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# **Characterisation of bitumenised waste in SFR 1**

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# Characterisation of bitumenised waste in SFR 1

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*Keywords:* bitumenised waste, properties, long-term stability, radionuclide migration, safety assessment.

This report concerns a study which was conducted for SKB. The conclusions and viewpoints presented in the report are those of the authors and do not necessarily coincide with those of the client.

## Abstract

The waste deposited in the Final Repository for Radioactive Operational Waste, SFR, consists in part of waste solidified in bitumen. Bitumen is considered to have favourable chemical and physical properties to act as a fixation material for radioactive waste. However, during interim storage and subsequent disposal bitumen's properties may change. This may influence the ability of the bitumen matrix to retain radionuclides. This report discusses different processes affecting the long-term performance of bitumenised waste, and an evaluation of these properties in waste deposited in SFR 1 is made. The possible effect of a bitumen barrier on the release rate of radionuclides from SFR 1 is assessed.

Based on leaching experiments reviewed in this study, it could take some thousand years, possibly more, to release all radionuclides in a 200-litre drum. The results are, however, extrapolated from experiments performed during a short period of time. Long-term deteriorating effects and the effect of a low temperature on the bitumen matrix are not very well documented. The literature focuses principally on bitumenised evaporator concentrate, but the bitumenised waste deposited in SFR 1 consists mainly of ion exchange resins. There are indications that the non-radioactive waste products usually investigated overestimate bitumen's ability to retain waste. Radiolytic effects has been estimated in this work to be negligible for waste categories F.17, F.20 and B.20 deposited in SFR 1, but for categories B.05, B.06 and F.18 the possibility of increased water uptake rate due to radiolysis can not be excluded. A more reasonable assumption is that bitumen will act as an effective barrier for radionuclide release during a time span from some hundreds to thousand of years.

Generally, the majority of the inventory of radionuclides in SFR 1 is not solidified in bitumen. By taking the bitumen barrier into account in the modelling of release of radionuclides from SFR 1, the total release rate from BMA and the Silo could be reduced at maximum by a factor of 2 and 1.5, respectively. In BLA organic  $^{14}\text{C}$  is only present in waste embedded in bitumen. Thus, taking the effect of a bitumen barrier into account can have an influence on the release rate of organic  $^{14}\text{C}$  from this repository part, but for the other nuclides in BLA the bitumen barrier is of little importance.

## Sammanfattning

Avfallet som deponeras i Slutförvaret för radioaktivt avfall, SFR, består delvis av avfall solidifierat i bitumen. Bitumen anses ha kemiska och fysikaliska egenskaper som är fördelaktiga för att innesluta radioaktivt avfall. Dessa egenskaper kan dock förändras negativt med tiden. Denna rapport diskuterar olika processer som påverkar långtids-egenskaperna hos bitumeniserat avfall. Effekten av dessa processer på det bitumeniserade avfallet i SFR 1 utvärderas. Slutligen uppskattas vilken effekt en bitumenbarriär kan ha på utsläppet av radionuklider från SFR 1.

Baserat på experimentella resultat som presenteras i litteraturen kan det ta ett par tusen år, kanske mer, innan alla radionuklider frigörs från en 200 liters tunna fylld med bitumeniserat avfall. Experimenten genomförs dock under en mycket kort tidsperiod (i bästa fall upp till några år). Försämringar i egenskaper under en mycket längre tidsperiod är mycket osäkra. Dessutom är det dåligt belagt vilken effekt en reducerad temperatur som råder på förvarsdjup har. Resultat som presenteras i litteraturen baseras ofta på bitumeniserade industrikoncentrat, men det bitumeniserade avfallet i SFR 1 består huvudsakligen av jonbytarmassor. Det finns resultat som tyder på att inaktiva avfallsprodukter som ofta studeras överskattar bitumens barriärförmåga. Radiolytiska effekter bedöms i denna rapport vara försumbara för avfallskategorierna F.17, F.20 och B.20 som deponeras i SFR 1. Däremot kan det inte uteslutas att ett ökat vattenupptag i kategorierna B.05, B.06 och F.18 erhålls på grund av sönderfall av radionuklider. En rimligare bedömning är att bitumen fungerar som en effektiv barriär för utsläpp av radionuklider under några hundra till några tusen år.

Huvuddelen av radionukliderna i SFR 1 förekommer i avfall som inte är solidifierat i bitumen. Om bitumen beaktas som en barriär i modelleringen av uttransporten av radionuklider från SFR 1 kan den totala utsläppshastigheten från BMA och Silon reduceras med en faktor 2 respektive 1,5 som mest. I BLA förekommer organiskt  $^{14}\text{C}$  endast i bitumensolidifierat avfall. Därmed kan bitumenbarriären påverka utsläppshastigheten av organiskt  $^{14}\text{C}$  från denna förvarsdel. För övriga nuklider i BLA är barriären försumbar.

# Contents

<b>Abstract .....</b>	<b>3</b>
<b>Sammanfattning.....</b>	<b>5</b>
<b>1 Introduction.....</b>	<b>9</b>
1.1 Background.....	9
1.2 Description of SFR 1 .....	9
<b>2 Description of bitumen and radioactive waste bitumenised.....</b>	<b>11</b>
2.1 Waste suitable for embedment in bitumen .....	11
2.2 Properties of bitumen .....	11
<b>3 Bitumenised waste in SFR 1 .....</b>	<b>13</b>
3.1 Amount of waste deposited in SFR 1 .....	13
3.2 Nuclide inventory in bitumen in SFR 1 .....	14
<b>4 Long-term behaviour of bitumenised waste .....</b>	<b>17</b>
4.1 Radiolytic effects.....	17
4.2 Biodegradation.....	19
4.3 Ageing .....	20
4.4 Water uptake .....	21
4.5 Leaching .....	25
<b>5 Evaluation of long-term properties of bitumenised waste at SFR 1 .....</b>	<b>31</b>
5.1 Radiolytic effects.....	31
5.2 Biodegradation.....	32
5.3 Ageing .....	32
5.4 Water uptake .....	32
5.5 Evaluation of the importance of the bitumen matrix as a barrier .....	34
5.6 Conclusions .....	37
<b>References.....</b>	<b>39</b>
<b>Appendix A</b> Estimation of absorbed energy caused by radiolytic decay	
<b>Appendix B</b> Rough estimate of the consequence of swelling of bitumenised waste in SFR 1 caused by water uptake	
<b>Appendix C</b> Estimation of transport times of radionuclides in BMA	

# 1 Introduction

## 1.1 Background

In 1988 the authorities gave SKB permission to take the SFR 1 repository in operation. SKB is enjoined to perform a safety assessment once every ten years. The last safety assessment made of the SFR 1 repository was presented in 1991 (SKB, 1991), and a new assessment is in progress.

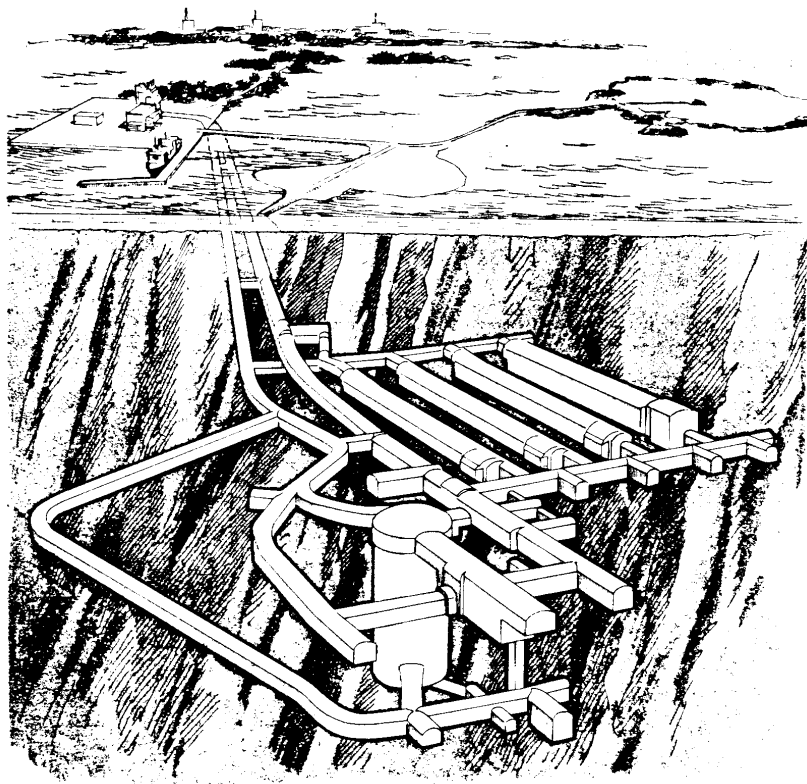
The present safety assessment will among other things contain an assessment of the consequences of radionuclides being released to the biosphere. In the previous assessment, it was assumed that the whole radionuclide inventory was instantaneously dissolved in the water within the waste packages. However, this may be an unnecessary conservative assumption for waste is embedded in bitumen. Instead, a model coupling the release of radionuclides from the bitumen matrix (waste embedded in bitumen) with water uptake of bitumen could be considered. One of the first steps in this process, which is presented in this report, is to characterise the bitumenised waste deposited in SFR 1.

After a short description of the SFR 1 repository below, follows a chapter describing the properties of bitumen. In Chapter 3 a compilation of bitumenised waste deposited in SFR 1 is given. Next, processes affecting the long-term properties of bitumenised waste are discussed, and finally an evaluation of these properties in bitumenised waste deposited in SFR 1 is made.

## 1.2 Description of SFR 1

SFR 1 is a final repository for low- and intermediate-level operational waste from the Swedish nuclear power plants, but also for small quantities of research, medical and industrial radioactive waste. The repository is located at a depth of 60 m in the bedrock in the neighbourhood of Forsmark NPP some 150 km north of Stockholm.

SFR 1 consists at present of four tunnels (BLA, BMA, 1BTF and 2BTF) and a Silo (Figure 1). Which repository part the waste is deposited in depends on physical and chemical properties as well as nuclide content and activity. The waste with the highest activity is deposited in the Silo and the lowest active waste in BLA. Bitumenised waste is deposited in the Silo, BMA and BLA.



*Figure 1 Schematic picture of the SFR 1 repository.*

## **2 Description of bitumen and bitumenised radioactive waste.**

This chapter gives a brief introduction to what type of material bitumen is and what it is used for within the nuclear industry.

### **2.1 Waste suitable for embedment in bitumen**

At present bitumen is used for solidifying low- and intermediate-level waste such as sludges, liquid concentrates and ion exchange resins. Sludges are generated when liquid waste streams are treated by chemical precipitation or flocculation in order to reduce the volume of waste contaminated with radionuclides. Examples of chemicals added to facilitate precipitation/flocculation are ferrocyanides, barium sulfate and metal hydroxides.

Bitumen is used for solidifying wet radioactive waste in several countries, e.g. Sweden, Finland, Denmark, France, Belgium, Germany, Switzerland, Lithuania, Russia and Japan. In Sweden bitumen is used for solidification of low- and intermediate-level radioactive wastes in the form of ion exchange resins and liquid concentrates at Barsebäck NPP and Forsmark NPP, but at Barsebäck the bitumen process will be finally closed down in year 2001.

### **2.2 Properties of bitumen**

Bitumen is a complex colloidal system consisting of a mixture of mainly high molecular aliphatic and aromatic hydrocarbons. Bitumen can be considered to consist of four different components; saturated hydrocarbons, cyclic hydrocarbons, resins and the dispersed particles called asphaltenes. The asphaltenes consists mainly of aromatic hydrocarbons with a molecular weight between 1,000 – 100,000 gram/mole, while the continuous phase consists of somewhat lighter molecules ( $M = 500 - 5,000$  gram/mole). The chemical composition of bitumen lies normally in the following ranges (Eschrich, 1980):

- 80 – 88 % carbon
- 8 – 11 % hydrogen
- 1 – 12 % oxygen
- 1 – 7 % sulphur
- 0 – 1.5 % nitrogen

The distribution among the different components in a bitumen varies as well. For two French bitumen grades referred to as 80/100 and 90/40 the following data are given (Table 1) (Lessart, 1993):



**Table 1 Chemical composition of two different bitumen grades (%).**

Bitumen	80/100	90/40
saturated	5.4	21.0
aromatic	72.5	53.0
resin	15.5	14.9
asphaltene	6.6	11.1

Natural deposits of bitumen have been found, but bitumen is normally manufactured by distillation of petroleum. The composition of bitumen obtained as a by-product in distillation of petroleum varies significantly depending on the origin of the petroleum and the distillation process. By blowing air through melted bitumen an oxidised, or blown, bitumen is obtained. This product has more plastic or elastic properties than the more viscous distilled, straight-run, bitumen.

Bitumen is a thermoplastic material, i.e. a solid material that at elevated temperatures becomes liquified. Physical data for the bitumen qualities used at Barsebäck NPP and Forsmark NPP are compiled in Table 2. The penetration depth may vary from 10 mm for the hardest bitumen to 110 mm for the softest (IAEA, 1993). The bitumen qualities used at Forsmark and Barsebäck is slightly below the centre of that range.

**Table 2 Physical data for bitumen qualities used at Barsebäck and Forsmark.**

Bitumen quality	IB45	IB55 <sup>a)</sup>	Mexphalt 40/50 <sup>b)</sup>
Type of bitumen	distilled	distilled	distilled
Penetration depth [mm]	35 – 50 <sup>c)</sup>	45 – 65 <sup>c)</sup>	40 – 50
Softening point [°C]	≥ 52	52 <sup>d)</sup>	52 – 60
Flash point [°C]	≥ 220 <sup>e)</sup>	≥ 220	> 250 <sup>f)</sup>
Density <sup>c)</sup> [g/cm <sup>3</sup> ]	1.03	1.02	1.01– 1.06
Water content [weight %]	0.5	≤ 0.5	not specified

a) Nynäs (1999)

b) Eschrich (1980)

c) at 25 °C

d) Roth (1996)

e) Pensky Martins

f) Cleveland Open Cup

### 3 Bitumenised waste in SFR 1

In this chapter a compilation of bitumenised waste deposited in SFR 1 is given. Both amount of waste (Section 3.1) and nuclide inventory (Section 3.2) are presented. The data given in this chapter is to a large extent based on an updated prognosis made for the waste volumes to be deposited in SFR 1 (Riggare and Johansson, 2001).

#### 3.1 Amount of waste deposited in SFR 1

The amount of bitumenised waste foreseen to be deposited in SFR 1 2030 is given in Table 3. The total volume of bitumenised waste to be deposited in SFR 1 is about 6,800 m<sup>3</sup>. The distribution of this volume between the different repository parts is approximately 500 m<sup>3</sup> in BLA, 2,900 m<sup>3</sup> in BMA and 3,300 m<sup>3</sup> in the Silo. The total volume foreseen to be deposited (including other waste categories than bitumenised waste) is about 18,000 m<sup>3</sup> (Silo), 12,000 m<sup>3</sup> (BMA) and 14,000 m<sup>3</sup> (BLA). The bitumenised waste will therefore constitute approximately 4 % of the total waste volume in BLA, 25 % in BMA and 20 % in the Silo.

**Table 3 Approximate amount of bitumenised waste deposited in SFR 1 2030 according to present prognosis (Riggare and Johansson, 2001).**

Waste category	Waste packaging	Number of packages		
		Silo	BMA	BLA
<b>B.05</b>	steel drum	–	4,200	–
<b>B.06</b>	steel drum	6,900	–	–
<b>B.20</b>	steel drums in ISO container	–	–	10
<b>F.05</b>	steel drum	–	1,700	–
<b>F.17</b>	steel container	–	900	–
<b>F.18</b>	steel mould	900	–	–
<b>F.20</b>	steel drums in ISO container	–	–	10

Reference packages have been defined specifying the content of a representative waste package (Riggare and Johansson, 2001). The waste composition of these reference packages is summarised in Table 4.

**Table 4 Waste composition [kg] of a reference package (Riggare and Johansson, 2001).**

Category	Bitumen	Ion exchange resin	Evaporator concentrate	Cellulose
F.05	95	130	–	–
F.17	820	650	120	3.6
F.18	960	600	–	–
F.20	3,420	4,680	–	–
B.05	150	50	–*	–
B.06	150	50	–*	–
B.20	5,400	1,800	–*	–

\* Present in small quantities in the waste from Barsebäck NPP.

### 3.2 Nuclide inventory in bitumen in SFR 1

The estimated radionuclide inventory embedded in bitumen in waste to be deposited in SFR 1 is compiled in Table 5. For the actinides and other nuclides within the decay chains 4N, 4N+1, 4N+2 and 4N+3, only four percent of the total activity content in the Silo is bitumenised. For the majority of the rest of the nuclides included in the inventory for the Silo between 10 and 20 % is in bitumenised waste. For a few nuclides, e.g.  $^3\text{H}$ , inorganic  $^{14}\text{C}$ ,  $^{60}\text{Co}$ ,  $^{59}\text{Ni}$  and  $^{63}\text{Ni}$ , about 33 % of the inventory is bitumenised.

In BMA, six percent of the total activity of the nuclides in the four chains is contained in bitumen. For the majority of the remaining nuclides the corresponding figure is between 25 and 50 %.

No waste located to 1BTF and 2BTF will be bitumenised.

For each nuclide but one in BLA, the activity in bitumenised waste is less than one percent of the total nuclide activity. The exception is organic  $^{14}\text{C}$ , which is only present in waste being bitumenised.

**Table 5 Estimated radionuclide inventory in bitumenised waste in SFR 1 year 2030 (Riggare and Johansson, 2001)**

<b>Nuclide</b>	<b>Silo</b>	<b>BMA</b>	<b>BLA</b>
H-3	1.9·10 <sup>11</sup>	1.3·10 <sup>10</sup>	2.7·10 <sup>6</sup>
Be-10	4.1·10 <sup>6</sup>	3.6·10 <sup>5</sup>	2.0·10 <sup>2</sup>
C-14 inorg	6.1·10 <sup>12</sup>	5.4·10 <sup>11</sup>	2.9·10 <sup>8</sup>
C-14 org	6.8·10 <sup>11</sup>	6.0·10 <sup>10</sup>	3.3·10 <sup>7</sup>
Cl-36	6.6·10 <sup>9</sup>	8.4·10 <sup>8</sup>	1.3·10 <sup>5</sup>
Fe-55	1.1·10 <sup>14</sup>	7.4·10 <sup>12</sup>	3.9·10 <sup>6</sup>
Co-60	5.2·10 <sup>14</sup>	3.3·10 <sup>13</sup>	9.2·10 <sup>8</sup>
Ni-59	6.8·10 <sup>12</sup>	6.0·10 <sup>11</sup>	3.3·10 <sup>8</sup>
Ni-63	1.1·10 <sup>15</sup>	9.8·10 <sup>13</sup>	4.8·10 <sup>10</sup>
Se-79	2.6·10 <sup>9</sup>	3.4·10 <sup>8</sup>	5.3·10 <sup>4</sup>
Sr-90	3.7·10 <sup>13</sup>	4.4·10 <sup>12</sup>	4.5·10 <sup>8</sup>
Zr-93	6.8·10 <sup>9</sup>	6.0·10 <sup>8</sup>	3.3·10 <sup>5</sup>
Nb-93m	2.5·10 <sup>12</sup>	1.8·10 <sup>11</sup>	4.8·10 <sup>7</sup>
Nb-94	6.8·10 <sup>10</sup>	6.0·10 <sup>9</sup>	3.3·10 <sup>6</sup>
Mo-93	3.4·10 <sup>10</sup>	3.0·10 <sup>9</sup>	1.6·10 <sup>6</sup>
Tc-99	3.3·10 <sup>12</sup>	4.2·10 <sup>11</sup>	6.6·10 <sup>7</sup>
Ru-106	1.3·10 <sup>9</sup>	1.5·10 <sup>8</sup>	4.3·10 <sup>-6</sup>
Pd-107	6.6·10 <sup>8</sup>	8.4·10 <sup>7</sup>	1.3·10 <sup>4</sup>
Ag-108m	3.9·10 <sup>11</sup>	3.4·10 <sup>10</sup>	1.8·10 <sup>7</sup>
Cd-113m	1.3·10 <sup>11</sup>	1.5·10 <sup>10</sup>	8.8·10 <sup>5</sup>
Sn-126	3.3·10 <sup>8</sup>	4.2·10 <sup>7</sup>	6.6·10 <sup>3</sup>
Sb-125	1.2·10 <sup>13</sup>	7.6·10 <sup>11</sup>	4.3·10 <sup>5</sup>
I-129	2.0·10 <sup>8</sup>	2.5·10 <sup>7</sup>	4.0·10 <sup>3</sup>
Cs-134	5.2·10 <sup>12</sup>	5.9·10 <sup>11</sup>	4.0·10 <sup>3</sup>
Cs-135	3.3·10 <sup>9</sup>	4.2·10 <sup>8</sup>	6.6·10 <sup>4</sup>
Cs-137	3.8·10 <sup>14</sup>	4.6·10 <sup>13</sup>	4.7·10 <sup>9</sup>
Ba-133	1.6·10 <sup>10</sup>	1.1·10 <sup>9</sup>	1.7·10 <sup>5</sup>
Pm-147	9.6·10 <sup>12</sup>	1.1·10 <sup>12</sup>	8.9·10 <sup>4</sup>
Sm-151	1.6·10 <sup>12</sup>	2.0·10 <sup>11</sup>	2.8·10 <sup>7</sup>
Eu-152	1.5·10 <sup>10</sup>	1.7·10 <sup>9</sup>	9.4·10 <sup>4</sup>
Eu-154	1.2·10 <sup>13</sup>	1.3·10 <sup>12</sup>	3.6·10 <sup>7</sup>
Eu-155	3.1·10 <sup>12</sup>	3.3·10 <sup>11</sup>	1.4·10 <sup>6</sup>
Ho-166m	2.7·10 <sup>10</sup>	2.4·10 <sup>9</sup>	1.3·10 <sup>6</sup>
Pb-210	3.6·10 <sup>-1</sup>	2.6·10 <sup>-2</sup>	1.6·10 <sup>-5</sup>
Ra-226	6.8	5.1·10 <sup>-1</sup>	4.0·10 <sup>-4</sup>
Ac-227	70	5.1	3.1·10 <sup>-3</sup>
Th-229	10	7.7·10 <sup>-1</sup>	6.0·10 <sup>-4</sup>
Th-230	3.1·10 <sup>3</sup>	2.3·10 <sup>2</sup>	1.8·10 <sup>-1</sup>
Th-232	3.4·10 <sup>-3</sup>	2.6·10 <sup>-4</sup>	2.0·10 <sup>-7</sup>
Pa-231	1.0·10 <sup>3</sup>	77	6.1·10 <sup>-2</sup>
U-232	8.3·10 <sup>5</sup>	6.1·10 <sup>4</sup>	45
U-233	6.9·10 <sup>2</sup>	51	4.0·10 <sup>-2</sup>
U-234	3.4·10 <sup>7</sup>	2.6·10 <sup>6</sup>	2.0·10 <sup>3</sup>
U-235	6.9·10 <sup>5</sup>	5.1·10 <sup>4</sup>	40
U-236	1.0·10 <sup>7</sup>	7.7·10 <sup>5</sup>	6.1·10 <sup>2</sup>
U-238	1.4·10 <sup>7</sup>	1.0·10 <sup>6</sup>	8.1·10 <sup>2</sup>
Np-237	1.4·10 <sup>7</sup>	1.0·10 <sup>6</sup>	8.1·10 <sup>2</sup>
Pu-238	1.2·10 <sup>11</sup>	8.6·10 <sup>9</sup>	6.4·10 <sup>6</sup>
Pu-239	1.1·10 <sup>10</sup>	8.6·10 <sup>8</sup>	6.7·10 <sup>5</sup>
Pu-240	2.3·10 <sup>10</sup>	1.7·10 <sup>9</sup>	1.3·10 <sup>6</sup>
Pu-241	1.3·10 <sup>12</sup>	9.0·10 <sup>10</sup>	4.6·10 <sup>7</sup>
Pu-242	1.0·10 <sup>8</sup>	7.7·10 <sup>6</sup>	6.1·10 <sup>3</sup>
Pu-244	24	1.8	1.4·10 <sup>-3</sup>
Am-241	3.3·10 <sup>10</sup>	2.5·10 <sup>9</sup>	1.9·10 <sup>6</sup>
Am-242m	3.1·10 <sup>8</sup>	2.3·10 <sup>7</sup>	1.7·10 <sup>4</sup>
Am-243	1.0·10 <sup>9</sup>	7.7·10 <sup>7</sup>	6.0·10 <sup>4</sup>
Cm-243	4.1·10 <sup>8</sup>	3.0·10 <sup>7</sup>	2.0·10 <sup>4</sup>
Cm-244	4.6·10 <sup>10</sup>	3.3·10 <sup>9</sup>	1.9·10 <sup>6</sup>
Cm-245	1.0·10 <sup>7</sup>	7.7·10 <sup>5</sup>	6.0·10 <sup>2</sup>
Cm-246	2.7·10 <sup>6</sup>	2.1·10 <sup>5</sup>	1.6·10 <sup>2</sup>

## 4 Long-term behaviour of bitumenised waste

Bitumen is considered to have favourable chemical and physical properties to act as a fixation material for radioactive waste. The diffusion of radionuclides in bitumen is insignificant and the diffusion of water vapour in bitumen is also slow. However, during interim storage and subsequent disposal bitumen's properties may change. This may influence the behaviour of the bitumen matrix, or other barriers in a repository, that has to be considered in a safety assessment. Processes normally studied are

- radiolytic effects
- biodegradation
- ageing
- water uptake
- leaching

The material presented in this chapter is a literature review giving the state of the art regarding the influence of these processes on the long-term behaviour of bitumenised waste.

### 4.1 Radiolytic effects

Radiation of the bitumen from the embedded radionuclides can cause generation of radiolysis gases. At high levels of radiation, the generated gases lead to bubble formation that causes swelling and fracturing of the bitumen matrix. Experiments on pure bitumen (Phillips *et al.*, 1984) have shown that irradiation of distilled bitumen (Mexphalt M35) lead to bubble formation and that oxidised bitumen (Mexphalt R85/40) is more prone to form pores or cracks when gamma irradiated than distilled bitumen. Crack formation in R85/40 under radiolytic swelling is confirmed by Burnay (1987).

Other effects of irradiation of the bitumen matrix is hardening of the bitumen and, in extreme cases, heating of the matrix. Irradiation of bitumenised waste products may also form organic degradation products (Valcke *et al.*, 1999). However, irradiation of bitumen may also, up to certain irradiation doses, have a positive effect on the physical properties of the bitumen. For instance, the softening point has been shown to increase (Phillips and Hitchon, 1983 and Kopajtic *et al.*, 1989).

The production of radiolytic gases depends upon the type of bitumen, the dose rate and the absorbed dose (IAEA, 1993). From experiments on external  $\gamma$ -irradiation of a non-radioactive bitumen/salt mixture, it is concluded that the hydrogen generation is proportional to the exposure dose (Duschner *et al.*, 1977). Burnay (1987) shows results where the increase in hydrogen generation is less than proportional to the dose. Gas generation is slightly higher for distilled bitumen than for oxidised (Kopajtic *et al.*, 1989). Duschner *et al.* (1977) come to the opposite conclusion, but the difference between different types of bitumen is small. The effect of temperature in the range 20 - 70°C is insignificant on the  $\alpha$ -radiolysis of bitumen (Kosiewicz, 1980).

The radiolysis gases consists of mainly hydrogen but also some carbon monoxide, carbon dioxide, nitrogen oxide and lighter hydrocarbons are formed (IAEA, 1993). Duschner *et al.* (1977), Kopajtic *et al.* (1989) and Kosiewicz, (1980) all conclude that about 95 % of the gas produced is hydrogen. Burnay (1987) also conclude that hydrogen is the main component in the formed gas, but state that at doses about 3 MGy and larger CO<sub>2</sub> production is comparable to that of H<sub>2</sub>.

Swelling due to irradiation of bitumen also depends on bitumen type, dose rate, absorbed dose, but also on size and surface-to-volume ratio of bitumenised product and waste loading (Phillips *et al.*, 1984). In Phillips *et al.* (1984) it is concluded that for the same total dose, swelling of Mexphalt M35 (distilled bitumen) is about 50 % greater than for Mexphalt R85/40 (oxidised bitumen).  $\gamma$ -irradiation of crushed samples of distilled bitumen (Mexphalt 80/100) and of oxidised bitumen (Mexphalt R90/40) to a total dose of 5 MGy cause intense swelling in both types of bitumen (Kopajtic *et al.*, 1989).

Radiolysis due to  $\alpha$ -radiation can be two to ten times more efficient than  $\gamma$ -radiation (Burnay, 1987). Measurements on small scale cans reveal that at the same dose rate (90 Gy/h)  $\alpha$ -loaded samples show significant swelling, but for  $\gamma$ -loaded samples no swelling is observed. While swelling is important for distilled bitumen when gamma doses exceed 2 MGy, gamma doses up to 200 MGy does not cause any increase in volume of oxidised bitumen (IAEA, 1993). The content of  $\alpha$ -emitting radionuclides in bitumenised waste is mostly limited ( $< 0.2 \text{ TBq/m}^3$ ) and is therefore of limited importance regarding swelling (IAEA, 1993).

A theoretical model has been developed (Burnay, 1987) to predict the swelling of full scale drums with bitumenised waste (concentrate containing a high proportion of NaNO<sub>3</sub> in oxidised bitumen). According to the model, for waste loadings of  $\alpha$ -emitters less than  $37 \text{ TBq/m}^3$ , swelling in both 200 and 500 litre drums was less than 1 %. Valcke *et al.* (1999) refer to studies where a swelling of 200 litre drums (Mexphalt 80/100 (distilled) and about 40 % precipitation sludge) of between 2 and 33 % during interim storage and that the swelling stopped after one to three years are reported. For a product consisting of 60 % Mexphalt R85/40 (oxidised) and 40 % evaporator concentrate, on the other hand, no significant swelling during interim storage has been observed. In IAEA (1993) it is stated that an absorbed dose of 10 MGy gives a radiolytic swelling of some 5 to 10 % in a 200 litres drum.

In another model it is assumed that for the same total dose similar swelling is obtained in canisters of different size provided that the product of dose rate and the height of the waste container raised to power two ( $I \cdot L^2$ ) is constant (Phillips *et al.*, 1984). The model correlated reasonably well with their experimental data. A more complicated model taking formation and growth of bubbles and gas diffusion in bitumen into account, has been shown to give a better agreement with experimental data (Phillips *et al.*, 1984). A much smaller swelling due to radiation was obtained for large samples (22 litres and 180 litres) of bitumenised waste than for small samples (up to 4 litres) irradiated under the same  $I \cdot L^2$  conditions (Johnson *et al.*, 1988). The “bubble model” was able to simulate the swelling of large samples better than the  $I \cdot L^2$  model.

Organic degradation products are formed by irradiation of bitumenised waste products. According to Valcke *et al.* (1999), the main degradation product formed under aerobic conditions is oxalate, but also formate and (bi)carbonate. According to Kagawa *et al.* (1999) the main component of radiolytic degradation is oxalic acid. Kagawa and co-workers conclude that the total organic carbon (TOC) concentration increases with increasing absorbed dose. An absorbed dose of 1 and 10 MGy results in a TOC concentration of 5 and 10 mg C/dm<sup>3</sup>, respectively. The effect of  $\alpha$ -radiolytic degradation of bitumen on plutonium solubility under conditions representative for those of a cement backfilled repository vault has been investigated (Greenfield *et al.*, 1997). It is concluded that it is unlikely that agents forming complexes with plutonium is formed. Savage *et al.* (2000) conclude that organic products formed by degradation of bitumen is insignificant in comparison to those formed by degradation of cellulose.

Properties related to the combustion behaviour of bitumen are the flash point and the self ignition point. Mexphalt R85/40 samples with a total  $\alpha$  content of 72 kBq/g and total  $\beta$  content of 5 MBq/g showed a 4 % increase in flash point and a 5 % decrease in self ignition points after six to seven years (Wilding *et al.*, 1991).

Based on experimental results the following judgement has been made of the effect of radiolysis (Eschrich, 1980). For absorbed doses of less than 0.1 MGy negligible effects on gas generation, swelling, hardening, heating and decrease in leach resistance are found. The amount of generated gases for absorbed doses between 0.1 and 2 MGy must be considered in packaging the bitumenised product, for instance by assuring that the gas can escape from the packaging and that there is enough volume for swelling. The changes in leachability and mechanical properties of the product is insignificant. For even higher absorbed doses, 2 – 10 MGy, a substantial swelling can take place and most likely also an increased leachability.

## 4.2 Biodegradation

Even though it has been concluded that a repository environment (low temperature, high salinity, high alkalinity, radiation) is all but ideal for promoting growth of micro-organisms (Buckley *et al.*, 1985a), there are micro-organisms which are well adapted to this particular environment. Chemicals required for micro-organisms growth origin from (Lessart, 1993):

- the waste (organic carbon and mineral compounds, e.g. NO<sub>3</sub><sup>-</sup>, SO<sub>4</sub><sup>2-</sup>)
- the embedding matrix (mineral compounds in cement, carbon in bitumen)
- the water (mineral nutrients).

Brodersen *et al.* (1991) state that some types of sludge particles seem to be toxic to micro-organisms.

Micro-organisms present in a repository can affect the waste for instance by production of gases (H<sub>2</sub>, CH<sub>4</sub>, CO<sub>2</sub> and H<sub>2</sub>S) and chemicals (e.g. organic liquids) (Barletta *et al.*, 1987). The rate of biological degradation of bitumen depends on factors such as type of microbes, environmental conditions and chemical composition of bitumen matrix.

As stated earlier in this report, bitumen can be considered to consist of four different components; saturated hydrocarbons, aromatic hydrocarbons, resins and asphaltenes. Different micro-organisms attack different components, why the components degrade at different rates. Under aerobic conditions it is the saturated fraction of bitumen that is degraded by micro-organisms under production of CO<sub>2</sub> (Lessart, 1993). For anaerobic conditions resins and asphaltenes are more or less unaffected by micro-organisms. The difference in CO<sub>2</sub> production between Mexphalt R90/40 and Mexphalt 80/100 caused by biodegradation reflects the lower content of easily degradable saturated hydrocarbons in the latter (Brodersen *et al.*, 1991).

A significant growth of microbes can be obtained on surfaces of bitumen exposed to flowing water (Pedersen, 2001). Microbial degradation of bitumen is a slow process under anaerobic condition, but is much faster under aerobic conditions. Swiss studies (Wolf, 1989) gave that bacterial degradation under aerobic conditions generated 15 – 40 l CO<sub>2</sub>/m<sup>2</sup>, year at normal temperature and pressure and that the degradation rate of bitumen was 20 – 50 g/m<sup>2</sup>, year. Under anaerobic conditions, the degradation rate was two orders of magnitude lower. This corresponds to a total gas production of between 30 and 80 m<sup>3</sup>/drum per 1,000 years (at Standard Temperature and Pressure) under aerobic conditions (Valcke *et al.*, 1999). Another investigation (Roffey *et al.*, 1987) reports a CO<sub>2</sub> production rate of 0.6 – 1.5 mmole CO<sub>2</sub>/g bitumen, month for aerobic conditions and of 1.1 – 1.5 mmole CO<sub>2</sub>/g bitumen, month for anaerobic conditions. Assuming a bitumen degradation rate of 50 g/m<sup>2</sup>, year the data reported by Roffey and coworkers corresponds to a CO<sub>2</sub> production rate of up to 20 l/m<sup>2</sup>, year under aerobic conditions. The data given for anaerobic conditions indicates a two orders of magnitude lower production rate.

Experiments on degradation rates for both pure bitumen and bitumen/sodium nitrate mixtures (Abdellah and Pedersen, 1984) have shown that under optimum conditions for microbial growth a thickness of 3 µm of non-radioactive test samples is affected after one year. In another study a value of 25 µm was reported for pure bitumen after three years (Jones, 1965).

Brodersen *et al.* (1991) state that with the present knowledge about biodegradation of bitumenised waste, biodegradation seems to be of minor importance for the long-term safety of bitumenised waste.

### 4.3 Ageing

It is known that the viscosity of bitumen increases with time why the material becomes harder. A possible explanation to this is that a redistribution of the molecules in the larger hydrocarbons take place or that more volatile hydrocarbons evaporates (NKA, 1985). In experiments performed on pure bitumen, bitumen with ion exchange resins and bitumen with sodium nitrate, a fourfold increase in viscosity after 3.5 years of storage was found (Brodersen and Nilsson, 1991). The increase in viscosity was approximately proportional to the square root of time.

Another process that makes bitumen harder is oxidising when in contact with oxygen. The oxidising reaction is much faster in the presence of light, but takes places also in darkness. The maximum depth to which the oxygen penetrates is estimated to be in the



range of 2.5 – 5 mm (Eschrich, 1980). The oxidising process accelerates with increased temperature and pressure, but there are no appreciable differences between different types of bitumen. The rate of hardening decreases in the course of time.

A bitumenised matrix consisting of 40 % ion exchange resin was leached for about two years, and the leaching rate and the effective diffusivity of  $^{60}\text{Co}$  and  $^{137}\text{Cs}$  was estimated (Perić *et al.*, 1992). After about 10 months it was found that the effective diffusivity of  $^{137}\text{Cs}$  and  $^{60}\text{Co}$  increased by two and three orders of magnitude, respectively. This effect was attributed to ageing.

Leaching of pure bitumen can also form organic degradation products. Kagawa *et al.* (1999) compare chemical and radiolytic degradation of pure bitumen. They come to the conclusion that the dominant degradation of bitumen is oxidation (chemical degradation by leaching under aerobic conditions), which leads to the formation of organic degradation products. After a leaching period of three years a total organic carbon (TOC) concentration of 0.6 – 1.2 g C/dm<sup>3</sup> was obtained in aerobic conditions. Leaching in anaerobic conditions resulted in a TOC concentration of less than 6 mg C/dm<sup>3</sup> after 50 days and not increasing. They analyse the content of carboxylic acids and come to the conclusion that formic acid and acetic acid are dominating. After leaching under aerobic conditions for a period of three years, the concentration of the two acids is about 0.5 – 1.5 g/dm<sup>3</sup> and 0.35 – 0.5 g/dm<sup>3</sup>, respectively. Formic acid, acetic acid and oxalic acid were also detected when bitumenised sludge were leached in deionised water as well as in mortar pore water (Yamaguchi *et al.*, 1999). They obtain a higher TOC concentration in mortar pore water than in deionised water, but they concluded that the concentration is very low.

#### 4.4 Water uptake

In spite of that bitumen is a hydrophobic material, water can be transported into the bitumen matrix. This process is usually described as diffusion of water vapour. Water uptake does not only take place in water saturated systems but also in humid air. In pure bitumen the water uptake is negligible. A diffusion coefficient for water in pure bitumen of  $2 \cdot 10^{-12}$  m/s at 20 °C is reported in NKA (1985). However, since the waste being bitumenised often is hygroscopic (e.g. salts and ion exchange resins), there is a driving force for water vapour in the bitumen matrix. The waste is dispersed in the form of particles in a continuous phase of bitumen. Water vapour can diffuse through the surrounding layer of bitumen to the waste particles. When the waste particles absorb water they begin to swell.

Table 6 gives the relative volume increase between dry cation resin and resin being saturated with water. Data is also given for two sodium-based salts. This gives an indication of a possible volume increase that can be obtained for the waste being bitumenised due to water uptake. However, swelling is very much dependent on the type of resin (cation or anion, powdered or bead resin, degree of cross-linking (i.e. DVB content) etc), the loading of the resin and pre-treatment of the resin. Residual water in the bitumenised resin prevents observable swelling (IAEA, 1993). Thus, a possible swelling will be closely related to the used waste process. It is normally not possible to estimate the swelling without making tests on the actual resin in the specific process it will be used. The results obtained in the different studies quoted below should be

considered as indications of possible swelling and not to be taken as actual facts about what will happen with a specific bitumenised waste matrix.

**Table 6 Relative volume of waste particles.**

Substance	Dry	Saturated solution
ion exchange resin <sup>a)</sup>	1	2.4
NaNO <sub>3</sub>	1	3.5
Na <sub>2</sub> SO <sub>4</sub>	1	14.5

*a) Based on cation exchange resin IR 120 on sodium form.*

The swelling of the particles could have several consequences:

- internal stresses will be generated within the matrix.
- the bitumen matrix may increase in size.
- the distance between the particles and thus the thickness of the bitumen layer between them could decrease.

When enough water has been taken up, an open communicating porosity will be generated. This can be caused in two ways:

- internal stresses cause cracks and fissures
- and possibly the waste particles increase in size until they touch each other

The process of water uptake depends mainly on the amount of waste in the matrix, the waste composition and the type of bitumen. Other factors affecting water uptake is the repository conditions for instance temperature, material surrounding the matrix and available volume for swelling. Fractures and open pores lead to an increased transport of water into the matrix. Literature data on swelling is not consistent, which can be attributed to differences in experimental set-up, waste composition and type of bitumen. The amount of water uptake in bitumenised resins has in general been found to be proportional to the square root of time (NKA, 1985). However, this may not be true for high waste loadings. The water uptake rate is an order of magnitude faster for irradiated bitumenised resins than for non-irradiated resins (Aittola and Kleveland, 1982). An increase in water uptake rate was also found when the resins contain decontamination chemicals.

An ion exchange resin can swell as much as 200 % (NKA, 1985). The ionic state of the resin affects the swelling ability, H<sup>+</sup> and OH<sup>-</sup> results in the largest swelling. Resins are therefore treated with sodium sulfate to reduce the swelling ability. Thermal treatment of an anionic resin reduces its swelling ability, while this is not the case for a cationic resin.

Brodersen *et al.* (1998) report on experiments performed at Risø with bitumenised waste products from the Risø waste processing plant. Waste containing 23 % easily soluble salts (Na<sub>2</sub>SO<sub>4</sub> and NaCl) resulted in a swelling after 224 days corresponding to

6 – 8 mm/year. Recalculated to a 216 litres drum with swelling only on the upper surface, this is equivalent to a 1 % increase in total volume. The swelling was still increasing at the end of the experiment why it is difficult to extrapolate the results from this work for longer time periods. The samples were in these experiments stored in water. When the samples were stored in high-humidity air, the swelling was reduced to 2 mm/year. The swelling is reduced somewhat when the samples also contained 4 % ion exchange resin. This may be due to the fact that salt is bound to the resin. Comparing with other experimental results obtained on “synthetic mixtures” made at the laboratory at Risø, these swelling rates are high. A more common swelling rate seems to be 0.1 – 0.5 mm/year. There are indications that the rate of swelling might increase in the presence of precipitated sludge material (BaSO<sub>4</sub>), especially when hydrophilic components (e.g. potassium soap) is present. This may possibly explain why a higher swelling is obtained for waste from the waste processing plant than for “synthetic mixtures”. Other explanations could be that the salt crystals in waste products from the waste processing plant are especially small. It may also be of importance that the waste consists of sodium sulfate and chloride instead of sodium nitrate.

The results discussed in the previous paragraph are for unconfined swelling of bitumen. When the bitumenised waste is confined in for example a drum, swelling is prevented and an osmotic pressure is generated. Confined swelling has been investigated at Risø and the results are presented in Brodersen *et al.* (1998). For soft bitumen the swelling rate is about the same for both confined and unconfined swelling. For hard bitumen the swelling rate is higher for confined than for unconfined swelling, and confined swelling of hard bitumen is similar to that of confined swelling of soft bitumen. The increase in water uptake in the presence of BaSO<sub>4</sub> indicated in the unconfined experiments discussed above results, in a confined environment, in a higher rate of pressure build-up than for similar materials without BaSO<sub>4</sub>. The rate of swelling of real bitumenised waste is high initially, but later it decreases to about 10 % of that obtained in unrestricted swelling.

Results of unconfined swelling of a mixture of bitumen, salt (mainly NaNO<sub>3</sub>) and sludge particles are also presented in Brodersen *et al.* (1991). Non-radioactive samples (0.75 ml) swell about 10 – 15 % in one year, while active samples of the same size swell to about twice the original volume. Decreasing the surface-to-volume ratio of non-radioactive samples decreases the swelling expressed as volume %, also shown by Valcke *et al.* (1999). Extrapolation of the results indicates that the swelling should be about 0.4 % after one year for a full scale drum. The authors stress that a higher swelling is expected for active products. Swelling and leaching experiments on full-scale drums with moderately active evaporator concentrate supports this. The swelling was about 15 % by volume after 455 days. Differences in swelling rate between radioactive and non-radioactive waste products were not found by Brodersen *et al.* (1998). The difference in comparison to the results presented by Brodersen *et al.* (1991) is attributed to the larger test samples, 20 cm<sup>3</sup>, used by Brodersen *et al.* (1998).

The swelling of Mexphalt R85/40 mixed with evaporator concentrate and reprocessing sludge (Eurobitum R85/40) was found to depend on temperature (Sneyers and van Iseghem, 1998). The volume increase at 40°C was three to seven times higher than at 23°C. Leachant composition (pH 7.0 – 7.5 or 12.5) had no influence on swelling rate

The effect of temperature and of waste-to-bitumen ratio on water uptake and swelling of cation exchange resin in bitumen (Mexphalt 40/50) immersed in water for up to one month has been studied (Brodén and Wingefors, 1992). For a matrix containing 40 weight % resin, almost no water uptake or swelling was observed at temperatures between 3 and 30°C. At higher resin concentrations, the water uptake increases with increasing resin concentration, but with no significant difference between a temperature of 20 and 30°C. However, at 3°C the water uptake was considerably higher. A possible explanation is cracking of the matrix at low temperatures. This is of importance since the temperature at repository depth is well below room temperature. The authors note that contradictory results are obtained in another study (Buckley, 1985b). It is not clear if different bitumen qualities are used in the two studies. Brodersen *et al.* (1998) conclude that swelling rate is higher at 40°C than at 23°C. Brodén and Wingefors (1992) conclude that the swelling rate obtained for waste loadings around 60 % or higher is so fast that it can not be controlled by diffusion.

The first steps in modelling the diffusion of water in a heterogeneous mixture of bitumen and ion exchange resins is reported in Häggblom and Wingefors (1987). Their results of some test calculations for a drum filled a bitumen matrix containing 30 % dry resins indicate that more than 10 % swelling is obtained in a layer of about 4 cm after 200 years. The authors state that the diffusivity of water in bitumen and resin has a significant influence on the results, and that a better knowledge of the diffusivities is needed before quantitative conclusions can be drawn from their calculations.

Aittola and Kleveland (1982) have compiled data from experiments in which bitumenised ion exchange resins have been exposed to water in order to study swelling and leaching of radionuclides. Different types of bitumen and resins have been analysed. The actual swelling of samples of bitumenised resins varies from a few percent to more than 100 % of the theoretical swelling of pure ion exchange resins. The highest swelling is obtained with the highest mixing ratio (55 volume %). The authors conclude that:

- type of bitumen and ion exchange resins has no noticeable effect on swelling
- for mixing ratio ion exchange resin and bitumen < 50 % by volume there is a slight increase in swelling with increasing amount of resin. When the ratio exceeds 50 % there is a strong effect on swelling
- addition of decontamination chemicals such as EDTA, citric acid, oxalic acid and ascorbic acid increases the swelling somewhat

The results also indicate that the sample size has no effect on swelling but that irradiation of the resin increases the swelling significantly. The data presented by Aittola and Kleveland (1982) in general corresponds to swelling rates from tenths of a millimetre per year to a few millimetres per year.

Significant variations in swelling of oxidised bitumen samples between triplicate samples have been found (Brodersen *et al.*, 1998). These differences may be due to variations in hardness of the bitumenised waste product. Furthermore, the degree of fracturing may vary between different samples.

## 4.5 Leaching

The term “leaching” is mostly used to refer to the release of radioactive components embedded in the bitumen matrix. However, other substances (e.g. salts) are simultaneously leached from the matrix. Leached ions and radionuclides may be kept dissolved or may sorb, precipitate or form complexes.

The diffusivity of radionuclides in undisturbed bitumen is negligible. Thus, a network of pores or fractures in the matrix is a necessity for release of nuclides. As discussed in the previous sections there are several mechanisms by which such an open porosity can be formed. Once a communicating porosity is established, the radionuclides dissolve and are released from the bitumen matrix by diffusion. Highly soluble radionuclides in evaporator concentrate can be assumed to be released at the same rate as highly soluble salts. Sparingly soluble radionuclides and nuclides in sparingly soluble salts can remain in the pore system for a long time. In a simplified model two different cases can be defined:

- the release of highly soluble radionuclides is governed by the rate by which open pores are formed.
- the release of sparingly soluble radionuclides and nuclides in sparingly soluble salt is governed by the nuclide dissolution rate.

The release rate is dependent upon type of bitumen, type of waste, waste loading, concentration of nuclides in solution and also all factors influencing the diffusivity of radionuclides (e.g. temperature). Complex formation may increase the release rate. On the contrary, sorption of radionuclides on bitumen or insoluble salts will reduce it. The leachability of a radionuclide can be characterised by its leach rate. The lower the leach rate the more difficult is it to release the nuclide from the matrix. Typical release rates are compiled in Table 7.

**Table 7 Approximate leach rates for bitumenised waste (IAEA, 1993).**

<b>Element</b>	<b>Leach rate [g/cm<sup>2</sup>, day]</b>
actinides and rare earth metals	$10^{-8} - 10^{-5}$
reprocessing waste nuclides	$10^{-6} - 10^{-5}$
alkali and alkaline earth metals	$5 \cdot 10^{-6} - 5 \cdot 10^{-3}$

Considerable efforts have been made to evaluate the release rate of radionuclides from a bitumen matrix. The experiments are most often leaching test on the laboratory scale. Data presented are based on experiments of a relatively short duration (up to a few years). However, the conditions determining the leaching rate may not be constant in time. For example, open pores may close, external conditions (e.g. temperature, pressure, water flow rate, water chemistry) may change and physical ageing of the bitumen matrix will result in a progressive hardening of the matrix. One therefore has to be cautious in applying the results in a safety assessment that extends over a much longer period of time. Extrapolation of results obtained from experiments on small-scale

samples to full-scale drums is also problematical due to the limited number of experimental data on larger samples. In general, it can be concluded that leaching tests are conservative and the results should be considered as an upper limit of the release rate (IAEA, 1993).

The leach rate is in general higher for bitumen matrices consisting of a hard bitumen than for those consisting of a soft bitumen. (Brodersen *et al.*, 1998, Brunel *et al.*, 1996, Sobolev *et al.*, 1999). In contrast to this, the bitumen types investigated by Aittola and Kleveland (1982) have no effect on the leach rate of caesium or of cobalt.

The leach rate increases with increasing salt loading (Sobolev *et al.*, 1999, Mattus *et al.*, 1989). This is probably due to a thinner layer of bitumen surrounding the salt crystals when the salt content increases. Increasing the amount of resin in a bitumen/resin mixture from 30 to 55 % by volume could double the leach rate of cobalt but it has a limited effect on that of caesium (Aittola and Kleveland, 1982).

The influence of pH on leaching of bitumenised evaporator concentrate and reprocessing sludge (Eurobitum) is discussed in Brodersen *et al.* (1998). The leaching of  $^{60}\text{Co}$ ,  $^{90}\text{Sr}$  and  $\beta$ -emitters is diffusion controlled, but  $^{137}\text{Cs}$  is probably not. There are indications that the extraction of  $^{137}\text{Cs}$ ,  $^{60}\text{Co}$  and  $\beta$ -emitters is higher in an environment with pH 12 than with pH 7 – 7.5, and that the leached fraction of the  $\alpha$ -emitters  $^{241}\text{Am}$ , Pu and total  $\alpha$  is lower at high pH. The leaching of  $\text{NaNO}_3$  is diffusion controlled and largely independent of pH, the latter supported by Yamaguchi *et al.* (1999). At a pH around 7 the leaching of  $\text{CaSO}_4$  is controlled by diffusion. At pH 12 the concentration of  $\text{Ca}^+$  and  $\text{SO}_4^{2-}$  in the leachate solution is controlled by the solubility of these ions. Experimentally estimated diffusion coefficients indicate that a full scale drum filled with Eurobitum R85/40 would be depleted in Na and  $\text{NO}_3^-$  in less than 13,000 and 20,000 years respectively (Sneyers and van Iseghem, 1998). The difference in leach rate between hard and soft bitumen is larger at a high pH (12) than at a lower pH (7 – 7.5) (Brodersen *et al.*, 1998).

Based on experiments on CEA bitumen (simulated bitumenised reprocessing sludge from La Hague) (Brodersen *et al.*, 1998) it can be concluded that:

- The leach rate of  $\text{NO}_3^-$  depends on the type of bitumen. Higher release rates are obtained for hard Mexphalt R90/40 than for the soft Mexphalt 80/100. This is explained by the formation of cracks in hard bitumen.
- Ni, Co, Fe and Ba are present in phases with very low solubility, e.g.  $\text{BaSO}_4$  and  $\text{Ni}_2[(\text{Fe},\text{Mn})(\text{CN})_6]$ , and the release of these species is therefore not diffusion controlled.
- The leaching of  $^{241}\text{Am}$  and  $^{238}\text{Pu}$  in alkaline conditions is controlled by their low solubility. No difference between hard and soft bitumen can be proved.
- The leaching of  $^{137}\text{Cs}$  seems to be uninfluenced by the type of bitumen.
- The release of  $^{90}\text{Sr}$  from soft bitumen is probably diffusion controlled. This behaviour can not be shown for hard bitumen due to large intersample variations. A

high pH environment suppresses the release rate in comparison to that in a neutral environment.

- Experiments performed at 0.1 MPa and 4.0 MPa demonstrate that the leaching of  $\text{NO}_3^-$  and  $\text{SO}_4^{2-}$  is independent of pressure.

Buckley and co-workers (Buckley *et al.*, 1985a) investigated the effect of micro-organisms on the release rate of bitumenised  $^{137}\text{Cs}$  and  $^{60}\text{Co}$ . A sodium nitrate solution containing the two nuclides was solidified in oxidised bitumen. The matrix was leached with different solutions, some containing micro-organisms and some not. The presence of micro-organisms did not influence the release rate. However, the salt content of the groundwater had a significant influence on the release rate. The diffusivity of  $^{60}\text{Co}$  and  $^{137}\text{Cs}$  was an order of magnitude less in salt laden groundwater than in distilled water. The leaching rate of  $^{137}\text{Cs}$  is higher than that of  $^{60}\text{Co}$ . This is supported by results obtained by Kopajtic *et al.* (1989), who also state that a possible explanation is that there is no appreciable interaction between caesium and bitumen, while cobalt forms insoluble hydrolysis products. Aittola and Kleveland (1982) conclude that the leach rate for caesium in bitumenised resin generally is higher than for cobalt and for strontium. However, when the resins contain decontamination chemicals the leach rate for cobalt is equal to or even higher than for caesium.

The leach rate for a matrix containing coarse  $\text{NaNO}_3$ -crystals (200 – 300  $\mu\text{m}$ ) is lower than that containing finer particles (20 – 100  $\mu\text{m}$ ) (Brodersen *et al.*, 1998). This could be due to the thicker bitumen film surrounding larger particles, but an influence of settling of the larger particles can not be excluded.

Experiments performed at Risø on bitumenised evaporator concentrate (Brodersen and Nilsson, 1991), have shown that the release of  $\text{NaNO}_3$  is between two and three times lower in samples enclosed in a container than for samples that can swell unhindered.

Leaching tests on full-scale bitumen cylinders consisting of sludge embedded in bitumen have been performed (Bernard *et al.*, 1982). The waste contains  $^{90}\text{Sr}$ ,  $^{239}\text{Pu}$  and  $^{241}\text{Am}$  or  $^{60}\text{Co}$ ,  $^{90}\text{Sr}$ ,  $^{137}\text{Cs}$  and  $^{239}\text{Pu}$ . The results reveal that the cumulative fraction of  $^{239}\text{Pu}/^{241}\text{Am}$  released after eighteen months is about  $10^{-5}$  and of  $^{90}\text{Sr}/^{137}\text{Cs}$  after one year is about  $10^{-4}$ .

The scale effect on leaching of waste cylinders has been examined (Brodersen *et al.*, 1998). Samples of 0.2, 2, 20 and 200 litres have been leached for 455 days. The leaching rate of  $^{137}\text{Cs}$  from 20 and 200 litre samples is one order of magnitude larger than that from 0.2 and 2 litre samples. For  $^{60}\text{Co}$ , on the other hand, the leaching rate seems to be higher for 0.2 and 2 litre samples.

Irradiation of bitumen increases the leaching rate of both  $^{137}\text{Cs}$  and  $^{60}\text{Co}$ . This is explained by the formation of cracks (Kopajtic *et al.*, 1989). An increased leaching of cobalt and caesium when the bitumenised resins are irradiated is indicated by the results presented by Aittola and Kleveland (1982) as well. Burnay (1987) reports a lowering of  $^{137}\text{Cs}$  release in  $\gamma$ -irradiated samples. This appears to be due to an increase in the distribution coefficient,  $K_d$  for sorption on the evaporator concentrate. Irradiation of bitumenised sludges has been investigated by Sobolev *et al.* (1999). Absorbed doses up to  $10^6$  Gy at dose rates from 10 Gy/h to  $10^3$  Gy/h have no effect on the leach resistance.

Two different programs concerning long-term leaching tests of bitumenised sludge (dominated by caesium) performed under “natural disposal conditions” are reported in Sobolev *et al.* (1999). In the first program, samples of a few litres or few tens of litres were filled in containers with the upper part open to the atmosphere. The containers were placed in the open. In the second program, waste moulds were buried at a depth of about 2 m. Rainwater and groundwater, respectively, contacting the waste were collected for chemical and radiometric analyses for nearly three decades. Parts of the results are summarised in Table 8. The authors conclude from the results of the programs that the major part of the radioactivity is released during the first year of the test period, and that soft bitumen is better than harder bitumen.

**Table 8 Results from long-term leaching tests (Sobolev *et al.*, 1999).**

Parameter	Time [years]	Open area		Near-surface repository			
		Sample 1 Pr 10	Sample 2 Pr 11	Sample 3 K 27	Sample 4 K 4	Sample 5 K 5	Sample 6 K 6
Waste type		Radon-73	Kursk NPP	Kursk NPP	Institu- tional	Institu- tional	Institu- tional
Bitumen type		BN-II	BN-IV	BN-IV	BN-III	BN-IV	BN-II
Waste load [%]		31	31	31	28	27	44
Leaching rate [g/cm <sup>2</sup> , day]	1	1.0·10 <sup>-7</sup>	7.3·10 <sup>-5</sup>	7.1·10 <sup>-7</sup>	2.2·10 <sup>-6</sup>	4.1·10 <sup>-5</sup>	1.8·10 <sup>-7</sup>
	11	2.6·10 <sup>-7</sup>	4.6·10 <sup>-5</sup>	1.3·10 <sup>-7</sup>	8.7·10 <sup>-7</sup>	1.5·10 <sup>-5</sup>	5.9·10 <sup>-8</sup>
	24	2.6·10 <sup>-7</sup>	-	-	4.9·10 <sup>-7</sup>	8.1·10 <sup>-6</sup>	3.4·10 <sup>-8</sup>
Leached fraction [-]	1	4·10 <sup>-4</sup>	1·10 <sup>-1</sup>	1·10 <sup>-3</sup>	4·10 <sup>-3</sup>	7·10 <sup>-2</sup>	2·10 <sup>-4</sup>
	11	1·10 <sup>-2</sup>	6·10 <sup>-1</sup>	2·10 <sup>-3</sup>	5·10 <sup>-3</sup>	8·10 <sup>-2</sup>	3·10 <sup>-4</sup>
	24	2·10 <sup>-2</sup>	-	-	7·10 <sup>-3</sup>	8·10 <sup>-2</sup>	4·10 <sup>-4</sup>

Sobolev and co-workers also report on another experiment where two samples of bitumenised sludge with different waste loading (29 % for S-I and 5 % for S-II) were buried in loamy soil without any additional barrier, and the radionuclide concentration in the soil was measured. The concentration in the surrounding soil after eleven years is shown in Figure 2. The maximum radionuclide concentration was measured underneath the waste blocks and was higher for S-II ( $2.7 \cdot 10^6$  Bq/kg) than for S-I ( $2.8 \cdot 10^4$  Bq/kg). However, the average radionuclide migration rate was higher for S-I, 36 mm/year for S-I and 24 mm/year for S-II. The difference in migration rate indicates the influence of salt content. A higher salt content leads to a higher release of Na<sup>+</sup>. Since the radionuclides compete with Na<sup>+</sup> for the available sorption sites, the increased release of Na<sup>+</sup> leads to a reduced possibility for the radionuclides to sorb in the soil.



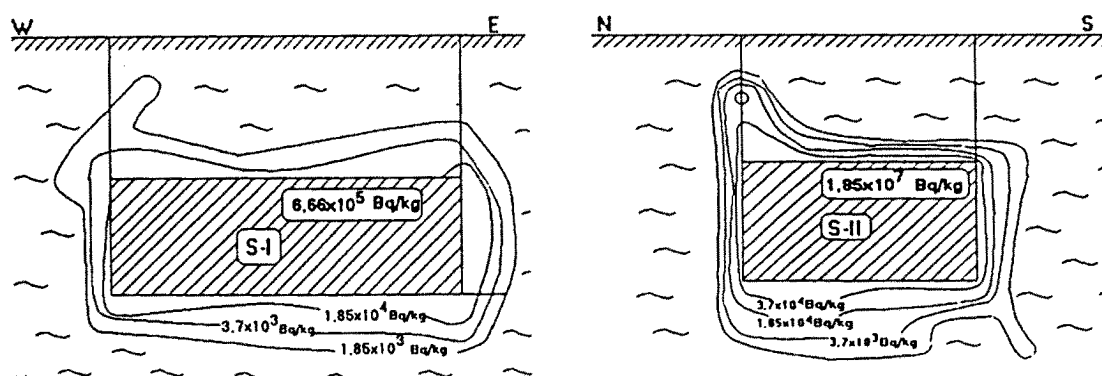


Figure 2 Measured radioactivity around buried bitumenised waste blocks after 11 years of deposition (Sobolev et al., 1999)

Comparatively few data is available on leaching of bitumenised resins. The work of Aittola and Kleveland (1982) has been discussed earlier in this section. Berntsson (1991a and 1991d) reports some results from two different experiments performed by Sydkraft (Harfors, 1978). The first experiment is on leaching of a perforated steel drum (200 l) containing bitumenised waste immersed in drinking water for 130 days. The results are shown in Table 9.

Table 9 Leaching [% of initial inventory] from steel drum with bitumenised waste (Harfors, 1978).

Nuclide	Amount leached after 130 days
$^{58}\text{Co}$	$2.3 \cdot 10^{-4}$
$^{60}\text{Co}$	$1.5 \cdot 10^{-4}$
$^{54}\text{Mn}$	$7.1 \cdot 10^{-4}$
$^{65}\text{Zn}$	$4.2 \cdot 10^{-4}$

The other experiment is a laboratory scale experiment in which the influence of water composition on leaching of grinded granular ion exchange resin embedded in bitumen is studied. The results are summarised in Table 10. It can be concluded that the highest release rate is obtained with drinking water and saline water.

Table 10 Leach rate [m/s] from bitumenised ion exchange resin (Harfors, 1978).

Water	Leach rate	
	$^{60}\text{Co}$	$^{137}\text{Cs}$
De-ionized water	$6 \cdot 10^{-15}$	$4 \cdot 10^{-13}$
Drinking water	$1 \cdot 10^{-12}$	$2 \cdot 10^{-12}$
Saline water	$3 \cdot 10^{-12}$	$6 \cdot 10^{-12}$
Concrete water	$2 \cdot 10^{-14}$	$2 \cdot 10^{-15}$

## 5 Evaluation of long-term properties of bitumenised waste at SFR 1

The material given in the previous chapters should form the basis for deciding which processes that has to be accounted for in the safety assessment. In this final chapter a judgement is made whether the different processes has to be accounted for or not.

### 5.1 Radiolytic effects

The effect of radiolysis of bitumen depends among other things on the energy absorbed by the bitumen matrix. The bitumen qualities used for the bitumenised waste deposited in SFR 1 are so called distilled bitumen, which is more prone to form bubbles than cracks. An extensive formation of cracks in the bitumen matrix is therefore not foreseen. Bubble formation that leads to swelling is more likely.

The absorbed energy due to radiolytic decay has been estimated for different waste categories with bitumenised waste (see Appendix A). When the calculations are based on the mean activity of nuclides within each category it is concluded that:

- the categories B.05 and F.05 deposited in BMA correspond to an integrated absorbed energy that is less than 0.01 MGy within a period of  $10^4$  years
- decay of radionuclides in F.17, also deposited in BMA, results in an integrated absorbed energy of 0.02 MGy within the same period of time
- for B.06 and F.18 deposited in the Silo the integrated absorbed energy is estimated to 0.05 MGy and 0.1 MGy, respectively
- the integrated absorbed energy for waste within BLA (B.20 and F.20) will be less than that for category B.05 and F.05

Using the database on waste already produced and deposited in SFR 1 (TRIUMF, 2000) a rough estimate indicates that the content of  $^{60}\text{Co}$  corresponds to an absorbed energy exceeding  $10^5$  Gy in a few percent of the packages in category B.05 and B.06. The rule of thumb given in the last paragraph in Section 4.1 says that negligible effects on gas generation, swelling, hardening, heating and decrease in leach resistance are found when the absorbed energy is less than 0.1 MGy. For energies about 2 – 10 MGy and higher, a substantial swelling can take place and most likely also an increased leachability (Eschrich, 1980). This indicates that the properties of at least some packages in B.05 and B.06 can be affected by internal radiation. For F.18 the corresponding factor is about fifty percent.

Formation of organic degradation products by irradiation of bitumen has been demonstrated (e.g. Valcke *et al.*, 1999). However, this effect is considered to be of limited importance for the long-term performance of the repository in comparison to degradation of cellulose.

From these results it is determined that radiolytic effects can be neglected in the safety assessment except for waste in categories B.05, B.06 and F.18. For these categories the possibility of an increased rate of water uptake due to radiolysis can not be excluded.

## **5.2 Biodegradation**

The rate of biodegradation of bitumen is generally low. This is especially true for anaerobic conditions foreseen to prevail in SFR 1. The influence of biodegradation on the release rate of radionuclides from a bitumen matrix will therefore most likely be small. This is supported by work on the long term stability of natural bitumen and natural bitumen analogues indicating that bitumen is stable over periods for more than  $10^4$  up to  $10^7$  years (IAEA, 1993). It is concluded that the effect of micro-organisms on the long-term properties of the bitumenised waste can be neglected.

## **5.3 Ageing**

The main effect of ageing is that the bitumen becomes harder and more brittle, which can lead to fracturing. The most important ageing process in the short-term seems to be oxidation. The conditions at repository depth are, however, not favourable (darkness and anaerobic). Oxygen may penetrate into a thin region close to the matrix surface only. The volume of the bitumen matrix that is affected by oxidation is small in comparison to that affected by water uptake.

In contrast to oxidation, the hardening of bitumen caused by a redistribution of molecules or evaporation of hydrocarbons affects the whole bitumen volume. The water uptake rate is not influenced by the hardening process at first, but as the bitumen hardens a reduced ability to deform when the waste swell can be obtained. Small cracks may be formed in the material instead. This in turn increases the rate of water uptake, swelling and release of radionuclides. The effect of this ageing process on the long-term performance of a bitumen barrier is unknown.

## **5.4 Water uptake**

### **5.4.1 Evaluation of water uptake in bitumenised waste deposited in SFR 1**

The bitumenised waste deposited in SFR 1 contains mainly ion exchange resins and relatively small amounts of evaporator concentrate (see Chapter 3). Of the bitumenised waste produced at Forsmark NPP, F.17 is the only waste category containing evaporator concentrate. According to the composition of a reference package, the content of evaporator concentrate in this category is about 3 volume %. The bitumenised waste from Barsebäck NPP may contain some evaporator concentrate. In addition, sodium sulfate is added in the bitumenisation process. The content of free sodium sulfate in the end product is, however, less than 1 % (Berntsson, 1991a, 1991d and 1992).

A fundamental difference between a resin and a concentrate is that the concentrate dissolves upon water uptake and will diffuse out from the waste matrix but the former

remains in the bitumen matrix. Swelling caused by water uptake of easily soluble salts present in the SFR 1 waste can therefore be expected to be small in comparison to the swelling that can be obtained for bitumenised waste with large amounts of ion exchange resins. It can therefore be concluded that water uptake in the bitumen matrix is governed by the ion exchange resins.

The experiments performed by Brodén and Wingefors (1992) indicate that water uptake in a bitumen matrix containing 40 weight % non-radioactive ion exchange resins is negligible, but that the water uptake increases significantly at higher waste loads (up to 70 %). When the amount of resins is 60 weight % or more the water uptake is so fast that it can not be described as a diffusive process. Aittola and Kleveland (1982) conclude that the water uptake rate is slow when the amount of ion exchange resins in bitumenised waste is 58 weight % or less, and that the water uptake rate increases one order of magnitude when the resins are irradiated. Note, however, that the experiments by Brodén and Wingefors (1992) were conducted for one month only and those by Aittola and Kleveland (1982) between four months and four years. Extrapolating the results for very long time periods is uncertain.

The bitumenised waste produced at Barsebäck NPP contains 25 weight % ion exchange resins (Table 4). The data given in the paragraph above indicates that water uptake rate should be a very slow process why it should take a very long time before the waste matrix is severely affected by penetrating water. The amount of ion exchange resins in waste produced at Forsmark NPP is between 40 and 60 weight %. For part of the waste from Forsmark NPP the water uptake rate should be very slow. The amount of resins in waste categories F.05 and F.20 is so high (60 weight %) that a very rapid water uptake can not be excluded. In addition, the waste loading in category F.17 (50 weight % resins and concentrate) is above the limit for which water uptake is negligible according to Brodén and Wingefors (1992).

It is important to keep in mind that there are several factors of importance for the long-term performance of a bitumen barrier that is not accounted for in the data presented in the literature. The main issue is the time dependence. The effect of ageing of bitumen and the effect of deterioration of the bitumen matrix due to water uptake is unknown. The possibility of an increased water uptake rate in and nuclide release rate from the bitumen matrix after a longer period of time can therefore not be overlooked.

Furthermore, the effect of a low temperature on the bitumen barrier is not very well documented. After repository closure, the temperature in the near-field rock of SFR 1 will be approximately the same as the mean temperature of the water in Öregrundsgrepen (about 6 – 7 °C). Most of the experimental data presented in the literature are obtained at a temperature significantly above that. However, Brodén and Wingefors (1992) report a much faster water uptake rate at 3°C in comparison to that at 20°C and 30°C.

In addition, the water uptake rate depends on the composition of the bitumenised waste. Brodersen *et al.* (1998) report a much higher swelling rate for bitumenised waste products from the Risø waste processing plant in comparison to that obtained using synthetic mixtures made in the laboratory.

It is therefore possible that the water uptake rate can be of importance for the long-term performance even though a certain waste category has a waste content below the upper limit for which the literature conclude that water uptake is limited. The effect of water uptake in bitumenised waste in SFR 1 therefore has to be evaluated in more detail before it can be concluded for how long time the bitumen matrix will be an effective barrier for release of radionuclides.

#### **5.4.2 Theoretical maximum volume increase of bitumenised waste in SFR 1 due to water uptake**

The theoretical maximum volume increase due to water uptake of ion exchange resins and evaporator concentrate in the waste has been estimated (Appendix B). In order to get an estimate of the consequence of swelling of bitumenised waste, the maximum theoretical volume increase is compared with the available voids within the different waste packagings.

It is estimated that the volume increase for bitumenised waste stored in BLA is well below the available void inside an ISO container.

For B.05, F.05 and F.17 stored in BMA the theoretical maximum volume increase is comparable to or larger than the available void inside the packages. As stated earlier in this report, about 20 % of the total volume available for waste deposition in BMA is foreseen to consist of bitumenised waste. The rough estimate of volume increase indicates that the concrete structure located inside BMA should not be backfilled. If it is, it can not be ruled out that the pressure built up inside the structure when the bitumenised waste swell will affect other barriers in BMA (e.g. concrete walls of the structure).

The theoretical maximum swelling of waste in category B.06 stored in the Silo is estimated to be comparable to the available void inside the drum and the expansion box deposited together with the drums. For category F.18 on the other hand, the volume increase is roughly twice the available void. The amount of waste in F.18 to be deposited in the Silo is estimated to be only a few per cent of the total volume inside the Silo. The bitumenised waste is, however, concentrated to the central part of the Silo and can therefore locally constitute a significant fraction of the waste. Local effects due to swelling can therefore arise.

### **5.5 Evaluation of the importance of the bitumen matrix as a barrier**

#### **5.5.1 Estimation of transport times of radionuclides in SFR 1**

The ability of the bitumen matrix to retain radionuclides is only of interest if the nuclides are retained for times comparable to or longer than the time needed for the nuclides to migrate through subsequent barriers. This time has been roughly estimated for BMA for the hydrological conditions prevailing during the first 1,000 years after repository closure (Appendix C). The results indicate that it takes 600 – 800 years for the nuclides to be transported out from BMA when the barrier effect of the bitumen

matrix is neglected. Note, however, that no retarding mechanisms such as sorption of radionuclides have been accounted for. The concrete structure built inside BMA is assumed to be intact. If there is an increased water flow through one or several sections of the structure due to fracturing of the structure, the transport time through the structure will decrease. However, it is the transport through the backfill on top of the structure that gives the largest contribution to the transport time, and that is to a large extent unaffected by fractures in the structure. Theoretically, fracturing of the concrete structure could reduce the transport time to some 500 years.

The construction of the Silo is such that the water flow through the Silo is very low, and the transport will be dominated by diffusion. The time needed for the nuclides to diffuse from the Silo to the surrounding rock has not been evaluated. It is, however, reasonable to assume that the time for an intact Silo will not be less than that for BMA.

In BLA the only barrier to nuclide transport is the packages in which the waste is deposited. For bitumenised waste this is a 216 litres steel drum in which the bitumen matrix is filled and a 19 m<sup>3</sup> ISO container made of steel in which a group of drums are placed before deposition in BLA. The ability of the ISO container and the steel drum to function as a barrier for radionuclide transport is neglected in the safety assessment. This means that the bitumen matrix is the only material retarding the near-field release of radionuclides. Taking the barrier effect of bitumen into account could therefore have a significant influence of the radionuclide release rate from BLA.

### 5.5.2 Comparison of transport times of radionuclides in SFR 1 and release rates of radionuclides from a bitumen matrix

Data on leach rate for different radionuclides and salts from bitumen can be found in the literature. Estimated leach rates varies by several orders of magnitude (Table 7). Assuming that these leach rates are constant in time and neglecting radioactive decay, the time needed to leach the whole nuclide inventory from bitumenised waste in a 200 litres drum can be calculated and are given in Table 11. The leach rate given for alkali and alkaline earth metals are such that it can not be excluded that they are released from the bitumen matrix faster than the estimated times for nuclide transport in the rest of BMA and the Silo. On the other hand, the leach rate for actinides, rare earth metals and for reprocessing waste radionuclides is so low that the release from the bitumen matrix could determine the release rate of radionuclides to the far-field.

**Table 11** Calculated time needed for leaching different elements from a 200 litre drum.

Element	Leach time [years]
actinides and rare earth metals	$2 \cdot 10^3 - 2 \cdot 10^6$
reprocessing waste nuclides	$2 \cdot 10^3 - 2 \cdot 10^4$
alkali and alkaline earth metals	$5 - 5 \cdot 10^3$

Experiments have been performed on real waste products from the waste management plant at Risø in order to investigate the leaching of soluble salt incorporated in a bitumen matrix (Brodersen *et al.*, 1998). Extrapolating their results in time indicates that it takes about 700 years to leach a 0.1 m thick layer of bitumen matrix on sodium and about 6,000 years to leach a thickness equivalent to half the diameter of the drum. For synthetic samples prepared in laboratory the leached thickness after 700 years is about 0.01 m to 0.05 m. The release rate of easily soluble salt (e.g. sodium) can be assumed to be of the same order of magnitude as for easily soluble radionuclides.

The diffusivity of  $\text{Na}^+$  and  $\text{NO}_3^-$  in bitumenised sludge and bitumenised mixtures of evaporator concentrate and sludge from the former Eurochemic facility has been estimated (Sneyers and van Iseghem, 1998). Based on these experiments it is evaluated that a 200 litres drum with bitumenised concentrate and sludge will be depleted in  $\text{Na}^+$  and  $\text{NO}_3^-$  in less than 13,000 and 20,000 years, respectively. For nuclides with a low solubility the depletion rate will be even lower.

### 5.5.3 Summary

The discussion in this section is to a large extent generic in terms of radionuclides. However, whether the bitumen matrix is of importance for the near-field release rate or not is nuclide dependent. The initial inventory of the nuclide at repository closure is of course of importance, but also the distribution between bitumenised and non-bitumenised waste. If only a small part of the total inventory of a nuclide within a repository part is embedded in bitumen, the delaying effect of bitumen will have a small or even negligible effect on the near-field release rate of that nuclide.

Other factors of importance are the radionuclide's chemical properties (e.g. ability to sorb and form complexes, solubility) and half-life. The chemical properties affect the transport rate through the barrier and thus when the radionuclide is released from the barrier, and the half-life the rate at which the initial inventory is reduced due to radioactive decay.

There is also a variation between different repository parts. For instance, a nuclide with a short half-life (e.g.  $^3\text{H}$  and  $^{90}\text{Sr}$ ) will decay within the near-field regardless of whether the bitumen matrix is accounted for or not when there are several other barriers in the repository besides the waste package. But for a repository where bitumen is the only barrier the retaining effect of bitumen can be crucial. On the other hand, a bitumen matrix is foreseen to have a limited effect on the release rate of very long-lived, sorbing nuclides in any part of SFR 1.

Only four percent of the total waste volume in BLA is bitumenised. On the other hand, all organic  $^{14}\text{C}$  in BLA is embedded in bitumen. The total inventory of organic  $^{14}\text{C}$  in BLA is three to four orders of magnitude lower than that for the Silo, BMA and 1BTF/2BTF. However, the barriers in BLA are less sophisticated than in the other repository parts. The total release of organic  $^{14}\text{C}$  from SFR 1 may therefore be dominated by the contribution from release of organic  $^{14}\text{C}$  from BLA in the short-term after repository closure, especially if the bitumen barrier in BLA is neglected. For all other nuclides in BLA the inventory in bitumenised waste is less than one percent of the

total nuclide inventory in BLA. The bitumen barrier will therefore be of no significance for the release of these radionuclides from BLA.

For BMA and the Silo the importance of a bitumen barrier is more difficult to judge since the other barriers in these repository parts have to be accounted for as well. It can, however, be concluded that short-lived nuclides will decay to a large extent within BMA and the Silo even if the bitumen barrier is neglected. A bitumen matrix will also have a limited effect on the release rate of very long-lived, sorbing nuclides from these two repository parts.

Approximately one fourth of the volume to be deposited in BMA is expected to be bitumenised. For many nuclides between 25 and 50 % of the total nuclide inventory in BMA is contained in bitumen. The difference in total release rate from BMA between a case where bitumen is accounted for and a case where it is not can therefore at maximum affect the release rate of between 25 to 50 % of these nuclides.

About one fifth of the volume to be stored in the Silo is foreseen to be conditioned in bitumen. The nuclide inventory in waste embedded in bitumen is at maximum about one third of the total nuclide inventory in the Silo. Examples of nuclides are  $^3\text{H}$ , inorganic  $^{14}\text{C}$ ,  $^{60}\text{Co}$ ,  $^{59}\text{Ni}$  and  $^{63}\text{Ni}$ . A reduction in release rate from the Silo can therefore influence 33 % of the nuclides by taking the effect of a bitumen barrier into account.

## 5.6 Conclusions

The experimental data available maintain that bitumen is an effective barrier against release of radionuclides. Based on leaching experiments reviewed in this study, it could take some thousand years, possibly more, to release all radionuclides in a 200-litre drum. Since the surface-to-volume ratio is the same for drum and a  $1.7\text{ m}^3$  steel container, it is reasonable to assume a similar time scale to leach a container.

The results are, however, extrapolated from experiments performed during a short period of time. Long-term deteriorating effects and the effect of a low temperature on the bitumen matrix are not very well documented. The literature focuses principally on bitumenised evaporator concentrate, but the bitumenised waste deposited in SFR 1 consists mainly of ion exchange resins. There are indications that the non-radioactive waste products usually investigated overestimate bitumen's ability to retain waste. Radiolytic effects has been estimated to be negligible for waste categories B.20, F.17 and F.20 deposited in SFR 1, but for categories B.05, B.06 and F.18 the possibility of increased water uptake rate due to radiolysis can not be excluded. A more reasonable assumption is that bitumen will act as an effective barrier for radionuclide release during a time span from several hundreds to thousand of years.

Generally, the majority of the inventory of radionuclides in SFR 1 is not solidified in bitumen. By taking the bitumen barrier into account the total release rate from BMA and the Silo could be reduced at maximum by a factor of 2 and 1.5, respectively. In BLA organic  $^{14}\text{C}$  is only present in waste embedded in bitumen. Thus, taking the effect of a bitumen barrier into account can have an influence on the release rate of organic  $^{14}\text{C}$  from this repository part, but for the other nuclides in BLA the bitumen barrier is of little importance.



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## **Appendix A: Estimation of absorbed energy caused by radiolytic decay**

# 1 Background

The energy absorbed when radionuclides decay affects the properties of the bitumen matrix. The result of a significant absorption of energy can for instance be an increased leachability. This document describes how the amount of energy absorbed is estimated. Results are also presented for the bitumenised waste deposited today at SFR 1.

## 2 Nuclide inventory

The activity in newly embedded waste is needed in order to estimate the total amount of energy that has been absorbed by the bitumen matrix since the waste was produced. A mean value of the nuclide inventory in newly embedded waste has been estimated (Riggare and Johansson, 2001). For waste category F.05 Riggare and Johansson (2001) give data on activity of transuranic elements at repository closure, but not for newly embedded waste. The activity of transuranic elements in newly embedded waste in waste category F.05 has been estimated as follows.

The ratio in activity between  $^{239}\text{Pu}$  and any long-lived nuclide (e.g.  $^{129}\text{I}$ ) is assumed to be equal for newly embedded waste and at repository closure. This is reasonable since the time between waste production and repository closure is short in comparison to the half-life of  $^{239}\text{Pu}$  and  $^{129}\text{I}$ . The activity of  $^{239}\text{Pu}$  in newly embedded waste is finally obtained by multiplying the activity of  $^{129}\text{I}$  in newly embedded waste with the obtained ratio in activity at repository closure. The same approach is made for  $^{240}\text{Pu}$ . The activity of other transuranic elements (and some of their daughters) in newly embedded waste is estimated using correlation factors (Lindgren *et al.*, 1998). The activity of each nuclide is estimated from the sum of the activity of  $^{239}\text{Pu}$  and  $^{240}\text{Pu}$  in newly embedded waste and the correlation factors compiled in Table 1.

The estimated mean activity of the nuclides in newly embedded waste in categories B.05, B.06, F.05, F.17 and F.18 is compiled in Table 2.

**Table 1 Correlation factors used for estimating the nuclide inventory of transuranic elements and some of their daughters in newly embedded waste in waste category F.05 (Lindgren *et al.*, 1998).**

<b>Nuclide</b>	<b>Correlation factor</b>	<b>Correlated to</b>
Pb-210	$2.0 \cdot 10^{-11}$	Pu-239/240
Ra-226	$2.0 \cdot 10^{-10}$	Pu-239/240
Ac-227	$4.0 \cdot 10^{-9}$	Pu-239/240
Th-229	$3.0 \cdot 10^{-10}$	Pu-239/240
Th-230	$9.0 \cdot 10^{-8}$	Pu-239/240
Th-232	$1.0 \cdot 10^{-13}$	Pu-239/240
Pa-231	$3.0 \cdot 10^{-8}$	Pu-239/240
U-232	$3.0 \cdot 10^{-5}$	Pu-239/240
U-233	$2.0 \cdot 10^{-8}$	Pu-239/240
U-234	$1.0 \cdot 10^{-3}$	Pu-239/240
U-235	$2.0 \cdot 10^{-5}$	Pu-239/240
U-236	$3.0 \cdot 10^{-4}$	Pu-239/240
U-238	$4.0 \cdot 10^{-4}$	Pu-239/240
Np-237	$4.0 \cdot 10^{-4}$	Pu-239/240
Pu-238	4.0	Pu-239/240
Pu-241	$1.0 \cdot 10^2$	Pu-239/240
Pu-242	$3.0 \cdot 10^{-3}$	Pu-239/240
Pu-244	$7.0 \cdot 10^{-10}$	Pu-239/240
Am-241	1.0	Pu-239/240
Am-242m	$1.0 \cdot 10^{-2}$	Pu-239/240
Am-243	$3.0 \cdot 10^{-2}$	Pu-239/240
Cm-243	$2.0 \cdot 10^{-2}$	Pu-239/240
Cm-244	3.0	Pu-239/240
Cm-245	$3.0 \cdot 10^{-4}$	Pu-239/240
Cm-246	$8.0 \cdot 10^{-5}$	Pu-239/240



**Table 2 Estimated activity of nuclides in newly embedded waste.**

Element	Half-life [yr]	Inventory [Bq/package]				
		B.05	B.06	F.05	F.17	F.18
H-3	1.23·10 <sup>1</sup>	8.7·10 <sup>5</sup>	6.3·10 <sup>6</sup>	4.0·10 <sup>5</sup>	9.2·10 <sup>6</sup>	8.2·10 <sup>7</sup>
Be-10	1.51·10 <sup>6</sup>	5.2	3.8·10 <sup>1</sup>	2.4	5.5·10 <sup>1</sup>	4.9·10 <sup>2</sup>
C-14	5.73·10 <sup>3</sup>	8.7·10 <sup>6</sup>	6.3·10 <sup>7</sup>	4.0·10 <sup>6</sup>	9.2·10 <sup>7</sup>	8.2·10 <sup>8</sup>
Cl-36	3.01·10 <sup>5</sup>	7.2·10 <sup>3</sup>	5.1·10 <sup>4</sup>	4.4·10 <sup>2</sup>	1.7·10 <sup>5</sup>	8.6·10 <sup>5</sup>
Fe-55	2.73	8.7·10 <sup>9</sup>	6.3·10 <sup>10</sup>	4.0·10 <sup>9</sup>	9.2·10 <sup>10</sup>	8.2·10 <sup>11</sup>
Co-60	5.27	8.7·10 <sup>9</sup>	6.3·10 <sup>10</sup>	4.0·10 <sup>9</sup>	9.2·10 <sup>10</sup>	8.2·10 <sup>11</sup>
Ni-59	7.60·10 <sup>4</sup>	8.7·10 <sup>6</sup>	6.3·10 <sup>7</sup>	4.0·10 <sup>6</sup>	9.2·10 <sup>7</sup>	8.2·10 <sup>8</sup>
Ni-63	1.00·10 <sup>2</sup>	1.7·10 <sup>9</sup>	1.3·10 <sup>10</sup>	7.9·10 <sup>8</sup>	1.8·10 <sup>10</sup>	1.6·10 <sup>11</sup>
Se-79	1.13·10 <sup>6</sup>	2.9·10 <sup>3</sup>	2.0·10 <sup>4</sup>	1.7·10 <sup>2</sup>	6.6·10 <sup>4</sup>	3.4·10 <sup>5</sup>
Sr-90	2.88·10 <sup>1</sup>	7.2·10 <sup>7</sup>	5.1·10 <sup>8</sup>	4.4·10 <sup>6</sup>	1.7·10 <sup>9</sup>	8.6·10 <sup>9</sup>
Zr-93	1.53·10 <sup>6</sup>	8.7·10 <sup>3</sup>	6.3·10 <sup>4</sup>	4.0·10 <sup>3</sup>	9.2·10 <sup>4</sup>	8.2·10 <sup>5</sup>
Nb-93m	1.61·10 <sup>1</sup>	8.7·10 <sup>6</sup>	6.3·10 <sup>7</sup>	4.0·10 <sup>6</sup>	9.2·10 <sup>7</sup>	8.2·10 <sup>8</sup>
Nb-94	2.03·10 <sup>4</sup>	8.7·10 <sup>4</sup>	6.3·10 <sup>5</sup>	4.0·10 <sup>4</sup>	9.2·10 <sup>5</sup>	8.2·10 <sup>6</sup>
Mo-93	4.00·10 <sup>3</sup>	4.4·10 <sup>4</sup>	3.2·10 <sup>5</sup>	2.0·10 <sup>4</sup>	4.6·10 <sup>5</sup>	4.1·10 <sup>6</sup>
Tc-99	2.11·10 <sup>5</sup>	3.6·10 <sup>6</sup>	2.6·10 <sup>7</sup>	2.2·10 <sup>5</sup>	8.3·10 <sup>7</sup>	4.3·10 <sup>8</sup>
Ru-106	1.01	3.6·10 <sup>6</sup>	2.6·10 <sup>7</sup>	2.2·10 <sup>5</sup>	8.3·10 <sup>7</sup>	4.3·10 <sup>8</sup>
Pd-107	6.50·10 <sup>6</sup>	7.2·10 <sup>2</sup>	5.1·10 <sup>3</sup>	4.4·10 <sup>1</sup>	1.7·10 <sup>4</sup>	8.6·10 <sup>4</sup>
Ag-108m	4.18·10 <sup>2</sup>	5.2·10 <sup>5</sup>	3.8·10 <sup>6</sup>	2.4·10 <sup>5</sup>	5.5·10 <sup>6</sup>	4.9·10 <sup>7</sup>
Cd-113m	1.41·10 <sup>1</sup>	4.3·10 <sup>5</sup>	3.1·10 <sup>6</sup>	2.6·10 <sup>4</sup>	1.0·10 <sup>7</sup>	5.2·10 <sup>7</sup>
Sn-126	1.00·10 <sup>5</sup>	3.6·10 <sup>2</sup>	2.6·10 <sup>3</sup>	2.2·10 <sup>1</sup>	8.3·10 <sup>3</sup>	4.3·10 <sup>4</sup>
Sb-125	2.76	8.7·10 <sup>8</sup>	6.3·10 <sup>9</sup>	4.0·10 <sup>8</sup>	9.2·10 <sup>9</sup>	8.2·10 <sup>10</sup>
I-129	1.57·10 <sup>7</sup>	2.2·10 <sup>2</sup>	1.5·10 <sup>3</sup>	1.3·10 <sup>1</sup>	5.0·10 <sup>3</sup>	2.6·10 <sup>4</sup>
Cs-134	2.06	7.2·10 <sup>8</sup>	5.1·10 <sup>9</sup>	4.4·10 <sup>7</sup>	1.7·10 <sup>10</sup>	8.6·10 <sup>10</sup>
Cs-135	2.30·10 <sup>6</sup>	3.6·10 <sup>3</sup>	2.6·10 <sup>4</sup>	2.2·10 <sup>2</sup>	8.3·10 <sup>4</sup>	4.3·10 <sup>5</sup>
Cs-137	3.01·10 <sup>1</sup>	7.2·10 <sup>8</sup>	5.1·10 <sup>9</sup>	4.4·10 <sup>7</sup>	1.7·10 <sup>10</sup>	8.6·10 <sup>10</sup>
Ba-133	1.05·10 <sup>1</sup>	8.7·10 <sup>4</sup>	6.3·10 <sup>5</sup>	4.0·10 <sup>4</sup>	9.2·10 <sup>5</sup>	8.2·10 <sup>6</sup>
Pm-147	2.62	6.5·10 <sup>8</sup>	4.6·10 <sup>9</sup>	3.9·10 <sup>7</sup>	1.5·10 <sup>10</sup>	7.7·10 <sup>10</sup>
Sm-151	9.00·10 <sup>1</sup>	2.2·10 <sup>6</sup>	1.5·10 <sup>7</sup>	1.3·10 <sup>5</sup>	5.0·10 <sup>7</sup>	2.6·10 <sup>8</sup>
Eu-152	1.35·10 <sup>1</sup>	5.0·10 <sup>4</sup>	3.6·10 <sup>5</sup>	3.1·10 <sup>3</sup>	1.2·10 <sup>6</sup>	6.0·10 <sup>6</sup>
Eu-154	8.59	7.2·10 <sup>7</sup>	5.1·10 <sup>8</sup>	4.4·10 <sup>6</sup>	1.7·10 <sup>9</sup>	8.6·10 <sup>9</sup>
Eu-155	4.76	5.0·10 <sup>7</sup>	3.6·10 <sup>8</sup>	3.1·10 <sup>6</sup>	1.2·10 <sup>9</sup>	6.0·10 <sup>9</sup>
Ho-166m	1.20·10 <sup>3</sup>	3.5·10 <sup>4</sup>	2.5·10 <sup>5</sup>	1.6·10 <sup>4</sup>	3.7·10 <sup>5</sup>	3.3·10 <sup>6</sup>
Pb-210	2.23·10 <sup>1</sup>	1.0·10 <sup>-7</sup>	7.5·10 <sup>-7</sup>	2.3·10 <sup>-8</sup>	1.2·10 <sup>-5</sup>	1.1·10 <sup>-4</sup>
Ra-226	1.60·10 <sup>3</sup>	1.0·10 <sup>-6</sup>	7.5·10 <sup>-6</sup>	2.3·10 <sup>-7</sup>	1.2·10 <sup>-4</sup>	1.1·10 <sup>-3</sup>
Ac-227	2.18·10 <sup>1</sup>	2.1·10 <sup>-5</sup>	1.5·10 <sup>-4</sup>	4.7·10 <sup>-6</sup>	2.4·10 <sup>-3</sup>	2.2·10 <sup>-2</sup>
Th-229	7.34·10 <sup>3</sup>	1.6·10 <sup>-6</sup>	1.1·10 <sup>-5</sup>	3.5·10 <sup>-7</sup>	1.8·10 <sup>-4</sup>	1.6·10 <sup>-3</sup>
Th-230	7.54·10 <sup>4</sup>	4.7·10 <sup>-4</sup>	3.4·10 <sup>-3</sup>	1.0·10 <sup>-4</sup>	5.5·10 <sup>-2</sup>	4.9·10 <sup>-1</sup>
Th-232	1.41·10 <sup>10</sup>	5.2·10 <sup>-10</sup>	3.8·10 <sup>-9</sup>	1.2·10 <sup>-10</sup>	6.1·10 <sup>-8</sup>	5.4·10 <sup>-7</sup>
Pa-231	3.28·10 <sup>4</sup>	1.6·10 <sup>-4</sup>	1.1·10 <sup>-3</sup>	3.5·10 <sup>-5</sup>	1.8·10 <sup>-2</sup>	1.6·10 <sup>-1</sup>
U-232	6.89·10 <sup>1</sup>	1.6·10 <sup>-1</sup>	1.1	3.5·10 <sup>-2</sup>	1.8·10 <sup>1</sup>	1.6·10 <sup>2</sup>
U-233	1.59·10 <sup>5</sup>	1.0·10 <sup>-4</sup>	7.5·10 <sup>-4</sup>	2.3·10 <sup>-5</sup>	1.2·10 <sup>-2</sup>	1.1·10 <sup>-1</sup>
U-234	2.46·10 <sup>5</sup>	5.2	3.8·10 <sup>1</sup>	1.2	6.1·10 <sup>2</sup>	5.4·10 <sup>3</sup>
U-235	7.04·10 <sup>8</sup>	1.0·10 <sup>-1</sup>	7.5·10 <sup>-1</sup>	2.3·10 <sup>-2</sup>	1.2·10 <sup>1</sup>	1.1·10 <sup>2</sup>
U-236	2.34·10 <sup>7</sup>	1.6	1.1·10 <sup>1</sup>	3.5·10 <sup>-1</sup>	1.8·10 <sup>2</sup>	1.6·10 <sup>3</sup>
U-238	4.47·10 <sup>9</sup>	2.1	1.5·10 <sup>1</sup>	4.7·10 <sup>-1</sup>	2.4·10 <sup>2</sup>	2.2·10 <sup>3</sup>
Np-237	2.14·10 <sup>6</sup>	2.1	1.5·10 <sup>1</sup>	4.7·10 <sup>-1</sup>	2.4·10 <sup>2</sup>	2.2·10 <sup>3</sup>
Pu-238	8.77·10 <sup>1</sup>	2.1·10 <sup>4</sup>	1.5·10 <sup>5</sup>	4.7·10 <sup>3</sup>	2.4·10 <sup>6</sup>	2.2·10 <sup>7</sup>
Pu-239	2.41·10 <sup>4</sup>	1.7·10 <sup>3</sup>	1.3·10 <sup>4</sup>	3.9·10 <sup>2</sup>	2.0·10 <sup>5</sup>	1.8·10 <sup>6</sup>
Pu-240	6.56·10 <sup>3</sup>	3.5·10 <sup>3</sup>	2.5·10 <sup>4</sup>	7.8·10 <sup>2</sup>	4.0·10 <sup>5</sup>	3.6·10 <sup>6</sup>
Pu-241	1.44·10 <sup>1</sup>	5.2·10 <sup>5</sup>	3.8·10 <sup>6</sup>	1.2·10 <sup>5</sup>	6.1·10 <sup>7</sup>	5.4·10 <sup>8</sup>
Pu-242	3.73·10 <sup>5</sup>	1.6·10 <sup>1</sup>	1.1·10 <sup>2</sup>	3.5	1.8·10 <sup>3</sup>	1.6·10 <sup>4</sup>
Pu-244	8.08·10 <sup>7</sup>	3.6·10 <sup>-6</sup>	2.6·10 <sup>-5</sup>	8.2·10 <sup>-7</sup>	4.2·10 <sup>-4</sup>	3.8·10 <sup>-3</sup>
Am-241	4.32·10 <sup>2</sup>	5.2·10 <sup>3</sup>	3.8·10 <sup>4</sup>	1.2·10 <sup>3</sup>	6.1·10 <sup>5</sup>	5.4·10 <sup>6</sup>
Am-242m	1.41·10 <sup>2</sup>	5.2·10 <sup>1</sup>	3.8·10 <sup>2</sup>	1.2·10 <sup>1</sup>	6.1·10 <sup>3</sup>	5.4·10 <sup>4</sup>
Am-243	7.37·10 <sup>3</sup>	1.6·10 <sup>2</sup>	1.1·10 <sup>3</sup>	3.5·10 <sup>1</sup>	1.8·10 <sup>4</sup>	1.6·10 <sup>5</sup>
Cm-243	2.91·10 <sup>1</sup>	1.0·10 <sup>2</sup>	7.5·10 <sup>2</sup>	2.3·10 <sup>1</sup>	1.2·10 <sup>4</sup>	1.1·10 <sup>5</sup>
Cm-244	1.81·10 <sup>1</sup>	1.6·10 <sup>4</sup>	1.1·10 <sup>5</sup>	3.5·10 <sup>3</sup>	1.8·10 <sup>6</sup>	1.6·10 <sup>7</sup>
Cm-245	8.50·10 <sup>3</sup>	1.6	1.1·10 <sup>1</sup>	3.5·10 <sup>-1</sup>	1.8·10 <sup>2</sup>	1.6·10 <sup>3</sup>
Cm-246	4.73·10 <sup>3</sup>	4.2·10 <sup>-1</sup>	3.0	9.3·10 <sup>-2</sup>	4.8·10 <sup>1</sup>	4.3·10 <sup>2</sup>

### 3 Absorbed energy due to radiolytic decay

The bitumen absorbs the energy radiated by nuclides embedded in the bitumen matrix. The energy absorbed by bitumen during a time period of 't' is given by:

$$q = \frac{k_1 \cdot k_2 \cdot C_0 \cdot (1 - e^{-\lambda \cdot t})}{\lambda} \cdot (E_\alpha + E_\beta + E_\gamma) \quad (1)$$

where	q	is the absorbed energy (Gy)
	C <sub>0</sub>	is the initial nuclide concentration (Bq/kg bitumen)
	λ	is the decay constant (years <sup>-1</sup> )
	t	is the time (years)
	E <sub>α</sub>	is the alpha energy per transformation of the nuclide (eV/decay)
	E <sub>β</sub>	is the beta energy per transformation of the nuclide (eV/decay)
	E <sub>γ</sub>	is the gamma energy per transformation of the nuclide (eV/decay)
	k <sub>1</sub>	is a constant = 3.15 · 10 <sup>7</sup> (s/year)
	k <sub>2</sub>	is a constant = 1.6021 · 10 <sup>-19</sup> (J/eV)

The energy emitted per transformation is nuclide dependent. <sup>60</sup>Co for instance mainly emits γ-radiation but <sup>137</sup>Cs emits β-radiation only. The amount of energy that a nuclide emits when it decays, E, is compiled in Table 3. Inherent in Equation (1) is that the bitumen absorbs all energy emitted by decaying radionuclides. i.e. the waste package is an infinite media. It is a conservative assumption which is reasonable for α, β and low-energy γ-irradiation since the dimensions of waste packages is much larger than the penetration depths of these types of rays (Porath *et al.*, 1981).

For single nuclides the emitted energy can be added to obtain the total absorbed energy. For nuclides that undergo chain decay estimation of absorbed energy is more complicated. One has to take into account the different energies emitted as well as the differences in half-life for the 'mother' and its 'daughters'. A simple and conservative approach has been used to estimate the energy absorbed due to radioactive decay of chains.

**Table 3** Emitted energy per transformation, E [MeV] (ICRP, 1983).

Element	$E_{\alpha}$	$E_{\beta}$	$E_{\gamma}$	Element	$E_{\alpha}$	$E_{\beta}$	$E_{\gamma}$
H-3	0	$5.68 \cdot 10^{-3}$	0.00	Eu-152	0.00	$1.36 \cdot 10^{-1}$	1.14
Be-10	0	$2.52 \cdot 10^{-1}$	0.00	Eu-154	0.00	$2.88 \cdot 10^{-1}$	1.22
C-14	0	$4.95 \cdot 10^{-2}$	0.00	Eu-155	0.00	$6.26 \cdot 10^{-2}$	$6.05 \cdot 10^{-2}$
Cl-36	0	$2.74 \cdot 10^{-1}$	$1.55 \cdot 10^{-4}$	Ho-166m	0.00	$1.28 \cdot 10^{-1}$	1.74
Fe-55	0	$4.20 \cdot 10^{-3}$	$1.69 \cdot 10^{-3}$	Pb-210	0.00	$3.80 \cdot 10^{-2}$	$4.81 \cdot 10^{-3}$
Co-60	0	$9.65 \cdot 10^{-2}$	2.50	Ra-226	4.86	$3.59 \cdot 10^{-3}$	$6.74 \cdot 10^{-3}$
Ni-59	0	$4.55 \cdot 10^{-3}$	$2.41 \cdot 10^{-3}$	Ac-227	$6.91 \cdot 10^{-2}$	$1.56 \cdot 10^{-2}$	$2.31 \cdot 10^{-4}$
Ni-63	0	$1.71 \cdot 10^{-2}$	0.00	Th-229	4.95	$1.14 \cdot 10^{-1}$	$9.54 \cdot 10^{-2}$
Se-79	0	$5.58 \cdot 10^{-2}$	0.00	Th-230	4.74	$1.46 \cdot 10^{-2}$	$1.55 \cdot 10^{-3}$
Sr-90	0	$1.96 \cdot 10^{-1}$	0.00	Th-232	4.07	$1.25 \cdot 10^{-2}$	$1.33 \cdot 10^{-3}$
Zr-93	0	$1.96 \cdot 10^{-2}$	0.00	Pa-231	5.04	$6.28 \cdot 10^{-2}$	$4.76 \cdot 10^{-2}$
Nb-93m	0	$2.84 \cdot 10^{-2}$	$1.91 \cdot 10^{-3}$	U-232	5.40	$1.74 \cdot 10^{-2}$	$2.19 \cdot 10^{-3}$
Nb-94	0	$1.68 \cdot 10^{-1}$	1.57	U-233	4.89	$6.08 \cdot 10^{-3}$	$1.31 \cdot 10^{-3}$
Mo-93	0	$5.54 \cdot 10^{-3}$	$1.07 \cdot 10^{-2}$	U-234	4.84	$1.32 \cdot 10^{-2}$	$1.73 \cdot 10^{-3}$
Tc-99	0	$1.01 \cdot 10^{-1}$	0.00	U-235	4.47	$4.80 \cdot 10^{-2}$	$1.54 \cdot 10^{-1}$
Ru-106	0	$1.00 \cdot 10^{-2}$	0.00	U-236	4.58	$1.14 \cdot 10^{-2}$	$1.57 \cdot 10^{-3}$
Pd-107	0	$9.26 \cdot 10^{-3}$	0.00	U-238	4.26	$1.00 \cdot 10^{-2}$	$1.36 \cdot 10^{-3}$
Ag-108m	0	$1.59 \cdot 10^{-2}$	1.62	Np-237	4.84	$6.85 \cdot 10^{-2}$	$3.43 \cdot 10^{-2}$
Cd-113m	0	$1.85 \cdot 10^{-1}$	0.00	Pu-238	5.58	$1.06 \cdot 10^{-2}$	$1.81 \cdot 10^{-3}$
Sn-126	0	$1.71 \cdot 10^{-1}$	$5.65 \cdot 10^{-2}$	Pu-239	5.23	$6.65 \cdot 10^{-3}$	$7.69 \cdot 10^{-4}$
Sb-125	0	$9.93 \cdot 10^{-2}$	$4.30 \cdot 10^{-1}$	Pu-240	5.24	$1.06 \cdot 10^{-2}$	$1.73 \cdot 10^{-3}$
Te-125m	0	$1.09 \cdot 10^{-1}$	$3.55 \cdot 10^{-2}$	Pu-241	$1.22 \cdot 10^{-4}$	$5.24 \cdot 10^{-3}$	$2.54 \cdot 10^{-6}$
I-129	0	$6.38 \cdot 10^{-2}$	$2.46 \cdot 10^{-2}$	Pu-242	4.97	$8.72 \cdot 10^{-3}$	$1.44 \cdot 10^{-3}$
Cs-134	0	$1.63 \cdot 10^{-1}$	1.55	Pu-244	4.65	$7.05 \cdot 10^{-3}$	$1.22 \cdot 10^{-3}$
Cs-135	0	$6.73 \cdot 10^{-2}$	0.00	Am-241	5.57	$5.19 \cdot 10^{-2}$	$3.24 \cdot 10^{-2}$
Cs-137	0	$1.87 \cdot 10^{-1}$	0.00	Am-242m	$2.52 \cdot 10^{-2}$	$4.39 \cdot 10^{-2}$	$5.11 \cdot 10^{-3}$
Ba-133	0	$5.38 \cdot 10^{-2}$	$4.01 \cdot 10^{-1}$	Am-243	5.36	$2.16 \cdot 10^{-2}$	$5.59 \cdot 10^{-2}$
Ba-137m	0	$6.51 \cdot 10^{-2}$	$5.96 \cdot 10^{-1}$	Cm-243	5.88	$1.37 \cdot 10^{-1}$	$1.34 \cdot 10^{-1}$
Pm-147	0	$6.20 \cdot 10^{-2}$	$4.37 \cdot 10^{-6}$	Cm-244	5.89	$8.59 \cdot 10^{-3}$	$1.70 \cdot 10^{-3}$
Sm-147	2.31	0.00	0.00	Cm-245	5.45	$6.50 \cdot 10^{-2}$	$9.55 \cdot 10^{-2}$
Sm-151	0	$1.97 \cdot 10^{-2}$	$1.34 \cdot 10^{-5}$	Cm-246	5.47	$8.00 \cdot 10^{-3}$	$1.51 \cdot 10^{-3}$

### 3.1 Estimation of nuclides that has to be accounted for in estimating the absorbed energy

As a first step in estimating the energy absorbed by the bitumen matrix, the energy emitted by each nuclide during an infinite long time is calculated. This corresponds to the maximum energy that a nuclide can emit for a given initial inventory. The emitted energy for each nuclide (sum of  $E_{\alpha}$ ,  $E_{\beta}$  and  $E_{\gamma}$ ) is recalculated to absorbed energy and the result is compiled in Table 5. The result in this table is based on activities given in Table 2 and the amount of bitumen shown in Table 4.

**Table 4** Amount of bitumen in different waste categories [kg/package].

Waste category				
B.05	B.06	F.05	F.17	F.18
150	150	95	820	960

**Table 5 Absorbed energy during an infinite time.**

Element	Half-life [yr]	Absorbed energy [Gy]				
		B.05	B.06	F.05	F.17	F.18
H-3	1.2·10 <sup>1</sup>	3.0·10 <sup>-3</sup>	2.1·10 <sup>-2</sup>	2.1·10 <sup>-3</sup>	5.7·10 <sup>-3</sup>	4.4·10 <sup>-2</sup>
Be-10	1.6·10 <sup>6</sup>	9.7·10 <sup>-2</sup>	7.0·10 <sup>-1</sup>	6.9·10 <sup>-2</sup>	1.9·10 <sup>-1</sup>	1.4
C-14	5.7·10 <sup>3</sup>	1.2·10 <sup>2</sup>	8.7·10 <sup>2</sup>	8.6·10 <sup>1</sup>	2.3·10 <sup>2</sup>	1.8·10 <sup>3</sup>
Cl-36	3.0·10 <sup>5</sup>	2.9·10 <sup>1</sup>	2.1·10 <sup>2</sup>	2.8	1.2·10 <sup>2</sup>	5.4·10 <sup>2</sup>
Fe-55	2.7	6.8	4.9·10 <sup>1</sup>	4.9	1.3·10 <sup>1</sup>	1.0·10 <sup>2</sup>
Co-60	5.3	5.8·10 <sup>3</sup>	4.2·10 <sup>4</sup>	4.2·10 <sup>3</sup>	1.1·10 <sup>4</sup>	8.6·10 <sup>4</sup>
Ni-59	7.5·10 <sup>4</sup>	2.2·10 <sup>2</sup>	1.6·10 <sup>3</sup>	1.6·10 <sup>2</sup>	4.3·10 <sup>2</sup>	3.3·10 <sup>3</sup>
Ni-63	9.6·10 <sup>1</sup>	1.4·10 <sup>2</sup>	1.1·10 <sup>3</sup>	1.0·10 <sup>2</sup>	2.8·10 <sup>2</sup>	2.1·10 <sup>3</sup>
Se-79	6.5·10 <sup>4</sup>	8.8	6.3·10 <sup>1</sup>	8.4·10 <sup>-1</sup>	3.7·10 <sup>1</sup>	1.6·10 <sup>2</sup>
Sr-90	2.9·10 <sup>1</sup>	2.0·10 <sup>1</sup>	1.4·10 <sup>2</sup>	1.9	8.3·10 <sup>1</sup>	3.7·10 <sup>2</sup>
Zr-93	1.5·10 <sup>6</sup>	1.3·10 <sup>1</sup>	9.2·10 <sup>1</sup>	9.1	2.4·10 <sup>1</sup>	1.9·10 <sup>2</sup>
Nb-93m	1.4·10 <sup>1</sup>	2.1·10 <sup>-1</sup>	1.5	1.5·10 <sup>-1</sup>	4.0·10 <sup>-1</sup>	3.1
Nb-94	2.0·10 <sup>4</sup>	1.5·10 <sup>2</sup>	1.1·10 <sup>3</sup>	1.1·10 <sup>2</sup>	2.9·10 <sup>2</sup>	2.2·10 <sup>3</sup>
Mo-93	3.5·10 <sup>3</sup>	1.4·10 <sup>-1</sup>	1.0	9.9·10 <sup>-2</sup>	2.7·10 <sup>-1</sup>	2.0
Tc-99	2.1·10 <sup>5</sup>	3.7·10 <sup>3</sup>	2.7·10 <sup>4</sup>	3.6·10 <sup>2</sup>	1.6·10 <sup>4</sup>	7.0·10 <sup>4</sup>
Ru-106	1.0	1.8·10 <sup>-3</sup>	1.3·10 <sup>-2</sup>	1.7·10 <sup>-4</sup>	7.5·10 <sup>-3</sup>	3.3·10 <sup>-2</sup>
Pd-107	6.5·10 <sup>6</sup>	2.1	1.5·10 <sup>1</sup>	2.0·10 <sup>-1</sup>	8.9	3.9·10 <sup>1</sup>
Ag-108m	1.3·10 <sup>2</sup>	1.7·10 <sup>1</sup>	1.3·10 <sup>2</sup>	1.2·10 <sup>1</sup>	3.3·10 <sup>1</sup>	2.6·10 <sup>2</sup>
Cd-113m	1.4·10 <sup>1</sup>	5.5·10 <sup>-2</sup>	3.9·10 <sup>-1</sup>	5.2·10 <sup>-3</sup>	2.3·10 <sup>-1</sup>	1.0
Sn-126	1.0·10 <sup>5</sup>	4.0·10 <sup>-1</sup>	2.8	3.8·10 <sup>-2</sup>	1.7	7.4
Sb-125	2.8	6.2·10 <sup>1</sup>	4.5·10 <sup>2</sup>	4.4·10 <sup>1</sup>	1.2·10 <sup>2</sup>	9.1·10 <sup>2</sup>
I-129	1.6·10 <sup>7</sup>	1.5·10 <sup>1</sup>	1.0·10 <sup>2</sup>	1.4	6.2·10 <sup>1</sup>	2.7·10 <sup>2</sup>
Cs-134	2.1	1.2·10 <sup>2</sup>	8.8·10 <sup>2</sup>	1.2·10 <sup>1</sup>	5.2·10 <sup>2</sup>	2.3·10 <sup>3</sup>
Cs-135	2.3·10 <sup>6</sup>	2.7·10 <sup>1</sup>	1.9·10 <sup>2</sup>	2.6	1.1·10 <sup>2</sup>	5.1·10 <sup>2</sup>
Cs-137	3.0·10 <sup>1</sup>	2.0·10 <sup>2</sup>	1.4·10 <sup>3</sup>	1.9·10 <sup>1</sup>	8.3·10 <sup>2</sup>	3.7·10 <sup>3</sup>
Ba-133	1.1·10 <sup>1</sup>	2.0·10 <sup>-2</sup>	1.5·10 <sup>-1</sup>	1.5·10 <sup>-2</sup>	3.9·10 <sup>-2</sup>	3.0·10 <sup>-1</sup>
Pm-147	2.6	5.1	3.6·10 <sup>1</sup>	4.9·10 <sup>-1</sup>	2.2·10 <sup>1</sup>	9.6·10 <sup>1</sup>
Sm-151	9.0·10 <sup>1</sup>	1.9·10 <sup>-1</sup>	1.3	1.8·10 <sup>-2</sup>	7.9·10 <sup>-1</sup>	3.5
Eu-152	1.3·10 <sup>1</sup>	4.2·10 <sup>-2</sup>	3.0·10 <sup>-1</sup>	4.0·10 <sup>-3</sup>	1.8·10 <sup>-1</sup>	7.9·10 <sup>-1</sup>
Eu-154	8.8	4.5·10 <sup>1</sup>	3.2·10 <sup>2</sup>	4.3	1.9·10 <sup>2</sup>	8.5·10 <sup>2</sup>
Eu-155	5.0	1.4	1.0·10 <sup>1</sup>	1.4·10 <sup>-1</sup>	6.1	2.7·10 <sup>1</sup>
Ho-166m	1.2·10 <sup>3</sup>	3.8	2.8·10 <sup>1</sup>	2.7	7.3	5.6·10 <sup>1</sup>
Pb-210	2.2·10 <sup>1</sup>	4.8·10 <sup>-15</sup>	3.5·10 <sup>-14</sup>	1.7·10 <sup>-15</sup>	1.0·10 <sup>-13</sup>	7.9·10 <sup>-13</sup>
Ra-226	1.6·10 <sup>3</sup>	3.9·10 <sup>-10</sup>	2.9·10 <sup>-9</sup>	1.4·10 <sup>-10</sup>	8.4·10 <sup>-9</sup>	6.4·10 <sup>-8</sup>
Ac-227	2.2·10 <sup>1</sup>	1.9·10 <sup>-12</sup>	1.4·10 <sup>-11</sup>	6.6·10 <sup>-13</sup>	4.0·10 <sup>-11</sup>	3.1·10 <sup>-10</sup>
Th-229	7.3·10 <sup>3</sup>	2.9·10 <sup>-9</sup>	2.1·10 <sup>-8</sup>	1.0·10 <sup>-9</sup>	6.1·10 <sup>-8</sup>	4.7·10 <sup>-7</sup>
Th-230	7.7·10 <sup>4</sup>	8.2·10 <sup>-6</sup>	5.9·10 <sup>-5</sup>	2.9·10 <sup>-6</sup>	1.7·10 <sup>-4</sup>	1.3·10 <sup>-3</sup>
Th-232	1.4·10 <sup>10</sup>	1.5·10 <sup>-6</sup>	1.1·10 <sup>-5</sup>	5.1·10 <sup>-7</sup>	3.1·10 <sup>-5</sup>	2.4·10 <sup>-4</sup>
Pa-231	3.3·10 <sup>4</sup>	1.3·10 <sup>-6</sup>	9.3·10 <sup>-6</sup>	4.5·10 <sup>-7</sup>	2.7·10 <sup>-5</sup>	2.1·10 <sup>-4</sup>
U-232	7.2·10 <sup>1</sup>	2.8·10 <sup>-6</sup>	2.1·10 <sup>-5</sup>	1.0·10 <sup>-6</sup>	6.0·10 <sup>-5</sup>	4.6·10 <sup>-4</sup>
U-233	1.6·10 <sup>5</sup>	3.9·10 <sup>-6</sup>	2.9·10 <sup>-5</sup>	1.4·10 <sup>-6</sup>	8.4·10 <sup>-5</sup>	6.4·10 <sup>-4</sup>
U-234	2.4·10 <sup>5</sup>	3.0·10 <sup>-1</sup>	2.2	1.1·10 <sup>-1</sup>	6.4	4.9·10 <sup>1</sup>
U-235	7.0·10 <sup>8</sup>	1.7·10 <sup>1</sup>	1.2·10 <sup>2</sup>	5.9	3.5·10 <sup>2</sup>	2.7·10 <sup>3</sup>
U-236	2.3·10 <sup>7</sup>	8.1	5.9·10 <sup>1</sup>	2.9	1.7·10 <sup>2</sup>	1.3·10 <sup>3</sup>
U-238	4.5·10 <sup>9</sup>	1.9·10 <sup>3</sup>	1.4·10 <sup>4</sup>	6.8·10 <sup>2</sup>	4.1·10 <sup>4</sup>	3.1·10 <sup>5</sup>
Np-237	2.1·10 <sup>6</sup>	1.1	7.8	3.8·10 <sup>-1</sup>	2.3·10 <sup>1</sup>	1.7·10 <sup>2</sup>
Pu-238	8.8·10 <sup>1</sup>	5.0·10 <sup>-1</sup>	3.6	1.8·10 <sup>-1</sup>	1.1·10 <sup>1</sup>	8.1·10 <sup>1</sup>
Pu-239	2.4·10 <sup>4</sup>	1.1·10 <sup>1</sup>	7.7·10 <sup>1</sup>	3.8	2.3·10 <sup>2</sup>	1.7·10 <sup>3</sup>
Pu-240	6.5·10 <sup>3</sup>	5.8	4.2·10 <sup>1</sup>	2.1	1.2·10 <sup>2</sup>	9.5·10 <sup>2</sup>
Pu-241	1.4·10 <sup>1</sup>	2.0·10 <sup>-3</sup>	1.4·10 <sup>-2</sup>	6.9·10 <sup>-4</sup>	4.2·10 <sup>-2</sup>	3.2·10 <sup>-1</sup>
Pu-242	3.8·10 <sup>5</sup>	1.4	1.0·10 <sup>1</sup>	5.0·10 <sup>-1</sup>	3.0·10 <sup>1</sup>	2.3·10 <sup>2</sup>
Pu-244	8.3·10 <sup>7</sup>	6.7·10 <sup>-5</sup>	4.8·10 <sup>-4</sup>	2.4·10 <sup>-5</sup>	1.4·10 <sup>-3</sup>	1.1·10 <sup>-2</sup>
Am-241	4.3·10 <sup>2</sup>	6.2·10 <sup>-1</sup>	4.5	2.2·10 <sup>-1</sup>	1.3·10 <sup>1</sup>	1.0·10 <sup>2</sup>
Am-242m	1.5·10 <sup>2</sup>	2.6·10 <sup>-5</sup>	1.9·10 <sup>-4</sup>	9.4·10 <sup>-6</sup>	5.6·10 <sup>-4</sup>	4.3·10 <sup>-3</sup>
Am-243	7.4·10 <sup>3</sup>	3.0·10 <sup>-1</sup>	2.2	1.1·10 <sup>-1</sup>	6.5	5.0·10 <sup>1</sup>
Cm-243	2.9·10 <sup>1</sup>	9.0·10 <sup>-4</sup>	6.6·10 <sup>-3</sup>	3.2·10 <sup>-4</sup>	1.9·10 <sup>-2</sup>	1.5·10 <sup>-1</sup>
Cm-244	1.8·10 <sup>1</sup>	8.1·10 <sup>-2</sup>	5.9·10 <sup>-1</sup>	2.9·10 <sup>-2</sup>	1.7	1.3·10 <sup>1</sup>
Cm-245	8.5·10 <sup>3</sup>	3.6·10 <sup>-3</sup>	2.6·10 <sup>-2</sup>	1.3·10 <sup>-3</sup>	7.7·10 <sup>-2</sup>	5.9·10 <sup>-1</sup>
Cm-246	4.7·10 <sup>3</sup>	5.2·10 <sup>-4</sup>	3.8·10 <sup>-3</sup>	1.9·10 <sup>-4</sup>	1.1·10 <sup>-2</sup>	8.6·10 <sup>-2</sup>

$^{60}\text{Co}$ ,  $^{99}\text{Tc}$  and  $^{238}\text{U}$  are the nuclides dominating the absorbed energy given in Table 5. However,  $^{238}\text{U}$  is very long-lived why the contribution is negligible for a time period up to at least  $10^4$  years. All other nuclides in the chains  $4N$ ,  $4N+1$ ,  $4N+2$  and  $4N+3$  (written in *Italics* in Table 5) give a negligible contribution to the total absorbed energy. Many of the other nuclides in Table 5 can also be neglected. Nine nuclides ( $^{14}\text{C}$ ,  $^{60}\text{Co}$ ,  $^{63}\text{Ni}$ ,  $^{94}\text{Nb}$ ,  $^{99}\text{Tc}$ ,  $^{125}\text{Sb}$ ,  $^{134}\text{Cs}$ ,  $^{137}\text{Cs}$  and  $^{154}\text{Eu}$ ) give together about 98 % or more of the integrated absorbed energy for times up to  $10^4$  years.

Energy emitted by daughters has not been accounted for in the data presented in Table 5 since chain decay of nuclides makes estimation of absorbed energy somewhat complicated. For example, each mole of  $^{244}\text{Pu}$  decays to one mole of  $^{240}\text{U}$  that in turn decays to one mole of  $^{240\text{m}}\text{Np}$  that decays to  $^{240}\text{Pu}$  (see Figure 1). The half-life of these three daughters is significantly shorter than that of  $^{244}\text{Pu}$ . The next nuclide in the chain,  $^{236}\text{U}$ , has a half-life that is comparable to that of  $^{244}\text{Pu}$ , but the following nuclide,  $^{232}\text{Th}$ , has a significantly longer half-life. It is therefore reasonable to assume that  $^{240}\text{U}$ ,  $^{240\text{m}}\text{Np}$ ,  $^{240}\text{Pu}$  and  $^{236}\text{U}$  formed by decay of  $^{244}\text{Pu}$ , decay simultaneously as  $^{244}\text{Pu}$ . Consequently, the energy emitted for each decay of  $^{244}\text{Pu}$  is the sum of the energy emitted by  $^{244}\text{Pu}$ ,  $^{240}\text{U}$ ,  $^{240\text{m}}\text{Np}$ ,  $^{240}\text{Pu}$  and  $^{236}\text{U}$ . Thus, in order to estimate the energy absorbed due to radioactive decay of nuclide in chains, the energy emitted as a nuclide decay is the sum of the energy emitted by the nuclide in question and of all its daughters having a half-life shorter than or comparable to the nuclide in question. This is summarised in Table 6.

**Table 6**      **Compilation of nuclides whose energy is added to the mother nuclide.**

Mother	Daughter nuclides whose energy is added to the mother nuclide
$^{90}\text{Sr}$	$^{90}\text{Y}$
$^{93}\text{Zr}$	$^{93\text{m}}\text{Nb}$
$^{93}\text{Mo}$	$^{93\text{m}}\text{Nb}$
$^{126}\text{Sn}$	$^{126\text{m}}\text{Sb}$ , $^{126}\text{Sb}$
$^{125}\text{Sb}$	$^{125\text{m}}\text{Te}$
$^{137}\text{Cs}$	$^{137\text{m}}\text{Ba}$
$^{244}\text{Pu}$	$^{240}\text{U}$ , $^{240\text{m}}\text{Np}$ , $^{240}\text{Pu}$ , $^{236}\text{U}$
$^{232}\text{Th}$	$^{228}\text{Ra}$ , $^{228}\text{Ac}$ , $^{228}\text{Th}$ , $^{224}\text{Ra}$ , $^{220}\text{Rn}$ , $^{216}\text{Po}$ , $^{212}\text{Pb}$ , $^{212}\text{Bi}$ , $^{212}\text{Po}$ , $^{208}\text{Tl}$
$^{232}\text{U}$	$^{228}\text{Th}$ , $^{224}\text{Ra}$ , $^{220}\text{Rn}$ , $^{216}\text{Po}$ , $^{212}\text{Pb}$ , $^{212}\text{Bi}$ , $^{212}\text{Po}$ , $^{208}\text{Tl}$
$^{245}\text{Cm}$	$^{241}\text{Pu}$ , $^{241}\text{Am}$ and $^{237}\text{U}$
$^{241}\text{Am}$	$^{237}\text{U}$
$^{237}\text{Np}$	$^{233}\text{Pa}$ , $^{233}\text{U}$ , $^{229}\text{Th}$ , $^{225}\text{Ra}$ , $^{225}\text{Ac}$ , $^{221}\text{Fr}$ , $^{217}\text{At}$ , $^{213}\text{Bi}$ , $^{213}\text{Po}$ , $^{209}\text{Tl}$ , $^{209}\text{Pb}$
$^{233}\text{U}$	$^{229}\text{Th}$ , $^{225}\text{Ra}$ , $^{225}\text{Ac}$ , $^{221}\text{Fr}$ , $^{217}\text{At}$ , $^{213}\text{Bi}$ , $^{213}\text{Po}$ , $^{209}\text{Tl}$ , $^{209}\text{Pb}$
$^{229}\text{Th}$	$^{225}\text{Ra}$ , $^{225}\text{Ac}$ , $^{221}\text{Fr}$ , $^{217}\text{At}$ , $^{213}\text{Bi}$ , $^{213}\text{Po}$ , $^{209}\text{Tl}$ , $^{209}\text{Pb}$
$^{242\text{m}}\text{Am}$	$^{242}\text{Am}$ , $^{242}\text{Cm}$ , $^{238}\text{Pu}$
$^{238}\text{U}$	$^{234}\text{Th}$ , $^{234\text{m}}\text{Pa}$ , $^{234}\text{U}$ , $^{230}\text{Th}$ , $^{226}\text{Ra}$ , $^{222}\text{Rn}$ , $^{218}\text{Po}$ , $^{214}\text{Pb}$ , $^{214}\text{Bi}$ , $^{214}\text{Po}$ , $^{210}\text{Pb}$ , $^{210}\text{Bi}$ , $^{210}\text{Po}$
$^{234}\text{U}$	$^{230}\text{Th}$ , $^{226}\text{Ra}$ , $^{222}\text{Rn}$ , $^{218}\text{Po}$ , $^{214}\text{Pb}$ , $^{214}\text{Bi}$ , $^{214}\text{Po}$ , $^{210}\text{Pb}$ , $^{210}\text{Bi}$ , $^{210}\text{Po}$
$^{230}\text{Th}$	$^{226}\text{Ra}$ , $^{222}\text{Rn}$ , $^{218}\text{Po}$ , $^{214}\text{Pb}$ , $^{214}\text{Bi}$ , $^{214}\text{Po}$ , $^{210}\text{Pb}$ , $^{210}\text{Bi}$ , $^{210}\text{Po}$
$^{226}\text{Ra}$	$^{222}\text{Rn}$ , $^{218}\text{Po}$ , $^{214}\text{Pb}$ , $^{214}\text{Bi}$ , $^{214}\text{Po}$ , $^{210}\text{Pb}$ , $^{210}\text{Bi}$ , $^{210}\text{Po}$
$^{210}\text{Pb}$	$^{210}\text{Bi}$ , $^{210}\text{Po}$
$^{243}\text{Am}$	$^{239}\text{Np}$ , $^{239}\text{Pu}$
$^{235}\text{U}$	$^{231}\text{Th}$ , $^{231}\text{Pa}$ , $^{227}\text{Ac}$ , $^{227}\text{Th}$ , $^{223}\text{Fr}$ , $^{223}\text{Ra}$ , $^{219}\text{Rn}$ , $^{215}\text{Po}$ , $^{211}\text{Pb}$ , $^{211}\text{Bi}$ , $^{211}\text{Po}$ , $^{207}\text{Tl}$
$^{231}\text{Pa}$	$^{227}\text{Ac}$ , $^{227}\text{Th}$ , $^{223}\text{Fr}$ , $^{223}\text{Ra}$ , $^{219}\text{Rn}$ , $^{215}\text{Po}$ , $^{211}\text{Pb}$ , $^{211}\text{Bi}$ , $^{211}\text{Po}$ , $^{207}\text{Tl}$
$^{227}\text{Ac}$	$^{227}\text{Th}$ , $^{223}\text{Fr}$ , $^{223}\text{Ra}$ , $^{219}\text{Rn}$ , $^{215}\text{Po}$ , $^{211}\text{Pb}$ , $^{211}\text{Bi}$ , $^{211}\text{Po}$ , $^{207}\text{Tl}$

Taking decay of daughters into account, the energy absorbed by bitumen will for some nuclides increase by as much as between two and three orders of magnitude in comparison to the data given in Table 5. However, it is still the nine nuclides specified above that dominate the energy absorbed by bitumen.

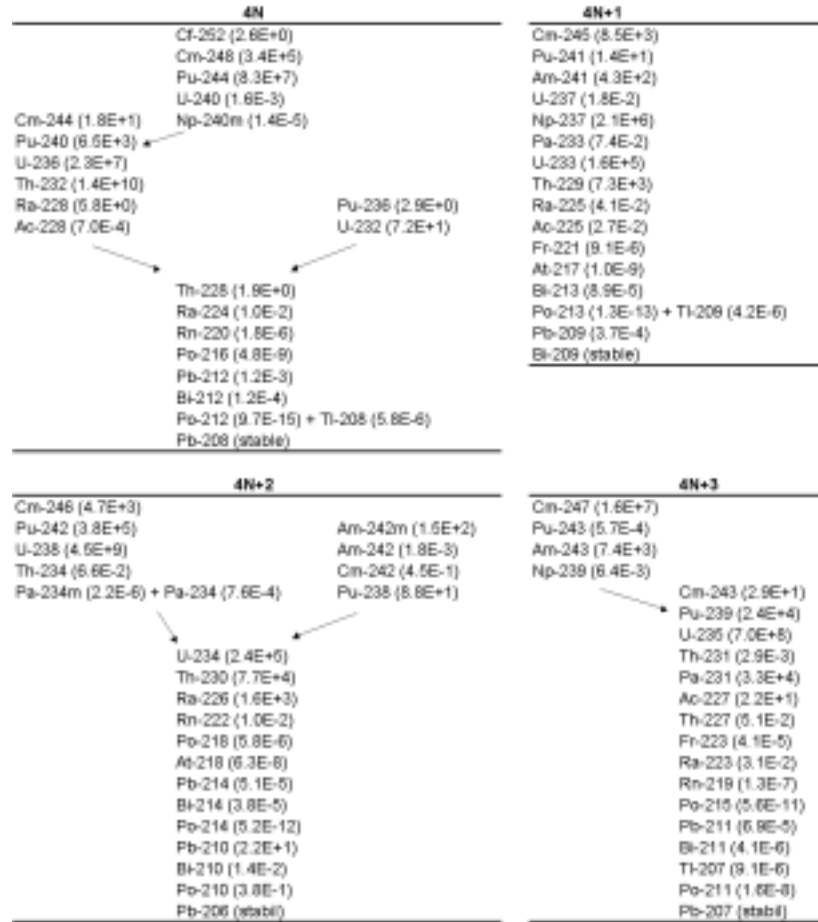


Figure 1 Decay chains

### 3.2 Estimation of nuclide specific energies emitted

Of the nine nuclides giving a significant contribution to the absorbed energy,  $^{137}\text{Cs}$  is the only radionuclide that decays to another radionuclide ( $^{137\text{m}}\text{Ba}$ ).  $^{137\text{m}}\text{Ba}$  has a half-life that is significantly shorter than that of  $^{137}\text{Cs}$ . The mother nuclide and the daughter nuclide will therefore decay more or less simultaneously. It is therefore reasonable to neglect the daughter except for that the energy emitted by the daughter is added to its mother. 94.6 % of  $^{137}\text{Cs}$  decays to  $^{137\text{m}}\text{Ba}$  that emits both  $\beta$ - and  $\gamma$ -radiation when it decays. The  $\beta$ -energy emitted by  $^{137}\text{Cs}$  is therefore assumed to be:

$$E_{\beta}^{Cs-137} = E_{\beta}^{Cs-137} + 0.946 \cdot E_{\beta}^{Ba-137\text{m}}$$

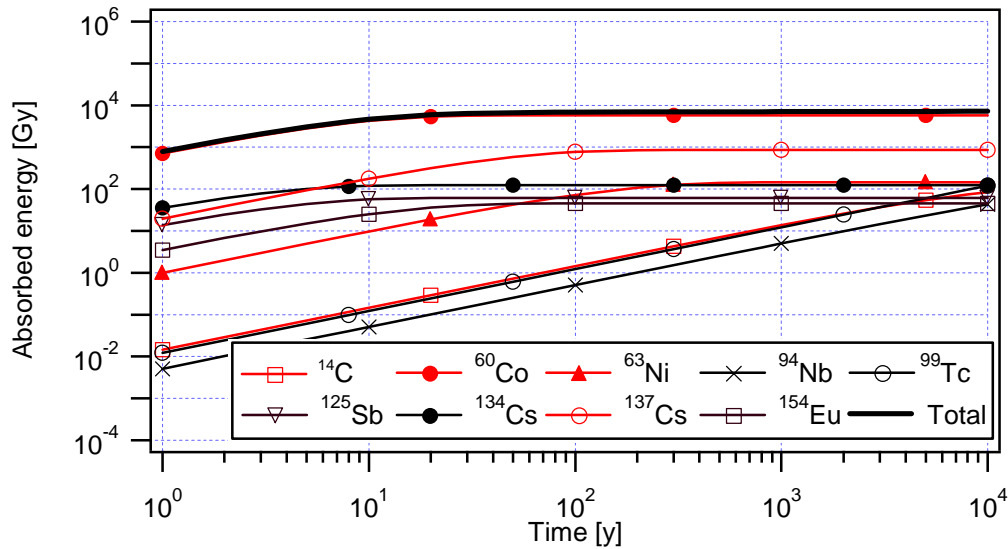
The  $\gamma$ -energy is calculated in the same way replacing  $E_{\beta}$  with  $E_{\gamma}$ .

**Table 7** Total energy emitted by  $^{137}\text{Cs}$  (MeV/decay).

nuclide	$E_\alpha$	$E_\beta$	$E_\gamma$	$E_{\beta, \text{tot}}$	$E_{\gamma, \text{tot}}$
$^{137}\text{Cs}$	0	$1.87 \cdot 10^{-1}$	0	$2.49 \cdot 10^{-1}$	$5.64 \cdot 10^{-1}$
$^{137\text{m}}\text{Ba}$	0	$6.51 \cdot 10^{-2}$	$5.96 \cdot 10^{-1}$		

### 3.3 Estimation of absorbed energy for waste in SFR 1

The amount of energy absorbed by bitumen due to radiolytic decay of  $^{14}\text{C}$ ,  $^{60}\text{Co}$ ,  $^{63}\text{Ni}$ ,  $^{94}\text{Nb}$ ,  $^{99}\text{Tc}$ ,  $^{125}\text{Sb}$ ,  $^{134}\text{Cs}$ ,  $^{137}\text{Cs}$  and  $^{154}\text{Eu}$  has been calculated for the bitumenised waste deposited in Silo and BMA. The calculations are based on the inventory given in Table 2, the amount of bitumen shown in Table 4, the decay energies compiled in Table 3 and Table 7 and the assumption that all energy is absorbed by the bitumen. The absorbed energy is plotted in Figure 2 to Figure 6 as a function of time.



*Figure 2* Absorbed energy due to irradiation of bitumen in B.05 as a function of time.

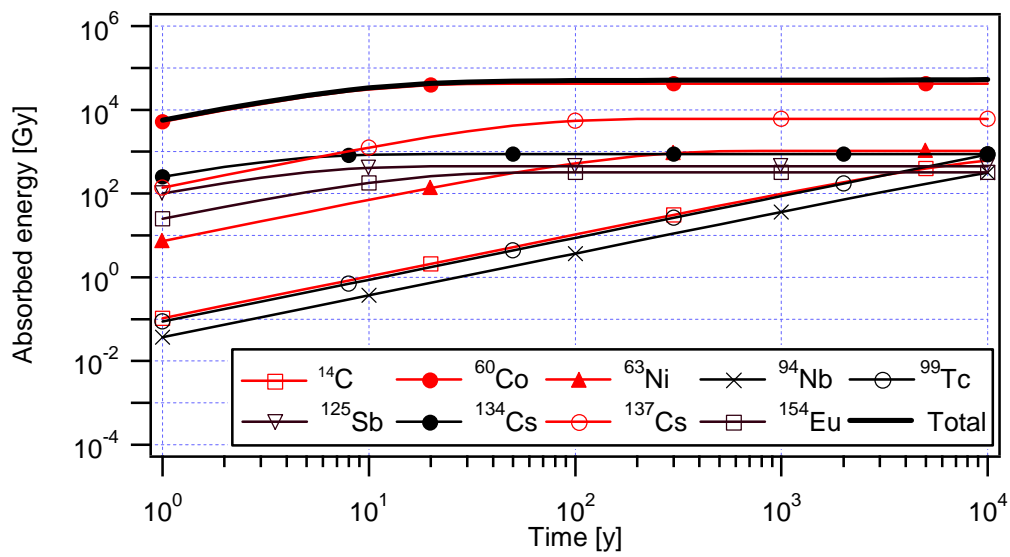


Figure 3 Absorbed energy due to irradiation of bitumen in B.06 as a function of time.

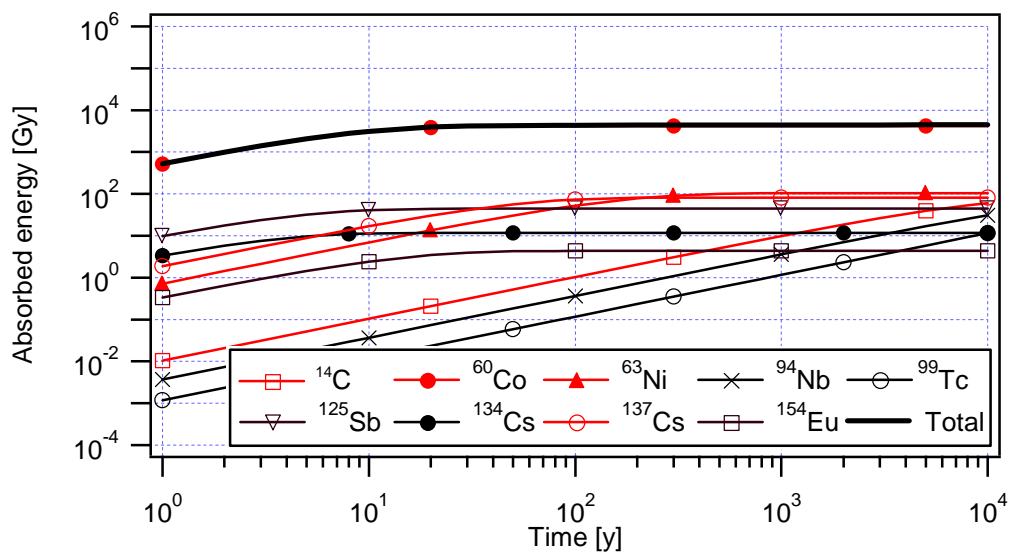


Figure 4 Absorbed energy due to irradiation of bitumen in F.05 as a function of time.



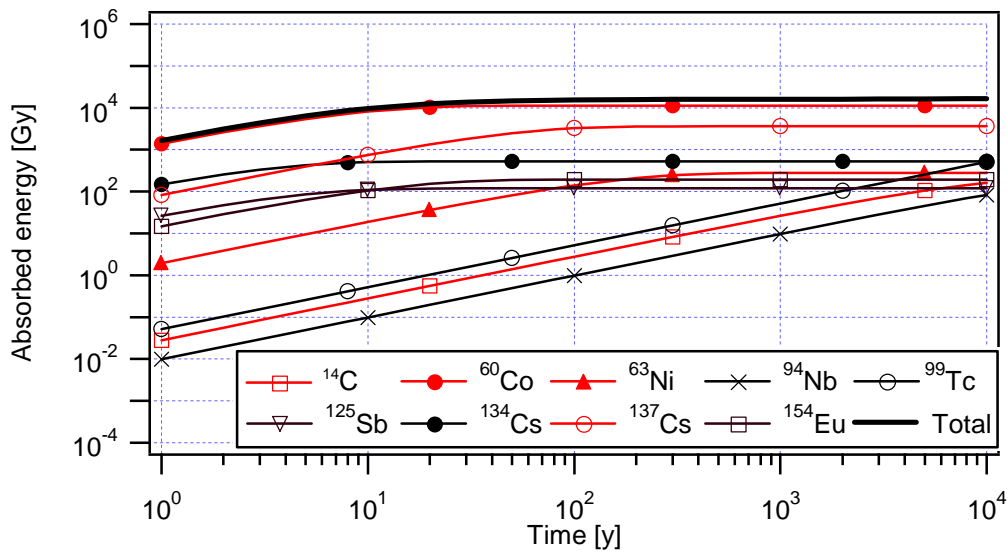


Figure 5 Absorbed energy due to irradiation of bitumen in F.17 as a function of time.

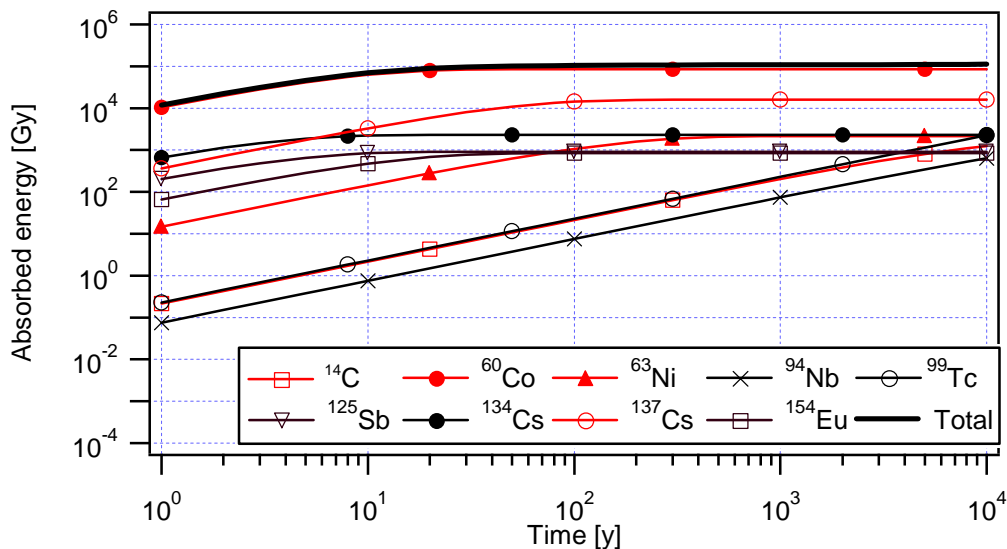


Figure 6 Absorbed energy due to irradiation of bitumen in F.18 as a function of time.

$^{60}\text{Co}$  is the dominating nuclide in terms of energy absorbed by bitumen, and is in general followed by  $^{137}\text{Cs}$ .

It has been concluded that negligible effects on gas generation, swelling, hardening, heating and decrease in leach resistance are found when the absorbed energy is less than 0.1 MGy (Eschrich, 1980). For energies about 2 – 10 MGy and higher, a substantial swelling can take place and most likely also an increased leachability. The categories B.05 and F.05 deposited in BMA correspond to an integrated absorbed energy that is less than  $10^4$  Gy within a period of  $10^4$  years. Decay of radionuclides in F.17, also deposited in BMA, results in a total absorbed energy of  $2 \cdot 10^4$  Gy. For B.06 and F.18 deposited in the Silo the total absorbed energy is estimated to  $5 \cdot 10^4$  Gy and  $1 \cdot 10^5$  Gy, respectively.

## 4 Discussion

The estimated absorbed energy is based on the assumption of an infinite media. Even though this is a reasonable assumption for the waste packages disposed of in SFR 1, the results for nuclides like  $^{60}\text{Co}$  and  $^{137}\text{Cs}$  are overestimated. Porath *et al.* (1981) compare the absorbed energy obtained for an infinite media with those for a finite media. They conclude that for nuclides emitting higher  $\beta$  energies, the absorbed energy is overestimated by up to 60 % assuming an infinite media. This is based on the results for  $^{60}\text{Co}$ . For  $^{137}\text{Cs}$  the absorbed energy is overestimated by 33 %.

The analysis presented in this appendix is based on an estimated mean value of the radionuclide activity in newly embedded waste (Riggare and Johansson, 2001). Using the database on waste already produced and deposited in SFR 1 (TRIUMF, 2000) a rough estimate indicates that the content of  $^{60}\text{Co}$  corresponds to an absorbed energy exceeding  $10^5$  Gy in a few percent of the packages in category B.05 and B.06. This indicates that the properties of at least some packages in these categories can be affected by the internal radiation. For F.18 the corresponding factor is about fifty percent.

The analysis made here does not include waste categories B.20 and F.20 stored in BLA. These two categories contain waste similar to B.05 and F.05, respectively, but with a lower radionuclide content. From the estimated results for B.05 and F.05 it is concluded that the absorbed energy for waste categories B.20 and F.20 will be well below 0.1 MGy.

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**Appendix B: Rough estimate of the  
consequence of swelling of  
bitumenised waste in SFR 1  
caused by water uptake**

# 1 Background

Water uptake in bitumenised waste leads to swelling of the bitumen matrix. This in turn may affect the packaging in which the matrix is contained or other barriers surrounding the waste packages (e.g. the concrete structure in BMA). A rough estimate of the swelling of bitumenised waste deposited in SFR 1 has therefore been made.

## 2 Estimate of the theoretical maximum volume increase caused by swelling

The theoretical maximum volume increase due to water uptake of ion exchange resins and evaporator concentrate in the waste has been estimated (Table 1). The estimation is based on the waste content in a reference package defined for each waste category (Riggare and Johansson, 2001). For ion exchange resins a maximum volume increase of a factor of two is assumed (Nilsson *et al.*, 1988) and for evaporator concentrate a factor of 3.5 corresponding to the volume increase when dry  $\text{NaNO}_3$  is dissolved and a saturated solution of  $\text{NaNO}_3$  is formed. The mass of waste given for a reference package (Riggare and Johansson, 2001) is recalculated to volumes assuming a density of  $1,030 \text{ kg/m}^3$  for bitumen and  $1,400 \text{ kg/m}^3$  for ion exchange resins. The density of dry ion exchange resins examined by Nilsson *et al.* (1988) is between  $1,140$  and  $1,520 \text{ kg/m}^3$ . Berntsson (1992) specifies the density of dry ion exchange resin to between  $1230$  and  $1500 \text{ kg/m}^3$ . The density of evaporator concentrate is approximated with the density of dry sodium sulfate, which is  $2,680 \text{ kg/m}^3$  (Weast, 1985).

In order to get an estimate of the consequence of swelling of bitumenised waste, the maximum theoretical volume increase is compared with the available voids within the different waste packages. The initial void in the package (not including the porosity of the materials) is given in Table 1. Drums stored in the silo are grouped in four on a metal plate and a metal box is inserted in the middle of the plate (see Figure 1). The metal box has a volume of 42 litres (Roth, 1996). When the waste is backfilled with concrete the volume inside the metal box will form an additional void equal to the volume of the box. The void given in Table 1 for a drum in category B.06 includes the void in a drum as well as one fourth of the volume of a metal box.

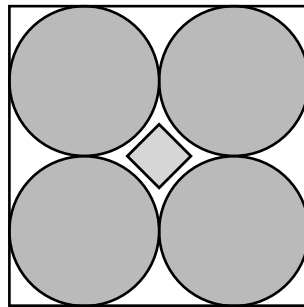


Figure 1 Schematic figure of drums and metal box on a plate seen from above.

**Table 1 Total void and volume increase due to water uptake for a reference package in each waste category with bitumenised waste.**

Waste category	Repository part	Waste packaging	Waste		Void [m <sup>3</sup> ]	Volume increase [m <sup>3</sup> ]
			[kg]	[m <sup>3</sup> waste/ m <sup>3</sup> bitumen product]		
B.05	BMA	drum	50	0.2	0.032	0.036
B.06	Silo	drum	50	0.2	0.043 <sup>a)</sup>	0.036
B.20 <sup>b)</sup>	BLA	drums in ISO-container	50	0.2	0.25 <sup>c)</sup>	0.036
F.05	BMA	drum	130	0.5	0.022	0.093
F.17	BMA	1.7 m <sup>3</sup> container	650 + 120	0.4	0.085	0.58
F.18	Silo	1.7 m <sup>3</sup> container	600	0.3	0.17	0.43
F.20 <sup>b)</sup>	BLA	drums in ISO-container	130	0.5	0.24 <sup>c)</sup>	0.093

a) Void includes void in one drum and ¼ of the void in a metal box.

b) Data is given for one drum.

c) Void includes void in one drum and 1/36 of the void in an ISO-container.

According to the data given in Table 1, the volume increase for bitumenised waste stored in BLA is well below the available void inside an ISO container.

For B.05, F.05 and F.17 stored in BMA the volume increase is comparable to or larger than the available void inside the packages. About 25 % of the total volume available for waste deposition in BMA is foreseen to consist of bitumenised waste, half of it is F.17. The estimated volume increase indicates that the concrete structure located inside BMA should not be backfilled. If it is, it can not be ruled out that the pressure built up inside the structure when the bitumenised waste swell will affect other barriers in BMA (e.g. concrete walls of the structure).

The swelling of waste in category B.06 stored in the silo is estimated to be comparable to the available void inside the drum and the expansion box deposited together with the drums. For category F.18 on the other hand, the volume increase is roughly twice the available void. The amount of waste in F.18 to be deposited in the silo is estimated to be only a few per cent of the total volume inside the silo. The bitumenised waste is, however, concentrated to the central part of the silo and can therefore locally constitute a significant fraction of the waste. Local effects due to swelling can therefore arise.

The amount of waste varies somewhat between different packages due to variations in the bitumenisation process at Barsebäck (Roth, 1996). In waste produced until 1992 the ratio of dry ion exchange resin and bitumen varies between 1:1.5 kg/kg and 1:14 kg/kg. The bitumenisation process was reconstructed, and from 1993 onwards this ratio is between 1:2.3 kg/kg and 1:5 kg/kg. The maximum theoretical volume increase for these mixing ratios are given in Table 2, and should be compared with the available void for B.05, B.06 and B.20 given in Table 1. It can be concluded that for the maximum ratio of

resin and bitumen obtained before the reconstruction, the estimated volume increase exceeds the available void for both B.05 and B.06 but not for B.20. After the reconstruction the maximum volume increase is still exceeding the void for B.05 but for B.06 it is comparable to the available void. This indicates that swelling of waste in categories B.05 and B.06, especially packages produced before 1993, can be a problem.

**Table 2 Estimated volume increase based on minimum and maximum ratio of bitumen and ion-exchange resins at Barsebäck.**

Mixing ratio resin/bitumen [kg/kg]	Waste		Volume increase [m <sup>3</sup> ]
	[kg]	[m <sup>3</sup> waste/ m <sup>3</sup> bitumen product]	
1:14	13	0.05	0.009
1:1.5	85	0.33	0.060
1:5	33	0.13	0.024
1:2.3	62	0.24	0.045

Variations in waste content between different packages within a waste category produced at Forsmark NPP exist as well. However, the waste content in the reference package used for the results presented in Table 1 for F.05, F.18 and F.20 is close to the upper limit. The estimated volume increase given in Table 1 for these three categories therefore gives an indication of the upper limit. Category F.17 may, however, contain some 50 volume % of waste in comparison to the 40 volume % in the reference package, and thus the volume increase could be even larger than that given in Table 1.

The data on maximum swelling presented by Nilsson *et al.* (1988) is related to dry resins. After bitumenisation, the resins are not completely dry but contain some residual water, indicating that the maximum swelling will be less than this theoretical estimate. In addition, the data given by Nilsson and co-workers are based on experiments using new resins and not on resins taken from any nuclear power plant, and the resins are not included in a bitumen matrix. Aittola and Kleveland (1982) have shown that there is a significant variation in swelling between different samples of bitumenised resins. The actual swelling of samples of bitumenised resins varies from a few percent to more than 100 % of the theoretical swelling of pure ion-exchange resins. The highest swelling is obtained with the highest mixing ratio (55 %). Their data also indicate that irradiation of the resin increases the swelling significantly, i.e. 'real' bitumen/resin mixtures could swell more than in 'laboratory mixtures'.

It is important to note that this is a theoretical estimation of the swelling. Swelling is very much dependent on the type of resin (cation or anion, powdered or bead resin, degree of cross-linking (i.e. DVB content) etc), the loading of the resin and pre-treatment of the resin. Residual water in the bitumenised resin prevents observable swelling (IAEA, 1993). Thus, a possible swelling will be closely related to the used waste process. It is therefore difficult to estimate the swelling without making tests on the actual resin in the specific process it will be used.



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## **Appendix C: Estimation of transport times of radionuclides in BMA**

It is unclear if the ability of the bitumen matrix to retain nuclides is of importance for the release rate of radionuclides from SFR 1 or if the matrix can be neglected. The bitumen matrix is only of interest if the nuclides are retained for times comparable to or longer than the time needed for the nuclides to migrate through subsequent barriers. A rough estimate of the time needed for the nuclides to be transport through BMA has therefore been made.

The analysis is based on the assumption that the water flow is directed upwards through BMA and that the volume inside the encapsulation not occupied by waste packages is not backfilled, i.e. consists of water. This water flow is in accordance with the conditions foreseen to prevail at the time of closure of SFR 1 (Holmén and Stigsson, 2001a). The water flow will be more and more horizontally directed with time. It is, however, reasonable to assume a vertical flow for the first 1,000 years from repository closure.

The resistance for diffusive transport and for convective transport at steady-state has been calculated. The resistance for diffusive transport is given by the ratio of diffusion length and effective diffusivity ( $L/D_e$ ) and for convective transport by the inverse of the water velocity ( $1/u$ ). The convective transport is based on data given in Holmén and Stigsson (2001b). The total flow through the different barriers in BMA today (2000 AD) and at 3000 AD is compiled in Table 1. Based on this, the total flow given in Table 2 has been assumed. It is also assumed that no water is flowing through the waste packages or in the walls of the encapsulation. This is a reasonable assumption since the waste packages and the walls will have a much lower hydraulic conductivity than the unfilled space inside the encapsulation.

The cross section area (Table 2) is estimated from data given in Holmén and Stigsson (2001b) except for 'Water around waste packages'. The area for the water is estimated from the floor area of the encapsulation in BMA and the number of containers placed on the floor. Diffusion length is with one exception based on data in Appendix A6-6 in SKB (1993). The exception is 'Water around waste packages' for which the diffusion length is calculated from the volume of water and an estimation of the contact area between waste packages and water.

**Table 1 Total water flow through the different barriers in BMA (Holmén and Stigsson, 2001b).**

	Total flow [m <sup>3</sup> /year]	
	2000 AD	3000 AD
Top filling	5.9	20.6
Waste encapsulation	0.07	0.13
Filling at sides	6.8	14.9
Sand floor	7.7	12.8
Loading areas	2.8	22.4

**Table 2 Water flow, area, diffusion length, diffusivity and porosity used in this analysis.**

	Total flow [m <sup>3</sup> /year]	Area [m <sup>2</sup> ]	Diffusion length [m]	Effective diffusivity [m/s]	Porosity [-]
Top filling (gravel)	10	2700	6	6·10 <sup>-10</sup>	0.3
Filling at sides (gravel)	10	270	4.5	6·10 <sup>-10</sup>	0.3
Ceiling of construction	0.1	2160	0.8	1·10 <sup>-11</sup>	0.15
Walls of construction	0.0	–	0.4	1·10 <sup>-11</sup>	0.15
Water around waste packages	0.1	100	1.4·10 <sup>-2</sup>	2·10 <sup>-9</sup>	1

The estimated water velocity and resistance is given in Table 3. A comparison of the convective and the diffusion resistance as calculated in this analysis shows that the main transport mechanism through the encapsulation and inside it (in the water) is diffusion. In the gravel backfill between the rock and the sides of the encapsulation the migration is dominated by convection. In top filling the resistance is of the same order of magnitude for both mechanisms.

**Table 3 Estimated resistance for convective and diffusive transport.**

	Pore velocity [m/year]	Resistance [year/m]	
		1/u	L/D <sub>e</sub>
Top filling (gravel)	1.2·10 <sup>-2</sup>	8.1·10 <sup>1</sup>	9.5·10 <sup>1</sup>
Filling at sides (gravel)	1.2·10 <sup>-1</sup>	8.1	7.1·10 <sup>1</sup>
Ceiling of construction	3.1·10 <sup>-4</sup>	3.2·10 <sup>3</sup>	3.8·10 <sup>2</sup>
Walls of construction	0.0	–	1.9·10 <sup>2</sup>
Water around waste packages	9.9·10 <sup>-4</sup>	1.0·10 <sup>3</sup>	2.2·10 <sup>-1</sup>

From the estimated resistances and transport lengths (diffusion length), the time needed to flow and diffuse through the different barriers have been calculated. The results are shown in Table 4. From these results, the total time needed to transport nuclides from the source term to the surrounding rock can be evaluated. The short transport length assumed for the water inside the encapsulation results in a transport time being negligible in comparison to that through the other barriers. The total transport time for nuclides migrating through the roof of the encapsulation is 7.9·10<sup>2</sup> years (3.0·10<sup>2</sup> + 4.9·10<sup>2</sup>). Transport through the walls of the encapsulation takes 6.0·10<sup>2</sup> years (7.6·10<sup>1</sup> + 3.6·10<sup>1</sup> + 4.9·10<sup>2</sup>).

These results indicate that it takes 600 – 800 years to transport radionuclides through BMA. Retarding mechanisms such as sorption of radionuclides have not been taken into account. The resistance within the waste package is also neglected.

**Table 4 Estimated transport time.**

	Transport time [years]	
	Convection	Diffusion
Top filling (gravel)	$4.9 \cdot 10^2$	$5.7 \cdot 10^2$
Filling at sides (gravel)	$3.6 \cdot 10^1$	$3.2 \cdot 10^2$
Ceiling of construction	$2.6 \cdot 10^3$	$3.0 \cdot 10^2$
Walls of construction	–	$7.6 \cdot 10^1$
Water around waste packages	14	$3.2 \cdot 10^{-3}$

The discussion above is valid for steady-state conditions. However, it is possible that the initial, non-stationary phase of the release of nuclides from the repository is long in comparison to the transport time at stationary conditions. A measure of the non-stationary transport is the break-through time. This is usually defined as the time needed to obtain a concentration on the outside of a body that is 5 % of that on the inside. The non-stationary diffusion through a finite slab is evaluated in order to get a measure of the break-through time for the non-stationary transport through the encapsulation. The solution to this problem is given in for instance Crank (1975) and Bird *et al.*, (1960).

The break-through time for the different parts of the encapsulation is given in Table 5. These times are in the same order of magnitude as the transport times estimated for diffusion under steady-state conditions.

**Table 5 Break-through time for the encapsulation.**

	Break-through time [years]
Ceiling of construction	$2.0 \cdot 10^2$
Walls of construction	$5.1 \cdot 10^1$

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