Written Portion of Prelims Defense:

Do a critical reading of two journal articles and submit a brief report on your assessment

"Isotopic Signatures of Weapon-Grade Plutonium from Dedicated Natural Uranium-Fueled Production Reactors and Their Relevance for Nuclear Forensic Analysis"

Assessment:

The first article looked at the ability to distinguish plutonium produced in three different Pu production reactors. The three reactors are listed below.

- 1. Hanford-type (graphite-moderated, light-water cooled) reactor [US]
- Calder Hall-type (graphite-moderated, gas-cooled) reactor [United Kingdom North Korea]
- 3. NRX-type heavy-water-moderated and light-water-cooled reactor [Canada India, Pakistan] It is alluded to that these reactors were chosen because of their Pu production status and their

All three have nuclear weapons existence in states not generally accepted as weapons states (India, Pakistan, North Korea). Infinite

lattice calculations were completed with MCNP and coupled to ORIGEN2 with another code called MCODE. The power density for all reactors were set at 40 W/cm³ and Pu isotopics were studied for these reactors under the conditions of natural uranium and recycled natural uranium loadings and a ²³⁹Pu wt% of 93.8.

> It is concluded that these three reactors cannot be distinguished from each other based on Pu isotopics in either uranium loading condition, the recycled uranium loading scenario having more ²³⁸Pu due to ²³⁶U. Two reasons for this are similar single group cross sections and uncertainties in reactor models and mode of operation. The author maintains that differences between statements that differences between the statement of the s LWRs with LEU, and these reactors can be distinguished with ²³⁸Pu/totPu and ²⁴²Pu/²⁴⁰Pu ratios, without mentioning uncertainties in ²⁴²Pu measurements. Further the author states that the lack of

Seems unlikely

actual sample data hinders the ability to distinguish between these productions reactors because, with these samples, specifics of irradiation are known and because there are differences in material properties. Finally the author concludes that a comprehensive database for nuclear forensic analysis, containing actual samples, should be created.

This paper highlights two Pu ratios that could be helpful for distinguishing reactor type and could help narrow fast-to-thermal ratio calculations for my project. In the paper, low concentrations of boron were added to the moderator to each reactor to simulate the presence of control rods, which wouldn't necessarily simulate control rods due to distributed versus localized flux depressions. The author also discusses how actual Pu samples are a potential solution to the inability of distinguishing these reactors, but does not provide any a concrete example for how.

"TRI-n-BUTYL Phosphate as an extracting solvent for inorganic nitrates - III"

The second article explored D-values for the three different oxidation states of Pu, (III) (IV) and (VI), in nitric acid or sodium nitrate, with TBP diluted in kerosene. Samples were prepared from tetravalent Pu in 6 M HNO₃ at a presumed concentration of 0.02 M Pu (the highest concentration of Pu reported in the paper). Pu(IV) was reduced with Fe(II) in order to run experiments for Pu(III) and Pu(VI) was prepared with anodic oxidation. Dependencies of nitric acid, TBP, Pu, and uranyl concentrations on D-values for Pu in its three oxidation states were explored with dependencies related to the power of the number of molecules needed for reactions (equilibrium constant). Pu was measured with alpha spec and nitric acid concentrations were determined through titration. Errors for this work were due to residual Pu(IV) during Pu(VI) experiments (up to 9%) and reproducibility of final results were ±10%.

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Pu(NO₃)₄ V(IV), was most readily extracted by TBP in kerosene, followed by Pu (VI), and Pu (III), each trailing the previous by an order, and two orders of magnitude, respectively. Increasing nitric acid concentrations increased extractability due to increased availability of nitrate ions. The D-value for these reactions should be related to the fourth power of the nitrate ion concentration, and this experiment yielded results something closer to a third power dependency because Pu(IV) could come complexed with a single nitrate.

had shown a quadratic increase?

Increasing TBP concentration for Pu(IV) had an showed a square law increase in D-values with constant nitric acid concentration because two TBP are needed to form the extractable complex. The authors also tested sodium nitrate as the salting solution for extraction and found that, mole for mole, sodium nitrate was more effective at extracting across the board. This was compared with Al(NO₃)₃, which is even more effective than sodium nitrate.

Uranyl nitrate was shown to decrease D-values for Pu, simply because uranyl loads the organic phase, reducing the amount of free TBP, which actually lowers D-values for all other dissolved species. The author notes that plutonyl nitrate is less extractable than uranyl nitrate by a factor of about 10, which is unusual because increasing atomic number usually associates with increasing extractability for the lanthanides and actinides.

The authors defend a point that would be useful for my dissertation, namely that the shape of D-value versus nitric acid concentration would remain the same for different concentrations of TBP. This is useful because my dissertation results could be extrapolated out to different TBP loadings.