2 glove box with O2 and H2O below 100 PPM. The sample was glued onto the experimental

chamber's sample holder and sealed in said chamber inside the glove box. The sample was analyzed on the ellipsometer after approximately 20 hours. Transmission electron microscopy on thin samples prepared in this manner and analyzed after having been held for a day in the glove box atmosphere show an oxide thickness of a few nm.

Modeling of experimental data

The material was analyzed in the ellipsometer after mechanical and electrochemical polishing, and had minimal surface roughness, and no surface layers. It was therefore modeled as a bulk metal.

Results

The measured Δ and Ψ (corrected for window effects) were fitted to a metal model. The ellipsometer's software was used to derive the optical constants n and k that are shown in figure 4 together with the model fit.

Discussion

Cerium and uranium are frequently used for comparison with plutonium. Cerium metal data are not available. The optical constants of uranium metal have been measured with the same instrument, albeit over a smaller range of energies and are depicted in figure 5. Comparison of figure 4 and 5 shows that plutonium has at higher photon energies a lower extinction coefficient and index of refraction. The plutonium data acquired here will be

used to model the ellipsometric response of an oxide covered plutonium surface and to determine the optical constants of surface oxides of plutonium.

Acknowledgements

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Figure Captions

Figure 1. Assembly drawing of double-windowed ellipsometer chamber for plutonium analysis.

Figure 2. Experimental data Δ and Ψ measured in air on a $1cm^2$ oxidized silicon wafer. A model consisting of bulk silicon, a 1 nm thick Si/SiO_2 interface layer and a SiO_2 layer of adjustable thickness provides the best fit at a SiO_2 thickness of 25.5 nm.

Figure 3**.** Experimental data Δ and Ψ measured on a $1cm^2$ oxidized silicon wafer mounted in the experimental chamber after correcting for window effects. A model consisting of bulk silicon, a 1 nm thick Si/SiO_2 interface layer and a SiO_2 layer of adjustable thickness yields the best fit at a SiO_2 thickness of 25.8 nm.

Figure 4. Index of diffraction n and extinction coefficient k of plutonium metal with an oxide thickness of a few nm.

Figure 5. Index of diffraction n and extinction coefficient k of uranium metal with an oxide thickness of a few nm.

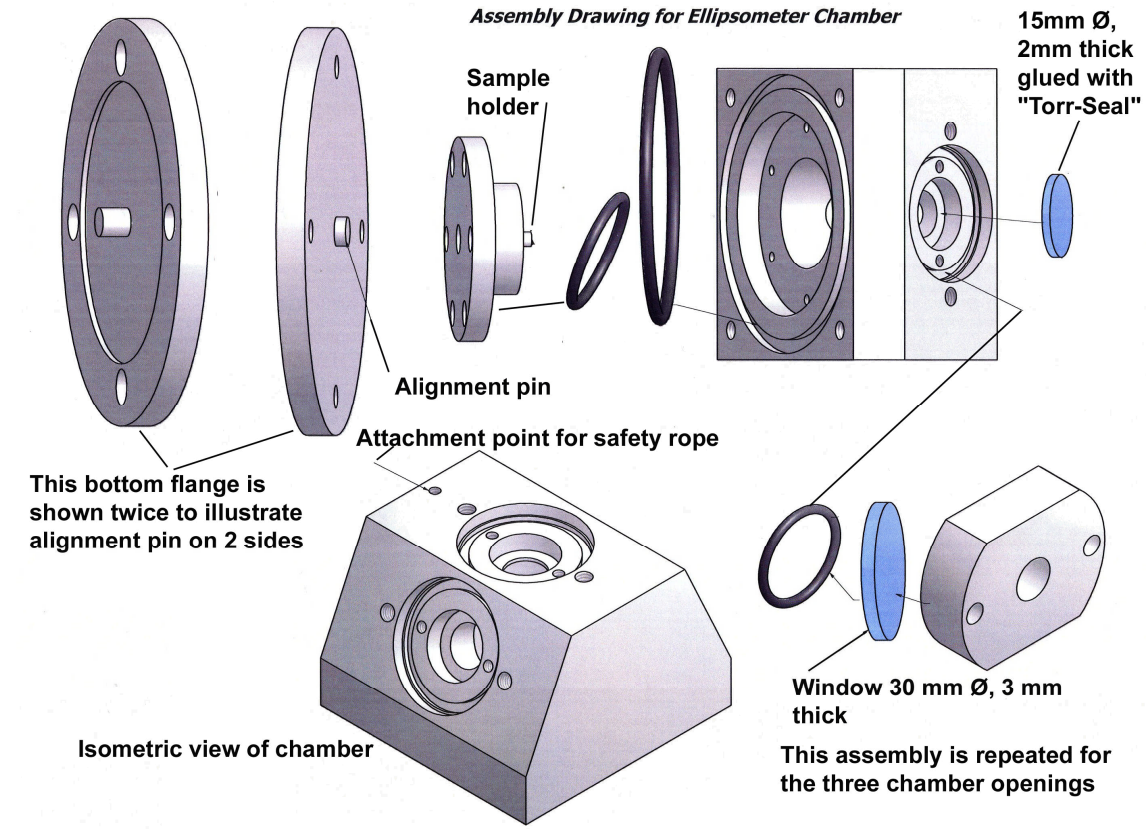

Figure 1.

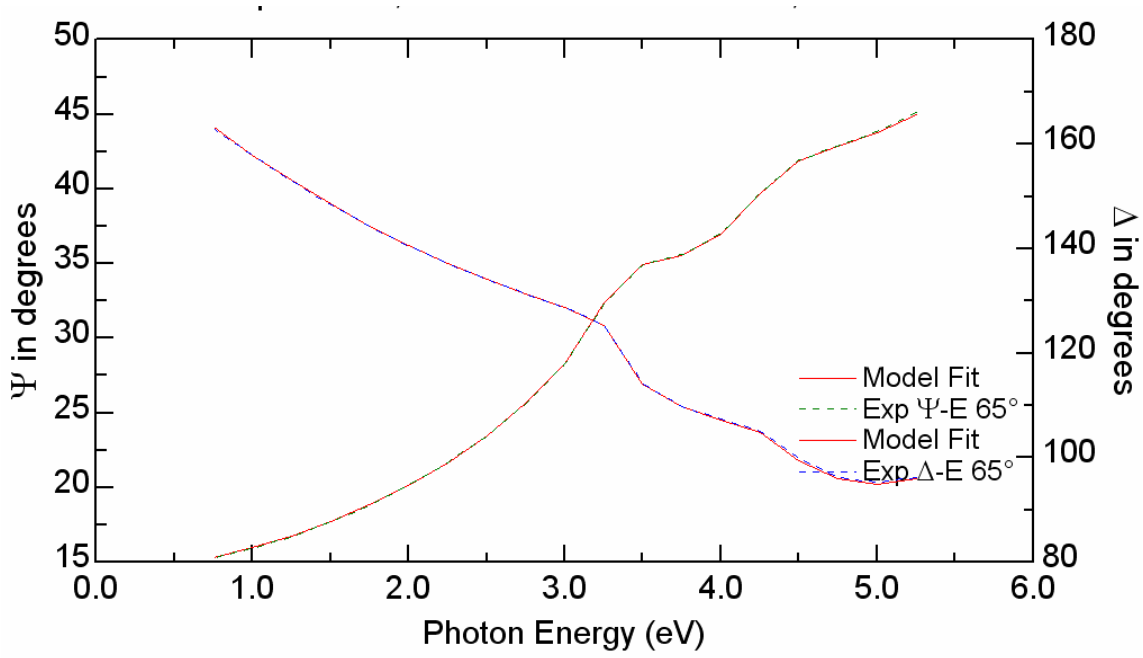

Figure 2.

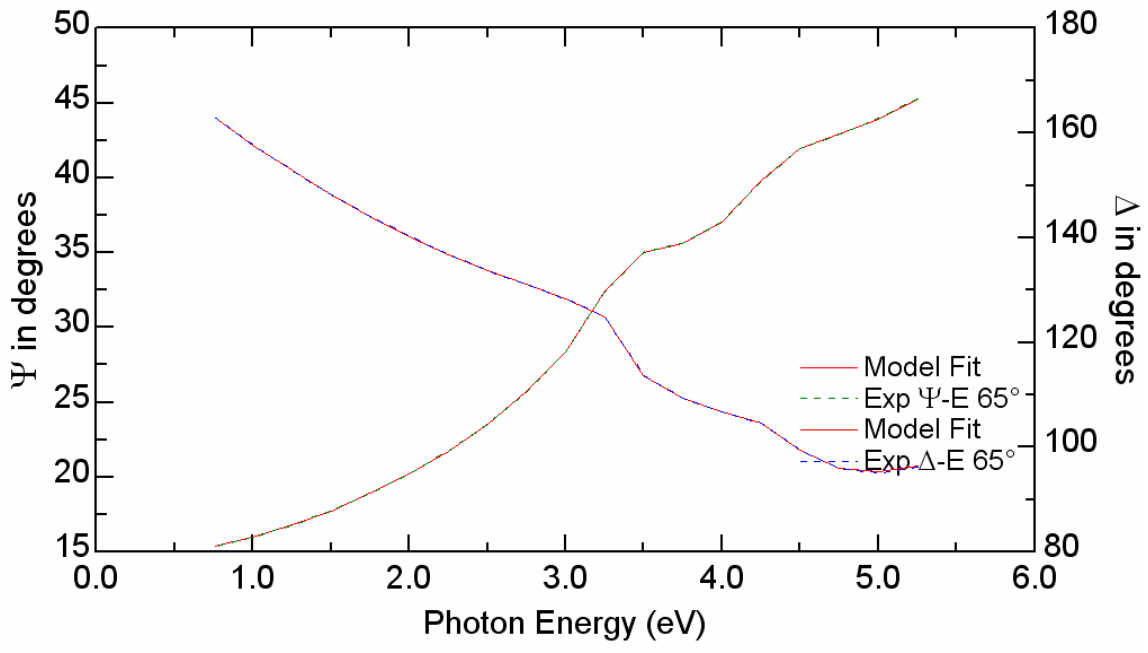

Figure 3.

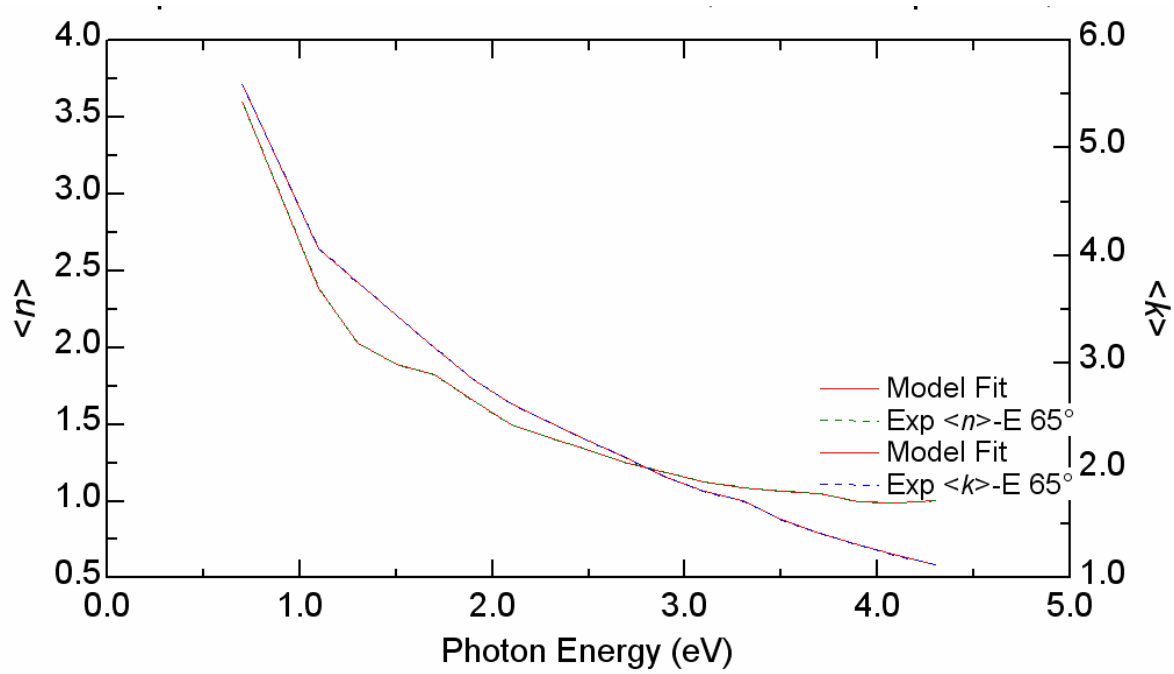

Figure 4.

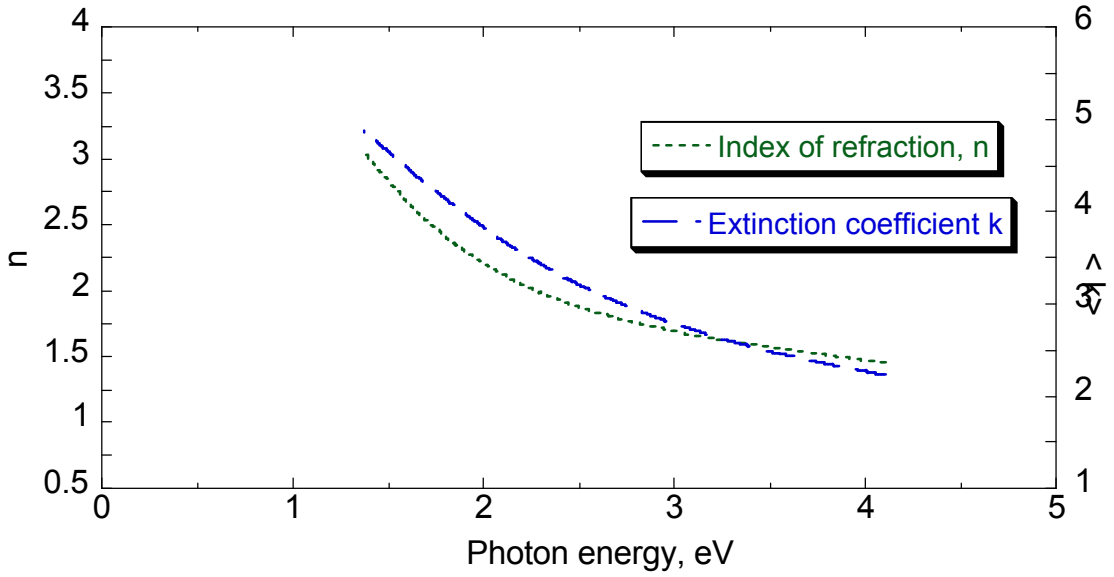
Index of refraction, n
Extinction coefficient k
n
k
Photon energy, eV
4
3.5
3
2.5
2
1.5
1
0.5
6
5
4
3
2
1
0
1
2
3
4
5

Figure 5.