Long term aspects of landfilling and surface disposal - lessons learned from nuclear and non-nuclear decommissioning, remediation and waste management

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Abstract

The fields of landfilling of conventional waste and that of surface disposal of nuclear waste have developed quite independently and also partly out of phase with each other. The paper analyses what knowledge and experience might be mutually beneficial as well as what further knowledge may be needed.

It is found that even though knowledge may exist, and information from lessons learned elsewhere be available, action may be subject to considerable initiation or incubation times. Legislation on financial reporting is summarized and its implications for early technical and financial planning are assessed. Prerequisites for long-term behaviour are analysed for the waste forms as well as for the seals and covers. The rationale for using natural and anthropogenic analogues is compiled, and alternative seals for landfills are analysed based on this information. Lessons learned from nuclear decommissioning are presented, and the difficulties encountered when the decommissioning takes place long times after commissioning and operation of a facility are illuminated. Comparison is made with contaminated soil in which area openly available domestic publications are lass abundant in some areas. The differences between end of license and end of responsibilities are clarified.

Uranium-containing waste is presented as an example. Prerequisites are presented for natural uranium together with its progenies and for depleted uranium, initially without any daughters. It is found that both alternatives are associated with a number of issues to consider, and that both call for long-term containment for conventional chemical hazard and radiological hazard reasons.

Keywords: Landfilling, surface disposal, shallow land burial, long-term, leaching, legislation, financing, financial reporting, prediction, natural analogues, decommissioning, contaminated soil, end of license, uranium, mill tailings, Ranstad, depleted uranium.

1. Introduction and objective

1.1 Introduction

Substantial efforts are presently being made on research in the areas of nuclear as well as non-nuclear (conventional) waste management. In many respects, the prerequisites and needs are different, e g with regard to nuclear criticality and reduction of the potential for detriment to man and the environment by radioactive decay. In other respects, and especially in the area called surface disposal in the nuclear area as well as landfilling in the conventional area, the needs for knowledge and for pertinent strategies with regard to long term aspects may be rather similar.

Consequently, one would expect comprehensive co-ordination of the efforts in the two areas. However, in many respects the various efforts are out of phase as well as incoherent.

For example, after more than a decade of intensive leach testing in the nuclear area, (Chapman and McKinley, 1987) concluded that "*the only reliable means of providing data for release modelling*" is to replicate disposal conditions in terms of chemical environment and time, and that leaching using pure water for a short time can only be used in order to "*compare the overall quality of waste forms*".

Nonetheless, twelve years later, the European Union issued a (EU Directive, 1999) stating that waste should be qualified for disposal according to a leach test using distilled water and lasting for 24 hours. After the Swedish ban on landfilling of organic matter, the most abundant waste form is probably ash from combustion and incineration. The appropriate standard as well as the Swedish implementation of the Directive state that the test is not applicable to materials that react with the leachant, and that equilibrium conditions should be sought for in such cases. Nonetheless, the accredited laboratories routinely carry out the test anyway without even making a comment about it in their analysis protocols.

Of course, ash, and especially fly ash, age on contact with water and air, and it has recently been shown (Sjöblom, 2011) that just one week of ageing - but preferably longer - may give rise to an order of magnitude decrease in the leach-rates for copper, lead and zinc.

The incoherence is particularly noticeable in cases where waste is

- conventionally hazardous as well as radioactive, and
- where the level of radioactivity is low and short-lived such that surface disposal / landfilling (as opposed to geological disposal) may appear as the preferable alternative

In concordance, there is a need to compare the approaches to surface disposal and to landfilling and to identify mutually beneficial knowledge and experience. Experience shows that there are two areas in particular that warrant attention, namely long term performance and long-term planning. Reasons for this include the following:

1. Comparison between allowed concentration in waste that may be deposited on a landfill for non-hazardous waste according to the (EU Directive 2008) with allowed leaching according to (EU Directive 1999) may give the numerical result, for a 10 metres high landfill, that depletion with regard to a hazardous substance may take up to a million years. (This figure is hypothetical since the next glaciation is expected to appear within 100 000 years).
2. Planning for long-term decommissioning and remediation has proven to be notoriously difficult from a financial as well as from a technical perspective (Lindskog and Sjöblom, 2008, 2009).

1.2Objectives and scope

The objective and scope of the present paper are to compare nuclear and non-nuclear waste management with the aim to:

- identify and describe circumstances and events leading to the present differences in approach between conventional and nuclear waste management
- identify what knowledge can be shared
- share some lessons learned
- present and discuss the example of uranium-containing waste
- identify areas in which further research is warranted
- identify pertinent strategies

2. The significance of trends

Human versatility implies ability to act as individuals as well as in cohorts. Especially the latter may, however, be associated with considerable initiation or incubation times. Trends are by no means any monopoly of fashion design. For instance, the greenhouse effect has been documented in scientific as well as in popular and well circulated literature (Arrhenius, 1919) for more than 100 years, but it is only in recent years that it has become a general concern.

X-rays were discovered in the 1990's and came to wide-spread practical use in medicine within only a few years. Unfortunately, high doses of ionising radiation may cause cancer long after the exposure, and this was discovered decades later in a number of tragic instances. Consequently, the hazards of ionising radiation were quite well known when the first (anthropogenic) nuclear reactor was started in 1942. However, there was essentially no experience of man-made radio-nuclides, and it took decades before their implications became studied with any intensity.

During the early years ending in the mid-70's, AB Atomenergi was responsible for a rather ambitious nuclear technology development programme to which our Government alone contributed around 1,55 G€ (Lindskog and Sjöblom, 2008). A total of 517 reports were published during the years 1956 to 1977, two of which deal with nuclear waste. A shift of paradigm came with a Government enquiry in the mid1970's (Swedish Government, 1976), and subsequently, the users of nuclear electricity have paid a total of around 2 G€ on nuclear waste research (see further below).

Studies of the events (Lindskog and Sjöblom, 2008) show that much of the knowledge base required to deal with the waste problem existed already in the 1950's, and also that the Government enquiry (Swedish Government, 1976) stands the test of time surprisingly well.

Consequently, it is important to consider that time is likely to catch up on issues not adequately dealt with at present, but where financing and research is warranted. The findings below in the present paper support a conclusion that long-term aspects of landfilling may constitute an attractive candidate in this regard.

3. The requirements from society

3.1The Environmental Code

The general requirements from society are summarized in the Swedish Environmental Code, and the following can be found in Chapter two.

- Take adequate protective measures and other precautions to avoid detriment to health and the environment
- Sufficient knowledge

- Compliance with the Polluter Pays Principle (PPP)
- Use of Best Available Technology (BAT)
- Comply with (all other) legislation

A first observation is that is the operator / owner / license holder that has the full and undivided responsibility for protection of health and the environment. The duty of the Authorities and Courts is to instigate and oversee compliance. Licenses to operate do not lift any of these basic requirements.

It should also be noted regarding PPP that the law does not indicate any upper level for the costs involved, nor does it indicate any limit in time. Moreover, the responsibility is a collective one among the parties involved.

The statements apply collectively. Thus, if BAT is not sufficient for adequate protection of health and the environment, then new knowledge will have to be found and technology developed.

3.2 Requirements on research and development

Since 1987, and under the Act on Nuclear Activities, the nuclear industry in Sweden has been obligated to submit, every third year, a comprehensive programme for the research and development needed for the safe management of the nuclear waste and for the decommissioning of the nuclear facilities. The programmes have been reviewed by the Swedish National Council for Nuclear Waste, the competent Authorities, and others, and input has been provided to support completeness and quality of the knowledge base. The research and development work has been conducted by the Swedish Nuclear Fuel and Waste Management Company (SKB), who in March this year (2011) submitted an application to build a final repository for spent nuclear fuel. It is presently under Authority review (Dverstorp et al., 1011).

No similar legislation process for ensuring sufficiently comprehensive research has been identified in the areas of landfilling and remediation of contaminated soil. It can be observed, however, that the level of financing on the non-nuclear side is more than one order of magnitude lower in Sweden.

3.3 Legislations on financing

In Sweden, the financing of the nuclear waste management as well as that of decommissioning of the nuclear facilities is assured under the Nuclear Liability Act. According to this Act, funds are set aside whilst the nuclear reactors and other facilities are in operation in order to cover all future costs. Moreover, securities are also provided in order to cover the uncertainties in the estimates. Such legislation has been in force since around the late 70's.

Financing of the final environmental liabilities associated with landfills is covered under the Environmental code by means of securities.

3.4 Legislation on financial reporting

All environmental liabilities as well as their precise levels must be reported under the Swedish Annual Reports Act, which for larger enterprises refers to the International Financial Reporting Standard (IFRS/IAS) issued by the International Accounting Standards Board (IASB). The international reporting standard (IASB, 2010) as well as an ASTM standard (ASTM, 2007a) provide relatively detailed instructions as to how the liabilities are to be evaluated. They require that exact figures be provided even for complex undertakings perhaps decades into the future. But they do also include the possibility of uncertainty, in which case alternative outcomes must be identified and evaluated together with their relative probability.

The penal law requires that an "*essentially correct financial situation*" be presented. Noncompliance may lead to up to six years in prison for those responsible, see (Lindskog and Sjöblom, 2009) for further detail.

4. Long-term performance and planning

4.1 The complexity of the issue and requirements on early planning

The requirements from society summarised in Section 3 imply that the following objectives will have to be achieved:

1. Methods for landfilling, including installation of final covers, have to be available. The installations must function during the length of time needed with regard to the content of hazardous substances and acceptable leach rates to the environment. This is something that the implementer will have to be able to show in such a manner that it will be accepted by the authorities and the courts.
2. Technical as well as financial planning have to be carried out in such a way that objective (1) above will be met. This must apply also in cases where the final closure may lay decades ahead into the future.

The objectives (1) and (2) above are closely interlinked. Achievement of objective (1) presupposes that the planning according to objective (2) be sufficiently elaborate in order for such cost estimates to be made that are sufficiently precise in order to allow accumulation of adequate funding to cover all future costs during the time when the benefits of the facility in question are reaped. This frequently implies that technical planning, including the associated research required, has to be made long before the operations required to achieve end of responsibility status are to be carried out.

Thus, the two issues of long term performance and long term planning are closely interlinked and are therefore dealt with together in the following.

4.2 Prerequisites for predicting long-term behaviour

In general, landfilling of organic matter is prohibited in Sweden, and consequently most of the waste deposited comprises oxides of metallic elements. It is frequently assumed among environmental chemists that the minor elements - some of which are regarded as "contaminants" - form phases in which they are major elements. It has, however, been well known among inorganic chemists, mineralogists and geologists for decades that this is often not the case and that the "contaminants" instead are incorporated into the phases formed by the major elements in the form of solid solution. This makes them far less accessible, especially after ageing, than what might be assessed using standard thermodynamic calculations software. Further information on this issue can be found in (Sjöblom and Noläng, 2011).

The kinetics aiming towards thermodynamic equilibrium are even much more difficult to estimate from theory or by semi-empirically based methods. Although there are co-variations between bonding energies and rates of reactions, at least for similar cases, there exists no general correlation. For example, the activation barrier to forming and breaking of hydrogen bonds in monomethylammonium chloride decreases from about 32 to 4 kJ/mole in a phase transition while the strength of the bonding - as evidenced by infrared data - remains approximately the same. (Sjöblom, 1975)

Rates of reaction are frequently assumed to follow a relationship given by the Arrhenius' equation. A number of conditions have to be met in order for this to be valid, including that the reaction depends on only a single mechanism. Moreover, the mechanism or mechanisms governing the rates may be very different for different ranges of parameters. Consequently, one needs to be able to prove that the mechanisms are the same if extrapolation is to be made outside the range of parameters. This is typically very difficult to prove in practice.

The considerations should also include some treacherous phenomena such as stress corrosion and depletion of inhibitors, in which cases little may be observed for a long time after which catastrophic break downs may take place.

The present state of knowledge regarding kinetics in chemistry thus strongly indicates that knowledge bases intended to be used for predictions on long-term behaviour should preferably include natural and anthropogenic analogues.

4.3 Natural and anthropogenic analogues

Literature searches on natural and anthropogenic analogues unveils a wealth of scientific papers in the area of nuclear waste, but few responses on disposal of conventional waste. Only some brief points are made in the following.

An outline of the general strategy applied can be found in (Poinssot and Gin, 2012), see also references therein. It is put forward that a specific approach has been progressively determined by the scientific community in order to understand and describe the evolution of waste forms and barrier materials. The approach is labelled "*long-term behaviour science*" and relies on a combination of experimental and modelling approaches. Natural and anthropogenic analogues are essential for the identification of key mechanisms as well as for benchmarking. Recommendations in this regard can be found in an ASTM standard (ASTM, 2007b).

Papers related to the Swedish nuclear waste programme include (Liu et al., 1996; Smellie et al.; 1997, Smellie et al.; 1999, Bruno et al., 2002). According to the Swedish nuclear waste programme, the spent fuel is to be put in composite canisters having iron on the inside for mechanical strength and copper on the outside for corrosion resistance. The canisters are to be put in holes in crystalline rock with bentonite clay in between.

Some countries have policies for reprocessing of the spent nuclear fuel, and in such cases the fission products are stabilised into a glass form (Poinssot and Gin, 2012). Reference (Sjöblom et al., 2011a) deals with nuclear waste glass as well as with glass from melting of conventional incinerator ash, and puts forward vitrified forts as an anthropogenic analogue.

Examples of nuclear waste forms and barrier materials used are shown in Table 1 together with the associated types of natural and anthropogenic analogues.

Table 1.

4.4 Landfilling

In Sweden, there is a ban on landfilling of organic materials. At the same time, about half of our domestic waste is being incinerated with recovery of the energy (Flyhammar, 2011). Thus much of the waste being deposited at present comprise inorganic material.

The long-term behaviour of a landfill depends on the combined developments in the waste and in the barriers. So far, focus has been on the most immediate concerns, namely emissions to air and water, but there is also a growing interest in the evolution of the waste form over time, see e. g. (Brännvall, 2010) and references therein.

Of course, it should be discussed which waste may require the most long-term containment, nuclear or conventional. Nuclear waste may decay over time and thus lose its potential for harming health and the environment. Contaminants in conventional waste may become stabilised, especially in cases where the "matrix" is reactive such as is frequently the case for ashes. It can be concluded, though, that long term containment may be required in either case depending on the particular circumstances.

It cannot generally be expected that the waste forms alone can provide the containment necessary in order to protect health and the environment for the length of time required. Consequently, landfills are supplied with covers and seals that are intended to provide the protection required, e. g. against percolating water. A few examples may be as follows (Sjöblom et al., 2011b; Rihm et al., 2009):

- Geomembranes are made of e. g. polyethylene or polyvinyl chloride. Presence of antioxidants is frequently important for their stability, and the rate of deterioration may increase considerably when the antioxidants have become consumed. Stress corrosion is also an issue as well as brittle behaviour for low loads over long times.
- Geo clay liners are made of two sheets of synthetic fabric with bentonite clay in-between. The two sheets are joined by either needle punching or stitching. The bentonite clay contains the mineral montmorillonite (sodium rich type) which swells strongly on contact with water, thus forming an efficient seal. The long

term shear strength depends on the ageing properties of the polymer material joining the two sheets. Bentonite itself is sensitive to chemicals, including salt. The installations are usually sensitive to differential settlements of the underlying waste. See e. g. (Brundin et al., 2001; Meer, 2007).

- Natural clays can provide a considerable chemical buffer capacity, but have in most cases higher hydraulic conductivity as compared to bentonite, Natural clays can show variations in properties. Sources for suitable clays are scarce in Sweden.
- Ashes from combustion of wood based fuels are recycled materials that may be compared with natural clays in terms of chemical buffer capacity and hydraulic conductivity. Details can be found in (Sjöblom and Tham, 2009). It might be added that no literature has been found on the influence of salt in a landfill seal, but general literature on soil suggests that the hydraulic conductivity might increase if the salt is lost. The following considerations may be required in order to meet the requirements on imperviousness (Tham and Andreas, 2008): selection of ash (e. g. grain size distribution), additive (e. g. water), compaction procedure and possible storage time.
- Mixtures of ashes and activated sewage sludge constitute recycled materials and may form tight seals in the short term. However, claims of long-term stability have been repudiated based on anthropogenic and natural analogues (Sjöblom and Tham, 2009).

Availability to natural and anthropogenic analogues for the waste forms listed above can be found in Table 2.

Table 2.

4.5 Decommissioning of nuclear facilities

It has been known since at least the mid-70' that nuclear facilities will have to be decommissioned, and that the cost may be on the order of 15 % of that for new build. It is only relatively recently, however, that decommissioning has become an integral part of the planning prerequisites for new nuclear facilities.

There are many lessons learned in this area including the necessity to perform radiological surveying, method selection, and identification of potential complications and cost raisers. The accumulated experience is that it is considerably more treacherous to make appropriate technical and financial planning for decommissioning of a facility as compared to new build.

Special attention is warranted in cases where there is a substantial difference in time between the operation of a facility and the subsequent decommissioning. It is often a challenge for technically oriented people to realise that it may be necessary to make technical plans now for financial purposes although they may not be needed for technical reasons for perhaps decades, see e. g. (Lindskog and Sjöblom, 2008, 2009).

It is essential that design prerequisites and requirements be well known before the waste is being deposited. This enables and facilitates appropriate waste management in terms of e. g. sorting, treatment and packaging. Insufficiencies in this regard might necessitate waste archaeology, in which case our clear experience is that it is usually much easier to do it all correctly from the beginning, see e. g. (Lindskog and Sjöblom, 2008, 2009).

4.6 Landfilling and contaminated soil

No case has been found in Sweden in which landfill covers constructed in accordance with modern legislation have had to be remediated. This situation is expected since all such installations are quite new, and since defects might be difficult to identify.

However, Sweden as many other countries, has a considerable legacy in terms of contaminated sites that need remediation. The Swedish Environmental Protection Agency (EPA) is responsible for the financing of such remediation that refers to pollution that has occurred before the year 1969. Around 50 M€ are paid each year for such purposes. A literature search was conducted in order to find out if the experience here was similar to that in the nuclear area, but no comprehensive reporting was found. This is hardly surprising in view of the fact that the Swedish National Audit Office, see (Riksrevisionen, 2011), found no reporting with comparison between predicted and incurred costs for the remediation projects.

International sources (Fogleman, 2005), see Chapter 15, unveil that in 1979, US EPA estimated that remediation of sites posing a significant risk to health and environment would cost around 6 G US $. Today, according to the same source, some estimates exceed 1 T US $.

The Swedish EPA has, however, commissioned a study (Carlsson, 2004) with the purpose of facilitating estimation of costs for covering landfills. Such estimates are needed in order for appropriate levels of securities to be decided, c. f. Section 3.3. The Carlsson report refrains from making any prognoses for costs in the long term on the grounds that it is too difficult.

It appears in (Carlsson, 2004) that many companies in Sweden do not declare any environmental liabilities in spite of the fact that they may be obligated by law. Curiously, those who do declare use taxed assets. The reason for this, according to (Carlsson, 2004), is fear for the tax authorities. This practice is surprising in view of the fact that there is no support for it in the tax legislation. The issue was dealt with by the Swedish Government already in 1977 when it concluded in a proposition that money set aside to cover environmental liabilities should not generate taxation. (Lindskog and Sjöblom 2008; Söderberg 2005) The proposition became law during the subsequent year, and still today there is no taxation in our system for covering nuclear liabilities using segregated funds. Another possible reason for the setting aside of taxed assets may be that no infringement is made to the bonuses of the managements.

4.7 End of license versus end of responsibilities

It was mentioned in Section 4.1 that the environmental liability is a collective responsibility. This means that the legal system is free to sue anyone involved (e. g. operator or owner) for all or part of the liability. It might therefore be tempting to conclude that the requirements on early planning are moderate.

Lessons learned tell a different story as is further described in (Lindskog and Sjöblom, 2011). A glance at the list of the enterprises on the stock market today and a couple of decades ago clearly indicates that there might not be anyone around to sue. Early planning is also required for other purposes, not least for the assurance of adequate funding.

It is important to realise that end of licence is not the same as end of responsibilities. End of license can take place as a result of licence expiration, decision of the owner / operator, or as a result of Authority action when license conditions have not been met.

End of responsibility may be decided by the competent Authority when all the environmental liabilities have been dealt with appropriately and in full.

5. Uranium and uranium containing waste

5.1 Chemical toxicity and radio-toxicity

Uranium containing waste has been generated in the form of tailings from uranium mining and beneficiation, and from use of munitions that contain uranium. Uranium and uranium compounds may be harmful to man through their chemical toxicity as well as through the radio-toxicity of uranium together with that of its progenies (daughter radio-nuclides). Limits for exposure of soluble uranium compounds may be based on chemical toxicity, and those for insoluble compounds on radio-toxicity (Lewis, 1996). Chemical effects of uranium and uranium-containing compounds include kidney damage, which may not be reversible.

The legislation in Sweden on classification of waste into hazardous and non-hazardous waste is at the time of writing (December 2011) still based on the national legislation under the European Union Dangerous Substances Directive (DSD) and Dangerous Preparations Directive (DSD). Here, the oxides UO_2, U_3O_8 and UO_3 all have the risk phrases R 26/28 (very toxic to inhalation and if swallowed). This implies that these compounds, together with equivalent other forms, may occur at most at a level of 0.1 %, or else the waste must be regarded as hazardous (Sjöblom et al., 2005, 2006; Sjöblom and Noläng, 2011, and references therein). In addition, the compounds have the risk phrases R33 (danger of cumulative effects) and R 51/53 (toxic to aquatic organisms, may cause long-term adverse effects in the aquatic environment).

It can be expected that the classification of waste will soon instead be based on the new European Union regulation on labelling, CLP. Under CLP, the same oxides all have the hazard statements H300, H330, H373 and H411 (which have about the same meaning as the risk phrases above). It is not to be expected that the change will lead to any less strict classification of waste.

Natural uranium comprises the isotopes U-234, U-235 and U-238 which have abundances of 0.005, 0.720 and 99.274 %, respectively. The half-life of U-235 is $0,70 \cdot 10^9$ years, and that of U-238 is $4.47 \cdot 10^9$ years. U-235 is the only naturally fissile isotope, and consequently, natural uranium is enriched in U-235 before it is used in our modern reactors utilising fission by means of thermal neutrons. Depleted uranium, low in U-235, is generated as a residue. The high density of 18950 kg/m^3 makes it attractive for use in munitions.

There are three naturally occurring decay series:

- Thorium series, starting with Th-232,
- Uranium series, starting with U-238, and
- Actinium series, starting with U-235

The actinium series is of little significance to health and environment in relation to that of the uranium series and is therefore not dealt with further in the following. The thorium series may contribute, and thorium frequently occurs in nature together with uranium. It is, however, generally less mobile. The thorium series is not dealt with further on the grounds of brevity and simplicity of the present paper. The long-term features of the U-238 series are shown in Table 3.

Table 3.

At equilibrium between the daughters, there is the same rate of the decays from each of the radio-nuclides involved (except for cases with alternative paths). Thus, the radio-nuclides in the tables, as well as the intermediates between them in the series, should contribute equally in terms of number of decays per second. But there are other important differences, such as type of radiation, energy involved and type of organ in a human that becomes irradiated. Some of these aspects are dealt with in practice by means of so-called dose factors. Moreover, the chemistry may vary considerably between mother and daughter.

5.2 Uranium mill tailings

An individual in a critical group living in the vicinity of uranium mine tailings receive a dose that depends on the half-lives of the nuclides in the decay chains, the chemical properties of the radio-nuclides involved, the character and energy of the radiation, the transport, and the form of uptake. In many or even all cases, it is concluded that the mother U-238 makes only a minor contribution to the total dose, and e. g. (Nair et al., 2010) conclude in one case that Rn-222, Po-210, Pb-210 and Ra-226 give rise to no less than 99,75 % of the total dose.

Estimation is treacherous. Alpha decays are associated with recoil effects implying that the decaying atom is thrown away from its position in its crystal structure and put e. g. in the pore water where it may end up in as a solute.

Uranium ore is frequently associated with reduced sulphur, typically in the form of pyrites. Pyrites exposed to air may oxidise to form sulphuric acid, which may give rise to acid mine drainage and associated widespread distribution of various heavy elements including uranium and its decay products. The phenomenon is largely due to microbial action and is autocatalytic in character.

All pyrite-containing tailings do not form acid drainage, and there are ways to prevent such developments. The requirement is that the rate of neutralisation must exceed that of oxidation of sulphur. Limestone is known to be very reactive in this regard. Consequently, addition of pH-buffering material in combination with covering or flooding to prevent oxidation constitute efficient remediation against acid mine drainage. See (Sjöblom and Noläng, 2011; Blowes, et al., 2005) and references therein.

It has been reported (Sjöblom et al., 1987) that strong complexing agents like gluconic acid can stabilize uranium in a dissolved form, and that even dilute chemistries containing this agent may dissolve uranium oxide fuel pellets in a couple of hours. It has been shown more recently that complexing agents with similar properties may be formed from organic matter by microbial action (Kalinowski et al., 2004; Edberg et al., 2010).

Microbes might influence mine tailings also in the absence of oxygen from the atmosphere since iron-III can be utilized as an oxidant (Landa et al., 1991). It should be noted, however, that microbial action typically requires inoculation and incubation, and that growth of microorganisms may be slow and their activity moderate under strongly reducing conditions.

For most radio-nuclides and transport situations, it can be assumed that transport takes place only for the radio-nuclides that are relative long-lived. There is an important exception, however. Radium-226 decays to radon-222 which has a half-life of only 3.825 days (Choppin et al., 2002). Radon is a noble gas and therefore associates itself with the atmosphere. If the gas phase in some tailings move, so will the radon until it decays, after which the radon daughters will transfer to any solid available.

These characteristics imply that uranium mill tailings may need not only protection with regard to percolating water, but also that transport of oxygen into the waste and of radon away from the waste may have to be hindered.

5.3 The Ranstad uranium mining and beneficiation facility

The Ranstad uranium mining and beneficiation facility was in full operation during 1965-1969. A total of 215 tonnes of uranium were produced from leaching of alum shale with sulphuric acid and subsequent liquid-liquid extraction. The ore contained on the average only 0.03 % uranium and consequently a million cubic metres of tailings were generated. It has been estimated that the residues contain about 100 tonnes of uranium and $5 \cdot 10^{12}$ Bq of radium-226. They cover an area of 25 hectares.(Sundblad, 1998)

According to (Strandell, 1998), a 2 metres high horizon in the rock contains about 1,5 % of the accessory mineral kolm. Kolm appears in fist-sized lumps and contain as much as 70-75 % organic matter and 0,3 % uranium. Thus, according to the modern legislation, and if kolm was waste, it should be assessed to be hazardous waste. See decision M 4532-04 from our highest environmental court for further illumination of this issue.

During 1990-1992, the tailings (6-10 metres) were covered (from bottom to top) with 0.3 metres clay-moraine mixture, 0.2 metres of crushed limestone, 1.2 metres of moraine, and 0.2 metres of a soil-moraine mixture (Hultgren and Olsson, 1993; Sundblad, 1998). The tests were carried out on a mixture of moraine and bentonite clay, giving rise to 0.3-4.4 % of the precipitation percolating through the seal. The actual installations were, however, made using a local clay and the resulting rate of percolation became 10-15 % of the precipitation.(Sundblad, 1998) Assuming an annual rainfall of 700 millimetres, this corresponds to 70-105 litres per square metre and year, thus exceeding the present limits for hazardous and non-hazardous waste which are 5 and 50 litres per square metre and year, respectively.

It appears from (Sundblad, 1998) as if although that pilot scale tests included mixing of the tailings with limestone, this was not done for the main operation. Allard (Allard et al., 1991) reports that the calcium content in alum shale is only 0.9 % while that of sulphur is 7.0 %. It thus appears that the dimensioning hardly includes long-term buffering of the acid generated during the (at least initially) slow oxidation of the pyrite.

Some of the international experiences can be found in (Rofer and Kaasik, 1998; IAEA, 2004a, 2004b; Franklin and Fernandes, 2011).

5.4 Depleted uranium from munitions

The prerequisites are somewhat different for the case of depleted uranium (containing almost exclusively uranium-238). As is apparent from Table 3, hundreds of thousands of years will pass before appreciable amounts of radium have formed, and associated radon is being generated. This length of time is sufficient in order for the soil at the surface to either have been removed by wind and rain or have been covered by substantial layers of deposited soil.

The issue is thus largely limited to that of the chemical and radiological toxicity of uranium and with time also the less mobile thorium (see Table 3).

There are also further issues to consider. Uranium metal is pyrophoric, and its use in munitions imply diminution into very fine particles. Perhaps the comparison is overly cautious, but the experience from the Tjernobyl accident is that the fine particles in the fallout travelled through the soil together with the water from the rain and penetrated in a short time to a depth of one or more decimetres. After some decades, transport of cesium-137 in mineral soil takes place at a rate of only about one millimetre per year (Forsberg, 1999).

Thus, fine particles from the use of uranium containing munitions may blow away with the wind for some distance and also penetrate into soil in the presence of rain.

6 Conclusions and final remarks

The main conclusion are as follows.

- Awareness comes in trends. Actors in the area of landfilling and surface disposal need to foresee what may be reasonable bases for future trends.
- Long-term effects do not usually evidence themselves in the short-term, but have to be searched for in order to be found and identified sufficiently early.
- Timely action is essential, since "waste archaeology" and other unplanned remedial actions are usually much more costly than doing things right from the beginning.
- Identification of issues of interest and significance requires relatively detailed studies already at early stages.

- The fundamental difficulties of long-term predictions and the associated high value of comprehensive studies of anthropogenic and natural analogues should be fully realized.
- BAT may not be enough. There is also a requirement on sufficient knowledge.
- Lessons learned from completed projects in related areas (such as nuclear waste and decommissioning) can provide valuable input to the planning
- Frequently, the requirements on correct declaration of the financial situation are harsher than the technical ones with regard to detailed and early planning.
- In many cases, it should be the need for financial planning that determines the timing of the technical planning.
- Long-term environmental liabilities are debts that we owe to future generations. It is essential that such liabilities be correctly balanced against financial assets which can be used at the time when they are needed. Such assets do not represent any income and should consequently not be taxed.
- End of responsibilities takes place when all obligations have been fulfilled. It is entirely different from end of license.
- Uranium containing waste has chemical toxicity as well as radio-toxicity, both of which call for long-term containment.
- For tailings from mining and beneficiation, uranium-238 needs to be considered together with all of its daughters (including the noble gas radon-222).
- For depleted uranium is should be sufficient to consider uranium and thorium.
- The combination of cover and waste form should provide the long-term containment required.

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References

Allard, B., Arsenie, I., Håkansson, K., Karlsson, S., Ahlberg, A.-C., Lundgren, T., Collin, M., Rasmuson, A. and Strandell, E., 1991. Effects of weathering on metal releases from an engineered deposit for alum shale leaching residues. Water, air and soil pollution, 57-58, pp. 431-440.

ASTM, 2007a. Standard Guide for Disclosure of Environmental Liabilities. ASTM Standard E 2173 – 07, April 2007.

ASTM, 2007b. Prediction of the long-term behavior of materials, including waste forms, used in engineered barrier systems (EBS) for geological disposal of high-level radioactive waste, ASTM Standard CI 174-07, June 2007.

Arrhenius, S., 1919. Kemien och det moderna livet. Hugo Gebers Förlag, Stockholm. German translation: Die Chemie und das moderne Leben. Autoris. deutsche Ausgabe von B. Finkelstein. Lpz., Akadem. Vlgsanst., 1922. English translation: Chemistry in Modern Life. Translated from the Swedish and revised by Clifford Shattuck Leonard, New York, D. van Nostrand Co., 1925.

Blowes, D. W., Ptacek, C. J. and Jambor, J. L. and Weisner, C. G., 2005. The geochemistry of acid mine drainage. In Lollar, B. S., editor. Environmental geochemistry. Treatise on geochemistry, Volume 9. Elsevier.

Brundin, H., Kihl, A., Lagerkvist, A., Pusch, R., Sjöblom, R. and Tham G.,2001. Långtidsegenskaper hos tätskikt innehållande bentonit. (Long-term properties of seals containing bentonite. In Swedish). Avfall Sverige – Swedish Waste Management, RVF Rapport 01:12.

Bruno, J., Duroa, L., and Grive, M., 2002. The applicability and limitations of thermodynamic geochemical models to simulate trace element behaviour in natural waters.Lessons learned from natural analogue studies. Chemical Geology, 190, pp 371-393.

Brännvall, E., 2010. Accelerate ageing of refuse-derived-fuel (RDF) fly ashes. Licentiate thesis. Luleå University of Technology.

Carlsson, B., 2004. Ekonomisk säkerhet vid deponering. (Economic security for landfilling, in Swedish). Envipro Miljöteknik AB, on commission by the Swedish EPA.

Chapman, N. A. and McKinley, I. G., 1987. The geological disposal of nuclear waste. John Wiley & Sons.

Choppin, G. R., Liljenzin J-O, and Rydberg, J., 2002. Radiochemistry and nuclear chemistry. Butterworth - Heinemann.

Dverstorp, B., Strömberg, B. and Simic, E., 2011. Licensing review of a spent nuclear fuel repository in Sweden. 13th International High-level Radioactive Waste Management Conference, Albuquerque, New Mexico, United States, April 10-14, 2011.

EU Directive 1999/31/EC on the landfill of waste.

EU Directive 2008/98/EC on waste.

Flyhammar, P., 2011. One decade of dramatic changes of the Swedish management of household waste. Sardinia 2011, Thirteenth International Waste Management and Landfill Symposium, 3 - 7 October 2011, S. Margherita di Pula (Cagliari), Sardinia, Italy.

Fogleman, V., 2005. Environmental liabilities and insurance in England and the United States. Witherby & Co Ltd, London. ISBN 1-85609-303-4.

Franklin, M. R. and Fernandes, H. M., 2011. Identifying and overcoming the constraints that prevent the full implementation of decommissioning and remediation programs in uranium mining sites. Journal of Environmental Radioactivity, in press.

Edberg, F., Kalinowski, B. E., Holmström, J. M. and Holm, K., 2010. Mobilization of metals fromuranium mine waste: the role of pyoverdines produced by *Pseudomonas fluorescens*. Geobiology, 8, pp. 278-292.

Forsberg, S., 1999. Behaviour of ^{137}Cs and ^{90}Sr in agricultural soils. Doctoral thesis. The Swedish University of Agricultural Sciences, Agraris 212, 1999.

Hultgren, Å. and Olsson, G., 1993. Uranium recovery in Sweden, history and perspective. SKB Arbetsrapport 93-42. Swedish Nuclear Fuel and Waste Management Company.

IAEA, 2004a. Environmental contamination from uranium production facilities and their remediation. Proceedings of an international workshop, Lisbon, 11-13 February, 2004.

IAEA, 2004b. The long-term stabilization of uranium mill tailings. Final report of a co-ordinated research project. IAEA-TECDOC-1403.

IASB, 2010. International Financial Reporting Standards and International Accounting Standards (IFRS/IAS). International Accounting Standards Board.

Kalinowski, B. E., Oskarsson, A., Albinsson, Y., Arlinger, A., Ödegaard-Jensen, A., Andlid, T. and Pedersen, K., 2004. Microbial leaching of uranium and other trace elements from shale mine tailings at Ranstad. Geoderma 122, pp 177-194.

Landa, E. R., Phillips, E. J. P. and Lovley, D. R., 1991. Release of ^{226}Ra from uranium mill tailings by microbial Fe(III) reduction. Applied Geochemistry, Vol. 6, pp. 647-652.

Lewis, R. J. Sr., 1996 Sax's dangerous properties of industrial materials, 9th edition. Van Nostrand Reinhold.

Lindskog, S., and Sjöblom, R., 2008. Regulation evolution in Sweden with emphasis on financial aspects of decommissioning. Decommissioning Challenges: an Industrial Reality? Sept. 28 to Oct.2, 2008 – Avignon, France.

Lindskog, S. and Sjöblom, R., 2009. Radiological, technical and financial planning for decommissioning of small nuclear facilities in Sweden. Proceedings of the 12th International Conference on Environmental Remediation and Radioactive Waste Management, ICEM 2009, October 11-15, 2009, Liverpool, UK.

Lindskog, S. and Sjöblom, R., 2011. Division of nuclear liabilities between different license holders and owners. Proceedings of the 13th International Conference on Environmental Remediation and Radioactive Waste Management, ICEM 20011, September 25-29, 2011, Reims, France.

Liu, J., Yu, J.-W. and Neretnieks, I., 1996. Transport modelling in the natural analogue study of the Cigar Lake uranium deposit (Saskatchewan, Canada). Journal of Contaminant Hydrology, 21, pp 19-34.

Meer, S. L. and Benson, C. H., 2007. Hydraulic Conductivity of Geosynthetic Clay Liners Exhumed from Landfill Final Covers. Journal of geotechnical and geoenvironmental engineering, May, 550-563.

Nair R. N., Sunny, F. and Manikandan, S. T., 2010. Modelling of decay chain transport in groundwater from uranium tailings ponds. Applied mathematical modelling 34, pp. 2300-2311.

Poinssot, C. and Gin, S., 2012. Long-term behavior science: The cornerstone approach for reliably assessing the long-term performance of nuclear waste. Journal of Nuclear Materials, 420, pp 182-192.

Rihm, T., Svedberg, B., Eriksson, M. and Rogbeck, Y., 2009. Alternativa konstruktionsmaterial på deponier, vägledning. (Alternative construction materials for landfills, guidance). Avfall Sverige, Report U2009:08.

Riksrevisionen, 2011. Revisionsrapport 2011-01-18, Dnr 32-2010-0664. (Revision report regarding the Swedish Environmental Protection Agency, in Swedish).

Rofer, C. K. and Kaasik, T., editors, 1998. Turning a problem into a resource: Remediation and waste management at the Sillamäe site, Estonia. NATO Science Series. 1. Disarmament Technologies - Vol 28. Proceedings of the NATO Advanced Research Workshop on Turning a problem into a resource: Remediation and waste management at the Sillamäe site, Estonia, 5-9 October, 1998.

Sjöblom, R., 1975. Hydrogen Bond Studies 112. Molecular reorientations in some hydrogen bonded solids. Acta Universitatis Upsaliensis, Abstracts of Uppsala dissertations from the Faculty of Science 350.

Sjöblom, R., Olson, P. M., Parke, J. M. and Schneidmiller, D., 1987. Post-accident chemical decontamination method development, Final Report. EPRI Research Project RP 2012-8, EPRI Report number EPRI-NP-4999, January 1987 (Concerns the primary system in the reactor Three Mile Island 2).

Sjöblom, R., Tham, G., Haglund, J-E. and Sjöö, C., 2005. Environmental qualification of ash from wood-based recycled fuels for utilization in covers for landfills. Kalmar ECO-TECH ´05 and The Second Baltic Symposium on Environmental Chemistry. Kalmar, Sweden November 28-29.

Sjöblom, R., Tham, G., Haglund, J-E. and Ribbing, C., 2006. Classification of waste according to the European Union Directive 91/689/EEC on hazardous waste from a Swedish application perspective. CIWM Conference 12th – 16th June 2006, Paignton, Torbay, UK.

Sjöblom, R., 2011. Lämplig metodik för grundläggande karakterisering av aska för acceptans på deponi. (Appropriate methodology for basic characterisation of ash for acceptance for landfilling). Avfall Sverige, Rapport U2011:22.

Sjöblom, R. and Noläng, B., 2011. Betydelsen av fast löslighet i järn(hydr)oxider för fastläggning av potentiellt miljöstörande ämnen i askor. (The significance of solid solution of iron (hydr)oxides for stabilisation of elements in ash which are potentially harmful to health and environment. Värmeforsk, Report 1198, November 2011.

Sjöblom, R., Ecke, H. and Brännvall, E., 2011a. Long-term stability of vitrified waste in natural environments. Submitted to International Journal of Sustainable Development & Planning.

Sjöblom, R., Lindskog, S. and Andreas, L., 2011b. Lessons learned from nuclear decommissioning and waste management relevant to end of responsibilities for landfills. Sardinia 2011, Thirteenth International Waste Management and Landfill Symposium, 3 - 7 October 2011, S. Margherita di Pula (Cagliari), Sardinia, Italy.

Sjöblom, R. and Tham, G., 2009. Anthropogenic and natural analogues for the development over time of mixtures of wood-based ash and activated sewage sludge. Sardinia 2009, 12th international waste management and landfill symposium. Margherita di Pula, Cagliari, Sardinia, 5-9 October, 2009.

Smellie, J. A., Karlsson, F. and Russell Alexander, W. A., 1997. Natural analogue studies: present status and performance assessment implications. Journal of Contaminant Hydrology, 26, pp 3-17.

Smellie, J. A., Karlsson, F., 1999. The use of natural analogues to assess radionuclide transport. Engineering Geology, 52, pp 193-220.

Strandell, E., editor, 1998. Uran ur skiffer, Ranstadverket. (Uranium from shale, the Ranstad plant, in Swedish). Printing financed by AB SVAFO.

Sundblad, B., 1998. Remediation of the former uranium mine at Ranstad. In Rofer, C. K. and Kaasik, T., editors, 1998. NATO Science Series. 1. Disarmament Technologies - Vol 28. Proceedings of the NATO Advanced Research Workshop on Turning a problem into a resource: Remediation and waste management at the Sillamäe site, Estonia, 5-9 October, 1998.

Swedish Government, 1976. Spent nuclear fuel and radioactive waste. A summary report given by the Swedish Government committee on radioactive waste. AKA public investigation. (English summary of AKA Reports I-III) Department of Industry, SOU 1976:32. ISBN 91-38-02973-1.

Söderberg, O., 2005. In the shadow of the nuclear power debate around 1980 – thoughts on the birth of the finance system of today. (In Swedish). In Nuclear waste – costs and financing (Swedish title: Kärnavfall – kostnader och finansiering). Swedish National Council for Nuclear Waste. SOU 2005:83. ISBN 91-38-22439-9.

Tham, G. and Andreas, L., 2008. Utvärdering av fullskaleanvändning av askor och andra restprodukter vid sluttäckning av Tveta Återvinningsanläggning (Results from a full scale application of ashes and other residuals in the final cover construction of the Tveta landfill). Värmeforsk Report 1064.

Table 1. Examples of nuclear waste forms and barrier materials used together with the associated types of analogues.

Type of material	Natural analogue?	Anthropogenic analogue?
Uranium oxide fuel	yes	No
Waste glass	Yes	Yes
Iron	Yes	Yes
copper	Yes	Yes
bentonite	Yes	No
Crystalline and other rock	Yes	No

Table 2. Examples of materials for seals in covers for landfills the associated types of analogues.

<table>
<tr><td>Type of material</td><td>Natural analogue?</td><td>Anthropogenic analogue?</td></tr>
<tr><td>Geomembranes</td><td colspan="2">No. Geomembranes have been used for only a few decades and no analogues are available.</td></tr>
</table>

Geo clay liners	Natural analogues exist for bentonite clay. Geo clay liners depend for their function (shear resistance) also on polymers for which there are no analogues.	No
Natural clays	Yes	No
Ashes from combustion of wood based fuels	Yes. Natural cements and other natural high pH occurrences	Yes. Roman cement and mortar
Mixtures of ash and activated sewage sludge	Yes. Sea floors. Does not support claim on longevity, see main text.	Yes. Historic waste heaps. Does not support claims on longevity, see main text.

Table 3. The main long-term features of the uranium-238 series, after (Choppin et al., 2002).

Mother nuclide	Daughter nuclide	Decays	Effective half-life, years
U-238	U-234	$\alpha + 2\beta$	$4.5 \cdot 10^9$
U-234	Th-230	α	$2.5 \cdot 10^5$
Th-230	Ra-226	α	$7.5 \cdot 10^4$
Ra-226	Pb-210	$4\alpha + 2\beta$	$1.6 \cdot 10^3$
Pb-210	Pb-206	$2\beta + \alpha$	22