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Calculated Influence of a Niobium Reflector in Plutonium Systems

Tracy Stover

Savannah River Nuclear Solutions, LLC, Savannah River Site, Aiken, SC 29803, Tracy.Stover@srs.gov

INTRODUCTION

Savannah River Site's H-Canyon facility has been tasked with using its unique electrolytic dissolver to disposition assorted plutonium materials, primarily research reactor grade stainless-steel clad plates. The dissolver, developed in the late 1960s and used into 1980s, has historically been employed on to disposition stainless steel and zirconium clad uranium materials. Restart of this facility brings with it a new mission.

In order to withstand that intense electrical currents, on the order of 10,000 amps, developed in this nitric acid system, the dissolver has to be made of a robust material. Niobium is used as the primary material of construction for the basket containing the fissile material, the electrodes contacting the material, and the structural material of the charging apparatus.

The new mission therefore creates a rather unique arrangement in which research reactor grade plutonium will be in contact with loosely fitting niobium reflection, while submerged in acid. There is currently no such arrangement in the benchmark handbook (Ref. 1). Therefore, the motivation for this paper is to investigate the effects of niobium reflection on a plutonium system. Various geometric and chemical configurations are modeled which are in line with the real system that will be in used. KENO-VI in SCALE 6.1 is used to simulate the neutron multiplication conditions. SCALE's ENDF-B VII 238-group cross sections are used in the stimulation. This version of SCALE and this cross-section library are chosen because they are the ones currently validated for use at Savannah River Site (Ref. 2).

DESCRIPTION OF THE WORK

Dissolver Description

The electrolytic dissolver was described in detail upon its initial deployment (Ref. 3). Early scoping work for this new mission is presented concurrent with this work (Ref. 4). Therefore, only a brief description of the dissolver is given.

The dissolver vessel is a large stainless-steel tank, approximately 7.5 ft. in diameter and 8 ft. in height. It is surrounded by a water jacket to provide cooling in addition to cooling coils inside the vessel itself. A charging and dissolution apparatus is affixed along the vertical centerline of the tank and is constructed primarily of niobium with thin protective platinum coatings on the electrodes and aluminum oxide insulators composing a basket which holds the material to be dissolved between the electrodes.

Materials are charged into one of three wells of the basket and an electrical current passed between the electrodes across the material. This breaks down cladding such as stainless steel and zirconium allowing the nitric acid to dissolve the fuel material inside. The nitric acid also acts as a conductor for the current.

Configurations Simulated

Two geometries are considered in this work and the second geometry is modeled with three chemical configurations. The fissile material is assumed to be Pu-239 with an alpha phase density of 19.86 g/cm^3. This allows the study to focus on the effects of niobium by using a pure, well understood fissile material as the charge.

The first geometry is a sphere with spherical shell reflector of niobium (Fig. 1) in vacuum.

The second geometry is a right circular cylinder of a height to diameter ratio of 1 between two plates of niobium. (Fig. 2). Vacuum is applied beyond the plate boundaries.

In each geometry a thickness of the niobium is fixed and the fissile mass adjusted until the predicted k-effective plus two times the uncertainty (k-best estimate, k_{BE}) is less than or equal to 0.95. The thickness of the reflector is increased, and the fissile mass adjusted again to give $k_{BE} <$ 0.95. This is repeated until the reflector is effectively infinite. In the second geometry, the open space between the plates not occupied by the plutonium is filled with either air (void), normal density water, or a dilute plutonium nitrate solution containing 3 g/L Pu-239 and 4 M excess nitric acid. The acid solution introduces competing effects of nitrogen neutron poisoning and additional fissile material. The acid solution is similar to what the concentrations are predicted to be in the electrolytic dissolver.

Once the behavior of each configuration out to an effective infinite reflection condition are established, sensitivity is examined. The electrolytical dissolver will have niobium from 0.15 cm thick in contact with the material up to about 2.5 cm thick cumulative with the electrodes and other structural components. Sensitivity is examined in two manners.

First, reflector thickness of 0.15, 0.5, 1.5, and 2.5 cm are examined in a simple variation. An uncertainty of ±0.5, ±1.0%, ±2.5%, ±5.0%, and ±10.0% is applied uniformly to all of the multi-group cross sections of niobium. No change in the plutonium cross section is assumed. This is achieved in the input deck by scaling the density of niobium by these factors. The macroscopic cross section consists of the product of the microscopic cross section and the number density. The number density is directly computed from the density, atomic weight and Avogadro's number. Therefore, scaling density achieves the same effect as multiplying each microscopic cross section by the same scaler.

Second, the SCALE TSUNAMI sequence is run to perform sensitivity calculations by reaction type. The 3-D KENO VI TSUNAMI sequence is run and the 44-group covariance library included with SCALE is used. This sequence returns an uncertainty in k_{BE} due to cross section uncertainty and includes the top reaction contributors to that uncertainty. The TSUNAMI calculations examine reflector thicknesses up to 5.5 cm.

Determination of k_{SAFE}

A conservative k-safe of 0.95 is employed due to the unique nature of this configuration. A k-best estimate (k-calculated plus two times the uncertainty) is compared to this value.

RESULTS

Fig. 3 shows the calculated impacts of reflector thickness on a sphere of Pu-239. After approximately 12.5 cm (~ 5 inches) the niobium is effectively an infinitely thick reflector. The mass of Pu-239 to achieve a k_{BE} just under 0.95 drops from 5.38 kg to 3.31 kg.

Fig. 4 shows the calculated impacts of reflector thickness on a cylinder of Pu-239 between two plates of niobium. All three chemistry configurations are shown on Fig. 4. For the dry case, the reflectors again become effectively infinite near 12.5 cm. The fissile mass is reduced from 8.95 kg with no reflector to 6.71 kg with an effectively infinite reflector. For the flooded cases however, there is a slight change in that the reflectors are effectively infinite closer 10 cm (~4 inches). Addition of either water or fissile acid solution greatly reduces the fissile mass that can be in the system. The water configuration starts at a fissile mass of 6.64 kg unreflected and reduces to 4.49 kg fully reflected. The acid solution (called "PuN" the figures) with dilute fissile is for practical purposes the same as water. However, its affect changes with increasing niobium thickness. At thick reflection, the acid solution fissile mass allowance is slightly higher than the water case but after 2.5 cm the effect is negative, i.e. slightly less fissile mass is allowed in the acid system. Differences between the water and acid systems remain on the order of ones to tens of grams.

Fig. 5 shows the result of linearly varying the niobium density, and by virtue linearly varying the macroscopic cross section, for the first configuration. If in typical practice administrative margins are on the order of 0.01 Δk and the codes are run until statistical uncertainty is less than 0.005 Δk, the niobium cross sections would need to be at least 5% uncertainty to have a comparable effect.

Figures 6, 7, and 8 show the results of varying the niobium density in the second configuration with air/void, water, and dilute fissile acid solution as fill between the niobium plates, respectively. Using the same criterion as above, niobium cross sections would have to have uncertainty in excess of 10% to potentially have an appreciable effect on these systems.

Figure 9, 10, and 11 show the results of the TSUNAMI study for only Geometry 2 configurations. Geometry 1 configurations were run but are not presented here. The total k_{BE} uncertainty, as a function of thickness, introduced by cross section uncertainty is shown in Fig. 9. The uncertainty is actually dampened (no pun intended) by the presence of liquid and also by the thickness of the niobium. Both of those parameters introduce the potential for additional neutron reactions other than those with plutonium. The primary contributor to the uncertainty in Fig. 9 is the neutron per fission release for Pu-239. The largest niobium contributors are the elastic, inelastic, and capture reactions (in descending order of importance). The effects of elastic and inelastic scatter in niobium are shown in Fig. 10 and 11, respectively. Note that the scale of Figures 9, 10, and 11, have been expanded to show detail; pay attention to the absolute values.

CONCLUSIONS AND FUTURE WORK

Fundamentally, there is no benchmark experiment of a similar system to qualitatively or quantitatively compare these results to.

This work introduced two significant conservatisms by assuming a k-safe of 0.95 and assuming 100 wt.% Pu-239. In practice the intended material is research reactor grade plutonium between 65 and 92 wt.% Pu-239 only and a more realistic k-safe for plutonium metal/solution systems near 0.96 will likely be used.

As Ref. 4 indicates, the actual mission will likely incorporate excess administrative margin and a neutron poison to ensure the system is safely subcritical. The effects of niobium uncertainty can be said to at worst be on par with administrative margin that would commonly apply to the systems. So, it can be accommodated by standard industry practice and there is no safety concern.

Absent those aspects however, for a charge mass around 4.5 kg of plutonium per container uncertainties on the order of 0.005 to 0.015 Δk could easily mean the difference between charging one or two containers per batch or between repacking the material or not. In other words, the difference between exposing workers to additional dose and not exposing them and maintaining gloveboxes for repack and not maintaining them.

While this work does not speculate on the accuracy of the current niobium cross sections, an integral experiment would beneficial for this type of system. Otherwise additional controls and conservatism would be employed to make any mission with this type of system successful.

ACKNOWLEDGEMENTS

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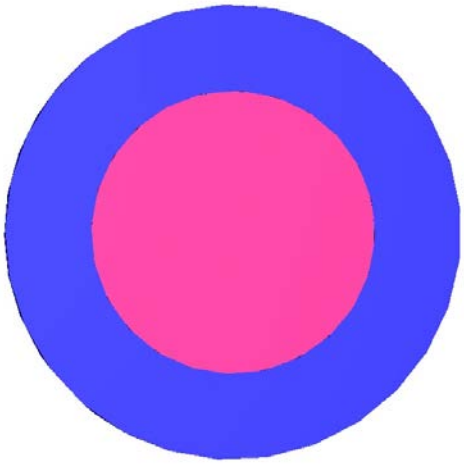

Fig. 1. Geometry 1 – plutonium sphere surrounded by a 2.5 cm niobium shell reflector.

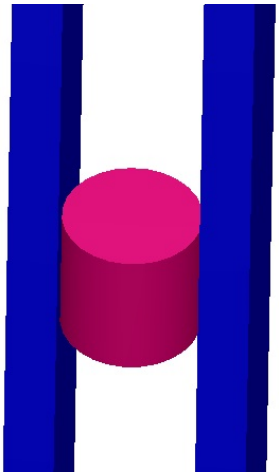

Fig. 2. Geometry 2 – plutonium cylinder between two 2.5 cm plates of niobium with various fill.

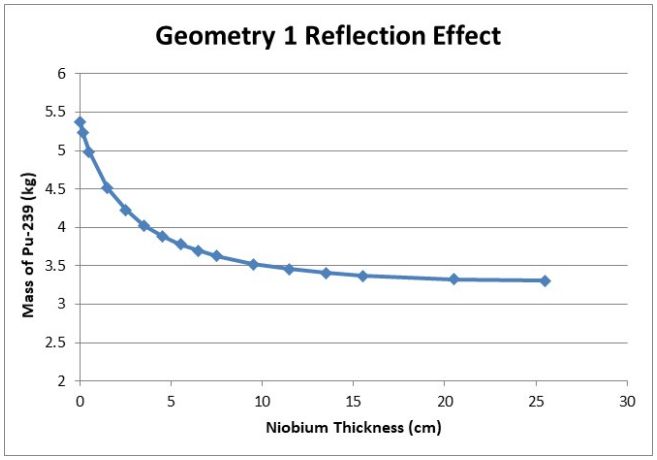

Fig. 3. Geometry 1 – fissile mass for k_{BE}=0.95 vs. niobium thickness.

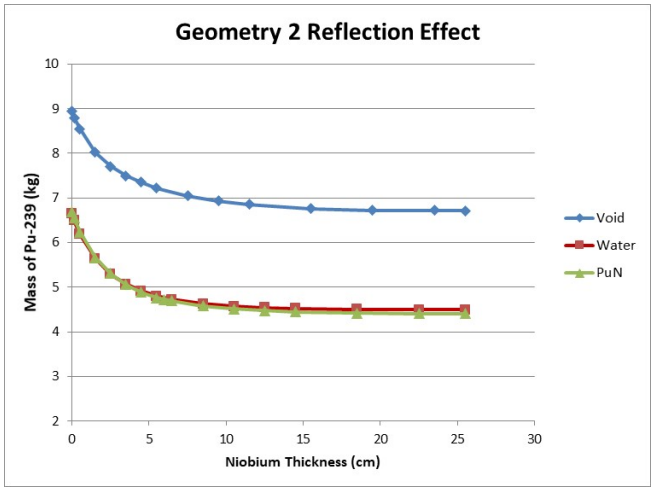

Fig. 4. Geometry 2 – fissile mass for k_{BE}=0.95 vs. niobium thickness.

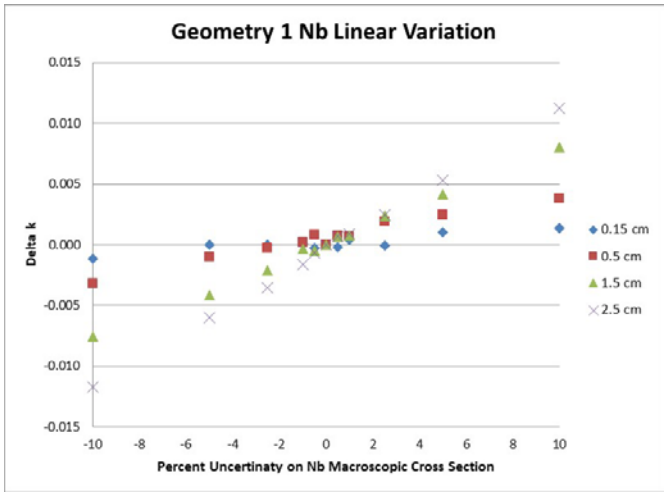

Fig. 5. Geometry 1 – Δk resulting from linear variation of niobium cross sections uncertainty

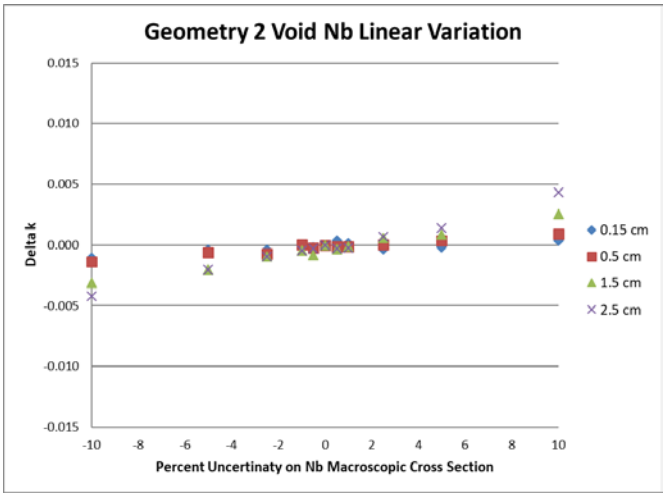

Fig. 6. Geometry 2 Void – Δk resulting from linear variation of niobium cross sections uncertainty

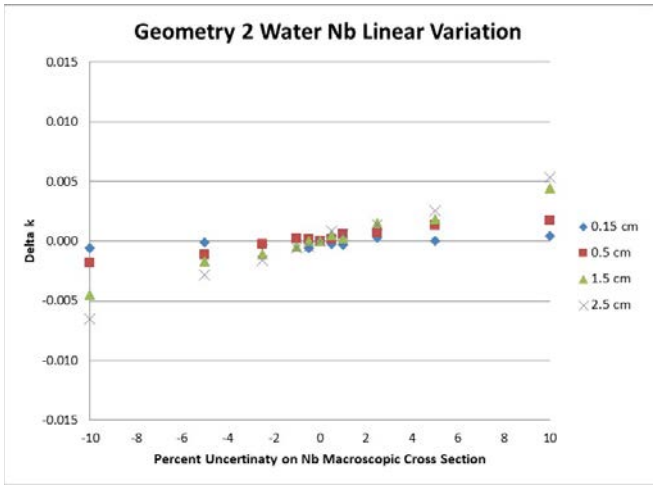

Fig. 7. Geometry 2 Water – Δk resulting from linear variation of niobium cross sections uncertainty

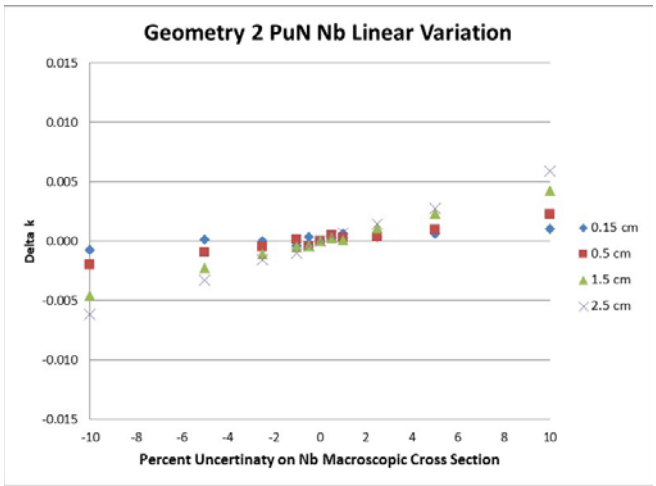

Fig. 8. Geometry 2 Plutonium Nitrate – Δk resulting from linear variation of niobium cross sections uncertainty

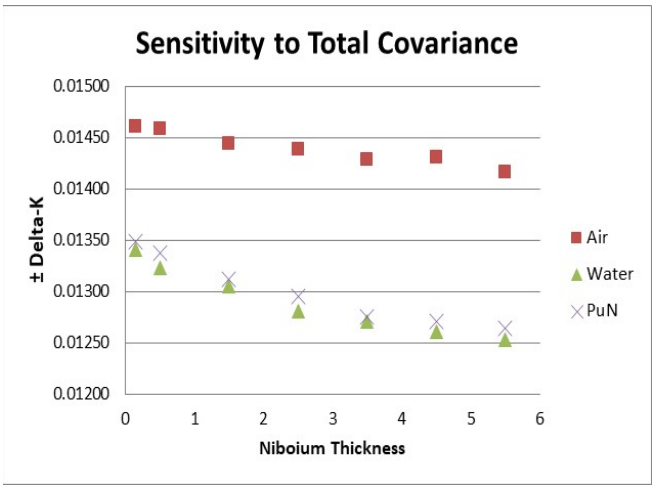

Fig. 9. Geometry 2 –Δk resulting from total cross section covariance versus niobium thickens

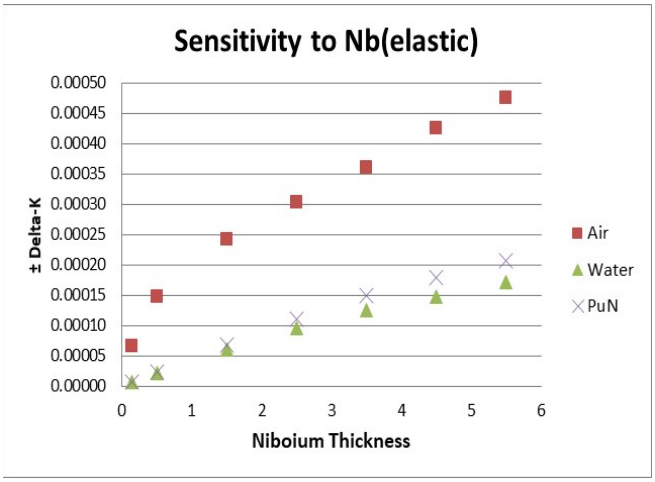

Fig. 10. Geometry 2 –Δk resulting from niobium elastic scatter cross section covariance versus niobium thickens

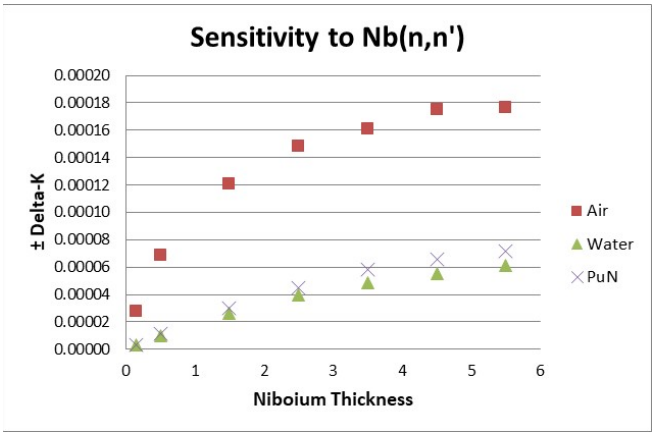

Fig. 11. Geometry 2 – total Δk resulting from niobium inelastic scatter cross section covariance versus niobium thicken