SOIL WASHING TREATMENT TRIALS AT UKAEA

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ABSTRACT

Laboratory and pilot scale soil washing trials have been carried out by UKAEA using radioactively contaminated soils from three of its sites. The radioactivity in these soils categorises them at the lower end of the Low Level Waste.

The results showed that soil washing by particle separation processes could only produce a radiologically "clean" bulk fraction (exempt from regulatory control) where the contaminant levels in the original soil were only 2-3 times higher than this level.

Soils with a higher contamination content could be remediated to:

- a risk based clean-up target, or
- a waste categorised as Very Low Radioactive Material. (A disposal route for this material is currently being considered as it is anticipated that it will be less costly than disposal of Low Level Waste).

The use of chemical leaching agents to enhance contaminant reduction from the partially cleaned products from the particle separation processes was generally ineffective – particularly for Cs-137 removal. Only when concentrated acids were used was appreciable contamination removed.

An estimate of the cost of treatment based on grain size separation and attrition scrubbing for a 5-10 tonne/h plant was found to be £80-£100 /tonne of soil treated. These costs include capital, labour, commissioning and decommissioning and analysis but, exclude the disposal of the contaminated fractions. In the cost estimate it is assumed that the soil would be treated in 1,000 m^3 batches and that at least 10 batches would be processed in the life of the plant.

In addition to washing soils, tests were also carried out to assess the removal of contamination from rocks using a tumbling mill as a scrubber. The surface activity of the rocks classifies them as LLW. The scrubbing mill was shown to very effective at removing surface contamination. The resulting products were (i) scrubbed "clean" rocks which were mainly free release, and (ii) abraded bulk material which is VLRM. Very little of the contamination transferred into the process water.

INTRODUCTION

UKAEA is responsible for managing the decommissioning of reactors and other redundant radioactive facilities from nuclear research and development (R&D) programmes.

This decommissioning includes the restoration of its sites which in turn includes dealing with the remediation of contaminated ground. As well as using "traditional" remediation methods for contaminated ground, UKAEA is assessing and evaluating other techniques through a targeted R&D programme.

Where radioactively contaminated ground is excavated, it results in radioactive waste which is very costly to dispose of. Technologies are therefore being assessed which might minimise the wastes produced and therefore reduce the overall disposal costs. One particular

technology being studied is soil washing. This has the potential to reduce the volume of material requiring disposal as Low Level Radioactive Waste (LLW). This paper presents an outline of laboratory and pilot scale soil washing trials carried out by UKAEA using radioactively contaminated soils from three of its sites.

APPROACH

Soil washing is based on either:

a) physical separation processes derived from the mineral processing industry in which the contaminated particles are segregated from the relatively uncontaminated bulk, or
b) chemical leaching processes in which the contaminants are selectively dissolved and then recovered from solution in a "concentrated" form, or
c) a combination of both (a) and (b).

The soil washing tests carried out by UKAEA therefore aimed to ascertain the degree to which these processes could clean the radioactively contaminated soils and rocks on its sites. The emphasis of the tests was primarily to use physical separation processes and then to assess enhancement by combining with chemical leaching processes.

The trials were assessed against "clean-up" targets shown in Table I:

The trials were carried out in three stages:

Stage 1: Initial laboratory tests on nine soils from different areas of three UKAEA sites. The aim of the tests was to assess contaminant segregation based on differences in particle grain size;

Stage 2: Pilot scale tests carried out at the UKAEA Soil Characterisation and Remediation Testing Pilot Plant facility based at Harwell. The aim of the tests was to ascertain contaminant reduction based on physical separation processes. The work was carried out on two bulk samples.
Sample 1: 800 kg of Cs-137 contaminated soil. Processes involved screening and settling velocity separation with intermediate attrition scrubbing.
Sample 2: 500 kg of Cs-137 contaminated rock. Processes involved scrubbing in a tumbling mill, followed by size separation of the products using screens;

Stage 3: Further laboratory tests on fractions collected from the pilot scale tests. These tests aimed to assess further contaminant reduction based on further processing.

Table I: Clean-up Targets for Soil Washing Trials

	Radioactive Waste Categorisation of Original Material	Clean-up Target	Comments
(i)	Low Level Waste (LLW)	Very Low Radioactive Material (VLRM)*	In this paper the boundary between LLW and VLRM is 40 Bq/g short-lived beta gamma activity and 1 Bq/g long lived alpha activity.
(ii)	LLW	Free Release Material (FRM)	Material <0.4 Bq/g total activity is exempt from UK regulation and therefore can be free released.
(iii)	LLW	Risk Based Clean-up Level (RBCL)	A level of <4 Bq/g short-lived beta-gamma activity and <1 Bq/g alpha activity was established as the risk based clean-up level for soils from the particular site.
(iv)	VLRM*	RBCL	As in (iii), clean-up <4 Bq/g short-lived beta-gamma activity and <1 Bq/g alpha activity.
(v)	VLRM*	FRM	As in (ii), clean-up to <0.4 Bq/g.

*It has been estimated that the disposal of VLRM in a purpose built facility could be significantly cheaper than disposal of this material as LLW. Indicative costs show a potential saving of at least a factor of four.

DETAILS OF THE STAGES

Stage 1 – Laboratory Tests

The nine soils tested at Stage 1 varied in both soil composition and in the degree of contamination. The most dominant radionuclide in each soil was Cs-137.

The laboratory tests were used to assess:

- the preferential distribution of the contaminants to particular soil fractions;
- the contaminant content of the contaminant-depleted fractions;
- the proportion of material in the contaminant-depleted fractions.

The laboratory tests at this stage used a "wet" screening method where the soil is mixed with water to form a slurry and the particles are then sized at 10 mm down to 0.038 mm using laboratory screens. The "wet" method ensures that the soil is disaggregated into its individual grains.

Stage 2 – Pilot Scale Tests

The Soil Characterisation and Remediation Testing Pilot Plant has been developed over a number of years. Its original function was as a mineral processing pilot scale plant for testing the beneficiation of mineral ores. The plant contains a wide variety of individual processing equipment which can be joined together in a treatment process using pumps and flexible pipe. In recent times the plant has been adapted for use as a soil washing testing facility. For radioactively contaminated soils, the plant is operated as a series of "batch" operations where the products from a particular process are collected either as an end product or for treatment by another downstream process or series of processes.

Pilot scale tests were carried out on two bulk samples. These were:

- Sample 1: contaminated soil; and
- Sample 2: contaminated rock.

In both cases the material tested was Low Level Waste where the majority of the contamination was Cs-137.

Sample 1
The process configuration used for the trial on Sample 1 is presented in Figure 1 together with a description of the equipment used and its purpose.

The process aimed to separate contaminated fractions based on differences in grain size and settling velocity as well as assessing the effects of attrition scrubbing on the partially cleaned "sands" fraction.

In addition, samples were collected for further testing at Stage 3 (as outlined below).

Sample 2
The main processing equipment used during the treatment of Sample 2 was a tumbling "wet" ball mill which was used as an attrition mill. No balls were added to the mill as the rocks themselves acted as an autogenous load.

Rocks up to 37 kg were loaded into the mill and tumbled for up to 60 minutes. Abraded material was withdrawn from the mill during this period through a series of peripheral openings.

Stage 3 - Further Laboratory Tests

Laboratory tests on products from the pilot scale tests for Samples 1 and 2 were further tested as follows:

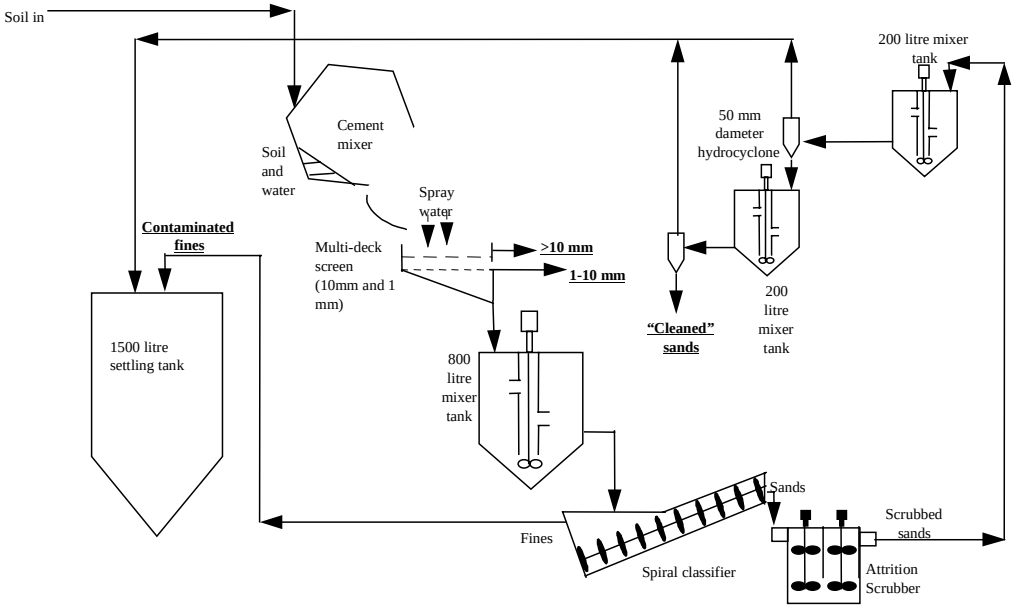
Sample 1

Sink-Float Tests Using Heavy Liquids:
Aimed at assessing the effect of removing contaminated light peaty matter from the scrubbed sands fractions.

Chemical Leaching Tests:
Aimed at assessing the removal of contamination from products from the soil washing trials by chemical leaching agents.

Sample 2

Sizing Tests:
Aimed at assessing the distribution of contamination on particular size fractions of the abraded material.



Equipment Description and Purpose

Equipment	**Purpose**
"Cement Mixer"	To mix the soil with water and to disaggregate lumps.
Multi-deck Vibratory Screen	To size material at 10mm and 1 mm.
Mixer-Storage Tanks:	To store slurry prior to processing with further downstream equipment.
Spiral (screw) classifier :	To separate contaminated fines from less contaminated "sands" and to reduce the water content of the sands so that they can be efficiently scrubbed in the attrition scrubber.
Attrition Scrubber:	To abrade potentially contaminated surface coatings from the sands.
Hydrocyclones	To remove the contaminated fine particles, generated during scrubbing, from the "cleaned" sands. Two stages of hydrocycloning were used with the second removing "entrained" fines from the hydrocyclone underflow (sands) product.
Settling Tanks:	To settle the fine solids from the process water aided by the addition of a coagulant.

Figure 1: Pilot Scale Treatment Process Used for Contaminated Soil – Sample 1

RESULTS

Laboratory Tests

Results from the laboratory tests showed that in nearly every soil examined, there is a marked elevation of the contaminant level in the finest fractions compared to the coarser fractions. From the three sites, there was only one exception to this. In soil from that particular site, the distribution of the radionuclides with size was erratic. It is suspected that this exceptional behaviour reflects the nature of the soil at the site which, when compared to the other three sites, is mostly chalk. The finest fractions in this soil tended to be fine weathered chalk rather than true clay.

Table II shows the degree of contamination depletion in the coarser fractions of the soils. Rather than choosing one specific cut-off size to mark the division between coarse and fine sizes, the criteria used was based on a 70:30 split between the proportion of coarse material to fine material in the particular soil. Thus, for some soils, this split might be at a size corresponding to 0.3 mm whilst in other soils this split might correspond to a size of 0.5 mm. In Table II, the coarse fractions are labelled "Treated" and the ratio of the original material to "treated" is termed the "Decontamination Factor"(DF).

Table II: Results from Laboratory Grain Size Analysis Tests

Site	**A**							**B**	**C**
Soil Sample Number	**A1**	**A2**	**A3**	**A4**	**A5**	**A6**	**A7**	**B1**	**C1**
Cs-137									
Original Bq/g	3.61	10.91	23.08	39.48	75.41	41.81	33.60	0.13	0.69
"Treated" Bq/g	0.75	4.85	17.78	31.11	35.80	26.00	20.55	0.13	0.09
DF	5	2	1	1	2	2	2	1	8
Sr-90									
Original Bq/g	1.27	0.25	0.52	-	0.34	0.25	-	-	-
"Treated" Bq/g	0.34	0.13	0.39		0.25	0.25			
DF	4	2	1		1	1			
Pu-238									
Original Bq/g	-	-	0.43	-	-	0.51	-	0.40	-
"Treated" Bq/g			0.24			0.37		0.35	
DF			2			1		1	
Pu-239									
Original Bq/g	-	0.34	1.51	-	0.31	2.31	-	3.32	-
"Treated" Bq/g		0.07	0.87		0.11	1.65		3.00	
DF		5	2		3	1		1	
Pu-241									
Original Bq/g	0.44	0.85	4.12	-	0.87	5.06	-	2.58	-
"Treated" Bq/g	0.18	0.19	2.12		0.27	3.60		2.58	
DF	2	5	2		3	1		1	
Am-241									
Original Bq/g	-	0.15	0.50	-	-	1.12	-	-	-
"Treated" Bq/g		0.03	0.33			0.78			
DF		5	2			1			
Co-60									
Original Bq/g	-	-	-	-		-	-	-	1.34
"Treated" Bq/g									0.26
DF									5

Note: no value indicates no activity of significance.

Other than the poor preferential distribution of contaminants to the chalk-rich soil, there is no other obvious trend which accounts for the variability of the DF with soil type, site type or level of contamination in the original material. In addition, as anticipated, the laboratory tests showed that the contaminants were mostly insoluble in water.

One of the soils examined had a very high proportion of material less than 0.038 mm (61% by mass). Particles >2.0mm in this soil showed a very marked depletion in Cs-137 contamination. However the proportion of material in this fraction is only 4% by weight making the volume reduction potential of this material poor.

Two of the soils examined, had short-lived radionuclide levels which were greater than 40 Bq/g (the boundary between VLRM and LLW – see * in Table I). The laboratory sieve analysis for one of these soils is shown in Tables III. This showed that a process based on grain size separation alone could potentially produce a bulk fraction 80% of the mass, with a short-lived nuclide content of <40 Bq/g.

Table III: Example of Size-Contaminant Distribution in one of the Soils Tested

(a) Fractional Distribution

Grain Size mm	Weight %	Cs- 137		Sr- 90		Pu- 239		Pu- 241	
		Bq/g	Distrib % in solids	Bq/g	Distrib % in solids	Bq/g	Distrib % in solids	Bq/g	Distrib% in solids
> 9.5 mm	6.4	65.65	5.6	0.40	7.4	0.07	1.5	0.18	1.3
2.0 - 9.5	17.6	31.04	7.3	0.34	17.4	0.14	7.8	0.35	7.1
0.5 - 2.0	44.9	33.41	19.9	0.19	24.8	0.10	14.1	0.25	13.0
0.3 - 0.5	10.7	43.73	6.2	0.26	8.1	0.18	6.2	0.55	6.9
0.063 – 0.3	11.8	90.03	14.1	1.20	41.3	0.38	14.3	1.19	16.2
0.038 – 0.063	2.4	165.30	5.3	0.02	0.1	1.05	8.2	2.85	7.9
<0.038	6.2	507.00	41.6	0.05	0.9	2.40	47.9	6.62	47.6
Soluble Fraction	-	0.10		0.05	-	<<0.01	-	<<0.01	-
TOTAL	100.0	75.51	100.0	0.39	100.0	0.31	100.0	0.87	100.0

(b) Cumulative Distribution of the Soil in (a)

Grain Size mm	Cumulative Weight %	Cs- 137		Sr- 90		Pu- 239		Pu- 241	
		Cum Bq/g	Cum Distrib % in solids	Cum Bq/g	Cum Distrib % in solids	Cum Bq/g	Cum Distrib % in solids	Cum Bq/g	Cum Distrib % in solids
>9.5	6.4	65.65	5.6	0.40	7.4	0.07	1.5	0.18	1.3
>2.0	24.0	40.25	12.8	0.36	24.9	0.12	9.3	0.30	8.4
>0.5	68.9	35.80	32.7	0.25	49.7	0.11	23.4	0.27	21.5
>0.3	79.6	36.87	38.9	0.25	57.8	0.12	29.6	0.31	28.3
>0.063	91.4	43.73	53.0	0.37	99.0	0.15	43.9	0.42	44.6
>0.038	93.8	46.86	58.4	0.36	99.1	0.17	52.1	0.48	52.4
All	100.0	75.41	100.0	0.34	100.0	0.31	100.0	0.87	100.0

Cum = Cumulative
Distrib = Distribution

Pilot Plant Tests

(i) Pilot Tests on Soil

The soil tested in the pilot scale tests had been analysed as containing 50 Bq/g beta-gamma activity and 15 Bq/g Cs-137 and was therefore classified as LLW. The results from soil washing tests with this soil are presented in Table IV.

The combined contaminant-depleted products were found to contain 1.78 Bq/g Cs-137 and 1.37 Bq/g Sr-90, whilst the contaminant-enriched products were found to contain 14.78 Bq/g Cs-137 and 6.91 Bq/g Sr-90. Activity levels in the process water were negligible. Although not reaching below the exemption level of 0.4 Bq/g, the contaminant-depleted fractions were VLRM and were below a Risk Based Clean-Up Level of 4 Bq/g.

It should be noted that an activity balance around the circuit showed that the original material must have been significantly less than the reported 15 Bq/g Cs-137. The reason this value was suspected of being too high is probably due to a sampling procedure which was biased towards sampling fines at the expense of the less contaminated coarser fractions.

Table IV: Pilot Plant Results

Product	Weight %	Cum Wt%	Cs-137 Bq/g	Cum Cs-137 Bq/g	Sr-90 Bq/g	Cum Sr-90 Bq/g
Contaminant Depleted Products						
>10 mm screen oversize	52.7	85.6	1.10	1.78	0.85	1.37
1-10 mm screen product	20.0		2.56		1.90	
Hydrocyclone Underflow (Cleaned Sands)	12.9		3.35		2.70	
Contaminant Enriched Products						
Spiral Classifier Overflow (Fines)	9.4	14.4	19.10	14.78	8.3	6.91
Total Hydrocyclone Overflows	5.0		6.64		4.3	

Cum = Cumulative

(ii) Pilot Tests on Contaminated Rocks

Two batches of rocky material were tested. In the first batch, the average activity was 411 counts per second beta-gamma activity per kg of rock (cps/kg), maximum of 1200 cps/kg. In the second batch activity levels were 703 cps/kg, maximum of 2400 cps/kg.

The results from the tumbling scrubber tests are presented in Table V.

Results from these rock scrubbing tests show that with both batches of material the contamination could be removed from the rocks such they could be free released or were below the target level for VLRM. The scrubbed rocks were 66%-67% of the mass of the original material. As the activity in the abraded products is distributed throughout its mass, this material was also below the target level for VLRM.

Table V: Results from Rock Scrubbing Tests

	Test 1	Test 2
Before Scrubbing		
Total Weight of Rocks Scrubbed (dry matter basis) kg	229	264
Activity Average: cps beta-gamma/ kg Maximum: cps	411 1200	703 2400
Waste Classification	LLW	LLW
30 minute Scrub		
Weight of Remaining Rocks	-	188
Activity cps beta-gamma Average/kg Maximum	-	56 200
Weight of Abraded Material kg	-	69
% material removed	-	29%
Waste Classification of Scrubbed Rocks	-	VLRM some Free Release
60 minute Scrub		
Weight of Remaining Rocks (dry matter basis) kg	155	174
Activity cps beta-gamma Average/kg Maximum	17 40	28 100
Weight of Abraded Material	71	84
% material removed	33%	34%
Waste Classification of Scrubbed Rocks	Free Release some VLRM	Free Release some VLRM
Waste Classification of Abraded Material	VLRM (3 Bq/g)	VLRM (6 Bq/g)

LLW – Low Level Waste
VLRM – Very Low Radioactive Material (<40 Bq/g beta-gamma activity)

Further Laboratory Tests

Further Laboratory Tests on Soil (Fractions from Sample 1)

Sink-Float Tests Using Heavy Liquids

Sink-float tests were carried out on some of the products from the pilot scale tests using a solution of sodium polytungstate (SPT). This was made up to a liquid of density 2.5 g/ml. The resulting sink and float products were weighed and analysed to ascertain whether the removal of peaty matter (<2.5 SG) could reduce the contamination in the remaining bulk fraction (SG >2.5).

The results showed that material <2.5 SG contained twice as much contamination as that in the >2.5 SG fraction. However, the proportion <2.5 SG in the total soil is relatively small (only 4 weight %). This indicates that relatively little more contamination could be removed from the contaminant-depleted fractions by adding further treatment stages aimed at removing the peat.

Chemical Leaching Tests

Products from the pilot scale tests were chemically leached with dilute solutions of the following reagents:

KCl, EDTA, NaOH, acetic acid, citric acid, oxalic acid, hydrochloric acid and nitric acid.
In general the results were poor with <16% of the activity leached.

These results were somewhat improved with concentrated acids. The best results were on the cyclone underflow sand and 1-10 mm fractions where concentrated HCl and HNO_3 leached 50% and 100% of the activity respectively.

Further Laboratory Tests on Abraded Rock (Fractions from Sample 2)

During the rock scrubbing pilot plant tests, abraded material was removed at 15 minutes, 30 minutes and 60 minutes. This material was subsequently sized in the laboratory and radiochemically analysed to ascertain the ease of contaminant removal. The results are presented in Table VI.

Table VI: Cs-137 Analysis of Size Fractions from the Abraded Material from the Rock Scrubbing Tests (dry matter basis)

Scrubbing Duration	Wt % Relative to the Feed	Cs-137	Cs-137 for the fraction	Cumulative Cs137 for the fraction
15 min				
>1mm	5.5	0.71		
0.3 - 1	1.6	1.6		
<0.3	5.8	12.51		
Sub-total	***12.9***		***6.09***	***6.09***
30min				
>1mm	3.7	0.24		
0.3 – 1	1.9	0.65		
<0.3	4.4	2.19		
Sub-total	***10.0***		***1.17***	***3.96***
60 min				
>1mm	3.5	0.09		
0.3 - 1	1.6	0.14		
<0.3	4.6	0.68		
Sub-total	***9.7***		***0.38***	***2.90***
TOTAL	**32.6**		**2.90**	

These results show:

- a gradual decrease in activity from the finest to the coarsest size fractions; and
- a decrease in activity as the duration of scrubbing increases.

The results are interpreted as reflecting the removal of contamination from the outer coatings of the rocks – with the majority of the contamination being associated with very fine material abraded from the rock surfaces. As the scrubbing duration increases there is less activity at the surface, so the abraded material decreases in contamination.

It is noteworthy that the original rocks were classed as Low Level Waste based on a surface analysis. All the abraded products are less than 40 Bq/g activity and could therefore be disposed as Very Low Radioactive Material. In addition, the cleaned rocks themselves are mostly free release.

DISCUSSION AND CONCLUSIONS

The results from laboratory and pilot scale tests have shown that:

For Soils:

- A radiologically "clean" bulk fraction (exempt from regulatory control in the UK) could only be produced where the contaminant levels in the original soil were only 2-3 times higher than this level;

- Soils with a higher contamination content could be remediated to:
  - a risk based clean-up target, or
  - a waste categorised as Very Low Radioactive Material. (A disposal route for this material is currently being considered as it is anticipated that it will be less costly than disposal of Low Level Waste).

- Laboratory work with specific gravity separation showed that low density peaty matter contained elevated levels of contamination. However, in the soil tested, its abundance was relatively low so that its removal from the relatively clean sands-sized fractions of the soil made little impact to the overall contamination content of the sands.

- Chemical leaching agents using mineral acids and complexants at low concentrations were generally ineffective in removing Cs-137. Only concentrated acids removed appreciable contamination.

- An estimate of the cost of treatment based on grain size separation and attrition scrubbing for a 5-10 tonne/h plant was found to be £80-£100 /tonne of soil treated. These costs include capital, labour, commissioning and decommissioning and analysis but, exclude the disposal of the contaminated fractions. In the cost estimate it is assumed that the soil would be treated in 1,000 m^3 batches and that at least 10 batches would be processed in the life of the plant.

For Rocks:

- A tumbling mill has been shown to very effective at removing surface contamination.

- The rocks tested were classified as LLW based on their surface activity. The resulting products from the scrubbing process are (i) scrubbed "clean" rocks which are mainly free release, and (ii) an abraded bulk material which is VLRM. Very little of the contamination was transferred into the process water. By processing contaminated rocks in this manner, significant cost savings could be made to the overall cost of disposal.

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