## Losses, Recycling, and Transmutation

Jan-Olov Liljenzin Professor emeritus Chalmers University of Technology

## Losses, recycling, and transmutation

Which criterion should we use in order to determine the needed actinide reduction in all wastes?

Probable future biological effects from a final repository: Most important are then element like iodine, technetium, and perhaps neptunium

Potential hazard of all waste in ALI:s or Sv: Most important are then the actinides Np, Pu, Am, and Cm

## Future biological effects from a repository

A real risk analysis is almost impossible because:

- Future scenarios leading to any large release of actinides or FPs from the repository can involve unforseen events
- The biological effect of radionuclides spreading in the biospere in a distant future is very hard to quantify

One solution th the second dilemma is to assume that biological evolution is arrested now. Then we can use data on radiation sensitivities and biological effects for the organisms known to exist today.

We have already observed microorganisms that have adapted to life in a highly radioactive solution.

## The use of future potential radiological hazard

- A hazard index can e.g. be constructed from the maximum permissible daily intake of radionuclides that is used in radiation protection today. However, this approach also asumes that the event releasing the potential hazard will occur rather soon, or we are trapped again by the changing nature of life.
- One often used method is to multiply the number of becquerels of each radionuclide by its specific dose factor in Sv/Bq (when ingested) and then sum all calculated values.
- If this method is chosen we need to define some kind of reference level. Often the potential hazard of a typical uranium ore is used, or that of the amount of U consumed in the reactor.

# A hazard index plot based on ALI and 1 kg PWR-UO<sub>2</sub> fuel at 44 MWd/kg



The data in the graph shows that the reduction factors needed for the hazard index of the actinide-free waste to reach the hazard index of U-burnt are:

Neptunium: time ~ 158 000 years, reduction factor ~ 2.5

Am+Cm: time: ~ 400 years, reduction factor ~ 124

Both values seems within reach of a well designed P&T system.

What requirements do this impose on the reprocessing and P&T system i.e. how do we calculate permissible losses?

There are in principle 2 ways to design a separation and transmutation system

1) Batchwise irradiation followed by reprocessing/ separation.

This is the classical way to operate the nuclear fuel cycle. The irradiation target/fuel can in principle be of any type.

2) On-line continuous separation from the target material under irradiation.
Often suggested for future systems.
Requires a liquid fuel e.g. Molten salt, or metallic melt, or perhaps pebbles.

Possible discontinuous P&T with FBR/ADS



It is possible to omit the FBR/ADS unit and recycle to the LWR. Then C feeds the LWR,  $\varepsilon$ C is the consumed actinide in the LWR, and D is the spent target/fuel coming out.

The problem is to determine what fraction of the incoming amount of an actinide that becomes waste

- A = incoming An amount in a single batch, first cycle
- B = (1-r) A
- C = (1-s) B = (1-r)(1-s) A
- $D = (1-\epsilon) C = (1-r)(1-s)(1-\epsilon) A$

(second cycle of An from same batch)

- $B = (1-r) D = (1-r)^2(1-s)(1-\varepsilon) A$
- $C = (1-s) B = (1-r)^2(1-s)^2(1-\varepsilon) A$
- $D = (1-\epsilon) C = (1-r)^2 (1-\epsilon)^2 A$

(third cycle of An from same batch) .... Etc.

 $D = (1-r)^{\infty}(1-s)^{\infty}(1-\varepsilon)^{\infty} A < --- After an infinite number of cycles$ 

#### The total amount lost to waste, R

• 
$$R = A (r + r (1-r)(1-s)(1-\varepsilon) + r(1-r)^2(1-s)^2(1-\varepsilon)^2 + ...) + A (s(1-r) + s(1-r)^2(1-s)(1-\varepsilon) + s(1-r)^3(1-s)^2(1-\varepsilon)^2 + ...)$$

- $R = Ar\Sigma(1-r)^{n}(1-\epsilon)^{n} + As(1-r)\Sigma(1-r)^{n}(1-\epsilon)^{n}(1-\epsilon)^{n}$
- $R = A(r+s(1-r))\Sigma(1-r)^{n}(1-s)^{n}(1-\varepsilon)^{n}$
- $R = A(r+s-sr) \Sigma (1-r)^{n} (1-s)^{n} (1-\varepsilon)^{n}$
- $R = A(r+s-sr)/(r+s+\epsilon-rs-r\epsilon-s\epsilon+rs\epsilon)$
- $R/A = (r+s-sr)/(r+s+\epsilon-rs-r\epsilon-s\epsilon+rs\epsilon) = 1/Q$

Maximum permitted loss, r, of e.g. Np

- s = 0.0001 (from MOX-fuel experience)
- $\epsilon = 0.05$  (in FBR estimated from SUPERFACT expts.)
- Q = A/R = 2.5 (requirement for Np from earlier diagram)

From these data and the earlier equations we get:

$$r = (Qs-s-\epsilon+s\epsilon)/(1-Q+sQ-s-\epsilon+s\epsilon) = 0.032$$

Hence, our assumptions and data gives a maximum permissible loss of 3.2% Np in one reprocessing cycle. This yields a reduction of the total Np-loss to waste by a factor of 2.5.

Requirements for Am, and Cm, are much harder.

### Continous on-line separation and recycle



## This scheme is much simpler to evaluate Mass balance equations, etc.

- A = Rate of addition of target atoms to reactor
- N = Number of target atoms in reactor
- S = Transfer rate to separation process
- $A = \varepsilon N + r S$ ; where  $\varepsilon$  is transmutation rate constant
- C = N / V; where V is irradiated volume
- v = k V = volume flow rate to separation; k=v/V
- S = v C; R = r S = loss rate to waste
- $\epsilon N = \phi_{avg} \sigma_{avg} N$ ;  $\epsilon = \phi_{avg} \sigma_{avg}$
- Q = A/R; needed reduction factor
- $r = \phi_{avg} \sigma_{avg} / [k (Q-1)]$

but  $\phi_{\text{avg}} \; \sigma_{\text{avg}}$  is of the magnitude 10-8 for fast neutrons, higher for thermal neutrons

## Problems with continuous on-line processing

- For most chemists, who have studied chemical thermodynamics, it is obvious that remixing of the cleaned flow with the bulk of fluid not yet cleaned involves an increase in entropy (loss of order).
- This is not the case when discrete units are removed, cleaned, and recycled. In that case we never lose the reduction in entropy from the separation.

This is also the phenomenon that hinders economic recovery of gold or uranium from sea water by a stationary plant.