

Physics of Nuclear Reactors

Tokaimura Accident

Daniele Timpano
Milica Krstovic

CALCULATIONS ON THE CRITICALITY ACCIDENT AT TOKAIMURA

- A.1. Influence of enrichment e on the k_{∞} factor
- A.2. Calculation of the k_{eff} factor in the dissolution column.
- B.1. Calculation of k_{eff} in the tank being filled
- B.2. Calculation of stationary flux in the critical tank
- C.1. Reflector effect of the water circuit
- C.2. Boron is added

Divide in groups of 5:

- *We will do Point A.1 together at the board*
- *I will leave you time for Point A.2 and B.1 (10 minutes)*
- *We will do Point B.2 and C.1 together*
- *I will leave you time for Point C.2 (10 minutes)*

Few concepts from:

1. Four factor formula
2. One-group reactor theory
3. Reactor kinetics

The Tokaimura Accident (1/3)

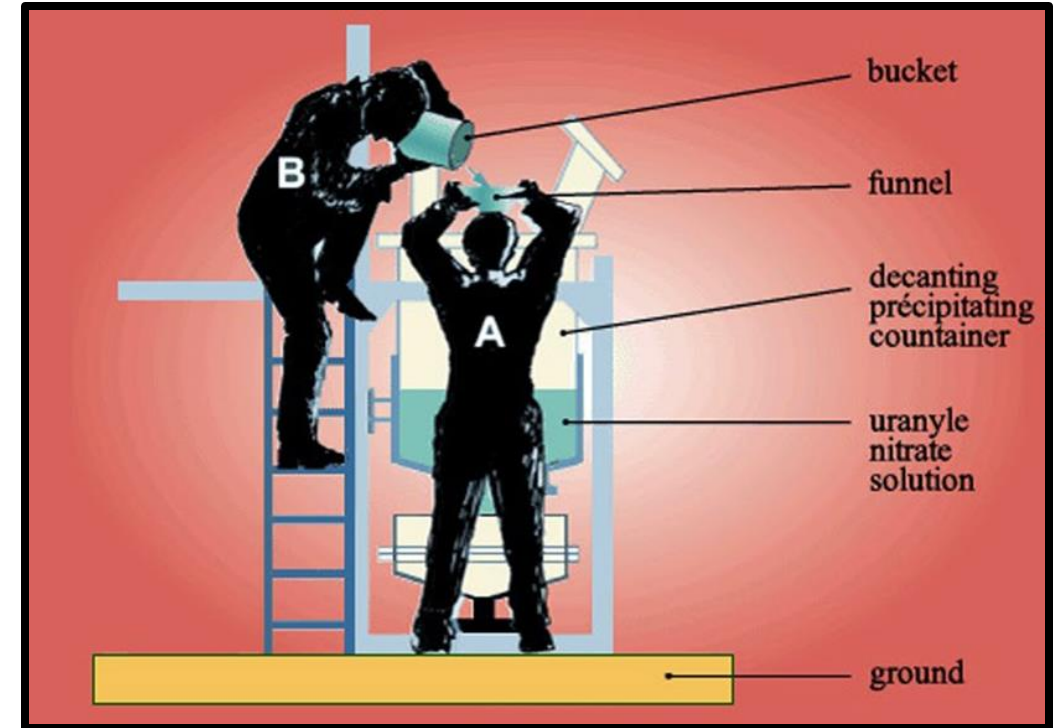
On 30 September 1999, a criticality accident occurred at the **Japanese fuel fabrication plant at Tokaimura**. Operators transferred an *aqueous uranium solution* (previously enriched from 5 to 20% in ^{235}U) from a dissolution column to a precipitation tank (with a much less safe geometry).

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.



The Tokaimura Accident (2/3)

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.



The Tokaimura Accident (3/3)

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 $\sigma_a^H = 0.3 \text{ b}$ for hydrogen. We give $\nu = 2.4$, $\sigma_f^5 = 580 \text{ b}$ and $\sigma_a^5 = 680 \text{ b}$ for ^{235}U . In the whole problem, the N_H/N_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 $N_5 = e.N_U$ in solution). L is the neutron diffusion length in solution (with $L^2 = D/\Sigma_a$) and $L_H = 2.8\text{cm}$ their diffusion length in light water (with $L_H^2 = \frac{D_H}{\Sigma_a^H} \approx \frac{D}{\Sigma_a^H}$).

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

- Retrieve $N_H = 6.69 \cdot 10^{22} \text{ cm}^{-3}$ 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_H/N_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?

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

- Retrieve $N_H = 6.69 \cdot 10^{22} \text{ cm}^{-3}$ approximately.

We know the water density and the molar mass of water ...

$$N_H = 2N_{H_2O} = 2 \times \frac{\rho_{H_2O} \cdot N_A}{M_{H_2O}} = 2 \times \frac{1 \times 6.022 \cdot 10^{23}}{18} = 6.69 \cdot 10^{22} \text{ cm}^{-3}$$

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

- Retrieve $N_H = 6.69 \cdot 10^{22} \text{ cm}^{-3}$ 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_H/N_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?

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

- 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.
 - *one thermal group* is good for uranium aqueous solution: homogeneous medium with fissile material mixed with moderator!!
 - *group reactor approximation*, no fast fission and no resonance escape probability (only thermal neutrons)!

or

- *in highly diluted uranyl solution*: negligible probability of fast fission and efficient moderation (no losses in resonances)

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

- Retrieve $N_H = 6.69 \cdot 10^{22} \text{ cm}^{-3}$ 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_H/N_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?

Exercise A.1 - Influence of enrichment e on the k_∞ factor

$$f = \frac{\Sigma_a^{fuel}}{\Sigma_a^{tot}}$$

$$\eta = \frac{\nu \Sigma^f}{\Sigma_a^{fuel}}$$

- 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_H/N_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).

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

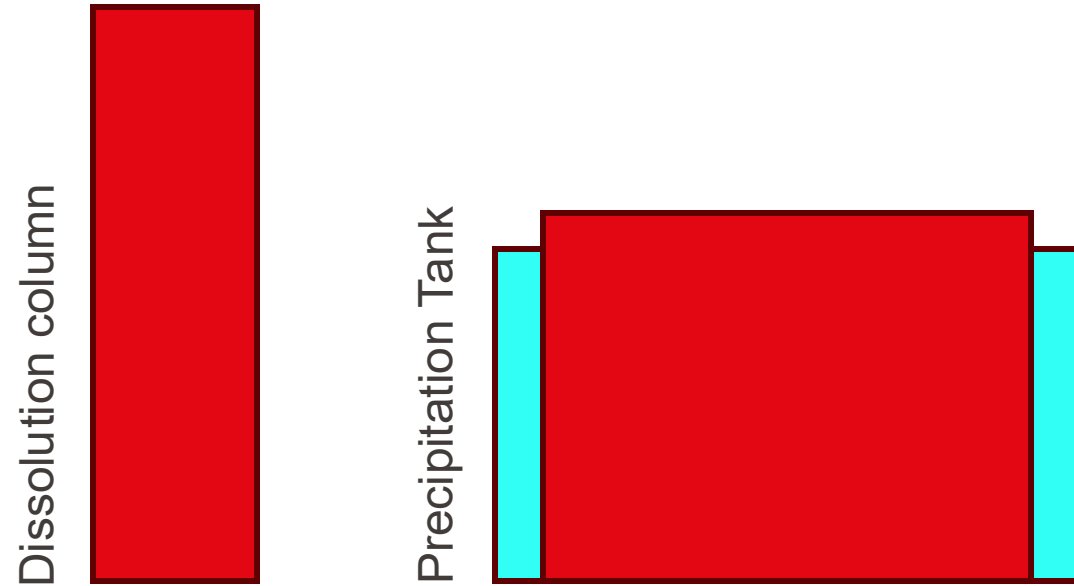
$$\eta = \frac{\nu \Sigma^f}{\Sigma_a^{fuel}} = \frac{2.4 \times eN_U\sigma_f^5}{eN_U\sigma_a^5} = 2.047$$

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

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

- Retrieve $N_H = 6.69 \cdot 10^{22} \text{ cm}^{-3}$ 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_H/N_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?**

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



- **Comment on the first risk** caused by uranium enrichment. Why is this risk limited in the dissolution column?

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

Express k_{eff} as a function of k_{∞} , L^2 and the geometrical buckling B_g^2 of the column. **What does the ratio $k_{\text{eff}}/k_{\infty}$ represent?** \longrightarrow

$k_{\text{eff}}/k_{\infty}$ represents the fast and thermal leakages.

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.** Deduct the k_{eff} value for $e = 5\%$ then 20% .

Assumption: diffusion coefficient is the same in the solution as in water $L_H^2 = \frac{D_H}{\Sigma_a^H} \approx \frac{D}{\Sigma_a^H}$

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

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

Bg: 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.

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

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

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

subcritical in the dissolution column

Exercise B.1 - Calculation of k_{eff} 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 $R = 20$ cm and height $H = 40$ 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.**

Exercise B.1 - Calculation of k_{eff} in the tank being filled

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

$$V_c = 25133 \text{ cm}^3 = h_0 \times \Pi \times (R/2)^2 \Rightarrow \frac{V_c}{V_t} = 0.5 = \frac{h_0}{4H} \Rightarrow h_0 = 2H = 20 \text{ cm}$$

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

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

When “filling continues until 4/5...”

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

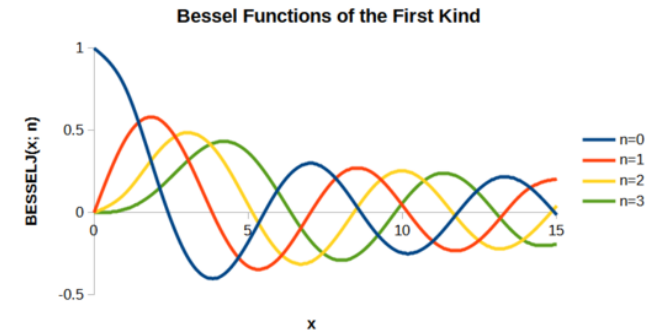
In this (almost) critical state, **the total fission power is only about 1W**. We note $\alpha = 2.505/R$ and $\beta = \pi/h$. Give the expression of the $\phi_{r,z}$ flux in the tank for $0 < r < R$ and $-h/2 < z < h/2$. At what point (r,z) in the tank is the flux maximum? **Calculate this value from 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.

Find where the maximum is:

We know that the solution in a cylindrical reactor:

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

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



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

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

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:**

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

Find what is the maximum

$$\text{Total fission power: } P_{\text{volumic}} = Q\Sigma_f\phi_{rz} \Rightarrow P \cdot dV = Q\Sigma_f\phi_{rz} \cdot 2\pi R dR dz$$

“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 $y = \beta z$:

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

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

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

Exercise C.1 - Reflector effect of the water circuit

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.



Infinite reflector: thickness about 2 to 3 L

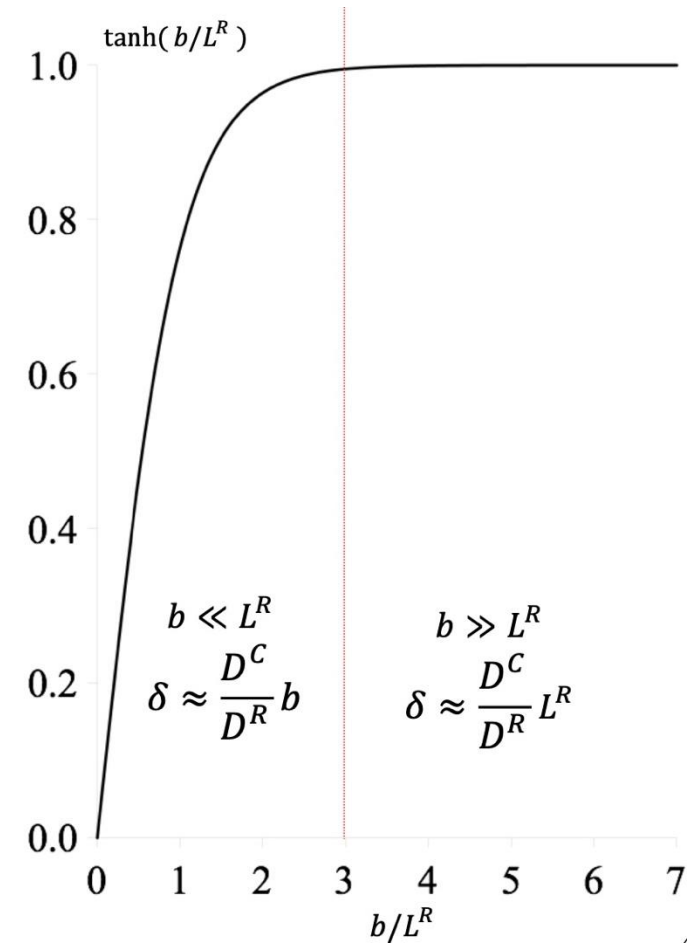
Why? In the reflector:

$$\nabla^2 \Phi - \frac{\Phi}{L^2} = 0$$

Imposing that the flux goes to 0 for $x \rightarrow \infty$:

$$\Phi(x) = B e^{-x/L} \rightarrow \text{reduced to } \sim 5\% \text{ of the flux when } x = 3L$$

(measured from beginning of reflector)



EPFL Exercise C.1 - Reflector effect of the water circuit

It is quite easy to show that, at an unchanged height, the radius of the **equivalent bare tank is about 23 cm (look at reflector savings and diffusion length)**. 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.

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

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

From reactor kinetics with a "simple" addition of delayed neutrons: $n(t) = n_0 \exp\left(\frac{k_{\text{eff}} - 1}{l_p + \beta_d \tau_d} t\right) = n_0 \exp(0.0187t)$

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 two generations of neutrons is $\tau = l_p + \beta_d \tau_d$. **What is the doubling time?**

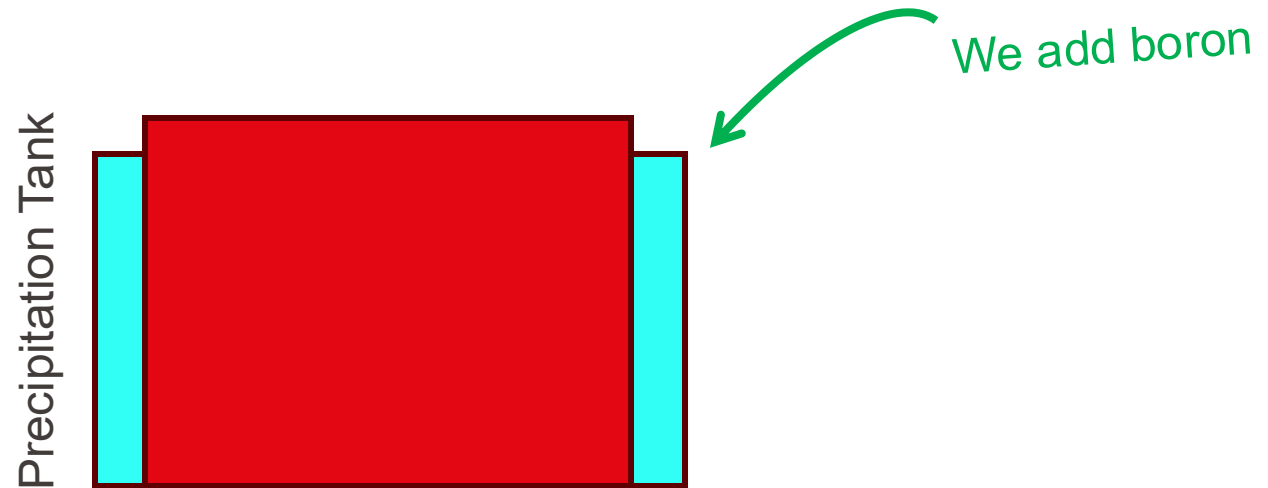
$$\Rightarrow \frac{n(t)}{n_0} = 2 = \exp(0.0187 t_{\text{double}}) \Rightarrow t_{\text{double}} = 37 \text{ s}$$

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

Exercise 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.



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

$$\begin{aligned}
 k_\infty = \varepsilon p f \eta = f \eta &= \frac{\Sigma_a^{fuel}}{\Sigma_a^{tot}} \cdot \frac{v \Sigma_f}{\Sigma_a^{fuel}} = \frac{v \Sigma_f}{\Sigma_a^{tot}} \\
 &= \frac{v N_5^{homogeneous} \sigma_f^5}{N_H \sigma_a^H + N_5^{homogeneous} \sigma_a^5 + N_{10}^{homogeneous} \sigma_a^{10}} = 0.3
 \end{aligned}$$

Where: $N_5^{homogeneous} = e N_U$ and $N_{10}^{homogeneous} = 0.2 N_B$ (only unknown)

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