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Spent nuclear fuel – how dangerous is it?

**A report from the project
"Description of risk"**

Allan Hedin

Swedish Nuclear Fuel and Waste Management Co,
Stockholm, Sweden

March 1997

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SPENT NUCLEAR FUEL - HOW DANGEROUS IS IT?

A REPORT FORM THE PROJECT "DESCRIPTION OF RISK"

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FOREWORD

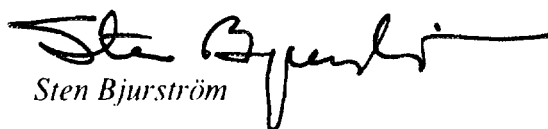
Our extensive contacts with the general public, political and community leaders, and large parts of the world's scientific community have made us at SKB acutely aware of the need for a better explanation of the ways in which radioactive material can be harmful to human beings. These are complicated issues, and the tendency of different interest groups to present only one side depending on their standpoint has not made it easier to get a fair and balanced perspective on exactly what is dangerous, how large the safety margins are, etc. The demands that are made on assessment of the long-term consequences with large safety margins are unusually stringent and go far beyond what is normally required in society. Naturally, this can be difficult to understand.

At the same time, we who work with these matters have often presented solutions and taken it for granted that many people have a clear understanding of the fundamental issues. It has become apparent today that we must devote much more effort to explaining what risks and hazards our proposals are intended to protect against.

After many years of intensive and broad research in international cooperation, we are in a much better position today to describe risks. We have had an opportunity to give thorough consideration to the fundamental conditions and to gather a more complete and broad body of facts than before. This makes it possible to put different problems in proper perspective and give an indication of their magnitude. Perhaps the most important message is that most of the radioactive substances that are harmful in spent nuclear fuel decay over the course of a few hundred years. The few substances that remain harmful for a longer time have chemical properties that keep them tightly bound to the environment in a deep repository.

We at SKB have therefore decided to address the most important issues in a special project – "Description of risk". Responsibility for compiling the basic material and ensuring that the standpoints rest on a firm scientific footing lies with SKB's Development division, in particular its Research and Development department. Besides the author Allan Hedin, Per-Eric Ahlström and Tönis Papp have been actively involved in producing the present report. It is our hope that this report will help to highlight the central issues pertaining to management and disposal of the spent nuclear fuel.

Stockholm in March 1997


Sten Bjurström

President

SUMMARY

The questions surrounding radioactive waste, and in particular spent nuclear fuel, are of great concern to many people. There is therefore a great need for comprehensible, clear and correct information. The subject contains many difficult questions: How can a layman grasp what radiation from radioactive waste really is? In what way is radiation harmful? How can man and the environment be protected against radiation? How can we grasp the long time spans involved? This publication has been written to try to bridge the gap between the expert and the scientifically interested layman when it comes to the risks surrounding radioactive waste.

Risk – Radiotoxicity and accessibility

Risks associated with spent nuclear fuel are often described in terms of radiotoxicity and accessibility. Radiotoxicity describes the harm which radiation can cause if people are exposed to it. Accessibility describes the degree to which a person can be exposed to radiation in different situations, e.g. during transport, interim storage or deep disposal. The goal of spent fuel management is to minimize the risks by making the fuel inaccessible to man and the natural environment for as long as it is hazardous.

Radiotoxicity

External and internal radiation

Certain types of radiation from radioactive substances penetrate matter easily. These substances are therefore hazardous to man when the radiation source is *external*, i.e. outside the body. Spent fuel emits powerful radiation with high penetrating power. The radiation in the vicinity of the fuel quickly gives lethal doses to unprotected persons. This radiation decays rapidly with time, but still requires protective measures for hundreds of years.

Other types of radiation are hazardous above all if the radioactive materials enter the body and emit their radiation there, *internal radiation*. Radioactive substances can enter the body primarily via food and via inhalation. During the first few decades, this radiotoxicity is dominated in the case of nuclear fuel by elements such as caesium and strontium. After these elements have decayed, the radiotoxicity is dominated by the remaining long-lived elements americium and plutonium.

Time scale

Nuclear fuel is fabricated of natural radioactive uranium mineral. During operation in a nuclear power plant, the radioactivity of the fuel increases greatly. After operation, activity and thereby radiotoxicity decline as the radioactive substances (isotopes of radioactive elements, called radionuclides) decay. After interim storage for about 40 years, when the waste is emplaced in the deep repository, only a few percent of the original radioactivity in the spent fuel remains.

Most radionuclides in spent nuclear fuel decay in the course of a few hundred years. Subsequently, the remaining radiotoxicity is completely dominated by a few nuclides with extremely low accessibility in most situations, including in a deep repository for spent nuclear fuel. A few of these nuclides will remain for very long times, up to 100,000 years. As time passes, the spent fuel will come more and more to resemble the natural uranium mineral from which it was fabricated. The radiotoxicity of such mineral, as well as of the spent fuel in the long run, is dominated by radiation from daughters of uranium (radium, radon, polonium, lead, etc.). These elements have always existed in nature and will always exist due to the very long half-life of uranium: 4.5 billion years.

Heat output

The radiation in the fuel is rapidly converted to heat. Heat output during reactor operation is very great. It is this thermal energy that is exploited and converted to electrical energy. One tonne of fuel develops about 30,000,000 watts of thermal power during operation. It is the enormous heat output during operation which can, if it is not controlled, lead to rapid, violent processes with disastrous consequences for the environment. This is also what happened at Chernobyl. (Swedish reactors have a design which makes such a sequence of events impossible.)

At the time of the planned deep disposal, the heat output of the fuel will have declined to about 1,300 watts per tonne, roughly equal to that of an ordinary radiator. The heat output of the fuel in the repository is thus only a tiny fraction of what it was in the nuclear reactor and far below what is required to cause rapid, dramatic consequences in the surrounding.

Criticality

Nuclear fuel has another property that constitutes a potential danger: If it is mixed with suitable materials and in special proportions, a self-sustaining chain reaction, such as in a nuclear reactor, can be obtained. This phenomenon is called *criticality*. Spent nuclear fuel is discharged from the

reactor for the very reason that its ability to sustain chain reactions has fallen to a low level, but in very extreme situations it may nevertheless become critical. It is therefore important to ensure that criticality cannot occur during handling of the fuel and after deposition in the deep repository. The potential danger of criticality can be avoided in a simple and reliable manner by limiting the amount of fuel in each canister and by incorporating materials in the canister which prevent a chain reaction from occurring.

No matter how the spent fuel is mixed with other materials, however, the chain reaction can never be so violent that it leads to an explosion. In other words, spent nuclear fuel from Swedish reactors can never, under any circumstances, cause a nuclear explosion.

Accessibility

A common way to deal with hazardous substances is to render them inaccessible. This is, for example, what is done when hazardous chemicals are kept in special containers and/or sealed spaces, or when medicines are kept in locked medicine cabinets. As long as a substance is not accessible to anyone or anything that can be harmed, it doesn't matter if it's hazardous.

Spent fuel is hazardous in different ways during long periods of time, and must be prevented from causing injury. This is done by making the fuel inaccessible, usually by isolating it behind some form of barrier. The fuel is a porcelain-like ceramic material that has low solubility in water, which also contributes greatly to rendering it inaccessible.

Accessibility before deep disposal

Spent nuclear fuel is handled in a long sequence of steps before the planned final deep geological disposal. After operation, the fuel remains in the reactor pressure vessel for a week or so, after which it is stored at the nuclear power plant for just under one year. Then the fuel is transported to a central interim storage facility. After about 40 years of interim storage, the fuel will be encapsulated and transported to the final deep repository. In all of these handling stages, accessibility is limited by special containers, or casks, during transport and by storing the fuel in water pools during the storage periods. Both the transport casks and the water in the storage pools effectively shield off the radioactivity emitted by the fuel.

Accessibility during deep disposal

The deep repository will render the waste inaccessible to man and the environment for a long time. This is achieved by a system of barriers and other factors:

- The fuel is placed in corrosion-resistant canisters with a steel insert providing mechanical strength.
- The canisters are surrounded by a layer of bentonite clay that protects the canister mechanically from small rock movements and prevents corroding substances from entering the canister. The clay also effectively absorbs any radioactive substances that are released if the canisters should be damaged.
- The canisters with surrounding bentonite clay are emplaced at a depth of about 500 m in the bedrock, where long-term stable mechanical and chemical conditions prevail.
- The chemical properties of the fuel and the radionuclides, for example their low solubility in water, also greatly limit transport of radioactive elements from the repository to the surface. This is particularly true of the elements which are most radiotoxic in the long term, such as americium and plutonium.

Together, the properties of the fuel and the barriers constitute a system that limits the accessibility of the spent fuel. The system is engineered so that accessibility is sufficiently limited even if one barrier should be defective. This is the meaning of the *multiple barrier principle*.

Normally, the copper canister alone is expected to isolate the fuel for such a long time that its radiotoxicity will decline to levels well below the radiotoxicity of the radioactive minerals that were mined to fabricate the fuel.

If the canister should be damaged, the chemical properties of the fuel (such as low solubility in water) and the clay buffer and rock comprise very effective barriers, particularly to many of the elements that are most radiotoxic in the long term. In Figure 1, the radionuclides have been divided into two categories: Category 1 comprises nuclides with very low water solubility and mobility in the buffer and the groundwater in the rock. Category 2 contains all other radionuclides in spent nuclear fuel. Figure 1 shows how radiological toxicity declines with time for the two categories. Long-term radiotoxicity is dominated completely by the radionuclides with the lowest accessibility.

Thorough evaluations of the safety of the repository are made in *safety assessments*. The safety assessments that have been performed for the conceived Swedish system for deep disposal of spent nuclear fuel have shown that the repository limits accessibility to the degree required in relation to the radiotoxicity of the fuel, with ample margin.

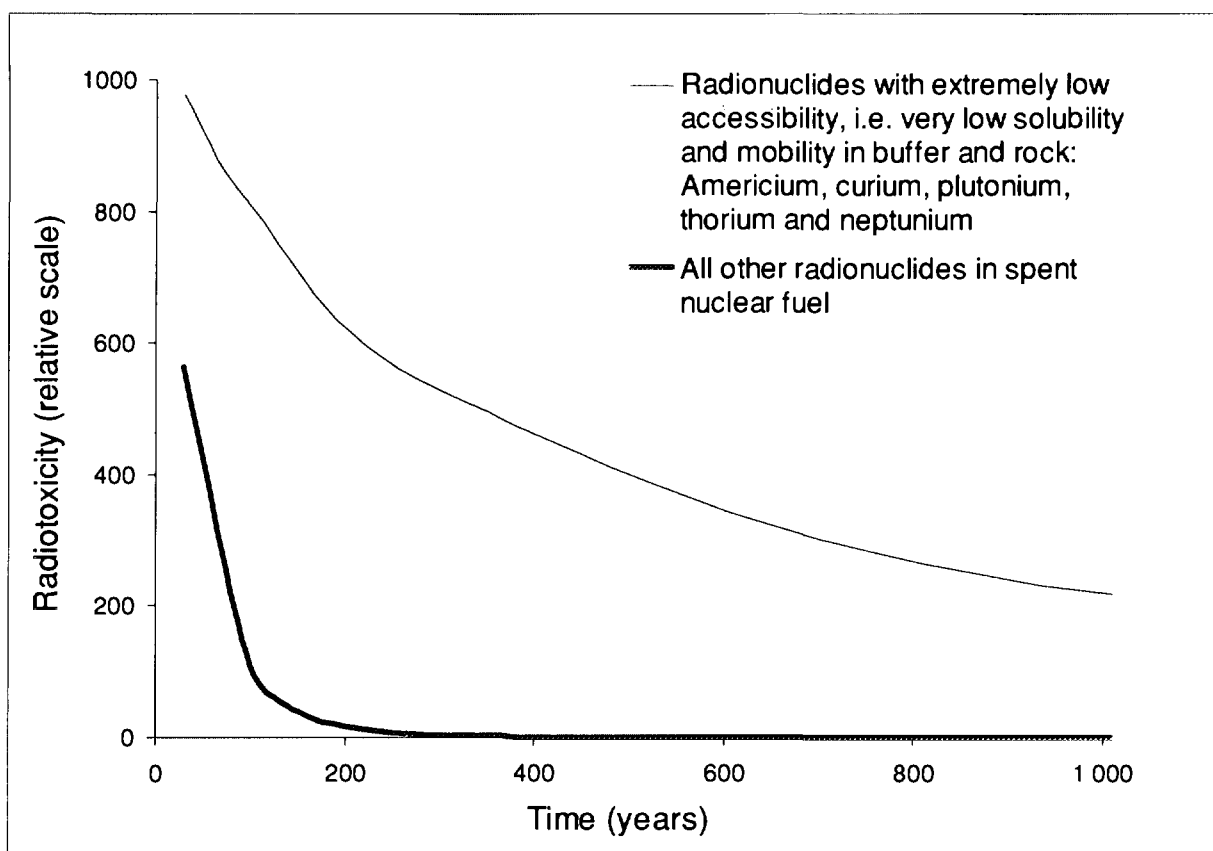


Figure 1. Radionuclides with extremely low accessibility in the deep repository dominate the radiotoxicity of the waste when it is deposited (emplaced). The blue line is dominated completely by plutonium and americium. The curves start at 30 years, since the spent fuel is planned to be kept in interim storage for 30–40 years before emplacement in a deep repository. By “radiotoxicity” is meant the harmful effects in a hypothetical situation where a person ingests the radionuclides via food. The radiotoxicity scale is the same as that used in Chapter 3, see e.g. Figure 3-11.

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INTRODUCTION

The issues surrounding radioactive waste, and in particular spent nuclear fuel, often arouse strong feelings in people. There are several reasons for this. We cannot perceive radiation from radioactive waste directly with our senses, which can make it difficult to form a notion of the risks – we tend to be disproportionately frightened and threatened by the unknown. The long time spans involved in discussions of spent nuclear fuel are also difficult for us to grasp. Without the facts, it can be difficult to understand why the risks posed by spent nuclear fuel from Swedish reactors have nothing to do with catastrophic events such as the accident at the Chernobyl nuclear power station or even nuclear weapons.

In addition to the fact that the issues surrounding radioactive waste are controversial and highly emotionally charged, nuclear power currently accounts for about 50 percent of Sweden's electricity supply. The management and disposal of the waste from nuclear power is therefore a matter of public concern about which everyone should have an opportunity to form an opinion.

Experts have access to a battery of technical tools, measuring instruments, mathematical models and concepts for describing and measuring radiation and hazards related to it. Much of this is difficult or impossible to understand for those without the right specialist knowledge.

The intention of this publication is to try to bridge the gap between the expert and the scientifically interested layman when it comes to understanding the risks surrounding radioactive waste. The report attempts to answer such questions as: What is ionizing radiation? In what way can radiation be harmful? How does radiotoxicity change over long spans of time? How can man and the environment be protected from radiation in the handling of spent nuclear fuel, now and in the future? The introductory chapter deals with a fundamental question for the whole approach of this report, as well as in the management of spent nuclear fuel: What exactly is a risk?

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2 CONCEPT OF RISK

2.1 INTRODUCTION

Risks of various kinds are a part of our everyday life. We assess and manage certain risks almost unconsciously, for example when we cross a busy road. Others require more careful analysis and consideration, for example when we are deciding whether to purchase a certain kind of insurance policy. Still others are not immediately visible and can only be identified and assessed by experts after advanced technical and scientific analyses. The latter category includes the risks associated with the depletion of the ozone layer and pollution by ecotoxins.

This report deals with the risks of radioactive waste. This chapter first discusses briefly what a risk really is. Some examples of risks and risk assessments within different fields are given, and the chapter is concluded with a brief general discussion of the risks of radioactive waste.

2.2 RISK – EXPOSURE AND HAZARD

What, then, is a risk, in the strict sense of the word? Certain characteristics always distinguish a risk, regardless of the type. A risk is always related to a threatening, or potential, *hazard*. A risk can further be said to consist of two components:

- a component that describes how often or to what extent the potential hazard occurs
- a component that describes the magnitude of the hazard.

The following example illustrates what we mean: You are faced with the temptation to park your car in an unauthorized spot for an hour. You consider two questions:

- How likely is it that a parking warden will happen by during the next hour?
- If one does, what would the consequences be?

The first question has to do with your *exposure* to the “hazard”, which in this case is a parking fine. The second is related to the *magnitude of the hazard*. The assessment of the *risk* is a weighing-together of exposure and hazard. If

parking wardens are numerous in the area (if the exposure is high), and if the amount of the parking fine (the magnitude of the hazard) is relatively low, the risk may be considered acceptable. If, on the other hand, the majority of the town's parking wardens are on strike at this particular time (low exposure), the risk may possibly be deemed reasonable, even if the fine would be very high (large hazard).

The above example illustrates the fact that a risk consists of two components: one that describes the exposure and one that describes the magnitude of the hazard. The exposure expresses the probability of a hazard occurring, or the extent to which it occurs. The component that describes the magnitude of the hazard is related to the consequences if the potential hazard occurs. The risk is a product of the two. The example also shows that we constantly make risk assessments in our everyday lives and let them guide our actions.

The terminology may vary between different types of risks, but at bottom a risk evaluation always consists of two components in one form or another. Further examples follow below.

Example 1 – PCBs in fish

The health risks associated with pollutants in the environment are normally expressed in terms of *exposure and hazard*. Exposure quantifies the extent to which a normal consumer of fish in Sweden is exposed to PCBs through his/her consumption of fish. Hazard (toxicity) describes the consequences of a given exposure, e.g. in the form of a measure of the carcinogenicity (cancer-causing property) of PCB compounds.

Risk is a product of exposure and hazard. A high exposure may be acceptable if the hazard is low, and vice versa. In this case, the risk can be reduced by reducing the exposure, in the short run by reducing consumption, in the longer run by reducing discharges of PCB into the environment. Hazard cannot be influenced in the same way; it is dependent on the chemical properties of PCB compounds.

Example 2 – Road traffic

Risks in road traffic can be described in a similar manner. Exposure can be expressed here as accident frequency. Exposure is higher on accident-prone sections of road, those with a high accident frequency. The magnitude of the hazard has to do with the consequences of an accident.

Risks in road traffic can be reduced by controlling both the frequency and the consequences of accidents. The accident frequency (exposure) can be reduced by building safer roads or grade-separated intersections, or by

sanding or salting when the road surface is icy. Legislation requiring motorists to drive with their headlights on is another example of a measure aimed at reducing the accident frequency.

The consequences of car accidents (the magnitude of the hazard) can be reduced by improving the safety design of the vehicle, for example by building more crash-proof cars. Legislation mandating the use of seat belts is another way to reduce the hazard in car accidents. Certain measures, for example lowering the speed limit, can reduce the risk by influencing both exposure and hazard.

Example 3 – Children and household chemicals

In many contexts, risk is discussed in terms of *accessibility* and *toxicity*. To prevent children from being injured by household chemicals, they are kept out of reach, for example behind locked doors or in child-safe packages. Exposure is limited by reducing *accessibility*. The *toxicity*, or biological hazard, of the chemical can be expressed, for example, as a measure of the corrosive properties of an acid.

The risk in this case is a product of accessibility and toxicity. The risk can be reduced by reducing exposure, by making the chemicals less accessible. Toxicity remains unchanged, since it is dependent on the properties of the chemical.

The more hazardous/toxic a chemical is, the more stringent are the demands on limiting accessibility – medicines are often kept in locked cabinets in the home.

All the above examples illustrate that a risk has two components: one related to exposure and one related to hazard or toxicity. The risk in question can be reduced by controlling one or both of the components. The next section shows how these principles are applied to the risks surrounding radioactive waste.

Different usages

It is worth noting that the word “risk” in everyday usage is often used in a different sense than the technical term discussed above. In the sentence “There is a great risk that it will rain tomorrow” the word “risk” is used synonymously with “probability” and thus has to do with exposure. Nothing is said about the consequences of the rain.

2.3 RADIOACTIVE WASTE

Risks associated with radioactivity fit in well with this way of describing risks. The terms *accessibility* and *radiotoxicity* are often used in this context.

Accessibility

Exposure is limited by keeping radioactive elements or nuclides isolated from man and the environment, in other words by keeping their accessibility low. This is done in different ways depending on the kind of radioactive material in question. Spent nuclear fuel, which is what this report is about, is managed in a series of steps after having been discharged from a nuclear reactor. Accessibility is limited by special casks during transport and by keeping the fuel submerged in water during an interim storage period. The planned final disposal in the bedrock greatly reduces accessibility by means of a series of engineered and natural barriers. The inherent properties of the fuel, for example its very low solubility, further limit accessibility.

Radiotoxicity

The toxicity of a radionuclide is dependent on its activity, and on what type of radiation its radioactive disintegration (decay) gives rise to. A distinction is made between two types of radiation: external and internal. External radiation is emitted by an external radiation source and penetrates the body from the outside. Internal radiation comes from radioactive substances that enter the body, via ingestion or inhalation, and emit their radiation there.

There are fairly precise and generally accepted measures of radiotoxicity. Radiotoxicity can therefore be described in more exact terms than many other hazards, for example those associated with many chemical compounds. Another distinguishing characteristic of radioactive substances is the fact that their toxicity declines with time as the substance decays.

Risk

Risk is once again a product of hazard (radiotoxicity) and exposure (accessibility). Since radiotoxicity varies with time, varying requirements are made on limitation of accessibility in various handling stages.

This report deals specifically with the radiotoxicity and accessibility of spent nuclear fuel. Chapter 3 describes how radiotoxicity can be measured and how it develops with time. Chapter 4 discusses how accessibility is reduced in various stages of handling and storage. The fifth and final chapter shows in general terms how the concept of risk can be applied to the long-term safety of a deep repository for spent nuclear fuel.

3 RADIOTOXICITY

3.1 INTRODUCTION

This chapter describes how the biological hazard or radiotoxicity of a typical spent fuel from a Swedish nuclear power plant develops after the fuel has been removed from the reactor.

The chapter begins with a brief review of the various steps in the production of nuclear fuel, from mining of uranium ore to finished fuel. The purpose is mainly to show how much uranium in the form of ore is consumed in the production of a given quantity of fuel. The radioactivity and radiotoxicity of the spent fuel can then be compared with values for an equivalent quantity of natural uranium.

The radiotoxicity of the fuel will decline with time and eventually be comparable to that of the natural uranium. Since the intention is that the fuel is to be disposed of deep down in the crystalline bedrock, a situation will eventually be reached reminiscent of the original one: a radioactive material in the bedrock with a radioactivity and a radiotoxicity comparable to that of the natural uranium material originally mined to fabricate the fuel.

After the description of nuclear fuel production, the way in which the *radioactivity* of the spent fuel changes with time is shown. Its activity is compared with that of the mined ore. The development of the decay heat, i.e. the heat output resulting from the radioactive decay of the fuel after discharge from the reactor, is also discussed. To describe the *radiotoxicity* of the fuel, radioactivity must be combined with a measure that describes the harmful effects of the radiation. This is done in section 3.5. The radiotoxicity of the fuel is also compared with that of the natural material.

In section 3.6, the properties of the spent nuclear fuel are briefly compared with the properties of the fuel in operation in the reactor. The chapter is then concluded with a brief discussion.

3.1.1 Basic facts about radionuclides

For the sake of the subsequent discussion, some basic facts about radioactivity are given below.

All matter consists of atoms. Atoms have a nucleus and a surrounding “cloud” of electrons. The nucleus consists primarily of protons and neutrons. There are around a hundred *elements* occurring in nature. An element is

distinguished by the fact that its atoms have a given number of protons in its nucleus. All atoms of the element oxygen, for example, have 8 protons, while atoms of lead have 82. The number of neutrons in the nucleus of an element can vary. A lead atom may, for example, have 124, 125 or 126 neutrons in its nucleus. We say that lead has several *isotopes*. The three isotopes are designated lead-206, lead-207 and lead-208. The numbers in the names indicate the sum of the number of protons and neutrons in the nuclei of the different isotopes. The word *nuclide* is often used as a synonym for isotope.

Most isotopes or nuclides that occur in nature are stable. A small number, however, have an excess of energy and strive to get rid of it by radioactive decay. These isotopes are called *radionuclides*. The rate at which a radionuclide decays is given by its *half-life*. The half-life is the time it takes for half of the original quantity of a radionuclide to decay. After a half-life, half of the original quantity remains, after two half-lives one-fourth remains, etc. Half-lives can vary from fractions of a second to billions of years. Two examples of radionuclides that exist in nature are uranium-238 and uranium-235, with half-lives of 4.5 and 0.7 billion years, respectively.

Almost all nuclides undergoing radioactive decay emit *alpha or beta particles*. Alpha particles consist of two protons and two neutrons (a helium atom) from the decaying nucleus. Beta particles are energetic electrons which the decaying nucleus emits at the same time as a neutron in the nucleus is converted to a proton. In both alpha and beta decay, *gamma rays* are also emitted, which are similar to X rays.

Uranium and certain other heavy radionuclides decay in a series of steps known as a *decay chain*. The decay chain that begins with uranium-238 is shown in Figure 3-1. There are four different chains for the heavy nuclides. Three end with the formation of stable lead, the fourth with the formation of stable bismuth.

Radioactivity is measured in a unit called a *becquerel* (Bq). One becquerel is equal to one disintegration per second.

A person exposed to ionizing radiation is said to be exposed to a *radiation dose*. Radiation dose is measured in a unit called a *sievert* (Sv). The concept of radiation dose is explained more thoroughly in section 3.5.

Uranium-238	4.5 billion years
↓ α	
Thorium-234	24 days
↓ β	
Protactinium-234	1.2 minutes
↓ β	
Uranium-234	246,000 years
↓ α	
Thorium-230	75,400 years
↓ α	
Radium-226	1,600 years
↓ α	
Radon-222	3.8 days
↓ α	
Polonium-218	3.1 minutes
↓ α	
Lead-214	27 minutes
↓ β	
Bismuth-214	20 minutes
↓ β	
Polonium-214	163 microseconds
↓ α	
Lead-210	22 years
↓ β	
Bismuth-210	5 days
↓ β	
Polonium-210	138 days
↓ α	
Lead-206	Stable

Figure 3-1. *Uranium-238 is transformed via natural decay in a series of steps to lead. The half-lives of the different nuclides are given, along with the type of decay, α or β .*

3.2 PRODUCTION OF NUCLEAR FUEL

Nuclear fuel is produced from uranium ore in a series of steps from mining of uranium ore to fabrication of fuel assemblies in a fuel fabrication plant. The radioactive substances in uranium mineral consist of the uranium isotopes uranium-235 and uranium-238, and uranium daughters, i.e. the radionuclides that are formed via decay of the uranium.

The different steps in the production of nuclear fuel are described in Figure 3-2 and are in brief as follows:

1. Mining of uranium ore under ground or in open-pit mines. The radioactive isotopes in the ore are uranium-238, uranium-235 and uranium daughters. The uranium content typically consists of 99.3% U-238 and 0.7% U-235. In addition, there is about 0.005% U-234, which is a decay product of U-238. The uranium daughters together are more radioactive than the uranium.
2. Extraction of uranium in the form of a uranium oxide, U_3O_8 , known as yellowcake, from the ore. This is done in a uranium mill, often adjacent to the mine. In this step the uranium daughters are separated as mill tailings, which are returned to the mine or deposited on the ground.
3. Conversion of U_3O_8 to uranium hexafluoride, UF_6 . This is done because UF_6 is a chemical form of uranium that is suitable for enrichment, the next step in the process.
4. Enrichment of the isotope U-235. It is U-235 that is used as fuel in a nuclear power reactor, so the object is to increase the concentration of U-235 by means of enrichment. The U-235 content is typically raised from 0.7% to 3.5% in enrichment. Due to the nature of the enrichment process, the concentration of U-234 is also raised to about 0.031%. The residue, depleted uranium, may in the future either be used for further energy production or treated as waste. The depleted uranium typically consists of 0.0013% U-234, 0.3% U-235 and 99.7% U-238. With these concentrations, 8 tonnes of natural uranium gives rise to 1 tonne of enriched uranium and 7 tonnes of depleted uranium, all in the form of uranium hexafluoride [3-1].
5. Fabrication of fuel assemblies. The enriched uranium hexafluoride is converted into uranium dioxide powder, UO_2 . The powder is compacted into cylindrical pellets, which are assembled into fuel rods. The pellets have a ceramic, porcelain-like structure.

The purpose of the above description, and Figure 3-2, is to provide an outline of the major mass and activity flows for radioactive elements in the different stages of production of nuclear fuel. The description does not give any details about, for example, the minor releases of radioactivity to water and air which occur in different processes.

In simplified terms, the radionuclide content of the uranium mineral is divided by the various processes into three fractions: uranium daughters, depleted uranium and enriched uranium.

Of 8 tonnes of uranium in uranium ore, 1 tonne of fuel is obtained. The remaining 7 tonnes are separated during enrichment as depleted uranium. Of the radioactivity originally present in the ore, most is now in the uranium daughters that are separated in the uranium mill. The rest of the activity is present in the depleted and the enriched uranium.

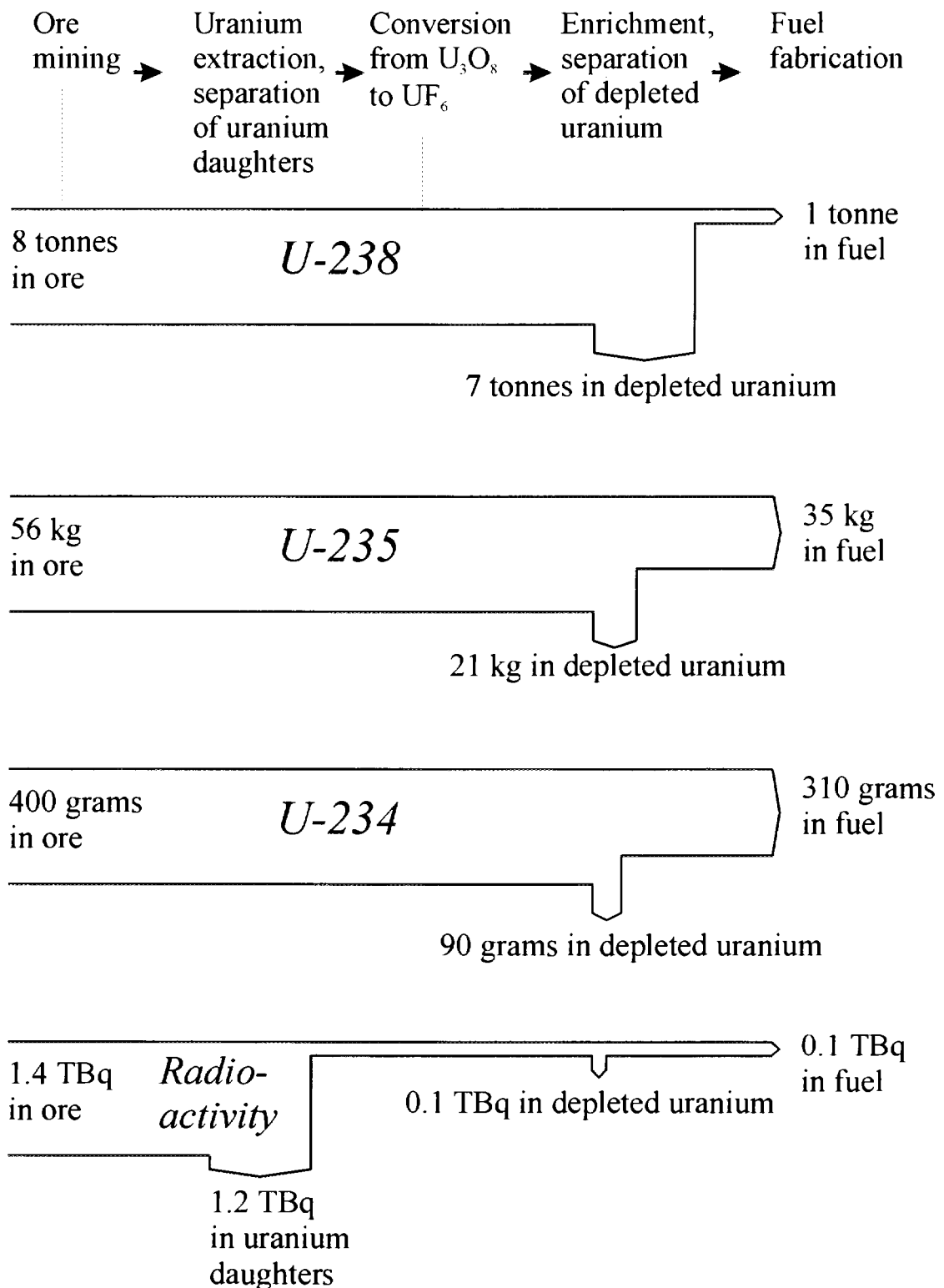


Figure 3-2. *Production of nuclear fuel. A certain quantity of ore contains 8 tonnes of uranium-238. The uranium is roughly divided in the various steps of processing into 1 tonne of enriched uranium (nuclear fuel) and 7 tonnes of depleted uranium. The isotopes U-235 and U-234 account for small portions of the masses in the different fractions. The uranium daughters separated in the uranium mill contain most of the radioactivity originally present in the ore.*

Figure 3-3 shows radioactivity versus time for the uranium daughters and for depleted uranium. The enriched uranium has also been included for the sake of completeness. The sum of the radioactivities of the depleted and the enriched uranium, plus that of the separated uranium daughters, is equal at any given time to the activity of the equivalent quantity of uranium ore. This activity level, which is virtually constant for billions of years, is also shown in the figure.

The curve for enriched uranium in Figure 3-3 is unrealistic. The enriched uranium is intended to be used as fuel in a reactor, which means its radioactivity will increase sharply and not follow the curve shown in the figure. This is discussed in the next section.

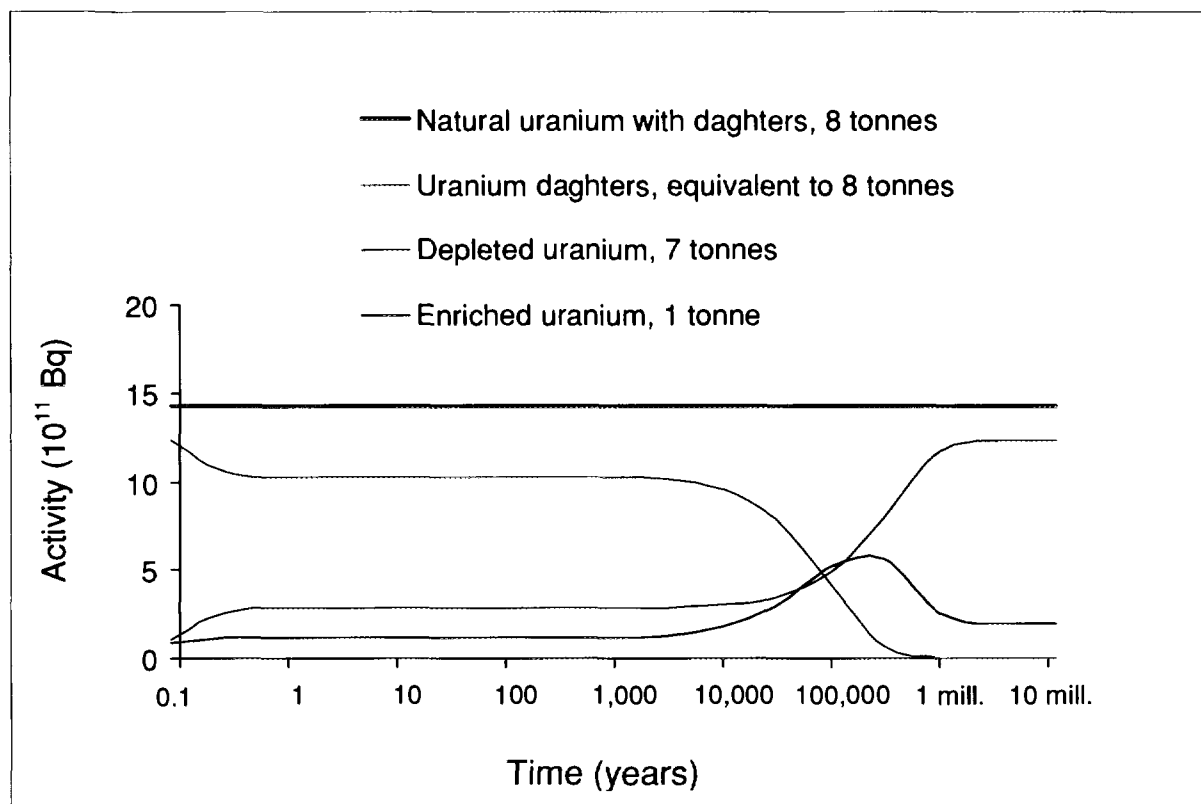


Figure 3-3. Development of the activity of the different fractions in Figure 3-2. For almost 100,000 years the uranium daughters account for the bulk of the activity. The activity of the depleted uranium increases as the uranium decays and new radioactive daughter products are formed. The activity of enriched uranium reaches a peak at around 100,000 years. The increase is due to the formation of daughter products primarily from the decay of uranium-234, which has been greatly enriched. Uranium-234 has a half-life of 246,000 years, which means that its activity declines after this time.

3.3 RADIOACTIVITY OF SPENT NUCLEAR FUEL

During operation in a nuclear power reactor, nuclei of the uranium isotope U-235 are split in a process called fission. New radioactive substances are formed in this process, and the radioactivity of the fuel material has increased sharply after operation in the reactor, see section 3.3.1 below. Most of these radionuclides have a very short half-life, but significant quantities of long-lived nuclides are also formed. This makes it necessary to handle the spent fuel in a safe manner for a long time to come.

The detailed radionuclide content of spent nuclear fuel is dependent on the fuel type and the conditions that have existed during operation. The *burnup*, i.e. the total energy extracted from each unit of fuel, is particularly important for the quantities of long-lived nuclides. For nuclides that are short-lived in comparison with the fuel's operating time, the *specific power*, i.e. the energy extracted every second from each fuel assembly during operation, is of greater importance. For the purpose of discussion in this report, fuel type and operating conditions have been chosen to illustrate typical conditions for the Swedish nuclear power programme.

The fuel type chosen for the presentation is SVEA 64, intended for boiling water reactors. The formation of radionuclides during operation has been simulated in a computer [3-2] with a burnup of 38 MWd/kg uranium and a specific power of 23.8 kW/kg uranium. The simulation has resulted in a calculated radionuclide content immediately after discharge from the reactor [3-3]. This radionuclide inventory has been used as a point of departure for calculations of the subsequent development of the inventory, see Appendix 1.

Radionuclide content is normally reported in the form of radioactivity. In other words, instead of giving the quantity of radionuclides directly (in kg or m³), the radioactivity to which they give rise is given in becquerels (disintegrations per second).

Figure 3-4 shows how radioactivity changes with time from about one month after discharge. To illustrate how activity declines in the long term, the figure has been divided into three parts with different activity scales.

3.3.1 Fission and activation products plus actinides

The spent fuel can be said to have two general categories of radionuclides: fission and activation products, plus actinides.

Fission products are the new materials that are formed when heavy atomic nuclei, such as uranium or plutonium, are split in the fission process in the nuclear reactor. Examples of fission products are iodine (I-129, I-131), caesium (Cs-134, Cs-135, Cs-137) and strontium (Sr-90). *Activation products* arise when electrically neutral particles, neutrons, which are formed in the fission process, are absorbed by different materials, chiefly

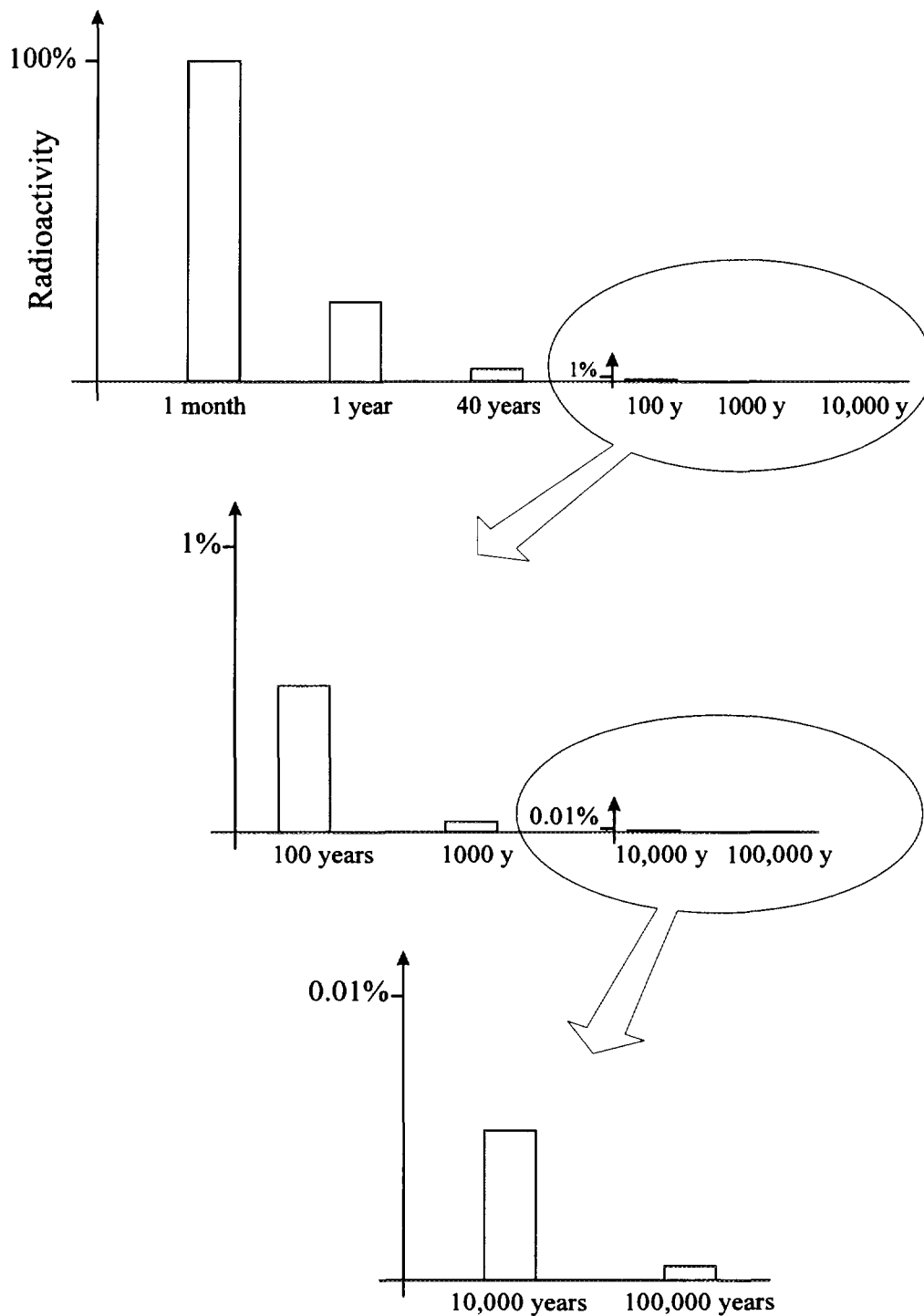


Figure 3-4. Radioactivity of a tonne of spent nuclear fuel of type SVEA 64 with a burnup of 38 MWd/kg U. To illustrate how the radioactivity declines over a long time perspective, the figure has been divided into three parts with different activity scales. (The 100% level on the top part corresponds to an activity of $3.35 \cdot 10^{17}$ becquerel.)

metals, in the fuel assembly. These substances can thereby become radioactive. Cobalt, nickel and niobium are examples of substances that are activated in this way.

The actinides consist of the uranium in the fuel plus the elements that are formed when uranium absorbs one or more neutrons without splitting. These elements, which are heavier than uranium as a result of the neutron absorption, are called transuranic elements, the most important of which is plutonium (Pu-238, Pu-239, Pu-240, Pu-241 etc.). Both uranium and transuranics are radioactive and decay in a series of steps in decay chains, see section 3.1.1. The quantity of radionuclides in the latter steps of the chains can thereby increase with time even after the fuel has been discharged from the reactor. This phenomenon is called *ingrowth*. An important example of ingrowth in spent fuel is the formation of americium-241 by β decay of plutonium-241.

Figure 3-5 shows the decline of the radioactivity in the fuel with time with logarithmic scales to allow longer time spans to be shown. The graph also shows how the total activity is distributed among fission products and actinides. The figure shows that the activity is dominated by fission and activation products during the initial 100 years, while actinides and actinide daughters dominate clearly after 500 years. This is a very important fact to bear in mind when considering the risks associated with the proposed deep disposal of spent nuclear fuel: As will be evident from Chapters 4 and 5, the actinides that dominate long-term activity have extremely low accessibility in a deep repository.

In a long time perspective, as the radioactive substances formed during operation decay the spent fuel will increasingly come to resemble the mineral that was originally mined to produce the fuel. What will remain will be the naturally occurring uranium isotopes U-238 and U-235, with half-lives of 4.5 and 0.7 billion years, respectively. The naturally occurring daughter products that are constantly formed by the chain decay of the uranium isotopes will also remain in the spent fuel far into the future.

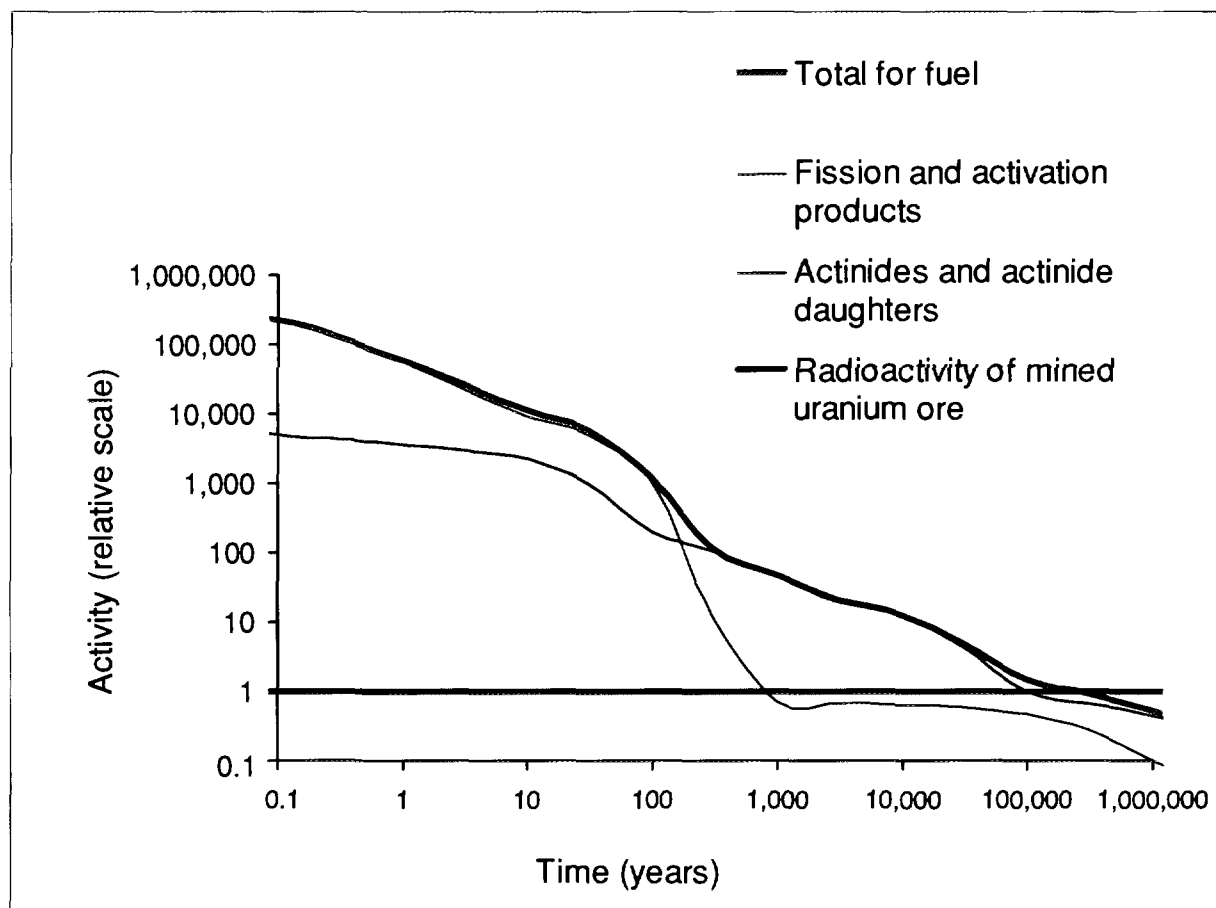


Figure 3-5. *Relative activity of spent nuclear fuel of type SVEA 64 with a burnup of 38 MWd/kg U. The activity is dominated during the first 100 years by fission products, thereafter by actinides.*

3.3.2 Comparison between mineral and the fuel cycle's fractions

Regard once again for a moment Figure 3-3, which shows the activities of the different fractions in fuel production. Then look at Figure 3-6, where the curve for enriched uranium is replaced with the curve for spent fuel. In addition, the activity scale is logarithmic to cover the wide range of activities presented in the figure. Let us now compare the radioactivity for two situations:

- The mineral is left in the uranium mine.
- The ore is mined, and the uranium is separated, enriched and used as fuel.

What do the total activities look like in the different cases? In case a (blue line), as we have seen in Figure 3-3, the activity remains virtually constant for times comparable to the half-life of uranium-238 (4.5 billion years). In case b (red line), the activity is much higher to begin with and declines

steadily for about 100,000 years. At that time the activity has fallen to a value near that of case a. During these 100,000 years the activity in case b is dominated completely by the contribution from spent fuel. After 100,000 years the activity declines more slowly and parallels case A.

Thus, the nuclear fuel cycle gives rise to considerably increased radioactivity during a period of about 100,000 years. To discuss what risks this entails, it is necessary to understand how activity is translated into radiotoxicity, as well as how accessible the radionuclides responsible for activity and radiotoxicity will be in the long term.

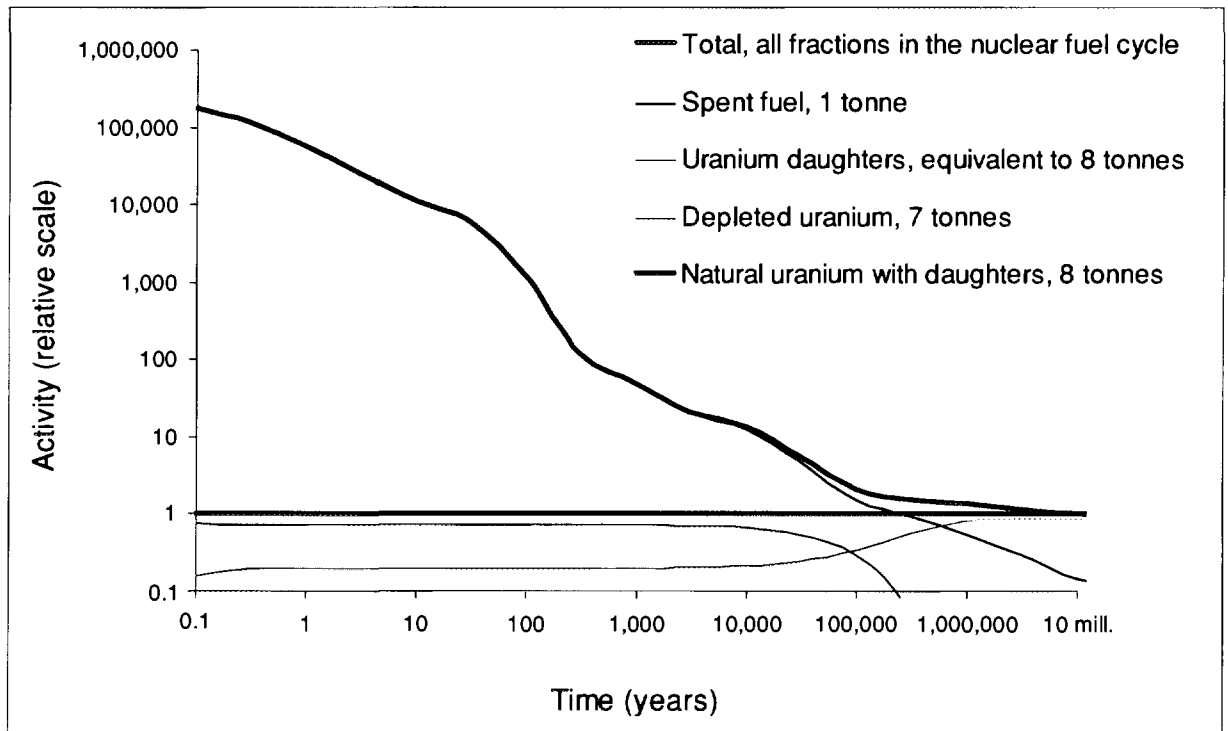


Figure 3-6. Radioactivity of uranium mineral (blue line) and all fractions that arise when the same quantity of uranium mineral is used in the nuclear fuel cycle (red line). The different fractions are the spent fuel, the depleted uranium and the uranium daughters that are separated in the uranium mill.

3.4 DECAy HEAT OF SPENT NUCLEAR FUEL

The radioactive disintegrations in the spent fuel liberate energy, which generates heat in the waste. It is important to know the magnitude of this heat output, since elevated temperatures accelerate chemical and other processes, for example during long-term deep disposal of spent nuclear fuel.

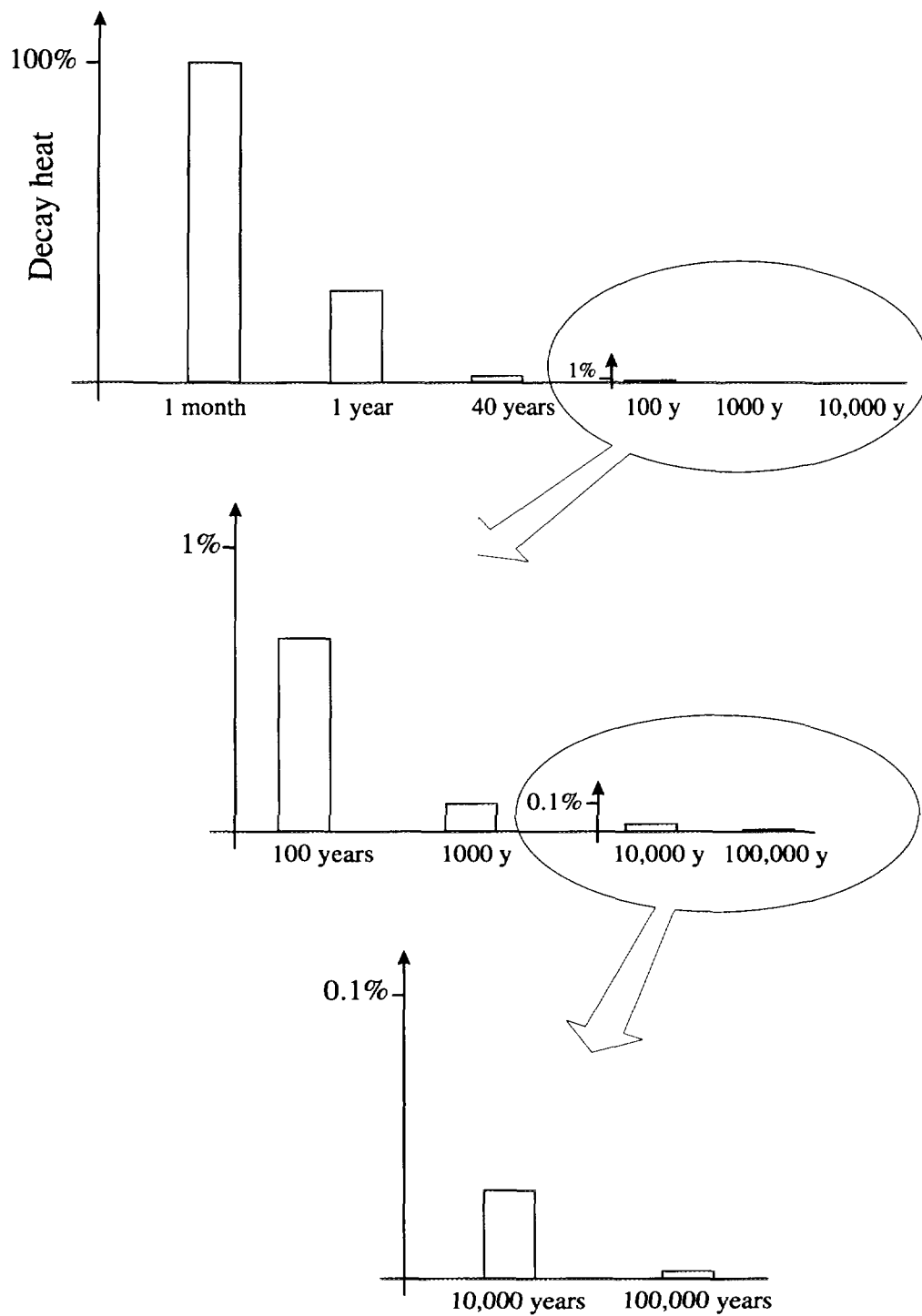


Figure 3-7. Decay heat for 1 tonne of spent nuclear fuel of type SVEA 64 with a burnup of 38 MWd/kg U.

The heat output in the fuel after discharge from the reactor is called *decay heat*. Figure 3-7 shows how the decay heat for one tonne of spent nuclear fuel declines with time.

When the fuel is to be emplaced in the deep repository after about 40 years, its heat output is just over 1300 watts per tonne of fuel, which is comparable to that of an ordinary radiator or a powerful toaster. This heat output continues to decline according to Figure 3-7. Decay heat is discussed further and compared with heat generation during operation in the reactor in section 3.6.1.

3.5 RADIOLOGICAL TOXICITY OF SPENT NUCLEAR FUEL

3.5.1 General about radiotoxicity of ionizing radiation

Radiation is harmful to living organisms because it is capable of damaging or killing biological cells. In man, high radiation doses during short spans of time can lead to death due to damage to the central nervous system, the gastrointestinal tract or the bone marrow. If the doses are very high, death may occur immediately. Lower radiation doses can above all cause cancer and chromosomal damage.

The unit of measurement for radiation doses to man is the sievert (Sv). The unit is a measure of harmful effect in the cells of the body, taking into account the fact that certain organs are more sensitive than others. We are constantly surrounded by natural background radiation, which in Sweden is of an order of magnitude of a thousandth of a sievert per year, 1 mSv/y. In addition, we are exposed to radiation by medical treatments and by radon in homes. These doses vary widely from one person to another. On average, a Swede is exposed to about 4 mSv/y if we add up these three principal sources [3-5].

A radiation dose of about 5,000 mSv is lethal if it is received all at once. Only in very extreme situations have people been exposed to lethal doses, such as when the atomic bombs were dropped in Japan during the Second World War and during the extinguishing work following the nuclear power accident in Chernobyl.

What radiation dose is needed to cause cancer? This is a much-disputed question. It has not been clearly established whether the likelihood of contracting cancer stands in direct proportion to the size of the dose when it comes to small doses. Experimental observations only exist for relatively high doses, hundreds of mSv and upward. Various national and international regulatory bodies who work with radiation protection matters normally assume that the probability of contracting a fatal cancer is *roughly five percent after exposure to a radiation dose of 1,000 mSv* [3-6].

What about lower doses? Here we have no actual observations to refer to. By assuming that the probability of cancer is proportional to the dose even down to very small doses, we can be sure of not underestimating the

probability at low doses. A dose of 100 mSv would then be assumed to give a probability of 0.5 percent for a fatal cancer, a dose of 10 mSv 0.05 percent, etc.

The assumption of proportionality between dose and probability of cancer is often called the *linear hypothesis*. This hypothesis is disputed at small doses. In this report, dose will be used as a direct measure of radiotoxicity. Double the dose is assumed to cause double the radiotoxicity, even at small doses. In other words, the linear hypothesis is assumed in this report.

Alpha, beta, gamma and neutron radiation

Radioactivity is measured in disintegrations per second, with no distinction between different types of radiation. When we discuss the hazard or toxicity of radiation, we must also take into account the type of radiation.

Almost all radionuclides in spent nuclear fuel are transformed by either alpha or beta decay. When an atomic nucleus undergoes alpha decay, it emits positively charged particles consisting of two protons and two neutrons. In the case of beta decay it emits negative electrons. In both cases, the nucleus also emits gamma rays, similar in nature to X rays.

A few elements in the spent fuel also emit neutrons when they decay. Neutrons are electrically neutral particles which occur in all atomic nuclei, except hydrogen.

External and internal irradiation

The radiation dose to which a given type of radiation gives rise in the human body is strongly dependent upon whether the radiation is emitted inside or outside the body. A distinction is therefore made between *internal* and *external* irradiation.

Only gamma rays and neutrons are able to penetrate the body to any great extent. Alpha particles are stopped by the outermost layers of skin, while beta particles can penetrate one or two centimetres. If alpha- or beta-emitting substances enter the body, e.g. by inhalation or food ingestion, their harmful effects are that much greater.

The external radiation to which humans can be exposed from spent nuclear fuel is therefore dominated completely by gamma rays and neutrons. As far as internal radiation is concerned, it is above all alpha and beta particles that need to be protected against.

Radiotoxicity of some naturally occurring isotopes

Radioactive minerals containing the elements uranium and thorium exist in nature. As was evident from section 3.1.1, these elements decay in a series of steps in which radioactive daughter products are formed. It is above all certain of the daughter products (radium, radon, lead, polonium, etc.) that are hazardous in the minerals. Pure uranium and thorium are much less hazardous (radiotoxic), since they have very long half-lives, i.e. low activities.

The radioactive gas radon is formed by the decay of uranium and thorium. The isotope radon-222 is formed in the decay chain that starts with uranium-238, see Figure 3-1. Radon-220 is formed by the decay of thorium-232. Radon and its daughter products can, if they enter the lungs, adhere to the lung tissue and cause lung cancer. Radon constitutes a radiation risk in mines, and particularly in combination with smoking. Some homes may also have such high levels of radon in the indoor air that the radiation protection authorities recommend corrective measures.

Radium-226 is formed by the decay of uranium-238 (Figure 3-1). If it enters the body, radium accumulates in the bone marrow, where it can cause a form of cancer known as osteosarcoma. This has sometimes afflicted individuals who have worked with radium paint, which was previously used e.g. in luminescent wristwatch dials.

3.5.2 External irradiation

External irradiation is often given as a dose rate. The dose rate tells how large a dose a person is exposed to by a radiation source per unit time and is often expressed in the unit mSv/h.

Figures 3-8a and b show the external radiation dose to which a person is exposed at a distance of 1 metre from one tonne of spent nuclear fuel. The contribution from both gamma and neutron radiation at different times after discharge from the reactor is shown. Approximately one year after the fuel has been discharged from the reactor, the dose rate is around 1,000,000 mSv/h. This means that a lethal dose, about 5,000 mSv, is received in about 20 seconds. The dose is dominated completely by the contribution from gamma rays. The radiation declines with time, but the dose rate after 40 years, when the spent fuel is to be emplaced in the deep repository, is still as high as 65,000 mSv/h. Protective measures are therefore required against external radiation in all handling of spent nuclear fuel, from discharge from the reactor until final deep disposal. Chapter 4 explains how this is done. Figure 3-8 also shows that the dose from neutron radiation is always much less than that from gamma radiation, but that neutron radiation declines more slowly.

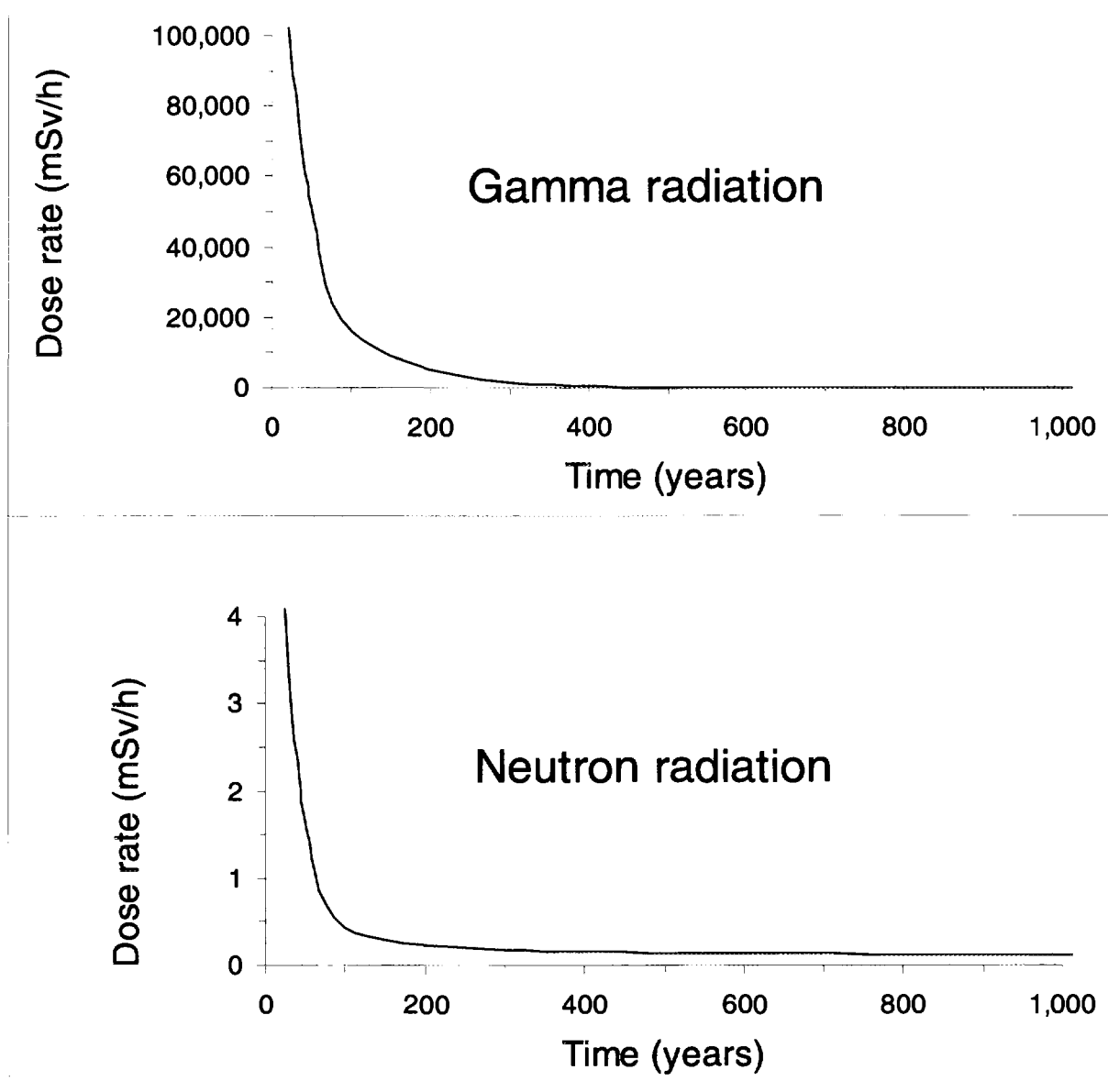


Figure 3-8a. Dose rate at a distance of 1 metre from one tonne of spent nuclear fuel at different times after discharge from the reactor. The data were taken from [3-4]. The same data with logarithmic axes, permitting longer time spans to be covered, are shown in Figure 3-8b.

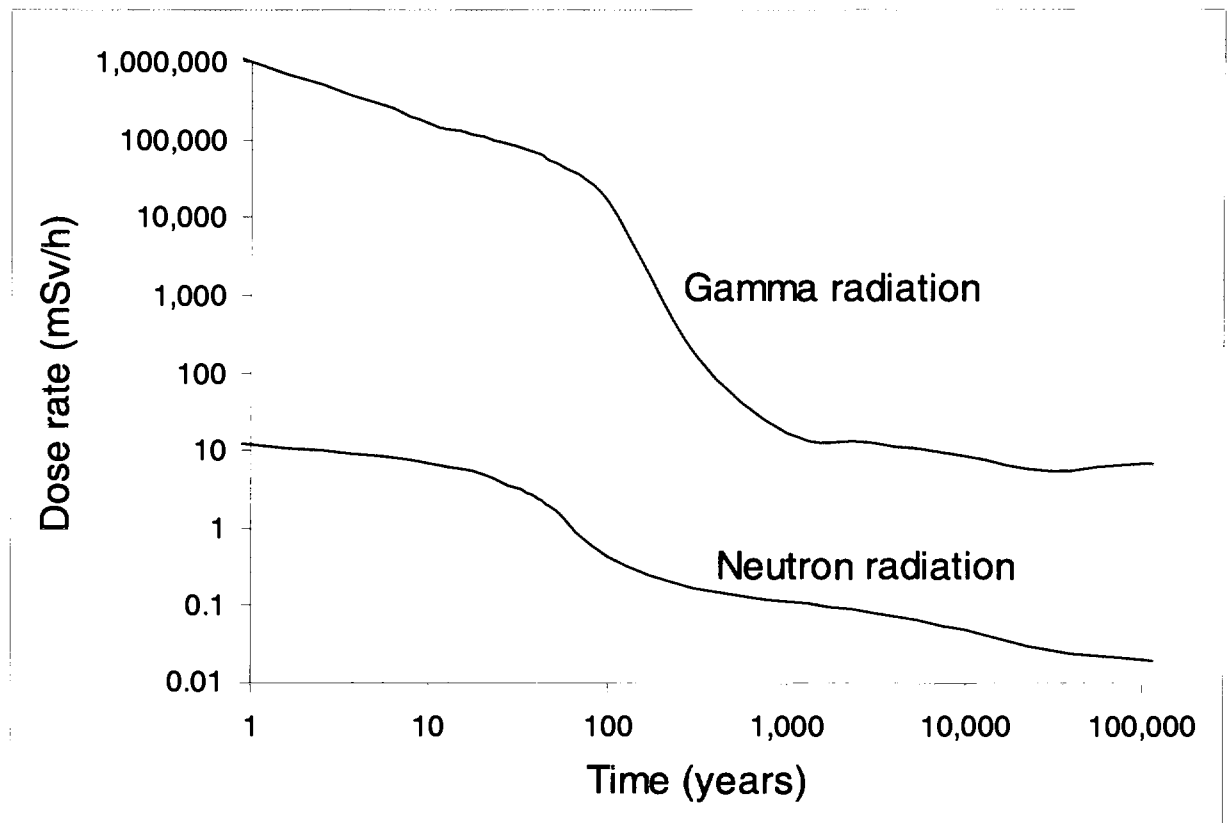


Figure 3-8b. Same data as Figure 3-8a but with logarithmic axes so that longer time periods can be illustrated.

3.5.3 Internal irradiation

How hazardous is a radioactive material if it gets inside the body?
Radiotoxicity is dependent on three factors:

- the quantity of the radioactive material
- the decay rate of the radionuclides
- the harmful effects caused in the body by the radioactive disintegrations

The third factor, harmful effects of internal irradiation, is generally expressed as the radiation dose to which a given activity, 1 becquerel, of a material, gives rise if it enters the body. Radiotoxicity is expressed for each radionuclide by a dose factor in the unit sievert/becquerel, Sv/Bq. The dose factors take into account the fact that the nuclides migrate and sometimes accumulate in different ways in the body.

The migration pathways for the material in the body and thereby its harmful effects are dependent on, for example, whether the material has been inhaled or ingested (swallowed). For this reason, two different sets of dose factors are used: one for inhalation and one for ingestion. Most radionuclides are more toxic if they are inhaled than if they are ingested.

Radiotoxicity of ingested materials

Figure 3-9 shows the radiotoxicity of spent nuclear fuel (if ingested) at different times after discharge from the reactor. The height of the bars is a measure of radiotoxicity, and the relative dominance of different nuclides at different times is also shown. The figure is divided into three parts with different scales for different time periods, since radiotoxicity declines sharply with time. All radiotoxicity measures are in percent of total radiotoxicity one month after discharge, the first bar in the figure.

Short-lived nuclides dominate radiotoxicity after short times. Because they are short-lived, these nuclides have a high level of activity, since they have to decay in less time. This means their radiotoxicity is generally also higher, since it is dependent on activity.

As short-lived isotopes disappear, their radiotoxicity will be dominated by more long-lived nuclides. They have always been present in the fuel, but have represented a smaller fraction of the total radiotoxicity during the periods when short-lived nuclides have been dominant. After 10,000 years, half of the radiotoxicity comes from the plutonium isotope Pu-239 with a half-life of 24,000 years. More than 75 percent of the original quantity of Pu-239 remains after 10,000 years. This means that the radiotoxicity caused by Pu-239 has been almost unchanged during this period, but has for short periods only comprised a fraction of the total radiotoxicity.

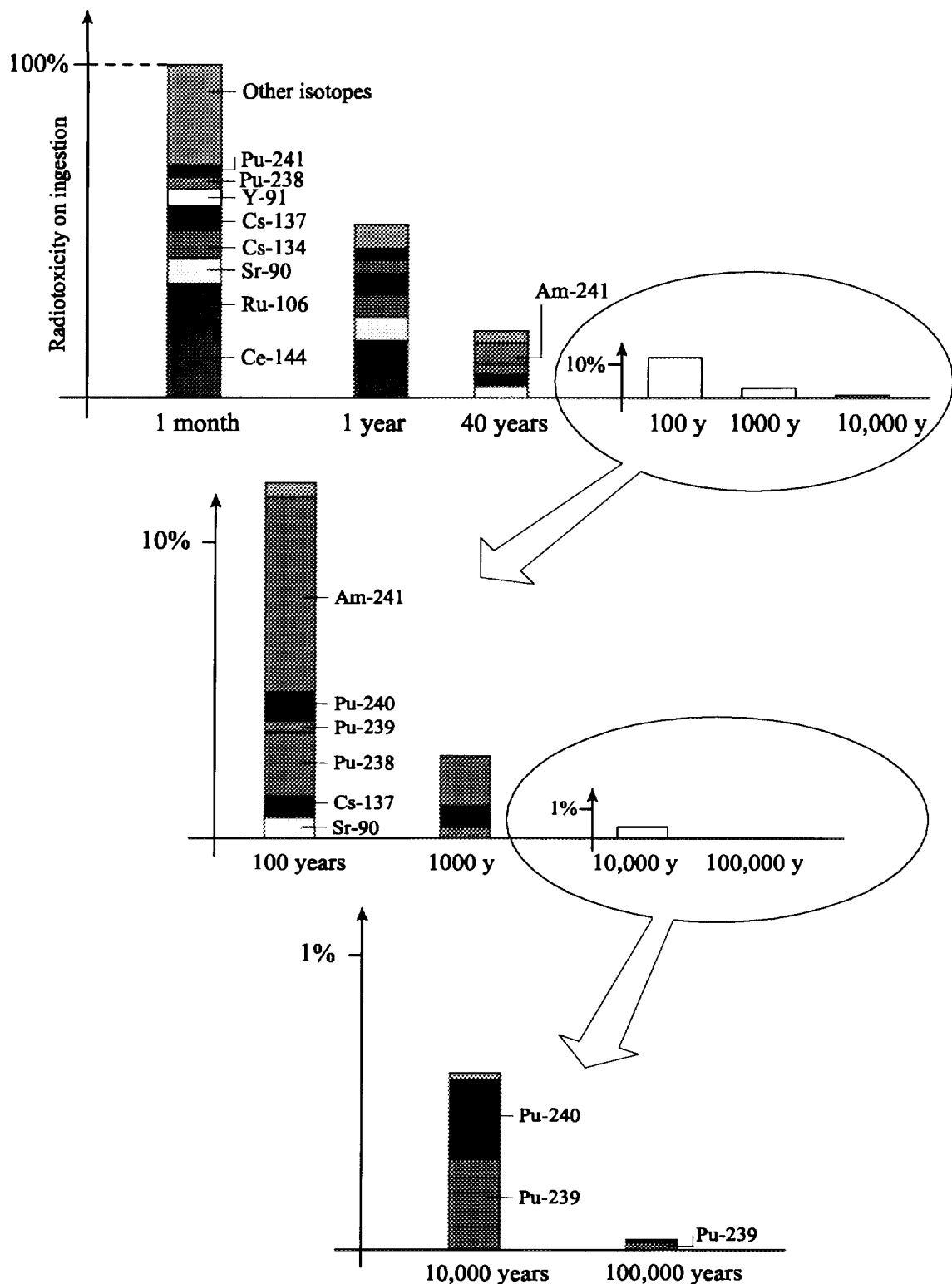


Figure 3-9. Radiotoxicity on ingestion of spent nuclear fuel of type SVEA 64 with a burnup of 38 MWd/kg U. Radiotoxicity is expressed in percent of the radiotoxicity one month after discharge from the reactor. Short-lived nuclides dominate in the beginning, when the total radiotoxicity is also greatest. After 40 years, when the fuel is to be disposed of in a deep repository, and more than 1,000 years in the future, the radiotoxicity is dominated by americium-241, formed by the decay of plutonium-241.

Radiotoxicity on inhalation

Figure 3-10 shows how radiotoxicity on inhalation develops with time. As in some previous figures, the radiotoxicity of the spent fuel is related to the radiotoxicity of the quantity of uranium that was originally mined to produce the fuel. Radiotoxicity on inhalation is dominated completely by actinides and actinide daughters.

The figure is perhaps primarily of theoretical interest. It is not reasonable to assume that radionuclides in spent nuclear fuel could become accessible via inhalation, given the way spent nuclear fuel is managed in Sweden today. In the case of deep disposal in particular, it is difficult to image any situation where this is likely to occur.

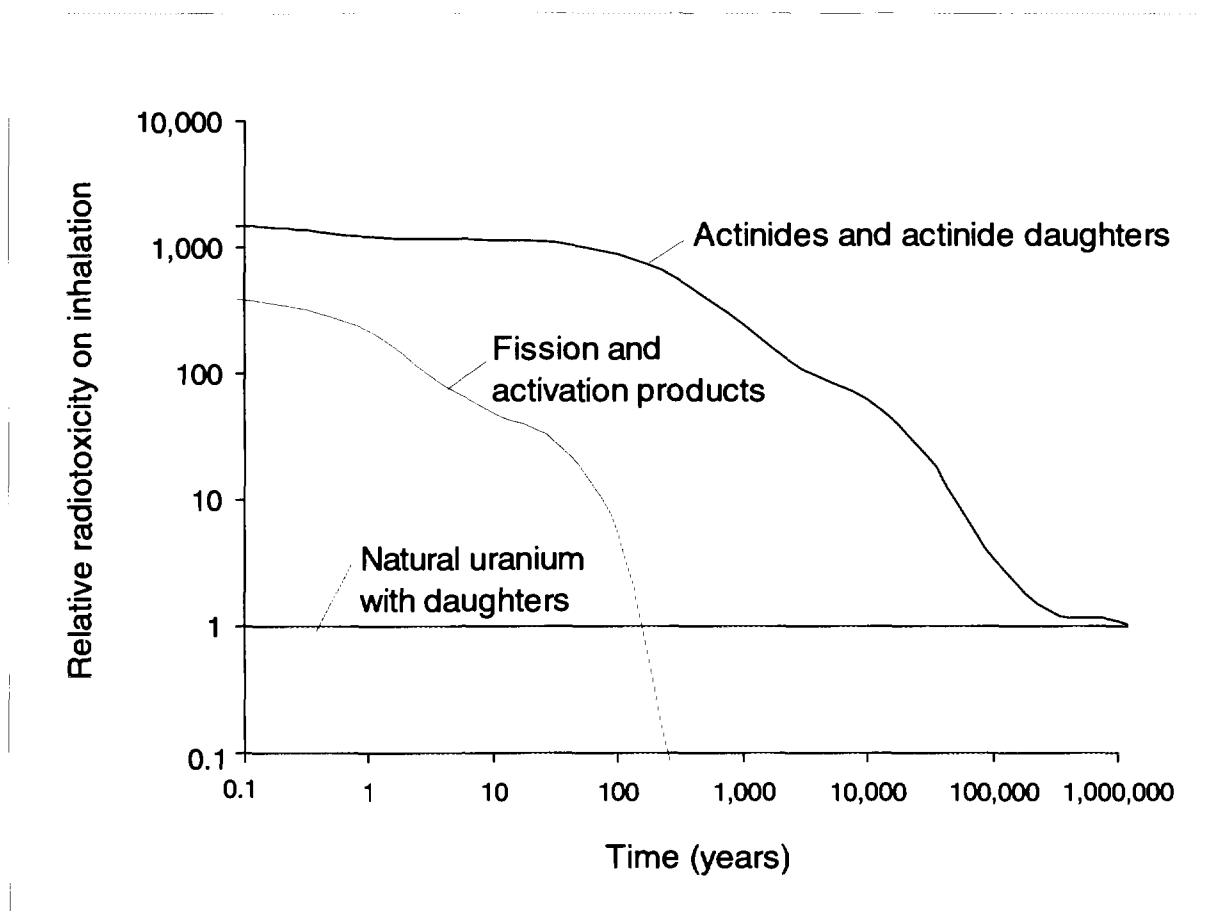


Figure 3-10. *Relative radiotoxicity on inhalation of spent nuclear fuel of type SVEA 64 with a burnup of 38 MWd/kg U. Radiotoxicity is related to the radiotoxicity of the quantity of uranium ore that was originally mined to produce the fuel, the horizontal line in the figure. Radiotoxicity on inhalation is dominated completely by actinides and actinide daughters.*

3.5.4 Comparison between mineral and the fuel cycle's fractions

The radioactivity of the quantity of fuel that is utilized to produce 1 tonne of fuel was compared with the sum of the radioactivities in all fractions in the nuclear fuel cycle in section 3.2.2. The equivalent comparison with respect to radiotoxicity is made in Figure 3-11. The radiotoxicity of the sum of the fractions in the nuclear fuel cycle has also come down to the levels of the ore after about 100,000 years.

The radiotoxicity of spent nuclear fuel is dominated completely by actinides, by plutonium and americium to be more exact, from a hundred years or so and onward (see Figure 3-9). In the discussion of risk and accessibility in the long term (Chapters 4 and 5), it is thus these elements that are of interest.

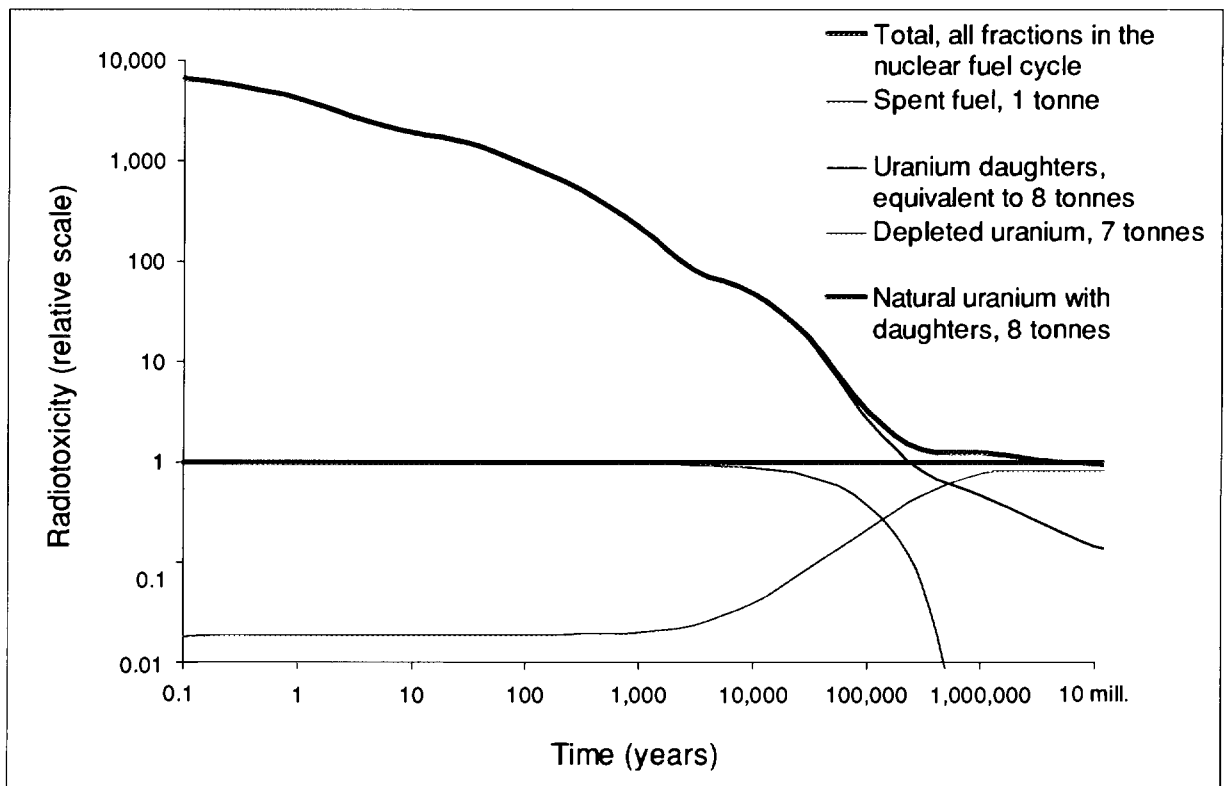


Figure 3-11. Radiotoxicity on ingestion of uranium ore (blue line), and of all fractions that arise when the same quantity of uranium mineral is used in the nuclear fuel cycle (red line). The different fractions comprise the spent fuel, the depleted uranium and the uranium daughters that are separated in the uranium mill.

3.6 COMPARISON OF FUEL DURING AND AFTER REACTOR OPERATION

This report is about spent nuclear fuel. To get a proper perspective in the discussion, it is useful to compare, for example, the heat output (decay heat) and radiotoxicity of the spent fuel with equivalent values for fuel during operation in a nuclear power plant.

3.6.1 Decay heat

The heat output of the fuel (decay heat) after different lengths of time following discharge from the reactor was shown in section 3.4. During operation, one tonne of fuel generates about 30,000,000 watts (30 megawatts) of thermal power (heat), see Figure 3-12.

This heat comes from two processes: the fission reactions in the fuel, and radioactive decay in those parts of the fuel that have already undergone fission. When the reactor is shut down, fission ceases and the heat output drops immediately to about 2 Megawatts per tonne of fuel. The heat then comes solely from radioactive decay in the spent fuel. A heat output of 2 Megawatts is comparable to the power developed by an electric locomotive.

The fuel is stored at the nuclear power plant site for about one year, after which it is stored at a central interim storage facility for around 40 years. After that the intention is for the fuel to be emplaced in a deep repository. By that time its heat output is down to just over 1300 watts/tonne. This means that the heat output at the time of disposal is around 23,000 times less than when the reactor is in operation and about 1500 times less than when the reactor has just been shut down.

It is the enormous heat output during operation which can, if it is not controlled, have disastrous consequences for the environment. This is also what happened at Chernobyl. (Swedish reactors have a design which makes such a sequence of events impossible.) In a deep repository, heat output is far below what is required to cause this type of violent outcome. As is explained in the second half of Chapter 4, it is other much slower and less dramatic processes that are important to consider when analyzing the safety of a deep repository.

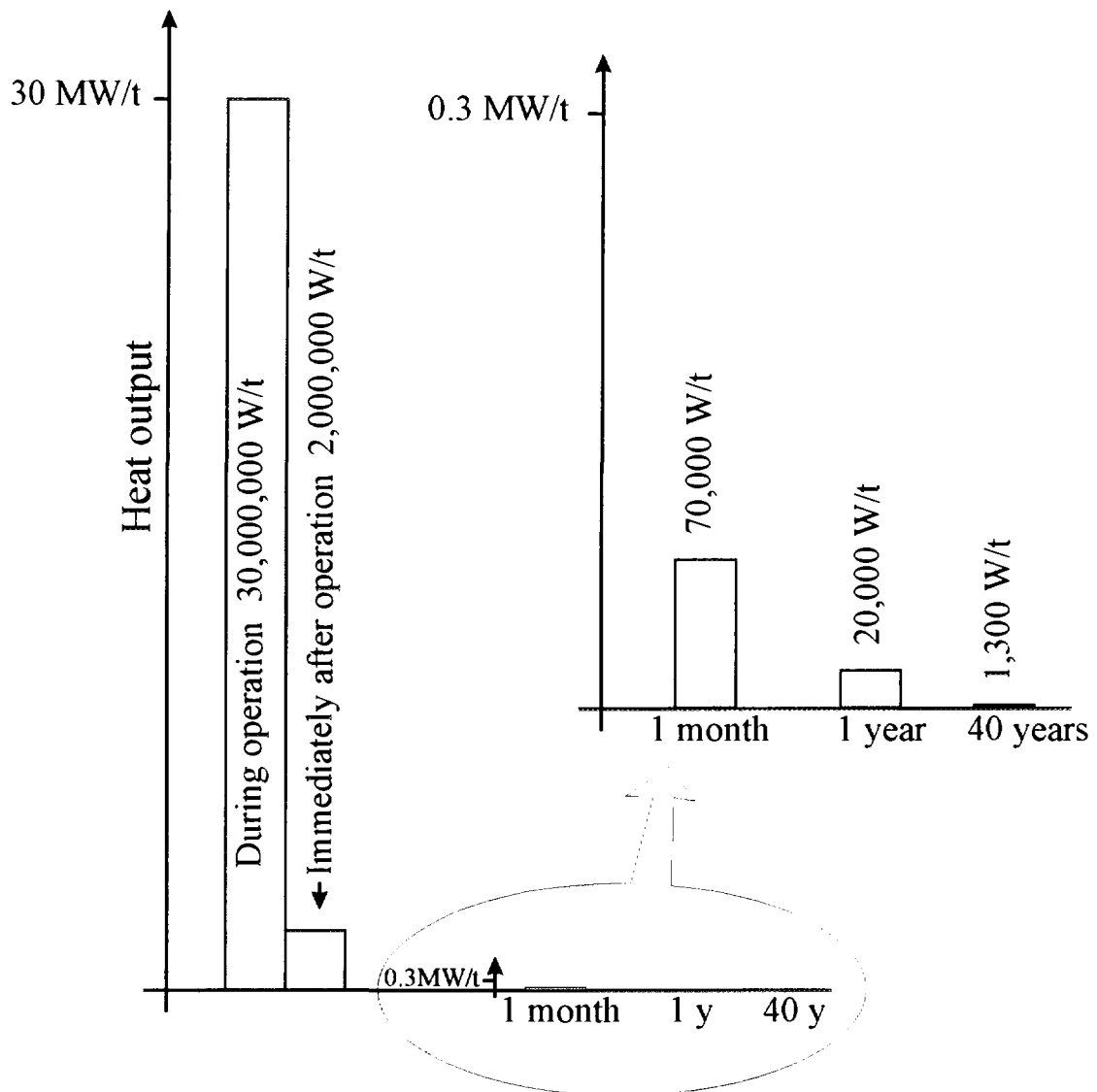


Figure 3-12. Heat generation in the fuel during operation and at different times thereafter. The heat output when the fuel is to be emplaced in the deep repository after 40 years is about 23,000 times less than the heat output in the reactor, counted per tonne of fuel.

3.6.2 Criticality

The fission in a nuclear reactor is a form of *chain reaction*: A neutron strikes a nucleus of uranium-235. The nucleus is split into two fission products and a few neutrons. These neutrons can in turn split other nuclei of uranium-235, whereby more neutrons are emitted which can split other nuclei, and so on. A prerequisite for such a chain reaction to occur and be self-sustaining is that the fuel is concentrated to a limited volume. The released neutrons must have plenty of uranium-235 nuclei “within reach” so that sufficiently many neutrons can strike uranium-235 nuclei to sustain the

chain reaction. This description is slightly simplified. Besides the concentration of uranium-235, the material in the space between the fuel assemblies is also of great importance for the occurrence of a chain reaction.

A composition of e.g. uranium-235 which is configured so that a chain reaction becomes self-sustaining in this manner is called *critical*. In a nuclear reactor, the fuel assemblies are arranged so that the reactor just barely becomes critical. In all other handling of nuclear fuel, criticality must be avoided. In slightly simplified terms, this is achieved by not concentrating the fuel assemblies too much and by the choice of suitable materials between the fuel assemblies. In a deep repository for final disposal, the spent nuclear fuel will be deposited in copper canisters each of which holds around two tonnes of fuel. The size, shape and material composition of the canisters are designed to inhibit a chain reaction, ensuring that criticality is avoided by a wide margin in the deep repository.

No matter how the spent fuel is mixed with other materials, however, the chain reaction can never be so violent that it leads to an explosion. In other words, spent nuclear fuel from Swedish reactors can never, under any circumstances, cause a nuclear explosion.

3.6.3 Radiotoxicity

How does the radiotoxicity of the fuel during and just after reactor operation compare with the radiotoxicity of the spent fuel during its subsequent handling? During operation the exact quantities of different radionuclides in the fuel are difficult to determine. Many nuclides have extremely short half-lives and disappear more or less instantaneously when the reactor is shut down. Shortly after shutdown, however, the radionuclide content and thereby radiotoxicity of the fuel can be determined with good accuracy. In Figure 3-13, radiotoxicity on ingestion ½ hour after operation is compared with radiotoxicity at later times.

Radiotoxicity is dominated by a number of short-lived nuclides, including iodine. Most of them have such short half-lives that they account for only a small fraction of the total radiotoxicity after one month, see Figure 3-13.

Which radionuclides pose the greatest risks for internal irradiation in the acute phase following an accident of the type which occurred in Chernobyl? To get an idea of the risks, one has to ask how accessible the different radionuclides become if they are dispersed in an uncontrolled fashion. There are great differences between different elements here. Iodine, for example, is very easily soluble in water and has high mobility in biological systems. In the human body, iodine accumulates in the thyroid gland. In the short term, the short-lived iodine isotopes could therefore cause high radiation doses in the thyroid if spent fuel nuclear were dispersed in an uncontrolled fashion in the environment following a nuclear power plant accident.

The only *long-term* health effect in the Chernobyl region demonstrated so far is cancer of the thyroid in children [3-7]. The cause is radioactive iodine, which is believed to have entered the body primarily via milk. The reason children have been selectively afflicted is in part due to the fact that they are more sensitive to radiation than adults, and in part because milk is a larger component in the diet of children. Irradiation took place shortly after the accident, after which it normally takes several years to develop cancer. One way to protect against radioactive iodine is to eat non-radioactive iodine in sufficient quantities to temporarily “saturate” the body with harmless iodine.

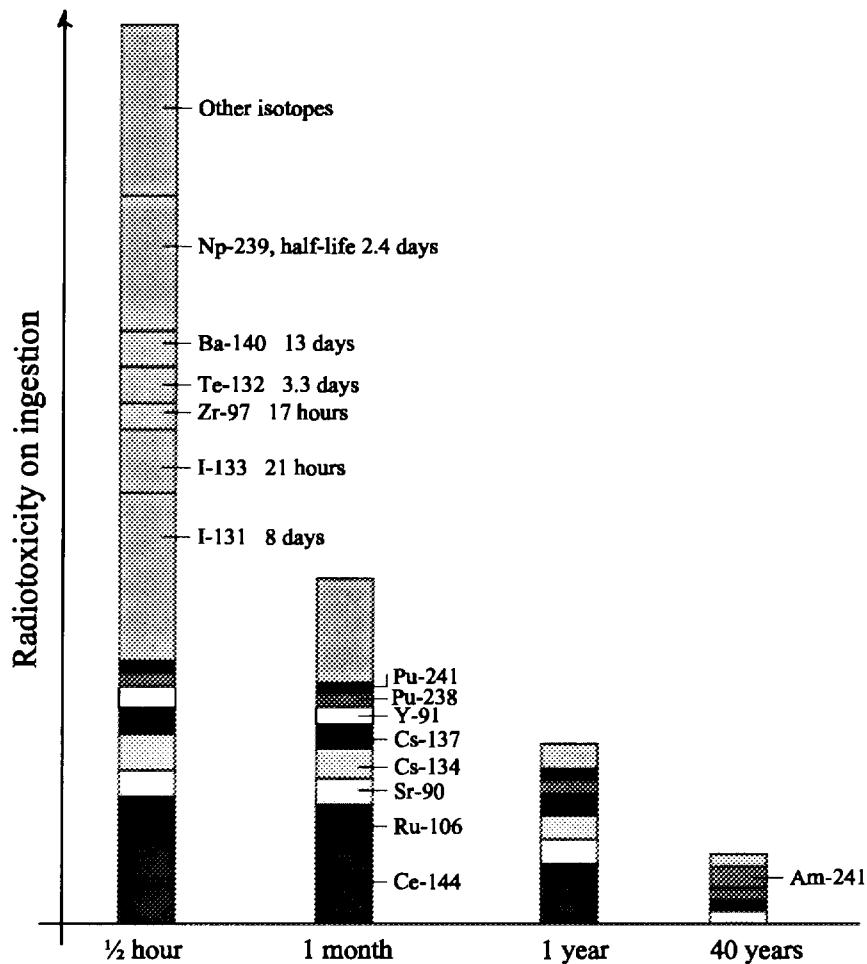


Figure 3-13. Radiotoxicity on ingestion 1/2 hour after reactor operation and at later times. Immediately after operation, radiotoxicity is dominated by a few nuclides with half-lives of up to a week or so.

Some of the personnel who worked with the extinguishing and clearing-up work following the Chernobyl accident were also exposed to high radiation doses, both internally and externally, particularly in the early phases.

Like decay heat, radiotoxicity during or just after reactor operation thus differs greatly from radiotoxicity at the planned time of disposal, about 40 years after discharge from the reactor. After 40 years, radiotoxicity via ingestion has fallen to about 7 percent of what it was immediately after discharge.

3.7 DISCUSSION

Which radionuclides are the most “hazardous” (radiotoxic) in spent nuclear fuel? There is no simple answer to this question. Different nuclides dominate during different eras. Comparisons can be made between different substances in the waste at a given point in time, as in Figure 3-9. Short-lived radionuclides dominate at the early stages. The long-lived nuclides that dominate when the short-lived ones have disappeared have a lower radiotoxicity at a given point in time, but are toxic for a longer period of time. It is perhaps less interesting to determine which nuclide or nuclides are most radiotoxic in any absolute sense. The total radiotoxicity at different times can be calculated with relatively high accuracy. This toxicity is also an indication of what protective measures must be adopted during different phases to minimize the risks.

The discussion of radiotoxicity from internal irradiation becomes somewhat academic if the question of how the radionuclides in the fuel might enter the body is not discussed simultaneously. The inherent chemical properties of the fuel and the radionuclides constitute in themselves a sharp restriction on accessibility. The fuel has very low solubility in water, something which is of paramount importance for how it might possibly be dispersed in different situations, not least during deep disposal. Furthermore, given how spent fuel is managed in Sweden today, it is difficult to imagine situations where the fuel could become gaseous or otherwise airborne and thereby accessible via inhalation. This leads us into the discussion of accessibility which is the subject of the next chapter.

4 ACCESSIBILITY

4.1 INTRODUCTION

As was explained in Chapter 2, a risk can generally be said to be made up of a component that describes hazard and another that describes exposure. This is also true of radioactive waste, where exposure is often expressed in terms of accessibility and hazard is expressed in terms of radiotoxicity. The radiotoxicity of the radioactive waste and its decrease with time are determined by the properties of the radionuclides and are very difficult to influence. The risks of radioactive waste are therefore reduced by keeping accessibility low.

Reduced accessibility is most important just after discharge from the reactor, and becomes less so as the radiotoxicity of the fuel declines. As explained in the preceding chapter, however, radiotoxicity remains high for long periods of time after discharge.

The difference between external and internal irradiation is crucial to the entire discussion of accessibility. Consider a person standing near the spent fuel without radiation shielding. This person is exposed to gamma rays and neutron bombardment in high doses for many years after the fuel has been taken out of the reactor, according to Figure 3-8 in the preceding chapter. Gamma and neutron radiation is “accessible” to this person, since it penetrates out of the fuel and into his or her body. Alpha and beta radiation, on the other hand, is not accessible in this situation, since the radionuclides must enter the body in order for these types of radiation to be toxic.

Due to their high penetrating power, gamma and neutron radiation act “remotely”, i.e. the injury can occur far from the radionuclide emitting the radiation. Alpha and beta radiation, on the other hand, act “locally”, i.e. very close to the radionuclide that is emitting them.

With this in mind, let us study accessibility and ways in which it can be limited, both during the handling of the spent fuel and during deep disposal after the repository has been closed and sealed. An overview of the Swedish system for management of spent nuclear fuel is shown in Figure 4-1 as a background to this review.

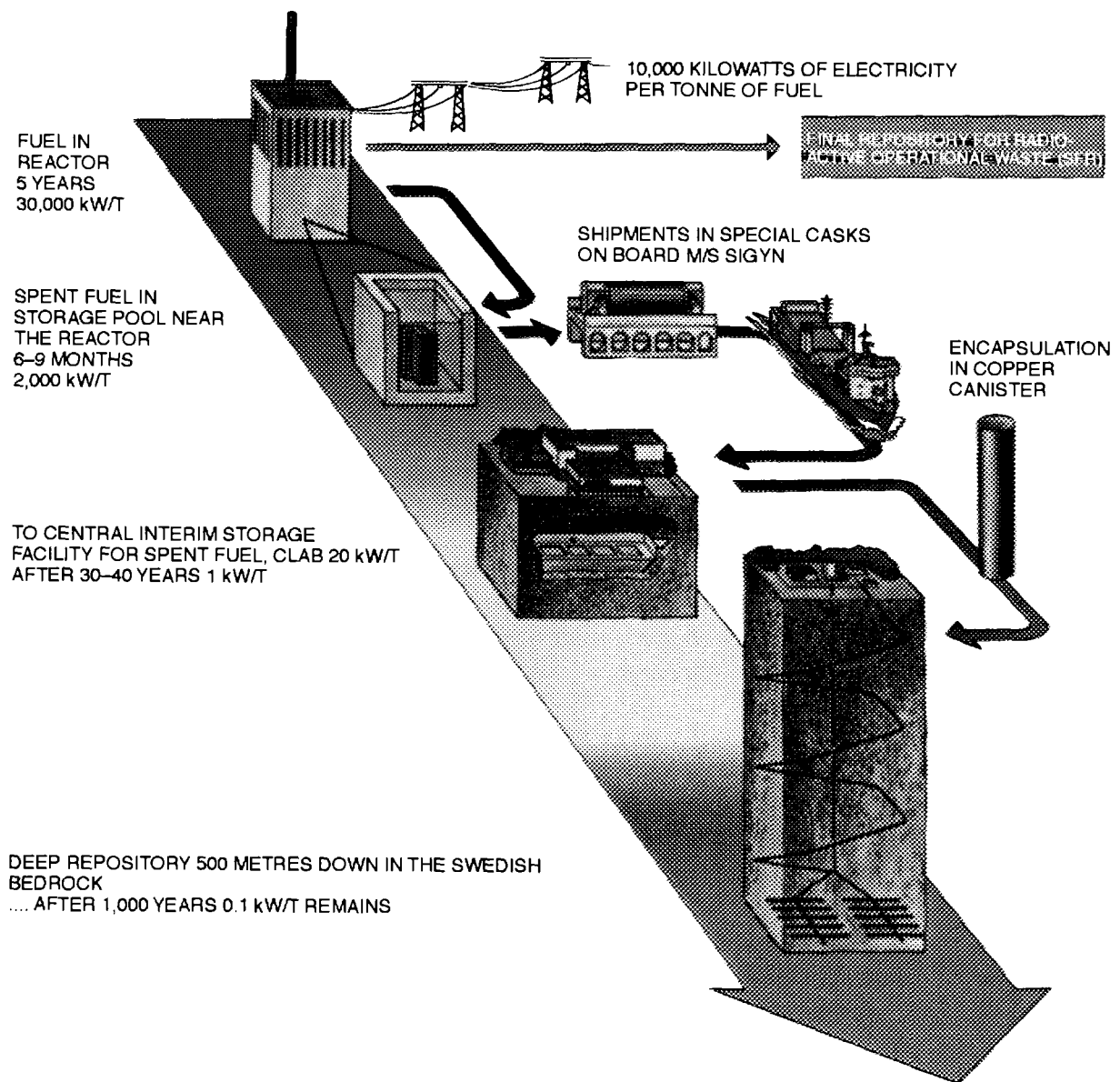


Figure 4-1. *The Swedish system for management of spent nuclear fuel. After the fuel has been discharged from the reactor it is stored at the nuclear power plant for 6 to 9 months. Then it is transported to the central interim storage facility. After about 40 years of interim storage the spent fuel will be encapsulated for final disposal.*

4.2 ACCESSIBILITY DURING HANDLING OF SPENT FUEL

4.2.1 External irradiation

Accessibility for external irradiation can be reduced by shielding off the spent fuel with a material that effectively attenuates the radiation. The denser a material is, the more effectively it protects against radiation. A centimetre-thick layer of lead, for example, attenuates gamma rays to half, i.e. half of the radiation is absorbed by the lead. Two centimetre-thick lead layers attenuate the radiation to one-fourth, and so on, see Figure 4-2. In this way, ten centimetres of lead “halves” the radiation ten times, i.e. to about one-thousandth. Water has an absorption capacity about one-tenth that of lead’s, which means about one metre of water is required to attenuate gamma rays to a thousandth.

Protection against external irradiation always consists of some suitable material that absorbs the radiation. When the fuel in a nuclear reactor has served out its useful life, the reactor is shut down and the fuel is removed from the reactor pressure vessel after a few days. It is placed in a storage pool adjacent to the reactor and stored there for six months to a year. All fuel handling takes place under water. The water in the storage pool constitutes an adequate radiation shield for the plant personnel. Accessibility is thus reduced by isolating the fuel’s radiation.

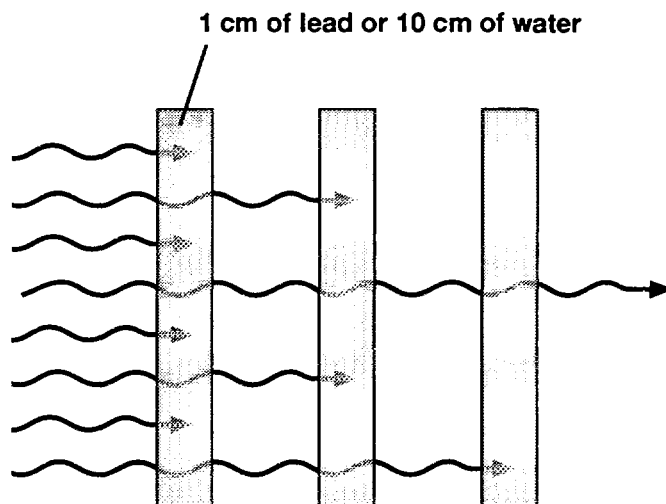


Figure 4-2. *Principle for protection against external irradiation. One cm of lead or 10 cm of water reduce the intensity of the gamma rays by half.*

During this period (as well as during reactor operation), accessibility of the fuel is also limited by careful surveillance of the premises on which the fuel is located and restriction of access to these premises. Both isolation and surveillance are methods used in many fuel handling steps.

After the initial at-plant storage of the fuel, it is transported to the central interim storage facility for spent nuclear fuel – CLAB – situated outside Oskarshamn. All Swedish nuclear power plants, as well as CLAB, are situated on the coast, and most fuel transport takes place by sea. The fuel is transported in special steel casks which protect the transport personnel against radiation in normal handling. The transport casks are also designed to withstand severe stresses in connection with more or less probable accidents and mishaps. Examples of situations the cask is designed to withstand are:

- Fire, 800°C for 30 minutes
- Fall from a height of 9 m onto a concrete slab. This is equivalent to a collision with rock wall at a speed of 50 km/h
- The water pressure at a depth of 200 m without water entering

Accessibility is thus reduced not only under normal conditions, but also if various unlikely events should occur.

In CLAB, the fuel assemblies are kept in a cooled water pool under ground. The fuel is submerged to a depth of about 8 metres, which is sufficient to absorb almost all gamma and neutron radiation. Access to CLAB is restricted and controlled in a similar manner as in the nuclear power plants, and the areas are TV-monitored.

The handling steps described thus far are the parts of the Swedish system for management of spent nuclear fuel that are already in operation. After an interim storage period of about 40 years, the intention is that the fuel will be placed in copper canisters for final disposal in a deep repository.

Encapsulation is planned to be done in a plant immediately adjacent to CLAB. Transport of the fuel assemblies between CLAB and the encapsulation plant, as well as emplacement of the fuel in the canisters, will be done by robots and be TV-monitored.

The copper canisters for deep disposal are about 5 cm thick and provided with a sturdy steel insert (inner container). The steel insert and the copper canister together provide protection against gamma rays and neutrons. The radiation level immediately outside the canister will nevertheless be so high that additional radiation protection is required during handling of the canisters. The dose rate from the gamma rays will dominate during the first 400 years or so, after which the neutron radiation will be strongest.

4.2.2 Internal irradiation

As has already been observed, to cause injury by internal irradiation the radionuclides must be released from the fuel and enter the body. The fuel consists of a solid ceramic material that is highly insoluble in water. It is encased in sealed tubes of Zircaloy, a metal alloy consisting of 98 percent zirconium. The alloy is designed to be corrosion-resistant, even under the extreme conditions prevailing during reactor operation. The Zircaloy tubes have a diameter of about one centimetre and are about four metres long.

Both the chemical form of the fuel and the Zircaloy cladding tubes protect effectively against release of radionuclides in all handling steps: during operation in the reactor, during interim storage in water and during transport. The fuel is transported in sturdy casks, which are a further obstacle to the escape of radionuclides.

The cladding tubes can be damaged during reactor operation. The condition of the tubes is therefore checked after fuel discharge. Approximately one tube out of two thousand has so far exhibited damage [4-1]. What happens to the fuel in damaged tubes during, for example, interim storage in water in CLAB? Both the fuel and most of the radionuclides embedded in the fuel matrix are highly insoluble in water. During the 40-year interim storage period, a small portion of the radionuclides in damaged cladding tubes will be dissolved in the water. This applies mainly to readily soluble elements such as caesium and iodine. The water is therefore continuously cleaned with filters that are regularly replaced and disposed of as radioactive waste. The chemical properties of the fuel and the radionuclides, such as solubility, and the importance of these properties for accessibility are dealt with more thoroughly in section 4.3.2.

To summarize, the measures that are adopted to protect against external irradiation, in combination with the chemical properties of the fuel and the radionuclides, very effectively limit the accessibility of spent fuel for internal irradiation in the various handling steps.

4.3 ACCESSIBILITY DURING DEEP DISPOSAL

The deep repository is supposed to make the waste inaccessible for man and the environment during a very long time. This is achieved by a system of barriers and other factors, Figure 4-3:

- The fuel is placed in corrosion-resistant canisters with a steel insert providing mechanical strength.
- The canisters are surrounded by a layer of bentonite clay that protects the canister mechanically from small rock movements and prevents corroding substances from entering the canister. The clay also effectively absorbs any radioactive substances that are released if the canisters should be damaged.
- The canisters with surrounding bentonite clay are emplaced at a depth of about 500 m in the bedrock, where long-term stable mechanical and chemical conditions prevail.
- The chemical properties of the fuel and the radionuclides, for example their low solubility in water, also greatly limit transport of radionuclides from the repository to the surface.

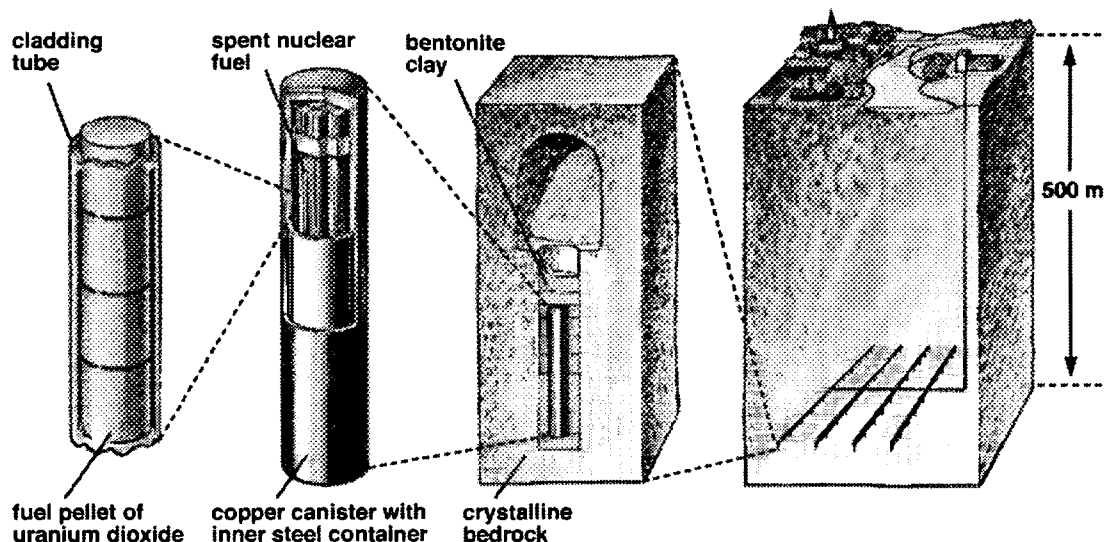


Figure 4-3. *Deep disposal of spent nuclear fuel according to the KBS-3 method.*

Together, the properties of the fuel and the barriers constitute a system that limits the accessibility of the spent fuel. The system is redundant and diversified, so that accessibility is sufficiently limited even if one barrier should be defective. This is the meaning of the *multiple barrier principle*.

The deep repository is designed to limit the accessibility of the spent fuel for such a long time that its radiotoxicity will decline to levels below the radiotoxicity of the quantity of ore that was mined to produce the fuel.

A brief description of how the repository protects against external and internal irradiation is provided in the following sections. For those who want to know more, there is a more detailed description in [4-2].

4.3.1 External irradiation

When the fuel is emplaced in the deep repository, approximately 40 years have passed since it was discharged from the reactor. During this time the radiotoxicity caused by external irradiation has declined considerably (Figure 3-8). However, protection against external irradiation is required for hundreds of years after emplacement in the deep repository. The canister, and above all the 500 metres of bedrock which separate man and the natural environment from the fuel in the deep repository, constitute a vastly oversized protection against external irradiation. But the repository is not designed with external irradiation in mind. It is instead designed to isolate and contain radionuclides that could cause injury by internal irradiation if they were to escape and reach man and the natural environment.

4.3.2 Internal irradiation

In order for radionuclides in the deep repository to cause injury by internal irradiation, they must somehow come into contact with man and the natural environment, 500 metres above the repository, and enter the body of an individual. Let us for the moment disregard the possibility of human intrusion into the repository and such extremely unlikely events as a meteorite impacting the earth in the vicinity of the repository. Let us instead ask: How might the processes that occur naturally in the crystalline basement lead to the transport of radionuclides to the surface?

In other words, what is required in order for the radionuclides to become accessible? Let us study this question briefly, barrier by barrier.

Canister

The copper canister with its steel insert is supposed to isolate the waste for long periods of time. As long as the canister is intact, it constitutes an absolute limitation on the accessibility of alpha- and beta-emitting

radionuclides. Great efforts have been made to determine what processes could violate the integrity of the canister in the repository. Under the conditions that are normally expected, chemical corrosion processes will be so slow that it would take millions of years before the copper canister has been penetrated. During this time the radiotoxicity of the fuel has decreased to levels that are comparable to that of natural uranium ore. The canister can also be subjected to mechanical stresses in a deep repository. The steel insert is designed to withstand heavy loads, but are there situations where the canister can fail due to mechanical stresses anyway? What happens, for example, in the event of an earthquake or an ice age? These questions are dealt with further in the report [4-2].

Chemical properties of the fuel and the radionuclides

As long as the canister is intact, the other barriers make no difference to accessibility. Let us therefore assume for the sake of argument that the canister is defective from the start, for example that it has a centimetre-large penetrating hole. The assumption of the hole and in particular its size is very unrealistic, but still useful in illustrating how other parts of the disposal system than the canister contribute towards limiting accessibility. A hollow canister in the repository would slowly fill with water, but let us assume here that the canister is water-filled from the start. We further disregard the Zircaloy cladding tubes surrounding the fuel and assume instead that the fuel is in direct contact with water.

Transport in water

Water plays a crucial role in transporting radionuclides away from the repository. In fact, all important transport mechanisms out of the repository involve some kind of movement of the radionuclides in the groundwater. This is equally true of transport from the fuel material out of a water-filled canister, transport through the wet clay buffer and transport with the flowing groundwater in the rock. In this context it is also of interest to know that the water flow at a depth of 500 m in the rock is so slow that a canister under typical conditions only comes into contact with a litre or so of “new” water every year.

What happens when the fuel comes into contact with water? In order for the radionuclides to move in the water, they must first dissolve in the water. Here the chemical properties of both the fuel material and the radionuclides embedded in the fuel matrix once again come into play.

Dissolution of fuel pellets

The solid fuel is a porcelain-like ceramic material composed of uranium dioxide, UO_2 . The chemical properties of the fuel material make it highly

insoluble in water. When the fuel comes into contact with water, the alpha radiation from the decay of the radionuclides will contribute to slow degradation of the fuel. Even with pessimistic assumptions regarding the rate of this process, only fractions of a tenth of a percent of the fuel will degrade each year if the fuel comes into contact with water [4-3]. The rate of degradation will then decline further as the intensity of the alpha radiation decreases with time.

Dissolution of radionuclides

Different substances have widely disparate solubilities in water. Solubility is determined by the interaction between the chemical properties of the substance and the water. Ordinary table salt, for example, has high solubility in water. Approximately 350 g of table salt can be dissolved in a container with a litre of water. If more salt is added, it sinks to the bottom of the container and remains in solid form. The water has become *saturated* with salt.

Most radionuclides in the spent fuel lie embedded in the fuel like raisins in a cake. This means they will become accessible for dissolution in water at the same slow rate the fuel is dissolved. After the radionuclides in the fuel have been exposed, they also have to dissolve in water in order to become accessible for transport. Most radionuclides also have very low solubility in water. This is particularly true of those nuclides that pose a long-term health hazard. Plutonium, for example, has a solubility of about 2 millionths of a gram per litre. The solubilities of americium and uranium are of a similar magnitude.

Several of the nuclides that dominate radiotoxicity in the short term, such as caesium and strontium, are more soluble. They are also often not as firmly embedded in the fuel as the actinides.

Table 4-1 shows the solubilities of a number of radionuclides that dominate the radiotoxicity of spent fuel during different eras. To provide an idea of what the solubilities mean from a risk viewpoint, the table also shows the amount of a solution saturated with the nuclide that must be drunk to obtain a dose of 1,000 mSv. This is the dose at which the probability of contracting fatal cancer is estimated at about five percent, see section 3.5.1.

Table 4-1. Solubilities of some radionuclides that dominate radiotoxicity during different eras. The volume of a saturated solution which, if drunk, gives a dose of 1,000 mSv of the isotope in question is also indicated. This is the dose estimated to have a 5 percent probability of causing fatal cancer.

Nuclide	Half-life (years)	Solubility in water (grams/litre)	Volume to dissolve 1000 mSv
Strontium-90	29	100	0.07 millionths of a litre
Cesium-137	30	100	0.1 millionths of a litre
Radium-226	1 600	0.000 2	5.4 decilitres
Americium-241	433	0.000 002	5 litres
Plutonium-238	88	0.000 002	1.3 litres
Plutonium-239	24 000	0.000 002	370 litres
Plutonium-240	6 700	0.000 002	100 litres
Plutonium-241	14	0.000 002	11 litres

Buffer

In our hypothetical example with an unrealistic early canister failure, where the canister has been assumed to be water-filled and the protective effect of the Zircaloy tubes around the fuel has been neglected, the chemical properties of the fuel and the radionuclides will limit the accessibility of the long-term radiotoxic nuclides in particular. The radionuclides that do dissolve must then pass through the clay buffer in order to escape from the repository. When the clay is placed in the repository it comes into contact with the groundwater and swells into a very dense material. The clay consists of impenetrable clay particles. Between the particles is a network of microscopic, water-filled pores. The water in these pores is virtually immobile. Different substances can nevertheless be transported with the water, albeit very slowly. This process is called *diffusion*.

On their way through the pore water, many radionuclides have a strong tendency to adhere to the surfaces of the clay particles. This phenomenon is called *sorption* and greatly prolongs the travel time for many of the most toxic radionuclides.

It is possible by means of simple rough calculations (Appendix 1) that take into account diffusion and sorption to arrive at approximate times for the transport of different radionuclides through the buffer. The results of such calculations are shown in Table 4-2. The calculations are based on a buffer thickness of 35 cm, which is the thickness that is planned to be used in the Swedish disposal system. The travel time can be related to the half-life of the respective radionuclide. In this way, it is possible to estimate how large a

percentage of the radionuclides that penetrated into the buffer from the canister will decay on their way out and therefore never reach the surrounding rock. As is evident from the table, this percentage is very great for many radionuclides. The results illustrate how the buffer contributes towards reducing accessibility in the deep repository.

Table 4-2. Typical travel times for different radionuclides in the buffer. The buffer is a very effective transport barrier for those radionuclides with the greatest long-term radiotoxicity.

Nuclide	Half-life (years)	Approximate travel time in buffer (years)	Percentage that decays in buffer
Strontium-90	29	2.6	6 %
Cesium-137	30	12.6	25 %
Radium-226	1600	126	6 %
Americium-241	430	190 000	100 %
Plutonium-238	88	3 100 000	100 %
Plutonium-239	24 000	3 100 000	100 %
Plutonium-240	6 600	3 100 000	100 %
Plutonium-241	14	3 100 000	100 %

Rock

Let us continue with our hypothetical example. If the canister is defective, and if portions of the fuel and its radionuclide content have dissolved, what happens with the nuclides that reach the rock without decaying during their passage through the buffer?

The repository will be built in granitic crystalline basement. Groundwater flows in the fracture systems that are always present in such rock and can transport various substances to the ground surface.

How long does it take for groundwater to move from a depth of 500 metres, where the repository is located, up to the surface? The answer depends on the configuration of the fracture system and the magnitudes and directions of the water streams. When the repository is built, the canister positions will be placed in blocks of rock with low water flows. Under typical conditions, 100 years can be assumed as a pessimistic value for water transport from such positions up to the surface. Age determinations of groundwater at these depths usually reveal that the water is thousands of years old.

The time required to transport radionuclides dissolved in the groundwater (travel time) is much longer for nearly all nuclides, however. As in the

buffer, radionuclides in the fracture systems in the rock have a strong tendency to adhere to the surfaces of the rock. Once again the phenomenon of sorption plays an important role for the transport of radionuclides in water. The nuclides can also penetrate into microscopic pores in the rock. The water in these pores is virtually immobile, and the consequence is that the nuclides are “withheld” from the flowing water. This phenomenon is called *matrix diffusion*, see Figure 4-4.

Due to sorption and matrix diffusion, the travel times in the rock for many radionuclides will be thousands of times longer than for the groundwater. This is once again particularly true of actinides such as plutonium and americium, which dominate the long-term radiotoxicity.

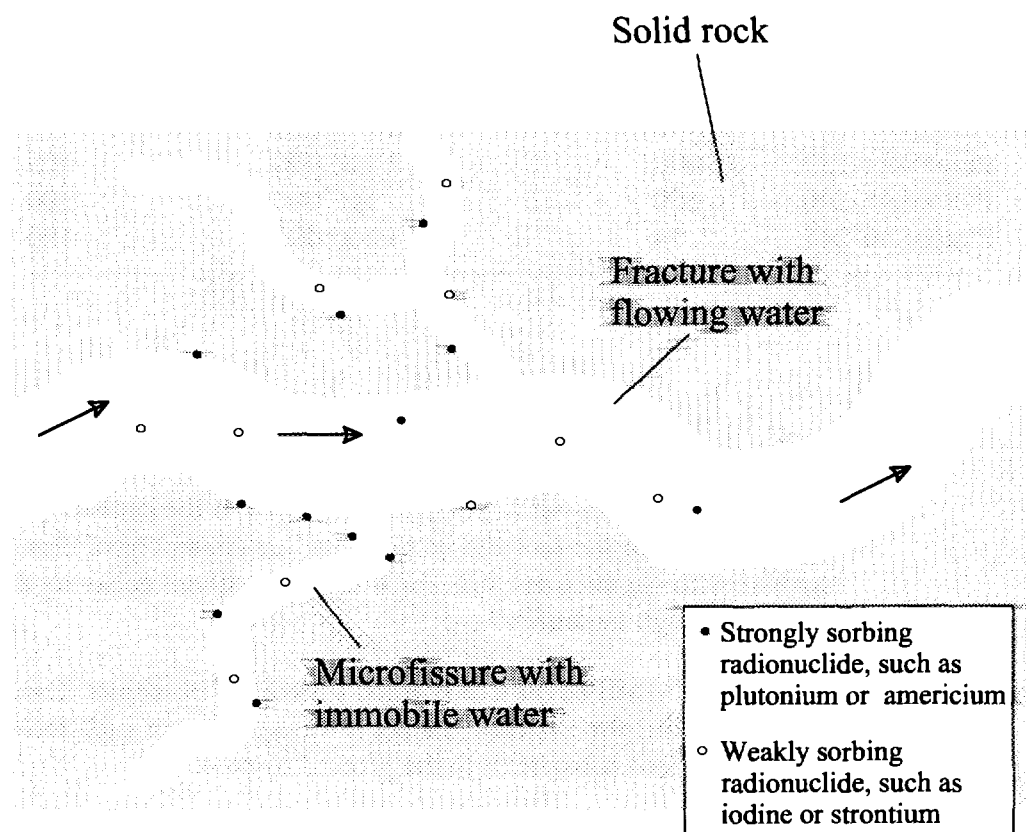


Figure 4-4. Many radionuclides are greatly retarded in the rock due to the fact that they adhere to the surfaces in the fracture systems in the rock, a mechanism known as sorption. Furthermore, all radionuclides can penetrate into microfissures in the rock and be “withheld” from the flowing water.

4.3.3 Summary

The copper canister is designed to isolate the fuel completely during such long periods of time that its radiotoxicity declines to levels below those for natural radioactive minerals.

If the canister should be damaged, the chemical properties of the fuel and the radionuclides, plus the clay buffer and the rock, will constitute very effective barriers to many of the most long-term toxic radionuclides. Long-term radiotoxic elements such as plutonium and americium are not just poorly soluble, they also have a very strong tendency to be “filtered” in the clay buffer and the fracture systems in the rock.

Elements such as strontium, caesium and iodine are readily soluble and some of them are not filtered as effectively in buffer and rock. Cesium-137 and strontium-90 account for a large portion of the radiotoxicity a hundred years or so after deposition, after which they disappear due to their relatively short half-lives. The function of the canister, which is to isolate all radionuclides for long periods of time, can thus be said to be most important in an early phase of disposal. Radiotoxicity is greatest then, and the ability of other parts of the repository to limit accessibility is best for the long-term radiotoxic nuclides.

In summary, accessibility in the deep repository is limited by a number of factors, both by engineered and natural barriers and by the inherent properties of the fuel and the radionuclides. Each barrier by itself provides a high degree of safety in the repository, which is the essence of the multiple barrier principle mentioned in the introduction to section 4.3.

In the last chapter, let us examine how radiotoxicity and accessibility can be combined to give an idea of the risk involved in the deep disposal of spent nuclear fuel.

5 THE CONCEPT OF RISK APPLIED TO THE DEEP REPOSITORY

5.1 PRODUCT OF RADIOTOXICITY AND ACCESSIBILITY

The two preceding chapters contain a relatively detailed discussion of the two components of a risk mentioned in Chapter 2: toxicity (hazard) and accessibility (exposure). Let us now tie together toxicity and accessibility in a simple way to illustrate how the concept of risk can be applied to the discussion of the long-term safety of a deep repository.

As is evident from Chapter 3, there are well-defined and widely accepted measures of the radiotoxicity of radionuclides in spent nuclear fuel. It is more difficult to find a simple and at the same time precise and accurate measure of accessibility. Let us instead make a compilation of the properties that determine the accessibility of different elements, see Table 5-1. The

Table 5-1. Important factors for the accessibility of different elements in a deep repository. The criteria for the classifications are defined in Appendix 1.

Element	Solubility in water	Mobility in buffer	Mobility in groundwater
Plutonium	Very low	Very low	Very low
Americium	Very low	Very low	Very low
Curium	Very low	Very low	Very low
Thorium	Very low	Very low	Very low
Neptunium	Very low	Very low	Very low
Uranium	Low	Very low	Very low
Radium	Low	High	Very low
Protactinium	Low	High	Very low
Strontium	High	High	Low
Caesium	High	High	Very low
Selenium	Very low	High	High
Technetium	Very low	Low	Very low
Iodine	High	High	High
Chlorine	High	High	High
Carbon	High	High	High

table includes the most important radioelements present in spent fuel during the proposed disposal in a deep repository. Solubility in water and mobility in the buffer and in the groundwater in the rock are given for each element. The criteria for the classifications in the table are defined in Appendix 1.

It is now possible with the aid of the table to subdivide the radionuclides in the spent fuel into two categories:

- Category 1 contains radionuclides with extremely low accessibility in a deep repository, those that have both very low solubility and low mobility in the buffer and the groundwater, according to Table 5-1. Radionuclides of the elements plutonium, americium, curium, thorium and neptunium are assigned to this category.
- Category 2 contains all other radionuclides in spent nuclear fuel, also those not included in Table 5-1.

The radiotoxicity of the radionuclides within each category can then be added together. The results are shown in Figure 5-1 a and b. Radiotoxicity pertains to ingestion via food and the radiotoxicity scale is the same as in Figure 3-11 in Chapter 3. The figures show that radiotoxicity is dominated by radionuclides with extremely low accessibility at the time of disposal, about 40 years after discharge from the reactor.

In terms of risk, i.e. a product of radiotoxicity and accessibility, it can be said that the risk declines very rapidly during the first couple of hundred years as radionuclides with high accessibility decay. The risk thereafter lies at a low level, since the radionuclides that dominate radiotoxicity have extremely low accessibility.

As noted above, it is difficult to find a simple and at the same time accurate measure of accessibility. The above line of reasoning can give a general idea of how the concept of risk can be applied to a deep repository. Furthermore, the reasoning is only valid *if a copper canister should be damaged*, something which in itself is very unlikely, especially during the initial period after closure of the repository. As long as the copper canister is intact, accessibility is zero.

In conclusion, let us take a brief look at what a more complete assessment of the long-term safety of a deep repository entails.

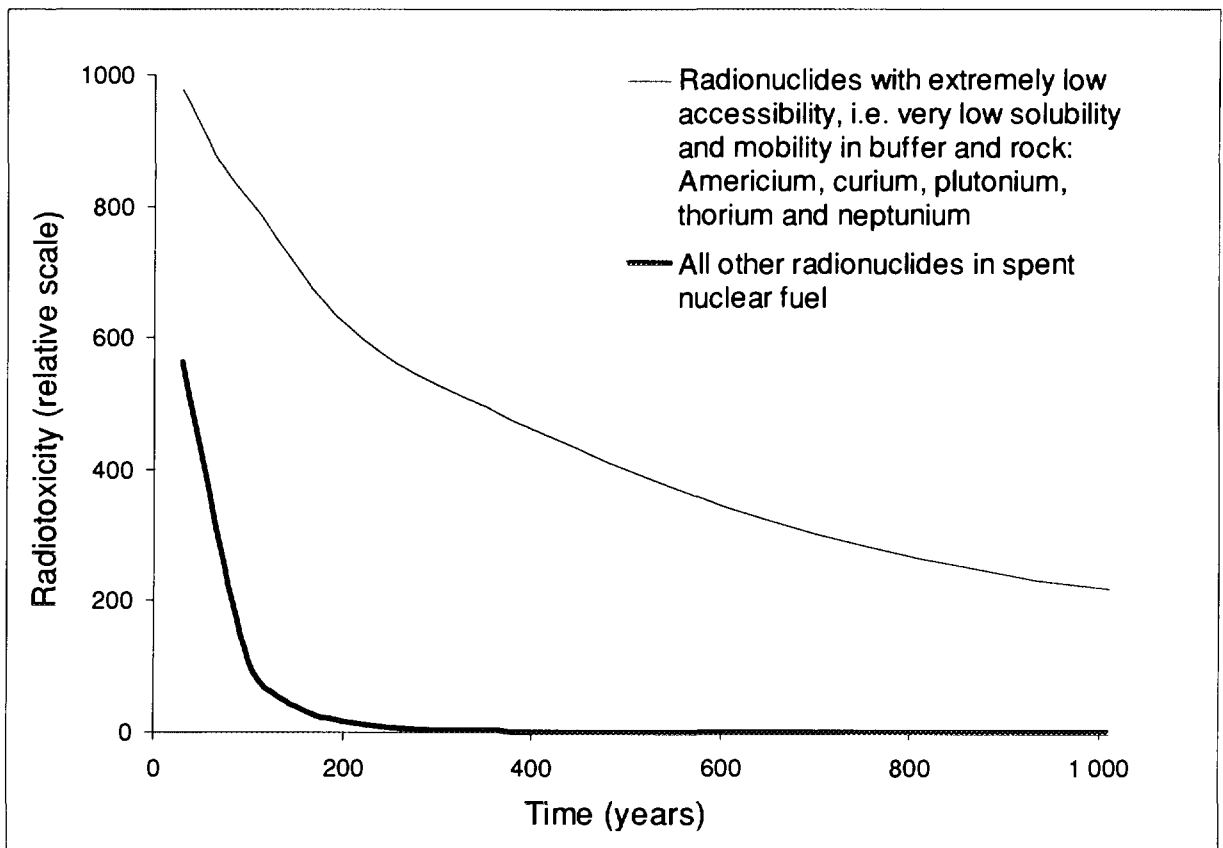


Figure 5-1a. To illustrate the concept of risk in connection with the deep disposal of spent nuclear fuel, the radionuclides can be divided into two categories: nuclides with extremely low accessibility (blue line) and others (red line). The figure shows that nuclides with low accessibility dominate radiotoxicity at the time of disposal. The blue line is heavily dominated by isotopes of plutonium and americium. The curves start at 30 years, since the spent fuel is intended to be stored for 30–40 years before being emplaced in a deep repository. The radiotoxicity scale is the same as the one used in Chapter 3, for example in Figure 3-11.

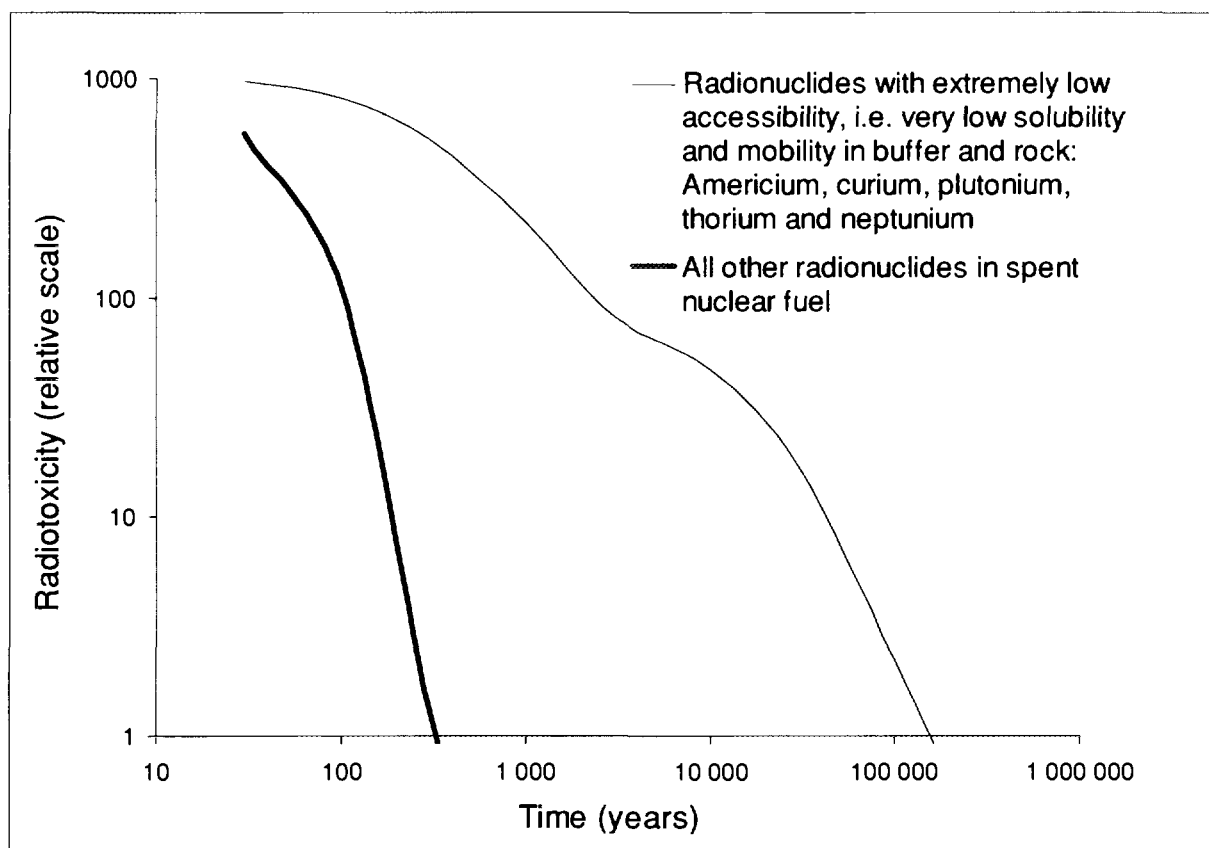


Figure 5-1b. The figure illustrates the same thing as Figure 5-1a, but with logarithmic scales to allow longer time spans to be shown.

5.2 SAFETY ASSESSMENT OF A DEEP REPOSITORY

The discussion in the preceding section is aimed at shedding light in general terms how a deep repository reduces the radiological risks caused by spent fuel. Much more careful and thorough evaluations of the safety of the repository are performed in *safety assessments*.

Safety assessments attempt to identify and analyze all the processes of change that are of importance for the long-term safety of the repository. This applies, for example, to the change in temperature in the repository as a result of the decay heat in the fuel, the corrosion of the copper canister and the long-term chemical stability of the buffer. The impact of external events such as earthquakes and ice ages is also explored, as are the consequences of various types of human intrusion in the repository.

Detailed calculations of radionuclide transport are carried out in order to demonstrate how other barriers would perform if the canister were to be damaged at different times. Calculations are also made of how radionuclides behave in the natural environment if they should escape from the repository, and what doses people in the vicinity of the repository might be exposed to.

Uncertainties exist in many of the processes dealt with in a safety assessment. These uncertainties can sometimes be considerable, and they grow larger as the time horizon considered grows longer. For this reason, the worst possible conditions within the limits of the uncertainties are often assumed in different parts of the assessment. Such pessimistic assumptions may pertain to, for example, how rapidly a process such as corrosion proceeds, or how strongly different radionuclides are sorbed in the rock. In this way, a lower limit can be assigned to the repository's ability to limit accessibility.

The results of the safety assessment are evaluated against established criteria for the safety of the repository. The assessments are reviewed by authorities and experts in Sweden and abroad. The assessments that have been carried out for the proposed Swedish system for deep disposal of spent nuclear fuel have shown that the repository provides, with ample margins, the restrictions on accessibility that are required in view of the radiotoxicity of the waste [4-4].

For those who would like to know more about how the repository performs under different conditions, there is more detailed material in [4-2].

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REFERENCES

- 3-1 Depleted Uranium from Enrichment – Valuable Energy Source or Waste for Disposal? Uranium Institute, London, 1996.
- 3-2 Calculations of Radionuclide Content and Decay Heat in Spent Fuel, SKB AR 90-17, Swedish Nuclear Fuel and Waste Management Co.
- 3-3 Bränslemängder, radionuklidinnehåll, resteffekter och typkapsel för SKB 91 (“Fuel quantities, radionuclide content, decay heats and typical canister for SKB 91”), SKB AR 90-41, Swedish Nuclear Fuel and Waste Management Co (in Swedish only).
- 3-4 Beräkning av radionuklidinnehåll, resteffekt, aktivitet samt doshastighet för utbränt kärnbränsle (“Calculation of radionuclide content, decay heat, activity and dose rate for spent nuclear fuel”), R Håkansson, Studsvik Nuclear AB, N(R)-96/079 (in Swedish only).
- 3-5 En liten faktabok om strålning (“Facts about radiation”), Swedish Radiation Protection Institute (in Swedish only).
- 3-6 1990 Recommendations of the International Commission on Radiological Protection, ICRP Publication 60; Annals of the ICRP Vol. 21 No. 1-3.
- 3-7 Bakgrund 1-96: Tio år efter Tjernobyl – vad vet vi idag om följderna? (“Background 1-96: Ten years after Chernobyl – what do we know today about the consequences?”), Kärnkraftsäkerhet och Utbildning AB, Nyköping (in Swedish only).
- 4-1 Överföring av skadade bränsleelement och enskilda stavar från kärnkraftverken till CLAB (“Transfer of damaged fuel assemblies and individual rods from the nuclear power plants to CLAB”). PM SoA 26-92, Swedish Nuclear Fuel and Waste Management Co (in Swedish only).
- 4-2 Använt kärnbränsle – Djupförvarets funktion och utveckling (“Spent nuclear fuel – Performance and development of the deep repository”). SKB report under preparation, in Swedish only.
- 4-3 SR 95 – Template for safety reports with descriptive examples. Swedish Nuclear Fuel and Waste Management Co, December 1995.

- 4-4 See e.g. a) SKI SITE-94, Deep Repository Performance Assessment Project, SKI Report 96:36, Swedish Nuclear Power Inspectorate, December 1996; b) SKI Project-90. SKI Report TR 91:23, Swedish Nuclear Power Inspectorate 1991; c) SKB 91 Final disposal of spent nuclear fuel. Importance of the bedrock for safety. Swedish Nuclear Fuel and Waste Management Co, May 1992; d) Final storage of spent nuclear fuel – KBS-3. Swedish Nuclear Fuel Supply Co/Div. KBS, May 1983.
- A-1 Annual Limits on Intake of Radionuclides by Workers Based on the 1990 Recommendations, ICRP Publication 61; Annals of the ICRP Vol. 21 No. 4.

APPENDIX 1 – CALCULATIONS

A. Chain decay

Theory

The exponential decay law applies to simple radioactive decays:

$$\frac{dN}{dt} = -kN \quad (1)$$

where

$N(t)$ is the quantity of the nuclide at time t ,

k is the decay constant, which is related to the half-life, $t_{1/2}$, by

$$k = \frac{\ln 2}{t_{1/2}}.$$

(1) has the solution

$$N(t) = N_0 e^{-kt}$$

where N_0 is the quantity at time $t = 0$.

For chain decay with n different nuclides where nuclide 1 decays to nuclide 2 which in turn decays to nuclide 3 etc., the following applies:

$$\frac{dN_i}{dt} = -k_i N_i + k_{i-1} N_{i-1} \quad (2)$$

where $N_i(t)$ and k_i are quantity and decay constant, respectively, for nuclide i , which is assumed to have an initial quantity $N_i(0)$.

By assuming the solution

$$N_i(t) = \sum_{j=1}^i a_{ij} e^{-k_j t} \quad (3)$$

i.e.

$$\frac{dN_i}{dt} = - \sum_{j=1}^i k_j a_{ij} e^{-k_j t}$$

and inserting it in (2) we get

$$-\sum_{j=1}^i k_j a_{ij} e^{-k_j t} = -k_i \sum_{j=1}^i a_{ij} e^{-k_j t} + k_{i-1} \sum_{j=1}^{i-1} a_{i-1,j} e^{-k_j t} \quad (4)$$

By identifying the coefficients in each term in (4) we get

$$a_{ij} = a_{i-1,j} \cdot \frac{k_{i-1}}{k_i - k_j} \quad \text{for } 1 \leq j < i. \quad (5)$$

By setting $t = 0$ in (3) we get

$$N_i(0) = \sum_{j=1}^i a_{ij}$$

which can be written as

$$a_{ii} = N_i(0) - \sum_{j=1}^{i-1} a_{ij} \quad (6)$$

All a_{ij} in (3) are now determined recursively by (5) and (6).

For the first nuclide in the chain,

$$a_{11} = N_1(0).$$

For nuclide 2 the following is obtained with the aid of (5):

$$a_{21} = a_{11} \frac{k_1}{k_2 - k_1} = N_1(0) \frac{k_1}{k_2 - k_1}$$

and with the use of (6)

$$a_{22} = N_2(0) - a_{21} = N_2(0) - N_1(0) \frac{k_1}{k_2 - k_1}$$

In a similar manner, the coefficients for nuclide 3 are obtained from those for nuclide 2. The recursive formulation provides good calculation economy, for example when calculation sheets are used.

Branched chains can be handled with simple modifications of the above.

Application

Chain decay has been calculated in this manner in the spreadsheet program Excel for different fractions in the nuclear fuel cycle. The initial radionuclide inventory for spent fuel has been obtained from calculations with the simulation program ORIGEN2 [3-2].

Verification

The development of the inventory with time has been calculated using the above method. The calculations have been verified by checking against results obtained with ORIGEN2 at various points in time up to 10^{10} years. The agreement is within one tenth of one percent, even for the final steps in chains with over 30 nuclides.

B. Radioactivity, decay heat and radiotoxicity

The activity of nuclide i , $a_i(t)$, is obtained simply from the quantity calculated above as $a_i(t) = k_i N(t)$.

The decay heat generated by a given nuclide is obtained by multiplying the activity by the nuclide's decay energy.

The radiotoxicity of a given nuclide is obtained by multiplying the activity by the dose factor for intake via food (ingestion) or inhalation. The dose factors, D , have been calculated by using the ALI values from [A-1] and the relationship

$$D = 0.02/ALI \text{ (Sv/Bq)}$$

where the ALI value is expressed in Bq. In cases where several ALI values are given for the same nuclide and exposure pathway, the lowest ALI value has been chosen, i.e. the one that gives the highest dose factor.

C. Retardation in the buffer

Retardation in the buffer, t_{Buff} , can be estimated with the formula

$$t_{\text{Buff}} = 0.05 L^2 (\epsilon_{\text{Buff}} + \rho_{\text{Buff}} \cdot K_{\text{dBuff}}) / D_{\text{eBuff}}$$

where

$$L = \frac{1}{2} (d_{\text{Out}} - d_{\text{In}}) / {}^{10}\log(d_{\text{Out}} / d_{\text{In}})$$

d_{Out} outside diameter of buffer 1.75 m

d_{In} inside diameter of buffer 1.05 m

ϵ_{Buff} porosity of buffer 0.25

ρ_{Buff} dry density of buffer 1600 kg/m³

D_{eBuff} effective diffusivity for the element in question, see table

K_{dBuff} K_{d} value for the element in question, see table

Table A-1. Chemical data for the most important elements in spent nuclear fuel. All data in the table are taken from [4-3].

Element	D_{eBuff} (m ² /y)	K_{dBuff} (m ³ /y)	t_{Buff} (y)	Solubility (mol/m ³)	K_{dRock} (m ³ /kg)
Am	$3.2 \cdot 10^{-3}$	3	187 000	$2 \cdot 10^{-5}$	0.2
Cm	$3.2 \cdot 10^{-3}$	3	187 000	$2 \cdot 10^{-5}$	0.2
Pu	$3.2 \cdot 10^{-3}$	50	3.1 mill.	$2 \cdot 10^{-5}$	0.2
U	$3.2 \cdot 10^{-3}$	3	187 000	$2 \cdot 10^{-4}$	2
Th	$3.2 \cdot 10^{-3}$	3	187 000	$2 \cdot 10^{-7}$	2
Ra	0.79	0.5	126	$1 \cdot 10^{-3}$	0.15
Np	$3.2 \cdot 10^{-3}$	3	187 000	$2 \cdot 10^{-6}$	2
Pa	0.79	3	756	$3 \cdot 10^{-4}$	1
C	$3.2 \cdot 10^{-3}$	0	9.7	>1	0.001
Cl	$7.9 \cdot 10^{-5}$	0	394	>1	0
Se	$3.2 \cdot 10^{-3}$	0.003	196	$1 \cdot 10^{-17}$	0.001
Sr	0.79	0.01	2.6	>1	0.015
Tc	$3.2 \cdot 10^{-3}$	0.1	6200	$2 \cdot 10^{-5}$	1
Pd	$3.2 \cdot 10^{-3}$	0.01	632	$2 \cdot 10^{-3}$	0.001
I	$7.9 \cdot 10^{-5}$	0	394	>1	0
Cs	0.79	0.05	12.6	>1	0.15

D. Characterization of accessibility

The following criteria have been used for the characterization of accessibility data that is done in Chapter 5 (data taken from Table A-1 on preceding page):

Solubility

Very low	$< 10^{-4} \text{ mol/m}^3$
Low	$10^{-4} - 10^{-2} \text{ mol/m}^3$
High	$> 10^{-2} \text{ mol/m}^3$

Mobility in buffer

Very low	$t_{\text{Buff}} > 10\,000 \text{ years}$
Low	$1,000 \text{ years} < t_{\text{Buff}} < 10,000 \text{ years}$
High	$t_{\text{Buff}} < 1,000 \text{ years}$

Mobility in rock

Very low	$K_d\text{Rock} > 0.1 \text{ m}^3/\text{kg}$
Low	$0.01 \text{ m}^3/\text{kg} < K_d\text{Rock} < 0.1 \text{ m}^3/\text{kg}$
High	$K_d\text{Rock} < 0.01 \text{ m}^3/\text{kg}$

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KBS Technical Reports 79-01 – 79-27

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The KBS Annual Report 1983

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Including Summaries of Technical Reports Issued during 1984. (Technical Reports 84-01 – 84-19)

Stockholm, June 1985

1985

TR 85-20

Annual Research and Development Report 1985

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Stockholm, May 1987

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Mark Elert

Kemakta Konsult AB

February 1997

TR 97-02

Äspö HRL – Geoscientific evaluation 1997/1. Overview of site characterization 1986–1995

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March 1997

TR 97-03

Äspö HRL – Geoscientific evaluation 1997/2. Results from pre-investigations and detailed site characterization. Summary report

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May 1997

TR 97-04

Äspö HRL – Geoscientific evaluation 1997/3. Results from pre-investigations and detailed site characterization. Comparison of predictions and observations. Geology and mechanical stability

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May 1997

TR 97-05

Äspö HRL – Geoscientific evaluation 1997/4. Results from pre-investigations and detailed site characterization. Comparison of predictions and observations. Hydrogeology, groundwater chemistry and transport of solutes

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June 1997

TR 97-06

Äspö HRL – Geoscientific evaluation 1997/5. Models based on site characterization 1986–1995

Ingvar Rhén (ed.)¹, Gunnar Gustafson²,

Roy Stanfors⁴, Peter Wikberg⁴

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May 1997

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Golder Associates Inc.

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Urban Svensson

Computer-aided Fluid Engineering AB

May 1997

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Uppsala University/Golder Associates AB

June 1997

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Forsyth Consulting

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Ji-Wei Yu, Ivars Neretnieks

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July 1997