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Criticality Safety Study of UF_6 and UO_2F_2 in 8-in.-Diameter Piping

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1. INTRODUCTION

The purpose of this report is to provide an evaluation of the criticality safety aspects of using up to 8-in.-inner-diameter (ID) piping as part of a system to monitor the ^{235}U enrichment in uranium hexafluoride (UF_6) gas both before and after an enrichment down-blending operation. The evaluated operation does not include the blending stage but includes only the monitors and the piping directly associated with the monitors, which are in a separate room from the blending operation. There are active controls in place to limit the enrichment of the blended UF_6 to a maximum of 5 weight percent (wt %) ^{235}U.

Under normal operating conditions of temperature and pressure, the UF_6 will stay in the gas phase and criticality will not be credible. The two accidents of concern are solidification of the UF_6 along with some hydrofluoric acid (HF) and water or moisture ingress, which would cause the UF_6 gas to react and form a hydrated uranyl fluoride (UO_2F_2) solid or solution. Of these two types of accidents, the addition of water and formation of UO_2F_2 is the most reactive scenario and thus limits related to UO_2F_2 will bound the limits related to UF_6.

Two types of systems are included in the monitoring process. The first measures the enrichment of the approximately 90 wt % enriched UF_6 before it is blended. This system uses a maximum 4-in.-(10.16-cm-) ID pipe, which is smaller than the 13.7-cm-cylinder-diameter subcritical limit for UO_2F_2 solution of any enrichment as given in Table 1 of American National Standard ANSI/ANS-8.1.[1] Therefore, this system poses no criticality concerns for either accident scenario.

The second type of system includes two enrichment monitors for lower-enriched UF_6. One monitors the approximately 1.5 wt % enriched UF_6 entering the blending process, and the second monitors the approximately 5 wt % enriched UF_6 coming out of the blending process. Both use a maximum 8-in.-(20.32-cm-) ID piping, where the length of the larger ID piping is approximately 9.5 m. This diameter of piping is below the 26.6-cm-cylinder-diameter subcritical limit for 5 wt % enriched UO_2F_2 solutions as given in Table 6 of ANSI/ANS-8.1. Therefore, for up to 5 wt % enriched UF_6, this piping does not present a criticality concern for either accident scenario.

Calculations were performed to determine the enrichment level at which criticality could become a concern in these 8-in.-ID piping sections. Both unreflected and fully water-reflected conditions were considered.

2. DESCRIPTION OF COMPUTER CODE AND INPUT MODEL

Nuclear criticality calculations were performed using the KENO V.a code within the SCALE 4.4a system[2] and the 238-group ENDF/B-V cross-section library. KENO V.a uses the Monte Carlo method to calculate a system's effective neutron multiplication factor (k_{eff}). Other modules within the SCALE 4.4a system perform cross-section processing to convert the ENDF/B-V data into a problem-specific working library. The BONAMI module performs resonance self-shielding for the unresolved resonance range using Bondarenko data. The NITAWL module performs resonance self-shielding for the resolved resonance range using the Nordheim integral transport method.

These computer codes and cross-section data have been used extensively to calculate many types of systems. For homogeneous low-enriched and high-enriched ^{235}U systems, no significant calculational biases have been detected as a function of either neutron energy or moderation level.[3,4] However, few critical experiments are available for validating calculations involving low-moderated UF_6 or UO_2F_2. Because of the limited number of applicable critical experiments, the calculated value of k_{eff} used as an upper safety limit was 0.92. This is consistent with values used for safety basis evaluations at the Oak Ridge National Laboratory.

The model used for these calculations began with a solid 9.5-m-long cylindrical deposit with an outer diameter of 20.32 cm. This deposit was surrounded by a 0.635-cm- (0.25-in.-) thick pipe wall made of carbon steel. For the water-reflected cases, this was further surrounded by a 30.48-cm- (12-in.-) thick water region. Vacuum boundary conditions were used at the ends of the pipe and outside the carbon steel pipe or water reflector. Calculations were also performed with deposits of shorter length and with deposits containing annular space to determine the sensitivity of k_{eff} to these parameters.

To show that limits related to UO_2F_2 will bound limits related to UF_6, calculations were done with UF_6 enriched to 90 wt % ^{235}U. This enrichment was chosen to bound actual conditions, even though highly-enriched uranium is not expected to enter the 8-in.-ID pipe. The moderation level was varied from a hydrogen to uranium ratio (H/U) of 0 to 2 in small increments by adding HF.

For the calculations with UO_2F_2, the enrichment in weight percent of ^{235}U was varied from 5 to 60%. The moderation level of the deposit was also varied from an H/U of 5 to 100 to determine the optimum moderation level for each enrichment. The highest enrichment for which k_{eff} did not exceed 0.92 for any moderation level was chosen as the enrichment limit.

The atom densities of the various materials were calculated using SCALE 4.4a. Standard Composition Library materials were used for the carbon steel and the water, which resulted in the atom densities shown in Table 1.

Mixtures of UF_6 and HF were created using densities for each compound as a function of the H/U ratio taken from Ref. 5. These material densities and resulting atom densities are given in Table 2.

Mixtures of UO_2F_2 and water were created using densities for each compound as a function of the H/U ratio taken from Ref. 6. These material densities and resulting atom densities are given in Table 3.

Table 1. Atom densities of steel and water

Element	Atom density (atoms/barn·cm)
Carbon steel	
Iron	8.35E–02
Carbon	3.93E–03
Water	
Hydrogen	6.68E–02
Oxygen	3.34E–02

Table 2. Composition of modeled UF_6 deposits

H/U	UF_6 density (g/cm^3)	HF density (g/cm^3)	H (atoms/ barn·cm)	F (atoms/ barn·cm)	U (atoms/ barn·cm)
0.0	5.0750	0.0000	0.00E+00	5.25E–02	8.75E–03
0.1	4.9390	0.0284	8.56E–04	5.19E–02	8.51E–03
0.2	4.7967	0.0552	1.66E–03	5.13E–02	8.27E–03
0.4	4.5352	0.1044	3.14E–03	5.01E–02	7.82E–03
0.6	4.3004	0.1486	4.47E–03	4.90E–02	7.41E–03
0.8	4.0887	0.1883	5.67E–03	4.80E–02	7.05E–03
1.0	3.8969	0.2244	6.75E–03	4.71E–02	6.72E–03
1.5	3.4889	0.3013	9.07E–03	4.52E–02	6.01E–03
2.0	3.1576	0.3636	1.09E–02	4.36E–02	5.44E–03

Table 3. Composition of modeled UO_2F_2 deposits

H/U	UO_2F_2 density (g/cm^3)	H_2O density (g/cm^3)	H (atoms/ barn·cm)	O (atoms/ barn·cm)	F (atoms/ barn·cm)	U (atoms/ barn·cm)
5	3.7519	0.5539	3.71E–02	3.32E–02	1.47E–02	7.34E–03
10	2.4130	0.7125	4.77E–02	3.33E–02	9.44E–03	4.72E–03
15	1.9105	0.7720	5.16E–02	3.33E–02	7.48E–03	3.74E–03
20	1.4080	0.8315	5.56E–02	3.33E–02	5.51E–03	2.76E–03
25	1.1646	0.8611	5.76E–02	3.34E–02	4.56E–03	2.28E–03
30	1.0870	0.8706	5.82E–02	3.34E–02	4.25E–03	2.13E–03
40	0.7670	0.9082	6.07E–02	3.34E–02	3.00E–03	1.50E–03
50	0.6260	0.9242	6.18E–02	3.34E–02	2.46E–03	1.23E–03
60	0.5272	0.9364	6.26E–02	3.34E–02	2.07E–03	1.04E–03
70	0.4559	0.9448	6.32E–02	3.34E–02	1.79E–03	8.96E–04
80	0.4013	0.9512	6.36E–02	3.34E–02	1.58E–03	7.89E–04
100	0.3250	0.9598	6.42E–02	3.34E–02	1.28E–03	6.39E–04

3. RESULTS

Each calculation included 200,000 neutron histories, with 1000 neutrons per generation and 203 generations, skipping a minimum of three generations. This was adequate to ensure source convergence for these models and resulted in an average Monte Carlo uncertainty of 0.0016.

The pipe models containing UF_6 deposits at 90 wt % ^{235}U enrichment did not approach a calculated k_{eff} of 0.92 under any of the moderation conditions from H/U of 0 to 2 (Fig. 1). The highest calculated k_{eff} was 0.83 at an H/U of 0.4, and at higher moderation levels the k_{eff} declined steadily. Therefore, it may be concluded that solid deposits of UF_6 do not pose a criticality hazard under the conditions evaluated in this report and that deposits of UO_2F_2 represent the bounding worst-case scenario.

The models containing UO_2F_2 and water were examined for various values of enrichment and moderation. This report presents data for fully water-reflected models with enrichments between 5 and 20 wt %, and unreflected models with enrichments between 50 and 60 wt %. The moderation level of the deposit was also varied from an H/U of 5 to 100 to determine the optimum moderation level for each enrichment. The respective enrichment and moderation ranges that are presented here are the culmination of a broader analysis that determined the range where the system k_{eff} peaked at a value around 0.92. The results of these calculations are shown in Table 4 and in Figs. 2 and 3.

For fully water-reflected pipe, the maximum enrichment for which the calculated value of k_{eff} remained below 0.92 for all moderation levels was 10 wt %. The maximum k_{eff} for 10 wt % enriched UO_2F_2 was 0.9173, which occurred at an H/U of 15 and a hydrogen-to-^{235}U ratio (H/X) of 150.

For unreflected pipe, the maximum enrichment for which the calculated value of k_{eff} remained below 0.92 for all moderation levels was 50 wt %. The maximum k_{eff} for 50 wt % enriched UO_2F_2 was 0.9135, which occurred at an H/U of 40 and an H/X of 80.

The system k_{eff} was more sensitive to changes in annular void space than it was for deposit length. Values of k_{eff} began to drop off significantly once the annular void space exceeded 1 cm in radius. However, calculations with progressively shorter deposits showed that the k_{eff} of the system is not significantly lowered until the deposit reached about 1.5 meters, reduced from the model baseline of 9.5 meters. These results imply that a deposit does not need to reach the full 9.5 meters in length to have substantial reactivity, but that an annular void space through a deposit can have a noticeable effect on reactivity.

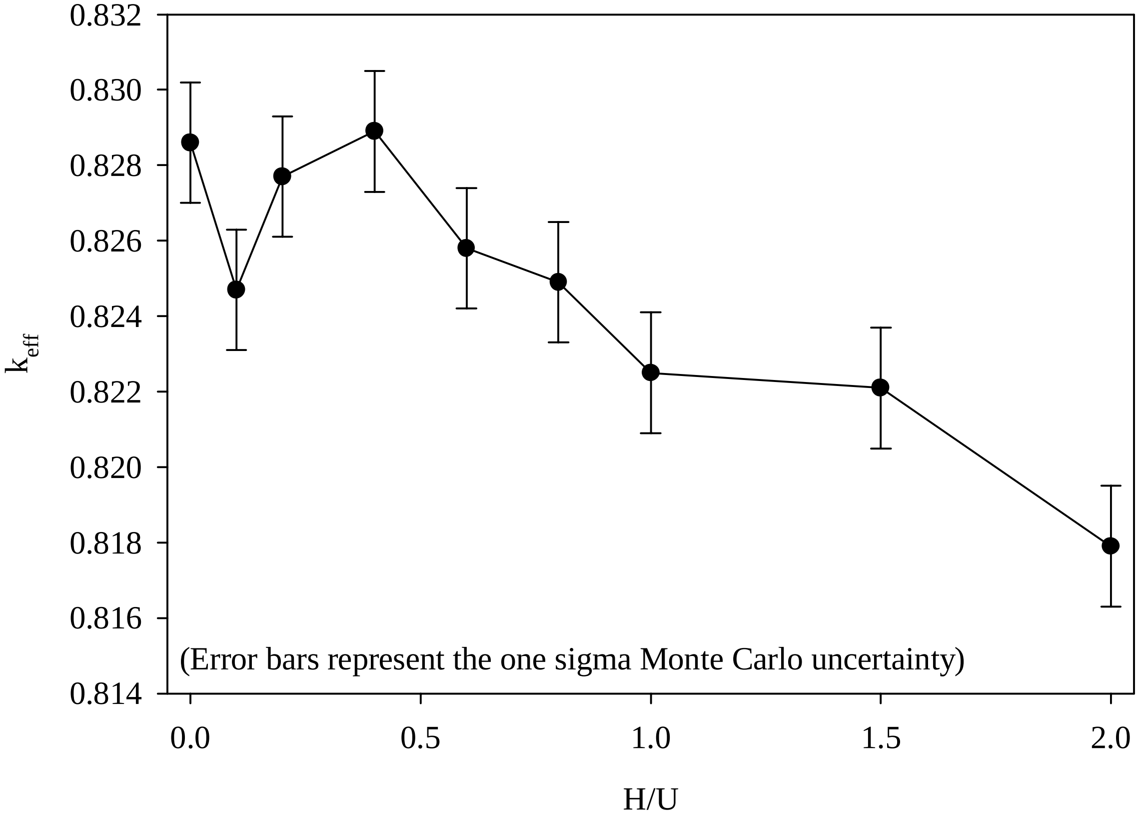

Fig. 1. Calculated k_{eff} for UF_6.

Table 4. Calculated values of k_{eff} for reflected and unreflected UO_2F_2 deposits

Enrichment	Reflection	H/U	H/X	k_{eff}
5	Water	5	100	0.7568
5	Water	10	200	**0.8065**
5	Water	15	300	0.8061
5	Water	20	400	0.7855
5	Water	25	500	0.7622
5	Water	30	600	0.7506
5	Water	40	800	0.6873
10	Water	5	50	0.8409
10	Water	10	100	0.9055
10	Water	15	150	**0.9173**
10	Water	20	200	0.9140
10	Water	25	250	0.9041
10	Water	30	300	0.8992
10	Water	40	400	0.8630
20	Water	5	25	0.9061
20	Water	10	50	0.9722
20	Water	15	75	0.9936
20	Water	20	100	1.0058
20	Water	25	125	**1.0064**
20	Water	30	150	1.0058
20	Water	40	200	0.9918
50	Unreflected	5	10	0.8034
50	Unreflected	10	20	0.8536
50	Unreflected	15	30	0.8645
50	Unreflected	20	40	0.8896
50	Unreflected	25	50	0.9063
50	Unreflected	30	60	0.9087
50	Unreflected	40	80	**0.9135**
50	Unreflected	50	100	0.9112
50	Unreflected	60	120	0.9087
50	Unreflected	70	140	0.9079
50	Unreflected	80	160	0.8989
50	Unreflected	100	200	0.8883

Table 4 (continued)

Enrichment	Reflection	H/U	H/X	k_{eff}
55	Unreflected	5	9	0.8171
55	Unreflected	10	18	0.8607
55	Unreflected	15	27	0.8806
55	Unreflected	20	36	0.8982
55	Unreflected	25	45	0.9100
55	Unreflected	30	55	0.9139
55	Unreflected	40	73	**0.9237**
55	Unreflected	50	91	0.9240
55	Unreflected	60	109	0.9211
55	Unreflected	70	127	0.9141
55	Unreflected	80	145	0.9136
55	Unreflected	100	182	0.9034
60	Unreflected	5	8	0.8260
60	Unreflected	10	17	0.8687
60	Unreflected	15	25	0.8883
60	Unreflected	20	33	0.9093
60	Unreflected	25	42	0.9177
60	Unreflected	30	50	0.9211
60	Unreflected	40	67	0.9310
60	Unreflected	50	83	**0.9320**
60	Unreflected	60	100	0.9270
60	Unreflected	70	117	0.9276
60	Unreflected	80	133	0.9277
60	Unreflected	100	167	0.9128

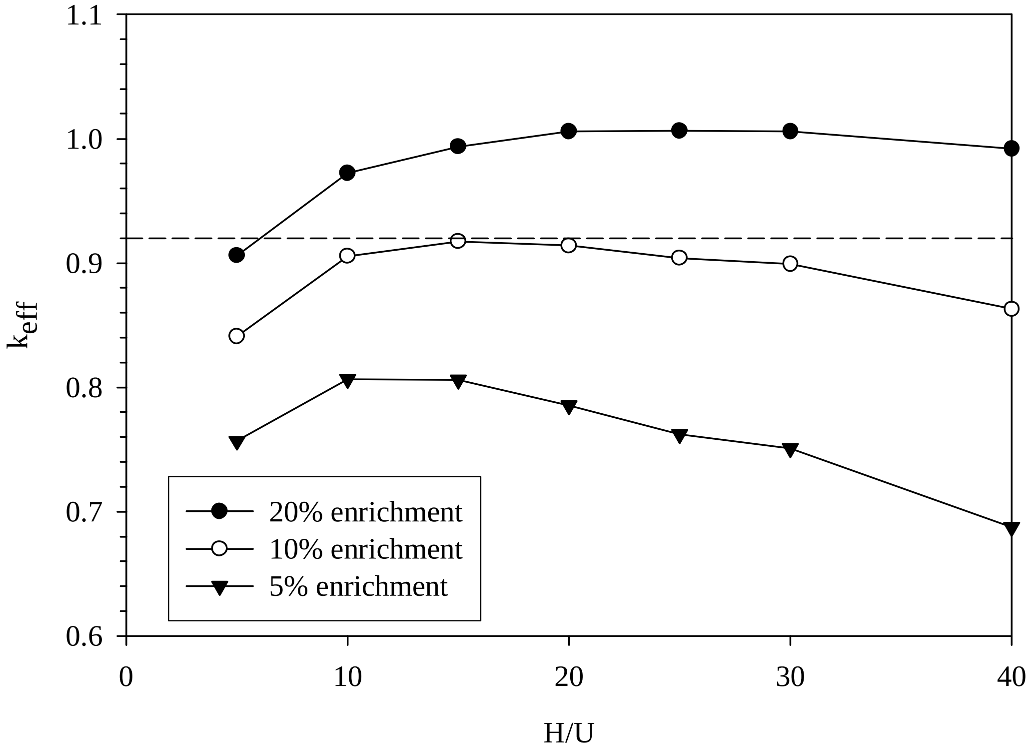

Fig. 2. Calculated k_{eff} values for water-reflected UO_2F_2 deposits.

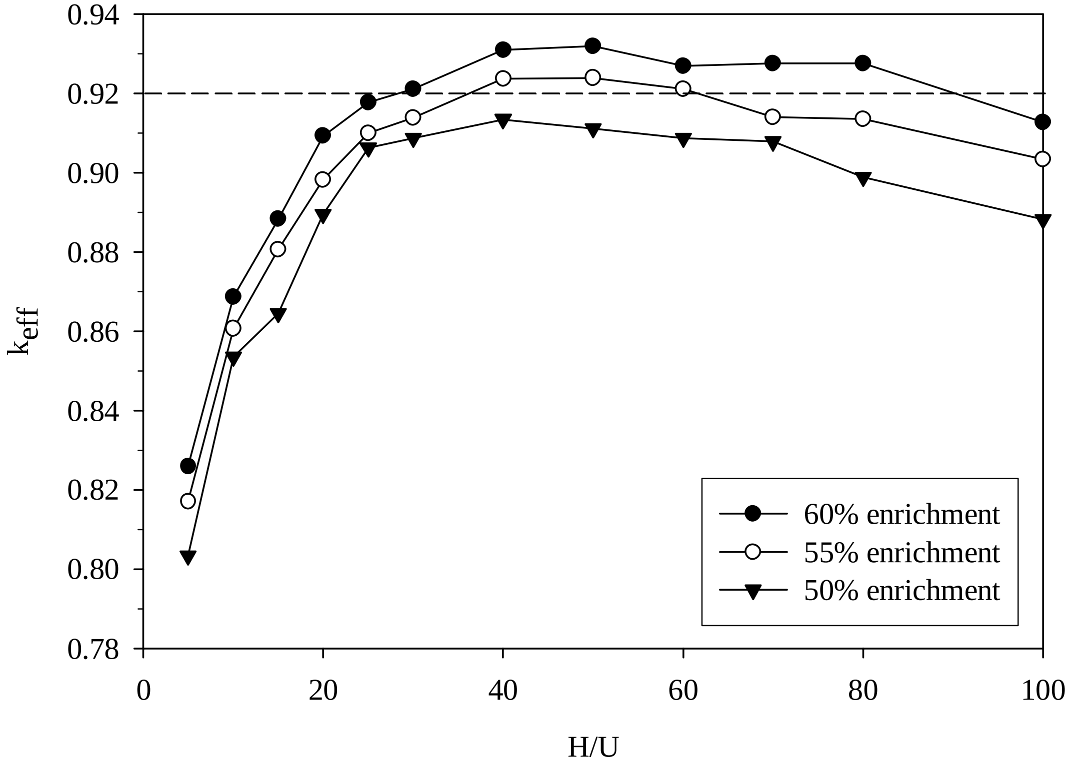

Fig. 3. Calculated k_{eff} values for unreflected UO_2F_2 deposits.

4. CONCLUSIONS

The results given in this report are based on the assumption that UF_6 enriched beyond 5 wt % ^{235}U cannot enter the 8-in.-ID piping sections. For the 8-in.-ID pipe evaluated in this report, solid deposits of UF_6 do not present a criticality hazard. Solid deposits of UO_2F_2 will not be a criticality risk if the enrichment stays below 10 wt % ^{235}U. Also, with the enrichment monitoring equipment in place, it is probable that the formation of a solid deposit would be detected before it became large. Other administrative controls, such as material balance, could also be used to ensure that a large solid deposit does not form.

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