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Project SAFE – Prestudy

Appendix A1:

Inventory

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1 INTRODUCTION

In order to calculate the long-term performance of a waste repository one has to start with a waste inventory. Therefore one can say that the waste inventory is the basis for all safety assessment.

In the licenses (SSI 1988 (b), SKI 1988 (b)) for the SFR-1 facility it is said that a renewed safety assessment should be carried out at regular intervals. In SSI 1988 (b) the interval is specified as at least each ten years. The latest safety assessment was published in 1993 (SKB 1993) which was an update of the 1987 final safety report (SKB 1987). In order to fulfil the demand in the license SKB has started a project; SAFE (Safety Assessment of Final Repository for Radioactive Operational Waste) which will carry out a renewed assessment which will be finished in year 2000.

The aim of this study is to identify the possible improvements in the waste inventory compared to former assessments. In the reviewed literature special attention has been directed towards the authorities review of the assessments.

In the SAFE project the waste inventory is defined as the amount of waste, waste matrix, engineered barriers and other construction materials which will be left in the repository at the time of closure.

In this report the inventory is divided in four parts, "general", "waste", "waste matrix" and "construction materials".

2 GENERAL

It is always possible to make waste inventory, or to use another word a source term, by making some rough estimate. The hard part is to make the uncertainties as small as possible. The aim for the inventory part of the SAFE project should be, with a reasonable cost, to make the uncertainties as small as possible.

2.1 REALISTIC VS. CONSERVATIVE INVENTORY

In reviewing the waste inventory of SFR-1 one notices there are significant differences between the inventory that has been used in the radionuclide transport calculations and what the prognosis over radionuclides, volumes and so on show. The reason is of course that in the designing of SFR-1 there was some safety margin included.

Recent work show that the margins are between prognosis and the design inventory is increasing. The reason are mainly the improvements that the nuclear power plant and other waste producers has been doing in their waste treatment and the continued low frequency of fuel failures. The use of shallow land disposal for VLLW has also meant much for the decreasing volumes.

The ambition of the SAFE project is to use two principle different cases for the radionuclide transport calculations. A conservative (pessimistic) case to use as official limiting case and a realistic case which should be used to calculate the probable future development of SFR-1. This means that two inventories must be defined.

The conservative inventory should (if possible) not differ from the one in SKB 1993 and SKB 1987. Changes can be done if the need for raised limits occur in some case or if the calculations show that some limit must be decreased in order to make the safety case. Both developments are unlikely to happen.

The realistic inventory should be based on the best knowledge at present time. Much of the work has already been done in different prognosis and annual reports to the authorities.

The approach with conservative and realistic inventories is applicable for mainly for radionuclides. For chemicals and other materials it is harder to state what a conservative inventory is.

The existing inventory has a level of detail with different inventories in the different rock vaults. In some cases this is a conservative assumption, in other cases a non-conservative assumption. Today, with the better computer technology and the new computerised database of the SFR-waste, there is a good opportunity to make a better differentiation, so the inventory can be divided on a waste type level. The ambition in the SAFE-project is to make radionuclide transport calculations where the starting point is the "waste type level".

In the end it is possible that one is not capable or does not have the need to do so detailed calculations, but the aim of the inventory should be to provide data detailed enough to make the detailed calculations possible.

2.2 WASTE VOLUMES

The volumes of waste are not interesting in it self for the long-term performance of SFR-1 as long as there are available space in the repository. The amounts or volumes of different waste types are nevertheless interesting as the foundation for calculating the amounts of radionuclides, chemicals and other materials.

The volumes of waste that is stated in SKB 1993 are mainly from a prognosis made in 1987. If one compares with the latest prognosis (Riggare 1995) one can conclude that there is room for great improvements regarding the relative amounts of different waste types

Since 1987 several changes has been made:

- Forsmark NPP nowadays uses only bitumen as matrix for stabilisation of ion-exchange resins.
- Ringhals NPP now uses a shallow land disposal at the site for very low-level waste.
- Barsebäck NPP has stopped sending their burnable low-level waste to Studsvik for incineration.
- New waste types, e.g. ashes from pyrolysis and cement solidified sludge, has been approved for disposal.
- No consideration of large scrap components was taken in SKB 1993.

In short, the inventory of waste amounts can be improved and if the SAFE project shall carry out realistic calculations it is a definite condition in order to calculate the amounts of nuclides, chemicals and other materials.

The last prognosis was made in 1995 and the next planned is to be done in 1998. In order to get the best possible estimate of future production the work should be performed as late as possible. Since the safety report should be published in 2000, it seems that the next prognosis should be postponed from 1998 to 1999. Until the new prognosis is finished the 1995 one should be sufficient for all foreseen needs in both project SAFE an in the ordinary work with SFR-1.

3

WASTE

The waste part of the inventory is quite complex. In order to give a more stringent review, this chapter is divided in three parts, "Radionuclides", "Chemicals" and other "Other materials".

3.1 RADIONUCLIDES IN SFR-1

The total allowed activity amount is 10^{16} Bq according to the licenses (SKI 1987 (b), SSI 1987 (b)). There are also limits on different nuclides, see Table 3-1.

Table 3-1. Allowed nuclide inventory in SFR-1.

Nuclide	Half-life (yr.)	Silo (Bq)	BTF (Bq)	BMA (Bq)	BLA (Bq)
^3H	12.3	$1.3 \cdot 10^{14}$	-	-	-
^{14}C	$5.7 \cdot 10^3$	$6.8 \cdot 10^{12}$	$1.3 \cdot 10^{11}$	$2.9 \cdot 10^{11}$	$2.6 \cdot 10^9$
^{55}Fe	2.7	$7.1 \cdot 10^{14}$	$1.7 \cdot 10^{13}$	$1.0 \cdot 10^{14}$	$2.3 \cdot 10^{12}$
^{59}Ni	$7.5 \cdot 10^4$	$6.8 \cdot 10^{12}$	$1.5 \cdot 10^{11}$	$1.0 \cdot 10^{12}$	$2.3 \cdot 10^{10}$
^{60}Co	5.2	$1.8 \cdot 10^{15}$	$4.0 \cdot 10^{13}$	$2.6 \cdot 10^{14}$	$5.8 \cdot 10^{12}$
^{63}Ni	100	$6.3 \cdot 10^{14}$	$1.5 \cdot 10^{13}$	$8.8 \cdot 10^{13}$	$1.9 \cdot 10^{12}$
^{90}Sr	28.8	$2.5 \cdot 10^{14}$	$2.7 \cdot 10^{12}$	$6.5 \cdot 10^{12}$	$7.1 \cdot 10^{10}$
^{94}Nb	$2.0 \cdot 10^4$	$6.8 \cdot 10^9$	$1.5 \cdot 10^8$	$1.0 \cdot 10^9$	$2.3 \cdot 10^7$
^{99}Tc	$2.1 \cdot 10^5$	$3.3 \cdot 10^{11}$	$3.6 \cdot 10^9$	$8.8 \cdot 10^9$	$1.1 \cdot 10^8$
^{106}Ru	1	$6.1 \cdot 10^{12}$	$6.2 \cdot 10^{10}$	$1.7 \cdot 10^{11}$	$2.1 \cdot 10^9$
^{129}I	$1.6 \cdot 10^7$	$1.9 \cdot 10^9$	$2.2 \cdot 10^7$	$4.7 \cdot 10^7$	$6.4 \cdot 10^5$
^{134}Cs	2.3	$8.1 \cdot 10^{14}$	$1.1 \cdot 10^{13}$	$2.2 \cdot 10^{12}$	$2.6 \cdot 10^{11}$
^{135}Cs	$3.0 \cdot 10^6$	$1.9 \cdot 10^{10}$	$2.2 \cdot 10^8$	$5.3 \cdot 10^8$	$6.4 \cdot 10^6$
^{137}Cs	30.2	$4.9 \cdot 10^{15}$	$5.3 \cdot 10^{13}$	$1.3 \cdot 10^{14}$	$1.4 \cdot 10^{12}$
^{238}Pu	87.7	$1.2 \cdot 10^{12}$	$1.7 \cdot 10^{10}$	$3.1 \cdot 10^{10}$	$4.7 \cdot 10^8$
^{239}Pu	$2.4 \cdot 10^4$	$3.8 \cdot 10^{11}$	$6.9 \cdot 10^9$	$1.2 \cdot 10^{10}$	$1.9 \cdot 10^8$
^{240}Pu	$6.6 \cdot 10^3$	$7.8 \cdot 10^{11}$	$1.1 \cdot 10^{10}$	$1.9 \cdot 10^{10}$	$2.9 \cdot 10^8$
^{241}Pu	14.4	$4.2 \cdot 10^{13}$	$5.4 \cdot 10^{11}$	$9.4 \cdot 10^{11}$	$1.5 \cdot 10^{10}$
^{241}Am	433	$1.0 \cdot 10^{12}$	$1.3 \cdot 10^{10}$	$2.4 \cdot 10^{10}$	$3.8 \cdot 10^8$
^{244}Cm	18.1	$1.2 \cdot 10^{11}$	$1.5 \cdot 10^9$	$2.8 \cdot 10^9$	$4.4 \cdot 10^8$
Total:		$9.3 \cdot 10^{15}$	$1.4 \cdot 10^{14}$	$5.9 \cdot 10^{14}$	$1.2 \cdot 10^{13}$

The latest prognosis over radionuclides content in SFR-1 was made in 1995 (Riggare 1995). The prognosis show that it is most probable that the 10^{16} Bq limit will probably never be achieved in SFR.

The existing inventory origins in the design (Thegerström 1981) and has not been changed for either SKB 1987 or SKB 1993. The reason is of course that it is a conservative inventory with considerable safety margins. There are considerable margins, according to the latest prognosis (Riggare 1995). The margins for both ^{60}Co and ^{137}Cs are factor a five. For the different plutonium nuclides the final margin is expected to be between a few up to thirty per cent. These margins are a well-known fact, this fact is mentioned in SKB 1987, SKI 1988, SSI 1988, SKI 1992, SSI 1992 and SKB 1993.

Nevertheless one should remember that the radionuclide prognosis is based on the assumption that all Swedish nuclear power plants are in use until 2010 and that the fuel failures are on a continuing low level.

As mentioned in Section 2.1 there should be two inventories. The one described above is the conservative one and should not be changed unless there is a need for it. The realistic one on the other hand must be based on some sort of prognosis of the radionuclides. It seems a good idea to do this at the same time as the prognosis of volumes in 1999.

All following suggested improvements refer to the realistic radionuclide inventory.

3.1.1 General comments

All nuclides are not equally easy to measure. Hard emitting gamma emitters like ^{60}Co and ^{137}Cs are very easy to measure, which means that it is very easy to keep track of the amounts in SFR-1. On the other hand, alpha and beta emitting nuclides can not be measured on the final waste package. Since these nuclides, especially the alpha emitters, are of great interest for the long-term performance of the repository, one has to make some indirect estimate.

One way to make this estimate is to use correlation coefficients or correlation factors, i.e. the relation between an easy to measure nuclide and the hard to measure nuclide. The coefficient can be generic or specific, for example power plant specific. Correlation factors are used for several nuclides in SFR-1. In SKB 1987 and 1993 there are general coefficients for ^{55}Fe , ^{63}Ni , ^{14}C , ^{94}Nb , ^{59}Ni , ^{106}Ru , ^{134}Cs , ^{90}Sr , ^{99}Tc , ^{135}Cs and ^{129}I towards ^{60}Co and ^{137}Cs . For transuranic nuclides (TRU) and ^{90}Sr plant specific factors are used. The generic factors are calculated in Thegerström 1981.

All correlations have a quite large uncertainty and new improved measurements and changed plant suggest that a review of the factors should

be performed. Unpublished material from the SKB project "Other waste" also shows that a review should give better correlations.

3.1.2 Specific nuclides

^{60}Co and ^{137}Cs

The nuclides ^{60}Co and ^{137}Cs are mentioned in SSI 1988, SSI 1992 and SKI 1992. These two nuclides will dominate the radionuclide inventory in the initial state and will in certain scenarios give a contribution to individual doses to the critical group. No additional investigations of the nuclides are needed at present since they are very easy to measure with gamma spectroscopy on the waste package. These nuclides are also measured by routine.

^{14}C

^{14}C is a nuclide that has been commented in SKI 1988, SSI 1988, SKI 1992 and SSI 1992. ^{14}C is a pure beta-emitter, and estimation of the activity from measurements on the waste package is therefore not possible. It is also hard to estimate ^{14}C since the correlation factor is very poor (Thierfeldt 1995). Another feature is that it is very hard to estimate the fraction of produced ^{14}C from the nuclear power plants that comes with the waste to SFR-1.

The chemical speciation is of great interest since organic and inorganic ^{14}C have very different physical and chemical properties. For example the sorption properties on concrete differ between the organic and the inorganic species.

The reviews mentioned above also point out that the ^{14}C -inventory is very uncertain but they also states that the safety margin is wide enough. At the same time the reviewers also says that SKB should try to acquire better knowledge in the subject.

The conclusion is that the ^{14}C should be reviewed, especially the questions of the fraction of ^{14}C that comes to SFR-1 and the chemical speciation.

^{59}Ni and ^{63}Ni

Nickel-59 and nickel-63 are two hard-to-measure nuclides that are mentioned in SKI 1992 and SSI 1992. Especially ^{59}Ni is interesting since it has a half-life of 75 000 years and thereby makes it significant in the long run dose-perspective.

A research program is currently working. The aim is to acquire a more sensitive measuring method by using Accelerator Mass Spectroscopy (AMS).

^{59}Ni is the limiting nuclide for metal components close to reactor core. This makes the nuclide interesting from an economical point of view. It is more expensive to deposit these scrap components in SFL than in SFR-1. It is therefore of great interest to take as much as possible of these waste components to SFR-1. A good estimate of the ^{59}Ni in the ion-exchange resins is then needed in order to find out how much of the limiting (conservative) inventory that must be set aside for ^{59}Ni in resins and to make better measurements of the near-core scrap components.

^{36}Cl

Chlorine-36 is a nuclide that has not been a nuclide of interest in the former safety assessments. ^{36}Cl is very hard to measure since it is a pure beta emitter. SKI 1988 and SSI 1988 highlights this and states that the inventory is between 1-100 GBq, based on the chlorine content in the reactor water. SSI 1988 says that the expected inventory of ^{36}Cl is quite low and should not give any significant contribution to the collective or individual doses.

Even if the expected doses from ^{36}Cl are small SKB should try to make a good estimate.

TRU and ^{90}Sr

Some other hard-to-measure nuclides are the TRU-nuclides and ^{90}Sr that are not directly measurable on the waste package. ^{90}Sr and the TRU-nuclides have great radiotoxicity.

Since these nuclides give a substantial part of the long-term release to the environment they are of great interest. A special program has been working since 1988 with the aim to increase the knowledge and to have better control of the TRU and ^{90}Sr inventory. There is nowadays a large database over TRU-nuclides and ^{90}Sr which can be used for more detailed assessment. For example there is a need to see how these nuclides are distributed between the different rock vaults in SFR and how to calculate the relations between nuclides that are hard or impossible to separate in the analysis, e.g. $^{239}\text{Pu}/^{240}\text{Pu}$ and $^{241}\text{Am}/^{238}\text{Pu}$. This work can be performed within the existing programme.

3.2 CHEMICAL SUBSTANCES

In this report "Chemical substances" are defined as chemically pure substances as, for example salts, solvents and organic substances. Also mixtures of what is usually defined as "chemical-technical" products are included in the definition, e.g. detergents and concrete admixtures.

Other substances, for example different metals, concrete, plastic and wood is defined as materials.

In general the waste in SFR-1 is well characterised and there are only small amounts of chemicals.

Some references to the safety reports (SKB 1987, SKB 1993) discuss questions about chemical substances. The main focus has been towards complexing agents, potentially environmental hazardous substances and some other chemicals that are well known to be in the waste like sulphate and borate/boric acid.

The increasing environmental awareness in the last years has influenced the whole society towards new, less environmentally hazardous, chemicals. This change of chemicals and the use of them have also affected the nuclear power plants. For example the power plants have exchanged their old detergents to new ones. These detergents often contain strong complexing agents, which if they are deposited in SFR-1 can seriously disturb the sorption properties of some radionuclides. Since these detergents and possibly some other substances can find their way into the waste there is a need to get better knowledge and SKB has therefore started a project to identify and, if the need arises, quantify chemicals in the waste.

3.2.1 Complexing agents

Strong organic complexing agents like EDTA, citric acid and oxalic acid can seriously disturb the sorption properties for many radionuclides and should therefore be avoided in SFR. (Complexing agents formed by degradation of cellulose are addressed in Section 3.6). All waste producers are aware of this fact and only minor amounts are deposited in SFR-1.

SKB should try to get better knowledge in this field since the complexing agents are used in decontamination and present in some detergents. The use of decontamination will probably increase in the future. Specifically there is a need to have a better knowledge about the amount of complexing agents that can be deposited in SFR-1 and what the consequences will be.

3.2.2 Potential environmental hazardous chemical substances

According to Scandiaconsult 1982 and Meijer 1987 the environmental impact is very small from environmentally hazardous substances. No action is foreseen except the above-mentioned investigation of chemicals in SFR waste.

3.2.3 Other Chemicals

There are chemicals in the waste that have no hazardous properties, but are still interesting due to some other property. For example enhanced deterioration of concrete by sulphate.

Concrete additives

Concrete additives are a group of chemicals of which increased knowledge is required, both regarding what kind of chemicals they contain and the effects they may have on repository performance. SKB has, on demand from SKI, started a project that aims to increase the knowledge in this field.

Borate / boric acid

Borate is discussed in SKB 1987 and SKB 1993. Borate / boric acid is used in PWR and ends up in certain waste types. Borate can already in small amounts affect the hardening of the concrete. This is of slight interest since it is neutralised with lime before solidification.

No action is foreseen in this area.

Sulphate

Sulphate is a species of interest since it can damage the concrete structure. An inventory of the amounts of sulphate was made in 1987 (Wiborgh 1987). The prognosis has changed since 1987 and therefore a simple check should be made to assure that the amounts are accurate.

3.3 OTHER MATERIALS

Two processes that can enhance the transport of radionuclides are complexation of nuclides and production of gas.

The inventory of complexing agents is discussed in 5.3.1 except for the degradation products of organic substances. The dominating contributor for complexing agents is cellulose. In an alkaline environment, cellulose will degrade under release of iso-saccarinic acid (ISA). ISA is a strong complexing agent with the strongest complexation to metal ions of the valence of three or four, e.g. different plutonium ions.

Gas is produced from three sources: corrosion of metals, microbial degradation of organic matter and radiolysis of water. The dominating source is the corrosion.

There are target values in SFR-1 on materials that can produce complexing agents and gas production, see Table 3-2 and 3-3. The gas production can, with some assumptions, be transformed into metal areas subject to corrosion, see Table 3-4. The assumptions are corrosion rates of 1 mm/year for aluminium and zinc and 3 µm/year for steel. These assumptions should be checked against the literature. The target values can be exceeded if an approval by SKI is given.

The inventory in SFR-1 is based on information in the waste type description on the average composition. Especially the packages with scrap metals and refuse is important in these estimates. Recent reports (Riggare 1997) show that these average compositions are inaccurate and a new improved average composition is presented. The uncertainties are still large in this area and a further look into this question should be beneficial.

Table 3-2. Limits of different materials in SFR-1.

Material	Silo (ton)	BMA (ton)	BTF (ton)	BLA (ton)
Ion-exchange resins	1500	1100	1600	50
Bitumen	920	1300	-	150
Cellulose	20	64	20	1000
Sludge	-	50	40	-
Other organic material	-	125	-	1000

Table 3-3. Limits on gas production in SFR-1.

Process	Silo (Nm³/yr.)	BMA (Nm³/yr.)	BTF (Nm³/yr.)	BLA (Nm³/yr.)
Corrosion	1700	4000	1500	3000
Radiolysis	50	-	-	-
Microbiologic activity	5	-	-	2000

Table 3-4. Calculated limits on gas producing materials.

Material	Silo	BMA	BTF	BLA
Steel (m ²)	1.2·10 ⁵	3.1·10 ⁵	1.2·10 ⁵	2.3·10 ⁵
Aluminium / Zinc (m ²)	450	1200	450	900

4

WASTE PACKAGE AND MATRIX

The waste packages and the matrix are well defined. In SKB 1993, SKB 1987 and in the waste type descriptions there are thorough descriptions of the waste package and types of matrix. With an accurate prognosis over different wastes there is no problem to estimate the amounts of materials the package and waste contribute with.

5 CONSTRUCTION MATERIALS

Only the construction materials that are left at time of repository closure is of interest in the safety assessment. The dominating materials that are of any concern will be the concrete constructions in the rock vaults, the shotcrete in the vaults (and possibly in the tunnels) and the backfill in the vaults and in the tunnels. The concrete additives may be of importance (see Section 3.5.3).

In Wiborgh et al 1987 there is a thorough account for the amounts of construction and barrier materials. This reference shows both a realistic inventory and also account for the inventory used for calculations. What is not in the reference is the amount of reinforcement bars. The near field assessment will show if there is a need to make an estimate of the content of iron in the reinforcement.

6 SUMMARY / CONCLUSIONS

One of the aims in the safety assessment of SFR-1 is to make radionuclide calculations to estimate the release to the environment. In order to make these calculations there is a need to be able to describe the inventory in greater detail. The improvements in computer technology and the new computerised database of waste in SFR-1 gives a good possibility to achieve this.

The aim for project SAFE is to make both conservative (pessimistic) and realistic radionuclide transport calculations. To achieve this goal there must be two inventories, a conservative and a realistic inventory. The conservative inventory is the inventory used in the design of the repository, which in most parts are identical with the limits in the licence for SFR-1. This inventory should not be changed unless there is a very strong need for it. Since it is not likely that there will be any changes in the conservative inventory, all changes discussed below regards the realistic inventory.

Volumes

Although the waste volumes are not interesting in themselves, the volumes are the foundation of the inventory. Therefore there is a great interest to have good estimates of the volumes of the different waste types. A thorough prognosis should be made in 1999, but until then the latest one from 1995 could be used in calculations.

Radionuclides

The total (actual) inventory of nuclides is calculated from the measurements of the easy-to-measure nuclides since, in principle, all hard-to-measure nuclides are calculated by correlation factors to ^{60}Co and ^{137}Cs . These factors should be reviewed since there are quite large uncertainties involved.

There are also some specific nuclides where better knowledge should be an advantage.

- ^{14}C is a nuclide that dominates the individual doses after a few hundred years and the collective dose in the inland-scenario. The amount of the nuclide is uncertain since the correlation factor is very uncertain. The chemical speciation of ^{14}C is also of interest due to different properties of organic and inorganic carbon.

- ^{36}Cl is a nuclide that is very hard to measure. Although the authorities in their reviews of the safety reports say that there probably are small doses from chlorine, the inventory should be improved.
- ^{59}Ni is a long-lived nuclide that limits the close-to-the-core metal scrap that can be taken to SFR-1. There is an ongoing research project that aims to provide a better measuring method. This should make it possible to improve the knowledge about ^{59}Ni inventory. The assumption that 90 % of the inventory is collected in the ion-exchange resins should be checked.
- TRU / ^{90}Sr are subjects to a special program. The database from this work should be used to calculate a better (more detailed) inventory. Calculations of the amount of different plutonium nuclides in SFR-1 should be done and an estimate of how TRU-nuclides are distributed in the repository vaults should also be performed.

Chemicals

Chemical substances, both in the waste, the waste package and in the construction materials are of great interest since even small amounts can seriously disturb the long term performance of SFR-1. SKB has already started a project to make a better inventory of what chemicals there are in SFR-1. There is also a special program about concrete admixtures going on. These two projects will hopefully improve the knowledge of the chemical substance inventory of SFR-1.

Other materials

Since the construction of SFR-1 and the materials used are well known there are no needs for improvement. The waste package and the matrix are also well defined in terms of which materials are present. The accuracy in the amounts of materials depends on how good the prognosis of volumes is.

Regarding knowledge of the inventory of the actual waste there is no need for action for ion-exchange resins waste. On the other hand there is room for improvement in the refuse and scrap metal inventory. A better average composition of different materials should decrease the uncertainties.

A literature review of corrosion rates of steel, aluminium and zinc in an alkaline environment should be performed.

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