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Impact of NDA Uncertainties on NCS at the K-25 Site

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INTRODUCTION

The K-25 Building at the East Tennessee Technology Park is relying on the use of Non-Destructive Assay (NDA) methods for characterizing hold-up materials in process equipment. The characterization data is used for many purposes including mass estimates for nuclear criticality safety (NCS) and waste disposition. This paper addresses the sensitivity of certain parameters in the NDA process to overall mass measurement results.

DESCRIPTION OF THE ACTUAL WORK

Passive Neutron Emission Method

One method for measuring uranium holdup in the gaseous diffusion cascade is by passive neutron emission. The dominant source of neutrons is from a (α, n) reaction between uranium and fluorine in the uranyl fluoride compounds. The alpha contribution from ^{234}U in high-enriched uranium dominates the other isotopes.

The compound form affects the neutron emission rate. Uranyl oxyfluoride (UO_2F_2) has a relatively low fractional weight of fluorine resulting in a lower (α, n) production rate over other uranyl fluoride compounds. However, the presence of water in UO_2F_2-nH_2O also reduces net (α, n) production. Original methods of NDA assumed a dry form of UO_2F_2 as the deposit compound, consistent with an operating cascade.

For all but very low enriched uranium, the mass of ^{235}U can be estimated by the following general equation:

$$M_{U\text{-}235} = \frac{CR \times RNDE \times R_{235/234}}{\varepsilon \times SA}$$

where: $M_{U\text{-}235}$ = Mass of ^{235}U (g)
CR = Net Neutron Count Rate
RNDE = Relative Neutron Detector Equivalent, which relates the efficiencies of the detector used to a "standard" detector
$R_{235/234}$ = ^{235}U /^{234}U Ratio (g ^{235}U / g ^{234}U)
ε = Geometry efficiency of the measurement related to the item being measured (cps/nps)
SA = Specific Activity or Neutron Emission Rate (n/s/g ^{234}U) (i.e., the α, n yield) for the compound

The ratio of ^{235}U to ^{234}U and the neutron emission rate are parameters that must be clearly defined. If the ^{235}U /^{234}U ratio is under predicted or if the neutron emission rate is over predicted, ^{235}U mass will be under estimated.

Examination of the $^{235}U/^{234}U$ Ratio

Actual $^{235}U/^{234}U$ ratios do not match theoretical ratios due to cascade equipment alignment. A non-linear curve-fit equation was developed to historic mass spectrometer data from 1960 through 1963 (representing the latter years of operation), along with an upper and lower 95% prediction curve.

An examination of the curve-fit equation and the data shows that when the curve-fit relation is compared to a 95% prediction curve, the maximum difference is 8.9%. Based on these results, an uncertainty of 10% should be applied to the $^{235}U/^{234}U$ ratios.

Alpha-Neutron Emission Rate

The historical neutron emission rate used in NDA measurements is 253.4 n/s per g ^{234}U for the dry form of UO_2F_2, with no uncertainty applied. This value is based on data for cylinders of UF_6 published by Sampson[1], which established a neutron emission rate of 576 ± 42 n/s per g ^{234}U for UF_6 and a factor of 0.44 to equate UO_2F_2 to UF_6. The 0.44 correction is based on using stopping power ratios to equate energy deposition in UF_6 fluorine to UO_2F_2 fluorine.

Six additional sources for (α, n) reactions were examined to identify the variability of data beyond the Sampson data. The sources examined were:

- ^{19}F(α, n) reaction cross sections measured by Balakrishnan et al.[2], Norman et al.[3], and Vukolov and Chukreev[4]
- Thick-target neutron yields measured by Bair et al.[5], Jacobs and Liskien[6], and Segre and Wiegand[1]

The first three references involve measuring alpha particle interaction cross sections for $^{19}F(\alpha, n)$ over varying energy ranges The measured cross-section data was incorporated into ORIGEN[7] for the purposes of calculating neutron emission rates for dry UO_2F_2 using cross section data from each of these references. The neutron yields in ORIGEN are calculated using stopping power methods adapted from the SOURCES code [8].

The next three references involve converting measured neutron emission rates from thick fluorine targets in various compounds and equating those compounds to UO_2F_2 also through use of ion stopping powers.

A statistical analysis of the data was performed to determine the appropriate neutron emission rate for dry UO_2F_2 to be used. The analysis resulted in a weighted mean of 217 n/s per g $^{234}U \pm 43$ n/s, which is substantially smaller than the historical value of 253.4 n/s per g ^{234}U, and will thus yield larger NDA estimates of ^{235}U mass.

Effect of Hydration Content of UO_2F_2

The presence of water in the UO_2F_2 compound similar to moisture conditions at K-25 will result in a lower neutron emission yield due to the additional energy deposition in the water molecule. Data from various studies show that UO_2F_2 systems will stabilize to a hydrated state of 1.6 – 2.0 moles of H_2O per mole of UO_2F_2 when in equilibrium at room temperature in air with relative humidity of 50 – 60%.

An assumed hydration level of 1.6 moles of water was used for the purpose of making NDA measurements for large deposits and is adequate to represent smaller deposits. Calculations show a reduction in neutron yields of about 83% for UO_2F_2 hydrated to 1.6 moles. This equates to a net neutron emission rate of 180 ± 36 n/s per g ^{234}U.

RESULTS

The basis for a generic (e.g., 50%) NDA measurement uncertainty needs to be documented so that the reliability of the data may be evaluated. This study shows that a historical generic uncertainty applied to mass measurements may not be sufficiently conservative.

From this study, an uncertainty of about 10% in the computed mass value is introduced from the $^{235}U/^{234}U$ ratio that was not previously accounted for. Additionally, use of the historical neutron emission value based on the data from Sampson for neutron NDA measurements may lead to an underestimation of the ^{234}U contents, and thus underestimate ^{235}U contents by as much as 40%.

Therefore, NCS evaluations must use conservative assumptions and conditions to account for the wide variability in NDA results.

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