GE-Hitachi Nuclear Energy
Global Laser Enrichment

An Estimate of Minimum Critical Water Content for 10% Enriched UF6 in 30-Inch and 48-Inch Cylinders

Lon Paulson and James Degolyer

Integration of Nuclear Criticality Safety into New Facility Design

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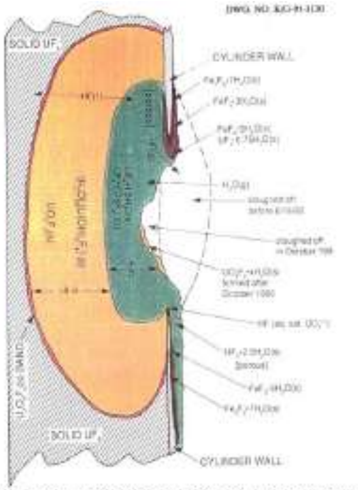
Wilmington Site

Introduction

- Planned GE Hitachi Global Laser Enrichment LLC (GLE) commercial facility will required outside pad storage of natural feed, enriched product, and tails involving both 30- and 48-inch cylinder designs.
- Small breaches adjacent to solid UF_6 result in chemical reactions which are self-plugging due to chemical reactions between UF_6, water, and metal.
- However, if large breach occurs adjacent to the void space at the top of a solid UF6 cylinder would be oriented to enable significant rainwater intrusion (worst case rainfall conditions).
- Cylinders with assays between 5-10% would require ~ 80 liters according to NUREG-1851 for a 10-ton cylinder (simple estimate based on safe slab layer per ANSI/ANS-8.1 and percent H2O consumed in reaction).
- Rather than a simple estimate, a more technical (computational) approach was desired to estimate the minimum mass of water to cause criticality.

Methodology

Chemistry of UF6 and Water:



Breached UF6 Cylinder Hydrolysis / Reduction Re

- The actual chemical reaction of UF_6(s) with excess H2O is a complicated affair (POEF-2086).
- For solid UF_6 and liquid water to form a solution, excess H2 is required (USEC-651):

$$UF_{6,solid} + (1602)H_2O_{liquid} \longrightarrow UO_2F_2(aq, 4HF/1600H_2O)_{liquid} + heat$$

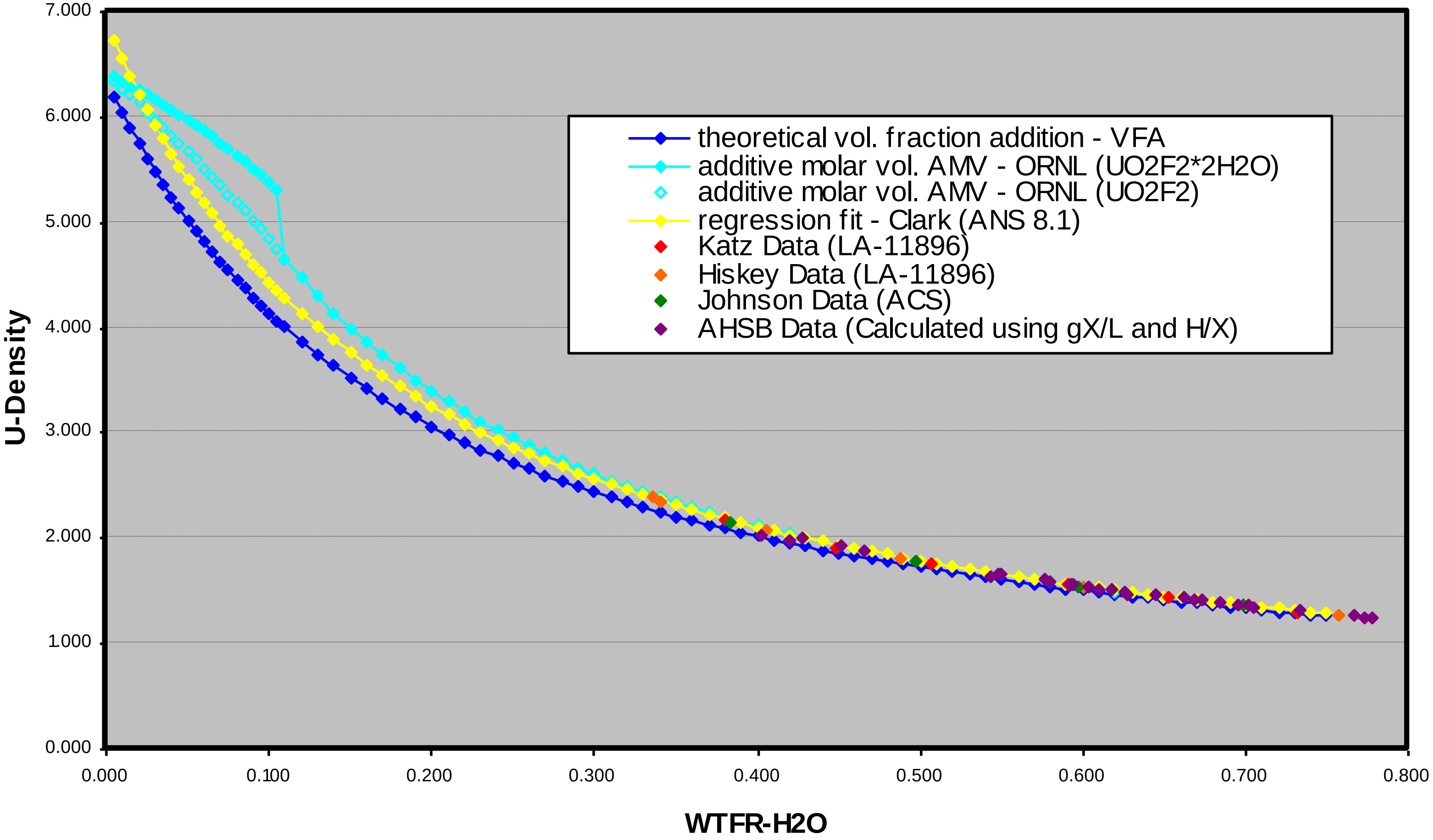
- To complicate matters further, we know that reacting UO2F2 with water results in hydrates; the resulting solution is not volume additive. Volume fraction addition (VFA) does not reflect dissolving effects.
- This work uses the more accurate additive molar volume (AMV) method; correlation developed by Barber, et. al. (ORNL/TM-12292)

$$\rho_U = 4.96 - 0.32 \cdot \frac{H}{U} \qquad \frac{H}{U} \le 4$$

$$\rho_U = \frac{M_U}{\frac{M_{UO_2F_2 \cdot 2H_2O}}{\rho_{UO_2F_2 \cdot 2H_2O}} + \left(\frac{H}{U} - 4\right)\frac{M_{H_2O}}{2 \cdot \rho_{H_2O}}} \qquad \frac{H}{U} > 4$$

Methodology

Chemistry of UO_2F_2 and Water: VFA vs. AMV



Methodology – Key Assumptions I

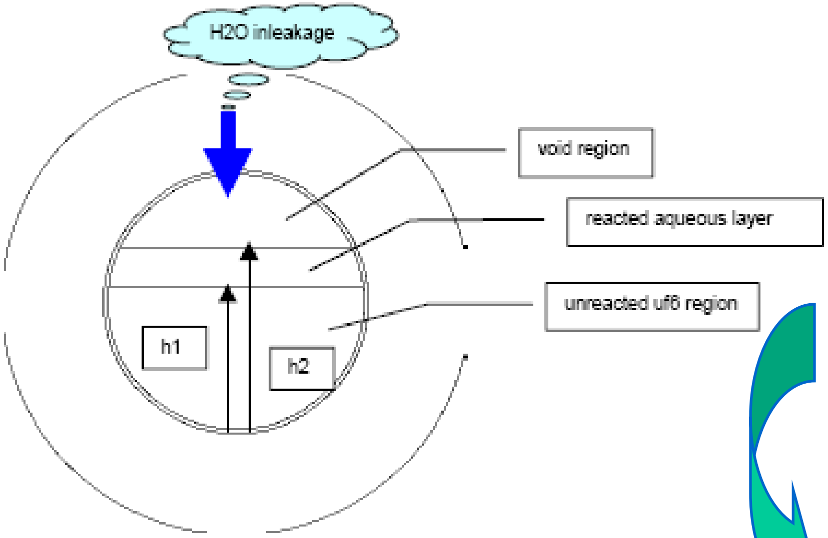
- **Cylinder**
  - A right circular cylinder (RCC) geometry [YCYLINDER] is used. The length of the cylinder is adjusted to correspond to the minimum certified volume in each respective cylinder while the inner/outer radius is assumed constant.
  - The initial content of the cylinder corresponds to the maximum net weight of 2,277 (30B) or 12,501 kgs (48Y) UF_6, respectively.
  - For all calculations, the enrichment is conservatively assumed 10.0 wt.% U235. The initial purity correspond to enriched commercial grade UF_6 consisting of 99.5 wt.% $U(10)F_6$ + 0.5 wt.% HF impurity

Methodology – Key Assumptions II

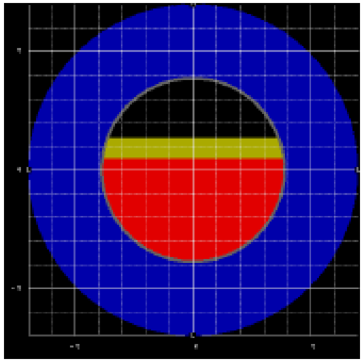
- **Chemical Reaction**
  - The chemical reaction between UF_6 (s) + H_2O reacts to completion (e.g. sufficient solid UF_6 exists inside cylinder)
  - The chemical reaction must stoichiometrically balance
  - The mass of water in-leakage, and weight fraction free water in products are variable.
  - The spatial distribution of the resultant chemical reaction products will form a “slab” layer on top of the unreacted UF_6 with variable thickness (depending on the weight fraction of free water/fraction of water reacted). The H/U ratio of the reacted layer containing $UO_2F_2{*}2H_2O$ is assumed to be ≥ 4 (fully hydrated) and homogeneous.
  - For each weight fraction free water in reacted products, the AMV method is used to compute the uranium density in the $UO_2F2{*}2H_2O$ + H_2O products
  - Once the AMV derived uranium density is computed, the HF (aq.) is then added to the mixture to compute the final mixture density characteristics (atomic number densities) using the VFA method. Solubility suppression effects of HF are ignored.

Methodology – Model Theory: H2O In-Leakage

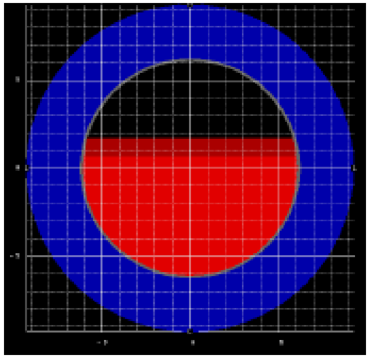
- GEMER Model Construct



how do you calculate h1 and h2 (for given h2o mass inleakage)?

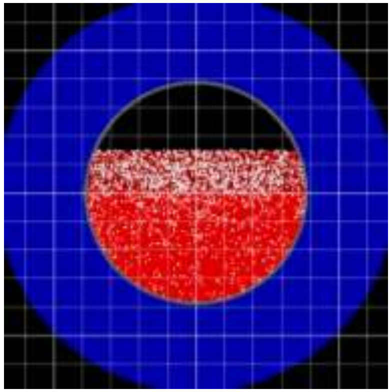


Model 30B (case 30b7020)



Model 48Y (case 48y20000)

example fission dist'n ➔



Methodology – Model Theory: H2O In-Leakage

- **Stoichiometric Balance: 2 Equations, 2 Unknowns**

If we denote (y) as the number of moles of solid UF6 that react, and (2y+x) as the total number of moles which correspond to the total mass of H2O inleakage that occurs, then the general stoichiometric balance (for excess water inleakage) can be written as,

(1) $$yUF_{6,solid} + (2y + x)H_2O_{inleakage} \longrightarrow yUO_2F_2 \bullet 2H_2O_{aq} + 4yHF_{aq} + (x - 2y)H_2O_{aq}$$

For a given mass of H2O inleakage (m_o), we can determine the weight fraction free H2O (w) in the aqueous layer from the following expression,

$$w = \frac{k^*{}_{h_2O}}{\left[k^*{}_{h_2O} + k_{uo2f2*2h2o} + k_{hf}\right]}$$

We now required one more equation to solve for unknown molar values of x, y [note: k_o=initial moles H2O inleakage]: $x = \mathrm{k_o} - 2y\{moles\}$

Solving for y in terms of the known initial mass of H2O inleakage (k_o) and variable weight fraction free H2O (w) in the reacted aqueous layer, the final general expression for y moles (after some algebra) can be shown to be,

$$y = \frac{k_o M_1 (1 - w)}{\left[4M_1(1 - w) + wM_2 + 4wM_3\right]}$$

Where subscripts 1, 2, 3 denote H_2O, $UO_2F_2{*}2H_2O$, and HF compounds. Once y moles is determined, then x moles may be determined from above.

Methodology – Model Theory: H2O In-Leakage

- **Compute final mixture density in reacted layer**

To summarize at this stage, at each assumed weight fraction (w) in the reacted aqueous layer, solve equation for y = f(w, k_o), then solve equation for x = f(k_o,y).

Given the molar values of x,y that satisfy the general chemical reaction stoichiometric equation, the moles of each constituent in the reacted aqueous layer may be determined.

First the density of uranium in the reacted aqueous layer is computed using the ORNL AMV method. This uranium density is used only as an intermediate result for the reacted aqueous products

Since the mass of the each of the constituents are known, and the AMV u-density is known, the volume of constituents may be computed. Likewise, from the molar solution of the general stoichiometry relation, the moles [mass] of HF generated is known; the volume of HF in the reacted aqueous layer may be computed.

The final mixture density is computed using this intermediate AMV result and the traditional VFA approach to add in the final mixture density effect of aqueous HF.

$$\rho^{VFA}{}_{mix} = \rho^{AMV}{}_{mix} + \rho_{hf}$$

Methodology – Model Theory: H2O In-Leakage

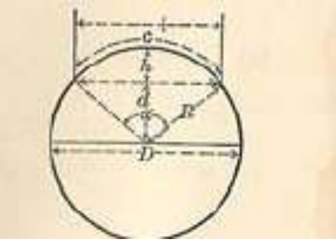
- **Compute unreacted / reacted aqueous layer mixture heights**

To compute h1, we must subtract the mass of UF6 consumed in the reaction from the original net weight of UF6 inside the cylinder. Since we know the moles of UF6 consumed (y), we know the mass consumed (kgs). The cross sectional area corresponding to the unreacted UF6 inside the cylinder can be used to determine the segment$_1$ area, representing void above unreacted UF6(s) where units include y=mole, R=cm, M=g/mole, rho=g/cm3, L=cm:

$$A_{segment} = A_{cyl} - A_{unreacted,uf6\_mix} = \pi R^2{}_{cyl} - [yM_{uf6} * 1000 / \rho_{uf6\_mix} / L_{cyl}]\{cm^2\}$$

Solve for h = h$_{void}$, using a root solver. The height (h1) corresponding to the top of the unreacted UF6 mass is then given by:

$$h_1 = h_{mix} = 2R - h_{void}$$



$$A_{segment} = R^2 \cos^{-1}[\frac{R-h}{R}] - (R-h)\sqrt{2Rh-h^2}$$

General circle mensuration formula (The Engineer's Manual, circa 1917).

To compute h2, we first compute the volume (L) of each of the constituents in the final reacted aqueous layer mixture, then add this volume to above unreacted volume;

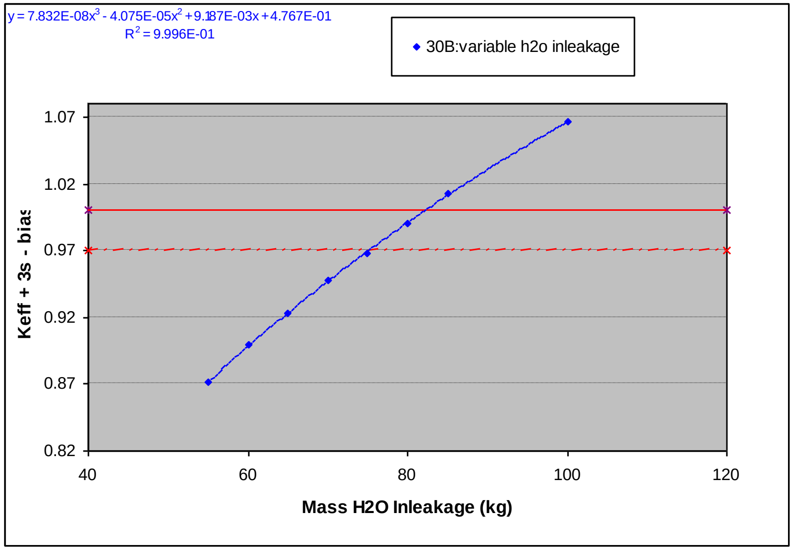
$$V_{mix,aq.layer} = V_{uo2f2*2h2o+h2o,aq.layer} + V_{hf,aq.layer}$$

$$A_{tot} = [A_{unreacted,uf6\_mix} + V_{mix,aq.layer} \bullet 1000 / L_{cyl}]\{cm^2\}$$

$$A_{segment} = A_{cyl} - A_{tot}\{cm^2\}$$

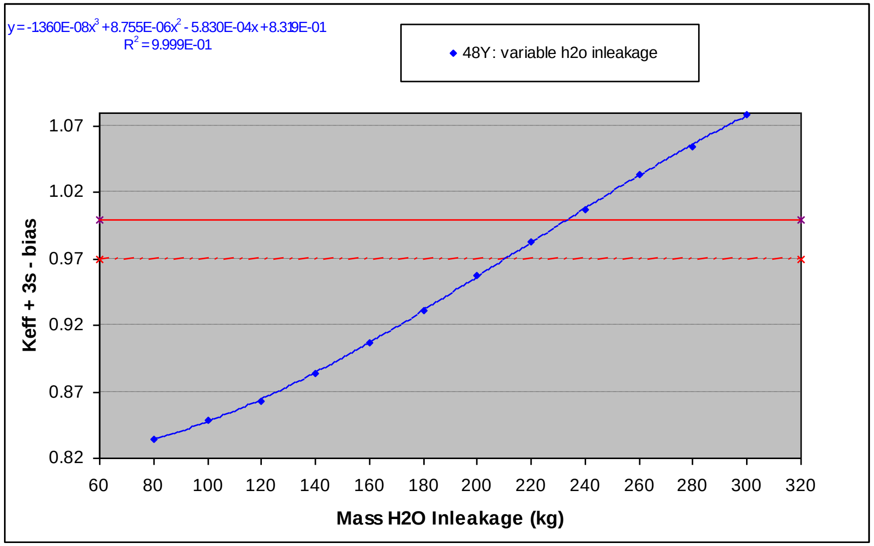
$$h_2 = h^*{}_{mix} = 2R - h_{void}$$

Results



MODEL 30B UF6 CYLINDER: 10% ENR

•the estimated maximum k+3σ-bias ≤ 0.97 safe limit intercepts occurs at 75.1 kgs H_2O
•the estimated maximum k+3σ-bias = 1.0 critical limit intercepts occurs at 82.2 kgs H_2O.

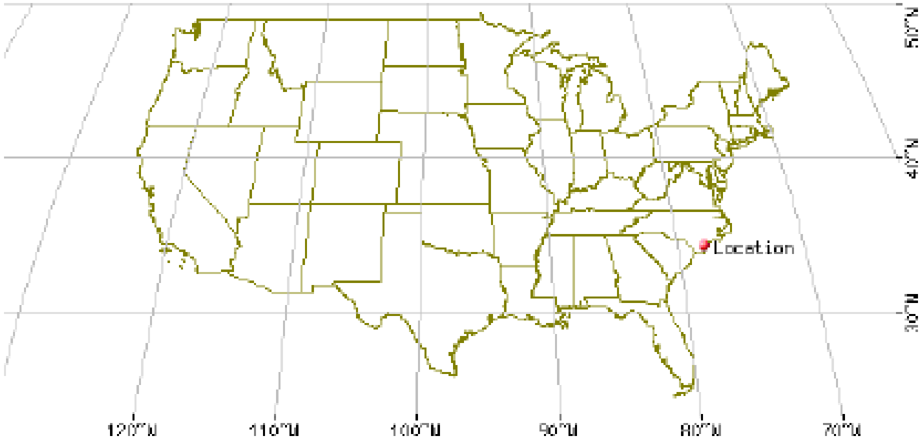


MODEL 48Y UF6 CYLINDER: 10% ENR

•the estimated maximum k+3σ-bias ≤ 0.97 safe limit intercepts occurs at 210.3 kgs H_2O
•the estimated maximum k+3σ-bias = 1.0 critical limit intercepts occurs at 233.5 kgs H_2O.

Application to ILM

- To evaluate the practical aspects of the above results, we need to establish a reasonable *response time* for operations under credible “cylinder breach” conditions. The first step is to determine maximum historical rainfall rate in a 24-hour period from available records for the Wilmington, NC



$$V_{h2o\{L\}} = A_{breach\{cm^2\}} \bullet Rate_{\{cm/hr\}} \bullet t_{\{hr\}} /1000$$

NOAA Point Precipitation Frequency Estimates:

Average Recurrence Interval (ARI) for Wilmington, NC over last 1000 years is ~22 inches in a 24 hour-period.

Application to ILM

- **Time interval evaluation summary (6 to12-inch hole size)**

Parameter Description	Model 30B Cylinder: 2,277 kgs U(10)F6				Model 48Y Cylinder: 12,501 kgs U(10)F6			
	Critical H2O Mass		**Safe H2O Mass**		**Critical H2O Mass**		**Safe H2O Mass**	
Water Volume (L)	82.2		75.1		233.5		210.3	
Worst case rainfall rate (inches per 24 hrs)	22		22		22		22	
Worst case rainfall rate (inches per hr)	0.9167		0.9167		0.9167		0.9167	
Worst Case Rainfall (cm per hr)	2.3283		2.3283		2.3283		2.3283	
Breach Diameter (inches)		Area (cm2)		Area (cm2)		Area (cm2)		Area (cm2)
Case 1	6.00	182.41	6.00	182.41	6.00	182.41	6.00	182.41
Case 2	8.00	324.29	8.00	324.29	8.00	324.29	8.00	324.29
Case 3	10.00	506.71	10.00	506.71	10.00	506.71	10.00	506.71
Case 4	12.00	729.66	12.00	729.66	12.00	729.66	12.00	729.66
Time Required for Water Volume (L) Intake	Hours	Days	Hours	Days	Hours	Days	Hours	Days
Case 1	193.54	8.06	176.82	7.37	549.77	22.91	495.15	20.63
Case 2	108.87	4.54	99.46	4.14	309.25	12.89	278.52	11.61
Case 3	69.67	2.90	63.66	2.65	197.92	8.25	178.25	7.43
Case 4	48.38	2.02	44.21	1.84	137.44	5.73	123.79	5.16

Conclusions – I

- A methodology to compute unreacted / reacted aqueous layers formed inside a UF6 cylinder has been established using hydrolysis chemical reactions and conservation of mass.

- Breach hold sizes of 6 to 12-inch diameters were evaluated; the corresponding time intervals under postulated "worst-case" rainfall conditions for ILM area are summarized as follows:

30B: critical mass of H_2O inleakage	requires 2-8 days.
30B: maximum safe mass of H_2O inleakage	requires 1.8-7.3 days
48Y: critical mass of H_2O inleakage	requires 5.7-22.9 days
48Y: maximum safe mass of H_2O inleakage	requires 5.1-20.6 days

- Upon discovery an administrative response time to "cover" cylinder within 8-hours is reasonably justified.

Conclusions - II

- Additional fire protection measures should be factored into emergency procedures to prohibit “high pressure” hose streams on cylinder storage pads.
- Chocked or saddle designs for 48-inch cylinder storage in 1-high or 2-high planar arrays should be engineered to provide sufficient spacing to prevent (7 ‘o clock position) contact of adjacent cylinder by lifting lug.