

Measuring the Density of Plutonium Surface Oxides

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Plutonium is known to have an extremely high oxygen affinity, growing a surface oxide in even low oxygen content atmospheres [1,2,3]. There are some measurements on the densities of mechanically packed oxide powders [4], for example for investigating powder sintering behaviour [5], and also on x-ray diffraction measured lattice parameters to calculate ideal oxide densities. However, there is very little data on *in-situ* grown surface oxides, necessary to fully understand growth kinetics.

Stanford et al [6] created a range of oxide thicknesses on bulk metallic plutonium, ranging from 35 nm to 400 nm as measured from focussed ion beam cross-sections. For each sample they used electron probe micro-analysis (EPMA) to measure the oxygen K α x-ray intensity relative to pure plutonium oxide reference materials, termed the O k-ratio to generate O k-ratio versus oxide thickness correlation curves.

The authors of this study applied thin film analysis algorithms on Stanford et al's comprehensive published data to calculate the densities of their grown oxides. Normally these algorithms are used to calculate layer thickness and/or composition from EPMA measured element k-ratios, using an assumed layer density. Utilising the known thicknesses and O k-ratios, and assuming stoichiometric PuO₂, the calculations were 'inverted' to output densities. The results from 3 different software packages, PENEPMA, DTSA-II and BadgerFilm, were compared and produced values ranging from 4.0 to 5.0 g.cm⁻³ (Table 1). This falls within the 1.5 – 5.8 g.cm⁻³ range of values published for mechanically compacted oxide powders [4] but is only half that of the 10.2 – 11.5 g.cm⁻³ expected from XRD-measured lattice parameters [7]. The authors discuss the potential causes for this discrepancy.

Table 1 Calculated mean densities and standard deviations.

Software	Mean density (g.cm ⁻³)	Standard Deviation (g.cm ⁻³)
PENEPMA	4.1	0.5
BadgerFilm	4.7	0.5
DTSA-II	5.0	0.6

References

- 1 Schnizlein and Fischer, 1967, *Journal of the Electrochemical Society*, **114**, 23
- 2 Larson and Cash, 1969, *Journal of Physical Chemistry*, **73**, 2814-2820
- 3 Ravat et al., 2018, *Corrosion Science*, **138**, 66-74
- 4 Moseley and Wing, 1965, *Properties of Plutonium Dioxide* - RFP-503.
https://digital.library.unt.edu/ark:/67531/metadc1028004/m2/1/high_res_d/4613823.pdf
- 5 Kutty et al., 2001, *Journal of Nuclear Materials* **297**, 120-128
- 6 Stanford et al. 2020, *Journal of Nuclear Materials*, **530**
- 7 Haire, R.G., 2019, *Plutonium Handbook 2nd Edition*, Vol. 3, Ch. 20, Editors Clark, D.L., Geeson, D.& Hanrahan, R.J., American Nuclear Society, La Grange, IL 60526 USA