

**Restoration Techniques:
Characteristics and Performances**

Restoration Strategies for Radioactively Contaminated
Sites and their Close Surroundings
RESTRAT - WP3

Theo Zeevaert
SCK· CEN, Boeretang 200, B-2400 Mol,
Belgium

and

Andrew Bousher
Westlakes Scientific Consulting Ltd, Westlakes Science and Technology Park,
Moor Row, Cumbria, CA24 3LN, UK

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Executive Summary

TD.3+4: Restoration Techniques: Characterisation and Performance

The aim of this report is to present the results of a literature survey which identifies and quantifies techniques whose application would be most appropriate to remediating sites that have been contaminated by radionuclides from European nuclear installations. The purpose of this exercise is to create a database, which can be used by other working packages within the RESTRAT framework, to determine the impact of applying these technologies to specific example sites. In addition, the database will provide applicable information to restoration projects outside RESTRAT.

Remediation techniques are selected if they have been demonstrated to be applicable for treating sites which have been contaminated by radionuclides. Speculative approaches have been rejected. The techniques encompass physical-, chemical- and biological-based approaches.

The remediation techniques which were selected have been characterised in terms of the following:

- Description: The means by which they are applied.
- Applicability: The contaminants and the media for which they are suited; the length of time for which they would be applicable; and the manpower required to apply them.
- Performance: The effectiveness against the contaminants (radionuclides) and the time during which they remain effective.
- Cost: Capitals, operational and maintenance costs.
- Side effects: In particular, the production of waste.

A summary of these characteristics are given in Table A.

The cost and performance values associated with each remediation technique, can vary over a large range. This reflects the fact that these techniques have been applied to a variety of wastes, on a variety of scales and under a variety of conditions. This gives rise to a large degree of uncertainty which are taken into account when quantifying the application of these techniques to the example sites used in the RESTRAT programme.

Table A
Performance, cost and workforce exposure values of various remediation technologies

Technology	Medium	Performance Indicator		Cost (EUR)		Workforce exposure (manh)	Service life
Removal of Source		<i>Decontamination factor</i>		<i>Extraction (per m³)</i>	<i>Disposal & transport (per m³)</i>	<i>(per m³)</i>	
Soil excavation	Solid	1 - 20		50 - 150	450 - 800	0.2 - 1	not applicable
Soil scraping	Solid	1 - 20		(per m ²) 1 - 3	450 - 800	(per m ²) 0.03 - 0.3	not applicable
Physical separation		<i>Decontamination factor</i>	<i>Waste reduction</i>	<i>Excavation & separation (per m³)</i>	<i>Disposal & transport of residue (per m³)</i>	<i>(per m³)</i>	
Soil washing	Solid	1 - 10	50 - 98%	200 - 650	2000 - 3000	0.25 - 1.5	not applicable
Flotation	Solid	1 - 10	28 - 97%	65 - 390	2000 - 3 000	0.25 - 1.5	not applicable
		<i>Decontamination factor</i>		<i>Separation from liquid (per m³)</i>			
Filtration	Liquid	2 - >100		0.1 - 3.8	2000 - 3000	0.4 - 1.4	not applicable
Chemical separation				<i>Excavation & separation (per m³)</i>			
Chemical solubilisation	Solid	1 - 20		180 - 820	2000 - 3000	1.2 - 3.5	not applicable
				<i>Separation from liquid (per m³)</i>			
Ion exchange	Liquid	3 - 100(U), 20 - 100(Cs)		1.3 - 2.5	2000 - 3000	0.4 - 1.4	not applicable
Biological Separation							
Biosorption	Liquid	2.5 - >100		1 - 3	2000 - 3000	0.4 - 1.4	not applicable
Containment		<i>Resultant permeability (m s⁻¹)</i>		<i>Total (per m² surface area)</i>		<i>(per m² surface area)</i>	
Capping	Solid	1 × 10 ⁻¹² - 1 × 10 ⁻⁹		30 - 45		0.03 - 0.3	1,000 y
Subsurface barrier	Solid					(per m ³ barrier volume)	
a) slurry walls		1 × 10 ⁻¹² - 1 × 10 ⁻⁸		510 - 710		0.06 - 0.4	100 - 1,000 y
b) grout curtains		1 × 10 ⁻¹² - 1 × 10 ⁻⁸		310 - 420		0.06 - 0.4	100 - 1,000 y
Immobilisation		<i>Mobility reduction factor</i>		<i>Total (per m³)</i>		<i>(per m³)</i>	
Cement-based solidification	Solid						
a) <i>ex-situ</i>		5 - 25		75 - 300		0.25 - 1.5	not known
b) <i>in-situ</i>		5 - 25		50 - 310		0.06 - 0.4	not known
Chemical immobilisation	Solid						
a) <i>ex-situ</i>		5 - 50		110 - 570		0.25 - 1.5	not known
b) <i>in-situ</i>		5 - 50		60 - 420		0.06 - 0.4	not known

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1. Terms of Reference

This report is submitted as the Technical Deliverables 3 and 4 against the requirements of Work Packages 3.1 and 3.2 for the Restoration Strategies for Radioactive Sites and their Close Surroundings (RESTRAT) project.

The RESTRAT project, which is funded by the European Commission under the Nuclear Fission Safety Programme, has an overall objective of developing generic methodologies for ranking restoration techniques as a function of contamination and site characteristics. The development of this generic methodology is based on an analysis of existing remediation methodologies and contaminated sites, and is structured in the following steps:

1. The characterisation of relevant contaminated sites.
2. The identification and characterisation of relevant restoration techniques.
3. The assessment of the radiological impact.
4. The development and application of a selection methodology of restoration options.
5. The formulation of generic conclusions and development of the manual.

The two Work Packages are specifically concerned with the characterisation of the restoration techniques; Work Package 3.1 deals with physical restoration techniques and Work Package 3.2 deals with chemical and biological restoration techniques. The aims of the two Work Packages are as follows:

1. To identify possible relevant restoration (remedial) techniques, with relation to the base case.
2. To determine the characteristics (quantitatively if possible) for use in the risk assessment methodology and in the selection methodology. Characteristics considered are applicability; performance; economical costs and side-effects.

This Technical Deliverable summarises the findings of the two Work Packages. The results from both Work Packages have been combined into a single report for clarity and to allow a comprehensive characterisation of the different restoration techniques.

2. Introduction

A number of European nuclear installations are reaching the end of their designed life expectancies. Whilst the controlled decommissioning of the main installations is planned, the problem of contaminants dispersed throughout the site has not been properly addressed. In addition, contaminants may be contained by methods which are unsuitable for long-term storage. Many of these sites will require restoration in order to reduce the level of risk to the workforce, the public and the environment.

The types of sites, their sizes and the nature of their contamination varies widely. Also, the restoration techniques that can be applied, vary widely.

To date, there is no systematic method for identifying the most appropriate restoration technique for a contaminated site. It is the aim of the RESTRAT project to develop a methodology for achieving this, taking into account the principles of radiological optimisation. This means that the extent to which a site is to be restored and the means by which this is carried out, will be determined as a function of not only the radiological impact or risks to population and workers, but also economic (e.g. cost of the restoration, possible future land uses) and social factors (e.g. discomfort to the population, anxiety). Consequently, one of the main tasks in this project is to characterise the available restoration techniques in terms of:

- Description: the means by which they are applied.
- Applicability: the contaminants and the media for which they are suited; the length of time for which they would be applicable, and the manpower required to apply them.
- Performance: the effectiveness against the contaminants (radionuclides) and the time during which they remain effective. This is expressed by one of the following terms: the decontamination factor (i.e. fraction by which the level of contamination is decreased), the reduction factor (i.e. the percentage reduction in the volume of waste), reduction in radionuclide mobility, or reduction in the permeability of the waste.
- Cost: capital, operational and maintenance costs;
- Side effects: in particular, the production of waste.

The restoration techniques considered for a given contaminated site, will be assessed in terms of the above characteristics in order to determine their applicability and usefulness for that particular site. This will depend on the characteristics and future requirements of each site.

3. Classification of Restoration Techniques

In this report, restoration techniques are taken to be techniques (or measures) which prevent (or reduce) the radiological impact (or risks) to the population from the residual contamination of contaminated sites. Alternatively, it can designate techniques or measures which lessen the need for monitoring the environmental contamination.

Restoration, unlike remediation, does not include measures which affect human behaviour, dietary and living habits or working activities. In addition, the study excludes administrative measures such as restrictions of access to the site.

Measures or techniques suitable for dealing with large-scale agricultural or urban contamination are not considered in this study. They are covered by part of the concurrent TEMAS (TEchniques and MAnagement Strategies for environmental restoration and their ecological consequences) project within the Nuclear Fission Safety Programme of the EC, which is concerned with environmental restoration.

The framework of this report divides restoration techniques into four major categories:

- Removal of Sources: bulk removal of contaminated medium, sometimes followed by separation of the contaminants from the contaminated medium.
- Separation: techniques which separate radionuclides from the bulk of the waste.
- Containment: techniques which provide barriers between contaminated and uncontaminated media to prevent the migration of contaminants.
- Immobilisation: techniques which add material to the contaminated medium, in order to bind the contaminants and reduce their leachability or mobility.

Surface decontamination is not considered in this report despite the fact that it is a method for separating contaminants. This is because normally the technique is only efficient when applied at the early or intermediate phase of an accident and is largely restricted to urban contamination.

Table 1 subdivides these categories into specific technologies. The best-reported technologies for each category are identified below. These will be described and characterised in greater detail in later chapters.

3.1 Removal of Sources

These techniques are normally applied to contaminated soil. However, contaminated groundwater or surface water can also be removed (by pumping). The removal of the contaminated medium may be followed by a subsequent separation procedure (see Section 3.2).

Removal of contaminated soil can consist of:

- excavation (by excavators, bulldozers); or,
- removal of the upper layer(s).

The removal of the upper layers is mostly performed by soil scraping (with a shovel or a scraper) or by turf cutting (with a turf-cutter or harvester).

These techniques are well-developed and applied extensively in the field. They will be described in greater detail in Chapter 4.

3.2 Separation

Separation of contaminants from contaminated media can be carried out both *in-situ* and *ex-situ* (following excavation or removal of the contaminated medium).

Separation techniques may be subdivided into three main categories (Table 1):

- physical separation (mostly *ex-situ*);
- chemical separation (mostly *ex-situ*);
- biological separation (*ex-situ* and *in-situ*).

3.2.1 Physical Separation

Soil washing, flotation and filtration are well-developed physical separation techniques and have been applied extensively in the field (especially soil washing). They will be considered in Chapter 4.

The other physical separation techniques, listed in Table 1, have only been tested in some special cases and little relevant information (costs, efficiency) is available. For example, magnetic separation and electrokinetic separation have been applied in cases where conventional soil washing was found to be ineffective (mostly for heavy metals).

3.2.2 Chemical Separation

Solubilisation (including pH change, complexation, oxidation and reduction and solvent extraction) and ion exchange are well-developed techniques which have been used extensively. They will be considered in greater detail in Chapter 5.

Chemical leaching is a potentially effective separation technique. However, it is not considered further in this report as it is currently in the developmental stage and relevant data are not available.

3.2.3 Biological Separation

Phytoremediation, biosorption and bioleaching are all well-known biological separation techniques. These will be discussed in detail in Chapter 6.

3.3 Containment

Containment is primarily a physical technique which is achieved through the formation of physical barriers. Capping, using barriers placed on top of the surface, in order to prevent or reduce vertical infiltration, is very well developed and has been extensively documented (see Chapter 4).

Among the subsurface barriers, slurry walls and grout barriers have been applied with great success. They can be installed with the contaminated media in place or after excavation (encapsulation). They are discussed in detail in Chapter 4.

Vitrified barriers are not considered by this study. They are particularly useful for the containment of hazardous waste (e.g. high level waste, or mixed waste) because the vitrified mass is very resilient to weathering. However, the high cost inhibits their use for low level radioactive contaminants.

Cryogenic barriers are also not considered further in this study because of their high power costs and the fact that they are not passive protection systems (when the action of cooling is stopped, the barrier becomes ineffective).

Wet covers and dry barriers have only been used in special situations. Wet covers have been achieved through the inundation of mines. Dry barriers involve circulating dry air through subsurface soil which yields very low permeabilities. The latter technique is particularly applicable in arid environments. However, these techniques are not considered further because of their specificity and, also, because of the lack of relevant data.

3.4 Immobilisation

Immobilisation can be subdivided into three categories:

- physical immobilisation;
- chemical immobilisation;
- biological immobilisation.

3.4.1 Physical Immobilisation

Cement-based solidification/stabilization is a well developed physical immobilisation technique and has been well-documented. It is considered in greater detail in Chapter 4.

Vitrification is also a well-developed, mature technology, but will not be considered further in this report for low-level radioactive contaminants for the same reasons to those given in Section 3.3.

3.4.2 Chemical Immobilisation

Chemical immobilisation is a well-reported technique. It is considered in detail in Chapter 5.

Immobilisation techniques, such as chemical precipitation, adsorption and oxidation/reduction are not considered further by this report due to the lack of currently available data.

3.4.3 Biological Immobilisation

Biological immobilisation techniques, such as phytostabilisation, whilst offering potentially useful remediation techniques, are not considered further in this study. This is due to the lack of available data regarding their application, performance and cost.

Table 1 Classification of restoration techniques

<u>REMOVAL OF SOURCES</u>		
Solid	Excavation (bulk removal) Removal of upper layers:	Soil scraping Turf cutting
Liquid	Pumping	
<u>SEPARATION</u>		
Physical	Soil washing Flotation Filtration Magnetic separation Electrokinetic separation Thermal separation Aqueous biphasic separation	
Chemical	Leaching Solubilisation	pH change Complexation Oxidation/reduction Solvent extraction Ion exchange†
Biological	Ion-exchange‡ Phytoremediation Bioleaching Biosorption	
<u>CONTAINMENT</u>		
Physical	Capping with soil, clay, geomembrane, geosynthetic clay liner, asphalt, concrete Subsurface barriers Dry barriers Wet covers	Grout barriers Slurry walls Polymer gel Vitrification Land Encapsulation
<u>IMMOBILISATION</u>		
Physical	Cement-based solidification/stabilization Vitrification (<i>ex-situ</i> , <i>in-situ</i>)	
Chemical	Precipitation Adsorption Oxidation/reduction	oxide, carbonate, phosphate, sulphide clay, metal oxide, polymer
Biological	Phytostabilisation	

Notes: † Ion exchange as a chemical solubilisation technique refers to the cationic displacement of radionuclides from a solid through contact with a solution containing the replacing cation.

‡ Ion exchange as a separation technique refers to the removal of radionuclides from solution through contact with an ion exchange column.

4. Physical Remediation Techniques

4.1 Removal of Sources

a) Description

The most straightforward and simple method for the reduction of the radiological impact to the human population and environment is to remove the medium containing the contaminants to a discharge or a disposal site.

Contaminants are sometimes separated from uncontaminated medium in order to reduce the volume of material for disposal. The uncontaminated medium can be returned to the excavation area.

Usually, the contamination is near the surface. Therefore, scraping is often applied instead of bulk removal, or turf cutting (for soil).

b) Applicability

i) Means

Excavators (Valentich, 1994; Rice, 1994; Shirley and Schlessler, 1994) bulldozers (Kutlachmedov *et al.*, 1994; Blagoev *et al.*, 1996) vibration cutters and graders (Blagoev *et al.*, 1996) are mostly used for the bulk removal of soil.

A scraper (Blagoev *et al.*, 1996) and a shovel (Roed and Andersson, 1996) are reported to have been used for soil scraping.

Turf cutting is usually carried out with a turf cutter (Bondar *et al.*, 1995; Kutlachmedov *et al.*, 1994) or a turf harvester (Jouve *et al.*, 1994; Grebenk'ov *et al.*, 1994; Jouve *et al.*, 1993).

Groundwater can also be treated if pumping is carried out.

ii) Media

The media that can be treated by these techniques are primarily soil (possibly with grass), mud, tailings and buried waste.

iii) Contaminants

All radionuclide contaminated soil and water may be removed by this approach.

iv) *Work rates*

Values for the work rate with excavators for soil and buried waste were reported to lie between 6.6 and 14.4 m³ h⁻¹ (Rice, 1994) and between 27 and 39 m³ h⁻¹ (Valentich, 1994) respectively. In addition, the Federal Remediation Technologies Roundtable (1997) indicated a work rate of 9100 tonne month⁻¹ excavated (soil or sediment).

Work rates for soil scraping were reported by Brown *et al.* (1996) to be 5 to 10 m² h⁻¹ for 75 mm depth and 10 to 100 m² h⁻¹ for 50 mm depth if turf removal is also included (small to large areas).

Turf cutting with a 'turf crocodile' was reported by Kutlachmedov *et al.* (1994). The work rate was expressed as 40,000 ha in 20 machine year. Brown *et al.* (1996) reports a turf removal rate of 300 m² h⁻¹.

c) **Performance**

i) *Effectiveness*

Soil excavations, carried out in the Ukraine (Kutlachmedov *et al.*, 1994) and in the Chernobyl accident area (Blagoev *et al.*, 1996), reported DF (decontamination factor) values of 6 to 8 and 1.1 to 3.2, respectively.

Soil scraping in the Chernobyl accident area gave reported DF values of 1.2 to 1.7 (Blagoev *et al.*, 1996). Soil scraping at the Nevada test site gave DF values of 7 and 20 for plutonium and americium, respectively (Chilton and Pfuderer, 1989). DF values of 4.5 (5 cm depth) and 6.5 (10 cm depth) were obtained at Pripyat (Ukraine) (Roed and Andersson, 1996). Akinfiev and Radchuk (1993) reported a DF value of 5.3 at the Chernobyl NPP (Nuclear Power Plant).

Turf harvesting (upper 5 cm layer) in the zone close to Chernobyl NPP (Bondar *et al.*, 1995) showed a clear distinction in effectiveness between podzolic and peaty soils (see Table 2):

Table 2 Decontamination factors for radionuclides achieved with a turf harvester

Soil	Radionuclide			
	²³⁹ Pu-	⁹⁰ Sr	¹³⁷ Cs	¹⁴⁴ Ce
Turf-podzol	26-56	2.3-16	12-67	24-77
Peaty soil	2.6-16	2.4-15	2.5-9.3	2.8-13

From Bondar *et al.*, 1995

Values within the same ranges were obtained in Ukraine (Kutlachmedov *et al.*, 1994) and Belarus (Greibenkov *et al.*, 1994).

ii) *Service Life*

Not applicable.

d) **Costs**

In the USA the Federal Remediation Technologies Roundtable (1997) reported removal costs of between 283 and 482 EUR tonne⁻¹ (270 and 460 US\$ ton⁻¹), including excavation, transport and disposal (at a Resource Conservation and Recovery Act, RCRA, permitted facility). Also in the USA, Du Teaux (1996) observed excavation costs of 56 EUR m⁻³ (45 US\$ yd⁻³), whilst Gaylord and Klein

(1994) reported excavation costs for soil to be 140 EUR m⁻³ (110 US\$ yd⁻³). For disposal the latter indicated costs of 1680 EUR tonne⁻¹ (1600 US\$ ton⁻¹) for radioactive waste (excluding transport) and 68 EUR tonne⁻¹ (65 US\$ ton⁻¹) for conventional waste.

The IAEA (1992) reported transport and placement costs for the removal of tailings in the USA to be 0.6 and 1.2 EUR m⁻³ km⁻¹ (0.6-1.3 US\$ m⁻³ km⁻¹). Similarly, the costs for loading, transport (10-25 km) and unloading in Japan were reported to be between 1.9 and 3.6 EUR m⁻³ km⁻¹ (2.4 US\$ m⁻³ km⁻¹) (IAEA, 1992).

Brown *et al.* (1996) gave costs of 2.9 EUR m⁻² (1.95 £ m⁻²) for the removal (soil scraping) of the top 75 mm of soil and transport to the disposal site, and between 1.2 (for large areas, i.e. the size of a park) and 3.0 EUR m⁻² (for small areas, i.e. the size of a garden (0.8 and 2.0 £ m⁻²) for soil removal and turf removal to a depth of 50 mm, and between 4.8 and 6.6 EUR m⁻² (3.2 and 4.4 £ m⁻²) if re-turfing is also considered.

In the city of Espoo (Finland) (urban area) the cost for soil removal (soil scraping), including transport was reported to be between 0.5 (easy conditions, i.e. a flat terrain with dry, resistant soil) and 2.3 EUR m⁻² (difficult conditions, a undulating terrain with wet, breakable soil) (Lehto, 1994).

A turf removal cost of 0.45 EUR m⁻² (0.3 £ m⁻²) is cited by Brown *et al.* (1996).

Jouve *et al.* (1994) reported that the costs of applying a turf harvester to a 150 km² pasture, in CIS (Commonwealth of Independent States), were as follows:

$$\begin{array}{rcccccc}
 & 2.1 \times 10^{-3} \text{ EUR m}^{-2} & + & 1.0 \times 10^{-3} \text{ EUR m}^{-2} & + & 2.1 \times 10^{-3} \text{ EUR m}^{-2} & = & 5.5 \times 10^{-3} \text{ EUR m}^{-2} \\
 & (2.2 \times 10^{-3} \$ \text{ m}^{-2}) & & (1 \times 10^{-3} \$ \text{ m}^{-2}) & & (2.2 \times 10^{-3} \$ \text{ m}^{-2}) & & (5.4 \times 10^{-3} \$ \text{ m}^{-2}) \\
 \text{for} & \text{equipment} & & \text{operation} & & \text{waste transport} & & \text{total}
 \end{array}$$

In addition, Jouve *et al.* (1993) reported that the costs for turf harvesting 10,000 m² were:

$$\begin{array}{rcccccc}
 & 0.033 \text{ EUR m}^{-2} & + & 0.01 \text{ EUR m}^{-2} & + & 0.014 \text{ EUR m}^{-2} & + & 0.017 \text{ EUR m}^{-2} & = & 0.074 \text{ EUR m}^{-2} \\
 \text{for} & \text{investment} & & \text{maintenance} & & \text{waste transport} & & \text{manpower} & & \text{total}
 \end{array}$$

Costs to account for seed and equipment were also given by Jouve *et al.* (1993).

e) Side effects

The production (and disposal) of radioactive contaminated waste is a serious drawback to this technique.

4.2 Physical Separation

4.2.1 Soil Washing

a) Description

Soil washing is a separation process in which fine soil particles (silts and clay) are removed from the granular soil particles. It is assumed that contaminants are tightly bound to the fine soil particles rather than to the larger grained sand and gravel. Therefore, contaminated soil or debris may be decontaminated by first mixing them with water to form a slurry and then passing the slurry through a separation machine to remove the contaminated, fine particles.

Soil washing is essentially an *ex-situ* process, where the clean granular output stream (soil particles) can be returned to the excavation area. The remaining contaminated soil fines and process wastes are available for further treatment and/or disposal.

b) Applicability

i) Means

The techniques and apparatus which may be used to achieve the physical separation in the soil washing process, include:

- Washing and rinsing (Bondar *et al.*, 1995; Bovendeur and Pruijn, 1994; Dworjanyn, 1996; Chilton and Pfuderer, 1989; USEPA, 1996);
- Attrition scrubbing (Timpson *et al.*, 1994; Sadler and Krstich, 1994; Peng and Voss, 1994; USEPA, 1996; Groenendijk *et al.*, 1996);
- Centrifugal separation: by Campbell centrifugal jig (Goldberg *et al.*, 1994; Mathur *et al.*, 1996); by Knelson centrifugal concentrator (Mathur *et al.*, 1996); or by other apparatus (Mista *et al.*, 1995);
- Gravity separation: by Magstream density separator (Elless *et al.*, 1994) with magnetic separation or by other apparatus (Bovendeur and Pruijn, 1994; USEPA, 1996);
- Hydrocycloning (Peng and Voss, 1994; Bovendeur and Pruijn, 1994; Mathur *et al.*, 1996; Nechaev and Projaev, 1996).

ii) Media

Media which can be treated by this technique include soil, sediment and sludge. However the contaminants need to be closely associated with the fine particles.

Humus soils with a high, naturally occurring organic content can be difficult to clean, whereas sandy soils are very easy to clean.

iii) Contaminants

Soil washing has been used to remove a variety of radionuclides. In particular, separation of uranium and plutonium from soils has been extensively tested at a number of sites. These include: the Fernald site (Dworjanyn, 1996; Mathur *et al.*, 1996), the Nevada test site (Chilton and Pfuderer, 1989), at INEL (Idaho National Engineering Laboratory) and LANL (Los Alamos National Laboratory) (Mathur *et al.*, 1996), in Russia (Nechaev and Projaev, 1996) and at other sites.

The separation of americium, radium, caesium, strontium and cobalt from contaminated soils has also been reported and documented in the literature reviewed (see Section 4.2.1c).

iv) Work rates

Work rates varied from 1.5 tonne h⁻¹ for pilot plant tests up to between 18 to 90 tonne h⁻¹ in the VORCE (Volume Reduction/Chemical Extraction) plants (USEPA, 1996; Du Teaux, 1996).

The Federal Remediation Technologies Roundtable (1997) reported a work rate of 5.5 tonnes h⁻¹.

c) Performance

i) Effectiveness

Decontamination factors (DF) reported in the literature show a high degree of variability. Mathur *et al.* (1996) reported DF values for uranium and plutonium ranging from "ineffective" up to 2 and 5, respectively. When combined with other techniques (e.g. magnetic separation), the DF values went up to 10. However, DF values of up to 15 were reported for plutonium and americium (Chilton and Pfuderer, 1989) and of up to 17 for lead (Federal Remediation Technologies Roundtable, 1997) when chemical leaching was applied in combination with soil washing. Mista *et al.* (1995) reported DF values from 1 to 9 for uranium and not much greater than 1 for plutonium. However, when the soil washing was combined with flotation the latter value went up to 5.5. Dworjany (1996) observed a DF value of 2.1 for uranium in soil at the Fernald site.

DF values from 4 to 80 (plutonium contamination) have been reported for pilot-plant tests at Superfund Sites (USEPA, 1996).

DF values from 2.5 to 8 have been obtained for radium at the Superfund Site of Montclair (New Jersey) (Du Teaux, 1996).

DF values of between 1 and 3 have been reported for ¹³⁷Cs and ⁹⁰Sr (Bondar *et al.*, 1994; Bondar *et al.*, 1995). DF values higher than 3 and 3.5 were obtained with the VORCE plant for ¹³⁷Cs and ²³²Th, respectively (USEPA, 1996).

Another important aspect of separation techniques is the reduction in the volume of disposable waste. Depending on the initial contamination and the allowable residual contamination in the soil on the site, reductions in waste volume of between 50 and 98% are reported (Groenendijk *et al.*, 1996; Mathur *et al.*, 1996; Mista *et al.*, 1995; Moroney *et al.*, 1994). Reductions of around 65% were obtained with the VORCE plants in Tennessee and New Jersey (USEPA, 1996) and a value of 54% for the radium contamination in Montclair (Du Teaux, 1996). However the process/wash water may contain elevated levels of contamination and will, therefore, also require treatment and/or disposal.

ii) Service Life

Not applicable.

d) Costs

According to USEPA (1996) projected unit costs vary with the size of the site. Treatment costs with the VORCE plant amount to 116-140 EUR tonne⁻¹ (111-134 US\$ ton⁻¹) for work rates of 18-90 tonne h⁻¹. Total costs can be as high as 293 EUR tonne⁻¹ (280 US\$ ton⁻¹) when waste is transported off-site.

Du Teaux (1996) listed total costs from 26 EUR m⁻³ (21 US\$ yd⁻³) for a pilot plant demonstration with VORCE at Montclair up to 500 EUR m⁻³ (400 US\$ yd⁻³) for a demonstration at King of Prussia (Superfund Site with metal contamination).

For pilot tests of 1.5 tonne h⁻¹ an operational cost of 180 EUR tonne⁻¹ (200 US\$ ton⁻¹) has been reported at the Montclair Superfund Site (USEPA, 1996) with disposal and transport of radioactive soils costing 942 EUR tonne⁻¹ (900 US\$ ton⁻¹).

In Canada, at Ataratiri (Toronto) a cost of 68 EUR tonne⁻¹ (100 Cdn\$ tonne⁻¹) was reported for a soil contaminated with heavy metals (Du Teaux, 1996). At Port Granby 400,000 m³ of marginally contaminated soil (containing uranium and ²²⁶Ra) was processed at a cost of 142 EUR m⁻³ (209 Cdn\$ m⁻³) (Pollock and Feasby, 1996).

According to the Federal Remediation Technologies Roundtable (1997), the average cost would be of the order of 178 EUR tonne⁻¹ (170 US\$ ton⁻¹), including excavation.

Wood (1997) reported costs of between 80 and 380 EUR tonne⁻¹ (50-250 £ tonne⁻¹).

e) Side Effects

The residual soil fines and process/wash waters may require further treatment and/or disposal.

4.2.2 Flotation

a) Description

Flotation is a process in which contaminated soil fractions (usually fine soil particles such as silts and clays) are separated from the clean soil fractions (large granular soil particles and gravel) by means of the production of a foam that contains the contaminated soil particles.

It is essentially an *ex-situ* process, in which the contaminated soil has first to be excavated. It is then mixed with water to form a slurry. A flotation agent, which binds to the surface of the contaminated soil particles to form a hydrophobic surface, is added to this slurry. Small air bubbles are introduced in the solution and adhere to the hydrophobic particles and transport them to the surface. The foam is removed from the surface and may be subject to further treatment or disposal. The clean soil can be returned to the excavation area.

b) Applicability

i) Means

Types of flotation reported to have been carried out, include froth-flotation (Buckley *et al.*, 1995; Bovendeur and Pruijn, 1994; Palmer *et al.*, 1995), tall column flotation and automated mechanical flotation (Mathur *et al.*, 1996). The two former techniques have been tested at INEL, LANL, Fernald and the Mound Laboratory (see section on Effectiveness).

ii) Media

The contaminated media which may be treated by this process include soil and sediment.

Soil characteristics, such as particle size distribution, radionuclide distribution (with respect to particle size, clay, sand, humus and silt content), specific gravity, chemical composition and mineralogical composition may affect the effectiveness of the flotation treatment. Larger soil particles may require grinding or removal from the soil prior to applying flotation. Soils with high organic content (i.e. histosols) can be difficult to treat with this technology.

iii) Contaminants

Contaminants which can be removed by flotation include heavy metals and radionuclides such as uranium and plutonium. Flotation is used extensively in the mining industry to concentrate constituents such as uranium from ores.

iv) Work rates

No information found.

c) Performance

i) Effectiveness

Results from the mining industry indicate that a consistent and successful segregation of contaminated fines from clean soil can be achieved with flotation (USEPA, 1996). The technique was reported to be 95% effective in separating uranium oxide from sandstone ores (USEPA, 1996). Bench-scale tests revealed high removal efficiencies, ranging from 70 to 90% for soil contaminated with bismuth, as a surrogate for plutonium oxide (USEPA, 1996). For radium in uranium mill tailings DF values from 4 to 6 have been observed (USEPA, 1996). However, tests at INEL, LANL, Fernald and Mound Laboratory showed very varied results; with between 3 and 78% removal for uranium and plutonium (Mathur *et al.*, 1996).

The organic content of the soil can reduce the effectiveness of this technology.

Additional studies are needed in order to determine the effectiveness of separating radionuclide-contaminated fines from soil.

Flotation was reported to achieve reductions of between 28 to 97% in the volume of contaminated soil (Mathur *et al.*, 1996).

The presence of clay and silt will increase the volume of contaminated material removed in the foam. This leads to an increase in the volume of material requiring additional treatment.

ii) Service Life

Not applicable.

d) Costs

USEPA (1996) reported that the capital cost of a flotation unit varied between 24,000 to 152,000 EUR (25,000-160,000 US\$) depending on the size of the unit.

Operation and maintenance costs varied between 0.75 to 3.76 EUR m⁻³ (3 to 15 US\$ per 1,000 gallons) of slurry treated.

e) Side effects

The residual foams generated by the flotation process may require further treatment or disposal.

4.2.3 Filtration

a) Description

Filtration is a process in which contaminants in liquid media (e.g. groundwater) are separated from the liquid media by a porous membrane. The pore size of the membrane can be varied to remove particles and molecules of various sizes. Depending on what is fed into the filtration system, the process generates two waste streams: a filter cake of solid material, a filtrate of treated (clean) liquid.

The treated (clean) liquid can be reused or returned to its natural environment. The filter cake requires further treatment and/or disposal.

b) Applicability

i) Means

Two membrane filtration systems have been reported (Anderson *et al.*, 1994; USEPA, 1996). This includes an application on surface water at Rocky Flats (Anderson *et al.*, 1994).

ii) Media

This process can be applied to contaminated liquid media. This includes groundwater and surface water.

Filtration may be applied in situations where radionuclide contaminants are associated with suspended solids in a liquid medium or where the liquid medium has been pre-treated with a precipitating agent.

iii) Contaminants

Filtration has been tested and used for the removal of a variety of radionuclides from contaminated water. The only radionuclide which cannot be removed, because of its chemical characteristics, is tritium.

iv) Work rates

No information found.

c) Performance

i) Effectiveness

Micro-filtration membranes, which are currently available, can effectively remove particles as small as 0.1 micron from water.

In tests conducted to evaluate radionuclide removal from water, efficiencies exceeding 99% were obtained for uranium, plutonium and americium with no chemical pre-treatment (USEPA, 1996). Du Teaux (1996) reported efficiencies between 58 and 95% for uranium in a commercial scale demonstration at Rocky Flats.

Removal efficiency for gross alpha emitters and radium were reported to be 86% and 43%, respectively (USEPA, 1996).

ii) Service Life

Not applicable.

d) Costs

For the system tested by USEPA (1996), the treatment cost ranges from 0.13 to 3.76 EUR m⁻³ (0.50-15 US\$ per 1000 gallons). This cost is dependent on the duration of the treatment, the volume of water treated, the type of contamination and the concentration of the contaminants.

e) Side effects

The residual filter cake and/or liquid concentrate may require further treatment and/or disposal.

4.3 Containment

4.3.1 Capping

a) Description

Capping is a containment technology in which a barrier is provided between the contaminated media and the surface.

Capping radioactive contaminants can have several objectives:

- Minimising the migration of radioactive substances to the surface;
- Restricting the infiltration of surface water and, hence, leaching of contaminants from the site;
- Providing shielding, to humans on the site, from direct irradiation, and
- Controlling windblown transport of contaminants.

b) Applicability

i) Means

A cap is usually a combination of several layers of different materials. Most of the objectives of capping can be achieved with natural materials, such as layers of soil, gravel, rock or rip-rap. However, the prevention or restriction of infiltrating water which, in general, is the most important objective, is achieved only by introducing low-permeability layers. These may also be made up of natural low-permeability soils such as clay (McGregor, 1994), but artificial materials may be more appropriate. Such materials include plastics (e.g. high density polyethylene), geomembranes (made of polyvinyl chloride, high density polyethylene, very low density polyethylene, polypropylene, polyester, hypalon), geosynthetic clay liners (usually blankets of bentonite clay with geotextile), asphalt (asphalt-mix, asphalt concrete, asphalt rubber membrane) or cement (concrete).

A typical cap for containing radioactive media may consist of several feet of compacted filler, a geomembrane, a layer of compacted clay, another geomembrane and several feet of top soil (USEPA, 1996).

ii) *Media*

Capping can be used on a variety of solid materials. This includes: soil, mine tailings, sediment and bulk waste.

Air temperature, seasonal variations, topography and other site-specific and subsurface conditions may affect the implementation of this technique.

iii) *Contaminants*

Capping can be used to contain all types of (solid) waste, including radioactive waste materials found in the soil matrix, debris and radioactively contaminated landfills.

iv) *Work rates*

Lehto (1994) indicated that between 100 and 220 man hours were required for the construction of a clay layer, a soil layer, underdraining and the laying of a plastic sheet for a disposal of 10,000 m³. Similarly, between 235 and 450 man hours were required for the disposal of 50,000 m³ by the same method.

c) ***Performance***

i) *Effectiveness*

Most capping technologies are well developed and considered reliable.

An important parameter, characterising the short-term effectiveness of a cap with respect to infiltration of surface water is the permeability (k).

Materials with very low permeability include:

- Geomembranes (e.g. hypalon): $k = 2 \times 10^{-12} \text{ m s}^{-1}$ (Chilton and Pfuderer, 1989);
- Geosynthetic clay liners: $k = 1 \times 10^{-12} - 1 \times 10^{-10} \text{ m s}^{-1}$ (Daniel, 1994)
- Asphalt:
 - $k = 1 \times 10^{-13} \text{ m s}^{-1}$ for a combination of asphalt concrete and liquid applied asphalt at the Rocky Flats, solar evaporation pond (Nixon *et al.*, 1994);
 - $k = 7 \times 10^{-10} \text{ m s}^{-1}$ for asphalt concrete (Chilton and Pfuderer, 1989);
 - $k < 1 \times 10^{-9} \text{ m s}^{-1}$ for hot asphalt mix (Beck *et al.*, 1994);
- Clay: $k = 1 \times 10^{-9} \text{ m s}^{-1}$ for sodium bentonite (Chilton and Pfuderer, 1989).

Over the long term, site conditions such as air temperature, seasonal variation and topography may affect the integrity of the cap due to cracking, settling, erosion or groundwater intrusion. Once a cap is installed, monitoring of groundwater and radioactive gas emissions and of cap integrity, are required to ensure the effectiveness of this technology.

ii) Service Life

A long service life is reported (of the order of 1,000 years) for the multilayer caps at Rocky Flats, solar evaporation pond (Ogg *et al.*, 1995) and Hanford (Wing and Gee, 1994). For asphalt caps Freeman and Romine (1994) and Nixon *et al.*, (1994) indicate service lives of more than 1,000 years for asphalt concrete and liquid applied asphalt (5,000 years in anaerobic conditions).

For geomembranes the service life is strongly dependent on the materials used: 10-25 years for polypropylene, polyester and polyvinyl chloride (Parikh and Rattan, 1994; Daniel, 1994) and perhaps several 1,000 years for high density polyethylene (Daniel, 1994).

For concrete Brandstetter *et al.* (1994) and Porter (1995) reported a service life of between 100 and 1,000 years and of several 1,000 years, respectively.

d) Costs

The cost of capping depends on the type and size of the cap. According to USEPA (1996) a typical clay cap costs 11 to 17 EUR m⁻² (10 to 15 US\$ yd⁻²). A typical RCRA cap, with multiple layers, more likely to be used for radioactive waste, costs between 28 and 34 EUR m⁻² (25 and 30 US\$ yd⁻²), and Wood (1997) reported the costs to be between 30 and 45 EUR m⁻² (20-30 £ m⁻²). Almost all this cost is capital.

The operating and maintenance costs are low and are generated by monitoring and possible repair.

e) Side Effects

No important side effects.

4.3.2 Subsurface Barriers

a) Description

Subsurface barriers constitute a containment technology when the barriers are installed around the contaminated zone to confine the contaminated material (and groundwater). They may consist of vertical barriers reaching down to a less permeable natural horizontal barrier, such as a clay zone, to impede groundwater flow. Alternatively, they may completely confine the area, including the bottom as is the case for land encapsulation.

Subsurface barriers are frequently used in conjunction with capping since this would produce an essentially complete containment structure surrounding the waste mass.

Vertical barriers are mostly slurry walls or grout barriers (grout curtains).

Slurry walls consist of vertically excavated trenches that are filled with a slurry. The slurry hydraulically shores the trench to prevent the collapse of the side walls during excavation and produces a barrier to groundwater flow.

Grout barriers may be constructed in two ways: permeation grouting or jet grouting by mixing. Permeation grouting takes advantage of the soil's natural permeability by using pressure to inject the grout, which then flows into the soil. Jet grouting by mixing, uses a rotating drill during injection the grout. This fractures the soil and mixes it with the grout.

Grout curtains (narrow, vertical, grout walls) are constructed by pressure-injecting grout into the soil. They are placed at closely spaced intervals so that each "pillar" of grout intersects the next, thus forming a continuous wall or curtain.

In the case of land encapsulation, the area to be encapsulated is firstly excavated after which a liner or other impermeable material is installed in the excavated area, and the excavated material is put back. This technology is generally used at the disposal stage of radioactive waste management.

b) *Applicability*

i) Means

Slurry walls are generally a mix of bentonite and water, or Portland cement, bentonite and water. They are normally 0.6 to 1.2 m thick and placed at depths of less than 15 m (USEPA, 1996).

Subsurface grout barriers use various kinds of grouts. This includes Portland cement, alkali silicate grouts and organic and inorganic polymers.

Cement grout has, for example, been applied at the Fernald site (Pettit *et al.*, 1994) and in Japan (Banno and Yoshida, 1994). Grout barriers with polyacrylamide, sodium silicate and polyacrylate were reported to have been used for mill tailings (Chilton and Pfuderer, 1989).

Other grout barrier materials tested with positive results include microfine cement (Dwyer, 1994) and also organic polymers (vinylester styrene, polyester styrene, acrylic, furfuryl alcohol) and inorganic polymers (sulphur polymer cement) at the Hanford site (Heiser and Colombo, 1994; Heiser *et al.*, 1994).

Potentially useful grouts have been developed in Germany, consisting of a naturally occurring wax (Montan wax), water and bentonite, and in France a glyoxal-modified sodium silicate grout has been used (Dwyer, 1994; Voss *et al.*, 1994). They have been tested in the field at the Mixed Waste Landfill Integrated Demonstration site at Sandia National Laboratories (New Mexico).

Grout curtains used as barriers, mostly consist of polymer grouts and are used at shallow depths (9 to 12 m maximum). Currently, a barrier consisting of a conventional cement grout curtain with a thin lining of polymer grout is undergoing field testing (USEPA, 1996).

A polymer gel of polyacrylamide was used with excellent effect to contain an oil spill (Wilkins, 1996).

ii) Media

Media to which these technologies can be applied include soil, sediment, leachates, bulk waste (tailings) and groundwater.

Prior to the installation of barriers, the soil's physical and chemical characteristics need to be known.

iii) Contaminants

This technology provides subsurface containment for a wide variety of radionuclides (especially uranium, plutonium, strontium and caesium) as well as other metals. Organic materials which do not adversely affect these barriers may also be contained.

The characterisation of contaminants is necessary, particularly for liquid contaminants. For example, slurry walls and grout barriers have the potential to degrade or deteriorate over time, and certain grout materials can be prevented from setting due to chemicals contained in the waste, particularly organic chemicals (USEPA, 1996).

iv) *Work rates*

No information found

c) ***Performance***

i) *Effectiveness*

The primary aim of subsurface barriers is to prevent or slow down the migration of contaminants within the groundwater. The short-term effectiveness of these barriers may be characterised by their permeability (k).

Field tests with Portland cement grout (with calcium and aluminium silicates), carried out at INEL, gave a permeability of $10^{-10} \text{ m s}^{-1}$ (Shaw and Weidner, 1996). Measurements from pilot-scale tests at INEL gave a permeability of 10^{-8} m s^{-1} for cement-based grouts, with lime and fly ash or gypsum and calcite. Similarly, a permeability of $10^{-10} \text{ m s}^{-1}$ has been measured for apatite (magnesium phosphates) (Shaw and Weidner, 1996). However, the effectiveness depends on several soil characteristics, including void volume, soil pore size and permeability of the surrounding material.

Very low permeabilities have been reported for grout barriers containing organic and inorganic polymers:

at the Hanford site: $k = 10^{-11} - 10^{-10} \text{ m s}^{-1}$ (Heiser and Colombo, 1994)
for other tests: $k = 5 \times 10^{-13} - 4 \times 10^{-11} \text{ m s}^{-1}$ (Heiser *et al.*, 1994)

The grout barriers with polyacrylamide, sodium silicate and polyacrylate applied on mill tailings showed permeabilities of $3 \times 10^{-9} - 5 \times 10^{-7}$, $7 \times 10^{-9} - 5 \times 10^{-6}$ and $< 3 \times 10^{-9} \text{ m s}^{-1}$, respectively (Chilton and Pfuderer, 1989).

A barrier of a polymer gel of polyacrylamide gave low permeabilities of between 10^{-11} and $10^{-10} \text{ m s}^{-1}$ (Wilkins, 1996).

The effectiveness of vertical barriers (e.g. slurry walls) largely depends on the presence of a confining layer of clay or rock into which the barrier is set.

The long-term effects of weathering, groundwater infiltration and physical disturbance, associated with uncontrolled future land use, may affect the integrity of the vertical barriers and contaminant mobility in an unpredictable way. In addition, the long-term effectiveness may be reduced by specific contaminant types (particularly organic chemicals) which may lead to the deterioration of the barrier. It also depends on the type of barrier used. Materials such as HDPE membranes and polymer grouts show a better chemical resistance (USEPA, 1996).

ii) *Service Life*

An indication of the service life has only been reported for land encapsulation. This suggests a reliability of between 100 and 1000 years (USEPA, 1996).

d) Costs

Capital costs make the major contribution to the costs of vertical barriers. Operation and maintenance costs are generally low.

The costs for the design and installation of a standard slurry wall into a soft to medium soil vary between 510 and 710 EUR m⁻² (540-750 US\$ m⁻²) (USEPA, 1996). However, the use of an HDPE membrane increases the capital cost (USEPA, 1996).

Grout curtains generally cost 310 to 410 EUR m⁻² (30-40 US\$ ft⁻²) (USEPA, 1996). The capital and operating costs are low. However, the use of a close-coupled barrier, with a polymer grout lining, increases the capital cost (USEPA, 1996).

The material and installation costs for land encapsulation varies between 262 and 850 EUR m⁻³ (276-895 US\$ m⁻³) . The first year operation and maintenance costs are estimated to be 0.043 EUR m⁻³ (0.045 US\$ m⁻³) (USEPA, 1996).

e) Side Effects

No important side effects.

4.4 Physical Immobilisation

4.4.1 Solidification/stabilisation

a) Description

Solidification/stabilisation processes are immobilisation processes that reduce the mobility and solubility of contaminants by solidifying or stabilising them into a matrix.

Cement-based solidification processes involve the addition of cement or a cement-based mixture to limit the solubility or mobility of the waste constituents. Pozzolanic solidification uses fly ash (non-crystalline silica) and lime (calcium) to produce a concrete-like solid.

These techniques are accomplished *in-situ* by injecting the solidifying agent(s) directly into the contaminated material, or *ex-situ* by excavating the contaminated material, machine-mixing it with the solidifying agent(s) and returning the mixture to the excavated area. The objective of the process is to form a solid monolith that contains the waste material, increasing its physical stability and compressive strength, decreasing water intrusion into the waste and the leachability of waste constituents.

b) Applicability

i) Means

Types of solidifying/stabilizing agents include Portland cement, gypsum, modified sulphur cement (elemental sulphur and hydrocarbon polymers) and grout (consisting of cement and other dry materials, such as fly ash or blast furnace slag).

Portland cements have a number of disadvantages. Many contaminants do not chemically bond to the cement matrix but are only encapsulated. Hence, they are subject to potential release through

leaching and disturbance. The curing of Portland cements can be inhibited by the presence of metal salts, organic materials and soils with high clay or silt contents. For these reasons, Portland cements are normally blended with other materials, such as pozzolanic, when used as solidifying agents (USEPA, 1996; Malone and Lundquist, 1994).

The most commonly used pozzolanic materials in soil stabilisation are fly ash, finely ground blast furnace slag, fluidised bed furnace ash, cement and lime kiln dust, and sodium or potassium silicates. Calcium compounds (e.g. lime, cement, gypsum, limestone) are commonly added as setting agents. Bentonite or other clays may also be added with gypsum, to lower the permeability (IAEA, 1994; Malone and Lundquist, 1994).

ii) Media

Media on which these techniques may be applied include soil, sediment, sludge and refuse.

iii) Contaminants

Properly implemented, and in appropriate blending, cement solidification can be applied to most contaminants, including all classes of radioactive wastes and mixed wastes.

However, some concerns may be posed by some types of hazardous waste (organic chemicals) that may interfere with the solidification process.

iv) Work rates

Shallow and deep *in-situ* soil mixing technique processes, on average, 36-72 tonne h⁻¹ and 18-45 tonne h⁻¹, respectively (USEPA, 1996).

The Federal Remediation Technologies Roundtable (1997) describes process depths of up to 6 m.

c) Performance

i) Effectiveness

Quantitative information is lacking. However, in the short term, the leach resistance of most solidified waste is relatively high. According to the USEPA (1996), "*in-situ* processes have demonstrated the capability to reduce the mobility of contaminated waste by more than 95%". However, the long-term effects of weathering, groundwater infiltration and physical disturbance, associated with uncontrolled future land use, can significantly affect the integrity of the solidified mass and contaminant mobility in an unpredictable way.

Testing must be carried out in advance to verify the compatibility and performance of candidate treatment methods for each site and waste type (Malone and Lundquist, 1994).

ii) Service Life

The USEPA (1996) reported that cement solidification is highly reliable for periods of 100 to 1,000 years. However, Malone and Lundquist (1994) state experiences with man-made Portland cement structures exposed to weathering have indicated that stability beyond 100 years is questionable. Some vendors claim a very long-term stability for pozzolanic materials (millions of years) based on the geologic age of naturally occurring pozzolanic monoliths.

More large-scale tests and demonstrations are needed to determine the long-term effectiveness of cement-based solidification, in order to create a higher level of confidence.

d) Costs

The overall costs (including excavation) for *ex-situ* processes are reported to be less than 110 EUR tonne⁻¹ (<100 US\$ ton⁻¹) by USEPA (1996). However, Malone and Lundquist, (1994) reported the costs to be between 20 to 150 EUR m⁻³ (20 to 120 US\$ yd⁻³) of soil with average values of 81 to 93 EUR m⁻³ (65 to 75 US\$ yd⁻³).

Du Teaux, (1996) indicates total costs of 72 EUR tonne⁻¹ (80 US\$ tonne⁻¹) for solidification of soils with alumina, calcium and silica, and 85 EUR tonne⁻¹ (94 US\$ tonne⁻¹) for solidification of soils with Portland cement and silicates.

Cement-based solidification has been reported to cost between 30 and 260 EUR tonne⁻¹ (20-170 £ tonne⁻¹) (Wood, 1997).

The *in-situ* soil mixing/auger techniques an average cost of 50 to 70 EUR m⁻³ (40-60 US\$ yd⁻³) for shallow applications, and 190 to 310 EUR m⁻³ (150-250 US\$ yd⁻³) for deeper applications (USEPA, 1996; Malone and Lundquist, 1994).

Du Teaux (1996) indicates a total cost of between 116 and 203 EUR tonne⁻¹ (111 and 194 US\$ ton⁻¹) for soil solidification with slurry.

In-situ solidification costs of between 90 and 170 EUR tonne⁻¹ (60-110 £ tonne⁻¹) have also been reported (Wood, 1997).

The costs for cement-based solidification vary widely according to the materials or reagents used, their availability, project size and chemical nature of the contaminants (types, concentration levels, etc.).

e) Side effects

No important side effects.

5. Chemical Remediation Techniques

5.1 Chemical Separation

5.1.1 Chemical Solubilisation Techniques

a) *Description*

Solubilisation is a technique used to separate the radionuclide contaminant from the soil matrix and to collect it as a concentrated solution. It is achieved by passing a suitable solvent through the contaminated soil. Subsequent separation of the solvent containing the dissolved radionuclide from the soil provides an effective method for decontaminating soils. In order to achieve this, the process requires a method for collecting the soil, a vessel for containing it, a means for delivering the solvent to the soil, a means for ensuring good contact between the solvent and the soil, and a method for extracting and collecting the resulting contaminated solvent.

b) *Applicability*

i) *Means*

An important requirement for this approach is that a solvent can be found which will dissolve the contaminating radionuclide in a particular medium. The choice and effectiveness of the solvent will be dependent upon the chemistry of the radionuclide, its concentration and the nature of the contaminated soil. Therefore, the choice of solvent tends to be site-specific (USEPA, 1996).

Factors which have a significant effect on the solubility include:

- Choice of solvent,
- Adjustment in the pH of the solvent,
- Addition of complexation agents,
- Addition of ion exchange agents to replace the radionuclide in the solid medium,
- Addition of oxidation and reduction agents.

This technique may be used in a stand-alone manner with the resulting concentrate available for disposal. Alternatively, the technique is used in combination with other technologies, where the concentrate is further treated (e.g. through immobilisation or incineration).

This treatment of contaminated soils has been taken from bench-scale experiments to full-scale processes for radionuclides (USEPA, 1996).

ii) *Media*

This technology has been used extensively at a commercial level in the metal extraction industry (e.g. uranium from ores). It has been shown to be suitable for dry soils, sediments and sludges. Being a chemical process, the technique is generally carried out *ex situ*.

iii) *Contaminants*

This group of techniques has proven to be well-suited to the removal of radionuclides, heavy metals, inorganic and organic contaminants (USEPA, 1996).

iv) *Work rates*

A throughput range of between 1.8 and 4.5 tonne h⁻¹ (2 and 5 ton h⁻¹) has been reported (USEPA, 1996).

c) ***Performance***

i) *Effectiveness*

The effectiveness of this approach is a function of its ability to collect radionuclides in a high concentration, low volume residual. The solubility of a radionuclide contaminant will depend upon the composition of the solvent used in the chemical separation process. Consequently, the composition of the solvent may be adjusted to enhance the solubility of the radionuclide in the following ways:

- Solvent

The choice of solvent can greatly effect the solubility of the radionuclide. Whilst aqueous solvents are usually used, organic solvents can also be employed to extract radionuclides. For example, Grace *et al.* (1995) utilised triethylamine to extract 89% of oxidised plutonium from a soil contaminated with 3.3 kBq kg⁻¹ of the radionuclide. Similarly, a 10% diethylamine solution has also been found to be effective at removing oxidised metal ions (Palmer *et al.*, 1995).

- pH

The solubilities of radionuclide compounds tend to vary significantly with the pH. For example, a study by Palmer *et al.* (1995) compared solubilities of lead and zinc ions in 30% sulphuric acid and 10% sodium hydroxide solutions. The alkali solution removed up to 70% of the lead and 10% zinc, whilst the acid solution extracted 33% of the lead and up to 22% of the zinc.

- Complexation

Reactions between radionuclide ions and complexing agents can enhance the solubility of the contaminants. Complexing agents may be organic or inorganic. Examples of organic complexing agents which have been used include (ethylenediaminetetraacetic acid (Gall and Farley, 1994; USEPA, 1996) and acetic acid (Thöming and Calmano, 1995). The latter was shown to be capable of extracting up to 70% of a number of heavy metals.

Inorganic complexing agents are usually carbonate compounds. Complexation between carbonate and uranium, in particular, has been well-studied (Dander *et al.*, 1994; Elless *et al.*, 1994; Dworjany, 1996). The process can achieve uranium removal rates of up to 70% (Dworjany, 1996).

- Ion Exchange

Competition between cations in the solvent and the radionuclide for binding sites in the soil can be utilised to increase the solubility of the radionuclide. This includes the use of the ammonium ion and the hydrogen ion to replace strontium and caesium ions (Bondar *et al.*,

1995). Here it was reported that decontamination factors of up to 4.2 and 1.2 for caesium and up to 10 and 2.3 for strontium were respectively obtained with the hydrogen ion and the ammonium ion.

- Oxidation/Reduction

The solubility of different oxidation states of radionuclides can vary considerably. Inclusion of oxidising/reducing agents in the solvent can significantly increase the dissolution of the radionuclide. An example of this approach is the use of sodium hypochlorite to oxidise uranium to a more soluble state. Used in combination with carbonate complexation removed 70% of uranium from soils (Dworjanyn, 1996).

Overall, a wide range of efficiencies (13-100%) were obtained by these techniques for removing radionuclides from soils (USEPA, 1996). Factors which contribute to this include: particle size, pH, partition coefficient, cation exchange capacity, organic content, moisture content, contaminant concentration and solubility (USEPA, 1996). The large variation reflects the dependence of the technique on the radionuclide, the solvent and the demonstration conditions.

The mineral content of the soil is an important factor in determining the efficiency. Kavkhuta *et al.* (1994) examined the leaching of caesium-137 from different clay minerals using potassium and ammonium solutions. The results are summarised in Table 3.

Table 3 Leach tests for caesium-137 in different clay minerals

Mineral	Fraction of ¹³⁷ Cs leached	
	0.5 M KNO ₃	0.5 M NH ₄ NO ₃
Vermiculite	0.126	0.126
Kaolin	0.195	0.564
Biotite	0.466	0.494
Muscovite	0.541	0.585
Hydromuscovite	0.620	0.712

From Kavkhuta *et al.*, 1994

Estimates for the throughput rate for soil by this technique can be between 2 to 5 tonne h⁻¹ (USEPA, 1996) and 16 m³ h⁻¹ (Thöming and Calmano, 1995).

ii) Service Life

Not applicable.

d) Cost

Capital, operating and maintenance costs for the process are reported to be medium to high. Estimates for the costs of the operation include 110 to 420 EUR tonne⁻¹ (100-400 US\$ ton⁻¹) (USEPA, 1996), 200 to 230 EUR tonne⁻¹ (390-450 DM tonne⁻¹) (Thöming and Calmano, 1995) and 80 to 260 EUR tonne⁻¹ (50-170 £ tonne⁻¹) (Wood, 1997). Solvent extraction has been estimated to be 50 to 700 EUR tonne⁻¹ (30-600 £ tonne⁻¹) (Wood, 1997). These values will be dependent upon how much pre-treatment, extraction, and post treatment is necessary. In addition, a multiple stage process would

add significantly to the cost. There will also be operating and maintenance costs associated with the storage and treatment of the process waste.

e) Side Effects

Chemical solubilisation gives rise to liquid wastes containing high levels of radionuclides which require suitable disposal or further treatment. Where organic solvents are used, the volume may often be minimised by recovering the solvent by distillation for reuse.

5.1.2 Ion Exchange

a) Description

Ion exchange is a process where contaminating ions are removed from a contaminated liquid by replacing them with less harmful ions. The process makes use of materials that have binding sites which have a significantly higher affinity towards the contaminating ions than to ions which are already bound. Contact between the contaminated liquid and the ion exchange material enables the contaminating ion to displace the less harmful ion. Therefore, the contaminant is removed from the liquid, and is concentrated on the surface of the ion exchange material, while the less harmful ion is released into the liquid.

Treatment of the contaminated groundwater is typically achieved by first extracting the water from a well sunk into the contaminated soil by pumping or collecting as it discharges. The water is filtered and passed, under pressure, through a fixed bed of the ion exchange material where the radionuclide is collected and concentrated. The groundwater, thus depleted of the radionuclide, can be readily disposed of.

b) Applicability

i) Means

Ion exchange is an *ex-situ* technique which, for radionuclide treatments, has been taken from bench-scale experiments to pilot-scale projects (USEPA, 1996).

Two types of ion exchange material are available: reversible and irreversible materials. Reversible materials are usually resins which may be regenerated by stripping off the contaminant (usually with a strong acid). The contaminant can be collected as a highly concentrated, low volume waste, which is available for further treatment. Irreversible ion exchange materials (zeolites) are cheap substances which accumulate the contaminant for later disposal.

ii) Media

The technique has been shown to be appropriate for removing radionuclides from groundwater, surface water, and other aqueous waste streams, including those collected from solubilisation techniques (see Section 5.1).

iii) Contaminants

This technique is constrained by the need for the contaminant to be in an ionic form. Non-ionic substances cannot be removed. However, the range of materials which may be treated can be extended by modifying the contaminated liquid (e.g. altering the pH). The technique has been used to treat liquids contaminated with radionuclides, heavy metals, inorganic and organic materials.

iv) *Work rates*

No information found.

c) *Performance*

i) *Effectiveness*

Typically, the operating cycle for this process will last 5-7 days before it is necessary to either regenerate or replace the ion exchange material. The number of cycles necessary will depend on the level of contamination.

The effectiveness of this approach for different contaminants is summarised in Table 4.

Table 4 Efficiency of ion exchange resin in removing radionuclides

Nuclide	Fraction Removed
Uranium	0.94* 0.65 - 0.99
Radium	0.65 - 0.97
Caesium-137	0.95 - 0.99
Strontium-89	0.95 - 0.99
* After 8 cycles. 5-7 days per cycle. For an initial waste stream concentration of 0.1 kg m ⁻³ and an approximate uranium loading of 0.035 kg kg ⁻¹ of commercial resin.	

From USEPA, 1996

This technique is dependent upon having a ion exchange material which is sufficiently selective towards the radionuclide ion. In addition, the radionuclide must be in a form which enables it to be collected by the ion exchange material; insoluble particles, colloids, and neutral molecules and complexes must be treated. The removal of competing ions must also be considered. Often, ion exchange materials are only effective over a limited range of pH values (Tadesse *et al.*, 1994).

ii) *Service Life*

Not applicable.

d) *Costs*

The capital, operating and maintenance costs for this process are high. The capital and operating costs have been estimated to be 1.3 to 2.5 EUR m⁻³ (5-10 US\$ per 1000 gallons) (USEPA, 1996). There will also be a disposal cost associated with the resulting residue.

e) *Side Effects*

In some cases, after the radionuclide has been concentrated on the ion exchange material it may be stripped off (usually by using a strong acid) and collected as a highly concentrated, low volume solution which is available for disposal or further treatment.

Where ion exchange materials are only used once the saturated material must be treated as a solid waste.

5.2 Chemical Immobilisation

a) Description

Chemical immobilisation is a process which limits the movement of contaminants through leaching and may also trap and contain gases (e.g. radon) within the medium (USEPA, 1996). This is accomplished through increasing the chemical stability of the contaminant within the medium by the addition of an immobilising reagent which chemically binds the contaminant.

Chemical immobilisation may be achieved both *in-situ* or *ex-situ*.

- *In-situ* techniques involve injection of the immobilising reagents directly into the contaminated medium;
- *Ex-situ* techniques require excavation of the contaminated material and mechanically mixing it with the immobilising reagents. The solidified material is then stored in containers, for disposal, or buried at the site under a sufficient thickness of soil to adsorb any gamma radiation.

b) Applicability

i) Means

Chemical immobilising techniques are simple to implement, requiring conventional handling equipment and readily available reagents and additives. No pre-treatment is necessary.

The reagents which have been successfully used to achieve immobilisation are thermoplastic and thermosetting polymers (Chisholm, 1994; Kalb and Adams, 1994; USEPA, 1996). Thermoplastic reagents include asphalt bitumen, paraffin and polyethylene. Thermosetting reagents include vinyl ester monomers, urea formaldehyde and epoxy polymers.

ii) Media

These techniques can be applied to soils, sediments, sludge and refuse. They are best suited to sites containing fine grain material.

iii) Contaminants

The techniques are applicable to radionuclides, heavy metals, inorganic, organic and mixed wastes. Some classes of organic materials (e.g. slightly volatile organic compounds and pesticides) may inhibit the effectiveness of chemical bonding of stabilisers or the mechanical bonding of stabilising agents (USEPA, 1996).

iv) *Work rates*

In-situ techniques require auger/caisson head systems and injector head systems to apply reagents to the contaminated soil. The estimated throughput rate is 40 to 80 tonne hr⁻¹ for shallow soil mixing and 20 to 50 tonne hr⁻¹ for deep soil mixing (USEPA, 1996).

Ex-situ techniques require an excavator and mechanical mixer to combine the reagents with the contaminated material. A full-scale polyethylene extruder can process material on a scale of 900 kg hr⁻¹, consisting of 30% binder and 70% waste. The extruded material will cool and set within a few hours (USEPA, 1996).

c) *Performance*

i) *Effectiveness*

The efficiencies of the techniques are affected by the presence of other contaminants, and, as such, are site-specific. The presence of organic material can be a particular problem when polymers are added as they can prevent hardening. Tests would have to be carried out to ensure the suitability of these techniques. However, leach experiments carried out by Frissel (1996) showed that efficiencies of between 80% and 100% may be achieved. The unconfined compressive strength of the solidified material has been reported to be between 1.5 and 10.8 MPa (USEPA, 1996).

ii) *Service Life*

The long-term effects of weathering, groundwater infiltration, and physical disturbance with uncontrolled future land use cannot be predicted (USEPA, 1996). Further studies are required to quantify these effects and to determine the likely service life of this technique.

d) *Costs*

The cost of *ex-situ* processes have been estimated to be below 110 EUR tonne⁻¹ (100 US\$ ton⁻¹), including excavation (USEPA, 1996).

The costs of *in-situ* processes vary with depth. Mixing/auger techniques have average costs of 50 to 75 EUR m⁻³ (40-60 US\$ yd⁻³) for shallow depths and 190 to 310 EUR m⁻³ (150-250 US\$ yd⁻³) for deeper applications (USEPA, 1996).

Reagents and additives are widely available and likely to be inexpensive. However, transportation of bulk chemicals may dominate, particularly when dealing with sites in remote locations.

Overall, the costs associated with these processes are low in comparison with other technologies. This is supported by Pollock and Feasby (1996) who estimated *in-situ* chemical immobilisation techniques for soils containing uranium, radium and arsenic to be 46 EUR m⁻³ (68 Can\$ m⁻³). In addition, Wood (1997) estimated the cost of immobilisation through liming to be 30 to 50 EUR tonne⁻¹ (20-40 £ tonne⁻¹).

e) *Side Effects*

No important side effects.

6 Biological Remediation Techniques

6.1 Biological Separation

6.1.1 Phytoremediation

a) Description

Phytoremediation is based on the well-established abilities of certain plants to accumulate elevated concentrations of metal ions. In most cases the metal ions are of little benefit to the plant. The plant responds to high levels of the metal by storing them in a form which minimises their toxicity. The radionuclide can be stored in the leaves and stems of the plant where it may be harvested along with the biomass. The biomass may be incinerated thereby reducing the volume of contaminated waste.

b) Applicability

i) Means

Phytoremediation is primarily applicable to contaminated soils. Contaminant removal is carried out *in-situ*. The technology required will be very similar to that of conventional crop-growing and harvesting practices. This approach has the advantage of causing minimal soil disturbance when compared with conventional remediation technologies.

Studies have been carried out at both laboratory and field scale. However, considerable development work is still required to assess the full potential of these techniques.

ii) Media

These techniques depend on the growth of plants and, as such, are comparatively slow. They are confined to soils which do not prove toxic to the plant and where remediation can take place over a long period of time. The technique is particularly applicable to treating the top 0.4 m of soils where plant roots actively accumulate elements.

iii) Contaminants

The technique is particularly effective for removing radionuclides and heavy metals.

iv) Work rates

No information found.

c) Performance

i) Effectiveness

A wide array of plant species accumulate large amounts of radionuclides from contaminated soils. The maximum capacities of forest and meadow plants were reported to be 28-1500 Bq g⁻¹ of ¹³⁷Cs and 300-1100 Bq g⁻¹ of ⁹⁰Sr (Nifontova *et al.*, 1989; Salt *et al.*, 1992). However, the uptake of radionuclides by plants is dependent on numerous environmental, physiological, and soil management factors. Cation exchange capacity, base saturation, base cations (e.g. potassium and calcium) and soil pH influence the amount of ¹³⁷Cs and ⁹⁰Sr that will be available for plant uptake (Entry *et al.*, 1996).

Soil type significantly effects the availability of radionuclides and their subsequent up-take by plants. Uptake of ^{137}Cs and ^{90}Sr was greater when plants were grown on sphagnum peat than on sand, silt or clay (Paasikallio, 1984). In general, plants growing in soils containing high amounts of organic matter will accumulate higher amounts of radionuclides (Entry *et al.*, 1996).

Fertilisation practices affect radionuclide uptake. For example, nitrogen fertilisation in nitrogen-limited soils increases plant growth and density which leads to greater ^{137}Cs accumulation from soils. However, the addition of phosphorus and potassium fertilisers reduced radionuclide uptake (Oulianenko *et al.*, 1995).

An accumulation rate of 0.42 MBq of ^{137}Cs per m^2 in 8 months has been from sediment of less than 4.4 kBq m^{-2} of ^{137}Cs (Dahlman *et al.*, 1969). In addition, a removal rate of 36% of ^{137}Cs and 44% of ^{90}Sr from a sand growth medium was achieved after five monthly cuttings of switchgrass (Entry *et al.*, 1996).

Trees accumulate substantial quantities of radionuclides, including ^{244}Cm , ^{137}Cs , ^{238}Pu , ^{226}Ra and ^{90}Sr (Entry *et al.*, 1995). A removal rate of 31% of ^{137}Cs and 11% of ^{90}Sr after one month, from sphagnum peat soil, was achieved with eucalyptus seedlings with bioconcentration ratios of 54:1 and 13:1, respectively (Entry and Emmingham, 1995).

The main disadvantage to phytoremediation techniques is that they require growth and harvesting over several seasons. For some species, such as woody shrubs, this could entail years.

ii) Service Life

Not applicable.

d) Costs

These techniques are considered to be a low cost technologies. However, very little information about the cost of this technique is available. Costs are likely to be similar to the costs of cultivating agricultural crops. Costs will be specific to the plant used and the site.

e) Side Effects

Care has to be exercised in introducing exotic plant species to a site in order to solve a problem. The introduced plant may become an aggressive weed replacing native species and considerable sums of money may be required to eradicate it. Containment of seed and pollen dispersal may also present problems. The use of plants which are native to the site is preferable.

The control of grazing animals (e.g. fencing or selecting unpalatable plants) also needs to be addressed.

Disposal of the contaminated biomass needs to be considered. High temperature incineration offers a practical solution to reducing the volume (Entry *et al.*, 1996). However, this will have to be carried out under controlled conditions.

6.1.2 Biosorption Techniques

a) *Description*

Biosorption treatments utilise the well-established ability of biomass to accumulate metal ions from solution. The biomass achieves this through accumulation, using a variety of mechanisms (Beveridge, 1989), such as physical adsorption, chemical bonding or production of precipitating agents (e.g. phosphate and sulphide). Both living and non-living biomass can be used to extract metal ions. For living organisms, accumulation can be both extracellular and intracellular (Beveridge, 1989). However, non-living biomass can retain many of the surface properties of the living organisms and, in addition, adsorption may be enhanced by chemically modifying the surface of the material.

Treatment of the contaminated water is accomplished by first filtering and then passing it through a vessel containing the biomass. Here the contaminant is extracted from the solution by biosorption onto the biomass (Volesky, 1990). The water, depleted of the contaminant, may then be returned to the environment. The contaminated biomass is then available for further treatment.

Biosorption can be a fairly rapid process with equilibrium times as low as a few minutes (Singleton and Simmons, 1996). It is accomplished *ex-situ* and may be used to treat contaminated water extracted from a site or the contaminated solutions from soil washing (see Section 4.2.1).

b) *Applicability*

i) *Means*

In biosorption processes, the solution is filtered and passed through a vessel containing the biomass. In the case of living biomass the vessel is usually a bioreactor. However, non-living biomass is normally immobilised by encapsulation or cross linking and the vessel essentially acts as a column.

ii) *Media*

The techniques are applicable to the treatment of contaminated groundwater and washings from contaminated soils.

iii) *Contaminants*

The technique is suitable for the removal of radionuclides, heavy metals, and also inorganic and organic contaminants. However, the technique is prone to interference from other material in the contaminated solution which may complex the contaminant or compete for binding sites on the biomass.

iv) *Work rates*

No information found.

c) *Performance*

i) *Effectiveness*

A wide range of microbial and fungal biomass have been shown to accumulate metal ions (Volesky and Holan, 1995). Typical capacities of the biomass are 25-400 mg g⁻¹. However, the capacity is significantly affected by factors such as the choice of biomass, the oxidation state of the metal ion, the pH, complex formation, hydrolysis of the metal ion, the presence of competing metal ions and

temperature (Volesky and Holan, 1995; Singleton and Simmons, 1996). Nuclides sorbed by biomass include: uranium(VI) (Galum *et al.*, 1983; Bengtsson *et al.*, 1995), thorium(IV) (Tsezos and Volesky, 1981; Strandberg *et al.*, 1981), strontium (Averby and Tobin, 1992), silver (Singleton and Simmons, 1996) and caesium (Averby, 1995). Phosphate-producing organisms have been found to accumulate radionuclides such as uranium(VI), americium(III), plutonium(VI), neptunium(IV), lanthanum(III) and thorium(IV) (Macaskie *et al.*, 1996).

The rate of removal of metal ions from solution can be very rapid. Often equilibrium is achieved within a matter of minutes. Biosorption can achieve removal efficiencies exceeding 90% for uranium and plutonium (Shumate *et al.*, 1978).

The main disadvantage to this type of approach is that living biomass will be constrained to a narrow range of physiological conditions which may not be optimal to metal accumulation. In addition, other nutrients may be required. Non-living biomass avoids this, but metabolic processes can no longer be utilised.

Whilst studies have been carried out on both a laboratory and pilot scale considerable further development is needed to assess the full potential of these techniques.

ii) Service Life

Not applicable.

d) Costs

The costs of this process are poorly defined and will vary considerably between sites.

The extraction of contaminated groundwater will require similar technology to that of ion-exchange (see Section 5.1.2). Therefore, many of the operation and maintenance costs associated with the application of ion exchange resins will be similar to those incurred by biosorption. Differences will be largely reflected in the cost of materials and how they are applied. The costs of biomass is likely to be considerably cheaper than ion exchange resins. However, metal uptake by biomass is likely to be significantly less efficient than by an ion exchange resin. Biosorption process will also be confined to a narrow range of conditions. Therefore, the estimated costs of this whole process is taken to be similar to that of ion exchange, i.e. 1 to 3 EUR m⁻³.

Treatment of the 'wash waters' from a soil washing process (see Section 4.2.1) could be made considerably cheaper if the biosorption process is integrated with the soil washing process. The cost of treatment will be the cost of operating and maintaining a filter plus operating a reactor or column containing the biosorbent. The cost of this would be low.

There will also be an operation and maintenance cost associated with the disposal of waste from the process.

e) Side Effects

In some cases, the radionuclide which has been accumulated by the biomass, may be chemically stripped from the biomass (usually by means of a strong acid) and collected as a concentrated, low volume waste for disposal or further treatment. This technique is often used with non-living biomass so that the biomass may be reused. Alternatively, the contaminated biomass can be disposed of directly as a contaminated solid. However, it is more likely that the volume of the waste would be reduced (usually by incineration) before disposal.

6.1.3 Bioleaching Techniques

a) *Description*

This technique utilises the ability of certain micro-organisms to create conditions in contaminated soils where the solubilities of relatively insoluble metal ions are increased. Micro-organisms capable of achieving this include *Thiobacillus thiooxidans* and *Thiobacillus ferrooxidans*. These convert sulphide or sulphur to sulphuric acid. This has two beneficial effects: it converts insoluble sulphides to the more soluble sulphates and it reduces the pH of the soil, thereby increasing the solubilities of radionuclide compounds.

The technique is normally carried out *ex situ*. The contaminated soil is collected and stored in a container through which water is slowly passed by either drip or spray irrigation. The micro-organisms in the soil utilise the sulphur and solubilises the radionuclide. The resulting leachate is collected for further treatment such as precipitation of the radionuclide with hydrated lime (Seidel *et al.*, 1995).

b) *Applicability*

i) *Means*

Microbial digestion of sulphide is a relatively slow process, with treatments lasting a number of weeks (Seidel *et al.*, 1995).

Studies have been carried out on both a laboratory and pilot scale (Seidel *et al.*, 1995). However, most of these have been involved solubilisation of heavy metals rather than radionuclides. In all cases, considerable further development is needed to assess the full potential of the technique.

ii) *Media*

Contaminated media which can be treated by this process include soils, sediments and mine tailings (Seidel *et al.*, 1995; Gourdon and Funtowic 1995).

iii) *Contaminants*

The technique is particularly useful for treating the insoluble sulphides of radionuclides and heavy metals.

iv) *Work rates*

No information found.

c) *Performance*

i) *Effectiveness*

Bioleaching depends on the generation of sulphuric acid to lower the pH. Therefore, the buffering capacity of the contaminated sediment has an important influence on the effectiveness of this technique (Seidel *et al.*, 1995). Often an additional source of sulphur to promote microbial activity and to overcome the buffering capacity of the soil.

A pilot scale project, using 60 tonne of sediment containing 0.5% sulphur, showed that *Thiobacilli* would oxidise sulphur at a rate of $70 \text{ g m}^{-3} \text{ h}^{-1}$. After 4 weeks, the pH of the sediment was reduced from pH 6 to about pH 2.

To date, little work has been carried out on bioleaching of radionuclides. However, the approach is likely to be well-suited to removing radionuclides from contaminated media, on the basis that it is suitable for heavy metals.

Laboratory based studies suggest that after 60 days, bioleaching would typically remove 63% of heavy metals (cadmium, nickel, copper and zinc) from sediments containing 1% sulphur (Seidel *et al.*, 1995). Similar results were obtained by Gourdon and Funtowic (1995).

ii) Service Life

Not applicable.

d) Costs

The costs associated with this technique are poorly defined. However, the costs will include: collection of the contaminated material, the application of water to the material, collection of the resulting leachate, the cost of sulphur and the cost of mixing the sulphur with the contaminated material.

The costs associated with this technique are likely to be similar to those of chemical separation (see Section 5.1.1), i.e. 100 to 400 EUR tonne⁻¹.

e) Side Effects

The leachate generated by this process will be acidic and contain a high concentration of radionuclides. It may be necessary to neutralise the leachate before disposal of the contaminated solution.

7. Application of Remediation Techniques to RESTRAT Example Sites

An examination of the information, given in Sections 3, 4 and 5, shows that the cost and performance values associated with each remediation technique, can vary over a large range. This reflects the fact that these techniques have been applied to a variety of radioactive wastes, on a variety scales and under a variety of conditions. The application of these techniques to a specific site must reflect this variability. This can be achieved through specifying uncertainty ranges for the values used to calculate the impact of each remediation technique.

The approach employed in assessing the impact of the remediation techniques for a specific site is to use the full ranges of reported costs and performances (except for pilot plant tests) as ranges of uncertainty for all the example sites. Triangular (for ranges of less than one order of magnitude) and log-triangular (for ranges of greater than one order of magnitude) distribution functions are assumed for each site. The maximum and minimum of the range is independent of the site, however, the mode depends on the site itself, and takes account of factors such as accessibility, the nature of the contaminated waste and local conditions. Hence, the maximum and minimum values for the performance and cost of remediation will be common to all sites, but the choice of mode will be site-specific.

The range of performance, cost and exposure times (to restoration workers), extracted from the information given in this report, are summarised in Tables 5 to 7, respectively. Where values could not be obtained from the literature then these were estimated on the basis of their similarity to other for which values were available.

Table 5 The performance of remediation techniques

Remediation Technique	Unit [†]	Value
<p>Source Removal</p> <p>Soil excavation</p> <p>Soil scraping</p> <p><u>NB</u> DF values will depend on the nature of the contamination (at depth or superficial), the depth of the excavation and the radionuclide considered.</p> <p>With a turf-harvester higher DF values were reported for turf-podzols.</p>	<p>DF</p> <p>DF</p>	<p>1 - 20</p> <p>1 - 20</p>
<p>Physical separation (<i>ex-situ</i>)</p> <p>Soil washing</p> <p><u>NB</u> In some pilot plant tests of soil washing also higher values are reported (between 4 and 80). However when separation and excavation are considered to be carried out on the same contaminated soil quantity it is unreasonable to expect that separation (with the less contaminated fraction left in place) would yield a DF values as high or higher than excavation.</p> <p>With a higher RF value, a lower DF value needs to be associated (negative correlation)</p> <p>Flotation</p> <p><u>NB</u> With a higher RF value, a lower DF value needs to be associated (negative correlation)</p> <p>Filtration</p> <p><u>NB</u> Range indicated for all radionuclides</p> <p>Fraction of U, Pu, Am reported removed</p> <p>Fraction of Ra reported removed</p> <p>Fraction of α-emitters reported removed</p>	<p>DF</p> <p>RF</p> <p>DF</p> <p>RF</p> <p>DF</p>	<p>1 - 10</p> <p>50 - 98 %</p> <p>1 - 10</p> <p>28 - 97 %</p> <p>2 - >100</p> <p>>99%</p> <p>43%</p> <p>86%</p>
<p>Chemical Separation (<i>ex-situ</i>)</p> <p>Chemical solubilization</p> <p><u>NB</u> This large range reflects the number of possible variations available for this technique, e.g. changes to pH, changes to solvent, etc. However, the technique is applicable to the types of radionuclide found.</p> <p>A RF value is not meaningful here, given that a liquid waste is produced.</p> <p>Ion exchange (liquids)</p> <p><u>NB</u> A RF value is not meaningful here, given that a liquid waste is produced.</p>	<p>DF</p> <p>DF</p> <p>DF</p>	<p>1 - 20</p> <p>20 - 100 (Cs)</p> <p>3 - 100 (U)</p>
<p>Biological Separation</p> <p>Biosorption (liquids)</p> <p><u>NB</u> Range indicated: for all radionuclides</p> <p>Fraction of U reported to be removed</p>	<p>DF</p>	<p>2.5 - >100</p> <p>>99%</p>
<p>Containment</p> <p>Capping</p> <p><u>NB</u> For most used capping materials (impermeable layers) the permeability lies within the indicated range, with probably, for asphalt layers k-values at the lower side and for clay layers at the higher side, also depending on soil conditions.</p> <p>Subsurface barriers</p>	<p>k</p> <p>k</p>	<p>1×10^{-12} - 1×10^{-9} m s⁻¹</p> <p>1×10^{-12} - 1×10^{-8} m s⁻¹</p>
<p>Physical Immobilization</p> <p><u>NB</u> Mobility reduction factor (MRF) factor derived from a single value (20).</p>	<p>MRF</p>	<p>5 - 25</p>
<p>Chemical Immobilization</p> <p><u>NB</u> Leaching tests show a reduction in mobility of between 80 and 100%.</p>	<p>MRF</p>	<p>5 - 50</p>

[†] DF = decontamination factor; RF = waste reduction factor; k = permeability coefficient; MRF = mobility reduction factor.

Table 6 The costs of remediation techniques

Remediation Technique	Cost
Source Removal	
Soil Excavation	
Excavation costs (including transport)	50 - 150 EUR m ⁻³
Excavation costs (including transport and RCRA disposal)	450 - 800 EUR m ⁻³
<u>NB</u> This type of disposal would be appropriate for the disposal of the total contaminated soil	
Disposal cost for radioactive material (including transport)	2000 - 3000 EUR m ⁻³
<u>NB</u> This type of disposal would be appropriate for the disposal of the most contaminated fraction	
Soil Scraping	
Scraping costs (including transport)	1 - 3 EUR m ⁻²
<u>NB</u> This is for the top 50 - 75 mm layer	(surface area)
Physical Separation (<i>ex-situ</i>)	
Soil washing	
Soil washing costs	150 - 500 EUR m ⁻³
Cost of excavation and transport (prior to washing)	50 - 150 EUR m ⁻³
Cost of disposal of radioactive residue (including transport)	2000 - 3000 EUR m ⁻³ (residue)
Flotation	
Capital costs	15 - 240 EUR / m ³
<u>NB</u> Very dependent on the size of the flotation unit and on the volume of soil to be treated.	
Derived from the capital cost of 150 kEUR for a large unit (assumed for 10000 m ³ of soil) and the capital cost of 24 kEUR for a small unit (assumed for minimum 100 m ³ of soil).	
Operational costs	0.8 - 3.8 EUR m ⁻³
Cost of excavation and transport (prior to flotation)	50 - 150 EUR m ⁻³
Cost of disposal of radioactive residue (including transport)	2000 - 3000 EUR m ⁻³ (residue)
Filtration (liquids)	0.1 - 3.8 EUR m ⁻³ (liquid)
Costs of disposal of radioactive residues (used filters)	2000 - 3000 EUR m ⁻³ (residue)
Chemical Separation (<i>ex-situ</i>)	
Chemical solubilization	
Separation costs	130 - 670 EUR m ⁻³
Cost of excavation and transport (prior to solubilization)	50 - 150 EUR m ⁻³
Cost of disposal of radioactive residue (including transport)	2000 - 3000 EUR m ⁻³ (residue)
Ion exchange (liquids)	1.3 - 2.5 EUR m ⁻³ (liquid)
Costs of disposal of radioactive residues	2000 - 3000 EUR m ⁻³ (residue)
Biological Separation	
Biosorption (liquids)	1 - 3 EUR m ⁻³ (liquid)
Costs of disposal of radioactive residues	2000 - 3000 EUR m ⁻³ (residue)

Table 6 (cont.)

The Costs of Remediation Techniques

Remediation Technique	Cost
<u>Containment</u>	
Capping	30 - 45 EUR m ⁻² (surf. area)
Subsurface Barriers	
Slurry walls	510 - 710 EUR m ⁻² (barrier)
Grout curtains	310 - 420 EUR m ⁻² (barrier)
<u>Physical Immobilization</u>	
<i>Ex-situ</i>	
Cost of excavation and transport (prior to immobilization)	25 - 150 EUR m ⁻³ 50 - 150 EUR m ⁻³
<i>In-situ</i>	50 - 310 EUR m ⁻³
<u>Chemical Immobilization</u>	
<i>Ex-situ</i>	
Cost of excavation and transport (prior to immobilization)	60 - 420 EUR m ⁻³ 50 - 150 EUR m ⁻³
<i>In-situ</i>	60 - 420 EUR m ⁻³

Table 7 The exposure times (restoration workers) of remediation techniques

Remediation Technique	Exposure Time (Restoration Workers)
<p>Source Removal</p> <p>Soil excavation <u>NB</u> Time for soil removal (including loading), dependent on surface area, terrain conditions</p> <p>Soil scraping <u>NB</u> For the top 50 - 75 mm layer</p>	<p>0.2 - 1 man h m⁻³</p> <p>0.03 - 0.3 man h m⁻² (surf. area)</p>
<p>Physical separation (ex- situ)</p> <p>Soil washing <u>NB</u> Labour rates reported were: 5.5 - 90 tonne h⁻¹</p> <p>Excavation and transport of the soil (prior to separation)</p> <p>Flotation Excavation and transport of the soil (prior to separation) <u>NB</u> As a first approximation, these values are taken to be the same as those for soil washing.</p> <p>Filtration (liquids) <u>NB</u> As a first approximation, these values are taken to be the same as those for ion exchange</p>	<p>0.05 - 0.5 man h m⁻³</p> <p>0.2 - 1 man h m⁻³</p> <p>0.05 - 0.5 man h m⁻³</p> <p>0.2 - 1 man h m⁻³</p> <p>0.4 - 1.4 man h m⁻³ (liquid)</p>
<p>Chemical Separation (ex-situ)</p> <p>Chemical solubilization <u>NB</u> Throughput was 2 - 5 tonne h⁻¹ (assuming 3 workers)</p> <p>Excavation and transport of the soil (prior to separation)</p> <p>Ion exchange (liquids) <u>NB</u> Working rate was 50 - 200 m³ day⁻¹ (assuming 3 workers)</p>	<p>1 - 2.5 man h m⁻³</p> <p>0.2 - 1 man h m⁻³</p> <p>0.4 - 1.4 man h m⁻³ (liquid)</p>
<p>Biological Separation</p> <p>Biosorption (liquids) <u>NB</u> As a first approximation, these values are taken to be the same as those for ion exchange</p>	<p>0.4 - 1.4 man h m⁻³ (liquid)</p>
<p>Containment</p> <p>Capping <u>NB</u> According to Lehto, (1994)</p> <p>Subsurface barriers <u>NB</u> As a first approximation the same values (per m³ of barrier) as those for physical immobilization (<i>in-situ</i>) were assumed</p>	<p>0.03 - 0.3 man h m⁻² (surf. area)</p> <p>0.06 - 0.4 man h m⁻³ (barrier volume)</p>
<p>Physical Immobilization</p> <p><i>Ex-situ</i> <u>NB</u> As a first approximation the same values were used as those for physical separation (<i>ex-situ</i>)</p> <p>Excavation and transport of the soil (prior to immobilization)</p> <p><i>In-situ</i> <u>NB</u> Values of 18 - 72 tonne h⁻¹ are reported, dependent on depth (shallow - deep soil)</p>	<p>0.05 - 0.5 man h m⁻³</p> <p>0.2 - 1 man h m⁻³</p> <p>0.06 - 0.4 man h m⁻³</p>

Chemical Immobilization	
<i>Ex-situ</i>	0.05 - 0.5 manh m ⁻³
<u>NB</u> As a first approximation the same values were used as for physical immobilization (<i>ex-situ</i>)	
Excavation and transport of the soil (prior to immobilization)	0.2 - 1 manh m ⁻³
<i>In-situ</i>	0.06 - 0.4 manh m ⁻³
<u>NB</u> as a first approximation the same values were used as for physical immobilization (<i>in-situ</i>)	

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