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ABSTRACT

The disposal of excess ^{233}U as waste is being considered. Because ^{233}U is a fissile material, a key requirement for processing ^{233}U to a final waste form and disposing of it is the avoidance of nuclear criticality. For many processing and disposal options, isotopic dilution is the most feasible and preferred option to avoid nuclear criticality. Isotopic dilution is dilution of fissile ^{233}U with nonfissile ^{238}U. The use of isotopic dilution removes any need to control nuclear criticality in process or disposal facilities through geometry or chemical composition. Isotopic dilution allows the use of existing waste management facilities that are not designed for significant quantities of fissile materials to be used for processing and disposing of ^{233}U.

The amount of isotopic dilution required to reduce criticality concerns to reasonable levels was determined in this study to be approximately 0.53 wt % ^{233}U. The numerical calculations used to define this limit consisted of a homogeneous system of silicon dioxide (SiO_2), water (H_2O), ^{233}U, and depleted uranium (DU) in which the ratio of each component was varied to learn the conditions of maximum nuclear reactivity. About 188 parts of DU (0.2 wt % ^{235}U) are required to dilute 1 part of ^{233}U to this limit in a water-moderated system with no SiO_2 present. Thus for the U.S. inventory of ^{233}U, several hundred metric tons of DU would be required for isotopic dilution.

I. BACKGROUND

With the recent ending of the cold war, the U.S. Government is examining options for disposing of excess fissile materials, which potentially include ^{233}U. Part of this material will be retained for research, medical, and industrial uses. However, part of the inventory may be declared excess and consequently may require disposal.

If ^{233}U is declared a waste, there are economic incentives to use existing waste processing facilities to prepare the material for disposal. However, these facilities were not designed for significant concentrations of fissile materials, nor for addressing the resulting nuclear criticality control issues. Also, because fissile materials can be used for nuclear weapons, materials with high fissile concentrations were not considered for disposal before the end of the cold war. Consequently, disposal of such fissile materials imposes the addition of criticality control to other requirements for safe disposal.

At reported[1] water-moderated solutions and metal densities between about 0.06 and 4.0 g/cm^3, ^{233}U has smaller measured and calculated critical dimensions (i.e., spherical masses/diameters, infinite-cylinder diameters/linear densities, or infinite-slab thicknesses/areal densities) than does either ^{235}U or ^{239}Pu. Because of significantly different fissile properties, the critical mass of a fully water-reflected and -moderated 0.4-g ^{233}U/cm^3 solution is about 1.3-kg ^{233}U, whereas solutions of ^{235}U and ^{239}Pu at similar densities have critical masses of about 2.6 kg and 2.7 kg, respectively. This paper addresses the unique criticality issues associated with processing and disposal of ^{233}U and suggests the use of isotopic dilution to reduce nuclear criticality control problems.

II. CHARACTERISTICS OF PROCESSING AND DISPOSAL OF ^{233}U

The potential quantities of ^{233}U requiring disposition are small, and some ^{233}U contains ^{232}U and its highly radioactive daughter products sufficient to require hot-cell processing of the material to an acceptable waste form. Strong incentives exist to (1) use existing facilities and (2) avoid complex criticality control and other licensing issues associated with the high-level waste (HLW) spent nuclear fuel repository program. These goals can be realized if nuclear criticality control is achieved by isotopic dilution, which is the addition of sufficient ^{238}U to lower the ^{233}U enrichment level below that at which nuclear criticality can occur.

Existing U.S. Department of Energy (DOE) HLW vitrification facilities and proposed transuranic waste processing facilities may be able to process ^{233}U. However, these facilities are not designed for significant concentrations of fissile materials. If such facilities are to be used to maintain nuclear criticality safety controls for ^{233}U, it is not possible to rely on traditional geometry or chemical (e.g., neutron absorbers or fissile concentration) controls without substantial modifications of plant equipment and operations.

If neither geometric nor chemical control is practicable for nuclear safety in a processing facility, isotopic dilution (enrichment) is the best remaining criticality control option. Noting that all uranium isotopes have the same chemical characteristics is important; therefore, the ^{238}U used isotopically to dilute the ^{233}U will not separate from the fissile uranium in any normal chemical process.

Relying on geometry or chemical composition alone within disposal facilities to control criticality over geological time frames is also difficult. Several mechanisms can cause changes in waste geometry and chemistry, including groundwater transport of uranium and mechanical disturbances of the waste. If criticality control is to be ensured for thousands of years by either geometric control or chemical control (including neutron absorbers), system performance must be predictable for these lengths of time. Such prediction has proven quite problematic. No such difficulties exist when isotopic dilution is used for criticality control.

III. LEGAL AND INSTITUTIONAL CONSIDERATIONS

An expanding series of laws, regulations, recommendations, and actions by the U.S. Government address nuclear criticality concerning disposal facilities. A trend is developing to use isotopic dilution as the preferred method of criticality control for fissile materials following disposal. The environmental impact statement[2] and record of decision[3] for the disposition of excess high-enriched uranium (HEU) recommended isotopic dilution of the fissile ^{235}U if any HEU was disposed of as a waste. The same considerations apply to the disposition of excess ^{233}U. Also, the U.S. Nuclear Waste Technical Review Board, in its recommendations to the U.S. Congress and the Secretary of Energy, recommended consideration of the use of DU to isotopically dilute fissile materials to prevent the potential for nuclear criticality in geological repositories containing fissile material.

IV. CONTROL OF NUCLEAR CRITICALITY BY ISOTOPIC DILUTION

This work determined that to ensure control of nuclear criticality in ^{233}U by isotopic dilution with ^{238}U, the ^{233}U concentration must be reduced to less than 0.53 wt %. For nuclear criticality safety, this concentration is equivalent to ^{235}U at an enrichment level of ~1.0 wt %—a level that will not result in nuclear criticality under conditions found in processing or disposal facilities. These uranium isotopic concentrations avoid the need to control other parameters to prevent nuclear criticality; that is, the ^{233}U can be treated as another radioactive waste. After isotopic dilution to these concentrations, nuclear criticality will not occur in a geological environment, over time, nor in waste processing operations that have not been designed for fissile materials.

V. COMPUTATIONAL APPROACH

Computational evaluations were conducted to define simple algebraic relationships that could be used for calculating the quantity of DU (i.e., 0.2 wt % ^{235}U and 99.8 wt % ^{238}U) needed for isotopic dilution of ^{233}U to ensure subcriticality in waste processing and disposal facilities. Specifically, mixtures composed of DU, enriched uranium, and quartz sand (SiO_2) were examined for variable degrees of neutron moderation by SiO_2, H_2O, and artificially by oxygen (O_2) to define infinite-material neutron-multiplication constants (k_∞) equal to or less than the k_∞ for 1 wt % ^{235}U and ~99 wt % ^{238}U.

SiO_2 and H_2O were selected as the most restrictive materials for subcriticality that are naturally occurring in large process systems and geological environments. Other neutron-absorbing compounds consisting of iron, calcium, and sodium cannot be assured to be present in any specific proportion; consequently, they were not considered in this study. Therefore, only combinations of ^{233}U, SiO_2, H_2O, and DU were evaluated.

The Standardized Computer Analyses for Licensing Evaluation (SCALE)[4] system, specifically the AJAX code and the CSAS1X sequence (BONAMI, NITAWL, XSDRN), along with the 238-energy group ENDF/B-V neutron cross-section library, were used to evaluate subcritical mixtures of these materials. Historic validation studies[5,6] using ENDF/B-V neutron cross sections have shown that water-moderated, homogeneous, single- and multi-unit, ^{233}U critical systems have calculated k_{eff} values greater than 0.95 (average $k_{eff} \approx 0.99$). Therefore, the CSAS1X sequence was executed for various combinations of SiO_2, H_2O, ^{233}U and depleted uranium, DU (i.e., 0.2 wt % ^{235}U and 99.8 wt % ^{238}U) to calculate subcritical infinite-homogeneous medium multiplication factors, k_∞ values, approximating 0.95 (0.98 for some systems). The use of a k_∞ acceptance value of 0.95 for this ^{233}U scoping study is not fully justified (i.e., integral experimental data for SiO_2–H_2O–^{233}U mixtures are not available for data testing and validation). Additionally, specific validation and analytical studies using configuration-controlled hardware and software, for these type systems and materials, are necessary to satisfy criteria for computational safety evaluations. Obtaining experimental benchmark evidence is a primary hurdle in completing such a specific validation.

The selected subcritical value for the infinite-media neutron multiplication factor (k_∞) for optimally moderated ^{233}U mixtures was $k_\infty \le 0.95$. The limiting subcritical enrichment for optimally moderated homogeneous aqueous systems is well defined[1] to be 1 wt % ^{235}U and 99 wt % ^{238}U. Though 1 wt % ^{235}U is well defined to be subcritical for an optimally water-moderated system, the calculated k_∞ is typically between 0.997 and 1.003, depending upon codes and neutron cross-section libraries that are used. The 1 wt % ^{235}U value was used to define the subcritical DU dilution relationship for uranium enriched in ^{235}U. Using the results of the computational study for ^{233}U dilution and the knowledge about the subcriticality of aqueous homogeneous 1 wt % ^{235}U enriched uranium, a simple equation was developed to define the necessary DU dilution to ensure the subcriticality of ^{233}U and uranium enriched in ^{235}U. The developed relationship for the most restrictive combinations of ^{233}U, enriched uranium, and DU is based upon the commonly accepted "unity rule" concept[7] that two or more mixtures of optimally water-moderated, subcritical (i.e., maximum $k_\infty \le 1.0$), infinite-media fissile materials may be homogeneously combined and remain subcritical if the composition of the materials remains homogeneous.

Because the physical and chemical conditions of ^{233}U and ^{235}U for some types of process and disposal options cannot be guaranteed, the results of this isotopic dilution study were reduced to the most restrictive possible combination of materials (i.e., H_2O, DU, ^{233}U, ^{235}U, and ^{238}U) that will ensure subcriticality. The effects of oxygen moderation and significant weight percents of ^{234}U and ^{236}U were examined and determined to be of no safety consequence in typical process and disposal environments.

VI. RESULTS

As determined from the computational studies and published data, the most restrictive combination of the considered materials is a homogeneous mixture of uranium and water. For this study, the mixture was assumed to be a mixture of water molecules and uranium atoms.

Limited results, not considering SiO_2, are as follows:

Computational Results for 187.7 g DU/g ^{233}U

g SiO_2/g ^{233}U	g H_2O/g ^{233}U	k_∞
0.0000	26.9434	0.9447
0.0000	25.6252	0.9463
0.0000	24.3979	0.9474
0.0000	23.2525	0.9482
0.0000	22.1810	0.9486
0.0000	21.1764	0.9487
0.0000	20.2327	0.9484
0.0000	19.3445	0.9479
0.0000	18.5071	0.9471
0.0000	17.7162	0.9460

A simple equation was developed to ensure the subcriticality of ^{233}U and uranium enriched in ^{235}U by dilution with DU. The equation defines the quantity of DU that must be blended with ^{233}U and various enrichments of ^{235}U. The mass of DU is expressed in terms of ^{233}U and enriched uranium masses as:

$$gDU = 188(g\ ^{233}U) + \left(\frac{E-1}{0.8}\right)(g\ of\ enriched\ uranium)$$

where

$g\ DU$ = g of depleted uranium (i.e., 0.2 wt % ^{235}U),
E = the wt % of ^{235}U in enriched uranium,
g of enriched uranium = total U - ^{233}U.

In the above relationship, ^{234}U and ^{236}U may be considered ^{238}U providing the atom number ratio of the (^{234}U + ^{236}U)/^{235}U does not exceed 1.0. If the calculated quantity of g DU using the above equation is negative, the uranium material already contains sufficient ^{238}U to ensure subcriticality, and no additional DU is needed.

VII. APPLICATION OF DILUTION EQUATION

Oak Ridge National Laboratory (ORNL) possesses a large quantity of contaminated ^{233}U in temporary storage produced from the Consolidated Edison Uranium Solidification Program (CEUSP). It is in a monolithic form of uranium, gadolinium, and cadmium oxides. Information regarding the material follows.

CEUSP Material

Uranium inventory		
^{232}U	<0.01 wt %	<0.1 kg
^{233}U	9.69 wt %	101.1 kg
^{234}U	1.39 wt %	14.5 kg
^{235}U	76.52 wt %	797.8 kg
^{236}U	5.59 wt %	58.3 kg
^{238}U	6.80 wt %	70.9 kg
Chemical inventory		
UO_2	64.37 wt %	1084.5 kg
CdO	19.46 wt %	328.0 kg
Gd_2O_3	2.20 wt %	36.8 kg
Metal contaminants	13.98 wt %	235.5 kg
Total material inventory		1684.8 kg

Because one cannot ensure that the cadmium, gadolinium, or other neutron-absorbing elements will remain intimately mixed with the uranium, no credit can be taken for their presence in the application of the dilution equation. Only the mass of elemental uranium can be applied in the dilution equation.

The 101.1 kg of ^{233}U is applied to the dilution equation separately from the remaining mass of uranium. Converted to grams, the mass of ^{233}U is 101.1 kg ^{233}U × (1000 g/kg) = 101,100 g ^{233}U. The remaining mass of uranium is then 1042.6 – 101.1 kg = 941.5 kg U or 941.5 kg U × (1000 g/kg) = 941,500 g U. Therefore, the effective enrichment of the remaining uranium is (100) × (797.8 kg ^{235}U)/(941.5 kg U) = 84.74 wt %. Substituting into the dilution equation,

$$g\,U(D) = 188(g\ ^{233}U) + \left(\frac{E-1}{0.8}\right)(g\,enriched uranium)$$

$$g\,U(D) = 188(101{,}100) + \left(\frac{84.74-1}{0.8}\right)(941{,}500)$$

$$g\,U(D) = 1.18 \times 10^{8}\ .$$

This is to say that it will require a dilution of about 1.18 × 10^8 g DU per 1.04 × 10^6 g CEUSP uranium to denature the CEUSP material such that no geological condition of the material can result in criticality. This amounts to increasing the mass of CEUSP uranium by a factor of about 113. This evaluated subcritical mixture is predicated upon the condition that the depleted uranium is of the same chemical composition as the CEUSP UO_2 such that no chemical separation of the mixture can occur.

Using the same computer codes and cross sections, test calculations were done with the above diluted mixture of CEUSP uranium oxides (omitting all other cadmium, gadolinium and metal contaminants) with various proportions of water combined in an infinite homogeneous media. The resulting infinite-media neutron multiplication factors, k_∞, for various water proportions within the mixture are as follows.

Water Volume Fraction	**k_∞**
0.65	0.9817
0.67	0.9867
0.70	0.9921
0.73	0.9943
0.74	0.9943
0.75	0.9937
0.76	0.9926
0.80	0.9817

The large k_∞ values are the result of the fissile material being predominantly ^{235}U (subcritical acceptance criterion for 1 wt % ^{235}U having a calculated $k_\infty \approx 1.00$) as compared with systems that are predominantly ^{233}U ($k_\infty \leq 0.95$ for a subcritical acceptance criterion for optimally moderated ^{233}U).

VIII. CONCLUSIONS

It is judged that the developed depleted-uranium dilution equation provided above is a good first approximation for diluting ^{233}U and enriched uranium providing the mixture is homogeneous and consists of uranium compounds (excluding compounds of carbon, beryllium, and deuterium) and water. The presence of other fissionable materials or non-neutron-absorbing, highly neutron-moderating elements such as carbon, beryllium, or deuterium has not been considered in this work. Though other scattering or absorbing nuclides may be present in a mixture, their effects have not been accounted for in the reduction of required U(D) mass for dilution of ^{233}U and enriched uranium.

Because the dilution equation uses depleted uranium as the diluent to approximate an equivalent 1 wt % ^{235}U enriched uranium and water-moderated system, the potential for an autocatalytic criticality accident[8] is rendered impossible. This results because low-enriched uranium, less than about 5.1 wt % ^{235}U, cannot become critical[1] without the presence of water and because it is judged that homogeneous systems of 1 wt % ^{235}U or ~0.53 wt % ^{233}U cannot be made critical as a mixture of U-H_2O.

Though other elements found in geological deposits may be effective neutron scatterers (e.g., silicon, aluminum, oxygen) or somewhat effective neutron absorbers (e.g., iron, sodium, calcium), no assurance can be provided that such elements will remain with the uranium during hydrogeochemical processes over geological time spans. Isotopic dilution of ^{233}U and ^{235}U with depleted uranium in an identical compound and form provides the only method to ensure that natural hydrogeochemical processes cannot transform the ^{233}U and ^{235}U into a critical configuration over geologic time spans.

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