

CALCULATIONS ON THE CRITICALITY ACCIDENT AT TOKAI MURA

On 30 September 1999, a criticality accident occurred at the Japanese fuel fabrication plant at Tokai Mura. Operators transferred an aqueous uranium solution (previously enriched from 5 to 20% in ^{235}U) from a dissolution column to a precipitation tank (with a much less safe geometry). During the filling process, the reflective effect of the water circuit around the tank brought the whole system into a prompt super-critical state, causing a strong power excursion. It was not until about 20 hours after the start of the accident that the water circuit was emptied and a concentrated boric acid solution was added.

Here, we aim at reconstructing the evolution of the effective multiplication factor k_{eff} (before, during and after the accident) by simple calculations based on the one-group diffusion approximation diffusion in a homogeneous environment. This exercise is very similar to the work carried out by the neutron experts sent by the Japanese safety authority that day, who had only a few hours to understand and then neutralise the accident. In the aqueous solution of uranyl nitrate, only the reactions of neutrons on the hydrogen of water and the ^{235}U of uranium are to be taken into account. We give $\sigma_a^{\text{H}} = 0.3$ b for hydrogen. We give $\nu = 2.4$, $\sigma_f^5 = 580$ b and $\sigma_a^5 = 680$ b for ^{235}U . In the whole problem, the $N_{\text{H}}/N_{\text{U}}$ ratio of the atomic density of hydrogen to that of enriched uranium in solution is fixed at 400. "e" is the enrichment of uranium to ^{235}U (atomic density $N_5 = e \cdot N_{\text{U}}$ in solution). L is the neutron scattering length in solution (with $L^2 = D/\Sigma_a$) and $L_{\text{H}} = 2.8$ cm their scattering length in light water (with $L_{\text{H}}^2 = \frac{D_{\text{H}}}{\Sigma_a^{\text{H}}} \approx \frac{D}{\Sigma_a^{\text{H}}}$).

A. Enrichment of uranium in the dissolution column.

A.1. Influence of enrichment e on the k_{∞} factor

Retrieve $N_{\text{H}} = 6.69 \cdot 10^{22} \text{ cm}^{-3}$ approximately. Justify the validity of the one-thermal-group diffusion approximation, and that the fast fission factor ϵ and the resonance escape probability factor p are equal to 1 in this case. Give the expression of the thermal utilization factor f , the reproduction factor η and the infinite multiplication factor k_{∞} according to the microscopic data provided, $N_{\text{H}}/N_{\text{U}}$ and e . Give the value of k_{∞} for $e = 5\%$ and then 20% (the values of k_{∞} and k_{eff} will only be rounded to the nearest 100 pcm in the following). Comment on the first risk caused by uranium enrichment. Why is this risk limited in the dissolution column?

Solution

$$N_{\text{H}} = 2N_{\text{H}_2\text{O}} = 2 \times \frac{\rho_{\text{H}_2\text{O}} \cdot N_{\text{A}}}{M_{\text{H}_2\text{O}}} = 2 \times \frac{1 \times 6.022 \cdot 10^{23}}{18} = 6.69 \cdot 10^{22} \text{ cm}^{-3}$$

- uranium aqueous solution: homogeneous medium with fissile material

- $\epsilon = 1$, because of the one-group approximation so only the thermal neutrons are fissioning.

- $p = 1$, because we are considering all n to be thermal

$$-f = \frac{\Sigma_a^{fuel}}{\Sigma_a^{tot}} = \frac{eN_U\sigma_a^5}{eN_U\sigma_a^5 + N_H\sigma_a^H} \rightarrow f_{5\%} = 0.2208, f_{20\%} = 0.5314$$

$$-\eta = \frac{\nu\Sigma_f}{\Sigma_a^{fuel}} = \frac{2.4 \times eN_U\sigma_f^5}{eN_U\sigma_a^5} = 2.047$$

$$k_\infty = \epsilon p f \eta \rightarrow k_{\infty,5\%} = 0.451, k_{\infty,20\%} = 1.088 > 1$$

The risk is limited in the dissolution column which is prone to high leakage.

A.2. Calculation of the keff factor in the dissolution column.

Express k_{eff} as a function of k_∞ , L^2 and the geometrical buckling B_g^2 of the column. What does the ratio k_{eff}/k_∞ represent? Simply express L^2 as a function of L_H^2 and f , then do the numerical application for $e = 5\%$ and then 20% (we will take $L^2 = 3.69 \text{ cm}^2$ for $e = 20\%$ in the following). The column (which is assumed to be completely filled with the solution) is a finished cylinder with a radius of 10 cm and a height of 80 cm. Express B_g^2 according to radius and height and then apply digitally. Deduct the k_{eff} value for $e = 5\%$ then 20% .

Solution

$$k_{eff} = \frac{k_\infty}{1 + L^2 B_g^2}, \text{ valid if } > 1$$

B_g : geometrical buckling

k_{eff}/k_∞ represents the fast and thermal leakages.

L : diffusion length of neutrons in solution.

L_H : diffusion length of neutrons in light water.

$$L^2 = \frac{D}{\Sigma_a^{tot}}, L_H^2 = \frac{D}{\Sigma_a^H} \Rightarrow \frac{L^2}{L_H^2} = \frac{\Sigma_a^H}{\Sigma_a^{tot}} = 1 - f \Rightarrow L^2 = L_H^2(1 - f) = 3.69 \text{ cm}^2$$

$$B_g^2 = \chi_R^2 + \chi_H^2 = \left(\frac{2.405}{R}\right)^2 + \left(\frac{\pi}{H}\right)^2 = 593.8 \text{ m}^{-2}$$

$$\Rightarrow k_{eff} = \frac{k_\infty}{1 + (1 - f)L_H^2 B_g^2} = 0.310 (5\%), 0.892 (20\%)$$

B. Approach to criticality in the precipitation tank.

B.1. Calculation of keff in the tank being filled

In the following $e = 20\%$. We forget for the moment that the tank is surrounded by a water circuit. The operators start by transferring the entire volume of the column into the tank of radius $R = 20 \text{ cm}$ and height $H = 40 \text{ cm}$. Up to what height is the tank filled in this way and

what is k_{eff} worth? Filling continues until 4/5 (height h). Check that the k_{eff} is very close to 1 at this point.

Solution

$$V_t = 51522 \text{ cm}^3 = H \times \Pi R^2$$

$$V_c = 25133 \text{ cm}^3 = h_0 \times \Pi(R/2)^2 \Rightarrow \frac{V_c}{V_t} = 0.4878 = \frac{h_0}{4H} \Rightarrow h_0 = 20 \text{ cm}$$

$$k_{eff} = \frac{k_\infty}{1 + (1-f)L_H^2 Bg^2} \text{ with } Bg^2 = \left(\frac{2.405}{R}\right)^2 + \left(\frac{\pi}{h_0}\right)^2 = 403.84 \text{ m}^{-2}$$

So, with $e = 20\%$, $k_{eff} = 0.950$

-“filling continues until 4/5...”

$$\Rightarrow h = \frac{4}{5} \cdot H = 32 \text{ cm} \Rightarrow Bg^2 = 240.98 \text{ m}^{-2} \Rightarrow k_{eff} = 0.999$$

B.2. Calculation of stationary flow in the critical tank

In this (almost) critical state, the total fission power is only about 1W. We note $\alpha = 2.505/R$ and $\beta = \pi/h$. Give the expression of the ϕ_{rz} flux in the tank for $0 < r < R$ and $-h/2 < z < h/2$. At what point (r,z) in the tank is the flow maximum? Calculate this value from the total fission power, integrating the expression of the power density over the whole tank. We will integrate according to the variables $x = \alpha r$ and $y = \beta z$, knowing that the primitive of $xJ_0(x)$ is $xJ_1(x)$ (J_0 and J_1 Bessel functions) and that $J_1(2.405) = 0.5$ approximately.

Solution:

$$\phi_{rz} = \phi(r, z) = \phi_{00} \cdot J_0(\alpha r) \cdot \cos(\beta z), \quad \text{with } 0 < r < R \text{ and } -\frac{h}{2} < z < \frac{h}{2}$$

$$\alpha = \frac{2.405}{R} \text{ and } \beta = \pi/H$$

$J_0(\alpha r)$ is max when $r = 0$

$$\frac{\partial}{\partial z} [\cos(\beta z)] = 0 \Rightarrow -\beta \sin(\beta z) = 0 \Rightarrow z = 0 \text{ or } \beta z = \pi \Rightarrow \frac{\pi}{H} z = \pi \Rightarrow z = H$$

But $-\frac{h}{2} < z < \frac{h}{2}$, so the only possible solution is $z = 0$.

Therefore, the flux is maximal at the center of the tank:

$$\phi_{max} = \phi_{00} = \phi(r = 0, z = 0)$$

-Total fission power: $P_{volumic} = Q \Sigma_f \phi_{rz}$

$$\Rightarrow P \cdot dV = Q \Sigma_f \phi_{rz} \cdot 2\pi R dr dz$$

-“the total fission power is only about 1W”:

$$1 [J \cdot s^{-1}] = \phi_{00} 2\pi Q \Sigma_f \int_0^R J_0(\alpha r) r dr \int_{-\frac{\beta H}{2}}^{\frac{\beta H}{2}} \cos(\beta z) dz$$

$x = \alpha R$ and $y = \beta z$:

$$1 = \phi_{00} 2\pi Q \Sigma_f \int_0^{\alpha R} J_0(x) \frac{x dx}{\alpha^2} \int_{-\frac{H}{2}}^{\frac{H}{2}} \cos(y) \frac{dy}{\beta} = \phi_{00} \frac{2\pi Q \Sigma_f}{\alpha^2 \beta} [x J_1(x)]_0^{\alpha R} \cdot [\sin(y)]_{-\frac{\beta H}{2}}^{\frac{\beta H}{2}}$$

$$= \phi_{00} \frac{2\pi Q \Sigma_f}{\alpha^2 \beta} \cdot \alpha R \times 0.5 \cdot 2 \sin \sin \frac{\beta H}{2} = \phi_{00} \frac{2Q \Sigma_f}{2.405} R^2 H$$

with $Q = 200 \text{ MeV}$ and $\Sigma_f = e N_U \sigma_f^5$: $\phi_{00} = 1.21 \cdot 10^8 \text{ cm}^{-2} \text{ s}^{-1}$

C. Divergence then extinction by adding boric acid.

C.1. Reflector effect of the water circ

Around the tank, there is in fact a cooling water circuit a few cm thick which acts as an infinite reflector. Knowing this, estimate a minimum value for this thickness and justify it. It is quite easy to show (and we admit it here) that at an unchanged height the radius of the equivalent bare tank is about 23 cm. From this, the new value of the k_{eff} can be deduced. Recalling the proportion of delayed neutrons among the fission neutrons of the ^{235}U , explain why this last k_{eff} value is problematic.

Solution

Infinite reflector: thickness about 2 to 3 L

$$R = 23 \text{ cm} \Rightarrow Bg^2 = 222.72 \text{ m}^{-2} \Rightarrow k_{\text{eff}} = 1.006$$

For U-235, $\beta_{\text{delayed}} = 0.00640$ so it corresponds to a +1\$ reactivity insertion.

$$n(t) = n_0 \exp\left(\frac{k_{\text{eff}} - 1}{l_p + \beta_d \tau_d} t\right) = n_0 \exp(0.0187t)$$

with $l_p = 10^{-4} \text{ s}$ which means that prompt neutrons are emitted 10^{-4} s after a neutron hit a U-235 nucleus, and $\tau_d = 50 \text{ s}$ being the delayed neutrons mean emission time.

So, the mean time between 2 generations of neutrons is $\tau = l_p + \beta_d \tau_d$

$$\Rightarrow \frac{n(t)}{n_0} = 2 = \exp(0.0187 t_{\text{double}}) \Rightarrow t_{\text{double}} = 37 \text{ s}$$

With such a reactivity insertion, the power is doubled every 37 seconds!

C.2. Boron is added

Once the water circuit is empty, the vessel has returned to the very slightly subcritical state calculated in B. Starting from this state, we wish to reduce the k_{∞} to 0.3 by completing the

volume of the tank with a solution of boric acid $B(OH)_3$ (B natural boron has 20% of ^{10}B with $\sigma_a^{10} = 3800$ b, and the rest of ^{11}B whose absorption is negligible). Based on the homogeneous solution that fills the whole tank after the operation, calculate the atomic density of natural boron N_B in the concentrated solution added to reach the targeted k_∞ . Deduce the total mass of boric acid required.

Solution:

We want $k_\infty = 0.3$ by adding Boric acid:

$$k_\infty = \epsilon p f \eta = f \eta = \frac{\Sigma_a^{fuel}}{\Sigma_{tot}^{fuel}} \cdot \frac{v \Sigma_f}{\Sigma_a^{fuel}} = \frac{v \Sigma_f}{\Sigma_a^{tot}}$$

$$= \frac{v N_5^{homogeneous} \sigma_f^5}{N_H \sigma_a^H + N_5^{homogeneous} \sigma_a^5 + N_{10}^{homogeneous} \sigma_a^{10}} = 0.3$$

Where:

$$N_5^{homogeneous} = e N_U \quad \text{and} \quad N_{10}^{homogeneous} = 0.2 N_B$$

$$\Rightarrow N_B = 10^{10} \text{ cm}^{-3}$$