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TUNGSTEN SPUTTER COATING DEVELOPMENT TO PRODUCE HIGH Z SHELLS

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We developed a production tungsten sputter coating process to uniformly deposit tungsten on 840 μm outer diameter GDP shells using a bounce coating technique. We were able to control the tungsten-coating rate and therefore coating thickness based on gravimetric analysis. At the end of our work we could routinely produce uniform 0.5 μm tungsten coatings on GDP shells with a Δ wall ≤0.04 μm. Techniques were developed and applied to measure coating uniformity based on x-radiography and x-ray fluorescence data. Typical surface roughness values for bounce coated shells having a 0.5 μm tungsten coating were 40 to 50 nm RMS. Stationary GDP shells were coated with 0.5 μm tungsten and found to have surface roughness approaching 10 nm RMS, which was similar to the roughness of the underlying GDP mandrel surface. This result indicates that coating processes with less agitation such as tap or roll coating may produce much smoother tungsten coatings.

I. INTRODUCTION

There are accounts of the sputter deposition of tungsten on various substrates for electronic and wear resistant applications in the literature.[1–3] Deposition of uniform high Z coatings on shells has been investigated using magnetron sputtering previously by others for applications in Inertial Confinement Fusion.[4,5] Over the last few years our group has investigated the sputter deposition of the high Z metals gold and palladium on shells for Inertial Fusion Energy applications.[6] More recently workers in our laboratory had carried out a limited study to sputter coat tungsten on glass shells to see if such shells could be permeation filled with D_2.[7] In this paper we report the development of tungsten coatings to produce high Z shells focusing on production and measurement of uniform tungsten wall thickness shells.

II. EXPERIMENTAL

During our current tungsten coating development studies we used magnetron sputter coating of pure tungsten initially on flat and then spherical substrates. Preliminary coating runs were on a combination of glass cover slips, sapphire spheres, PAMS shells, and GDP shells to understand and optimize the coating process. We arrived at an optimum coating distance between the tungsten sputter target and our substrates of 10 cm. During coating development we were able to determine coating rates for flats, stationary shells, and bounced shells. We used a piezoelectric bounce pan system in order to uniformly coat shells. A schematic diagram of our coating system is shown in Fig. 1.

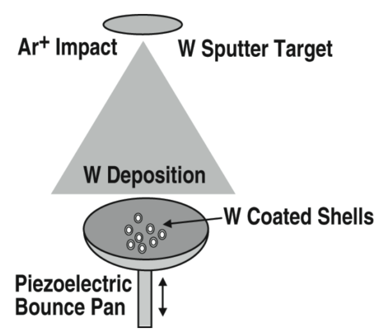

Figure 1. Schematic diagram of bounce coating system used in development of tungsten shells.

In our preliminary coating runs, we observed that PAMS shells often expanded during the sputtering process when we used a 10 cm coating distance. This made the coating process and also the analysis of coatings using PAMS shells problematic. The expansion was likely due to localized heat produced during sputter coating, which caused the PAMS to soften and expand from internal gas pressure within shells. Because of difficulties in tungsten coating of PAMS mandrels, all later development focused on tungsten coating of GDP mandrels which did not change size during sputter coating. Tungsten coated GDP shells provided us reproducible and unambiguous results for coatings.

The GDP shells used during tungsten coating development had a nominal outer diameter of 840 μm and a wall thickness of 14 μm. We typically bounce coated batches of 5 GDP shells at a time. In all we carried out eleven production runs of 5 shell batches. The last coating run carried out used a batch of 10 GDP mandrels, as a curiosity, to see if we could observe any relationship between batch size and coating surface finish. Average thickness for the tungsten coatings was determined gravimetrically by weighing shells before and after tungsten deposition. Shell diameters were measured by interferometry and shell weights were measured using a Cahn microbalance.

Coating rates for shells were determined from the average tungsten coating thickness. In addition, some shells were examined by SEM to verify coating thickness. A typical tungsten coating rate for bounced shells was ~0.40 μm/h once our coating process was under control. Surface roughness was measured for both stationary coated shells and bounce coated shells by using a Veeco Instruments WYKO Surface Profiler. A new quantitative radiography method was applied to tungsten bounce coated shells to determine tungsten wall uniformity.[8] In addition, an x-ray fluorescence analysis method was carried out on one of the shells analyzed by quantitative radiography to verify wall uniformity. These two methods will be described along with results later in this paper.

III. RESULTS

III.A. SEM Examination

Figure 2 shows a low magnification SEM photomicrograph of a 0.8 μm thick tungsten coated GDP shell. The coating quality obtained for the shell shown in Fig. 2 was typical for tungsten-coated shells once our coating process was under control. Figure 3 shows a high magnification SEM photomicrograph showing the coating cross-section. Observed in the cross-section is a columnar grain structure for the tungsten sputter coating. Also evident from the SEM photo is texture and scattered bumps or domes on the surface. There are also shallow sub-micron pits observed on the surface. The domes and pits are likely produced during the coating process from fine tungsten particles added or taken away from the coating during shell-shell and shell-pan collisions.

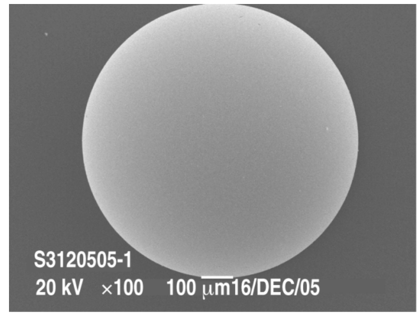

Figure 2. Low magnification SEM photograph of 0.8 μm tungsten coating on GDP shell.

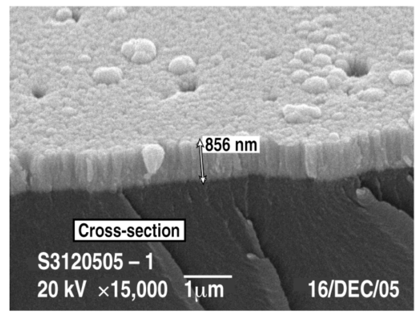

Figure 3. High magnification SEM photomicrograph cross-section for tungsten sputter coating showing columnar grain structure.

III.B. AFM Spheremap Data

AFM spheremap data was obtained for a starting GDP mandrel and a 0.8 μm tungsten bounce coated GDP shell. This data is presented in Figs. 4 and 5. The power spectra for both shells are similar up to ~ mode 10. After mode 10 the spectra for the tungsten coated shell rises and remains elevated to mode 1000. The difference in the two power spectra is indicative of greater surface roughness for the tungsten-coated shell. The greater surface roughness can also be plainly seen in the shell profile data shown in Figs. 4 and 5. An important observation from the power spectra data is that low modes (modes < 10) were not affected by the tungsten sputter coating process.

III.C. WYKO Surface Roughness Measurements

We carried out WYKO surface profile analysis for a variety of tungsten-coated shells during our coating development. Figure 6 presents examples of 3-dimensional surface profiles for a 0.5 μm tungsten coating on a stationary shell and a bounce-coated shell. The fine tungsten particles added or taken away from the coating during shell-shell and shell-pan collisions. Dramatic difference in the surface roughness is observed in the

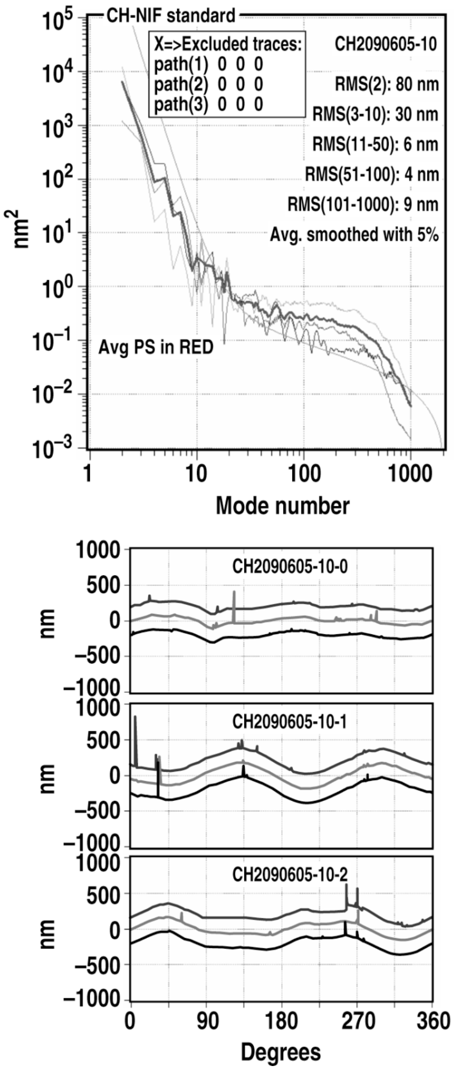

Figure 4. AFM spheremap power spectra and profiles for representative GDP mandrel used during tungsten coating development.

comparison. A summary of surface roughness results for tungsten coated shells and starting GDP mandrels is given in Table I.

III.D. Quantitative X-Radiography to Determine Wall Uniformity

We applied a new quantitative method developed by GA to determine tungsten wall uniformity for 0.5 μm tungsten coated GDP shells.[8] This analysis was the first example of x-ray transmission analysis for a high-Z metal coated shell. Previously we had only used x-ray transmission analysis to determine wall uniformity for low-Z materials such as beryllium and carbon.

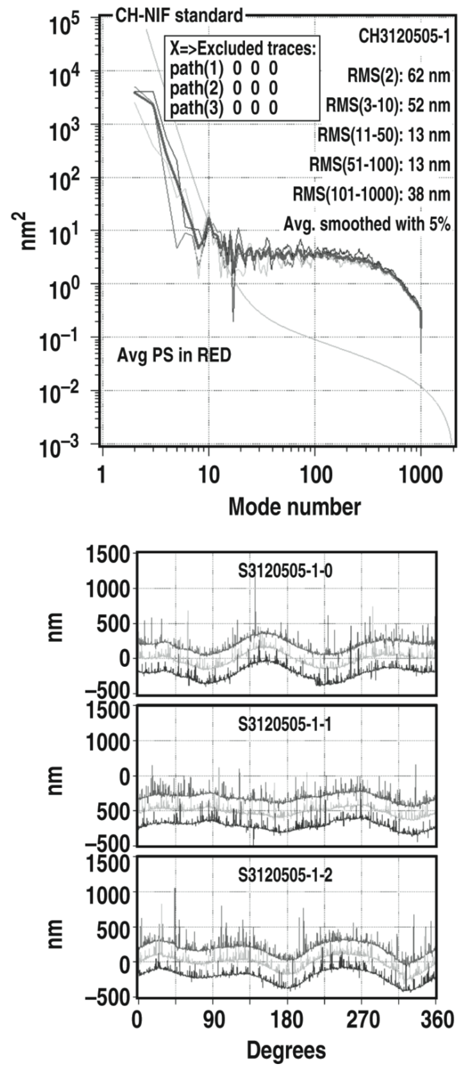

Figure 5. AFM spheremap power spectra and profiles for 0.8 μm tungsten bounce coated GDP shell.

During our analysis two tungsten coated GDP shells from the same coating batch and two uncoated GDP mandrels were x-rayed on a plate. The resulting x-ray images were digitized using a unique LabView program to remove distortion from the lens and plate. Next the x-ray transmission was determined for each shell. The transmission for the tungsten coated GDP shells was compared to that for the uncoated GDP shells. From this comparison a thickness profile for each shell was determined. Figure 7 shows the results for this analysis. For both tungsten-coated shells a thickness of 0.50±0.01 μm was calculated from the x-ray transmission data. Our thickness from gravimetric analysis for these shells was determined to be 0.5 μm.

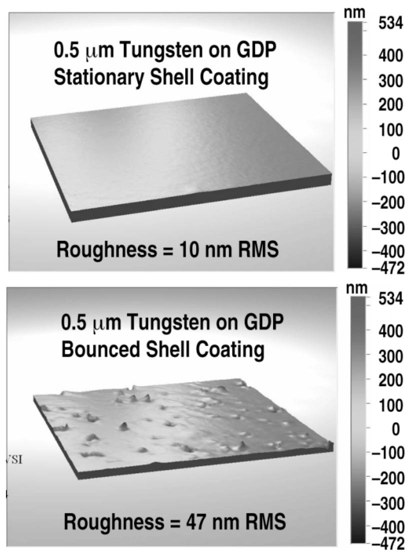

Figure 6. Comparison of surface roughness for 0.5 μm tungsten coating deposited on a stationary shell and a bounce coated shell. Sampled region is approximately 100 μm × 100 μm. Height scales are identical in each surface photo.

Table I. Surface Roughness Summary Data

Surface Measured	Shell Agitation	Batch Size	WYKO Average Roughness (nm RMS)
GDP shell	Bounced	30	10.3±2.8[a]
0.5 μm W on GDP shell	Stationary	5	11.2±3.2[a]
0.5 μm W on GDP shell	Bounced	5	45.0±7.3[b]
0.5 μm W on GDP shell	Bounced	10	66.5±9.0[a]
0.8 μm W on GDP shell	Bounced	5	84.5±4.5[b]

[a]Corresponds to average of individual measurements from 5 different shells.
[b]Corresponds to average of individual measurements from 4 different shells.

III.E. X-Ray Fluorescence Measurements to Determine Wall Uniformity

We also used x-ray fluorescence (XRF) as a tool to verify the tungsten wall uniformity for tungsten coated GDP shells. In this method, one of the shells analyzed by quantitative x-radiography was examined by XRF. This tungsten coated shell was determined to have an average thickness of 0.5 μm by gravimetric and by quantitative x-ray analysis. Figure 8 is a schematic diagram showing key features of the XRF analysis technique.

In the XRF technique the coated shell was held between two plastic 4 μm thick XRF foils inside a holder. To focus and isolate on different regions of the coated

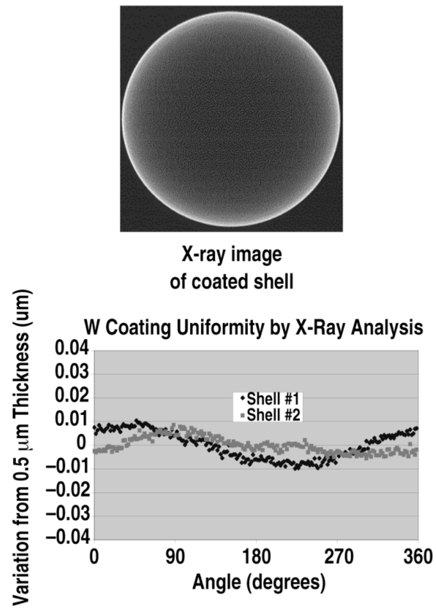

Figure 7. X-ray image of 0.5 μm tungsten coated GDP shell and x-ray transmission analysis data showing tungsten coating uniformity within a shell, and between two different shells.

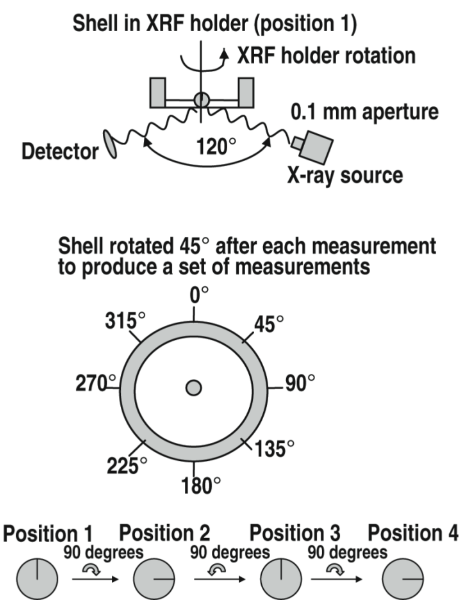

Figure 8. Schematic diagram showing key features of XRF analysis technique used to determine wall uniformity for tungsten coated GDP shells.

shell, we used a 100 μm aperture to limit the x-ray beam and the region of the shell measured for tungsten XRF counts. The shell examined had a outer diameter of 840 μm. In our technique we measured tungsten XRF counts in different positions for an annulus that circled the equator (horizontal mid-plane) of the shell. We rotated the XRF holder in 45° increments in the horizontal plane that was aligned with the collimated x-ray beam. With this technique we were able to measure tungsten counts around a horizontal annulus for the shell (position 1). We next opened the holder and rotated the shell 90° perpendicular to the holder plane (rotation 1) to measure the tungsten counts in an annulus around the shell that was orthogonal to the first position. After measuring tungsten counts for rotation 1, we then rotated the shell in the holder back 90° to the original position (rotation 2) to make a third set of tungsten count measurements. We then rotated the shell one last time back 90° (rotation 3) for a final set of tungsten counts. This treatment for changing the shell position for the various XRF measurements is detailed in Fig. 8.

Figure 9 is a summary of the XRF data for the treatment described. The plot presents tungsten XRF counts and thickness as a function of radial position around the tungsten coated shell. The reproducibility for tungsten counts for a given measurement was ±10 counts/s. Since we independently determined the average tungsten coating thickness for this shell gravimetrically and by quantitative radiography, we were able to convert the raw tungsten count data to a thickness for the various radial positions around the shell.

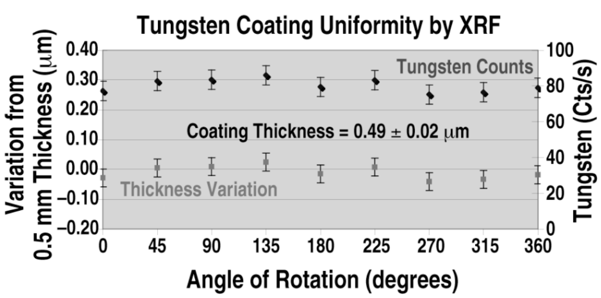

Figure 9. Summary of x-ray fluorescence data for 0.5 µm tungsten coated GDP shell previously analyzed by quantitative radiography.

IV. DISCUSSION

IV.A. Coating Thickness Control

During our work on tungsten coating development, the coating rate and therefore thickness deposited on substrates was well controlled based on gravimetric data. Checks done during this work on flats to measure coating rates and thickness by interferometry were consistent with thickness calculated for flats and shells using gravimetric data. In addition the new quantitative x-ray analysis method used to determine tungsten coating thickness was an independent check of thickness control that supported the gravimetric results.

IV.B. AFM Spheremap Analysis

The AFM power spectrum presented in Fig. 5 for the tungsten bounce coated GDP shell shows elevated mid and high modes (modes >10). However modes <10, and particularly mode 2, appear unaffected by the tungsten sputtering process when the data is compared to the Fig. 4 power spectra which corresponds to a typical GDP mandrel. This result indicates that the shells are not being deformed during the coating process and that long-range shell uniformity is being maintained. The shell profiles shown in Fig. 5 are indicative of a rough surface with many small (~ micron diameter) domes and occasional pits. This was also observed in the SEM photo shown in Fig. 3, and the WYKO surface profile for the tungsten bounce coated shell shown in Fig. 6.

IV.C. WYKO Surface Roughness Analysis

The WYKO surface roughness data summarized in Table I shows various trends. When we tungsten sputter coated a stationary shell to 0.5 µm we observed an average surface roughness of 11 nm RMS which was very similar to the surface roughness for the GDP mandrel. When we bounce coated batches of 5 shells the average tungsten surface roughness for a 0.5 µm coating increased to 45 nm RMS likely due to shell-shell and shell-pan collisions.[9] When we increased the batch size to 10 shells the average tungsten surface roughness, again for a 0.5 µm coating, increased to 66 nm RMS. Even though the increase in shells is small (from 5 to 10 shells), and we only have data for one coating run, it is very likely that doubling the quantity of shells coated can correlate with increased surface roughness from more shell-shell collisions. This had been observed during development of GDP coatings using bounce agitation where coating of large shell batches correlated with increased production of domes and surface roughness.[9]

The surface roughness data of Table I also indicates an increase in surface roughness with increased tungsten coating thickness. This has often been observed for metal sputter coatings.[10] In our case where we have used a fixed coating distance, increased coating thickness means a longer required coating time. Since we use the bounce coating method to uniformly coat shells this means more shell-shell and shell-pan collisions which can further increase surface roughness for tungsten coatings. The increase in average surface roughness from 45 nm RMS to 84 nm RMS going from a 0.5 µm to a 0.8 µm tungsten coating supports this hypothesis.

There is one important observation to make for the roughness data obtained for the stationary coated shell. After coating, WYKO surface analysis showed the stationary coated surface to have roughness that remained at approximately 10 nm RMS. Since the sampling of the stationary shell was at the north pole this region is similar to a flat substrate and would yield the smoothest surface. Sampling the stationary shell near the equator would likely yield a rougher surface. This is an important result which shows that if one could coat shells uniformly with less agitation, the surface can be made very smooth with the measured roughness approaching that of the underlying substrate.

There are various ways to agitate shells during sputter coating to try to obtain uniform coatings; bounce coating, tap coating, and roll coating. In the work we present here, we used bounce coating, as this technique is the production method used to fabricate uniform coatings on the majority of small (<1 mm diameter) spherical ICF targets. However for sputter coating, the surface roughness may increase undesirably from the bouncing process.

Surface roughness can be decreased by tap or roll coating of shells.[9] Past experience for coating of GDP shells has shown that agitation by rolling or tapping can produce shell surfaces with less features when compared to agitation by bouncing for the same number of shells coated to the same thickness.[9] This data indicates that much improved surface roughness can be obtained using a tap or roll coating process. Therefore improved surfaces for tungsten sputter coated shells may be obtained in the future using tapping or rolling for shell agitation. The coating uniformity for coated shells would need to be monitored during trial of these other agitation methods, since smooth coating surfaces and uniform walls are not necessarily correlated.

IV.D. Tungsten Coating Uniformity

The coating uniformity of the tungsten-coated shells was determined in two ways. The first method used was quantitative x-radiography which determined the tungsten wall uniformity in one view (one plane) for a shell. In addition since we analyzed two shells from the same batch by this method we obtained an indication of uniformity from shell to shell. The second method used to determine wall uniformity was by XRF measurements on one of the shells analyzed by x-radiography. During the XRF analysis the shell was rotated 90° after each set of measurements which allowed shell coating uniformity for two orthogonal planes to be determined.

Quantitative x-radiography can be a very sensitive technique to characterize wall thickness uniformity.[8] Using spectra for x-ray transmission, the difference in transmission across 200 different radial lines for each shell was calculated. The sensitivity of the LabView program to read the transmission spectrum is 2.7%, meaning that for every 0.027 change in transmission there is a 0.01 μm change in thickness. The results of measurements for two different tungsten coated shells from the same batch shown in Fig. 7 indicates that the uniformity of the tungsten coating within the two shells was ±0.01 μm. In addition, comparison of the coating thickness results from the x-ray transmission data for the two shells showed them to have an average tungsten coating thickness within 0.01 μm of each other. Therefore the tungsten coatings were indicated to be uniform from shell to shell, inferred from the two sampled bounce coated shells. This result was consistent with gravimetric average coating thickness data for bounce-coated shells from the same batch.

Figure 9 is the coating thickness uniformity based on XRF measurements for one of the tungsten-coated shells that was analyzed by quantitative radiography. Evaluation of the radial data in the Fig. 9 plot shows the data to be relatively flat. This suggests that the tungsten coating on the bounce-coated shell analyzed is fairly uniform. When we take the average tungsten counts at each radial position and convert these to a thickness using the average coating thickness from gravimetric analysis, we observe that the coating thickness around the shell is 0.49 ±0.02 μm. This result and the waviness of the plotted data as a function of radial position are both similar to the quantitative radiography results presented in Fig. 7. These observations are not surprising since the same shell was analyzed by both methods. The rather good agreement between the two independent analysis methods, quantitative x-radiography and x-ray fluorescence measurements, suggests that both methods can be used to characterize tungsten coating uniformity.

V. SUMMARY

The first controlled production of thin wall tungsten shells was carried out. We successfully developed a production sputter coating process to deposit pure tungsten uniformly on GDP shells. Problems were encountered sputter coating tungsten on PAMS shells because of shell expansion due to shell heating during the coating process. The coating rate and thickness for deposited tungsten coatings on GDP shells was well controlled during our study based on gravimetric analysis.

New techniques were developed to complement the gravimetric analysis of tungsten coating thickness to determine uniformity for coatings. These new techniques were quantitative x-radiography and x-ray fluorescence measurements. The analysis techniques to determine wall uniformity indicated that we could produce uniform 0.5 μm tungsten coatings on GDP shells having a Δ wall ≤0.04 μm using bounce coating.

The best surface roughness for a bounce-coated shell having a 0.5 μm tungsten coating was approximately 40 nm RMS. Stationary coated shells were observed to have much better surface roughness. Stationary GDP shells that were coated with 0.5 μm tungsten were found to have surface roughness approaching 10 nm RMS, which was close to the roughness of the underlying GDP mandrel surface. This result indicates that coating processes with less agitation such as tap or roll coating may produce much smoother tungsten coatings. Previous studies during development of GDP coatings showed that tap and roll coating can produce very smooth shell surfaces. Future efforts for development of high quality tungsten coatings should focus on improved agitation methods during tungsten deposition, and investigations into the effect of batch size and coating thickness on final coating quality.

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