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# **Project SAFE**

## **Complexing agents in SFR**

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Kemakta Konsult AB

January 2001

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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

Low- and intermediate level radioactive waste, produced at Swedish nuclear power plants, will be deposited in an underground repository, SFR. Different substances in the waste or in degradation products emanating from the waste, and chemicals added during the building of cementitious barriers in the repository, may exhibit complexing properties. The complexation of radionuclides with such ligands may increase the mobility of the deposited radionuclides as sorption on the cement phases is decreased and solubility increased. This could lead to an increased leaching of the radionuclides from the repository to the geosphere and biosphere. To be able to evaluate the implications for the function and long-term safety of the repository a study has been performed on complexants in SFR. The study is a part of project SAFE (Safety Assessment of Final Repository for operational Radioactive Waste) at the Swedish Nuclear Fuel and Waste Management Co, SKB. Concentrations of complexants were calculated in different waste types in the repository and compared to critical levels above which radionuclide sorption may be affected. The analysis is based on recent research presented in international and national literature sources.

The waste in SFR that may act or give rise to substances with complexing properties mainly consists of cellulose materials, including cement additives used in waste conditioning and backfill grout. The radioactive waste also contains chemicals mainly used in decontamination processes at the nuclear power plants, e.g. EDTA, NTA, gluconate, citric acid and oxalic acid.

The calculations performed in this report show that the presence of complexants in SFR (ISA and other substances) may lead to a sorption reduction for some radionuclides in certain waste types. This may have to be considered when performing calculations of the radionuclide transport.

Concentration calculations of isosaccharinic acid (ISA), using a degradation yield of 0.1 mole/kg cellulose (2%), showed that the limit above which effects on sorption of radionuclides is apparent ( $>10^{-4}$  M) is exceeded mainly in three waste types in BMA (F.17, F.23/steel and S.21) and in one waste type in the Silo (S.22). The relatively high level of ISA is in part due to the fact that these waste types are not conditioned in cement (F.17 is bituminised), thus there is no sorption to cement. Both bi- tri- and tetravalent elements will probably be affected (S.21 and S.22). It should be pointed out that ISA will only be produced in these waste types if the integrity of the moulds is not maintained and alkaline water enters. Degradation of ISA has not been regarded in the calculations. Other conservative approaches have been adopted in the calculations, thus the concentration of ISA may be somewhat overestimated.

Calculations assuming a higher yield of ISA, corresponding to 1000 years of degradation (about ~2.5%), lead to relatively small changes in concentration. Calculations using a yet higher yield (10%), which corresponds to about 10 000 years of degradation, showed that critical levels of ISA may arise in all waste types. This is however a highly conservative approach.

Due to the presence of NTA, citric acid, sodium capryliminodipropionate and gluconate emanating from decontamination chemicals, critical concentrations may arise in some waste types in BMA, the Silo and BTF (e.g. F.17, B.05 and F.05). Elements probably to be affected is Mn and in some cases Ni and Pb. Na-gluconate may be present in such quantities that critical concentrations are obtained in waste packages containing unconditioned or bituminised waste (F.17, F.18, F.05 and O.07). Na-gluconate affects the sorption of Eu and potentially also other elements, but current knowledge is limited to Eu. For waste types where the waste is stabilised in cement, sorption of Na-gluconate on the cement will reduce the concentration below critical levels. EDTA will be present at relatively low concentrations but may exceed critical levels in a limited number (56) of waste packages of waste type S.09.

The presence of cellulose based additives in cement used as backfill was evaluated. Calculations resulted in a concentration of ISA below critical levels.

For the waste types in BLA the presence of complexing agents has not been studied as no credit is given in the safety assessment to sorption in this repository part (no cement/concrete).

The performed calculations have shown that, due to the presence of ISA and other complexants in SFR, a reduction in sorption may have to be applied for some elements in certain waste types when carrying out calculations of the radionuclide transport (the number of waste types depends on assumptions regarding the ISA yield).

It should be pointed out that the estimates of concentrations in different waste types are based on a number of conservative assumptions, thus the actual concentrations are probably lower than the calculated.

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Appendix B - Calculated concentrations of ISA using three different yields

Appendix C - Waste characteristics in SFR (BMA, Silo and BTF)

# 1 Introduction

Low- and intermediate level radioactive waste, produced at Swedish nuclear power plants, will be deposited in an underground repository, SFR. Different substances in the waste or in degradation products emanating from the waste, and chemicals added during the building of cementitious barriers in the repository, may have complexing properties. The complexation of radionuclides with such ligands may increase the mobility of the deposited radionuclides as sorption on the cement phases is decreased and solubility increased. This could lead to an increased leaching of the radionuclides from the repository to the geosphere and biosphere. To evaluate the implications for the function and long-term safety of the repository, a study has been performed on complexants in SFR.

This report is based on

- a review of the national and international literature related to complexing agents in underground repositories, including degradation of cellulose and effects on radionuclide sorption and solubility
- documents prepared by the Swedish Nuclear Fuel and Waste Management Co, SKB, presenting background information on sources to complexing agents in SFR (chemicals used at nuclear power plants, documents regarding cement additives, etc).

A summary of the complexing agents that may be deposited in a repository is made and an evaluation of their importance in SFR is performed. Concentrations and amounts are calculated in different waste types of the repository and the effect on radionuclide sorption and solubility is discussed.

The results of this review on complexing agents in SFR, and their effect on radionuclide mobility, will be used in the safety assessment when calculating the radionuclide transport.

## 2 State of the art

A literature review on complexing agents has been carried out covering both the Swedish and the international literature mainly after 1991 (when the previous assessment in Sweden was performed). As to the national sources, documents and reports by the Swedish Nuclear Fuel and Waste Management Co, SKB and related safety assessments (low- and intermediate level waste in SFL; Skagius et al., 1999) have been consulted for information. This include documents on complexing agents and chemicals used at the nuclear power plants (NPP) and in cement additives, cellulose degradation, waste characterisations, etc. Recent research performed at Swedish Universities is also reported (e.g. Pavasars, 1999). The international sources include work mainly performed in Switzerland and England (NAGRA, NIREX/AEA and PSI).

In this chapter a general presentation of complexants in low-and intermediate level waste is given. The effect on sorption of radionuclides is discussed. The presence of different substances in SFR is more thoroughly reported in Chapter 3.

### 2.1 Complexing agents in low- and intermediate level waste

Complexing agents generally present in low and intermediate level radioactive waste include both organic and inorganic compounds, of which the organic ligands are more abundant as well as important. They are constituted primarily by low weight organics, such as the decontamination agents EDTA (ethylene diamine tetraacetic acid), DTPA (diethylene triamine pentaacetic acid), NTA (Nitroltriacetate), oxalic, citric and tartaric acids, alkyl sulphonic acids and substituted phenols. Other organic complexing agents of varying importance are degradation products from polymers, bitumen (predominately oxalic acid), cation exchange resins (also mainly oxalic acid), anion exchange resins (amines), cellulose (isosaccharinic acid) and cement additives. Degradation of other high molecular weight organics (such as the polymers nylon, polystyrene, polyvinylchloride, etc) has, in comparison with cellulose degradation products, probably no significant influence on the radionuclide sorption (Bradbury and Van Loon, 1997).

The chemical structure of organic ligands contains many carboxylic acids or carboxyl groups. NTA, EDTA and DTPA are constituted by aminopolycarboxylic acids. These have strong complexing properties, mainly due to their ability to form chelates, i.e. complexes where the central metal atom has bonds to more than one ligand atom of the complexing agent. Organic ligands, and especially the chelat complexing agents, are the most important for the radionuclide mobility. The carboxylic acids are generally weak or moderately strong complexing agents. Carboxylic acids with only one carboxylic group (e.g. acetic acid) are weaker than those with several groups (e.g. oxalic acid). The relative strength follows the pattern DTPA > EDTA > NTA > tricarboxylic acids > di- > mono (Allard et al., 1985).

Inorganic ligands from the radioactive waste and the surrounding ground water, e.g. CO<sub>3</sub>, NO<sub>3</sub>, SO<sub>4</sub>, S, PO<sub>4</sub>, CN, B(OH)<sub>4</sub>, F, I, Cl, Br, will be present in underground repositories. It has been concluded that under the ambient high pH in a cementitious repository, only cyanide complexes would be of importance for the radionuclide (Ni,

Ag, Pb) sorption (Bradbury and Van Loon, 1997). However, cyanide present as ferrocyanide will play a minor role for the mobility of radionuclides since the hexacyanoferrate anion ( $\text{Fe}(\text{CN})_6^{4-}$ ) only form weak complexes with metal cations (Bradbury and Van Loon, 1997).

## **2.2 Degradation of different materials**

Organic material present in the waste in an underground repository can be degraded to products exhibiting complexing properties. The main components that will be subject to degradation processes are cellulose (in waste and cement additives), ion exchange resins and bitumen. However, only cellulose has been found to give rise to degradation products with strong complexing properties (SKB, 1991). The degradation of the three components are further described in Section 2.2.1-2.2.3).

The presence of microorganisms and their importance for the repository performance have recently been investigated for SFR (Pedersen, 2001). Microorganisms may increase radionuclide mobility by the degradation of cellulose and the production of isosaccharinic acid (ISA). According to Pedersen, the microbes may also affect the mobility by reducing or oxidising the nuclides or by physically transporting them through sorption to the microbial body.

Fatty acids and other organic acids are not end products and will finally form compounds such as carbon dioxide and methane. The degradation and stability of ISA is further discussed under Section 2.2.1, subsection "Sorption and stability of ISA"

### **2.2.1 Cellulose**

#### ***Degradation mechanisms***

Substantial amounts of cellulosic materials such as wood and paper, can be present in low- and intermediate level radioactive wastes (Wiborgh, 1995). In the highly alkaline cement pore water present in a cementitious underground facility for storage of radioactive waste, the cellulose is slowly degraded to water soluble, low molecular weights compounds. The mechanisms of alkaline degradation of cellulose are fairly well understood (Pavasars, 1999, Van Loon and Glaus, 1998 and Bradbury and Van Loon, 1997).

In alkaline media, the reducing glucose end group of a cellulose molecule is subject to a number of isomerizations. Depending on the elimination sequence, the glucose units can be split off one by one from the cellulose molecule, resulting in an endwise chain depolymerization, *the peeling-off reaction*. The hexose unit may also remain attached to the cellulose molecule, i.e. formation of a stable metasaccharinic acid end group, which terminates the chain depolymerization - *the chemical stopping reaction*. According to Van Loon and Glaus (1997), alkaline degradation also involves a *physical stopping reaction* and *alkaline hydrolysis* (or mid-chain scission). In the former, a reducing end group reaches the crystalline region of the cellulose, which means that the cellulose no longer is accessible (Van Loon and Glaus, 1998). In the alkaline hydrolysis, which is a slow process, the cellulose chain is split through cleavage of glycosidic bonds. This produces new reducing end-groups, leading to a continued peeling-off reaction which can result in total degradation of the cellulose. Together, the chemical and physical stopping reactions represent the *total chain termination reaction* (Pavasars, 1999). The degradation will initially be dominated by the peeling-off reaction (about 5% degraded after  $10^4$ - $10^5$  years). The rest of the cellulose will be degraded by a combination of alkaline hydrolysis and the peeling off reaction.

### **Degradation products**

The products formed during alkaline degradation of cellulose are to a great extent determined by the composition of the solution (Bradbury and Van Loon, 1997; Van Loon and Glaus, 1997). In the presence of  $\text{Ca}^{2+}$  in the cement pore waters, the formation of isosaccharinic acid (ISA) is favoured (Bradbury and Van Loon, 1997, Van Loon and Glaus, 1997/1998). ISA has been found to be the most common and abundant final degradation product of cellulose (>80 % of DOC). The substance exhibits strong complexing properties and may affect the radionuclide mobility (Pavasars, 1999, Glaus et al., 1999 and van Loon et al., 1999). Minor degradation components are short chain aliphatic acids such as formic acid, acetic acid and lactic acid, comprising less than 5-10 % of the dissolved organic carbon (Glaus et al., 1999 and Van Loon et al., 1999).

### **Degradation rates**

The literature presents different degradation rates depending on the environmental conditions used in the experiments. Many experiments have been performed under high temperatures or for a limited period of time. Extrapolation of the results to natural conditions is difficult and may lead to an overestimation of the degradation rate. The error introduced is enlarged when considering long time periods, as is the case for the degradation of cellulose in a waste repository.

In general, the degradation rate and the composition of degradation end products are dependent on pH, cations present, temperature and red-ox conditions (Pavasars, 1999, Bradbury and Van Loon and Glaus, 1997). The final amount of cellulose degraded is dependent on the amount of reducing end groups in a cellulose molecule and on the degree of polymerisation (Van Loon and Glaus, 1998). The degree of polymerisation (DP) affects the degradation so that the larger the cellulose molecule, the lower the mole fraction of reducing end groups and the lower the extent of degradation. The degree of polymerisation is lower for paper than for wood and cotton (Bradbury and Van Loon,

1997). DP for different cellulosic materials lies within a factor of approximately 3.  $(Gr)_0$  ( $(Gr)_0 = 1/DP$ ) can be used to estimate the number of reducing end groups in cellulose (Glaus et al., 1999).

The degradation processes can be described by Equation 2.1 (e.g. Van Loon and Glaus, 1998). For data used in calculations, see Van Loon and Glaus, 1998.

$$\text{Degraded cellulose} = 1 - \left( \frac{1 - \frac{k_1}{k_t} \cdot (Gr)_0 \cdot (1 - e^{-k_1 t})}{e^{k_{obs} x_n t}} \right) \quad \text{Equation 2.1}$$

where  $k_1, k_t =$  degradation rates for the alkaline degradation  
 $DP =$  degree of polymerisation  
 $0 \leq (Gr)_0 \leq 1/DP$   
 $x_n =$  number of glucose units peeled off  
 $k_{obs} =$  degradation rate for the alkaline hydrolysis

During the initial stages of cellulose degradation (up to 100 000 years), the relatively fast peeling-off mechanism will dominate and about 3-10 % of the cellulose will be degraded (Van Loon and Glaus, 1998). The remaining cellulose will degrade via the slow alkaline hydrolysis. This process leads to the production of new reducing end groups which will be subject to the peeling-off reaction. Thus, theoretically, alkaline hydrolysis can enable complete degradation of cellulose for times greater than 100 000 years. The degraded amount after 10 000 years is used in this report when calculating the concentration of ISA in different waste types and parts of the repository.

Previously, a yield of 0.1 mole ISA/kg cellulose has been used (Skagius et al., 1999). This yield, equal to about 2% of the cellulose being degraded, will also be used for comparison.

Experiments at the Linköping University (Pavasars, 1999) showed that after three years, 15-20% of the cellulose in cellulose powder was degraded to ISA (~1 mole ISA/kg cellulose), the rate decreasing with time. A model allowing long-term predictions indicated that the cellulose would degrade much faster than previously expected. A complete degradation was shown after 150-550 years assuming a constant pH of 13.3, instead of  $10^6$ - $10^7$  years according to Van Loon and Glaus (1997, 1998). However, in another experiment by Pavasars performed on sawdust, only 0.35 mole ISA/kg cellulose (~6%) was produced after three years. After 1-2 years the ISA concentration did not further increase with time, as in the experiment on cellulose powder (Pavasars, 1999).

## 2.2.2 Bitumen

Bitumen is a mixture of high molecular aliphatic and aromatic hydrocarbons, including components such as nitrogen, oxygen and sulphur (Allard et al., 1985). Chemical changes with time, including microbial degradation, would be expected under oxidising conditions. It has however not been demonstrated that bitumen would be degraded by

microorganisms under anaerobic conditions. According to a recent report (Pedersen, 2001) the microbial degradation of bitumen will be very slow under anaerobic conditions and should not affect the safety in SFR.

Different carboxylic acids could be formed during degradation of bitumen. However, those found are reported to be weak complexing agents and will have no significant influence on the sorption of radionuclides onto cementitious materials (Allard et al., 1985; Bradbury and Van Loon, 1997). On a site in northern Jordan with natural occurring bitumen and alkaline groundwater (conditions similar to SFR) no elevated groundwater concentrations of organic substances was found (SKB, 1991). Nor was the solubility of some analysed trace elements elevated, both indicating a minor degradation of bitumen and a limited effect on radionuclide transport in a repository.

### **2.2.3 Ion exchange resins**

Ion exchange resins consists of polystyrene chains with amines as active groups in anion exchangers and sulfonic acids in cation exchangers (Allard et al., 1985). In cation exchangers oxalic acid would be the major degradation product although only a minor fraction of low molecular compounds are likely to be formed (Allard et al., 1985). Degradation of the anion exchange resins may give rise to different forms of potentially complexing amines. However, according to several investigators, the degradation products from ion exchange resins have not been shown to have any significant influence on the sorption of radionuclides onto cementitious materials (Bradbury and Sarott, 1994; Bradbury and Van Loon, 1997; Allard et al., 1985).

## **2.3 Influence of ISA and other complexants on radionuclide mobility**

Isosaccharinic acid (ISA) forms strong water-soluble complexes with many radionuclides, especially tri- and tetravalent metals (e.g. Eu and Th). The presence of ISA in the pore water thus increases solubility and reduces the sorption of the radionuclides to cement.

Citric acid, oxalic acid, EDTA and NTA mainly affect the divalent elements Mn, Ni and Pb.

### **2.3.1 Sorption**

#### ***Presence of ISA***

Several investigators have studied the effects on sorption onto cement of different radionuclides under the presence of isosaccharinic acid. In Allard et al., 1995 ISA was found to form strong complexes, chelates, especially with three- and four-valent elements, but probably also with bivalent elements. Van Loon and Glaus (1998) observed effects on the sorption of Eu(III) at ISA concentrations of about  $1 \cdot 10^{-4}$  M. Wieland et al. (1998) found negligible effects at concentrations between  $10^{-5}$  -  $10^{-2}$  M (also in Bradbury and Van Loon, 1997). Only for ISA concentrations  $>10^{-2}$  M, reduction of the sorption of Eu(III) was significant. The difference between the two

studies is not clear. The sorption of Eu-ISA might be an explanation. Eu sorption on HCP surfaces was reduced by a factor 100 at equilibrium concentrations of  $\sim 10^{-1}$  M ISA (Bradbury and Van Loon, 1997).

No influence on Th sorption was found at equilibrium ISA concentrations of  $\leq 10^{-4}$  M (Bradbury and Van Loon, 1997). Wieland et al., 1988 found a significant effect on the sorption of Th(IV) on cement in the ISA concentration range  $10^{-5}$  -  $10^{-2}$  M. In a report by PSI, sorption of Th was said to be reduced by over four decades at ISA equilibrium concentrations of  $10^{-4}$  -  $10^{-2}$  M (Bradbury and Van Loon, 1997). The effect was much higher than the sorption effects described in Van Loon and Glaus, 1998. Experiments by Pavasars (1999) have shown that the addition of a high concentration of ISA ( $3 \cdot 10^{-2}$  M) results in a persistent reduction of radionuclide sorption which after 3 months was found to be reduced 400 times for Pm and at least 50 times for Th (Pavasars, 1999). The addition of  $\sim 6 \cdot 10^{-3}$  M ISA (1 g/l) caused a sixfold increase in the total alpha activity in the solution. Further increase of the ISA concentration did not increase the alpha activity significantly (Dahlén et al, 1999, in Pavasars, 1999).

Sorption reduction has been found for Ni as well, but significant effects will only be seen at ISA concentrations higher than  $10^{-2}$  M (Van Loon and Glaus, 1998). This indicates that the Ni-ISA complex is less stable than Th-ISA or Eu-ISA. Cr and Sr are two other metals unaffected by ISA concentrations below  $10^{-2}$ . Van Loon studied the degradation of Aldrich cellulose, Tela, cotton and paper, which had various effects on the sorption of different radionuclides. In a short-term test on hardened cement paste with ISA present in trace concentrations ( $10^{-12}$  M), no adverse effect on sorption of metals could be seen (Bradbury and Van Loon, 1997).

Experiments on waste in SFR (Silo1 from Studsvik) have been performed to evaluate the presence and effects of complexing ligands (Allard and Riggare, 1998). No rapid mobilisation of radionuclides was identified in the alkaline environment and presence of ISA could not be indicated. Adding high amounts of ISA lead to an increased mobilisation of alpha-activity (not Am, possibly Pu) but it was concluded that deposition in a cementitious matrix would be an acceptable method for the disposal of radioactive waste.

To summarise, three- and tetravalent elements (Eu, Am, Cm and Th, Np, Pa, Pu, U, Tc, Zr, Nb) will undergo a significant sorption reduction for ISA concentrations higher than  $10^{-4}$  -  $10^{-3}$  M. For bivalent elements (Ni, Co, Fe, Be and Pb), the concentration must be higher than  $10^{-2}$  M (Van Loon and Glaus, 1998). However, Pavasars (1999) and others suggests that in a long-term perspective, the effects of cellulose degradation products on radionuclide sorption will probably be negligible.

**Table 2.3.1 Summary of effects on sorption due to the presence of ISA and the magnitude of the sorption reduction**

ISA conc (M)	Effects on sorption of radionuclides onto cement*			Source
	Significant effects	Negligible effects	Reduction factors	
$10^{-12}$	no adverse effects			Bradbury and Van Loon, 1997
$1 \cdot 10^{-4}$	Effects observed for Eu(III)	Th(IV)		Van Loon and Glaus, 1998
$10^{-5} - 10^{-2}$	Th(IV)	Eu(III)		Wieland et al., 1988
$10^{-5} - 10^{-2}$	Th(IV)			Van Loon and Glaus, 1998
$>10^{-4} - 10^{-2}$	Th(IV)		Th: >40	Bradbury and Van Loon, 1997
$<10^{-2}$		Cr, Sr unaffected		Van Loon and Glaus, 1998
$3 \cdot 10^{-2}$	persistent reduction after 3 months		Th(IV): >50 Pm: 400	Pavasars (1999)
$>10^{-2}$	Significant effects for Eu(III), effects observed for bivalent (e.g. Ni)			Van Loon and Glaus, 1998
$>10^{-2}$	Effects observed for Eu(III)			Wieland et al., 1988
$>10^{-2}$	Eu(III)		Eu: >40	Bradbury and Van Loon, 1997
$\sim 10^{-1}$			Eu: 100	Bradbury and Van Loon, 1997

\* different results obtained in Wieland et al., 1988 and Van Loon and Glaus, 1998.

### **Presence of cement additives**

Although experiments have been carried out at different organisations (e.g. Nirex/Nagra), questions remain about the effect on mobility of the presence of cement additives. The uncertainties regard long-term effects and effects at realistic concentrations. Common cement additives include naphthalene sulphonic acid, lignine sulphonic acid, polycyclic acid polymers, gluconate, maleinic acid and melamine formaldehyde. In SFR most cement additives used are cellulosic (lignine, lignosulphonates, etc). The amount of cellulose added to cement is limited, but have been regarded in the analysis.

### **Presence of other complexing agents**

Chemicals used at the Swedish nuclear power plants include substances that could act as complexants with radionuclides. The most important ones are citric and oxalic acid, NTA, EDTA and Na-gluconate. The effects on sorption of some of these complexants have been reported in Bradbury and Sarott (1994). Based on speciation calculations simulating cement pore waters (pH = 11-13, Ca-concentration = 0.001-0.1 M) they conclude that Ni, Mn and Pb are the only elements to be affected, EDTA being the only complexant of significance. The sorption of Ni and Mn will be reduced at EDTA concentrations higher or equal to  $10^{-4}$  M and the sorption of Pb may be affected if the

concentration exceeds  $10^{-3}$  M (Table 2.3.2). NTA and citric acid may play a role at very high concentrations ( $>10^{-2}$  M). No effect on sorption is expected for other elements (Cs, Pu, Am, U, etc) at concentrations less or equal to 0.1 M. This is primarily due to the high amounts of  $\text{Ca}^{2+}$  and  $\text{OH}^-$  which will reduce the effects of EDTA, oxalic acid and citric acid due to complexes of Ca-organic ligands or hydroxyl-metals.

**Table 2.3.2 Sorption reduction factors for Ni, Mn and Pb in the presence of EDTA, citric acid and NTA (Bradbury and Sarott, 1994)**

Conc. of chemical M (mole/l)	Reduction factors in presence of different chemicals											
	Ni				Mn				Pb			
	EDTA	NTA	Citric	Oxalic	EDTA	NTA	Citric	Oxalic	EDTA	NTA	Citric	Oxalic
$\leq 10^{-4}$	1	1	1	1	1	1	1	1	1	1	1	1
$10^{-3}$	2	1	1	1	3	1	1	1	1	1	1	1
$10^{-2}$	10	1	1	1	20	1	1	1	2	1	1	1
$10^{-1}$	100	5	1	1	200	6	4	1	10	2	1	1

Other sources also report that the effects of EDTA, although being a strong complexing agent, are limited to a shorter time period due to a sorption recovery and  $K_d$ -values similar to those measured without complexing agents.

An initial sorption reduction of americium has been reported at EDTA concentrations of 0.5 g/l ( $1.5 \cdot 10^{-3}$  M) (Allard and Andersson, 1987). After a couple of weeks to months, sorption recovered and was the same as without the presence of EDTA. At EDTA concentrations of 10 g/l ( $3 \cdot 10^{-2}$  M), the recovery took up to one year. The recovery may be due to the fact that EDTA itself sorbs onto the cement.

In a report by PSI (Bradbury and Van Loon, 1997), Ni is said to be the only safety relevant radionuclide to be influenced by EDTA. Effects are only likely to be apparent at EDTA concentrations well in excess of  $10^{-3}$  M. PSI found no available data on the sorption of Ni on cement in the presence of EDTA. A reduction factor of 50 was chosen for Ni and chemical analogues based on an estimated concentration of Na-EDTA of  $4.6 \cdot 10^{-2}$  M in the pore water. Pb forms weaker complexes with EDTA and a sorption reduction factor of 5 was used (Bradbury and Van Loon, 1997).

Currently, experiments are being performed at the University of Örebro to further investigate the effects of citric acid and similar complexants on the mobility of radionuclides.

Experiments on the influence of gluconate on sorption have been carried out (Norden and Allard, 1994). Sorption of Eu decreased by approximately three orders of magnitude at gluconate concentrations above  $10^{-4}$  M. However, the sorption values recovered to their original values within approximately 200 days. Another author reported long-term reductions of only one order of magnitude for Eu at a gluconate concentration of 0.25 M (Bradbury and Van Loon, 1997). No effects could be seen on sorption of Cs, Sr, Ni, Eu, Th on CSH phases for gluconate concentrations up to  $10^{-4}$  M (Bradbury and Van Loon, 1997).

Cellulosic materials contain alkali soluble organics (e.g. hemicellulose) which, together with the degradation products of cellulose, will contribute to the dissolved organic matter in the pore solution. The presence of these organics has been shown to affect the sorption of metals, especially Ni (Van Loon and Glaus, 1998).

### ***Sorption onto cement and stability of ISA and gluconate***

Some processes may reduce the concentration of complexants in the pore water, e.g. the degradation, the formation of sparingly soluble compounds and the sorption to cement.

Experiments are currently being performed in the Nirex programme to evaluate the effect of stability of the main degradation product of cellulose, isosaccharinic acid, under alkaline conditions. Until recently, ISA was believed to be stable under alkaline conditions and microbial degradation has been considered negligible (Van Loon et al., 1997; Van Loon and Glaus, 1998). In Allard et al., 1995, ISA was reported to degrade chemically to carboxylic acids under high pH and aerobic conditions. Also biological degradation was reported to be relatively rapid in alkaline, aerobic environments. ISA would probably degrade under anaerobic conditions as well, but the rate of this process was not known (Allard et al., 1995). New experiments on mud and debris from naturally occurring alkaline lakes indicate that ISA can be microbially degraded under repository conditions (Askarieh et al, 2000; Greenfield et al, 1995). However, uncertainties remain about the development of microbial populations with time and if removal of complexants by microbial action would have a significant impact on the release of radionuclides (Askarieh et al, 2000).

According to experiments the calculated maximum concentration of sparingly soluble ISA compounds is very low (Van Loon et al, 1997; Van Loon and Glaus, 1998).

Therefore, sorption to cement seems to be the dominating process for the reduction of ISA in the pore water. Experiments have shown that the addition of  $3 \cdot 10^{-3}$ - $5 \cdot 10^{-3}$  M ISA significantly reduced sorption of Pm-147 and Th-234, but that the effect was only apparent during an initial period of 1 month. Thereafter, sorption of ISA to cement reduced the concentration in solution. When using yet higher concentrations of ISA ( $3 \cdot 10^{-2}$  M), the reduction of radionuclide sorption was more persistent still after 3 months (in Pavasars, 1999).

Experiments have also demonstrated that the total sorption capacity of the cement for ISA is approximately 0.3 mole/kg (Van Loon et al, 1997; Bradbury and Sarott, 1994). PSI (Bradbury and Van Loon, 1997) gives a value of about 0.2 mole/kg (ISA range  $10^{-5}$  to  $10^{-2}$  M) for hardened HTS cement paste. Experiments by Pavasars (1999) confirm the high sorptivity of ISA onto cementitious phases. Only trace amounts of ISA were detected in leachates of a cement body and no through-diffusion was observed through a concrete barrier after 700 days, indicating that ISA is retained in the cement body due to strong sorption.

Van Loon and Glaus (1998) suggest two different sorption isotherms to estimate the sorption of ISA to cement. The equilibrium concentrations of ISA,  $Q_{ISA}$ , are described as:

$$Q_{ISA} = \frac{K \cdot q \cdot C_{ISA}}{1 + K \cdot C_{ISA}} \quad \text{Equation 2.2}$$

where  $K = 690 \text{ l/mole}$   
 $q = 0.17 \text{ mole/kg}$   
 $C_{ISA} = \text{concentration of ISA after degradation of cellulose}$

$$Q_{ISA} = \frac{K_1 \cdot q_1 \cdot C_{ISA}}{1 + K_1 \cdot C_{ISA}} + \frac{K_2 \cdot q_2 \cdot C_{ISA}}{1 + K_2 \cdot C_{ISA}} \quad \text{Equation 2.3}$$

where  $K_1 = 1730 \text{ l/mole}$   
 $q_1 = 0.1 \text{ mole/kg}$   
 $K_2 = 12 \text{ l/mole}$   
 $q_2 = 0.17 \text{ mole/kg}$   
 $C_{ISA} = \text{concentration of ISA after degradation of cellulose}$

Van Loon and Glaus showed that the non-linear two-site model (Equation 2.3) gives a better agreement with measured data than the linear one-site model (Equation 2.2). In this report, both models have been used for comparison when calculating the concentration of ISA in the pore water of different waste packages in SFR. For results and calculated concentrations, see Section 3.2.

Also gluconate has been demonstrated to sorb to a significant extent on cementitious materials (Bradbury and Van Loon, 1997). Likewise to ISA, the sorption of gluconate can be described with a Langmuir type of sorption isotherm. The affinity for sorption to cement ( $K$ ) seems to be even higher for gluconate than for ISA.

### **Sorption of ISA complexes**

Although ISA forms strong chelate complexes with radionuclides, recent experimental results indicate that the formed ISA complexes also will sorb to the cement phases (Holgersson, 2000). The reduction in sorption above a critical ISA concentration is interpreted as the result of saturation of the sorption sites due to sorption of ISA and ISA complexes (Holgersson, 2000). Thus, even if isosaccharinic acid reaches critical concentration levels and reduces the sorption of radionuclides to cement in contact with this water, sorption of the entire ISA complex on cement may be unaffected in other parts of the repository where the ISA-concentration is lower and sites are still available for sorption.

### 2.3.2 Solubility

The presence of ISA will affect the solubility of trivalent and tetravalent elements. According to the literature, the solubility of U, Th and Pu increase with a factor of 250, 500 and  $2 \cdot 10^5$  respectively, at a concentration of ISA of  $2 \cdot 10^{-3}$  M (Allard et al., 1995). In experiments with a total ISA concentration of  $5 \cdot 10^{-3}$  M, the solubility of Th, Pm and Ni increased with a factor of 14000, 300 and 2 (Holgersson et al., 1999).

The solubility of Pu increases significantly at ISA concentrations above  $0.1 \cdot 10^{-3}$  M. At ISA concentrations of  $1 \cdot 10^{-3}$  M the solubility of Pu is in the order of  $10^{-5}$  M and increases to about  $10^{-4}$  M at ISA concentrations of  $5 \cdot 10^{-3}$  M (Allard et al., 1995).

In the coming radionuclide transport calculations, higher solubility constraints may be applied for some radionuclides. In SFR, the amount of radionuclides is low and only for a few elements, if any, solubility constraints will be of concern. International safety assessments have shown that the enhancement of radionuclide solubility by organic degradation products is in general not of concern in low- and intermediate level waste repositories. More important is the influence on these ligands on sorption (Van Loon and Glaus, 1998).

### 3 Complexing agents in SFR

The presence of complexing ligands in SFR has been identified and their amounts calculated. The documentation covers an investigation of

- the waste
- products emanating from the degradation of the waste
- the repository construction materials
- material, chemicals and additives used when producing waste packages

A list consisting of a wide variety of complexing ligands was set up (Appendix A). The list covers the substances that one may expect to find in radioactive waste repositories in general. The importance of each substance was evaluated from its strength and presence in SFR. Documents produced by SKB on waste types and materials deposited in SFR and information on the usage of chemicals at the nuclear power plants (NPP) were used. The relevance of the different substances is commented upon in the list in Appendix A.

The waste in SFR contains a substantial amount of organic substances, consisting mainly of ion exchange resins, bitumen, different plastics, rubber and cellulose materials. The cement, used in waste moulds and construction, contains organic additives of which most are constituted by cellulose or cellulose derivatives. The radioactive waste also contains chemicals used in different processes at the nuclear power plants. The substances are described in the coming sections.

The importance of the identified complexants for the long-term safety of SFR is evaluated from their strength and abundance in the different repository parts; BMA, Silo, BTF and BLA. For those present in great amount or those known to have a strong complexing character, calculations of concentrations in different waste types and different parts of SFR are performed (Section 3.2).

#### 3.1 Sources

##### 3.1.1 Cellulose in waste and cement additives

Cellulose is present in the waste in the form of paper and textile. ISA, the degradation product of cellulose, is the most important complexant with respect to both amounts and strength. Estimated amounts of cellulose in different waste types are reported in the tables below together with the amount of cellulose in cement additives.

Different cement additives are present in concrete packaging, in cement used as waste conditioning and in the grout used as backfill around the containers in the SFR repository. SKB investigated used additives and pointed out those exhibiting complexing properties (SKB 99/10). The most common cement additives were not found to be strong complexants (e.g. Silix GP and Sika AER). Sikament 10, containing maleinic acid, has however been subject to discussions and it is not yet clear if the product is an important complexant or not. In this report, Sikament 10 is not further regarded. Additives based on cellulosic materials are the most important potential sources for

complexing agents. The amounts used when calculating concentrations of ISA in different waste types are reported in tables below.

The additive Cements, containing 0.11 % cellulose based material per kg cement, is used in mould construction since 1998 (0.7 kg/mould is assumed for all waste packages, e.g. in R.01, R.02, O.02 and O.23). Some of the waste conditioned in cement contain Betokem (cellulose based substances in Betokem = 0.12% of dry weight or 0.11 % of total cement weight), see tables below.

Other cement additives (such as Barra 55, Peramin F and Peramin L), are either relatively weak complexants or are present at low amounts. No further consideration is given to these products.

**Table 3.1.1 Amount of cellulose (kg/package) present in different waste types in BMA from waste, cement additives and chemicals (tall fatty acid)**

	Waste	Cement additives		Chemicals	Total
		Cement waste matrix	Concrete packaging	Tall fatty acid	
F.17	3.6	–	–	–	3.6
F.23/steel	150	–	–	–	150
F.23/concrete	29	0.61	–	–	29.6
O.23	30	0.61	0.7	–	31.3
R.23/steel	44	–	–	–	44
R.23/concrete	11	–	0.7	–	11.7
S.21	7.9	–	–	–	7.9
S.23	29	–	–	–	29
S.09	–	–	–	–	–
O.01	–	1.6	0.7	–	2.3
R.01	–	–	0.7	–	0.7
R.10	–	–	–	–	–
R.15	–	–	–	–	–
B.05	–	–	–	0.03	0.03
F.05	–	–	–	–	–
F.15	–	–	–	–	–

**Table 3.1.2 Amount of cellulose (kg/mould) present in different waste types in the Silo from waste and cement additives (mould production and conditioning)**

	Waste	Cement additives		Chemicals	Total
		Cement waste matrix	Mould production	Tall fatty acid	
O.24*	35	–	–	–	35
O.24/concrete	36	1.1	–	–	37.1
S.22	8	–	–	–	8
S.11	3	–	–	–	3
O.02	–	1.6	0.7	–	2.3
R.02	–	–	10.7	–	10.7
R.16	–	–	–	–	–
B.06	–	–	–	$4.4 \cdot 10^{-4}$	$4.4 \cdot 10^{-4}$
F.18	–	–	–	–	–

\* steel drum in concrete mould

In BTF there is no cellulose except for waste type B.07. This mould contains tall fatty acid (0.01 kg/mould), which is assumed to degrade to ISA.

In BLA, large amounts of cellulose are present. However, as no cement is used for backfilling or conditioning, the environmental conditions do not allow ISA to be formed (cement is only present between two steel drums surrounding the waste package S.14). Moreover, due to the limited amount of concrete construction materials in BLA the potential sorption capacity is insignificant. Hence, although complexing agents would be present in BLA, they would not further influence the mobility of elements. Concentrations of radionuclides are low in BLA and the elements are not expected to be solubility limited.

In the backfill concrete grout in BMA, BTF and the Silo, Plastiment and/or Methocel has been used as cement additives. The amount of cellulose in these cement additives is approximately 0.4 % of the cement weight (for Methocel, 0.2 % cellulose in dry cement). According to SKB (Johansson C and Skogsberg M, 1999), Methocel and Plastiment were used from 1991 and 1998 respectively and will not be used after 1999-2000.

### 3.1.2 Chemicals used at Swedish nuclear power plants

In an investigation by the Swedish Nuclear Fuel and Waste Management Co, SKB (SKB 99/13), a summary of the chemicals handled at the Swedish nuclear power plants (NPP) is presented. Used amounts during 1998 of different substances, including those exhibiting complexing properties, were derived, see Appendix A (based on the 1998 annual report/"Miljörapport" for the different plants and interviews with employees).

The substances of greatest importance are mostly used in decontamination processes, e.g. tensides and acids. Those complexants that could be important for the radionuclide mobility are listed in Table 3.1.3.

**Table 3.1.3 Estimated total amounts used at NPP (BKAB= Barsebäck, FKA= Forsmark, OKG= Oskarshamn, RGAB= Ringhals, SVIK= Studsvik) during 1998 (kg/year) or in total (kg tot), SKB 99/13**

Complexants [kg in total]* or [kg/year]	Estimated amounts to SFR from NPP					Tot
	BKAB	FKA	OKG	RGAB	SVIK**	
EDTA* (total amounts)	2.5	2.5	2.5	2.5	–	10
Sodium capryliminodi- propionat, NKP	–	–	9.2	–	–	9.2
NTA	–	10	1.8	–	5	16.8
Citric acid	3.8	33	0.5	–	2	39.3
Oxalic acid	–	35	–	–	2	37
Na-gluconate	–	1.9	12	–	1	14.9
Picolinic acid* (total amounts)	–	–	–	2.5	–	2.5
Tall fatty acid (Na-salt)	0.3	–	–	–	–	0.3

\* no more EDTA to SFR after 1998, picolinic acid only used during 1998

\*\* EDTA and gluconate in waste package S.09 from Studsvik have been studied in a separate analysis by SKB (see Table 3.1.5)

The most abundant chemicals used are citric and oxalic acid. These complexants are however less strong than for example EDTA and NTA. EDTA is present in low amounts and has not been used at the power plants since 1998. At a maximum, 10 kg are estimated to have been deposited in SFR (Riggare P and Johansson C, 1999). Negligible amounts of EDTA have been used in research (at Studsvik/SVIK). However, the amount of EDTA and gluconate in waste package S.09 has recently been studied in a separate analysis by SKB. Preliminary results were presented in January 2001 (Johansson C, 2001). In total, 56 moulds (S.09) have been produced containing about 4 g EDTA and gluconate respectively, see notes in Table 3.1.5. Packages of this waste type that will be produced in the future will not contain any EDTA or gluconate.

An evaluation of the strength of different complexants is given in Appendix A. Tall fatty acid is conservatively assumed to exhibit a cellulosic character, therefore producing ISA when being degraded. The substance is present in small amounts but are found in two waste types not containing cellulose (B.05 and B.07).

Chemicals used at the nuclear power plants will end up in different waste types in SFR, see Table 3.1.4. The relative distribution to different parts of SFR of the amount of chemicals used was estimated for each power plant (Johansson C, 1999 and SKB 99/15).

**Table 3.1.4 Waste types receiving complexants from chemicals used at the power plants and their allocation in different repository parts in SFR (Johansson C, 1999)**

Waste type		Allocation in different parts of SFR (%)		
		BMA	BTF	Silo
BKAB	B.05, B.06, B.07	22	56	22
FKA	F.05, F.15, F.17, F.18	72	0	28
OKG	O.01, O.02, O.07, O.23	23	58	19
RGAB	R.10, R.15, R.16	18	0	82
SVIK	S.09, S.23	100	0	0

In the investigation by SKB, the annual production of different waste types was also determined (SKB 99/15). The amount of chemicals per mould was calculated by dividing amounts used during 1998 with the number of moulds produced per year (see Table 3.1.5). No annual production is given for O.01, F.05 and F.15 since these waste types are no longer produced. The amount in O.01 is assumed to be equal to the amount in O.23 (to BMA from Oskarshamn). The amount in F.05 and F.15 is assumed to equal F.17 (to BMA from Forsmark). As the amount of chemicals used during 1998 is distributed to fewer moulds, the amount per mould will be somewhat overestimated. For EDTA, the amount per mould was calculated using the number of moulds produced until 1998-12-31 and the total amount of EDTA deposited in SFR (10 kg). The amount of EDTA in 56 moulds of type S.09 is reported separately (Table 3.1.5).

**Table 3.1.5 Amount of chemicals per mould (kg/mould) in SFR**

Waste type	Moulds/year*	EDTA <sup>§</sup>	NKP**	NTA	Citric acid	Oxalic acid	Na-gluconate	Picolinic acid	Cellulose
<b>BMA</b>									
F.17	22	6.4·10 <sup>-4</sup>	–	0.33	1.1	1.1	0.06	–	–
O.23	17	6.4·10 <sup>-4</sup>	0.12	0.02	0.01	–	0.16	–	–
S.23	5	–	–	0.14	0.06	0.057	0.03	–	–
O01	–	6.4·10 <sup>-4</sup>	0.12	0.02	0.01	–	0.16	–	–
S.09	30	– <sup>§</sup>	–	0.14	0.06	0.057	0.03 <sup>§</sup>	–	–
R.10	4	6.4·10 <sup>-4</sup>	–	–	–	–	–	0.035	–
R.15	9	6.4·10 <sup>-4</sup>	–	–	–	–	–	0.035	–
B.05	2	6.4·10 <sup>-4</sup>	–	–	0.42	–	–	–	0.033
F05	–	6.4·10 <sup>-4</sup>	–	0.33	1.1	1.1	0.06	–	–
F15	–	6.4·10 <sup>-4</sup>	–	0.33	1.1	1.1	0.06	–	–
<b>SILO</b>									
O.02	59	8.6·10 <sup>-4</sup>	0.03	0.006	0.002	–	0.04	–	–
R.16	80	8.6·10 <sup>-4</sup>	–	–	–	–	–	0.026	–
B.06	150	8.6·10 <sup>-4</sup>	–	–	0.01	–	–	–	0.0004
F.18	17	8.6·10 <sup>-4</sup>	–	0.16	0.54	0.58	0.03	–	–
<b>BTF</b>									
O.07	12	4.8·10 <sup>-3</sup>	0.44	0.09	0.02	–	0.58	–	–
B.07	15	4.8·10 <sup>-3</sup>	–	–	0.14	–	–	–	0.011

\* Annual production; SKB 99/15. Assumptions: 0.01=O.23 (from Oskarshamn=>BMA) and F.05, F.15=F.17 (Forsmark=>BMA)

\*\* Sodium capryliminodipropionat

§ New values for S.09 (56 moulds in total): EDTA = 4.3·10<sup>-3</sup> kg/mould; gluconate = 3.6·10<sup>-3</sup> kg/mould (Johansson C, 2001); otherwise: 10 kg EDTA divided by number of moulds produced 1998-12-31.

When calculating concentrations and evaluating effects on sorption of radionuclides, some chemicals such as NKP and picolinic acid are compared to complexing agents with known strength and effects on sorption.

### 3.1.3 Important degradation processes

According to the discussion in Section 2.2, the degradation of cellulose to ISA can have a significant influence on the sorption of some radionuclides onto cement. Degradation of other materials such as bitumen or ion exchange resins (producing carboxylic acids and amines) has been shown to be limited or result in relatively weak complexes (Section 2.2).

The amount of ISA in different waste types is derived both using a yield of 0.1 mole/kg (~2% degraded cellulose; Skagius et al., 1999) and by using degradation Equation 2.1 in Section 2.2.1 (Van Loon and Glaus, 1998). The amount degraded after 1000 years using the equation is approximately 2.5%. After 10 000 years about 10% of the cellulose is

degraded. The production of ISA after 10 000 years using Equation 2.1 is 5-6 times higher than the yield of 0.1 mole ISA/kg cellulose. Concentrations of ISA based on 2% degraded cellulose (0.1 mole/kg) is a more likely estimation.

The production of ISA and calculated concentrations in different waste types and repository parts are presented in Section 3.2.

### **3.1.4 Summary**

Cellulose and some of the chemicals used at the nuclear power plants were found to be the main sources of complexing organic ligands in SFR. Estimated amounts and concentrations are listed in tables in Section 3.2. Of the inorganic ligands only cyanid complexes may be of importance for the radionuclide sorption. In SFR though, cyanide is present in very small or no amounts and will not play an important role for the assessment (Riggare P and Johansson C, 1999). Moreover, the likely chemical form of cyanide (ferrocyanide) will lead to weak complexes with metal ions (Skagius et al., 1999; Bradbury and Van Loon, 1997).

## **3.2 Concentration calculations**

In this section the concentration of identified complexants is calculated in different waste types. The effect on sorption of radionuclides is evaluated in Chapter 3.4.

### **3.2.1 Waste characteristics**

The extent of complexation of ISA and other complexing agents with radionuclides is affected by the concentration in the cement pore water. The concentration in the pore water for different waste types was calculated based on the amount of cellulose or chemicals in the waste, the extent of degradation of cellulose, the porosity of cement and the sorption of complexants onto cement (only for ISA).

When calculating the concentration of ISA, dilution and sorption is first studied for the (conditioned) waste alone. Sorption may also occur in the surrounding concrete packaging. For ISA, concentrations will be calculated taking into consideration the dilution and sorption in waste *and* concrete packaging. For substances such as EDTA and citric acid, sorption to cement has not been investigated in the literature. The concentrations of these substances are calculated using the inner volume of waste packages (mostly conditioned in cement or bitumen).

For used volumes and amount of cement, see Appendix C. For a more thorough description of the different waste types see SKB, 1993 and Riggare and Johansson, 2001.

### **3.2.2 Concentration of ISA**

The production of ISA through the degradation of cellulose was calculated from the amounts of cellulose and its derivatives using a yield of 0.1 mole/kg cellulose (see Section 2.2.1 and 3.1.3). This yield of ISA was previously used in another Swedish safety assessment (Skagius et al., 1999) and represents the degradation of cellulose after a few years. Calculations were also performed using an ISA yield after 1000 and

10 000 years respectively (Van Loon and Glaus, 1998). The latter is however a highly conservative approach and will probably lead to an overestimation of the calculated concentrations. Calculated concentrations of ISA based on 0.1 mole ISA per kg degraded cellulose are presented in Tables 3.2.1-3.2.3. Concentrations using all three yields are given in Appendix B.

The concentrations of ISA are adjusted taking into consideration the effects of sorption to cement using both the non-linear model (Equation 2.3) and the linear model (Equation 2.2), see Section 2.3.1. In the studied concentration ranges, the equilibrium concentration of ISA is approximately 20-50% higher using the non-linear equation. As experiments have shown better adjustment with the non-linear two-site model, this approach will be adopted. Concentrations in waste packages in different repository parts are given in the tables below (sorption data based on non-linear model). Degradation of ISA has not been regarded. This process may further reduce the pore water concentration. Waste types with potential critical ISA levels are shaded and will be discussed in more detail in Section 4.3.

**Table 3.2.1** *Calculated concentrations of ISA (originating from waste, cement additives and chemicals) in different waste types in BMA, using a yield of 0.1 mole ISA/kg cellulose. Waste and surrounding concrete packaging are treated separately with and without sorption to cement*

M (mole/l)	Waste		Waste and surrounding concrete packaging	
	No sorption	Sorption	No sorption	Sorption
F.17	$4.2 \cdot 10^{-3}$			
F.23/steel	$6.8 \cdot 10^{-2}$	$3.3 \cdot 10^{-4}$		
F.23/concrete	$2.8 \cdot 10^{-2}$	$1.2 \cdot 10^{-4}$	$1.4 \cdot 10^{-2}$	$4.7 \cdot 10^{-5}$
O.23	$2.9 \cdot 10^{-2}$	$1.2 \cdot 10^{-4}$	$1.5 \cdot 10^{-2}$	$5.0 \cdot 10^{-5}$
R.23/steel	$2.0 \cdot 10^{-2}$	$6.9 \cdot 10^{-5}$		
R.23/concrete	$1.0 \cdot 10^{-2}$	$3.9 \cdot 10^{-5}$	$5.4 \cdot 10^{-3}$	$1.8 \cdot 10^{-5}$
S.21	$1.6 \cdot 10^{-1}$		$5.4 \cdot 10^{-2}$	$2.1 \cdot 10^{-4}$
S.23	$2.7 \cdot 10^{-2}$	$3.1 \cdot 10^{-5}$	$1.3 \cdot 10^{-2}$	$2.2 \cdot 10^{-5}$
S.09	—*	—	—	—
O.01	$5.4 \cdot 10^{-4}$	$6.1 \cdot 10^{-7}$	$5.6 \cdot 10^{-4}$	$7.6 \cdot 10^{-7}$
R.01	—	—	$1.8 \cdot 10^{-4}$	$2.5 \cdot 10^{-7}$
R.10	—	—	—	—
R.15	—	—	—	—
B.05	$1.0 \cdot 10^{-4}$ **			
F.05	—	—	—	—
F.15	—	—	—	—

\* these waste types do not contain cellulose in waste or packaging

\*\* tall fatty acid only source of produced ISA. Concentration of tall fatty acid =  $8.3 \cdot 10^{-3}$  M.

The calculated concentrations of ISA in B.05 in BMA, B.06 in the Silo and B.07 in BTF, originates from the presence of tall fatty acids (Na-salt), originally used in chemicals at the nuclear power plants. The substance is assumed to degrade to ISA in the same way as cellulose, though this assumption is uncertain. The concentration of the fatty acid itself is  $8.3 \cdot 10^{-3}$  M,  $1.2 \cdot 10^{-4}$  M and  $1.8 \cdot 10^{-4}$  M in B.05, B.06 and B.07 respectively.

The concentration of ISA in some of the waste packages is also a result of the degradation of cellulose in cement additives (e.g. ligninesulphonates). The presence of additives in different waste packages is specified in Tables 3.1.1-3.1.2.

**Table 3.2.2** *Calculated concentrations of ISA (originating from waste, cement additives and chemicals) in different waste types in the Silo, using a yield of 0.1 mole ISA/kg cellulose. Waste and surrounding concrete packaging are treated separately with and without sorption to cement*

M (mole/l)	Waste		Waste and surrounding concrete packaging	
	No sorption	Sorption	No sorption	Sorption
O.24*	$1.9 \cdot 10^{-2}$	$4.0 \cdot 10^{-5}$	$1.2 \cdot 10^{-2}$	$2.8 \cdot 10^{-5}$
O.24/concrete	$3.3 \cdot 10^{-2}$	$7.8 \cdot 10^{-5}$	$1.6 \cdot 10^{-2}$	$4.3 \cdot 10^{-5}$
S.22	$1.6 \cdot 10^{-1}$		$5.5 \cdot 10^{-2}$	$2.2 \cdot 10^{-4}$
S.11	$1.1 \cdot 10^{-3}$	$9.4 \cdot 10^{-7}$		
O.02	$5.4 \cdot 10^{-4}$	$6.1 \cdot 10^{-7}$	$5.6 \cdot 10^{-4}$	$7.6 \cdot 10^{-7}$
R.02	–**	–	$2.7 \cdot 10^{-3}$	$3.8 \cdot 10^{-6}$
R.16	–	–	–	–
B.06	$1.5 \cdot 10^{-6}$ <sup>§</sup>			
F.18	–	–	–	–

\* steel drum in concrete mould

\*\* these waste types do not contain cellulose in waste or packaging

<sup>§</sup> tall fatty acid only source of produced ISA. Concentration of tall fatty acid =  $1.2 \cdot 10^{-4}$  M

The levels of ISA in F.17, F.23/steel and S.21 in BMA and S.22 in the Silo are higher than in the other waste packages. This is mainly due to the fact that there is no cement in these waste types, thus sorption of ISA to cement will not reduce the concentration in the pore water. The waste in F.17 is bituminised and S.21 and S.22 consist of two steel drums (where concrete is only present between the inner and outer drum).

**Table 3.2.3** *Calculated concentrations in BTF of ISA (originating from tall fatty acid in chemicals) in O.07 and B.07. A yield of 0.1 mole ISA/kg cellulose is used*

M (mole/l)	Waste		Waste and surrounding concrete packaging	
	No sorption	Sorption	No sorption	Sorption
O.07	–	–	–	–
B.07*	$2.2 \cdot 10^{-6}$		$1.0 \cdot 10^{-6}$	$<1.0 \cdot 10^{-6}$

\* tall fatty acid only source of produced ISA. Concentration of tall fatty acid =  $1.8 \cdot 10^{-4}$  M

Except for the above calculations for different waste types, an estimation of the concentration of ISA in the cement used as backfill in the repository was performed. The concentration was calculated to  $3 \cdot 10^{-3}$  M from a composition of 0.4% cellulose of the cement weight and a production of ISA based on a yield after 10 000 years of degradation (10%) (the volume of water in cement was calculated from a porosity of 30% and bulk density  $2000 \text{ kg/m}^3$ ). Sorption reduces the concentration to  $2 \cdot 10^{-5}$  M. The effects on sorption is further discussed in Chapter 3.4.

### 3.2.3 Concentrations of other substances

Concentrations of citric acid, EDTA etc from chemicals used at NPP, are not assumed to be reduced in solution due to sorption to cement. It is possible that the process occurs (as for ISA), but no references have been found treating the subject except for gluconate. This means that the concentrations reported in Tables 3.2.4-3.2.6 are probably overestimated. The substances will probably undergo degradation leading to yet lower concentrations. Another conservative approach is the distribution of chemicals to fewer moulds (see Section 3.1.2). The concentrations of the substances are listed in Table 3.2.4-3.2.6 (shaded concentrations exceeding critical levels are commented upon in Section 4.3).

**Table 3.2.4 Calculated concentration (M) of chemicals in different waste packages in BMA**

M (mole/l)	EDTA	NKP	NTA	Citric acid	Oxalic acid	Na-gluconate**	Picolinic acid	Tot
F.17	$2.6 \cdot 10^{-5}$	–	$2.0 \cdot 10^{-2}$	$6.6 \cdot 10^{-2}$	$1.5 \cdot 10^{-1}$	$3.7 \cdot 10^{-3}$	–	$2.4 \cdot 10^{-1}$
F.23/steel	–	–	–	–	–	–	–	–
F.23/concrete	–	–	–	–	–	–	–	–
O.23	$2.0 \cdot 10^{-5}$	$3.9 \cdot 10^{-3}$	$1.2 \cdot 10^{-3}$	$3.3 \cdot 10^{-4}$	–	$7.8 \cdot 10^{-3}$	–	$1.3 \cdot 10^{-2}$
R.23/steel	–	–	–	–	–	–	–	–
R.23/concrete	–	–	–	–	–	–	–	–
S.21	–	–	–	–	–	–	–	–
S.23	–	–	$7.0 \cdot 10^{-3}$	$2.8 \cdot 10^{-3}$	$6.0 \cdot 10^{-3}$	$1.4 \cdot 10^{-3}$	–	$1.7 \cdot 10^{-2}$
S.09	–*	–	$2.0 \cdot 10^{-2}$	$7.8 \cdot 10^{-3}$	$1.7 \cdot 10^{-2}$	$3.8 \cdot 10^{-3}$ *	–	$4.8 \cdot 10^{-2}$
O.01	$7.2 \cdot 10^{-6}$	–	$4.2 \cdot 10^{-4}$	$1.2 \cdot 10^{-4}$	–	$2.7 \cdot 10^{-3}$	–	$3.3 \cdot 10^{-3}$
R.01	–	–	–	–	–	–	–	–
R.10	$1.0 \cdot 10^{-5}$	–	–	–	–	–	$1.3 \cdot 10^{-3}$	$1.3 \cdot 10^{-3}$
R.15	$7.4 \cdot 10^{-6}$	–	–	–	–	–	$9.5 \cdot 10^{-4}$	$9.5 \cdot 10^{-4}$
B.05	$6.8 \cdot 10^{-5}$	–	–	$6.8 \cdot 10^{-2}$	–	–	–	$6.8 \cdot 10^{-2}$
F.05	$1.0 \cdot 10^{-4}$	–	$7.9 \cdot 10^{-2}$	$2.6 \cdot 10^{-1}$	$5.9 \cdot 10^{-1}$	$1.5 \cdot 10^{-2}$	–	$9.4 \cdot 10^{-1}$
F.15	$8.7 \cdot 10^{-6}$	–	$6.8 \cdot 10^{-3}$	$2.2 \cdot 10^{-2}$	$5.1 \cdot 10^{-2}$	$1.3 \cdot 10^{-3}$	–	$8.1 \cdot 10^{-2}$

\* calculations based on amounts from a recently performed study (Johansson C, 2001) resulted in concentrations of  $4.8 \cdot 10^{-4}$  M Na-gluconate and  $3.9 \cdot 10^{-4}$  M EDTA (in 56 of the S.09 moulds).

\*\* Sorption will reduce the calculated concentrations

In BMA the highest concentrations are found in F.17, S.09, B.05 and F.05 for citric acid, oxalic acid and Na-gluconate. Na-gluconate and also sodium capryliminodipropionate (NKP) are stronger complexants than the organic acids (concentrations of about  $10^{-3}$  M). Na-gluconate has been found to sorb strongly to cement. Calculations for ISA (Section 3.2.2) showed that the concentration will be reduced with at least four orders of magnitude due to sorption to cement. The reduction will be similar or even higher for gluconate, leading to concentrations of  $<10^{-6}$  M for all waste types in BMA where cement is used to stabilise the waste (O.23, S.23, S.09, O.01 and F.15).

In the waste packages deposited in the Silo, calculated concentrations are lower than in BMA. The highest concentrations are found in F.18 for citric and oxalic acid (about  $10^{-2}$  M). Due to sorption in the cement waste matrix, the concentration of Na-gluconate in O.02 will be reduced below the critical level. The concentration of gluconate in F.18 will not be reduced as this waste type is not conditioned in cement. The effects on sorption are further discussed in Chapter 3.4.

In BTF there are only two waste types receiving substances originating from chemicals used at NPP. The concentrations are about  $10^{-3}$ - $10^{-2}$  M.

EDTA is present at relatively low concentrations in the waste packages in all repository parts.

**Table 3.2.5** *Calculated concentrations (M) of chemicals in different waste types in the Silo*

M (mole/l)	EDTA	NKP	NTA	Citric acid	Oxalic acid	Na-gluconate	Picolinic acid	Tot
O.24	–	–	–	–	–	–	–	–
O.24	–	–	–	–	–	–	–	–
S.22	–	–	–	–	–	–	–	–
S.11	–	–	–	–	–	–	–	–
O.02	$9.7 \cdot 10^{-6}$	$3.2 \cdot 10^{-4}$	$9.9 \cdot 10^{-5}$	$2.8 \cdot 10^{-5}$	–	$6.5 \cdot 10^{-4}$	–	$1.1 \cdot 10^{-3}$
R.02	–	–	–	–	–	–	–	–
R.16	$9.9 \cdot 10^{-6}$	–	–	–	–	–	$7.0 \cdot 10^{-4}$	$7.1 \cdot 10^{-4}$
B.06	$9.8 \cdot 10^{-5}$	–	–	$9.7 \cdot 10^{-4}$	–	–	–	$1.1 \cdot 10^{-3}$
F.18	$1.7 \cdot 10^{-5}$	–	$5.1 \cdot 10^{-3}$	$1.7 \cdot 10^{-2}$	$3.8 \cdot 10^{-2}$	$9.4 \cdot 10^{-4}$	–	$6.0 \cdot 10^{-2}$

**Table 3.2.6** *Calculated concentrations (M) of chemicals in different waste types in BTF*

M (mole/l)	EDTA	NKP	NTA	Citric acid	Oxalic acid	Na-gluconate	Picolinic acid	Tot
O.07	$3.3 \cdot 10^{-5}$	$3.0 \cdot 10^{-3}$	$9.1 \cdot 10^{-4}$	$2.5 \cdot 10^{-4}$	–	$5.9 \cdot 10^{-3}$	–	$1.0 \cdot 10^{-2}$
B.07	$3.3 \cdot 10^{-5}$	–	–	$1.5 \cdot 10^{-3}$	–	–	–	$1.5 \cdot 10^{-3}$

### 3.3 Estimated amounts

Total amounts of identified complexants deposited in different parts of a filled repository were calculated. To obtain the total amounts of chemicals and cellulose (ISA) in the waste, the amounts per mould were multiplied with the total number of deposited moulds in a filled repository (BMA, Silo, BTF, BLA). The total number of moulds in the different parts of SFR has been estimated in a prognosis by SKB (Riggare and Johansson, 2001).

#### 3.3.1 Cellulose

The total amount of cellulose originating from the waste, cement additives and from chemicals are shown in Table 3.3.1 for the repository parts BMA and the Silo in SFR. In BMA the total amount of cellulose in the waste is  $1.1 \cdot 10^5$  kg and in the Silo  $9.0 \cdot 10^3$  kg. The cellulose loading in BLA is not reported as this cellulose will not form ISA due to the absence of cement (alkaline conditions). In BTF no cellulose is deposited. However, a limited amount of tall fatty acid from chemicals used at the nuclear power plants is present in waste packages B.07 (4 kg in total).

**Table 3.3.1 Estimated total amounts (kg) of cellulose to be deposited in BMA and Silo**

	Total number of moulds*	Waste	Cement additives		From chemicals**
			Cement waste matrix	Concrete packaging	
F.17	879	3164	–	–	–
F.23/steel	270	40500	–	–	–
F.23/concrete	54	1566	32	–	–
O.23	784	23520	470	549	–
R.23/steel	486	21384	–	–	–
R.23/concrete	343	3773	–	240	–
S.21	1493	11795	–	–	–
S.23	61	1769	–	–	–
S.09	382	–	–	–	–
O.01	738	–	1205	517	–
R.01	1702	–	–	1191	–
R.10	136	–	–	–	–
R.15	295	–	–	–	–
B.05	4230	–	–	–	140
F.05	1718	–	–	–	–
F.15	11	–	–	–	–
<b>BMA tot (kg)</b>	<b>13582</b>	<b>108086</b>	<b>1716</b>	<b>2497</b>	<b>140</b>
O.24	150	5250	–	–	–
O.24	–	–	–	–	–
S.22	430	3440	–	–	–
S.11	96	288	–	–	–
O.02	3532	–	5766	2472	–
R.02	705	–	–	7544	–
R.16	3733	–	–	–	–
B.06	6902	–	–	–	3
F.18	938	–	–	–	–
<b>Silo tot (kg)</b>	<b>16486</b>	<b>8978</b>	<b>5766</b>	<b>10016</b>	<b>3</b>
<b>SFR tot (kg)</b>	<b>30068</b>	<b>117064</b>	<b>7482</b>	<b>12513</b>	<b>143</b>

\* Riggare and Johansson, 2001

\*\* Tall fatty acid (assumed to produce ISA as cellulose)

### 3.3.2 Other substances

The total amounts of substances originating from chemicals used at the Swedish nuclear power plants are reported in Table 3.3.2. Citric acid is present in greatest amounts. The amounts of complexants have been calculated using the estimated amount per mould and the total number of deposited moulds in SFR (Riggare and Johansson, 2001, see Table 3.3.1). EDTA has not been used after 1998 and picolinic acid only during 1998.

**Table 3.3.2 Estimated total amount (kg) of chemicals used at NPP and deposited in different parts of SFR**

	EDTA <sup>s</sup>	NKP	NTA	Citric acid	Oxalic acid	Na-gluconate	Picolinic acid <sup>s</sup>	Cellulose <sup>**</sup>	Tot
F.17	0.2	–	288	949	1007	55	–	–	2299
O.23	0.3	98	19	5	–	127	–	–	249
S.23	–	–	9	3	3	2	–	–	17
O.01	0.5	96	19	5	–	125	–	–	246
S.09	0.2	–	55	22	22	12	–	–	109
R.10	–	–	–	–	–	–	0.1	–	–
R.15	0.1	–	–	–	–	–	0.3	–	–
B.05	3	–	–	1768	–	–	–	140	1911
F.05	1	–	562	1855	1968	107	–	–	4494
F.15	–	–	4	12	13	0.7	–	–	29
<b>BMA tot</b>	<b>3</b>	<b>194</b>	<b>955</b>	<b>4621</b>	<b>3013</b>	<b>427</b>	<b>0</b>	<b>140</b>	<b>9353</b>
O.02	0.7	105	20	6	–	136	–	–	268
R.16	0.6	–	–	–	–	–	2	–	3
B.06	2	–	–	38	–	–	–	3	44
F.18	0.2	–	154	510	541	29	–	–	1235
<b>Silo tot</b>	<b>4</b>	<b>105</b>	<b>175</b>	<b>554</b>	<b>541</b>	<b>166</b>	<b>2</b>	<b>3</b>	<b>1549</b>
O.07	2	279	55	15	–	364	–	–	715
B.07	0.9	–	–	51	–	–	–	4	56
<b>BTF tot</b>	<b>3</b>	<b>279</b>	<b>55</b>	<b>66</b>	<b>–</b>	<b>364</b>	<b>–</b>	<b>4</b>	<b>771</b>
<b>SFR tot</b>	<b>10</b>	<b>481</b>	<b>600</b>	<b>3368</b>	<b>1573</b>	<b>725</b>	<b>3</b>	<b>147</b>	<b>6759</b>

<sup>s</sup> EDTA only used until 1998. For S.09 56 moulds in total assumed for total amounts of EDTA (Johansson C, 2001); Picolinic acid only used during 1998.

\* Sodium capryliminodipropionat

\*\* tall fatty acid

### 3.4 Effects on radionuclide solubility and sorption

In this section waste types with potential critical concentrations of ISA and other complexants are discussed in more detail.

#### 3.4.1 ISA

##### *Important waste types*

Effects on sorption of radionuclides (tetravalent) will be seen at concentrations of isosaccharinic acid above  $10^{-4}$  M. This limit is only exceeded in waste type F.17, F.23/steel, S.21 in BMA and in S.22 in the Silo (assuming a yield of 0.1 mole/kg cellulose). In B.05 in BMA the concentration is equal to the critical level ( $1 \cdot 10^{-4}$  M). Waste types F.17 and B.05 are bituminised and consequently do not contain cement as the other waste types. Neither S.21 nor S.22 is conditioned in cement. If the integrity of

the mould is not maintained, elements may form complexes with ISA leading to an increased mobility of elements. ISA will be produced after the intrusion of alkaline water. The concentration may be reduced to levels below effect levels if cement surrounding the waste is available for ISA to sorb onto. In S.21 and S.22 cement is present near the actual waste (in between steel drums). If cement phases are not available for sorption of ISA in S.21 and S.22, sorption reduction may be seen for bivalent nuclides as well, the concentration being higher than  $1 \cdot 10^{-2}$  M. This level is not exceeded in F.17.

In F.23/concrete and O.23 critical levels are slightly exceeded in the actual waste, but not when considering sorption and dilution in the concrete mould surrounding the waste (Table 3.2.1).

B.05 does not contain pure cellulose in the waste. The concentration of ISA has been calculated assuming that the tall fatty acid present is degraded as cellulose. This is a conservative approach. The concentration of the fatty acid itself is  $8.3 \cdot 10^{-3}$  M, which is below effect levels for organic acids with 2-3 carboxylic groups (compare with e.g. citric acid in Section 2.3.1).

The concentration calculations assuming a more conservative case of ISA production (using the yield after 1000 years of cellulose degradation; i.e about  $\sim 2.5\%$ ) lead to relatively small changes in concentration. Critical levels were exceeded in four new waste types, though only slightly (F.23/concrete, O.23 and R.23/steel in BMA and O.24 in the Silo). When considering sorption and dilution in the concrete mould surrounding these waste types, concentrations of ISA will stay below critical levels (except in R.23/steel). The concentrations are at a maximum twice as high as the concentrations based on the lower yield (see Appendix B).

The calculations performed for a yet higher yield, corresponding to about 10 000 years of degradation (10%), showed that critical levels of ISA may arise in all waste types. This is however a highly conservative approach (see Appendix B).

It should be emphasised that high concentrations of isosaccharinic acid will not develop in F.17, S.21 and S.22 unless alkaline water enters (the moulds are not conditioned in cement). Intrusion of water is only possible if the mould is damaged. Moreover, it will take some time for the water to reach the entire waste, especially in the bituminised matrix.

During the intrusion of water, there will also be a simultaneous leakage of ISA from the mould. This diffusion process will reduce the concentration in the pore water. For example, regarding a linear yield and diffusion during the first 100 years of degradation, the actual concentration in S.21 will be approximately  $3 \cdot 10^{-3}$  M (compare with  $1.6 \cdot 10^{-1}$  M).

When not taking sorption of ISA to cement into consideration, critical concentrations are exceeded for the majority of the waste packages.

The calculated concentrations of ISA are somewhat conservative due to several factors of which some have been described above. Assumptions probably leading to an overestimation of the levels of ISA or other complexants are:

- all cellulose is assumed to be available for degradation
- diffusion of ISA out of the moulds is not taken into consideration (see calculation for S.21 above).
- the chemical stability of ISA is not regarded. If ISA is subject to degradation, the concentration in the repository will be reduced, thus reducing negative effects on radionuclide sorption to cement.

Although already having formed complexes with ISA, sorption to cement of the ISA-metal complex itself may reduce the mobility of radionuclides. Sorption of ISA complexes to cement is not included in the calculations, but the process is reported to occur according to recent research.

### ***Leakage of ISA to other waste packages***

It is important to study if the leakage of ISA from waste types with critical levels may affect the mobility of elements in other parts of BMA. Calculations showed that addition of 2 kg cement around each waste package would be needed to reduce the concentration under effect levels (30% porosity assumed in cement). If no cellulose additives is present in the cement (0.4 % of the cement weight) about 1.5 kg cement is needed. Dilution of the ISA will further decrease the concentration, thus initially high concentrations of ISA in some waste types will only have a local effect on the radionuclide mobility.

### ***Cellulose based cement additives***

An analysis was performed to evaluate the effect on radionuclide sorption due to the presence of cellulose based additives in cement used as backfill. Calculations resulted in a concentration of ISA of  $1.6 \cdot 10^{-5}$  M in the cement, i.e. levels below critical concentrations.

### **3.4.2 Other complexants**

The calculated concentrations of EDTA, NTA, citric acid, oxalic acid, gluconate and other substances mainly emanating from decontamination chemicals, were compared with experimentally derived effect levels. Chemicals for which effects on sorption are not known (picolinic acid, NKP, etc) were related to complexing agents with known effects (citric acid, EDTA). Thus, sodium capryliminodipropionat (NKP), a relatively strong complexing ligand, will be compared to EDTA. Picolinic acid will be compared to citric acid.

Critical concentrations are exceeded in some waste types receiving chemicals. In BMA, levels are high in e.g. F.17, O.23, F.05 and B.05 for citric acid, NTA and gluconate (see Table 3.4.1). The calculated concentrations of gluconate are higher than  $1 \cdot 10^{-4}$  M, the level where effects have been seen for Eu. However, due to sorption to cement it is unlikely that concentrations this high will arise. Additional calculations demonstrated that the gluconate concentrations would be reduced under effect levels for all waste types except for F.17 and F.05 (bituminised waste) when taking sorption to cement into consideration.

NKP is believed to be a strong complexant (similar structure as EDTA) and the evaluation of sorption effects is made through comparison with effect levels for EDTA ( $>1 \cdot 10^{-4}$  M). In the Silo, the concentration of NKP in O.02 is in the range where sorption effects may occur. NKP in O.07 (BTF) will also exceed the critical concentration.

The calculated concentrations of chemicals in five of the waste packages in BMA are somewhat overestimated (F.05, F.17, F.15, O.01 and O.23). When distributing the amount of chemicals used at Forsmark and Oskarshamn during 1998, the annual production of only F.17 and O.23 was used (F.05, F.15 and O.01 are no longer produced, i.e. no annual production). Considering fewer moulds will lead to a higher amount per mould (see Section 3.1.2).

**Table 3.4.1 Waste packages with concentrations that exceed the critical levels  $1 \cdot 10^{-2}$  M and  $1 \cdot 10^{-4}$  M for different chemicals**

Waste package	Substance	Concentration (M) <sup>§</sup>	Risk of sorption reduction
F.17, F.05	NTA/Citric acid/ Na-gluconate	$>1 \cdot 10^{-2}$ / $>1 \cdot 10^{-2}$ / $>1 \cdot 10^{-4}$	Mn, Ni, Pb/Mn/ Eu <sup>***</sup>
O.23, O.02	NKP <sup>*</sup>	$>1 \cdot 10^{-4}$	Mn, Ni, Pb
S.09	EDTA <sup>**</sup> /NTA	$>1 \cdot 10^{-4}$ / $>1 \cdot 10^{-2}$ /	Mn, Ni/ Mn, Ni, Pb
B.05, F.15	Citric acid	$>1 \cdot 10^{-2}$	Mn
F.18	Citric acid/Na-gluconate	$>1 \cdot 10^{-2}$ / $>1 \cdot 10^{-4}$	Mn/Eu <sup>***</sup>
O.07	NKP <sup>*</sup> /Na-gluconate	$>1 \cdot 10^{-4}$	Mn, Ni, Pb/Eu <sup>***</sup>

\* assumed critical levels are the same as for EDTA

\*\* in 56 of the moulds

\*\*\* few experiments found in this literature search on Na-gluconate/gluconic acid

§ see Section 3.2.3 for the exact concentrations

The presence of EDTA will probably not reduce the sorption of radionuclides in any of the waste types. A newly performed study of S.09 presented a content of about 4 g EDTA in each of 56 moulds of this waste type (Johansson C, 2001). This amount results in an EDTA concentration of  $3.9 \cdot 10^{-4}$  M, i.e. slightly above the critical level where sorption of Ni and Mn may be affected.

Although the calculated concentrations exceed critical levels in some waste types, the literature only indicates effects on sorption for Ni, Pb and Mn (see Table 2.3.2). As to Na-gluconate, sorption of this substance to cement will reduce the calculated concentrations in Tables 3.2.4-3.2.6 under effect levels for those waste types that contain cement. Data is scarce on the effects of gluconate on sorption of different elements, although the sorption of Eu has been shown to be initially reduced.

To summarise, sorption may be reduced for some elements (mainly Mn) in some waste packages (mainly F.17, B.05 and F.05). Depending on the actual importance of the relatively high concentrations of NKP and gluconate, yet more elements may be affected, especially in waste package O.23 and O.07. However, it should again be pointed out that the estimated concentrations in F.17, F.05 and O.23 may be overestimated.

## 4 Conclusions

The calculations performed in this report show that the presence of complexants (ISA and other substances) in SFR may lead to such high concentrations that sorption of certain elements may be affected in some waste types. As to ISA, critical concentrations may arise mainly in four waste packages not conditioned in cement (F.17, F.23/steel and S.21 in BMA and S.22 in the Silo). The high ISA concentrations in S.21 and S.22 will probably affect both bi- tri- and tetravalent elements.

By adding a small amount of cement around the waste packages during deposition in BMA, any excess of ISA leaching from the mould will sorb to the cement phases, thus decreasing the concentration of ISA. This will prevent effects on the mobility of radionuclides present in other parts of the repository. Furthermore, recent experiments indicate that the ISA complexes sorb to cement and that the reduction in sorption at high concentrations of ISA may be due to saturation of sorption sites by uptake of ISA. This means that sorption on cement/concrete in the surrounding of the waste packages may be unaffected due to lower ISA-concentrations there. According to the calculations, the use of cellulose based cement additives will lead to ISA concentrations below critical levels, thus not affecting the sorption of radionuclides.

Due to the presence of complexants in chemicals (mainly sodium caprylimino-dipropionate, NTA and Na-gluconate), critical concentrations may arise in some waste types in BMA, the Silo and BTF (F.17, F.18, O.23, O.07, O.02, S.09, B.05, F.15 and F.05). Elements probably to be affected are mainly Mn and in some cases Ni, Pb and Eu. Na-gluconate may also affect the mobility of other elements, but current knowledge is limited to Eu. For the waste types in BLA the presence of complexing agents has not been studied as no credit is given in the safety assessment to sorption in this repository part.

It should be pointed out that the estimates of concentrations in different waste types are based on a number of conservative assumptions, thus the actual concentrations may be lower than the calculated.

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## Appendix A - Complexing agents in general and in SFR

*Table A-1 Potential complexing agents in SFR and comments on their relevancy and importance for the safety assessment in SFR*

Complexing agent/group\$	Source*	Comment	Usage/amounts to be deposited	Con- sidered
EDTA (Ethylene-diamine-tetra-acetate)	Chemicals NPP	Strong	Small amounts, no longer used, ~10 kg	yes
Citric acid	Chemicals NPP	Relatively strong (tri-)	1900 kg	yes
(Tall) fatty acid ("Tallfetttsyra")	Chemicals NPP	Cellulose based => ISA	15 kg, small amount in comparison to other org matter	Yes
NTA (Nitroltriacetate)	Chemicals NPP	Strong (weaker than EDTA and fosfonates)	800 kg	yes
Lignine, Lignosulphates, lignine sulphonates (Cementa Flyt M97, PA in Betokem)	Cement additives	Cellulose based => ISA	1000 kg	yes
Cellulose or hydroxypropylmethyl-cellulose (Plastiment BV40, Methocel)	Cement additives	Regarded as cellulose => ISA	5200 kg	yes
Oxalic acid/ iron oxalate	Chemicals NPP (degradation of bitumen, CER#)	Medium strength (dicarboxylic)	600 kg/1200 kg	yes
Gluconic acid/ Na-gluconate	Chemicals NPP	Strong	Not used/ 700 kg	yes
Picolinic acid	Chemicals NPP	Relatively weak (one carboxylic group)	3 kg	Yes
Cellulose or cellulose derivatives	waste	Degrades to ISA	See this report	yes
ISA	Deg. of cellulose	strong	See cellulose etc in this table	yes
DTPA (Diethylene-triamine-penta-acetate)	Decont.	strong	Not used	No
Maleinic acid (Sikament 10)	Cement additives	Investigations are carried out	10-12 kg/mould	no
Amines	Deg. of AER#	Negligible effects	-	no
Inorganic ligands (CO <sub>3</sub> , NO <sub>3</sub> , SO <sub>4</sub> , S, PO <sub>4</sub> , CN, B(OH) <sub>4</sub> , F, I, Cl, Br)		Little importance, except CN (see below)	In waste and water	no
Hydrazin	Corrosion inhibitors	No importance (form H <sub>2</sub> , H <sub>2</sub> O)	-	no
Acetic acid		weak	See ethylacetate	no
Ethylacetat		weak	Small amounts	no

Carboxylates	from degradation of exchange resins, etc	weak, especially in alkaline environment		no
Fumaric acid			Not used	no
Other organic acids	Degradation of bitumen, polymeres, etc in decont.	Relatively weak		no
CMC (carboxymethyl-cellulose)	-	-	Not used	no
Nitrit		No importance	only small amounts	no
"Vinsolharts"/ ligninesulfonates (Barra 55)	Cement additives	Relatively strong	Small amounts	no
Cyanid complexes		Relatively strong (ferrocyanides weak)	Small/no amounts	no
Melamine formaldehyde (Peramin F and L)	Cement additives	Relatively weak	Used, amount not known	no
DBP, MBP		No importance for waste to SFR (conditioning)	-	no
Akrylates	-	poly- relatively strong	Possibly used but not identified in SFR	no
Boric acid, Borax		no complexing properties		no
Akrylamides		no complexing properties		no
Na-citrat			discharge in ocean	no
Tartaric acids, Alkyl sulphonic acid, Substituted phenoles			Not identified in SKB 99/13	no
Phosphates	Chemicals NPP	Strong	Used, amounts not known	no

\* Some of the above mentioned complexants may have more sources than the tabulated

# CER = Cation exchange resins, AER = Anion exchange resins \$ Based on SKB 99/10, 99/13, personal communication, guideline to NPP from SKB (products with potentially complexing substances) and other national and international literature on complexants.

**Appendix B - Calculated concentrations of ISA in SFR using three different yields**

**Table B-1** *Calculated concentrations of ISA in waste types in SFR using three different yields of ISA. Sorption of ISA to cement in the conditioned waste and the concrete packaging (waste+mould) is considered*

Degradation	1-10 years		~1000 years		~10 000 years	
	0.1 mole/kg (1.8 %)		0.14 mole/kg (2.6%)		0.56 mole/kg (~10%)	
Yield	Waste	Waste + mould	Waste	Waste + mould	Waste	Waste + mould
<b>BMA</b>						
F.17	$4.2 \cdot 10^{-3}$		$6.1 \cdot 10^{-3}$		$2.4 \cdot 10^{-2}$	
F.23/steel	$3.3 \cdot 10^{-4}$		$6.1 \cdot 10^{-4}$		$6.2 \cdot 10^{-2}$	
F.23/concrete	$1.2 \cdot 10^{-4}$	$4.7 \cdot 10^{-5}$	$1.9 \cdot 10^{-4}$	$7.1 \cdot 10^{-5}$	$3.9 \cdot 10^{-3}$	$4.1 \cdot 10^{-4}$
O.23	$1.2 \cdot 10^{-4}$	$5.0 \cdot 10^{-5}$	$2.0 \cdot 10^{-4}$	$7.5 \cdot 10^{-5}$	$4.5 \cdot 10^{-3}$	$4.6 \cdot 10^{-4}$
R.23/steel	$6.9 \cdot 10^{-5}$		$1.0 \cdot 10^{-4}$		$8.0 \cdot 10^{-4}$	
R.23/concrete	$3.9 \cdot 10^{-5}$	$1.8 \cdot 10^{-5}$	$5.8 \cdot 10^{-5}$	$2.6 \cdot 10^{-5}$	$3.1 \cdot 10^{-4}$	$1.2 \cdot 10^{-4}$
S.21*	$1.6 \cdot 10^{-1}$	$2.1 \cdot 10^{-4}$	$2.3 \cdot 10^{-1}$	$3.6 \cdot 10^{-4}$	$8.8 \cdot 10^{-1}$	$2.6 \cdot 10^{-2}$
S.23	$3.1 \cdot 10^{-5}$	$2.2 \cdot 10^{-5}$	$4.5 \cdot 10^{-5}$	$3.2 \cdot 10^{-5}$	$2.2 \cdot 10^{-4}$	$1.5 \cdot 10^{-4}$
S.09	-**	-	-	-	-	-
O.01	$6.1 \cdot 10^{-7}$	$7.6 \cdot 10^{-7}$	$8.7 \cdot 10^{-7}$	$1.1 \cdot 10^{-6}$	$3.4 \cdot 10^{-6}$	$4.2 \cdot 10^{-6}$
R.01	-	$2.5 \cdot 10^{-7}$	-	$3.6 \cdot 10^{-7}$	-	$1.4 \cdot 10^{-6}$
R.10	-	-	-	-	-	-
R.15	-	-	-	-	-	-
B.05	$1.0 \cdot 10^{-4}$		$1.5 \cdot 10^{-4}$		$5.7 \cdot 10^{-4}$	
F.05	-	-	-	-	-	-
F.15	-	-	-	-	-	-
<b>SILO</b>						
O.24	$4.0 \cdot 10^{-5}$	$2.8 \cdot 10^{-5}$	$5.9 \cdot 10^{-5}$	$4.1 \cdot 10^{-5}$	$3.2 \cdot 10^{-4}$	$2.0 \cdot 10^{-4}$
O.24	$7.8 \cdot 10^{-5}$	$4.3 \cdot 10^{-5}$	$1.2 \cdot 10^{-4}$	$6.4 \cdot 10^{-5}$	$1.0 \cdot 10^{-3}$	$3.5 \cdot 10^{-4}$
S.22*	$1.6 \cdot 10^{-1}$	$2.2 \cdot 10^{-4}$	$2.3 \cdot 10^{-1}$	$3.7 \cdot 10^{-4}$	$8.9 \cdot 10^{-1}$	$2.8 \cdot 10^{-2}$
S.11	$9.4 \cdot 10^{-7}$		$1.4 \cdot 10^{-6}$		$5.3 \cdot 10^{-6}$	
O.02	$6.1 \cdot 10^{-7}$	$7.6 \cdot 10^{-7}$	$8.7 \cdot 10^{-7}$	$1.1 \cdot 10^{-6}$	$3.4 \cdot 10^{-6}$	$4.2 \cdot 10^{-6}$
R.02	-	$3.8 \cdot 10^{-6}$	-	$5.5 \cdot 10^{-6}$	-	$2.2 \cdot 10^{-5}$
R.16	-	-	-	-	-	-
B.06	$1.5 \cdot 10^{-6}$		$2.1 \cdot 10^{-6}$		$8.1 \cdot 10^{-6}$	
F.18	-	-	-	-	-	-
<b>BTF</b>						
O.07	-	-	-	-	-	-
B.07	$2.2 \cdot 10^{-6}$	$1.0 \cdot 10^{-6}$	$3.2 \cdot 10^{-6}$	$1.5 \cdot 10^{-6}$	$1.2 \cdot 10^{-5}$	$5.7 \cdot 10^{-6}$

\* Waste not conditioned, cement between two steel drums

\*\* These waste types do not contain cellulose in waste or packaging

**Appendix C - Waste characteristics in SFR (BMA, Silo and BTF): used volumes and amounts of cement**

**Table C-1 Waste characteristics in BMA**

	CEMENT				VOLUMES				TOTAL VOLUMES		
	Waste		Mould		Waste		Mould		Waste + Cond waste	Waste + mould	Total
	Cement + water	Cement	Cement	Cement	Waste cement	Porosity in cement	Porosity in mould material				
kg	m <sup>3</sup>	kg	kg	m <sup>3</sup>	m <sup>3</sup>	m <sup>3</sup>	m <sup>3</sup>	m <sup>3</sup>	m <sup>3</sup>	m <sup>3</sup>	
F.17 Bitumen	0	0	0	0	0.085	0	0	0.085	0	0.085	0.085
F.23/steel Cement	1356	0.68	407	0	0.085	0.136	0	0.221	0	0.221	0.221
F.23/concrete Cement	565	0.28	170	214	0.05	0.0565	0.109	0.107	0.109	0.107	0.216
O.23 Cement	565	0.28	170	214	0.05	0.0565	0.109	0.107	0.109	0.107	0.216
R.23/steel Cement	1356	0.68	407	0	0.085	0.136	0	0.221	0	0.221	0.221
R.23/concrete Cement	565	0.28	170	214	0.05	0.0565	0.109	0.107	0.109	0.107	0.216
S.21 *	96	0.048	0	29	0.005	0	0.0096	0.0050	0.0096	0.0050	0.0146
S.23 Cement	565	0.28	565	214	0.05	0.0565	0.1092	0.107	0.1092	0.107	0.216
R.01 Cement	1400	0.70	1400	214	0.15	0.14	0.1092	0.290	0.1092	0.290	0.399
R.10 Cement	1120	0.56	1120	214	0.1	0.112	0.1092	0.212	0.1092	0.212	0.321
R.15 Cement	2100	1.05	2100	0	0.085	0.21	0	0.295	0	0.295	0.295
B.05 Bitumen	0	0	0	0	0.032	0	0	0.032	0	0.032	0.032
S.09 Cement	280	0.14	280	0	0.01	0.028	0	0.038	0	0.038	0.038
F.05 Bitumen	0	0	0	0	0.0216	0	0	0.0216	0	0.0216	0.0216
F.15 Cement	805	0.40	805	0	0.17	0.081	0	0.251	0	0.251	0.251

\* Waste not conditioned, cement between two steel drums

**Table C-2 Waste characteristics in the Silo**

	CEMENT				VOLUMES				TOTAL VOLUMES			
	Waste		Mould		Waste		Mould		Waste		Total	
	Cement + water	Cement	Cement	Cement	Waste	Porosity in cement	Porosity in mould material	Cond waste	Waste + mould	Waste + mould	Waste + mould	
kg	m <sup>3</sup>	kg	kg	m <sup>3</sup>	m <sup>3</sup>	m <sup>3</sup>	m <sup>3</sup>	m <sup>3</sup>	m <sup>3</sup>	m <sup>3</sup>	m <sup>3</sup>	
O.24	Cement	1780	0.89	534	214	0.01	0.178	0.109	0.188	0.297	0.297	
O.24	Cement	1000	0.5	300	214	0.01	0.1	0.109	0.110	0.219	0.219	
S.22	*	96	0.048	0	29	0.005	0	0.0096	0.005	0.0146	0.0146	
S.11	Cement	1820	0.91	1820	0	0.085	0.182	0	0.267	0.267	0.267	
O.02	Cement	1540	0.77	1540	214	0.15	0.154	0.1092	0.304	0.413	0.413	
R.02	Cement	1400	0.7	1400	214	0.15	0.14	0.1092	0.290	0.399	0.399	
R.16	Cement	2100	1.05	2100	0	0.085	0.21	0	0.295	0.295	0.295	
B.06	Bitumen	0	0	0	0	0.03	0	0	0.03	0.03	0.03	
F.18	Bitumen	0	0	0	0	0.17	0	0	0.17	0.17	0.17	

\* Waste not conditioned, cement between two steel drums

**Table C-3 Waste characteristics in BTF**

	CEMENT				VOLUMES					TOTAL VOLUMES		
	Waste		Mould		Waste	Porosity in	Mould	Waste	Cond	Waste +	Total	
	Cement + + water	Cement water	Cement	Cement	Waste	cement	Porosity in mould material	Porosity in mould material	waste	waste	mould	mould
O.07	0	0	0	1656	0.5	0	0	0.6	0.5	0.5	1.1	1.1
B.07	0	0	0	1656	0.5	0	0	0.6	0.5	0.5	1.1	1.1

\* waste not conditioned, concrete mould