Nuclear Energy, the Energy Balance

Chapter 4

Radioactive wastes; conditioning and disposal

second revision

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Introduction

Should nuclear energy fulfil its claim of being a clean technology, all radioactive wastes have to be packed in appropriate containers and disposed of in a safe manner. This is not common practice, today.

Radioactive wastes are classified in different ways. Important factors are:

- specific activity, Bq/kg;
- half-life of the radionuclides in the waste;
- type of radiation;
- presence of alpha-emitting nuclides, usually wastes containing actinides such as U, Np, Pu and Am.
- biochemical properties of the radionuclides

Unfortunately, the global nuclear industry still has not been able to reach consensus about a waste classification with unambiguous standards. Probably the economic consequences of such a classification are the cause of this indecisiveness.

In an often used, but vaguely defined, classification there are three classes of radioactive wastes [IAEA-377, 1995]:

LLW Low level waste

ILW Intermediate level waste

HLW High level waste.

Particularly the classification of alpha-holding wastes is unclear. Should wastes containing uranium oxide U_3O_8 be classified as ILW or HLW? Sometimes this kind of waste seems to be considered LLW, based on low radiation doses at the outside of the waste container, but possibly not based on the biological hazard of uranium and actinides when accidentally discharged into the environment.

Not only are the standards uncertain, but there also circulate in the nuclear industry many concepts for packaging radioactive wastes. In order to make any estimation possible of the energy expenditure of the waste handling and disposal, some choices are made in this study. Critics of these choices should look first to the nuclear industry for failing as yet to define unambiguous international standards for waste conditioning and disposal of all types of radioactive waste.

In this study alpha waste is classified as ILW if containing solely U at low concentrations, or HLW if containing U and actinides such as Np, Pu and Am.

Waste containers

Except for the mill tailings (see further on in this chapter), all radioactive wastes are to be packed into containers. In this study five types of standard containers, V1 through V5, are used, depending on the type of waste. These container concepts belong to the frequently quoted types in the nuclear literature, e.g. [IAEA-355 1993].

Representative standard containers for packaging radioactive wastes from the nuclear process chain:

Table 14

container	waste type	capacity (m^3)	displaced volume (m^3)	mass loaded (Mg)	remarks
V1	LLW	0.20	0.20	$0.03 + m_{waste}$	not for final disposal
V2	LLW/ILWα	0.20	1.00	$1.80 + m_{waste}$	

V3	HLW + α	0.29	1.18	6.5 + m_{waste}	German Type II
V4	LLW + ILW	1.73	4.10	5.8 + m_{waste}	not for alpha wastes
V5	spent fuel	–	2.40	25	2Mg HM

Producing, filling, handling and transport of V2, V3 and V4 containers is assumed to take as much energy as construction of the power station (see chapter 3 for the derivation of this value): 157 GJ/Mg, with R = 4.8.

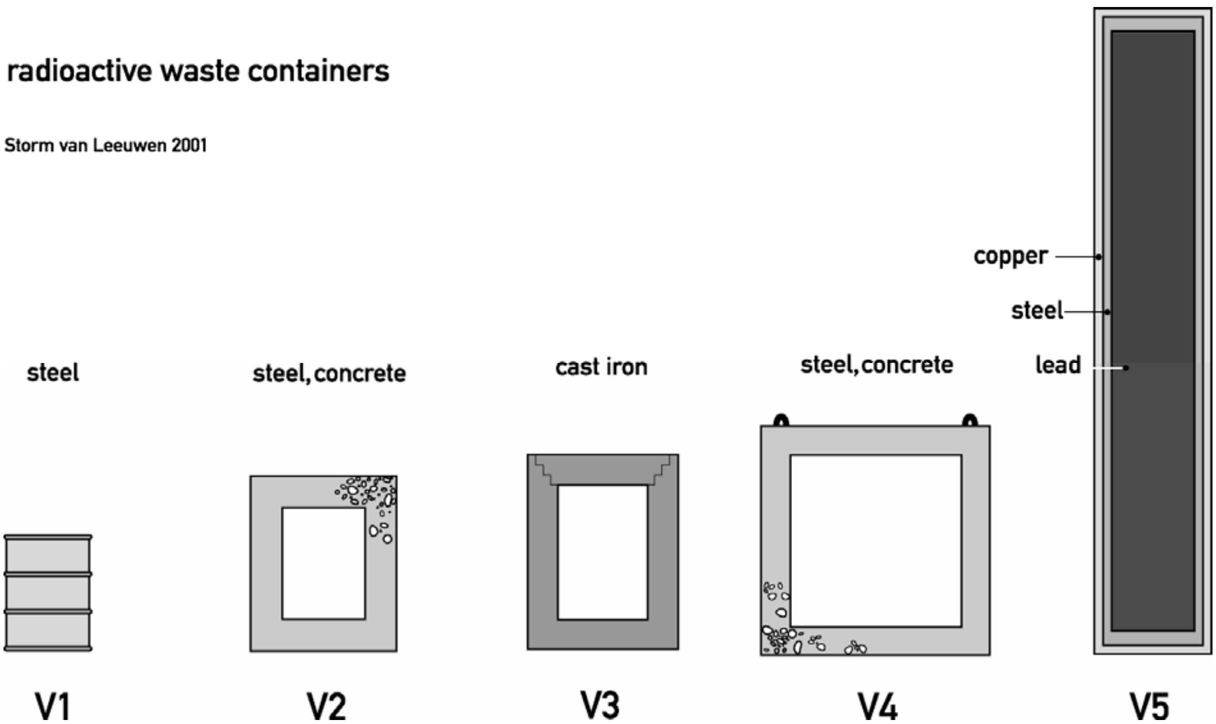

Figure 16. The various canisters, etc., used for storing radioactive materials are shown in this figure. They are described in Table 14.

Mill tailings

Up to the present day, mill tailings of the uranium mining and milling industry are simply discharged into the environment. In a clean nuclear cycle the mill tailings would have to be safely sequestered from the biosphere. Apart from the remaining uranium in the mill tailings (extraction yield = 0.98 maximal for the richest ores, dropping to less than 0.70 for lean ores, and even to 0.2 for the hypothetical exploitation of oil shales), the mill tailings contain the uranium decay daughters: ^{230}Th, ^{226}Ra, ^{220}Rn, ^{210}Pb and ^{231}Pa). These nuclides pose a serious long term environmental risk [NRC , 1996], [Andriesse, 1994], all the more so because the elements are chemically mobile after the milling process. The amount of mill tailings are calculated from Eq. 11, which follows from Eq. 2.1 (Chapter 2):

Amount of mill tailings:

$$m_{tail} = 1.18 \bullet m_U \bullet \left(\frac{100}{Y \bullet G} - 1 \right), \qquad \text{(Eq. 11)}$$

with $Y = 0.98 - 0.0723 \bullet (\log G)^2$ (Eq. 2.2)

m_{tail}	= mass of tailings	Mg
m_U	= mass of produced uranium	Mg
Y	= yield	fraction extracted uranium
G	= ore grade	mass-% U_3O_8 in ore body

The factor 1.18 comes from converting Mg U to Mg U_3O8: 1.18 Mg U_3O_8 contains 1 Mg U.

Restoring the uranium mine to green field conditions

The mill tailings - a sandy material saturated with several chemicals (e.g. sulphuric acid) – must be isolated from the biosphere. This process consists of several steps:

- neutralizing the acids in the tailings, in this study with limestone
- immobilization of the tailings
- transport of the immobilized tailings into the mine
- replacing the overburden
- replanting the area with indigenous vegetation.

Estimation of the energy expenditure of these activities is difficult, because despite its obvious necessity if one is to ever speak of a sustainable process, it has never yet been done. In this study we assume neutralizing and immobilization is done in two steps:

- the tailings are mixed with powdered limestone to neutralize the acids and a phosphate (e.g. sodium phosphate) to render the radioactive nuclides insoluble in water.
- the resulting mass is poured between thick layers of bentonite, in order to isolate the tailings from the ground water.

It should be noted that this procedure is a model to estimate the energy expenditure, not a real process.

The energy expenditure of above process is estimated to be four times that of mining. The mass of the tailings, including the limestone and bentonite, is about twice the mined mass. The limestone and bentonite have to be mined as well, the sodium phosphate has to be produced. The overburden has to be replaced and replanted.

Specific energy expenditure of mining (60% open pit, 40% underground, [Rotty et.al., 1975]) is:

J_{mining} = 1.06 GJ/Mg(ore), with R = 8.0

Based on the assumptions above, reclaiming the mining area will take:

$J_{tailings}$ = 4.2 GJ/Mg tailings, with R = 8.0

Operational wastes

Conditioning in preparation for final disposal

During normal operation, the following quantities of radioactive waste are produced, including the decommissioning and dismantling wastes of the facilities of each process [IAEA-293 1988], [Orita 1995], [IAEA-377 1995].

Table 15

process	quantity		waste type	container	number of containers	
conversion	54	m^3/GWe.a	LLW	V2	270	/GWe.a
fuel fabrication	75	m^3/GWe.a	LLW	V2	375	/GWe.a
reactor operation	50	m^3/GWe.a	ILW	V2	250	/GWe.a
200	m^3/GWe.a	LLW	V2	1000	/GWe.a	
diverse operational wastes:			1895	containers V2/GWe.a		
plus either:						
diffusion enrichment, or	59	m^3/Mg SWU	LLW	V2	295	/Mg SWU
ultracentrifuge enrichment	230	m^3/Mg SWU	LLW	V2	1150	/Mg SWU

Operational wastes usually contains alpha emitters, so this type of waste is packed in V2 containers. The unit of one gigawatt-year(electric) for the first four items of Table 15, corresponds to 1 full-power year of the reactor. The energy expenditure of waste packaging and interim storage before final disposal, is included in the specific energy expenditure of each process.

From the literature (above) we find that the conditioning (packaging) of 1895 V2-containers requires an annual energy expenditure of 0.536 PJ with the ratio of thermal to electrical energy = 4.8.

Final disposal of operational wastes

We have assumed that all radioactive wastes except the much more dangerous spent fuel elements, which are treated separately below, are disposed of in a geological repository similar to the Swedish SFR concept [IAEA-349, 1993]. The wastes are stored in large caverns, mined in a stable rock formation. In this concept, 13.8 Mg rock has to be removed for each m^3 of disposed waste. Since the repository is back-filled with bentonite, which has to be mined, prepared and transported as well, the total specific energy expenditure of disposal is taken three times the energy expenditure of mining.

J_{disp} = 45.5[1] GJ/m^3 waste, with R = 5.

Depleted uranium

The main waste product of the enrichment process is depleted uranium, as uranium hexafluoride UF_6. Currently, the depleted uranium is stored as UF_6 in metal containers. In the USA about 500,000 Mg depleted UF_6 is stored above ground, most of it out-of-doors [NRC, 1996]. Large quantities have been converted into the metal, for use as ballast in airplanes and in armor-penetrating munition. When the depleted uranium is used in munition, the metal is effectively discharged into the environment - a highly undesirable situation, as recent history has amply proven.

Needless to say that in a sustainable scenario, if such is indeed possible for nuclear energy, the depleted uranium, as all well as all other radioactive wastes, has to be removed from the biosphere. Since the compound UF_6 is volatile (it sublimes at 56.5 degrees C) and is extremely reactive, it has to be reconverted to triuranium octaoxide U_3O_8, before it can be packed in containers for final disposal in a geological repository.

Assumed reconversion consumes as much energy as conversion, the specific energy expenditure is:

J_{reconv} = 1.48 GJ/kgU, with R = 27

The amount of depleted uranium is calculated from Eq. 12, which can be derived from Eq.4.1, Chapter 2.

$$W = F - P = \left(\frac{x_p - x_f}{x_f - x_t} \right) \qquad \text{(Eq. 12)}$$

W	= waste mass	kg U
F	= feed mass	kg U
P	= product mass	kg U
x_f	= feed assay = 0.0071	fraction ^{235}U
x_p	= product assay	fraction ^{235}U
x_t	= tails assay = 0.0020	fraction ^{235}U

We assume that the depleted tri-uranium octa-oxide is packed in V2 containers, with fill factor 1. One Mg of depleted uranium corresponds to 1.18 Mg tri-uranium octa-oxide. As the density is 8.30 Mg/m^3, 1 Mg U corresponds to 0.142 m^3 , or 0.711 V2 containers per Mg U(depleted). This comes to 0.71 m^3/Mg U (the volume of a V2 container is 1 m^3). For the conditioning of operational waste (above), somewhat analogous to depleted uranium, we derived a value of 0.534 PJ for the annual requirement of

1895 containers. This comes to 282 x 10^{-6} PJ per container. Multiplying this cost per container by the figure of 0.71 containers per MgU we find the cost for conditioning the depleted U_3O_8 to be 201 x 10^{-6} PJ/MgU, with a ratio of thermal to electric energy of 4.8.

The disposal of this waste entails the same processes as for operational waste. This was found to cost 45.5 GJ/m^3 in the section on operational waste above. Multiplying this cost by the same figure as for the conditioning (.71 m^3/MgU) we find an energy cost for the final disposal of depleted uranium to be 32 PJ/Mg U.

Adding up these energy costs:

Reconversion	1430 GJ(th)/MgU	+ 53 GJ(e)/MgU
Conditioning	166 GJ(th)/MgU	+ 35 GJ(e)/MgU
Disposal	28 GJ(th)/MgU	+ 4 GJ(e)/MgU

Giving a total (rounded) energy expenditure of:

1700 GJ/MgU, with R = 18

Spent fuel elements

Interim storage

After removal from the reactor, the spent fuel elements are stored for 30-60 years in a heavily shielded building, where the elements are cooled by air or water [Konings & Dodd 1999], [Nachrichten, 1997].

Specific energy expenditure includes:

- operational energy expenditure (maintenance, surveillance during 30-100 years)
- transport
- construction of the facility
- dismantling

These costs depend on the capacity of the storage facility. We found the range of costs to be

J_{int} = from 1.86 to 16.5 GJ/kgHM

The lower value applies for a very large facility, the higher value for a relative small one. The specific energy expenditure is calculated from cost data (151 - 1340 $(2000)/kgHM), with the energy intensity rule (12.34 MJ/$(2000)).

In this study the average value, J_{int} = 9.5 GJ/kgHM, is used, with R = 11. It should be noted that the figures in the literature are based on estimates, not on operational data. Real costs and energy expenditures will always be higher, not lower.

Conditioning for final disposal

In a once-through cycle, the spent fuel elements are packed in special canisters of type V5, after a cooling period of 30-60 years in interim storage. Canister V5 stems from the Swedish SKB-3 concept [Papp ,1998a], [IAEA-349, 1993]. Packing the highly radioactive fuel elements and sealing off the canisters has to be done under remote control. The canisters are fabricated from high quality materials, because they have to last for thousands of years, e.g. only exceedingly pure electrolytic copper is used. It is reasonable to assume the energy intensity of this process is as high as the construction of the reactor, 157 GJ/Mg (see under construction in Chapter 3). Based on cost estimates [Konings & Dodd 1999] and the energy intensity rule, a value of about 120 GJ/Mg is found. The higher figure is used in this study, because it seems te be more reliable. Costs estimates of new technologies always are underestimated and spent fuel packaging is a complety new technology. No operational data are available at the time of writing (spring 2001).

A V5 canister has a mass of about 25 Mg and contains about 2 Mg HM in spent PWR fuel. So the energy expenditure of this process can be estimated at: J_{cond}= 2.0 GJ/kgHM with r = 4.8.

The Swedish SKB-3 concept is the most elaborate and advanced concept for final disposal of spent fuel

[Papp, 1998a], [Papp, 1998b], [IAEA-349 1993]. A Swiss concept is quite similar. In this study the SKB-3 concept serves as a model for final disposal of spent fuel and all other radioactive wastes, except the mill tailings, which we treated above.

Final disposal in a stable geological repository

In the SKB-3 concept galleries are mined in granite or other very stable rock strata. The spent fuel canisters are placed in boreholes in the floor of the galleries by remotely piloted vehicles. The holes are filled up with bentonite, as is the gallery itself after filling the holes. The total energy expenditure of this process can be estimated on the basis of the projected cost, using the same money to energy conversion factor as was used in Chapter 3 to estimate the energy expenditure of the construction of the power plant. The result is:

J_{disp} = 8.7 GJ/kg HM.

We have not used this value, but have calculated the estimated energy expenditure on the basis of standard mining practice, as follows:

To store 7000 MgHM, about 5.8 x 10^6 m^3 granite has to mined, or about 16 x 10^6 Mg [Papp, 1998a], or 2.3 Mg rock/kgHM.

Underground mining consumes about J_{mining} = 4.0 GJ/Mg rock with R = 5 (see above). This results in:

J_{disp} = 9.2[2] GJ/kgHM.

However, the mine must be backfilled with bentonite, and the bentonite itself has to be mined, prepared and transported as well. All operations after mining the galleries are remotely piloted, including the backfilling. We estimate the total energy expenditure to be about four times higher than for mining alone:

J_{disp} = 10.0[3] GJ/kgHM

Decommissioning and dismantling wastes of the power plant itself

Present practice indicates that about 4-10% of the construction materials of a nuclear power plant has to be considered radioactive waste, with the lower value applying for a reactor with a very short active lifetime. Undoubtedly, this percentage depends on the total neutron flux and therefore on the full-power time of the reactor, as more materials will be more heavily activated and contaminated, the longer the operation time. Approximating an average value, a constant 7.2% of the construction materials is assumed (37000/516000 = 0.072), divided as follows:

Table 16

material	quantity		class	container type	fill fraction	number of containers
cleanup wastes	1000	m^3	ILW	V2	0.5	10000
reactor + ass. systems	400	Mg	HLW	V3	0.2	800
steel	400	Mg	ILW	V3	0.2	800
steel	4000	Mg	LLW	V4	0.2	1500
concrete	16000	m^3	LLW	V4	0.5	18500

The energy costs of the packaging of this material are lumped together with the other dismantling costs discussed in detail in Chapter 3.

The total volume of dismantling wastes to be disposed off in a geological repository is summarized in

Table 17.

Table 17

container	number	volume, m^3
V2	10000	10000
V3	1600	1900
V4	20000	82000
total	31600	93900

Applying the figure of 45.5 GJ(total)/m^3 used above for operational wastes, we find that 4.3 PJ will be necessary for this disposal. Because of the large uncertainties in all of the dismantling costs this item has also been lumped together in Chapter 3 with the rest of the dismantling costs and not considered as a separate cost.

Enrichment wastes

As stated above, the main waste enrichment is the depleted uranium. There are other wastes, however, and these should also be properly disposed of. These are given, for diffusion enrichment, as 295 V2 containers per Mg SWU.

Using the figure derived above for the conditioning of low-level radioactive waste in V2 containers of 282 x 10^{-6} PJ/container, we find a total energy cost for the 295 containers of 73.3 x 10^{-3} PJ/Mg SWU. To calculate this we must know the mass and enrichment of the uranium used in the reactor. The equation to calculate this is found in Chapter 2 (Eq. 4.1).

The number of SWU per kg product mass (i.e. the mass of enriched uranium) is given by S, which is a function of the fraction ^{235}U in the feed, the product and the tail. (Eq. 4.2).

The energy expenditure for conditioning and disposal is then:

Conditioning:	6.5 x 10^{-3} PJ(e)/Mg SWU + 66.8 x 10^{-3} PJ(th)/Mg SWU;
Disposal	1.3 x 10^{-3} PJ(e)/Mg SWU + 10.5 x 10^{-3} PJ(th)/Mg SWU;
The total is thus:	7.8 x 10^{-3} PJ(e)/Mg SWU + 77.3 x 10^{-3} PJ(th)/Mg SWU.