Vitrification and dilution of radioactive waste

Jan Willem Storm van Leeuwen
Independent consultant

member of the Nuclear Consulting Group

November 2019
storm@ceedata.nl

Note

In this document the references are coded by Q-numbers (e.g. Q6). Each reference has a unique number in this coding system, which is consistently used throughout all publications by the author. In the list at the back of the document the references are sorted by Q-number. The resulting sequence is not necessarily the same order in which the references appear in the text.

Contents

FIGURES

Nuclear waste

Definition

An obvious answer may may be: 'nuclear' means 'related to nuclear power', and 'waste' means 'materials not usable for activities of the nuclear system, or not usable for any industrial process. It turns out to be not as simple as it may seem. The question has two components: what is meant by 'nuclear', and what is meant by 'waste'? Generally 'nuclear' is associated with radioactive materials, however, 'nuclear' is not always the same as 'radioactive', as will explained below.

The system of industrial processes needed to convert the potential energy in uranium into useful energy generates waste streams, as any industrial system. Commonly, some waste streams are classified as non-radioactive, for example carbon dioxide CO_2, other waste streams are called 'radioactive'. Caveat. Virtually all materials found in nature contain naturally occurring radionuclides and therefore should strictly be classified as 'radioactive'. The natural specific radioactivity of the human body is 143 Bq/kg body weight, mainly caused by the naturally occurring radionuclides potassium-40 and carbon-14 [Charpak & Garwin 2002] Q300. People living in areas with relatively high concentrations of uranium en thorium in the ground likely will have a higher specific radioactivity than the global average. The same holds true for people living in areas contaminated by human-made radionuclides as a consequence of nuclear accidents, for example Chernobyl and Fukushima.
To distinguish between 'radioactive' and 'non-radioactive' a criterion must be applied, for example: natural materials are 'non-radioactive', if they do not contain radionuclides at concentrations higher than the human body.

Publications of the International Atomic Energy Agency (IAEA) and the nuclear industry point to different views on the notions 'nuclear' and 'waste'.
At one hand some waste streams of the nuclear process chain containing radioactive materials are classified as 'non-nuclear', on the other hand radioactive materials generated by the nuclear process chain are often not classified as 'waste', but as a 'potential energy source', see section below.

Spent nuclear fuel: waste or resource?

[IAEA-wmdb-st-1 2001] Q656 p 31-34
Some IAEA Member States have policy of direct disposal, once through fuel cycle model,
other have a policy to reprocess their spent nuclear fuel: resource, closed fuel cycle.
Spent fuel typically: 94.3% U, 1,15% Pu and 4.55% waste products (other actinides, fission products and unwanted impurities).
Waste products HLW; vitrification is a well established opertion that has been rigorously examined and approved by regulatory authorities in several countries. Removal of U and Pu reduces the volume of HLW, but leads to the production of LILW.
Orininally reprocessing was the only considered management option for spent fuel. Later on direct disposal was recognised as an attractive alternative for various reasons:

- non-proliferation aspects
- limited market for MOX fuel
- cancellation of fast breeder programmes
- expected cost benefit
- technical and economical difficulties with reprocessing smaller quantities of some fuel types
- public concern over reprocessing facilities.

View of the IAEA and the nuclear industry

The nuclear process chain starts with the recovery of uranium from uranium ore. This process involves materials with high concentrations of uranium and its decay products. The mining and milling wastes, called mill tailings, are to be classified as radioactive, see report m41 *Uranium mine rehabilitation*.
Notable is the following statement of the nuclear industry [WNA-04 2011] Q271:

'Strictly speaking these (*mining and milling wastes*) are not classified as radioactive wastes'.

Is this view based on the fact that the uranium mining wastes contain exclusively natural occurring radionuclides?

Criteria determining the health hazards of a radioactive material are not only the specific activity of that material, measured in Bq/kg, but also the isotopic composition of that material.
Are short-lived radionuclides less hazardous than long-lived radionuclides?
Are the biological properties of radionuclides inside living organisms taken into account?
Are all pathways of contamination by radioactive materials taken into account? How about chronic exposure to radionuclides via air, drinking water and food?
Are synergetic effects of contamination by a number of radionuclides simultaneously taken into account?
The radiological models applied by the nuclear industry have a limited scope, empirical evidence from recent decades is not included, see report m11 *Health effects of radioactivity*.

Publications of the IAEA and the nuclear industry are not clear about their definition of 'nuclear waste'. Obviously this notion is not unambiguously defined. Within the nuclear industry numerous systems of waste classifications are used, different countries may apply different systems. The nuclear industry often distinguishes between materials containing long-lived radionuclides and materials containing only short-lived radionuclides. Probably this distinction is based on economic arguments.
Within the framework of its Joint Convention project the IAEA published a series of reports *Radioactive Waste Management Data Base - Status and Trends*, for instance [IAEA-*wmdb-st-4* 2005] Q659, discussing envisioned international agreements on waste management. In these reports the IAEA describes numerous regulations and waste classifications.
These WMDB reports do not mention contributions other than from the USA, Europe and Japan. It remains unclear wether the non-contributing countries would comply with the regulations proposed by the IAEA. For more details see report m31 *Industrial views on radioactive waste*.
Many descriptions of the various waste classes are vague and some descriptions are susceptible to economic considerations. Quantified standards are missing in the descriptions of the waste categories. Some examples:
How is the classification 'Below Regulatory Concern' defined?
Descriptions of notions as: 'Exclusion', 'Exemption', 'Clearance' [IAEA-wmdb-st-1 2001] Q656 are unclear, even for an independent physicist. No numerical standards are mentioned. The descriptions suggest that these notions may be interpreted according to local financial considerations.

The World Nuclear Association (WNA), which may be seen as a representative of the nuclear industry, states in its publication *Radioactive Waste Management* [WNA 2016a] Q540:

- Nuclear power is the only large-scale energy-producing technology which takes full responsibility for all its wastes and fully costs this into the product.
- The amount of radioactive wastes is very small relative to wastes produced by fossil fuel electricity generation.
- Used nuclear fuel may be treated as a resource or simply as a waste.
- Nuclear wastes are neither particularly hazardous nor hard to manage relative to other toxic industrial wastes.
- Safe methods for the final disposal of high-level radioactive waste are technically proven; the international consensus is that this should be geological disposal.

These statements are questionable in view of the present practice. After nearly seven decades of civil nuclear power all hazardous nuclear waste is still stored at vulnerable temporary storage facilities within the human environment. For comments on the above statements see report m31 *Industrial views on radioactive waste*.

A major drawback in the proposed regulations and waste classifications by the IAEA and WNA might be the reliability of the inspections and measurements needed to assign the correct classification to each unit of nuclear waste. Questions arise, such as:

- Would it be technically possible to know exactly the content of radionuclides in each container of radioactive waste?
- Would it be possible to inspect and classify all radioactive wastes?
- How reliable would the inspections be? Which guarantees could be given for independent inspections, not compromised by financial and/or political interests?
- Would it be possible to preserve the documentation of the classified wastes over decades of time?
- Could be guaranteed that a skilled workforce remains available during the coming decades?

Dilution of radioactive waste

View of the IAEA

According to [IAEA-wmdb-st-1 2001] Q656 there are two basic strategies for radioactive waste disposal:

- 'isolate and confine'
- 'dilute and disperse'.

The first strategy involves the emplacement of waste into a disposal facility that is intended to isolate the waste from humans and the environment and to prevent or limit releases of potentially harmful substances (toxic metals, radionuclides, organics) such that human health and the environment are protected.
The second strategy involves deliberately dispersing the waste into the environment in a manner intended to dilute harmful contaminants in the waste to levels that are considered 'acceptable' according to internationally agreed standards.
The IAEA does not make clear how 'internationally agreed standards' define the classification 'acceptable'.

Theoretically it may seem viable: diluting radioactive matter with an amount of non-radioactive matter will dilute the activity to a low concentration and will make radioactive waste 'harmless'. In practice this option is less simple, in view of the actual masses and specific activity of the radioactive wastes generated by nuclear power.

Lifetime radioactive waste generation of one NPP

Lifetime radioactive waste generation of one advanced reference nuclear power plant (NPP):

uranium mill tailings	7730 Gg chemically toxic and radioactive waste
front-end processes	11 Gg
OMR operation	188 Gg
spent fuel	584 Mg
decommissioning and dismantling	100 Gg (= $1*10^8$ kg)
discharges into the environment	unknown, must be significant
radioactive cooling water	unknown, likely tens of Gg
contaminated soil	unknown, likely tens of Gg

See also reports:

m04 *Decommissioning and dismantling*
m11 *Health effects of radioactivity*
m12 *Human-made radioactivity*
m19 *Advanced reference reactor and EPR*
m26 *Uranium mining + milling*
m32 *Geologic repositories and waste conditioning*
m36 *Materials for nuclear power*
m40 *Radioactive waste management - future CO2 emissions*.
m41 *Uranium mine rehabilitation*

Numerical example of waste dilution

During its operational lifetime the reference advanced reactor generates an amount of human-made radioactivity in the order of magnitude of 10 EBq (= $1*10^{19}$ Bq), embedded in the spent fuel. Assumed

that about 1% of the human-made radioactivity would be embedded in the waste materials (100 Mg) resulting from decommissioning and dismantling of the power plant, then the average specific activity of those materials would be 1 GBq/kg (= $1*10^9$ Bq/kg). Dilution of 100 Gg radioactive materials to the level of the human body (143 Bq/kg) would require billions of tons of sand or other non-radioactive material. The resulting material could be released into the public domain without restrictions

Obviously this is not practically viable. In practice the standard of 'harmless', or 'acceptable' specific activity would be defined at a much higher level than the human body, resulting in a specific activity of the diluted waste millions of times higher. The IAEA and nuclear industry have not specified notions as 'acceptable' and 'below regulatory concern', see Introduction and report m31 *Industrial views on radioactive waste*. In practice te strategy of dilution might lead to uncontrolled releases of massive amounts of dangerous radioactive materials into the human environment.

Global legacy of radioactive waste

By 2019 all radioactive materials generated during seven decades of civil nuclear power are still awaiting final disposal in a permanently safe repository, and are stored in temporary storage facilities. These materials include the radioactive wastes from about 650 nuclear power stations and more than 30 reprocessing plants. In 2019 about 350 Gg of spent nuclear fuel were stored in temporary facilities, according to figures from [IPFM 2011] Q513.
The amounts of radioactive waste from reprocessing plants, including decommissioning and dismantling wastes, can only be guessed, but are likely to be counted in milions of tons. Debris and scrap from reprocessing plants are contaminated with all kinds of radionuclides from spent fuel, fission products and actinides. In addition large volumes of soil are contaminated wth radionuclides., also to be counted in millions of tons.

Health risks

Larger waste volume increase the chances humans will come into contact with that waste or that it will enter the food chain. This may raise ethical questions like: What would be more acceptable to individuals: a certain chance of ingesting 100 lethal doses or a million times greater chance of ingesting one lethal dose?

Practice

Military nuclear facilities did dilute nuclear wastes by soil in the past, simply by letting the liquid wastes leak into the ground and ground water, via 'storage' ponds or otherwise. Besides, large amounts of nuclear wastes, including complete reactors, have been dumped into the sea: diluting by sea water.

Diluting radioactive wastes by air and (sea) water is common practice in the civil nuclear industry. Large amounts of radioactive materials routinely are discharged by nuclear power stations, for example cooling water containing tritium, carbon-14 and other man-made radionuclides..
Reprocessing plants discharge even larger amounts of radionuclides, including fission products and actinides, than nuclear power plants. This practice is an essential part of their 'waste reduction' policy. It is not by chance that the reprocessing plants of France (La Hague) and the UK (Sellafield) are situated at the sea shore.

Vitrification of high-level radioactive waste

Concept

According to [WNA 2012b]Q541 a typical 1 GWe reactor produces each year about 700 kg high-level wastes, contained in about 23 Mg (metric tons) of spent fuel. With 'high-level wastes' WNA likely refers to the fission products plus actinides in spent fuel, but does not mention them explicitely. After separation from spent fuel in a reprocessing plant the liquid high-level wastes are evaporated to solids, mixed with glass-forming materials, melted and poured into stainless steel canisters which are then sealed by welding. The vitrified waste from the operation of a 1 GWe reactor for one year would fill about twelve canisters, each 1.3 m high and 0.4 m diameter and holding 400 kg of glass, according to WNA.
The canisters are to be placed in a geological repository for permanent disposal. In another process, called Synroc, the wastes are calcined and mixed wich several metaloxides for conversion at high temperatures into a crystalline ceramic material Synroc (synthetic rock).

Applying the vitrification concept the mass holding the highly radioactive materials would be reduced from 23 Mg spent fuel to 4.8 Mg borosilicate glass, a reduction of a factor of less than five. Would this marginal mass reduction justify the astronomical cost of reprocessing and the introduction of additional health hazards?
As a means of volume reduction of high-level waste the vitrification concept turns out to be a fallacy: the radioactive waste volumes increase enormously by reprocessing, as will explained below.

In spent fuel fission products and actinides constitute 3.5 and 1.4 mass% respectively. The solidified waste may contain up to about 30 % fission products plus actinides IAEA-187 1979] Q268, so apparently a significant volume reduction might be achieved.
In this story the zircalloy cladding hulls (0.6 - 2 Mg per Mg fuel) seem to be ignored, which also are highly radioactive with long-lived radionuclides.
Severe problems arise with the borosilicate glass, e.g. radiolytic reactions, heat generation, (re)crystallization and segregation of elements.

Fallacies and misconceptions

The waste volume reduction by vitrifiation concept is implicitely based on a fallacy, namely the feasibility of 100% complete separation of all chemical elements constituting spent fuel, without losses. This will be explained in the following sections

The amount of radioactivity in spent fuel does not change by the mechanical and chemical treatments in the reprocessing plant, it simply means a redistribution of the radionuclides from one material flow to several other. Inevitably, mixing an amount of radionuclides, compacted in a solid (spent fuel), with nonradioactive fluids or other substances increases the volume of the radioactive waste, heavily complicating the waste disposal problems, see also figure 3.

In thermodynamic terms: the latent entropy of the dangerous radioactive mass in spent fuel increases enormously by dissolving the spent fuel in a liquid. The controllability of the system, the radioactive species from the spent fuel, decreases rapidly with increasing entropy. This implies that an input of high-quality energy is needed to keep the system at the same level of controllability. This energy input increases exponentially with the entropy increase.

Separation of the elements in a solid or solution never can be complete, partly due to the chemical

properties of the components of a mix, partly due to the inherent properties of chemical and physical extraction equilibria, partly due to technical imperfections. Economic considerations are left aside here, but will doubtlessly play a part. The difficulties increase with the number of compounds or elements in the mix which are to be separated.

The nuclear industry states the hazards of nuclear waste being easily controllable, because of the relatively small masses and volumes involved, see also report m31 *Industrial views on radioactive waste*.

As a result of the fission of uranium nuclei in the reactor the level of radioactivity of the nuclear fuel rises by a factor of 1 billion, due to the generation of artificial radionuclides. During the disasters of Chernobyl and Fukushima jointly an amount of radioactivity was been dispersed into the biosphere equivalent to the annual production of one nuclear power plant, 0.01% of the global nuclear legacy. This amount was contained in about 20 Mg (metric tons) of spent fuel.

The view of the nuclear industry suggests as if the hazards posed by the radioactive waste from nuclear power would be proportional to the volume or the mass of the vitrified waste.
Above assertion of the nuclear industry is seriously misleading for several reasons, such as:

- the hazards are determined by the amounts of radioactivity, the biochemical behaviour and the radiotoxical properties of the radionuclides
- only a part of the high-level waste can be immobilized in boroslicate glass or Synroc, because:
  - a significant part of the radionuclides in the waste is chemically incompatible with a glass or a ceramic matrix
  - a part of the radionuclides escapes into the environment during processing.
- the huge masses and volumes of intermediate-level and low-level waste are negected, see also the first section *Nuclear waste*.

The safest way to handle the high-level nuclear waste is to keep the spent fuel elements intact, to pack them in the most durable containers and to store the containers in a geologic repository.

Radioactive decay of spent nuclear fuel

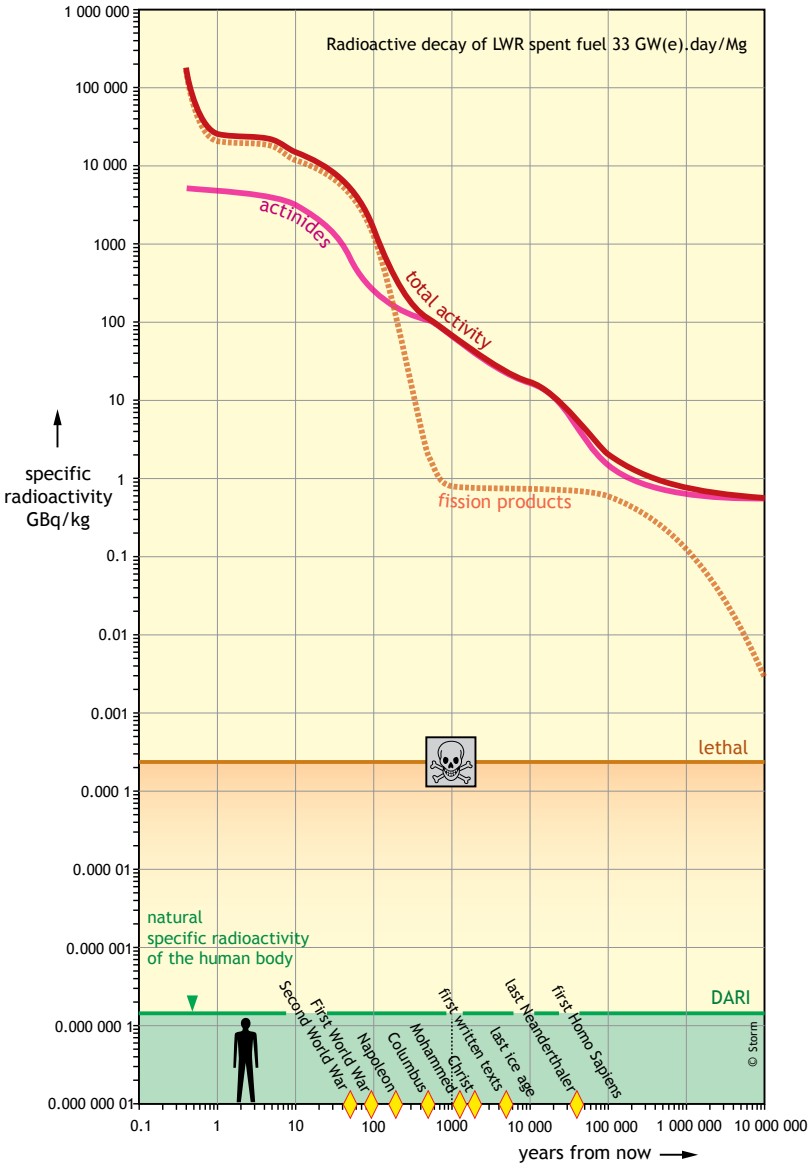

Figure 1

The specific radioactivity, in gigabecquerel per kilogram (GBq/kg), of spent fuel at a burnup of 33 GWe.day/Mg (gigawatt electric per metric tonne uranium) charged into the reactor. Nuclear fuel from current types of nuclear reactors usually has higher burnup (40-50 GWe.day/Mg) than the fuel this diagram is based on and consequently its specific radioactivity is higher. The contributions of tritium and carbon-14 are not included in these curves. Note that both axes have logarithmic scales. Each scale division denotes a factor ten. With linear time scales the horizontal axis would be about 100 kilometers long and the vertical axis some 100 million km.

On the horizontal axis a reverse historic timescale is indicated, to give an idea of the time frames involved. The green line indicates the natural radioactivity of the human body (143 Bq/kg). Sources: Bell 1973 [Q264], Hollocher 1975 [Q262], JPL-77-69 1977 [Q263], Charpak & Garwin 2002 [Q300].

The radioactivity of spent fuel at a given moment in an operating reactor is largely set by the fission products. During the fission process some thousand different nuclides are formed and a significant part of these are radioactive. During the first month after the fission process has been shut down, intentionally or by accident,

the radioactivity of spent fuel decreases sharply, due to the decay of very short-lived fission products. After the first months the radioactivity decreases slowly and is chiefly set by radionuclides with longer half-lifes. After some 300 years the radioactivity of spent fuel is chiefly set by the actinides, not to say that the very long-lived fission products are unimportant after the crossover.

From the viewpoint of health hazards it is important, among other parameters, to know which radionuclides are released from spent nuclear fuel, by whatever cause; Report m17 *Pathways of radioactivie contamination* addresses the pathways along which radioactive materials from spent fuel may enter the human environment. The radioisotopic composition of the released radioactive material depends on the time period between the end of the fission process and the moment of release.

The half-lifes of the radionuclides present in used nuclear fuel vary from milliseconds to millions of years. To gain some insight into in this matter the fission products are grouped according their half-lifes in Tables 1-6. Table 1 for example lists a selection of radionuclides which have almost completely decayed within 0.1 year (37 days) after shutdown of the fission process. After 10 half-lifes only one thousandth of the original amount of a radionuclide remains, the rest has decayed to the decay daughter of the radionuclide, mostly a stable nuclide. For example, iodine-133 decays to stable xenon-133 with a half-life of 20.8 hours. Within 37 days some 42 half-lifes have gone by and about $3*10^{-13}$ of the original amount is left.

The highly hazardous actinides generally have long to very long half-lifes (see Table 7 in report m12 *Human-made radioactivity*). The activation products, a minor but stil important contribution to the total radioactivity of spent fuel at the moment of discharge from the reactor, have half-lifes varying from days to thousands of years (see Table 8 in report m12 *Human-made radioactivity*).

The decay curves of Figure 1 show that after about 4 centuries the specific radioactivity of spent fuel is mainly set by the actinides. After about 1000 years the radioactivity of the fission products remains nearly constant at a level of slightly less than 1 GBq/kg for about 100000 years.This level is still more than a million times as high as the specific activity of the human body. Assume the 'short-lived' waste contains no actinides, accordingly to the advocated concept of the vitrification – which is not possible as explained –, which radionuclides are present in that waste?
In its communication with the public the nuclear industry seems to suggest that this level of specific activity is not harmful to humans anymore.

During the 1000 years before the specific activity of the waste reaches that 'harmless' level, the waste containers are not storaged in a deep geologic repository, according to the nuclear industry, but in above-ground facilities, which are less expensive. One can be sure that the waste containers will deteriorate by natural degrading processes, not counting human actions, to such an extent that much, if not all, radioactivity will end up in the environment.

Reprocessing of spent nuclear fuel

Mass composition of spent fuel

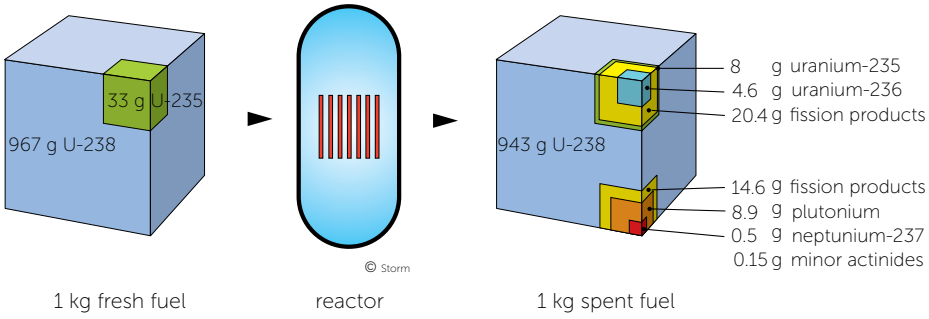

Figure 2.
Mass composition of fresh and spent nuclear fuel after three years in the reactor, corresponding with the current world average burnup. The amounts of the components other than U-238 in fresh and spent fuel of the newest types of LWRs are some 40% larger.
In this diagram the various components of fresh and spent fuel are shown separately. but actually the atoms of the present isotopes and elements are dispersed on molecular level.Complete separation is not possible. All new components, represented by the small cubes, are strongly radioactive. The total mass remains nearly constant, a minute fraction is converted into energy during fission. The small cubes at top right represent the mass of the nuclides originating from U-235, the cubes on the lower right corner are formed from U-238.
In addition to the fuel, about 2 - 0.6 kg zircalloy cladding + spacers and about 25 g silver-indium-cadmium alloy control rods are loaded and discharged with each kilogram of fuel.

In spent fuel fission products and actinides constitute roughly 3.5 and 1 mass-% respectively, see Figure 2. The solidified waste would ideally contain up to about 30 % fission products plus actinides. A significant volume reduction of the waste seems possible if the weakly radioactive uranium could be separated from the other, high-level radioactive components of spent fuel. However, this concept is flawed by serious misconceptions and fallacies.

Reprocessing of spent fuel

In the reprocessing plant the spent fuel is chemically treated to separate it into several fractions: unfissioned uranium, newly formed plutonium, fission products and actinides. The radioactivity, in the spent fuel present in a very condensed form, is during the reprocessing spread over large volumes of liquids and solids. All gaseous fission products, such as tritium, carbon-14, the radioactive noble gases and a significant part of iodine-129 are released into the air. Complete separation of a mix of different species into its pure consituents is a delusion, as a consequence of the Second Law (see report m38 *Nuclear power and the Second Law*). Separation processes never go to completion, the more so the greater the number of constituents and the higher the radioactivity of the mix. This follows from the Second Law. Consequently a significant fraction of the radionuclides is discharged from the plant with the waste water into the sea. Not by chance the European reprocessing plants (La Hague in France and Sellafield in the UK) are located at the sea coast.

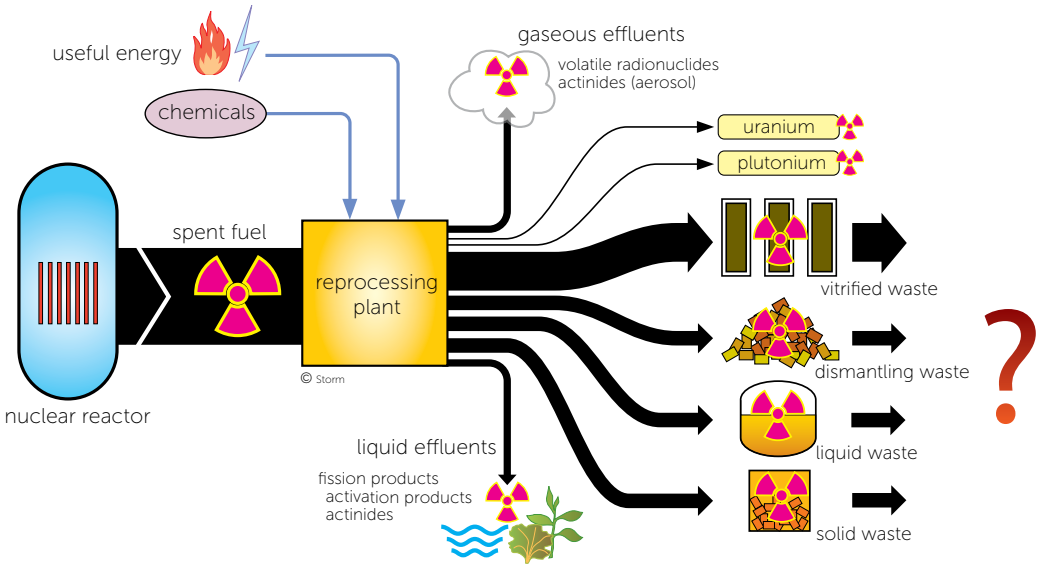

Figure 3

Outline of the radioactive waste streams from reprocessing of spent fuel. In order to recover uranium and plutonium from spent fuel, the radioactivity from the spent fuel is partitioned into a number waste streams. Significant amounts of radionuclides are discharges into the environment. Only a part of the radionuclides from the spent fuel can be vitrified. How to safely isolate the radioactive waste streams from the biosphere is still an open question.

Practice

Issues

By reprocessing the radioactive contents of the spent fuel are distributed among large volumes of different materials, only a part of it ends up in the vitrified waste. This greatly enhances the chances of dispersal and of severe accidents involving massive amounts of radioactivity. A significant part of the radioactive contents is released into the environment, as pointed out in reports m17 *Pathways of radioactive contamination* and m20 *Reprocessing of spent nuclear fue*l.

In its communication with the public on the waste vitrification concept the nuclear industry conveniently overlooks some practical aspects of this technique, such as:

- All gaseous fission products and a substantial fraction of other fission products and of the actinides are inevitably discharged into the environment by the reprocessing plant.
- Generation of large volumes of other radioactive wastes during reprocessing because substantial fractions of the radionuclides from spent fuel are spread among non-radioactive solids and liquids and do not end up in the glass. Actually the radioactive waste volume increases to a great multiple of the volume of spent fuel.
- Immense amounts of radioactive waste will result from decommissioning and dismantling of nuclear power plants and reprocessing plants.
- Some nuclides are discharged during solidifying the liquid waste stream (calcination) and subsequent vitrification of the solid residu. Not all nuclides (e.g. Se-79, Ru-106, I-129) can be effectively fixed into a glass, since they hardly form stable compounds with the borosilicate matrix, or become volatile during the calcination process.

The nuclear industry does not discuss the large volumes of the remaining radioactive waste which cannot be vitrified, nor about the releases of large quantities of radionuclides into the biosphere. Nothing is said about the massive volumes of radioactive wastes resulting from the decommissioning and dismantling of the reprocessing plant at the end of its operational lifetime: immense amounts of debris and scrap (order of magnitude: a million of tonnes) are heavily contaminated with all kinds of radionuclides from the processed nuclear fuel.

The assumption that 100% complete separation of all chemical elements constituting spent fuel is feasible is in conflict with the Second Law of thermodynamics. Report m38 *Nuclear power and the Second Law* explains this observation.

Furthermore the vitrification concept is (implicitely) based on the questionable assumption that the borosilicate glass with will remain stable for hundreds or even thousands of years and that no severe problems will arise with the borosilicate glass, caused by radiolytic reactions, heat generation, (re)crystallization and segregation of elements.

Deep geological repositories are even more needed in case of vitrification than in case of direct disposal of spent fuel, because the volumes of highly radioactive waste containing long-lived radionuclides are larger. These other wastes are liquids and solids, including the cladding hulls of the fuel elements. No concepts of deep geological repositories for these kinds of radioactive waste are known.

No safe final disposal facility for the highly radioactive glass is operational, only paper concepts and experiments exist.

The energy consumption of spent fuel reprocessing is exceedingly high, particularly if the energy consumption of the cleanup and dismantling of the reprocessing plant are included. For that reason the energy balance of once-though mode with vitrification is negative, even including the recycling plutonium and uranium as MOX fuel.

Unnamed problems

The concept of vitrification as a means for waste reduction is flawed by serious misconceptions and fallacies.

- Not included are the xircalloy cladding hulls (0.6 - 2 tonne per tonne fuel), which also are highly radioactive, containing long-lived radionuclides.
- A considerable number of radionuclides. present in substantial amounts, cannot be vitrified. In spent fuel nearly the full Periodic System of the elements is represented. Not all elements form stable oxides, or oxides that can be incorporated into a stable glass matrix, nor in Synroc, for example tritium and carbon-14.
- Separation of the components of spent fuel is inherently incomplete. This implies that all fractions from the separation process will be contaminated with undisirable nuclides, see report m20 *Reprocessing of spent fuel*.
- In the first step of the separation process - the chopping of the fuel elements into small pieces - the gaseous and volatile elements are set free, such as tritium H-3, carbon-14, iodine-129 and the noble gases (e.g. krypton-85). These radionuclides are virtually completely discharged into the environment.
- Massive amounts of low-level and medium-level radioactive waste originating from the nuclear chain are not accounted for.
- Severe problems arise with the borosilicate glass by radiolytic reactions, heat generation, (re)-crystallization and segregation of elements. These phenomena may cause a desintegrating of the glass matrix and consequently a high leachability by water of the solid mixture.
- To utilize the specific properties of Synroc, the fission product stream has to be fractionated again. Some nuclides, but not all, can be immobilized in boroslicate glass, other, but not all, can be immobilized in Synroc.

Assumptions and fallacies

The waste volume reduction concept by vitrification, as communicated with the public, may be based on starting points and assumptions which are questionable or even in conflict with basic laws of nature, such as:

- The view that short-lived radionuclides, which decay within a number of centuries (see Figure 1) would be not dangerous or less dangerous than long-lived.
- The view that waste containing less radioactive material per kg than spent fuel is nothing to worry about.
- The assumption that all long-lived radionuclides can be vitrified.
- The assumption that 100% complete separation of all chemical elements constituting spent fuel is feasible.
- The assumption that the borosilicate glass with the long-lived radionuclides will remain stable for thousands of years and that no severe problems will arise with the borosilicate glass, caused by radiolytic reactions, heat generation, (re)crystallization and segregation of elements.
- Neglect of the substantial fraction of the fission products and actinides which are discharged into the environment by the reprocessing plant, see also report m20 *Reprocessing of spent fuel*.
- Neglect of the large amounts of other radioactive wastes released during reprocessing (see Figure 3).

- Neglect of the immense amounts of radioactive waste resulting from decommissioning and dismantling of nuclear power plants and reprocessing plants.
- Neglect of an ever present consequence of the Second Law of thermodynamics: the unavoidable ageing and degradation of materials and structures, see report m38 *Nuclear power and the Second Law*.
- The assumption that waste containing short-lived radionuclides, however defined, could be stored safely in temporary above-ground facilities for more than four centuries and could be kept free of natural disasters, human failings, armed conflicts, criminal or terroristic actions.
- The assumption that future generations will have the economic means to maintain the storage facilities for the 'short-lived' radioactive waste adequately, without any revenue for them.
- The assumption that future generations will have the knowledge of the exact locations and properties of the stored 'short-lived' radioactive wastes from centuries before and will have the expertise and economic means to safely handle the wastes.

As pointed out above spent fuel contains many tens of different kinds of elements and only a limited number of these elements has the appropriate properties to be chemically immobilised.

Entropy generation by reprocessing

Reprocessing of spent fuel is an extremely costly and polluting process, greatly enhancing the health hazards posed by nuclear power, and raising severe security problems. These security problems can be limited by keeping the spent fuel elements from nuclear power stations intact. In the fuel elements all dangerous fissile and radioactive materials generated in the fission process are compacted in the smallest possible volume. Safe disposal of intact fuel elements in a geologic repository is the least hazardous way of dealing with this dangerous material and will require the least effort and financial investments.

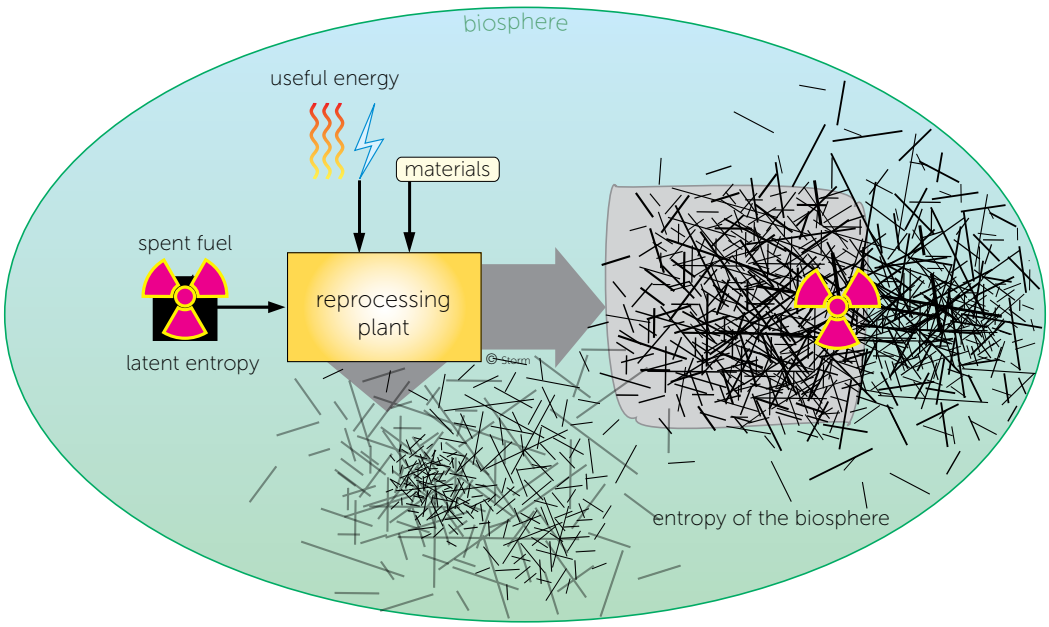

Figure 4

Symbolic representation of the entropy production by reprocessing of spent fuel. In spent fuel is a large amount of latent and delayed entropy confined to the volume of the spent fuel elements. In the reprocessing plant a substantial part of the content of the spent fuel is released into the environment and its latent entropy becomes an acute entropy increase of the biosphere. The other part is distributed over large volumes of originally non-radioactive materials, greatly increasing the entropy of that systems

n the reprocessing sequence the contents of the spent fuel are distributed over large volumes of non-radioactive materials (see figure 6), greatly increasing the entropy of the spent fuel contents. A signicant part of the radionuclides are discharged into the biosphere via aerosols, gaseous effluents and liquid effluents, causing the conversion of latent entropy into entropy of the biosphere.
New entropy is generated, according to the Second Law, by the consumption of energy and ordered materials in the separation processes. Entropy is also generated by the construction of the reprocessing plant.
In addition to the above mentioned entropy increases, reprocessing generates another form of latent entropy present in the construction materials that became contaminated during the operation of the plant by all kinds of radionuclides from the spent fuel. At issue is to prevent this latent entropy becoming entropy of the biosphere as a result of decommissioning & dismantling of the reprocessing plant itself.
The total quantity of radioactivity does not change by processes in the reprocessing plant, only the extent of dispersion changes.

Reprocessing offers no solution of radioactive waste problems, on the contrary, as is explained in the previous sections on P&T and vitrification. The amount of radioactivity in the nuclear waste streams is not

influenced by the mechanical and chemical operations in a reprocessing plant. Discharging a significant part of the radioactive substances into the environment cannot be conceived as a 'solution' to the nuclear waste problem. Actually the radioactivity from the spent fuel is dispersed over large volumes and masses of non-radioactive substances. Instead of volume reduction, the vitrification option results in a huge volume increase, worsening the waste problems beyond control.

The best way to handle spent fuel might be keeping the spent fuel elements intact, - in the fuel elements fissile and other radionuclides are compacted in the smallest possible volume -, to pack them in very durable containers and to dispose of in a safe geological repository (see report m32 *Geologic repositories*). Direct disposal poses the least risks and consumes the least materials and energy, prevents latent entropy in the spent fuel becoming entropy of the biosphere and prevents entropy generation by construction, operation and dismantling.
A number of countries, among other USA, Sweden, Finland and Canada, has chosen for this option.
It is a fallacy to believe in 'retrievable' storage of spent fuel. In no way it is possible to extract net usable energy from it, when all industrial processes needed to achieve reuse of spent fuel are accounted for. The Second Law is relentless.

References

Q262
Hollocher 1975
Hollocher T C,
Storage and disposal of high-level radioactive wastes, in:
The Nuclear Fuel Cycle,
Union of Concerned Scientists,
MIT Press, Cambridge, Mass., 1975.

Q263
JPL-77-69 1977
An analysis of the technical status of high-level waste and spent fuel management systems,
JPL-77-69,
Jet Propulsion Laboratory, Pasadena, CA, December 1977.

Q264
Bell 1973
Bell M J,
ORIGEN, the ORNL isotope generation and depletion code,
ORNL-4628,
Oak Ridge National Laboratory, Oak Ridge, Tenn, May 1973.
‹3445600501482.pdf›
web.ornl.gov/info/reports/1973/3445600501482.pdf
ORNL Research Libraries
Building 4500N, MS-6191
PO Box 2008
Oak Ridge, TN 37831-6191
USA

Q268
IAEA-187 1979
Characteristics of solidified high-level waste products,
Technical Report Series No 187,
International Atomic Energy Agency, Vienna, 1979.

Q271
Replaced by Q540

Q300
Charpak&Garwin 2002
Charpak G & Garwin RL,
'The DARI'
Europhysics News (2002) Vol 33 No.1
formerly:
www.europhysicsnews.com/full/13/article4/article4.html
‹ epn02104 ›
http://www.europhysicsnews.org/articles/epn/pdf/2002/01/epn02104.pdf
download 14 Febr 2016

Q513
IPFM 2011
Editors Feiveson H, Mian Z, Ramana MV & von Hippel F,
Spent fuel from nuclear power reactors. An overview of s new study by the International Panel on Fissile Materials,
International Panel on Fissile Materials IPFM, June 2011,
‹ipfm-spent-fuel-overview-june-2011.pdf›
http://fissilematerials.org/library/ipfm-spent-fuel-overview-june-2011.pdf
retrieved October 31, 2012.

Q540
WNA 2016a
Radioactive Waste Management,
World Nuclear Association.
http://www.world-nuclear.org/information-library/nuclear-fuel-cycle/radioactive-waste-management/.aspx
updated July 2016, retrieved August 2016

Q541
WNA 2012b
Waste Management: Overview
World Nuclear Association.
http://www.world-nuclear.org/info/Nuclear-Fuel-Cycle/Waste-Management-Overview/
updated December 2012, retrieved Nov 2015

Q656
IAEA-wmdb-st-1 2001
Radioactive Waste Management Status and Trends,
IAEA, August 2001
‹ IAEA-WMBD-ST-1.pdf ›
https://newmdb.iaea.org/library
retrieved 13 Jan 2016

Q657
IAEA-wmdb-st-2 2002
Radioactive Waste Management Status and Trends-issue #2,
IAEA, September 2002
‹ IAEA-WMBD-ST-2.pdf ›
https://newmdb.iaea.org/library
retrieved 13 Jan 2016

Q658
IAEA-wmdb-st-3 2003
Radioactive Waste Management Status and Trends-issue #3,
IAEA, August 2003
‹ IAEA-WMBD-ST-3.pdf ›
https://newmdb.iaea.org/library
retrieved 13 Jan 2016

Q659
IAEA-wmdb-st-4 2005
Radioactive Waste Management Status and Trends-issue #4,
IAEA, March 2005
‹ WMDB-ST-4.pdf ›
http://www-pub.iaea.org/MTCD/publications/PDF/WMDB-ST-4.pdf
and: https://newmdb.iaea.org/library
retrieved 12 Jan 2016