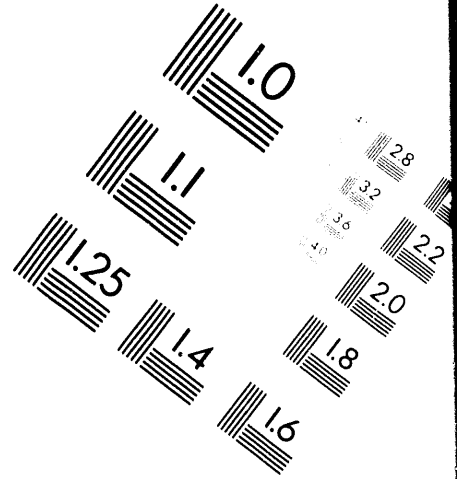
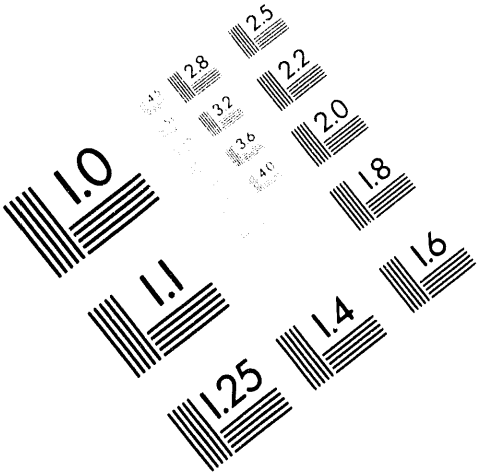




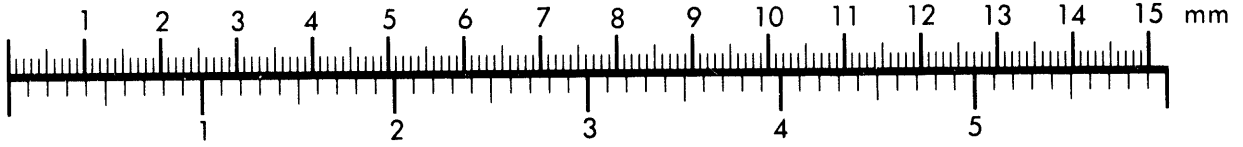
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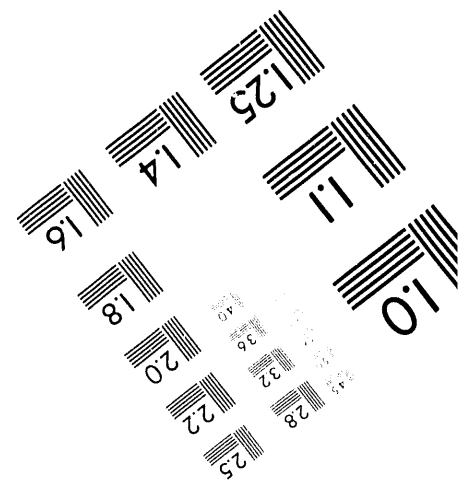
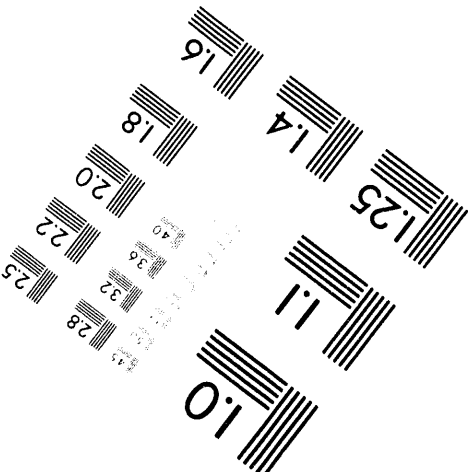
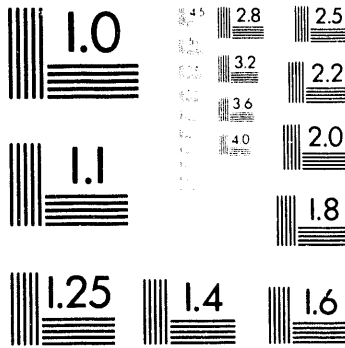
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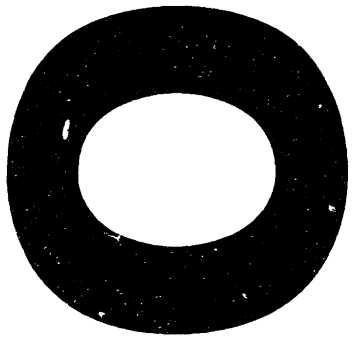
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100 Area Soil Washing: Bench-Scale Tests on 116-F-4 Pluto Crib Soil

Prepared for the U.S. Department of Energy
Office of Environmental Restoration and
Waste Management



Westinghouse
Hanford Company Richland, Washington

Hanford Operations and Engineering Contractor for the
U.S. Department of Energy under Contract DE-AC06-87RL10930

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5. Key Words treatability, CERCLA, autogenous grinding, chemical extraction, attrition scrubbing APPROVED FOR PUBLIC RELEASE <i>6/7/94 N. Sales</i>		6. Author Name: J. G. Field <i>J. G. Field 6/6/94</i> Signature Organization/Charge Code 85910/P71AA	
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ACRONYMS AND ABBREVIATIONS

ASTM	American Society for Testing Materials
BSE	back-scattered electrons
CB	centrifugal barrel
CEC	cation exchange capacity
CERCLA	<i>Comprehensive Environmental Response, Compensation, and Liability Act</i>
DOE-RL	U. S. Department of Energy, Richland Operations Office
DOT	U. S. Department of Transportation
EDE	effective dose equivalents
EPA	U. S. Environmental Protection Agency
FS	feasibility study
HP	horse power
ICP-MS	inductively-coupled plasma mass spectrometry
JCPDS	Joint Committee on Powder Diffraction Standards
MCL	maximum contaminant level
MDA	minimum detectable activity
NPL	National Priorities List
NTU	nephelometric turbidity units
OU	operable unit
PNL	Pacific Northwest Laboratory
RI	remedial investigations
RPM	revolutions per minute
SE	secondary electrons
SEM-EDS	scanning electron microscopy-energy dispersive spectrometry
TCLP	toxicity characteristic leaching procedure
TOC	total organic carbon
TPg	test performance goals
WHC	Westinghouse Hanford Company
XRD	x-ray diffraction
XRF	x-ray fluorescence

EXECUTIVE SUMMARY

The Pacific Northwest Laboratory conducted a bench-scale treatability study on a pluto crib soil sample from 100 Area of the Hanford Site. The objective of this study was to evaluate the use of physical separation (wet sieving), treatment processes (attrition scrubbing, and autogenous surface grinding), and chemical extraction methods as a means of separating radioactively-contaminated soil fractions from uncontaminated soil fractions. The soil washing treatability study was conducted on a soil sample from the 116-F-4 Pluto Crib that had been dug up as part of an excavation treatability study.

Particle size distribution data indicated that this soil was a poorly graded gravel in which the gravel fraction (4.75- to 75-mm) constituted about 76% of the soil mass. The soil also contained about 17% sand (0.074- to 4.75-mm), and minor amounts of cobbles (>75-mm) and silt-clay fractions (<0.074-mm).

The pH of the soil was relatively alkaline (8.84) and total organic carbon content was relatively low (0.25 %). The cation exchange capacity of the soil (<2-mm-sized fraction) was 19.58 meq/100 g, and the dominant exchangeable cation was Ca. Trace element analyses of this soil (<2 mm-sized fractions) showed no elevated concentrations above typically uncontaminated soil background levels. Total chromium (a potential inorganic contaminant) concentrations in this soil was very low (16 mg/kg). The toxicity characteristic leaching procedure (TCLP) extraction of this soil indicated that all regulated inorganic constituents (silver, arsenic, barium, cadmium, chromium, lead, and selenium) were well below the regulatory levels.

Data on the distribution of radionuclide in various size fractions indicated that the soil-washing tests should be focused on the gravel and sand fractions of the 116-F-4 soil. The radionuclide data also showed that ¹³⁷Cs was the only contaminant in this soil that exceeded the test performance goal (TPG). Therefore, the effectiveness of subsequent soil-washing tests for 116-F-4 soil was evaluated on the basis of activity attenuation of ¹³⁷Cs in the gravel- and sand-size fractions.

Two types of tests (physical and chemical) were conducted to reduce the activities of ¹³⁷Cs in the particle-size fractions of 116-F-4 soil. The physical tests consisted of attrition scrubbing (2- to 0.25-mm-sized fraction) and autogenous grinding of gravel fractions. Chemical extractions were also conducted on the sand fraction.

The results of autogenous surface grinding experiments using a centrifugal barrel processor showed that 94% to 97% of total ¹³⁷Cs activity in the gravel fractions could be removed if grinding was conducted in a water medium. The data indicated that grinding was less effective when conducted in an electrolyte medium. Following autogenous

surface grinding, the gravel fractions containing initial ^{137}Cs activities ranging from 186 to 391 pCi/g were found to contain an average residual activity of 19 pCi/g. This value is well below the TPG of 30 pCi/g for ^{137}Cs . The autogenous surface grinding data indicated that the bulk of the contaminant activity (about 74%) was located in the first millimeter of the gravel particle surface. The grinding data also showed that it is necessary to grind approximately a 3-mm surface layer of gravel particles to reduce the residual ^{137}Cs activity below the TPG. On average about 30% by weight of fines (<0.25-mm) were generated during the autogenous surface grinding experiments. The residual ^{137}Cs activity in the treated gravel fraction was functionally related to the quantity of fines generated.

It should also be noted that because of a limited number of experiments, factors that influence autogenous surface grinding such as consistency, uniformity of grinding, and energy requirements were not evaluated. These additional data may be needed to evaluate in detail the scale-up factors for conducting pilot or field-scale autogenous surface grinding.

Based on the data from previous attrition-scrubbing tests on 116-D-B soil from the 100 Area, optimized attrition scrubbing tests were conducted on the sand fraction (2- to 0.25-mm) of 116-F-4 soil. Two-stage and three-stage attrition scrubbing was conducted in the presence of an electrolyte at an optimum pulp density of about 79% and an energy input of 1.5 HP min/lb. The two-stage and the three-stage attrition scrubbing removed on average 50% and 60% of ^{137}Cs activity, respectively. The residual ^{137}Cs activities in scrubbed samples, ranging from 75 to 114 pCi/g, were well above the TPG for this radionuclide.

Chemical extraction experiments were also conducted on both untreated and attrition-scrubbed sand fractions from 116-F-4 soil. Previous extraction experiments indicated (DOE-RL 1993b) that a proprietary extractant (Extractant II) was the most effective of all extractants tested in removing substantial amounts of radionuclides including ^{137}Cs from Hanford soils. The chemical extraction data showed that one-quarter and one-half formal concentrations of Extractant II removed from 72% to 79% of the total ^{137}Cs activity from sand fractions resulting in residual activities that ranged from 52 to 77 pCi/g. Chemical extraction tests conducted on two-stage attrition scrubbed samples showed that the residual ^{137}Cs activity can be reduced to 27 pCi/g, a value below the TPG. These data indicated that a combination of two-stage scrubbing in electrolyte followed by chemical extraction can reduce initial ^{137}Cs activities of 210 to 260 pCi/g in sand fraction to below the TPG with concomitant generation of 2.3% contaminated fines (on bulk soil basis).

Preliminary flocculation tests were conducted on waste-water streams generated from wet sieving, two-stage attrition scrubbing with an electrolyte, and autogenous grinding. These tests showed that it was feasible to settle the suspended solids in these liquid wastes using commercially available flocculents. Using these flocculents on wash waters resulted in final turbidities that were very low (10 to 20 NTU). The activities of ^{60}Co and ^{152}Eu in all supernatants from treated or untreated waste solutions were both below minimum detectable activities (MDA) of 300 pCi/L. The activity of ^{137}Cs in all

supernatants (except in supernatant from attrition-scrubbing spent electrolyte) were well below the purgewater criterion of 2,000 pCi/L. Only, the supernatants from attrition-scrubbing with electrolyte contained soluble ^{137}Cs activities (23,400 pCi/L in untreated, and 5700 pCi/L in treated) that exceeded the purgewater limit of 2,000 pCi/L.

The data obtained from these treatability tests indicate that the soil from the 116-F-4 Pluto Crib has the potential to be soil washed by using autogenous surface grinding for gravel fractions and two-stage attrition scrubbing with electrolyte and chemical extraction of the sand fraction.

Based on this study, it appears that one of the two following options can be used if soil washing is considered as the appropriate remediation technique for the 116-F-4 soil:

- The first processing option involves treating only the water-washed gravel-size (>13.5-mm) material (about 81% of the soil mass) by autogenous surface grinding in a water medium. Using this treatment option, about 55% of the clean soil with residual ^{137}Cs activity of about 19 pCi/g can be recovered as backfill material. This treatment option involves disposing of about 21% of the untreated soil (<13.5-mm fraction) with about 24% of contaminated fines (<0.25-mm) generated during autogenous surface grinding.
- The second option includes the treatment of both sand and gravel fractions. The gravel fractions are treated by autogenous surface grinding in a water medium, and the sand fraction (2- to 0.25-mm) is treated with two-stage attrition scrubbing with an electrolyte followed by chemical extraction. These soil-washing treatments generate nearly 55% treated gravel fraction and about 11% treated sand fraction as backfill material. The recovered soil material (about 66% of the soil mass) would contain an average ^{137}Cs activity of about 20 pCi/g. This treatment option results in disposal of about 34% of the soil mass (8% untreated soils and approximately 26% contaminated fines generated during the treatment processes).

Based on process knowledge, this and the previous soil-washing studies (DOE-RL 1993b) showed that choosing appropriate soil-washing schemes may result in appreciable volume reduction of material for disposal. The cost effectiveness of the technology and issues relating to the secondary waste streams generated in the treatment process will require further evaluation.

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1.0 INTRODUCTION

The 100 Area of the Hanford Site contains nine inactive nuclear reactors that were operated for the production of fissionable material (Figure 1-1). All these water-cooled reactors (B, C, D, DR, F, H, KE, KW, and N) situated along the southern bank of the Columbia River have been shut down and currently are being evaluated for decommissioning. Waste streams that were generated during the operation of these reactors were disposed of in the vicinity, resulting in wide-spread contamination of both soil and groundwater. The bulk of the contamination originated from the disposal of very large volumes of reactor cooling water containing both fission and activation products.

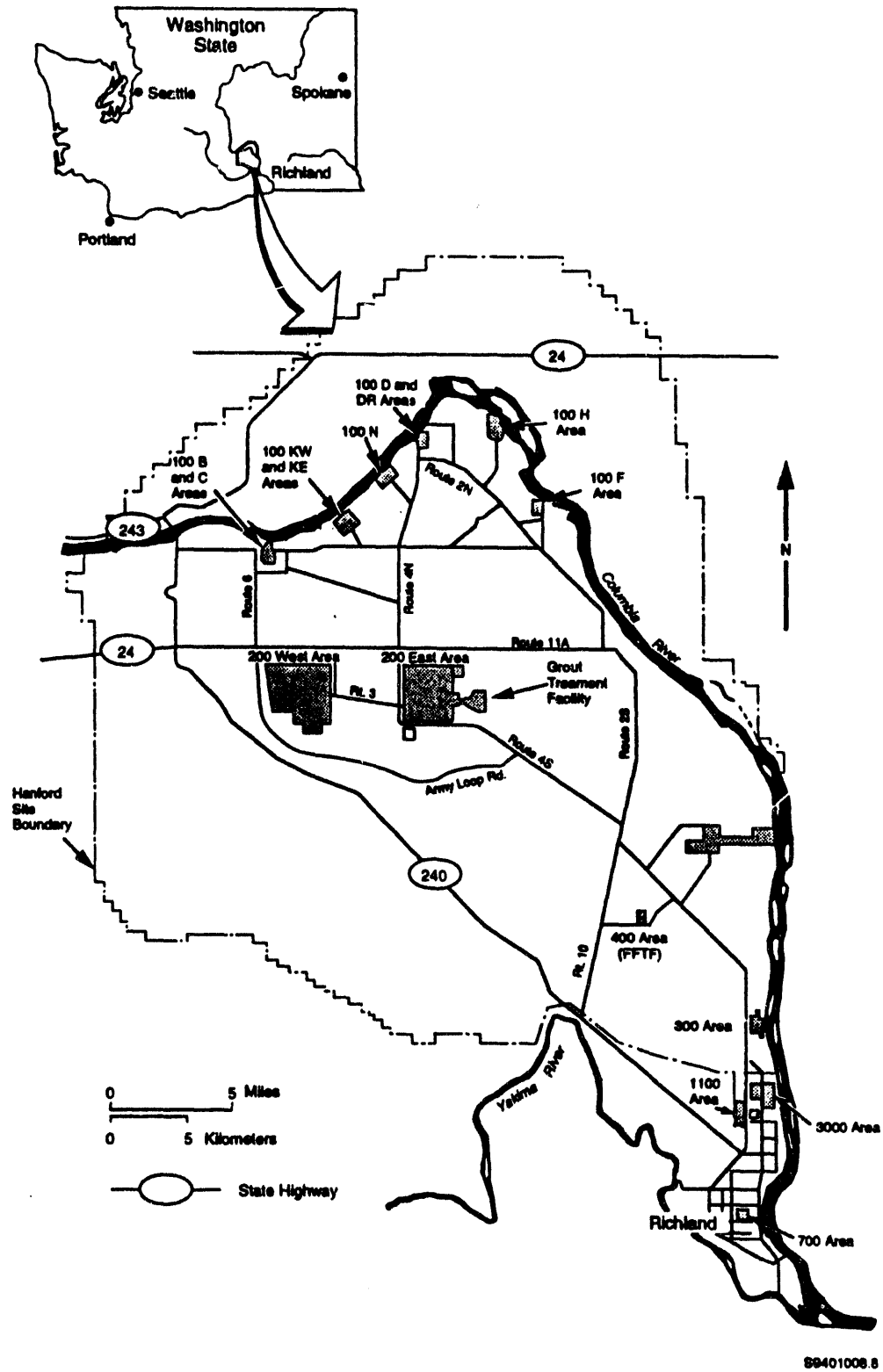
The contaminated cooling water was discharged into engineered trenches and cribs that resulted in substantial volumes of soil being contaminated with radionuclides. Additionally, leaks in the cooling water transfer systems also contaminated the soil and groundwater. The extent of the contamination resulted in the inclusion of the 100 Areas of the Hanford Site in the National Priorities List (NPL) compiled under the *Comprehensive Environmental Response, Compensation, and Liability Act* (CERCLA) by the U.S. Environmental Protection Agency (EPA).

The types and the extent of environmental contamination in the 100 Areas have been investigated as part of remedial investigation and feasibility studies (RI/FS) (DOE-RL 1992a). Based on these data, several remedial measures have been examined and documented in a report (DOE-RL 1992d). Because of the large volumes of contaminated soils that need remediation, treatment technologies that reduce the volume of soil to be disposed of as waste may offer significant savings in remedial costs. One of the more promising remedial measures is soil washing. Therefore, soil-washing treatability studies were conducted on the 100 Area soils to derive specific data for evaluating the applicability of this technology.

A test plan was developed (DOE-RL 1992b) to conduct laboratory-scale tests on two representative 100 Area soils contaminated during cooling water disposal. These laboratory-scale bench tests were designed to fulfill the initial screening phase of an overall three-phase treatability study. The objective of this treatability study was to evaluate the use of physical separation systems and chemical extraction methods as a means of separating chemically and radioactively contaminated soil fractions from uncontaminated soil fractions.

The laboratory-scale screening tests to be performed on 100 Area soils were described as a set of procedures (Freeman et al. 1993). These procedures include physical, chemical, and mineralogical characterization of the soils; soil fractionation through wet-sieving to characterize particle mass and contaminant distribution; contaminant mobilization through physical (attrition scrubbing and autogenous grinding) and chemical treatments (extraction and leaching); and the treatment of wash water and spent extractants. A report on the treatability of these two soils was recently completed (DOE-RL 1993b). A soil-washing treatability study was also conducted on a third soil sample from the 100-F Area (from a pluto crib), and the results obtained are the subject of this report.

Figure 1-1. The Location of the Hanford Site.



2.0 SOIL-WASHING TECHNOLOGY

2.1 DESCRIPTION OF THE TECHNOLOGY

Soil washing is a remediation technology aimed at removing inorganic, organic, and radionuclide contaminants to the extent that the residual levels of these contaminants in coarse fractions (the bulk) of the treated soil are below selected safe levels. The remediated coarse soil is returned to the excavation site, and the smaller volume of soil containing the contaminants is disposed. Typically, soil washing is accomplished with physical or chemical techniques, or a combination of both techniques.

The effectiveness of soil washing as a remediation technique depends mainly on the type of binding processes that exists between the contaminants and the soil particles. Contaminants in soils may be found adsorbed or precipitated on the soil mineral surfaces or as distinct particles. If a contaminant exists mainly in the adsorbed form, the finer soil particle fractions tend to contain higher concentrations of that contaminant. This phenomenon can be attributed to the fact that finer soil particles possess greater surface areas per unit mass than the coarser sized fractions. If the contaminants exist mainly as coatings or precipitates on soil particles, washing with water and physical agitation through attrition, may dislodge and separate the contaminated fine material, or the precipitates may be dissolved by using chemical extractants. Contaminants that exist as distinct particles can also be separated if they possess significantly different specific gravities than the bulk minerals in a soil matrix.

Typically, the initial step in the soil-washing process consists of separating the soils into different size fractions. This is generally accomplished by using hydraulic classification techniques such as screening or hydrocyclones. If necessary, hydraulic jigs may also be used to separate contaminant-bearing particles based on their differences in size and specific gravity. Depending on the soil and the types of contaminants, this initial physical separation process may accomplish the goal of recovering clean soil that constitutes the major fraction of the soil mass and isolating the major fractions of the contaminants in a smaller volume of fine-grained soil that can be removed for disposal.

If the contaminant distribution among various particle fractions of a soil indicates that the levels of contamination in the coarser soil fractions exceed designated safe levels, additional physical and/or chemical treatments may be necessary to release the surface-bound contamination. The principal physical method used to release the contaminants, through grinding action, is attrition scrubbing. In this method, the moist soil particles are agitated vigorously and made to attrit against each other to dislodge the surface-bound contaminants. Such scrubbing will generate contaminant-laden fine material that is washed out of coarser material containing significantly lower concentrations of contaminants. Attrition scrubbing may also be conducted with surfactants or electrolytes to attenuate the re-adsorption of released contaminants onto the freshly exposed surfaces of the soil particles. Typically, the electrolytes may either be acidic or alkaline solutions that enhance the solubility of contaminants and may also contain ligands that chelate mobilized cationic contaminants.

Additionally, the electrolytes may include ionic components that reduce re-adsorption of released contaminants by blocking the adsorption sites on soil particle surfaces. Generally, attrition scrubbing with a suitable electrolyte increases contaminant removal through a synergistic combination of physical and chemical processes. Chemical treatments used to remove contaminants from coarse soil fractions typically consist of an extractive process that uses solvents (aqueous acids, alkalis, organic solvents) to desorb and dissolve the contaminant-bearing particles and coatings from the soils.

2.2 FEASIBILITY OF SOIL-WASHING RADIONUCLIDE-CONTAMINATED SOILS

A literature review indicated that a number of bench-scale tests have been conducted to assess the feasibility of using soil washing as a technique to remediate soils contaminated with various radionuclides (Table 2-1). A number of these studies were focused on the feasibility of soil washing to remediate soils contaminated with $^{238/239}\text{Pu}$ and ^{241}Am . The effects of scrubbing action on soil washing were studied by Horton and Albenesius (1976). They conducted tests on samples of plutonium-contaminated soils from the Savannah River Laboratory site in South Carolina by agitating these soils in a flow-through container system. Their data indicated that increasing agitation time resulted in decreasing levels of plutonium in the sand fraction with concomitant increase in the amount of contaminated silt-clay fraction.

Table 2-1. Soil-Washing Feasibility Tests of Radionuclide-Contaminated Soils.

Soil Source	Contaminant Radionuclide	References
Savannah River site, South Carolina	^{238}Pu	Horten and Albenesius (1976)
Rocky Flats site, Colorado	^{238}Pu , ^{241}Am	Navratil and Kochen (1982) Stevens and Rutherford (1982)
Rocky Flats, Colorado; Hanford Site, Washington; Mound, Ohio; Idaho Falls, Idaho; Los Alamos, New Mexico	$^{238,239}\text{Pu}$, ^{241}Am	Stevens et al. (1982)
Montclair/Glen Ridge, New Jersey	^{226}Ra , ^{230}Th	Richardson et al. (1989) Phillips et al. (1993)
Idaho Falls, Idaho	^{60}Co , ^{137}Cs	Gombert (1992); Murray (1993)
Fernald, Ohio	U	Soil Decon Task Group (1993)
Hanford site, Washington	^{235}U , ^{238}U	Serne et al. (1992); DOE-RL (1994a)
Hanford Site, Washington	^{60}Co , ^{137}Cs , ^{152}Eu	DOE-RL (1993b)

Navratil and Kochen (1982) investigated the effectiveness of wet and dry screening, attrition scrubbing, and chemical extraction for releasing ^{238}Pu and ^{241}Am from contaminated soils (total activity: 900 to 140,000 pCi/g) from the Rocky Flats Plant site in Colorado. The results showed that dry screening was not as effective as wet screening for isolating the radionuclides in the fine fraction. Using wet screening, about 65% by mass of the soil (>2.4-mm in size) containing low activities of these radionuclides (<6 pCi/g of Pu and <3 pCi/g of Am) was recovered. Additionally, using attrition scrubbing of the 2- to 0.42-mm soil fraction, the activity of Pu in this fraction was reduced from 300 pCi/g to <20 pCi/g. Among the chemical extractants tested, Calgon, oxalic acid, and a proprietary detergent removed major fractions (98% to 99%) of plutonium and americium from the 4- to 0.42-mm-sized fraction. Another study was conducted by Stevens and Rutherford (1982) on soils from the same site. They examined the effects of more intense agitation on chemical extractability of these radionuclides. The data indicated that the extractability of the contaminants was significantly increased by a combination of intense agitation and dissolution.

The effects of repetitive chemical extraction on the mobilization of plutonium and americium from contaminated soils from five different sites were investigated by Stevens et al. (1982). In this study, soils were successively extracted for a total of five times with three different extractants (sodium hydroxide, 2N hydrochloric acid, and a mixed extractant consisting of 2% nitric acid, 0.2% hydrofluoric acid, 2% pine oil, and 5% Calgon solution). The data showed that these three extractants differed in their effectiveness for removing the contaminants from each type of soil. For instance, hydrochloric acid was the most effective extractant for releasing the contaminants from the soils sampled from sites at Hanford, Idaho, and Mound, whereas the sodium hydroxide was the most effective extractant for the Rocky Flats soil. The mixed extractant was most effective for releasing plutonium and americium for the Los Alamos soil.

A bench-scale soil-washing study consisting of wet sieving was conducted by Richardson et al. (1989) on soils contaminated with radium and thorium from a site in Montclair/Glen Ridge, New Jersey. The results of these tests indicated that up to 56% of the mass (>74 μm in size) of soil could be separated with residual activity of ≤ 16 pCi/g. A recent pilot-scale study was conducted on the same soil by Phillips et al. (1993). The data from this test showed that at least 54% of the soil mass (>74 μm) containing residual activity of <12 pCi/g could be recovered as remediated material and returned to the excavation site.

Recently, the extractability of ^{60}Co and ^{137}Cs from contaminated soil from a waste pond in Idaho Falls, Idaho, was tested by Gombert (1992). This study consisted of using a sequence of different chemical extractants to assess the release of these two radionuclides from the finer fraction (<0.425 mm) of the contaminated soil. The results indicated that up to 84% of cobalt could be extracted from the soil. By contrast, only a minor fraction (about 20%) of cesium was extractable by this procedure. The same waste pond soils were used in a more extensive bench-scale soil-washing feasibility study conducted by Murray (1993). This test included a number of soil-washing unit operations such as wet sieving, attrition scrubbing, froth flotation, and chemical extraction. The effects of a number of parameters related to these processes (temperature, pulp density, retention time, the types and

concentrations of extractants) were also tested. The data from these tests showed that using a soil-washing system consisting of wet sieving, froth flotation, multiple-stage attrition scrubbing, and chemical extraction resulted in recovering about 92% of the soil mass with a residual cesium activity below the specified activity level of 690 pCi/g.

A set of soil-washing studies was conducted by the Soil Decon Task Group (1993) to check the feasibility of removing uranium from two contaminated soils from Fernald, Ohio. The data indicated that a number of chemical extractants such as carbonate solutions, citric acid, Tiron with sodium dithionite solutions, and a mixture of sodium salts of citrate, bicarbonate, and dithionite effectively removed substantial amounts of uranium from these soils. Attrition scrubbing with chemical extractants (sodium and ammonium carbonate, citric and sulfuric acids) also released significant amounts of uranium from these soils.

Results of laboratory- and pilot-scale tests conducted on uranium-contaminated soils from the 300 Area of the Hanford Site (Serne et al., 1992; DOE-RL, 1994a) showed that wet sieving and attrition scrubbing treatments were effective in separating soil fractions with lower ^{235}U and ^{238}U activities from the contaminated soil mass. These soil-washing tests showed that it was possible to recover 85% of the soil mass (>0.425-mm) containing ^{235}U and ^{238}U activities of <5 and <50 pCi/g respectively.

The feasibility of removing three contaminant radionuclides (^{60}Co , ^{137}Cs , and ^{152}Eu) from soils from the 100 Area of Hanford Site was examined recently in a series of bench-scale tests (DOE-RL 1993b). One of the soil samples from a contaminated trench (116-D-1B) contained ^{60}Co , ^{137}Cs , and ^{152}Eu activities of 6, 104, and 84 pCi/g respectively. Wet-sieving data showed that >2-mm size fraction (47% of the mass) contained low levels of radionuclides (1, 11 and 2 pCi/g of ^{60}Co , ^{137}Cs , and ^{152}Eu respectively) therefore needed no additional treatment. The data also showed that additional volume reductions could be achieved if radionuclide activity levels in the 2- to 0.25-mm size fraction were reduced through either physical or chemical treatments.

Attrition scrubbing tests with and without an electrolyte (consisting of ammonium citrate and citric acid) indicated that the removal of radionuclides was highest when two-stage scrubbing was conducted with an electrolyte present in both stages. During this two-stage process, >79%, 48%, and 94% of ^{60}Co , ^{137}Cs , and ^{152}Eu activity was removed, and about 14% by weight fines was generated. Chemical extraction tests on 2- to 0.25-mm size fraction conducted with a proprietary extractant showed that major fractions of all three radionuclides were consistently removed from this soil fraction (average of >90%, 85%, and >99%, of ^{60}Co , ^{137}Cs , and ^{152}Eu , respectively) resulting in very low residual activities.

These data showed that soil from the trench (116-D-1B) can be effectively ameliorated by fractionating the soil by using wet-sieving and treating the 2- to 0.25-mm size fraction using either two-stage attrition scrubbing with an electrolyte or chemical extraction with a proprietary extractant. For this soil, it was estimated that using one of these two treatments would result on average about 85% volume reduction with residual activities of <1, <4 and <30 pCi/g for ^{60}Co , ^{137}Cs , and ^{152}Eu respectively.

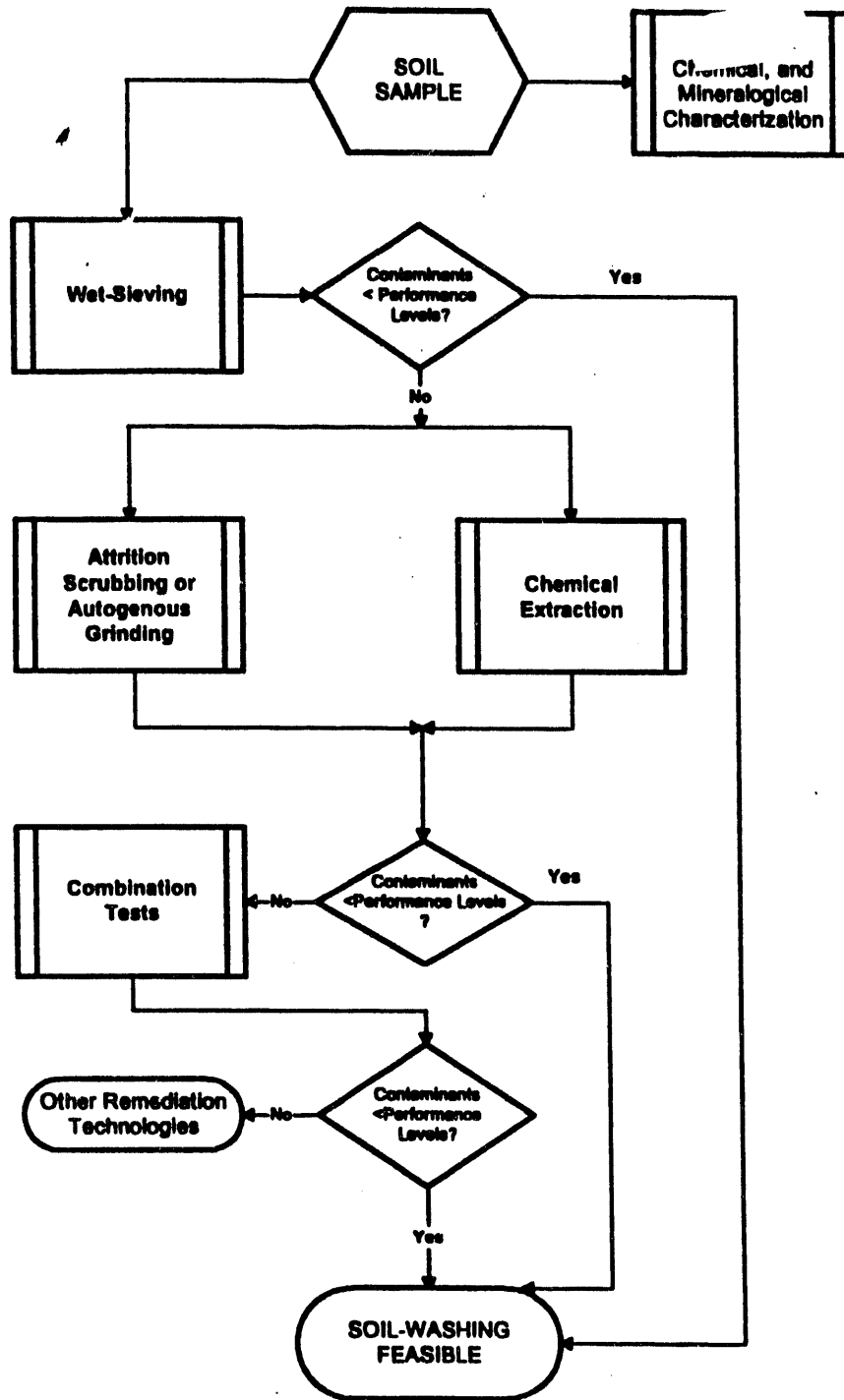
2.3 TEST PERFORMANCE GOALS FOR RADIONUCLIDES

The test plan for 100 Area soil-washing feasibility tests (DOE-RL 1992b) included a set of target performance levels (TPL) for various radionuclides. According to these criteria, treated soils were to be considered clean if the residual activities of radionuclides were found to be below these specified levels. Recently, based on effective dose equivalents (EDE) of 10 mrem/yr, a revised set of activity values were established. (See Table 6-2 in WHC, 1994). According to this manual, these values were established to assure that the EDE do not exceed the specified limits for any reasonable scenario (i.e., direct exposure, ingestion, inhalation, and consumption of food crops and animal products). These new conservative limits for radionuclides established from a pathway programming basis are listed in Table 3-1. The set of values specified for accessible soils in 100 Area of Hanford Site were used as the test performance goals (TPG) for these treatability tests. The TPG values therefore were used as a guide to assess the level of soil contamination and the effectiveness of soil-washing processes.

2.4 A SOIL-WASHING TEST SCHEME FOR 100 AREA SOILS

A review of published data from a number of laboratory-scale soil-washing tests on radionuclide-contaminated soils indicated that a test scheme for soil washing could be developed by combining a number of unit operations. Therefore, a scheme was formulated for testing the feasibility of soil washing 100 Area waste trench soils (Figure 2-1). This scheme included, as an initial step, physical, chemical, and mineralogical characterization. These characterization data are essential for selecting relevant treatment methods for each soil. The data regarding the mass distribution of particle sizes and the activities of contaminant radionuclides in these soil fractions were obtained from wet-sieving data. If the residual radionuclide activities of a substantial portion of the bulk soil were found to be less than the specified test performance goals (TPG), then wet sieving alone could be used to remediate that soil. By contrast, if the wet sieving data indicate that the coarser-sized fractions contain residual activities that exceed the TPG, additional physical (attrition scrubbing or autogenous grinding) and chemical (extraction or leaching) treatment tests were conducted. Remediation would be feasible if these treatments reduced the radionuclide activities in coarser fractions to below the TPG. If this goal was not accomplished, additional tests that combine both physical and chemical treatments were conducted. These combination tests typically included multi-stage attrition scrubbing or autogenous grinding with electrolytes or chemical extractants. Soil washing would be considered effective for a soil if the residual radionuclide activities in the treated coarser soil fractions were found to be less than the TPG and the percentage of clean coarse material represented a significant fraction of the original soil.

Figure 2-1. A Scheme for Soil-Washing Feasibility Tests on 100 Area Soils.



3.0 SOIL SAMPLES FROM THE 100 AREA

3.1 SITE DESCRIPTION

The 100 Area of the Hanford Site is located in a structural basin (Pasco basin) on the Columbia Plateau. A generalized geologic cross section through the 100-B/C Area (Figure 3-1) shows a sequence of unconsolidated surficial sedimentary deposits (Hanford formation). The topmost layer of this formation consists of a thin layer (1 to 15 ft thick) of light brown, fine, slightly silty, eolian deposits.

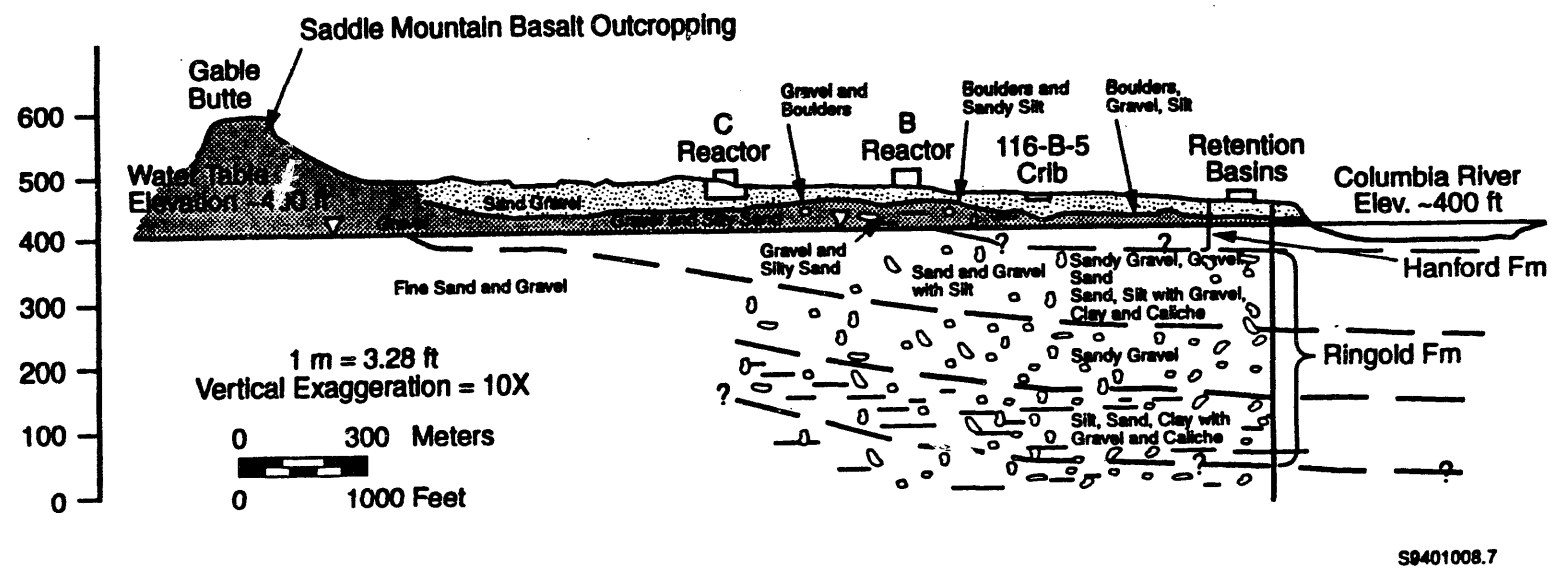
The principal sediments of the Hanford formation underlying the surficial layer range from twenty to several hundred feet in thickness and consist of poorly sorted, unconsolidated glaciofluvial material (Pasco gravels). The origin of these sedimentary deposits is believed to be the periodic cataclysmic floods that occurred during the late Pleistocene epoch (Baker 1981; Mullineaux et al. 1977). The Pasco gravels are variable mixtures of particle sizes that range from boulders to silt. The bulk of Pasco gravels are very coarse textured and are classified as silty-sandy gravels typically consisting of about 50% gravel, 40% sand, and 10% silt (DOE-RL 1988). Mineralogically, these deposits consist of quartz, feldspars, and ferromagnesian minerals (DOE-RL 1992c). All the trenches and cribs that were engineered for waste disposal in the 100 Area were located in the Pasco gravels.

The Ringold Formation, ranging up to several hundred feet in thickness, underlies the Hanford formation. This formation, late Miocene to late Pleistocene in age, consists of stratified deposits of material ranging in size from gravel to clay. The uppermost unconfined aquifer system in the Hanford area, ranging up to 300 ft in depth, is located within the Hanford and Ringold Formations. The Saddle Mountain Basalt Formation underlies the stratified Ringold deposits.

3.2 TEST SITE

The soil sample for soil-washing feasibility tests was obtained from the material excavated from the 116-F-4 Pluto Crib located in the 100-FR-1 Operable Unit (Figure 3-2). The 116-F-4 Pluto Crib consisted of a cobble-filled 55-gal steel container with sealed top and open bottom resting on one end of an 6-ft by 6-ft by 6-ft zone of cobble-size aggregate (DOE-RL, 1994b). This crib was one of the typical liquid disposal structures constructed below the ground surface with open bottoms (Figure 3-3). A distribution pipe discharged waste water into the rockfill to generate uniform flow over the crib bottom. The top of cribs were backfilled with soil to provide protection from radiation. This structure received water from process tubes in the 100-F Reactor that had failed fuel claddings. This crib was used only for a short time (1952 to 1954) and was contaminated with activation and fission products. Based on the soil sampling conducted in 1976, the total volume of contaminated soil in this trench was estimated by Dorian and Richards (1978) to be about 18,000 ft³ (30 by 30 by 20 ft). The average activities of radionuclides and an inventory of total activity for the 116-F-4 Pluto Crib (Dorian and Richards 1978) are listed in Table 3-1. These data indicate

Figure 3-1. Generalized Geologic Cross Section at 100-B/C Area, Hanford.



3-2

Figure 3-2. Location of the 116-F-4 Pluto Crib in the 100-FR-1 Operable Unit, Hanford.

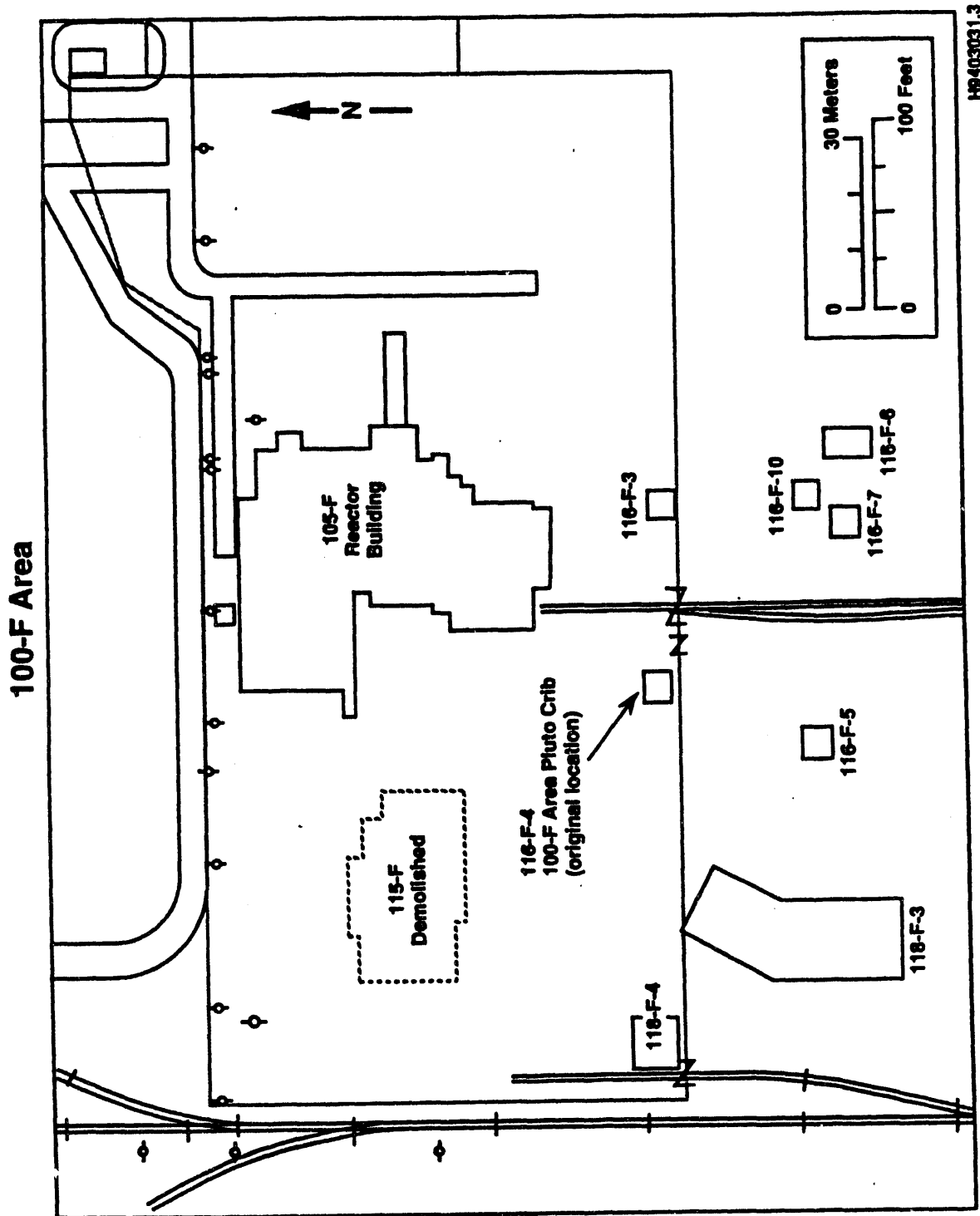
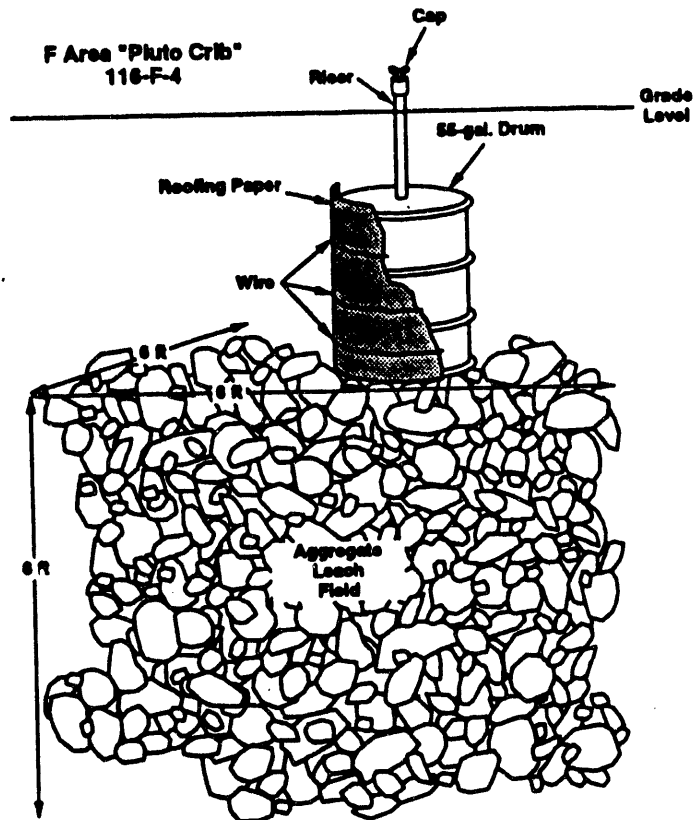
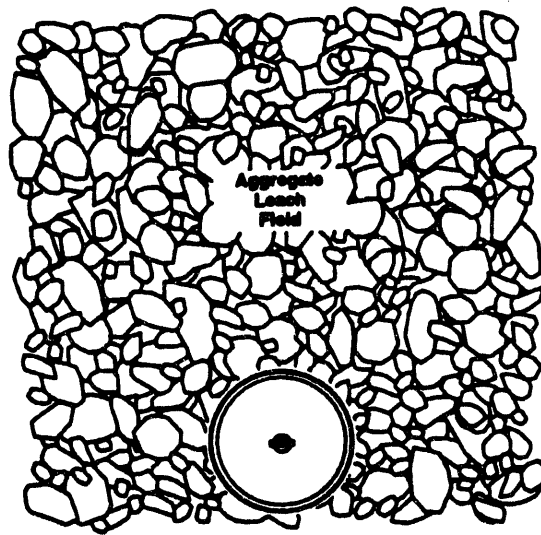


Figure 3-3. Pluto Crib (116-F-4)



Crib Design



Plan View

NS012028.2a

the soil excavated from this Pluto Crib may contain activity levels of ¹³⁴Cs, ¹³⁷Cs, and ¹⁵⁴Eu, that exceed the proposed TPG.

Table 3-1. Average Activities of Radionuclides in Soils from 116-F-4 Pluto Crib.

Radionuclide	116-F-4 Average (pCi/g)	Test Performance Goals (pCi/g)
⁶⁰ Co	0.47	7.1
¹³⁴ Cs	21	10
¹³⁷ Cs	1600	30
¹⁵² Eu	8.9	15
¹⁵⁴ Eu	40	14
¹⁵⁵ Eu	94	630
⁹⁰ Sr	1100	2800
²³⁸ U	1.3 ^a	370
^{239/240} Pu	35	190
Contaminated Soil Volume (ft ³)	18000	--
Contaminated Soil Mass (g)	1.2 x 10 ⁹	--
Total Activity (Ci)	3.5	--

Source: Dorian and Richards (1978).

^aMeasured as total U.

3.3 SOIL SAMPLE COLLECTION

About 4500 yd³ of soil including the material from the 116-F-4 Pluto Crib was excavated as part of the Excavation Treatability Tests (DOE-RL 1994b). About 500 yd³ of this material was classified as contaminated soil and stored in a modular storage unit. During excavation, the soils were treated with dust suppressants such as XDCA (a beet polysaccharide) and lignosulfate (a paper manufacturing byproduct). The contaminated soil samples were collected from the modular storage unit by WHC personnel and transported to the laboratory in Department of Transportation (DOT)-specified containers. A total volume of 40 gal of soil was collected and transported to the laboratory in eight 5-gal containers.

3.4 SOIL SAMPLE PREPARATION

The soil samples received in 5-gal-capacity containers were spread on drying trays, placed in fume hoods and, as recommended by the ASTM standard practice (D 421-85), thoroughly air dried. All air-dried samples were friable, and therefore easily disaggregated. Following disaggregation, each soil sample was dry-screened using a 13.5-mm screen to isolate gravel-sized material and a 2-mm screen to separate material finer than coarse sand.

Subsamples of air-dried <2-mm material were obtained by homogenizing, coning, and quartering the soil (ASTM C 702-87). Because of the large volume of the sample, subsampling was accomplished by compositing air-dried and dry-screened soil sample into four equal batches. Next, each of these four batches was coned and quartered, and one randomly selected quarter from each of the four batches was composited into a single batch. From this single batch, representative subsamples were drawn for subsequent work. All the remaining soil was combined and stored in plastic-lined 5-gal containers. If this single batch was used up during testing, additional batches were prepared by compositing the remaining soil into an appropriate number of batches and repeating the process of coning and quartering until a new single batch of soil was obtained.

4.0 CHARACTERIZATION OF SOILS

The principal objectives of soil characterization were to determine the properties (physical, chemical, and mineralogical) that govern the contaminant partitioning and release behavior of soils during the washing process. Typically, all the characterization tests can be conducted on <2-mm size fractions. However, there are no test protocols to measure characteristics such as total organic carbon (TOC), pH, cation exchange capacity (CEC), and toxicity characteristics leaching procedure (TCLP) of gravel and cobble fractions. Only a limited number of tests such as the particle size distribution, specific gravity, and radionuclide analysis can be conducted on very coarse (gravel and cobble) soil fractions. Therefore, this study included a complete suite of specified characterization tests for <2-mm size soil fractions, and only a limited number of applicable tests for the gravel fractions of 116-F-4 soil. The characterization tests were conducted according to the methods outlined in the test plan (DOE-RL 1992b) with exceptions noted in Appendix A.

4.1 PHYSICAL CHARACTERIZATION

4.1.1 Moisture Content

Gravimetric water contents of the soil samples were determined using a standard procedure (Gardner 1986). Air-dried soil samples (<2 mm) in tared containers were oven-dried at 105 ± 5 °C for 10 to 24 hr, cooled over a desiccant, and weighed. The gravimetric water content was computed as percentage change in soil weight before and after oven drying.

4.1.2 Particle Size Distribution

The particle size of 116-F-4 soil sample was determined according to ASTM method D 422-63. According to this method, the distribution of particle sizes larger than 2 mm (retained on No. 10 sieve) is determined by dry-sieving. Soil fractions finer than 2 mm were dispersed, and the distribution of particles smaller than 0.074 mm was determined by measuring the sedimentation rate using a hydrometer. Following the hydrometer measurements, the soil sample was washed through a 0.074-mm (No. 200) sieve and dried at 110 ± 5 °C, and material larger than 0.074 mm was dry-sieved through a set of sieves (Numbers 20, 40, 60, and 140). The weight percentage of soil finer than each specified size fraction was tabulated.

4.1.3 Specific Gravity

The specific gravity of soil samples was determined according to the ASTM standard test methods. For soil particle fractions larger than 4.75 mm, ASTM method C 127-88 was used. The specific gravity of soil fractions finer than 4.75 mm was measured by ASTM method D 854-83. The specific gravity value of the whole sample was computed as the

weighted average of both soil fractions, as specified in ASTM D 854-83. Additionally, specific gravities of particles smaller than 2 mm were determined and used in calculating particle sizes by the hydrometer method (ASTM D 422-63).

4.2 CHEMICAL CHARACTERIZATION

4.2.1 Total Organic Carbon

The total organic carbon (TOC) content of the soil sample was measured by the coulometric method (ASTM D 4129-88). In this method, soil-bound carbon is mobilized as carbon dioxide through combustion and acidification. The released carbon dioxide is absorbed into ethanolamine and measured by coulometric titration. The TOC value was calculated as percentage of the mass of soil.

4.2.2 Soil pH

The pH of <2-mm sized fraction of the 116-F-4 soil was determined by equilibrating the soil sample with deionized distilled water (1:1) for 10 min and decanting, and then measuring the pH of the supernatant with a calibrated glass electrode.

4.2.3 Cation Exchange Capacity

Cation exchange capacity of the soil was determined according to the ammonium acetate method (Thomas 1986). According to this method, the exchangeable cations were displaced by equilibrating about 5 g of soil with 25 mL of 1N ammonium acetate solution for 30 min. After centrifuging the soil suspension, the supernatant was decanted and saved. The equilibration was repeated a second time, and the displaced supernatant was composited with the supernatant from the first equilibration. Finally, the combined supernatant was analyzed for the exchangeable cation content (Ba, Ca, Mg, Sr, and Na) by inductively coupled plasma-mass spectrometric analyses (ICP-MS) (PNL-ALO-280). Cation exchange capacity was calculated as the milliequivalent sum of all exchangeable cations per 100 g of oven-dry soil.

4.2.4 Total Elemental Analyses

The total elemental composition of the soil sample (<2-mm-sized fraction) was measured in duplicate by x-ray fluorescence spectrometry (PNL 7-40-48, Rev. 1). This method uses an iron target for analyzing Al, Ca, K, and Si; a Zr target for measuring As, Co, Cr, Cu, Fe, Pb, Mn, Ni, Rb, Se, Sr, Ti, and Zn; a Ag target for analyzing U; and a Gd target to determine Ag, Ba, Cd, Sb, and Zr.

4.2.5 Radionuclide Analyses

The radionuclide content of the soil sample (<2-mm-sized fraction) was determined by specified standard procedures. Gamma-ray spectrometry (PNL-ALO-106; PNL-ALO-464) was used to determine the activities of ^{60}Co , ^{134}Cs , ^{137}Cs , ^{152}Eu , ^{154}Eu , and ^{155}Eu . Analyses of ^{90}Sr were conducted according to standard wet chemical separation and beta counting methods (PNL-ALO-106; PNL-ALO-463; PNL-ALO-465). The activities of Pu isotopes $^{239/240}\text{Pu}$ were measured by acid digesting the samples (PNL-ALO-106), separating Pu (PNL-ALO-417; PNL-ALO-466), electroplating Pu onto counting disks (PNL-ALO-468; PNL-ALO-496), and counting by alpha spectrometry (PNL-ALO-469). Uranium activities in these samples were determined by fusing and dissolving the soil samples (PNL 7-40-78), and then analyzing the solutions by ICP-MS (PNL-ALO-280; PNL-ALO-282).

4.2.6 Toxicity Characteristic Leaching Procedure (TCLP)

The TCLP test on 2-mm-sized fraction of the 116-F-4 soil was conducted according to Method 1311 (EPA 1990). The extract was analyzed for seven regulated metals (arsenic, barium, cadmium, chromium, lead, silver, and selenium) by ICP-MS (PNL-ALO-280).

4.2.7 Sequential Extraction

Sequential extractions of soils were conducted to gain some understanding of contaminant binding mechanisms with operationally defined groups of mineral forms in soils. The method (Belzile et al. 1989) consists of extracting soils sequentially with increasingly strong extractants; the fractions solubilized are characterized as "exchangeable," "carbonate-bound," "Mn-oxide bound," "Fe-oxide bound," "organic matter and sulfide bound," and "residual mineral bound." In this method, these extractants are used sequentially: (1) 1N magnesium chloride solution (pH 7) equilibrated with soil for 30 min to displace the exchangeable fraction, (2) 1M sodium acetate solution (adjusted to pH 5 with acetic acid) contacted with soil for 5 hours at room temperature to dissolve "carbonate-bound" fraction, (3) a solution of 0.1M hydroxylamine-hydrochloride and 0.1M nitric acid equilibrated with soil for 30 min at room temperature to dissolve "Mn-oxide bound" fraction, (4) a mixture of 0.04M hydroxylamine hydrochloride and 25% (v/v) acetic acid heated with soil for 6 hr at 96 °C to mobilize "Fe-oxide bound" fraction, and (5) digestion of the soil for 5 hr at 85 °C with 30% hydrogen peroxide (acidified to pH 2 with nitric acid) followed by a room temperature extraction with a solution of 3.2M ammonium acetate and 20% (v/v) nitric acid to release "organic-bound" or "sulfide-bound" fractions. These extraction steps were expected to provide information on specific affinities of contaminants for different types of mineral surfaces and matrices. The extract obtained from each step was counted for the activities of the contaminant radionuclide (^{137}Cs).

4.3 MINERALOGICAL CHARACTERIZATION

Radionuclides in soil can exist in several forms such as exchangeable, specifically adsorbed, surface-precipitated, and as part of substrate mineral structure. The ease of removal of a contaminant depends on the type of association between the contaminant and the mineralogical substrates. Sequential extraction techniques provide some information regarding the types of minerals that may exist in soils; however, these techniques do not directly identify specific minerals and their association with different contaminants.

Mineralogy of the soil was determined by using two different techniques. X-ray diffraction analyses (XRD) was used to determine the structural identity, and scanning electron microscopy with energy dispersive spectrometry (SEM-EDS) analyses was used to obtain chemical and morphological data on minerals present in the sand fraction (2 to 0.25 mm) of the soil. Minerals in the clay fractions ($<2 \mu\text{m}$) were identified by XRD.

The XRD of the sand fraction (2 to 0.25 mm) of the soil was conducted by grinding the sample into silt-sized material, packing this material into aluminum sample holders to obtain randomly oriented specimens, and scanning these samples using Cu or Co $K\alpha$ radiation. The minerals were identified by their characteristic diffraction patterns with standard diffraction data from Joint Committee on Powder Diffraction Standards (JCPDS). The XRD of clay fractions was conducted by preparing samples oriented on glass slides. These samples consisted of clays that were K-saturated, Mg-saturated, K-saturated and heated to 550 °C, and Mg-saturated and glycerated. Oriented sample specimens were scanned from 2 to 30 ° (2θ) using Co $K\alpha$ radiation. Clay minerals were identified on the basis of the typically known changes in the oriented diffraction patterns brought about by these treatments.

The SEM-EDS analyses of minerals in the sand and gravel fractions of 116-F-4 soil were conducted on selected mineral grains of diverse morphology. First, an optical binocular microscope was used to examine mineral particles under reflected light, and mineral grains with different morphologies were picked out for SEM-EDS analyses. These mineral grains were mounted on carbon specimen stubs, and coatings of electron-conductive carbon were vacuum deposited. Mineral grains were examined in a SEM, and images were obtained using both secondary (SE) and back-scattered electrons (BSE). The BSE images were obtained to detect different mineral inclusions in the major mineral matrix. The chemical composition of different minerals was ascertained from collected x-ray spectra.

Autoradiography was used to identify the distribution of low-energy electron (beta particles) emitting radionuclides on the surface of the gravel particles and on edges of mica particles from soil. The surfaces of the gravel particles were coated with emulsions that are sensitive to beta particles and the images were developed after 2 to 3 months and analyzed using a SEM. Also, particles of mica were mounted on edge and encased in polymeric resin. After the resin hardened, the surface was ground flat and coated with specially formulated photographic emulsion. The images were developed after 2 to 3 months and were examined using a SEM.

Additionally, optical microscopy was used to examine the surface morphology of particles from sand and gravel fractions. Also, the sand fractions of attrition-scrubbed material were analyzed for any changes in morphology and surface features.

4.4 RESULTS AND DISCUSSION

4.4.1 Physical Characterization

The air-dry gravimetric moisture content for <2-mm-sized fraction was 0.58%. The low moisture content was indicative of low contents of clay and organic matter in this soil.

The particle size distribution data are shown in Figure 4-1. The particle size distribution of this soil sample ranged over five orders of magnitude. Predominant fraction of this soil consisted of gravel-sized material (76.3%) with coarse gravel being the most abundant fraction resulting in a mass median diameter of 35 mm (Table 4-1). Even though the total sand fraction constituted 17.3% of the mass, the soil contained negligible quantity of coarse sand (0.3%). The clay and silt content of this sample was also very low (2%). This soil also contained a small fraction (4.4%) of cobble-size material. Using the engineering soil classification criteria (ASTM D 2487-90), the 116-F-4 soil material can be classified as poorly graded gravel (group symbol: GP).

For specific gravity, the tabulated data (Table 4-2) showed that the average of the bulk soil was 2.72. This value is typical of most mineral soils. The specific-gravity data are typically used to assess particle settling times from suspensions.

Table 4-1. Particle-Size Distribution Data for 116-F-4 Pluto Crib Soil

Particle-Size Fraction ^a	% Wt.
Cobble (300 mm - 75 mm)	4.4
Coarse Gravel (75 mm - 19 mm)	64.6
Fine Gravel (19 mm - 4.75 mm)	11.7
Coarse sand (4.75 mm - 2 mm)	0.3
Medium sand (2 mm - 0.425 mm)	4.4
Fine sand (0.425 mm - 0.074 mm)	12.6
Silt and clay (<0.074 mm)	2.0

^aParticle-size designations are based on standard classification method ASTM D 2487-90.

Figure 4-1. Particle-Size Distribution for 116-F-4 Pluto Crib Soil.

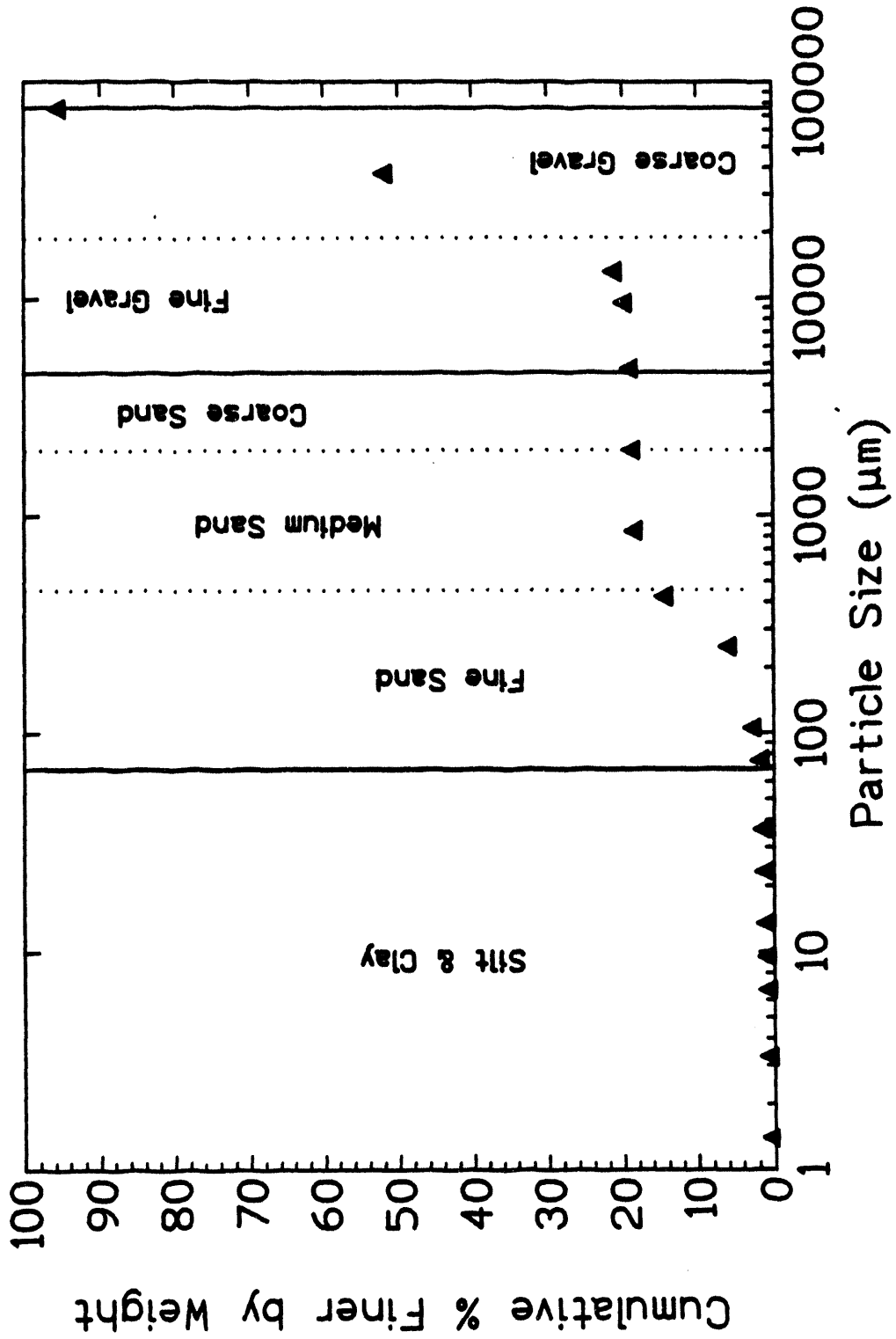


Table 4-2. Specific Gravity of 116-F-4 Pluto Crib Soil.

Particle-Size Fraction	Specific Gravity
Cobble (>75 mm)	2.73
Gravel (75 mm - 4.75 mm)	2.71
Medium sand, silt, and clay (<2 mm)	2.72
Bulk specific gravity	2.72

4.4.2 Chemical Characterization

The TOC content of 116-F-4 soil was 0.25% (Table 4-3). The low TOC value is typical of coarse-textured soils from the 100 Area of Hanford. The pH value indicated that the 116-F-4 was alkaline (Table 4-3) and was probably fully base-saturated with no exchangeable Al being present.

The cation exchange capacity (CEC) measurements (Table 4-3) indicate that the soil is fully base-saturated. These data agree with the conclusions drawn from soil pH measurement. The dominant exchangeable cation was Ca, which accounted for 66% of the total CEC. Minor amounts of exchangeable Mg and trace quantities of exchangeable Ba and Sr were present in this soil. Together, Ca and Na comprised almost all the exchange capacity of these soils (96%). Because of the predominance of Ca on the exchange sites, the soil is expected to flocculate during the soil-washing process.

Table 4-3. Total Organic Carbon Content, pH, and Cation Exchange Capacity of 116-F-4 Pluto Crib soil^a.

Chemical Parameter	Measured Value
TOC (mg/kg)	2500
pH (SU)	8.84
CEC (meq/100g)	19.58 ^b

^aMeasurements conducted on <2-mm material

^bBa: 0.02 meq/100 g, Ca: 12.97 meq/100 g, Mg: 0.75 meq /100 g, Sr: 0.02 meq/100 g, Na: 5.82 meq/100 g

The major and the trace element composition of the 116-F-4 Pluto Crib soil are listed in Tables 4-4 and 4-5, respectively. The major element content is typical of soils containing aluminosilicate minerals. The trace element content is within the range typically found in

uncontaminated soils. These measurements showed that the total concentration of Cr in this soil is about two orders of magnitude below the TPG of 1600 mg/kg.

Table 4-4. Major Element Concentrations in 116-F-4 Pluto Crib Soil^a.

Element	Wt %
Aluminum	5.67
Calcium	4.10
Iron	6.83
Potassium	1.15
Silicon	22.25
Titanium	1.02

^aMeasurements of duplicate samples conducted by XRF spectrometry using Ag, Gd, Fe, and Zr targets.

The radionuclide data for 116-F-4 (<2-mm fraction) soil (Table 4-6) indicated that the activities of all measured radionuclides (except ¹³⁷Cs and ¹⁵²Eu) were from one to three orders of magnitude below the respective TPGs. The measured activity of ¹⁵²Eu was about 5 times lower than the TPG for this radionuclide. These data indicated that the only contaminant radionuclide of concern in the <2-mm-size fraction of this soil is ¹³⁷Cs. The mass concentration of the contaminant radionuclide (¹³⁷Cs) in this soil was calculated from the specific-activity data to be about 7 µg/kg suggesting that ¹³⁷Cs in this soil is probably present in various adsorbed or coprecipitated forms and not as distinct pure solid phases.

The TCLP extraction data (Table 4-7) showed that the extract concentrations of all regulated elements were orders of magnitude lower than the regulatory limit. These data confirmed the conclusion drawn from the total trace element analyses that none of the trace elements are present at concentration levels to be considered a contaminant. Therefore, additional TCLP tests were not conducted on treated soil fractions. Also, the trace level leachability indicated that in these soils chromium is present in its reduced form (Cr III).

The results of different extractive treatments (Table 4-8) showed that the first extractive step removed a minor fraction (0.9%) of ¹³⁷Cs from the soil. The second step also mobilized a very small fraction of this contaminant radionuclide. The extractability of ¹³⁷Cs during the third step was as meager (0.5%) as the first two extractive steps. The reductive extraction step (Step 4) released about 3% of the bound contaminant. Finally, the oxidative acid extractive step released a maximum of 8.4% of ¹³⁷Cs from the soil. All the extractive steps in total released no more than 13% of the ¹³⁷Cs in the <2-mm size fraction of the 116-F-4 soil.

Table 4-5. Trace Element Concentrations in 116-F-4 Pluto Crib Soil^a.

Trace Element	Concentration (mg/kg)	Range of Concentrations (mg/kg) in Uncontaminated Soils ^b
Antimony	<8	0.2 - 10
Arsenic	3	0.1 - 40
Barium	718	100 - 3000
Cadmium	<7	0.01 - 2
Chromium (Total)	16	5 - 1500
Copper	28	2 - 250
Lead	12	2 - 300
Manganese	573	20 - 10000
Nickel	16	2 - 750
Rubidium	60	20 - 1000
Selenium	<1	0.01 - 12
Silver	<7	0.01 - 8
Strontium	445	4 - 2000
Uranium	<5	0.7 - 9
Vanadium	110	3 - 500
Zinc	56	1 - 900
Zirconium	143	60 - 2000

^aMeasurements of duplicate samples conducted by x-ray fluorescence spectrometry using Ag, Gd, Fe, and Zr targets.

^bBowen (1979).

Table 4-6. Radionuclide Data for 116-F-4 Pluto Crib Soil^a.

Radionuclide	Activity (pCi/g)	Test Performance Goals
⁶⁰ Co	0.13	7.1
¹³⁴ Cs	<0.85	10
¹³⁷ Cs	609	30
¹⁵² Eu	3.23	15
¹⁵⁴ Eu	0.28	14
¹⁵⁵ Eu	<0.63	630
⁹⁰ Sr	262	2800
²³⁵ U	0.16	170
²³⁸ U	3.44	370
^{239/240} Pu	18.5	190

^aAnalyses conducted on <2-mm-size material.

Table 4-7. Analyses of Extract from Toxicity Characteristics Leaching Procedure conducted on 116-F-4 Pluto Crib soil^a

Element	Concentration (mg/L)	EPA Regulatory Level (mg/L)
Arsenic	<0.20	5.0
Barium	0.37	100.0
Cadmium	0.09	1.0
Chromium	<0.04	5.0
Silver	<0.02	1.0
Lead	<0.10	5.0
Selenium	<0.20	1.0

^aExtraction conducted on <2-mm-sized soil fraction.

Table 4-8. Sequential Extraction Data for 116-F-4 Pluto Crib Soil.

Extraction Step	¹³⁷ Cs
I. MgCl ₂ extractable %	0.9
II. Acidic sodium acetate soluble %	0.9
III. Reductive-acid soluble %	0.5
IV. Reductive-acid soluble (heated) %	2.6
V. Oxidative-acid soluble (heated) %	8.4
Residual %	86.7
Initial Activity (pCi/g)	609

These sequential extraction data showed that only trace fractions of ¹³⁷Cs in the 116-F-4 soil (<2-mm-size fraction) were associated with "exchangeable," "carbonate-bound," and "Mn-oxide" phases. "Reducible acid-soluble" and "oxidizable acid-soluble" phases in this soil also contained minor fractions of the contaminant. By far the largest fraction of ¹³⁷Cs in this soil (about 87% of total) appeared to be associated with recalcitrant (insoluble) solid phases. These data indicated that the typical extractants used in these extractive steps were collectively capable of mobilizing only a minor fraction (about 13%) of ¹³⁷Cs bound in the mineral matrix.

4.4.3 Mineralogical Characterization

Plagioclase feldspar and quartz were the major minerals (88% by mass) found in the 2- to 0.25-mm-sized fraction and thus constitute the principal mineral matrix of the 116-F-4 soil (Table 4-9). Micaceous minerals (biotite, illite, and muscovite), kaolinite, hornblende, and Fe-titanate were present in minor quantities (1% to 5% by mass). Minerals present in trace quantities (<1%) were smectite, vermiculite, chlorite, Fe-oxide, potassium feldspar, sodium feldspar, magnetite, titanite, cassiterite, and zircon. SEM-EDS data showed that sand-and gravel-size mineral particles in general contained other minerals as inclusions. For instance, quartz, hornblende, and mica were found both as distinct particles and as inclusions in plagioclase feldspar grains (Figure 4-2). Similarly, sodium feldspar also existed as separate particles and as inclusions in potassium-feldspar matrix (Figure 4-3). Fe-titanate was found as inclusions in plagioclase (Figure 4-4) and potassium feldspar particles (Figure 4-5). Titanite and potassium feldspar inclusions were also found in magnetite particles (Figure 4-6). Trace minerals such as cassiterite and zircon were also found as inclusions (Figures 4-7 and 4-8). Optical microscopy showed that both sand and gravel particles had whitish and reddish-yellow coatings (Figures 4-9 and 4-10). Energy dispersive x-ray analyses indicated that these coatings consisted of kaolinitic and iron-oxide minerals.

Table 4-9. Mineralogy of 116-F-4 Pluto Crib Soil

Mineral Phases		
Major Phases	Minor Phases	Trace Phases
Plagioclase Feldspar, Quartz	Mica (biotite, illite, muscovite), kaolinite, Hornblende, Fe-titanate	K-Feldspar, Na-Feldspar, Smectite, Vermiculite, Chlorite, Fe-oxide, Titanite, Magnetite, Cassiterite, Zircon

Table 4-10. Types of ¹³⁷Cs-Mineral Associations.

Types of Sites	Types of Minerals
Exchangeable	Smectite, chlorite, kaolinite, Fe-oxide
"Wedge"	Micas, vermiculite
Structural	K-Feldspars, micas

The types of associations that are known to occur between the contaminant radionuclide (¹³⁷Cs) and the minerals that were identified in these soils are listed in Table 4-10. The exchangeable sites on these minerals typically consist of ionizable surface hydroxyls that adsorb ions. Increasing pH results in more negatively ionized exchange sites, thus resulting in increased cation exchange capacities of minerals. Additionally, smectites possess a number of interlayer exchange sites that are not affected by the pH. These sites originate as a result of isomorphous substitutions within the tetrahedral and octahedral cationic positions within the smectite structure. Depending on the strength of binding, fractions of cations occupying these exchange sites can easily be displaced by electrolytes. The sequential extraction data (see Table 4-8) showed that the easily exchangeable fraction of ¹³⁷Cs in this soil constituted only a minor fraction of the total activity.

Extensive studies have shown that wedge sites on micas and vermiculite have uniquely high affinities for Cs (Scott and Smith 1987). For instance, based on adsorption experiments on various clay minerals, Sawhney (1964) concluded that Cs is preferentially "fixed" into the wedge sites of micas and vermiculites and this fixed Cs was not exchangeable. Because of fixation, mobilization of Cs occupying these wedge sites can only be accomplished by disrupting and/or dissolving the mineral structures. The autoradiograph (Figure 4-11) showed that the edges of mica (possibly wedge sites) contained concentrations of radionuclides. Because these wedge sites have very high affinities for Cs, this autoradiograph might represent the ¹³⁷Cs distribution on these sites. Sequential extraction data (see Table 4-8) confirmed that only up to 11% of ¹³⁷Cs can be extracted by treating this soil with hot reductive and oxidative acids. The standard extractants used in this procedure appeared incapable of releasing the strongly-bound Cs ions (about 89%) that were apparently associated with wedge and structural sites of minerals.

Autoradiography of surfaces of gravel-size particles showed localized distribution of the principal radionuclide contaminant (^{137}Cs) (Figure 4-12). A closer examination of the surfaces indicated that the radionuclide is associated with specific minerals (Figure 4-13). Optical microscopy and EDS analyses indicated that these particle surfaces contain kaolinitic and iron oxide coatings that have high surface areas and mica minerals embedded in feldspathic matrix. Therefore it appears that the radionuclides on the surfaces of gravel particles are associated mainly with kaolinitic and iron oxide coatings and mica minerals. Because these gravels were washed previously, it appears that these coatings and their associated radionuclide activity cannot be mobilized to any significant degree by just water washing.

The data from radionuclide analyses (see Table 4-6), the sequential extraction (see Table 4-8), and mineralogical analyses (Table 4-9) show that the contaminant (^{137}Cs) is present in trace concentrations and appears to be bound with high affinity to specific mineral sites. These characterization data were used to formulate potential soil-washing treatments to effectively release ^{137}Cs and other radionuclides from selected soil size fractions.

Figure 4-2. A Scanning Electron Micrograph (Back-Scattered Mode) of a Plagioclase Feldspar Particle. The lighter inclusions are hornblende.

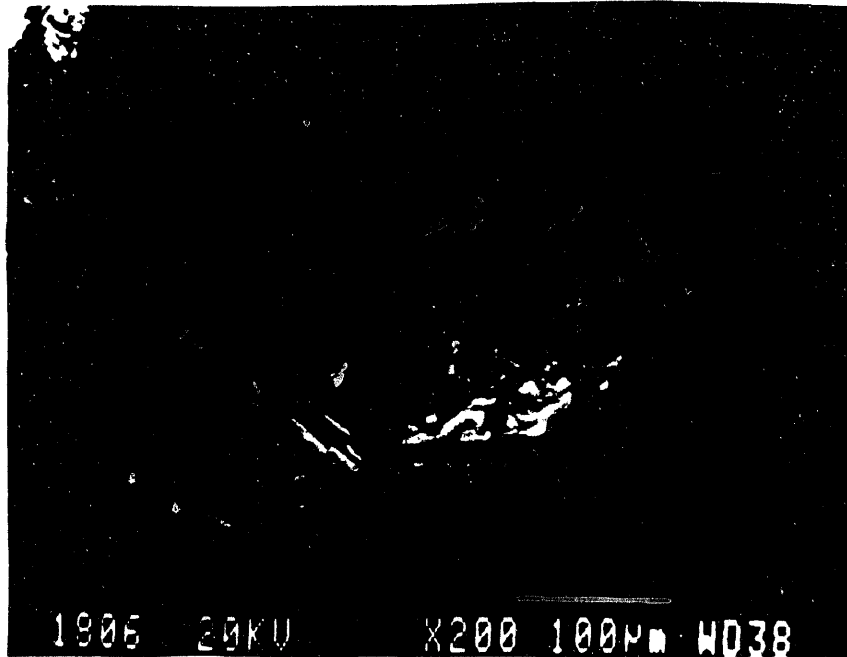


Figure 4-3. A Scanning Electron Micrograph (Back-Scattered Mode) of a Potassium Feldspar Fragment with Sodium Feldspar Inclusions (Darker Areas).

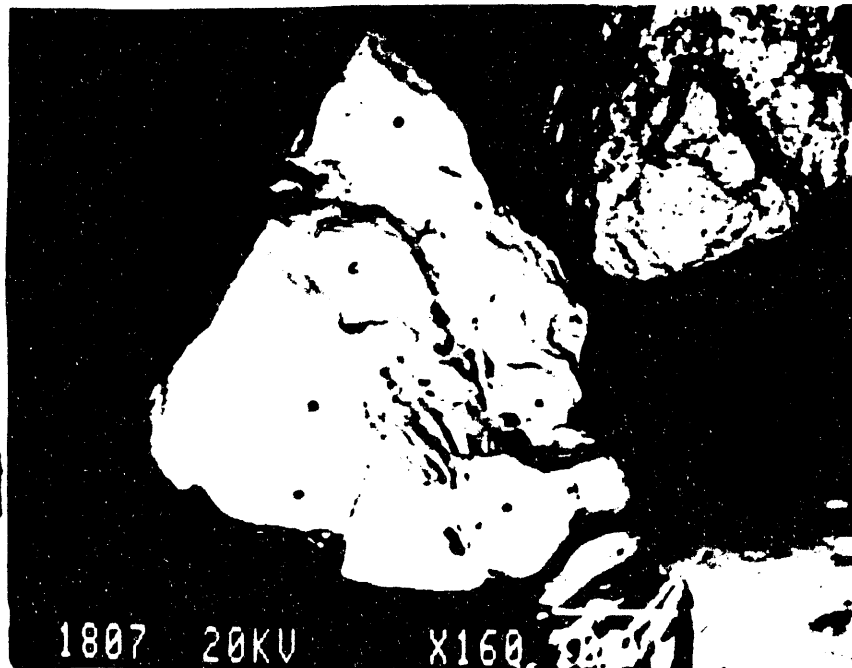


Figure 4-4. A Scanning Electron Micrograph of a Plagioclase Feldspar Grain with Inclusions of Hornblende (Light Areas), and Fe-Titanates (Very Bright Regions).

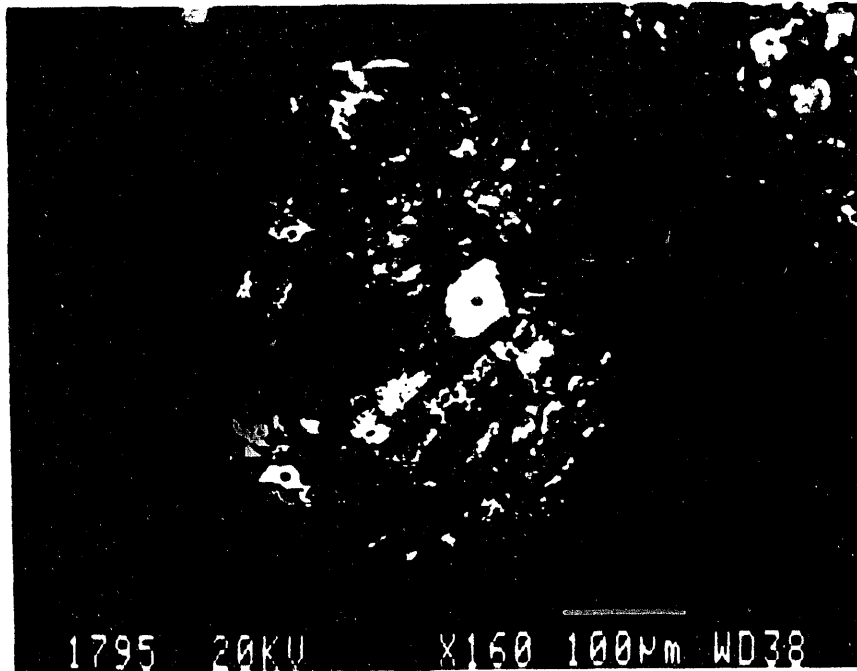


Figure 4-5. A Scanning Electron Micrograph of a Fe-titanate Particle with Potassium Feldspar Inclusions (Darker Areas). The dark particle on the right is quartz.



Figure 4-6. A Scanning Electron Micrograph of a Magnetite Particle with Inclusions of Titanagite (Gray regions) and Potassium Feldspar (Darker Areas). Two smaller potassium feldspar particles contain bright inclusions of Fe-titanate.

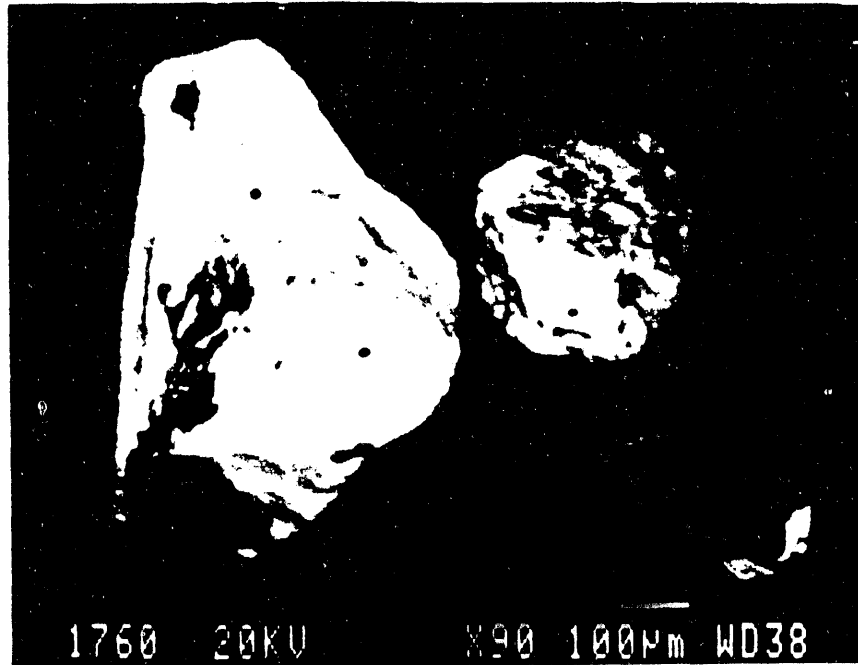


Figure 4-7. A Scanning Electron Micrograph of Cassiterite Inclusions (Bright Region) in Plagioclase Matrix

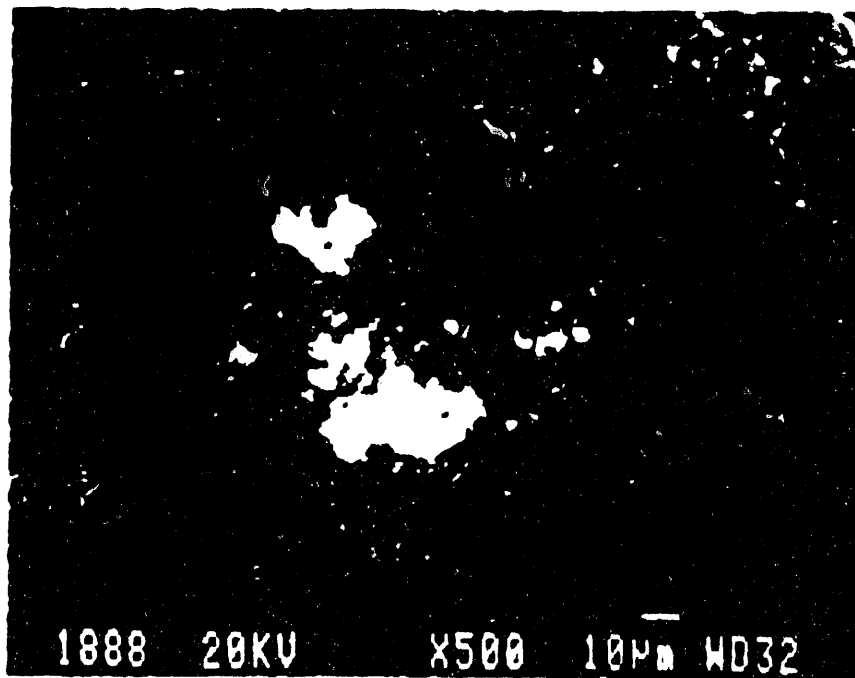


Figure 4-8. A Scanning Electron Micrograph of Zircon Inclusion (Bright Area) in a Quartz Particle.

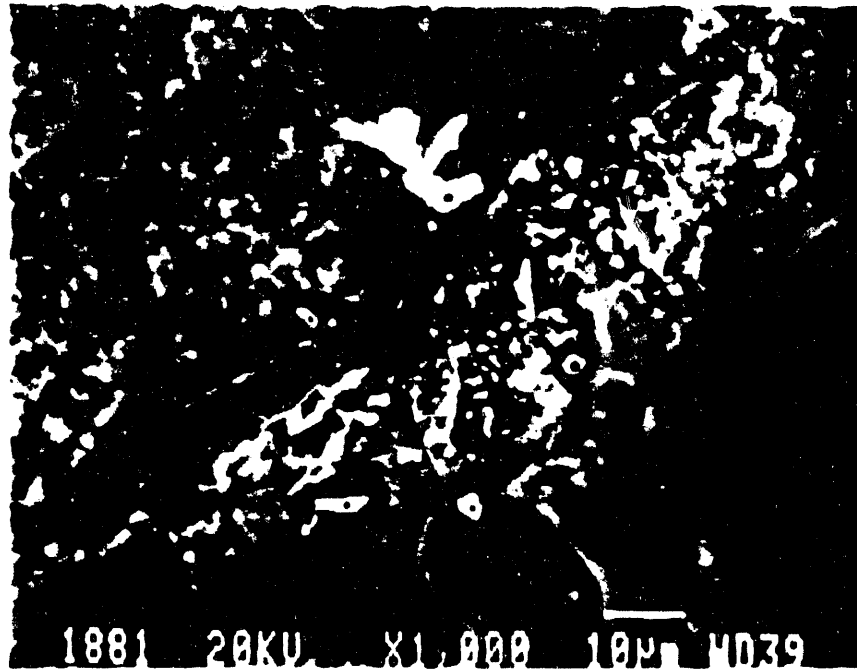


Figure 4-9. An Optical Micrograph of Sand-Sized Particles with Surface Coatings of Kaolinite and Iron Oxide.

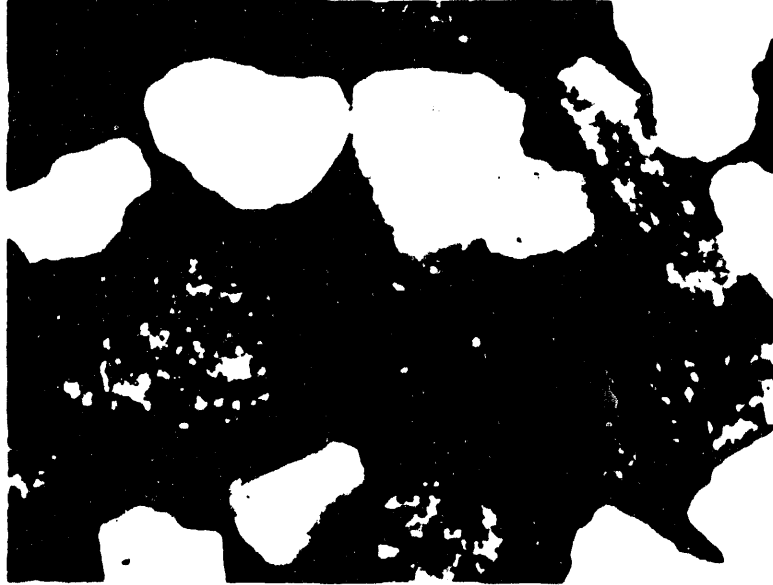


Figure 4-10. An Optical Micrograph of a Gravel Particle with Embedded Mica Mineral and Coatings of Kaolinite and Iron Oxide.

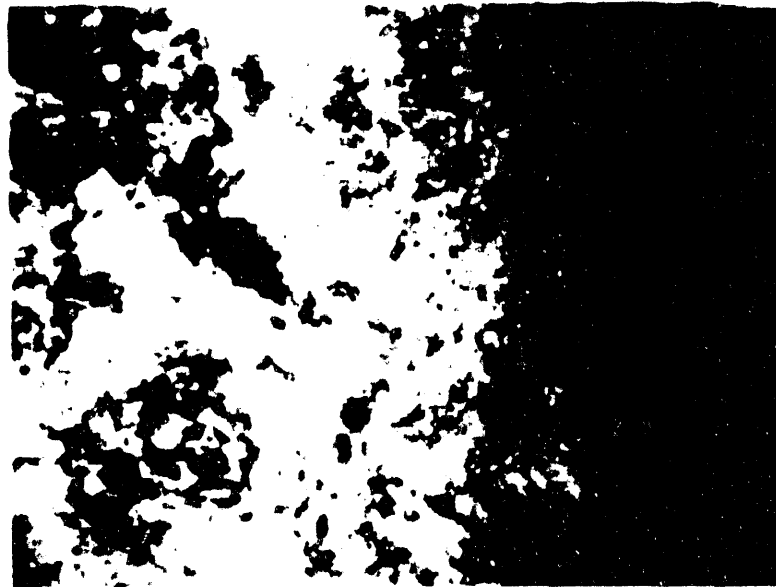


Figure 4-11. An Autoradiograph of Radionuclides Located on the Edges of a Mica Particle.

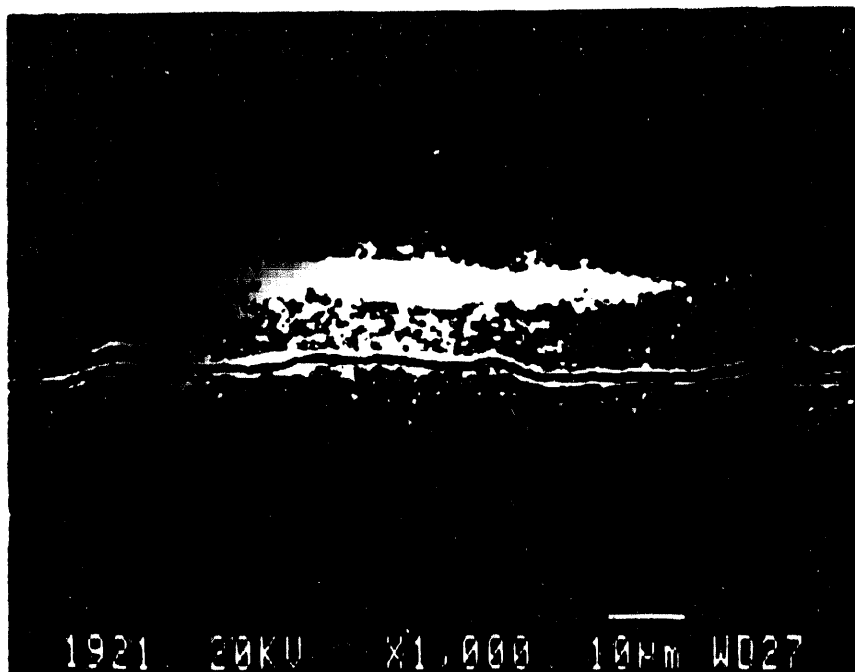


Figure 4-12. An Autoradiograph of the Surface of a Gravel Particle Showing Localized Distribution (Bright Spots) of Radionuclides.

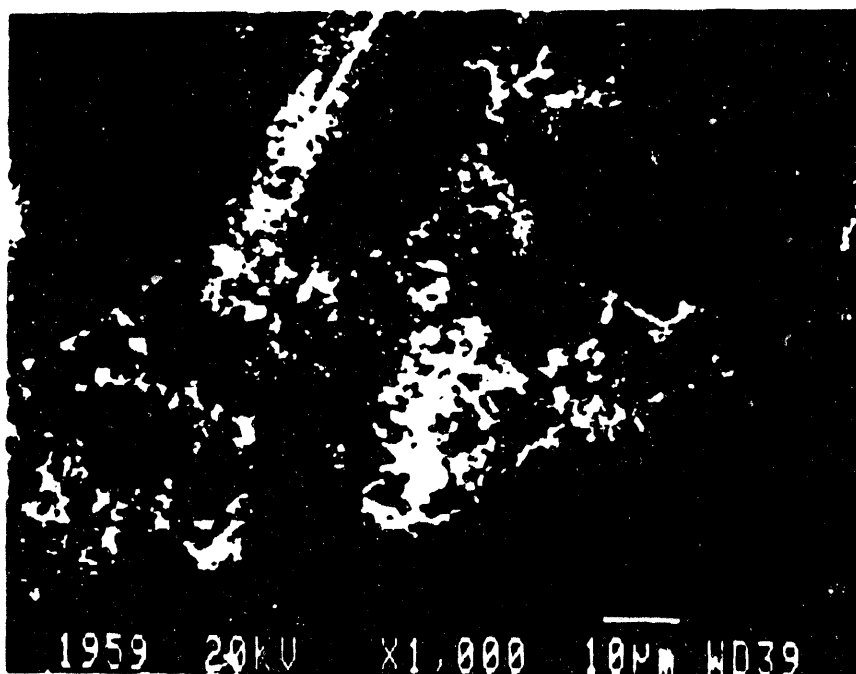
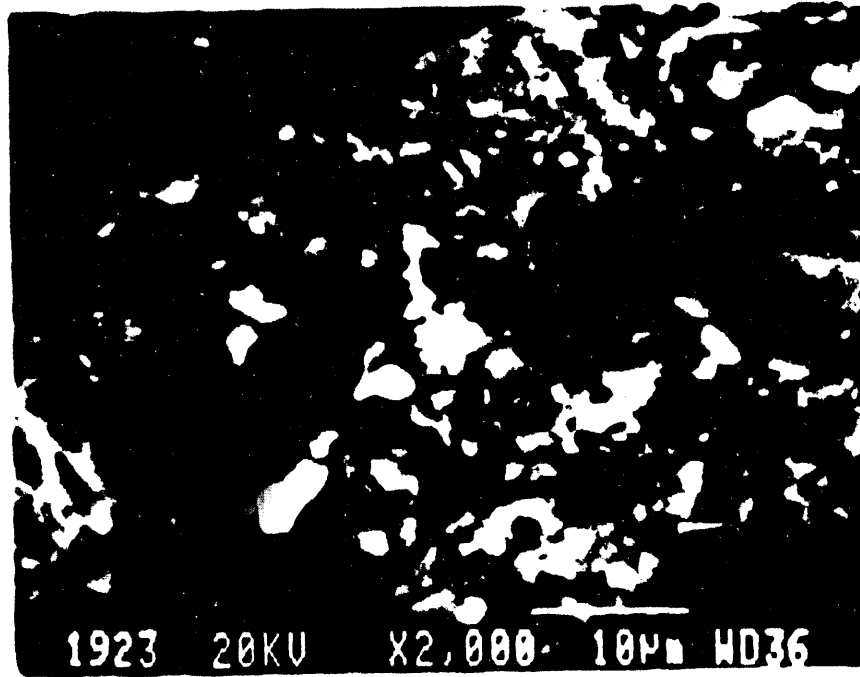


Figure 4-13. An Autoradiograph of Radionuclide Activity Associated with Minerals on the Surface of a Gravel-Size Particle.



5.0 WET SIEVING

5.1 OBJECTIVE

The goal of this test was to find the extent to which contaminants are associated with various particle size fractions of soils. In soils, the finer sized fractions, because of their larger surface areas per unit mass, usually contain larger fractions of contaminants. By preferential removal of fine fractions through wet sieving (i.e., soil washing), significant fractions of the total soil contamination can be isolated for disposal. The wet-screening test was conducted to determine the mass distribution of contaminants within various size fractions of the 116-F-4 soil sample. Also, radionuclide activities were measured in samples of water-washed gravel fractions. The data generated were necessary to assess the contaminant mass in each of the soil size fractions and the activity released into wash water during the sieving process, a proxy for physical soil washing. These data were used to identify the soil fractions that needed additional treatment such as attrition scrubbing and/or chemical extraction to reduce the contaminant activities to meet the contaminant-specific TPGs.

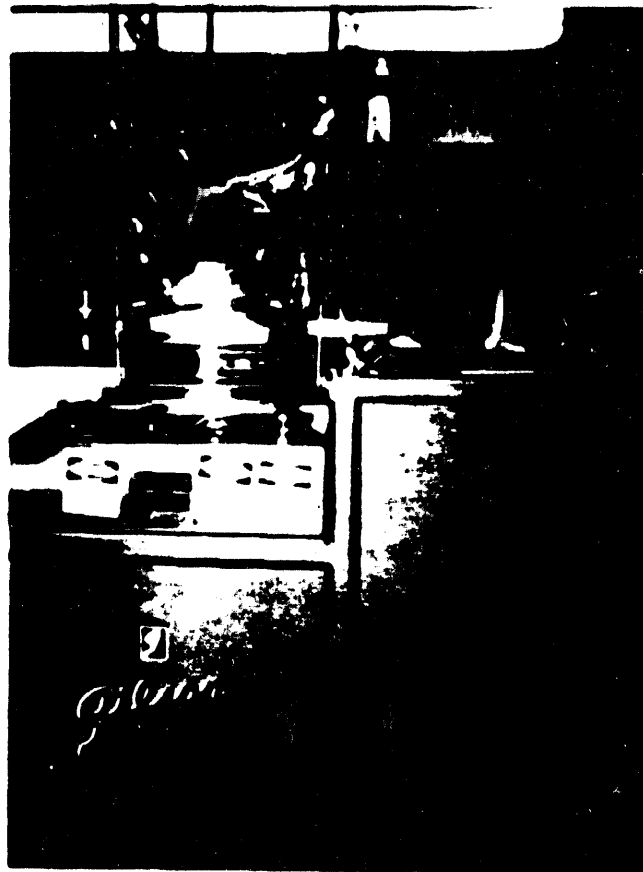
5.2 EQUIPMENT AND PROCEDURES

All wet-sieving experiments were conducted with a Gilson Wet-Vac unit. The unit consists of a sieve-nesting cylinder, filter holder, and a discharge cone assembly mounted on a vibrating frame (Figure 5-1). This unit also includes a water spray system that can be operated either in automated or manual mode. Additionally, vacuum can be applied to the sieve assembly to hasten the screening process. The Gilson wet-sieving system can be operated either by recirculating the wash water or by once-through use of water that drains into an external tank. When wet sieving was conducted in the water recirculating mode, it is necessary to use a filter to screen out the $<25\text{-}\mu\text{m}$ particles from wash water to prevent clogging the water recycling system. Wet sieving operations using this unit can be conducted at different vibration intensities to optimally agitate the wet soil particles.

The procedure used for wet-sieving was similar to the ASTM method D 422-63 except for the following modifications. Because the objective of this test was to examine the contaminant distribution among particle fractions, no dispersant was used. The suggested use of a mixture of sodium hexametaphosphate and sodium hydroxide was omitted because dispersants tend to release and redistribute the contaminants between soil and aqueous phases.

Wet sieving was conducted in once-through mode without the filter section. The once-through-mode sieving required only 3 to 4 L of water during 40 min to completely wet sieve about 0.5 kg of soil. A set of sieves consisting of 2-mm (No. 10), 0.420-mm (No. 40), 0.25-mm (No. 60), and 0.074-mm (No. 200) was used to conduct wet-sieving tests. At the end of the sieving cycle, soil fractions retained on each sieve were rinsed with fresh deionized distilled water until the wash water was clear. The soil fractions retained on the sieves were oven dried at 105 ± 5 °C and weighed. The oven-dried soil fractions were composited to represent $>2\text{-mm-}$, $2.00\text{- to }0.42\text{-mm-}$, $0.42\text{- to }0.25\text{-mm-}$, $0.25\text{- to }0.074\text{-mm}$ and, $<0.074\text{-mm-}$ sized fractions.

Figure 5-1. Gilson Wet-Vac Wet-Sieving Unit.



The quantity of soil that could be optimally sieved in each sieving cycle depends on the texture of the soil and ranges from 75 to 150 g. Exceeding the optimal sample weight for a soil causes sieve blinding. Therefore, wet sieving was conducted a number of times to accumulate sufficient quantities of soil in each sieve fraction. These soil fractions and the wash water were analyzed for the contaminants of interest (see section 10.0 for washwater data). Mass balances were computed for weight fractions, and the activity balances were computed from the radionuclide data. Manually water-washed gravel fractions were used for radionuclide determination.

5.3 RESULTS AND DISCUSSION

The wet-sieving data (Table 5-1) indicated that the particle size distribution in 116-F-4 soil was bimodally distributed. A major fraction of the soil material (about 79% of the mass) exists in >13.5-mm size fraction. About 13% of the soil mass consists of sand-size material. Measurements of radionuclide activity indicated that all particle size fractions contained ^{137}Cs activities that were above the TPG (Figure 5-2). However, the activities of ^{60}Co , and ^{152}Eu in all size fractions except in 4.76-mm - 2mm and <0.074-mm were well below their respective TPGs (Table 5-1). Additionally, the ^{90}Sr and $^{239/240}\text{Pu}$ activities measured in selected size fractions (constituting 97.7% of the total soil mass) indicated that these radionuclides were present in more than an order of magnitude lower activities than their respective TPGs.

Table 5-1. Contaminant Radionuclide and Mass Distribution Data for 116-F-4 Pluto Crib Soil.

Particle-Size (mm)	¹³⁷ Cs	⁶⁰ Co	¹⁵² Eu	⁹⁰ Sr	^{239/240} Pu	Wt%
	pCi/g					
> 38.1	337	<1	2	4.2	3.6	48.2
38.1 - 13.5	186	<1	<1	6.7	6.5	30.5
13.5 - 9.51	310	<1	<1	--	--	1.3
9.51 - 4.76	186	<1	<1	--	--	0.7
4.76 - 2	451	53	11	--	--	0.3
2 - 0.42	218	1.2	0.4	100	18.5	5.2
0.42 - 0.25	220	0.2	0.4			7.7
0.25 - 0.074	449	0.6	0.8			3.8
<0.074	2620	1.1	17			2.3
Bulk Soil	331	<1.1	<1.8	55 ^a	7.4 ^a	100

<2-mm size fraction contained 262 pCi/g of ⁹⁰Sr

^aActivity in 97.7% of the soil mass.

Except in the finest size-fraction, the radionuclide activities (¹³⁷Cs, ⁶⁰Co, and ¹⁵²Eu) in all other size-fractions did not show the typical particle size-activity relationship (i.e., increasing activity per unit mass with decreasing particle size). One possible explanation for this anomaly is that the bulk of the soil material in the crib (consisting of gravel and cobble size material) was subjected to the cooling water with higher activities of radionuclides thus acquired a higher fraction of the total burden of radionuclides. The lack of typical particle size-activity relationship for this soil results in uniform activity per unit mass for various cumulative particle-size composites (Table 5-2). These computations showed that wet sieving 116-F-4 soil cannot isolate any lower activity size fractions and that any chosen particle-size fraction has to be treated to reduce the ¹³⁷Cs activity to meet the TPG.

Calculations showed that about two-thirds of the total ¹³⁷Cs activity in the soil exists in gravel-size (> 13.5-mm) fractions (Figure 5-3). The sand fraction (2- to 0.25-mm fraction) contains about 9% of the total ¹³⁷Cs activity in the soil, and the silt and clay fraction (<0.074 mm) contains about 18% of the total ¹³⁷Cs activity. Together these size fractions account for about 93% of the total ¹³⁷Cs activity in the 116-F-4 soil. Typical ameliorative soil-washing processes (attrition scrubbing, autogenous surface grinding, and chemical extraction) are designed to treat sand and gravel fractions. The silt and clay fractions (separated by wet sieving) are usually disposed of without any treatment.

The wet-sieving data showed that the radionuclide contaminant of concern in 116-F-4 soil was ¹³⁷Cs. Also, the mode of contaminant distribution showed that wet sieving alone

cannot isolate particle-size fractions with activity levels below the TPG. Thus, the ¹³⁷Cs activity and distribution data indicated that to meet the TPG additional ameliorative treatments must be conducted on both the sand and gravel fractions of this soil.

Table 5-2. Calculated Activity of ¹³⁷Cs in Cumulative Particle-Size Fractions of 116-F-4 Pluto Crib Soil.

Particle Size (mm)	¹³⁷ Cs Activity of Composite (pCi/g)	Cumulative Wt %
> 38.1	337	48.2
> 13.5	278	78.7
> 9.51	279	80.0
> 4.76	278	80.7
> 2.00	279	81.0
> 0.42	275	86.2
> 0.25	271	93.9
> 0.074	280	97.7

Figure 5-2. Percent of Total ¹³⁷Cs Activity Distribution Among Different Particle-Size Fractions of 116-F-4 Pluto Crib Soil.

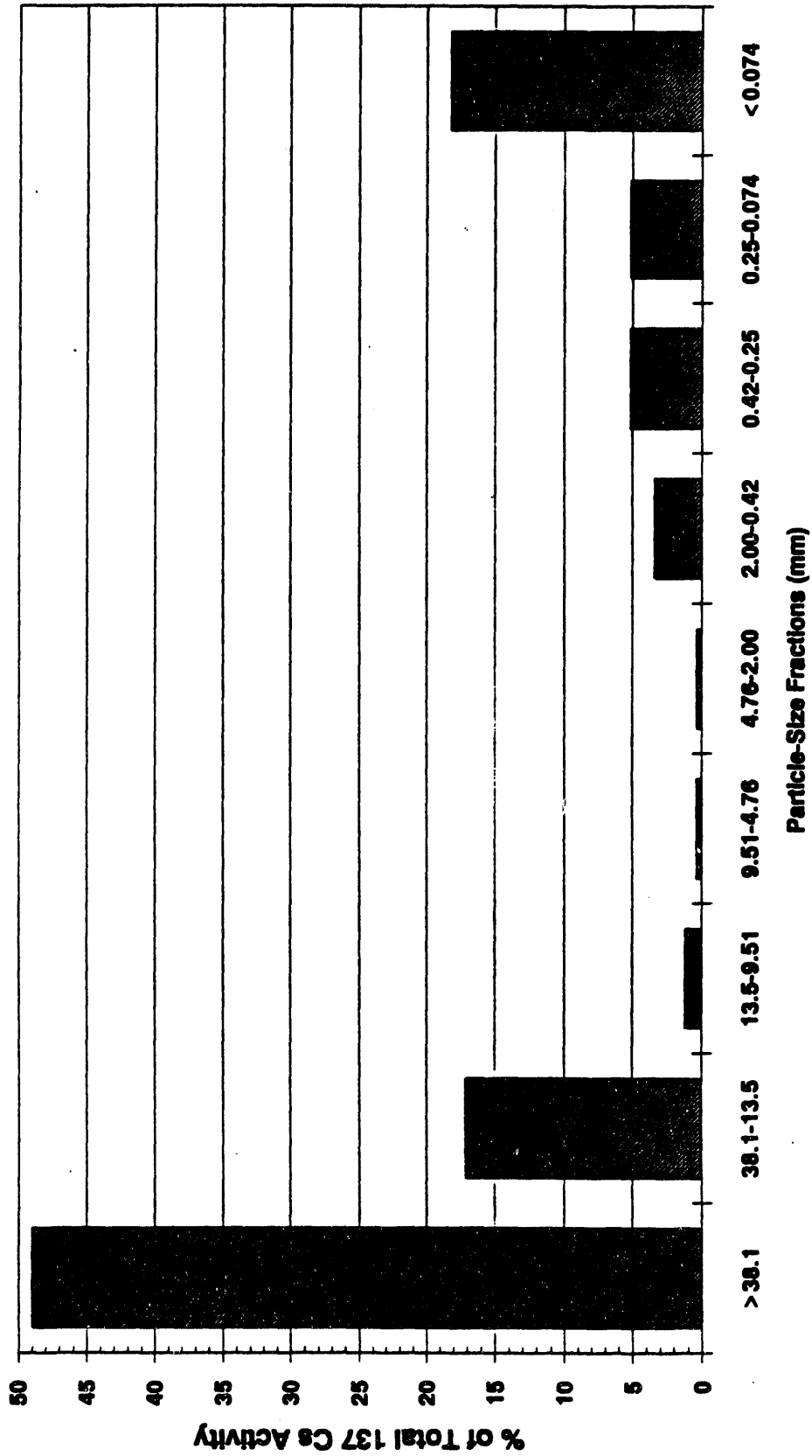
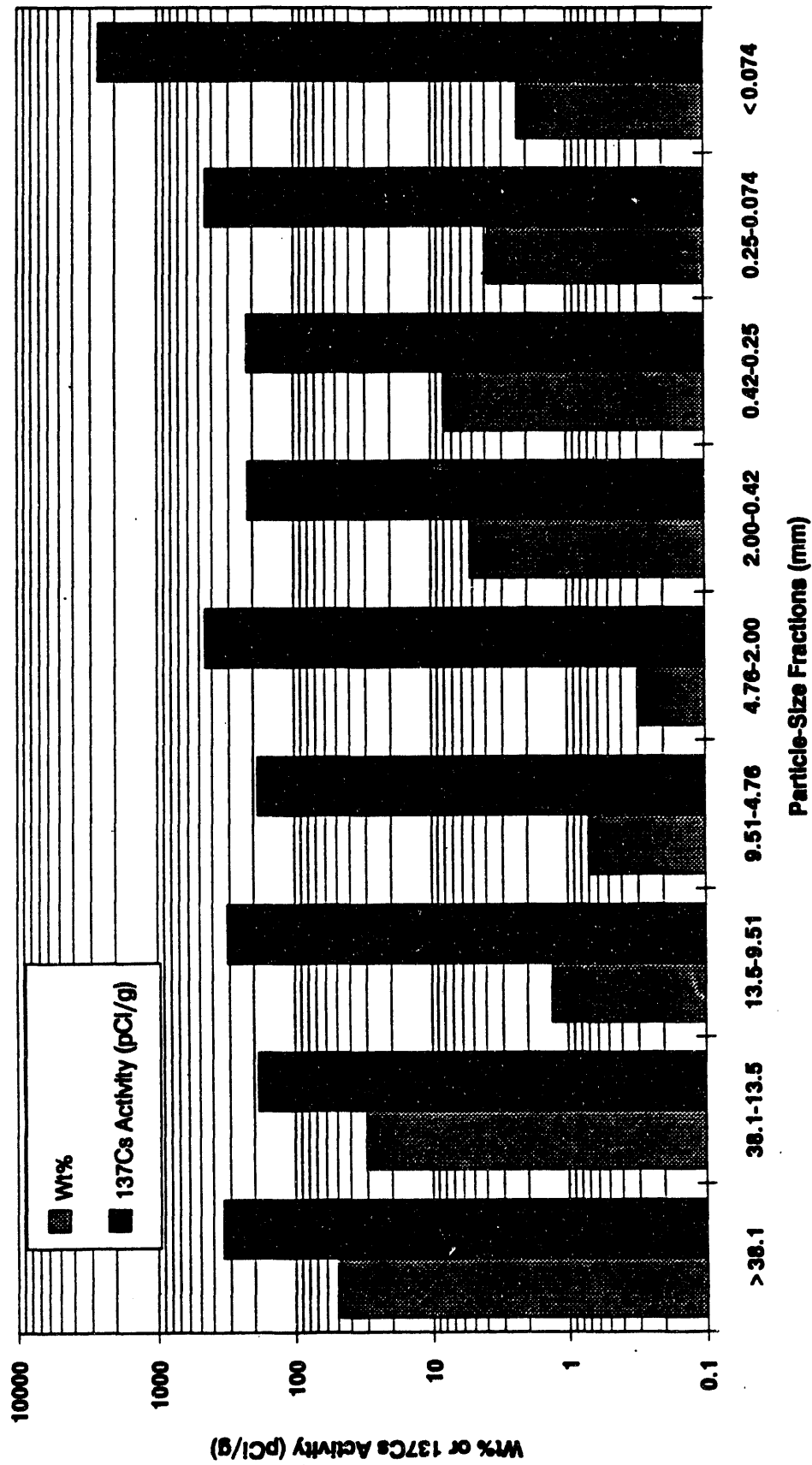


Figure 5-3. Distribution of ¹³⁷Cs Activity Among Different Particle-Size Fractions.



6.0 ATTRITION SCRUBBING

6.1 OBJECTIVE

The attrition scrubbing tests were conducted to examine whether contaminants can be removed from the surfaces of soil particles through scrubbing action. These tests were not necessarily designed to provide data to directly design full-scale equipment. However, the results should show whether some form of attrition scrubbing is beneficial in partitioning contaminants to the fine fraction. The parameters that affect attrition scrubbing, including solids density, impeller speed, residence time, and the use of electrolytes, have been previously evaluated using 2- to 0.25-mm fractions from 116-D-1B soil from Hanford Site (DOE-RL 1993b). Attrition scrubbing tests are usually conducted on sand-sized material. All the scrubbing tests reported in this section were conducted on 2- to 0.25-mm-sized fractions.

6.2 EQUIPMENT AND PROCEDURES

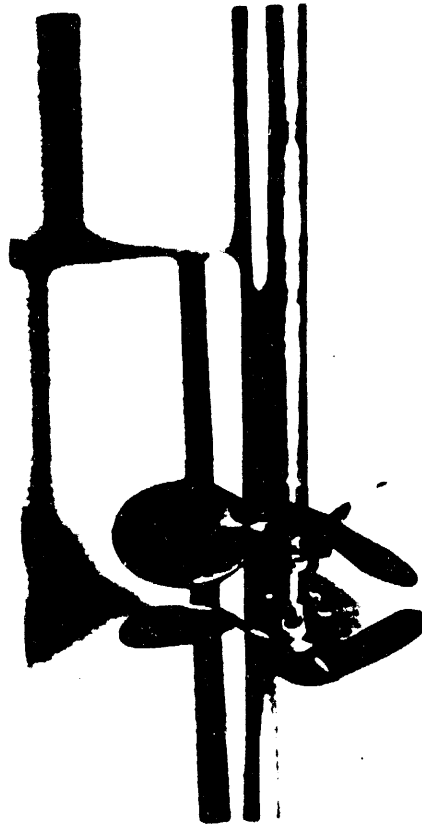
The attrition scrubbing tests were performed in a laboratory-scale attrition scrubber fabricated from a high-torque, servo-controlled, stir motor with a stainless steel shaft and two three-bladed 7.5-cm-diameter impellers (Figure 6-1). The impellers were fixed on the end of the steel shaft with the blades aligned with opposing pitch. This configuration maximizes the particle-to-particle contact and results in the desired scrubbing action. The motor speed can be continuously adjusted from 100 to 2000 rpm, and the speed is maintained by a servo-control loop to ensure reproducibility between tests. The motor controller also has a built-in timer to allow the contact times to be controlled precisely. The scrubber can be programmed to run at either a fixed rpm or specified torque. The mix containers used in these experiments were 1-L rectangular containers.

All scrubbing tests were conducted on approximately 500-g loads of previously wet-sieved and air-dried 2- to 0.25-mm-sized fractions of 116-F-4 Pluto Crib soil. Preliminary observations indicated that particles were moving effectively when scrubbing was conducted with impeller speed set at 900 -1200 rpm. Previous tests on 116-D-1B soil established the range of pulp densities and energy input necessary to achieve optimum scrubbing of the soil material (DOE-RL 1993b). These data were used to conduct optimized attrition-scrubbing tests on 116-F-4 soil material. During these tests, torque values were recorded at 5-min intervals to track the total energy input for each experiment. The energy input per unit mass (HP-min/lb) was computed by the relationship (Shigley and Mischke 1989):

$$\text{HP-min/lb} = (T \times n \times t) / (1.008 \times 10^6 \times M) \quad (6-1)$$

Where T is the torque in units of oz.in, n is the shaft speed in revolutions per minute (rpm), t is the scrubbing time in minutes, and M is the mass of soil being scrubbed.

Figure 6-1. Close-up View of Opposing Pitch Impellers.



The scrubbing tests were conducted in two stages in an electrolyte medium consisting of 0.5M ammonium citrate with enough citric acid added to bring the initial pH to 3.5. Scrubbing was conducted at 79% pulp density for 30 min with an average energy input of 1.5 HP min/lb. Attrition scrubbing with electrolyte was conducted because previous experiments indicated that such scrubbing enhanced the removal of radionuclides from soil (DOE-RL, 1993b). Such enhanced removal by electrolyte addition appeared to be a result of the synergistic combination of scrubbing action, the improved dissolution of radionuclide-bearing surface coatings, and the reduced reabsorption of solubilized contaminants onto freshly exposed surfaces of the coarse-grained soil.

The two-stage tests were performed by conducting the first stage scrubbing with electrolyte and wet sieving the scrubbed material through a 0.25-mm sieve to remove the fines that were generated. Removal of fines after scrubbing was necessary because accumulation of fines reduces scrubbing efficiency (DOE-RL 1993b). The second-stage scrubbing was conducted on washed and air-dried 2- to 0.25-mm- sized material, with electrolyte added to bring the pulp densities up to 79%. After completing the second-stage scrubbing, the material again was wet sieved through a 0.25-mm sieve, and the coarse fraction was oven dried at 105 ± 5 °C, and representative subsamples taken for gamma counting.

A three-stage attrition scrubbing experiment was also conducted to examine the effectiveness of additional scrubbing on cumulative removal of ^{137}Cs from the sand fraction (2- to 0.25-mm).

6.3 RESULTS AND DISCUSSION

The attrition scrubbing data (Table 6-1) showed that during the first stage, approximately 40% of the ^{137}Cs activity could be removed from the sand fraction (2- to 0.25-mm) with concomitant generation of about 6% to 15% fines. The activity of ^{137}Cs in first-stage scrubbed fractions ranged from 115 to 132 pCi/g well above the TPG. The second-stage attrition scrubbing on average reduced the activity an additional 11% (the overall reduction in activity was about 50%). The measured ^{137}Cs activities after scrubbing in two stages ranged from 91 to 114 pCi/g. These residual activities were also well above the TPG of 30 pCi/g. Optical microscopic examination of the attrition-scrubbed coarse material showed (Figure 6-2) significant reductions in whitish and yellow-red coatings that were observed on initial material (Figure 4-9). The scrubbed particles also presented a more rounded appearance as compared to the more angular morphology of unscrubbed material.

The third-stage scrubbing removed an additional 8% of the activity (for a total reduction in activity of 64%) resulting in residual activity of 75 pCi/g in the coarse fraction. This residual activity exceeds the TPG of 30 pCi/g indicating that even attrition scrubbing with electrolyte in three stages cannot reduce the ^{137}Cs activity in the sand fraction of 116-F-4 soil to the required TPG. Therefore, it appears that additional treatments such as chemical extraction may be necessary to reduce further the activity of ^{137}Cs to meet the TPGs.

Table 6-1. Attrition Scrubbing Data for 2- to 0.25-mm-Sized Fraction of 116-F-4 Pluto Crib Soil.

Test #	Attrition Scrubbing Stage	^{137}Cs Initial Activity (pCi/g)	^{137}Cs Final Activity (pCi/g)	% Reduction in ^{137}Cs Activity ^a	Wt % Fines	HPmin/lb
1	I	208	132	37	5.7	0.984
1	II	132	114	45	4.2	1.736
2	I	204	115	44	7.3	1.427
2	II	115	91	55	3.9	1.578
3	I	203	122	40	14.5	1.510
3	II	122	98	52	6.0	1.960
4	I	208	120	42	5.7	--
4	II	120	91	56	4.2	--
4	III	91	75	64	4.0	--

^aBased on initial activity.

Figure 6-2. Optical Micrograph of Twice Attrited Sand-Size Particles from 116-F-4 Soil.



7.0 AUTOGENOUS SURFACE GRINDING

7.1 OBJECTIVE

The autogenous surface grinding experiments were conducted to test the removal of surface contamination from gravel-sized material from 116-F-4 Pluto Crib soil. The goal of this treatment was to remove surface contamination through the self-grinding action of cobbles and gravels. Autogenous grinding is a process that uses ball or rod mills (without steel balls or rods) to promote crushing of the material. The objective of the experiments was to promote grinding of the contaminated particle surface and to minimize the pulverization of particles. The material from 116-F-4 consisted mainly (about 81% of total mass) of gravel-sized particles with ^{137}Cs as the main contaminant (Table 5-1). Because washing this material with water did not significantly reduce the ^{137}Cs activity, additional physical treatment such as autogenous grinding was tested as a means to reduce the surface-bound contaminants.

Previous autogenous surface grinding experiments on gravel fractions from 116-C-1 (Batch II) trench soil (DOE-RL 1993b) indicated that it was necessary to remove a large fraction of the surface through more intense grinding to achieve the TPG for the contaminant radionuclides. This could be achieved either through prolonged grinding in a rod mill or grinding for a shorter time period using a high-intensity grinding apparatus. Using a surrogate rod mill, a preliminary long-term experiment was conducted to assess the grindability of gravel surfaces from 116-F-4 soil. High intensity surface grinding experiments in water and in an electrolyte media were also conducted using a laboratory-scale centrifugal barrel (CB) processor.

Typical CB processors consist of two to six barrels mounted on the periphery of a turret. The turret rotates at a high speed in one direction, while the barrels spin at a relatively lower speed in the opposite direction (Figure 7-1). The rotation of the turret can exert up to 50 g force on the material in the drums. The counter rotating drums force the particles to grind intensely against each other. The intense grinding removes surface material in time spans that are significantly shorter than that achieved by a rod or a ball mill (Rhoades 1981). An additional advantage of the CB processor is that the particles in the barrels slide and grind against each other and do not fall or impact against each other as in ball or rod mills. This smooth action increases grinding action and significantly reduces breaking of particles.

The radial acceleration (A) on a particle within a drum of a CB processor can be computed as follows (Rhoades 1981),

$$A = V^2/R \quad (7-1)$$

Where V is the velocity of the particle and R is the distance of the particle from the center of the barrel. The velocity of the particle is computed as,

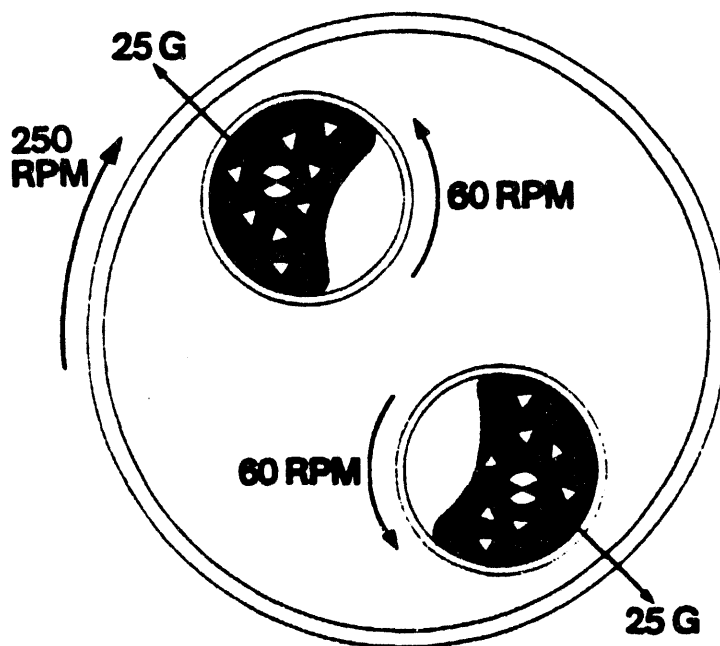
$$V = 2\pi R(\text{RPS}) \quad (7-2)$$

Where RPS is the speed of rotation of the turret in revolutions per second. The radial acceleration A is equal to 32G where G is the gravitational force. Therefore the gravitational force on a particle within the barrel of a CB processor can be calculated as,

$$G = 1.234R (\text{RPS})^2 \quad (7-3)$$

Clearly, the particles at the outer periphery of the drums (farther from the center of the turret) experience greater grinding force (larger R) than that experienced by the particles that are closer to the center of the turret (smaller R). Typically, CB processors are designed so that the force difference between particles at these two locations is approximately 50%, which results in smooth grinding action.

Figure 7-1. Schematic Diagram of a Centrifugal Barrel Processor.

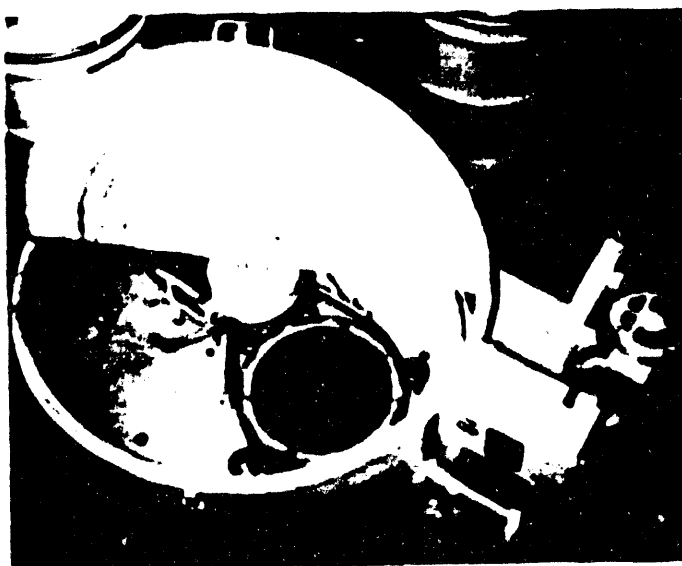


7.2 EQUIPMENT AND PROCEDURES

A long-term autogenous surface grinding experiment was conducted using a rock tumbler as a surrogate laboratory-scale rod mill. The grinding load consisted of a single 103-g contaminated (water-washed) gravel particle from 116-F-4 soil, 59 g of uncontaminated gravel pieces, 100 g of coarse carborundum powder, and about 235 mL of an extractant (see section 8.0). Spent carborundum medium was replaced with fresh material every 6-days. This scoping test was conducted over a period of 17 days. The residual ^{137}Cs activity in the contaminated gravel fragment was monitored at 1, 2, or 4 day intervals.

The equipment used for high-intensity autogenous surface grinding consisted of a Harper centrifugal barrel processor with a 3-ft-diameter turret that contained two barrels each 14.25 in. diameter and 6 in. high (Figure 7-2). Each of these barrels has a maximum capacity of 2.5 kg of material, and the grinding can be conducted at centrifugal force ranging from 6 to 19g force. Autogenous surface grinding experiments were conducted on gravel-sized material (approximately 2-in size) separated from 116-F-4 Pluto Crib soil. All CB surface grinding experiments were conducted on approximately 1.5- to 2-kg batches that were prewashed, air dried, and counted for ^{137}Cs activity. The surface grinding experiments were conducted in either water or an electrolyte medium consisting of 0.5M ammonium citrate with enough citric acid added to bring the initial pH to 3.5. The composition of electrolyte in these experiments was the same as that used in attrition scrubbing experiments. The experiments were conducted for the required time periods after which the gravel fractions were washed with water to remove the fines and the reduction in radionuclide activity was calculated from gamma counting the oven-dry fines. Grinding up to a total of 80 min (in 20- min increments) was conducted on prewashed gravel fractions. Continuous grinding for 40 to 60 min was also tested to examine the effects of grinding for longer time intervals. All grinding was conducted at a centrifugal force of 19g force.

Figure 7-2. Laboratory-Scale Centrifugal Barrel Processor Used for Autogenous Surface Grinding.



7.3 RESULTS AND DISCUSSION

The data from the long-term autogenous surface grinding experiment are listed in Table 7-1. The results indicate that the first three days of grinding removed a major fraction (about 81%) of ¹³⁷Cs activity from the gravel particle. The data show that the cumulative reduction in ¹³⁷Cs activity correlated with the cumulative amount of fines generated from autogenous surface grinding (Figure 7-3). The initial 10% of the fines contained about 80% of the activity associated with the gravel particle. Subsequent 10% of the fines contained about 10% of the total activity.

The residual ¹³⁷Cs activity in the gravel fragment indicated a very rapid initial decline followed by a declining rate of activity reduction (Figure 7-4). At the conclusion of the experiment, the residual ¹³⁷Cs activity had declined from an initial activity of about 1007 pCi/g to a relatively very low 35 pCi/g.

The approximate thickness of material ground off from the surface of the gravel particle was computed to examine any relationship that may exist between this parameter and the mobilized and residual activities. The calculations were conducted with a first

Table 7-1. Long-Term Autogenous Surface Grinding Data.

Cumulative Grinding Time (days)	Total Residual ¹³⁷ Cs Activity (pCi)	Cumulative % Reduction in Total Activity	¹³⁷ Cs Activity per Unit Mass (pCi/g)	Cumulative Wt % Fines
0	103902	0	1007	0
1	37252	64.1	373	3
2	25021	75.9	262	7
3	19462	81.3	212	11
4	15197	85.4	171	14
5	13225	87.3	153	16
6	11527	88.9	136	18
8	6022	94.2	79	26
9	4610	95.6	63	29
10	3983	96.2	56	31
11	3694	96.4	53	32
12	3279	96.8	48	34
15	2466	97.6	38	37
17	2214	97.9	35	39

Figure 7-3. Cumulative Reduction in ^{137}Cs Activity of the Gravel Fragment as a Function of Cumulative Mass of Fines.

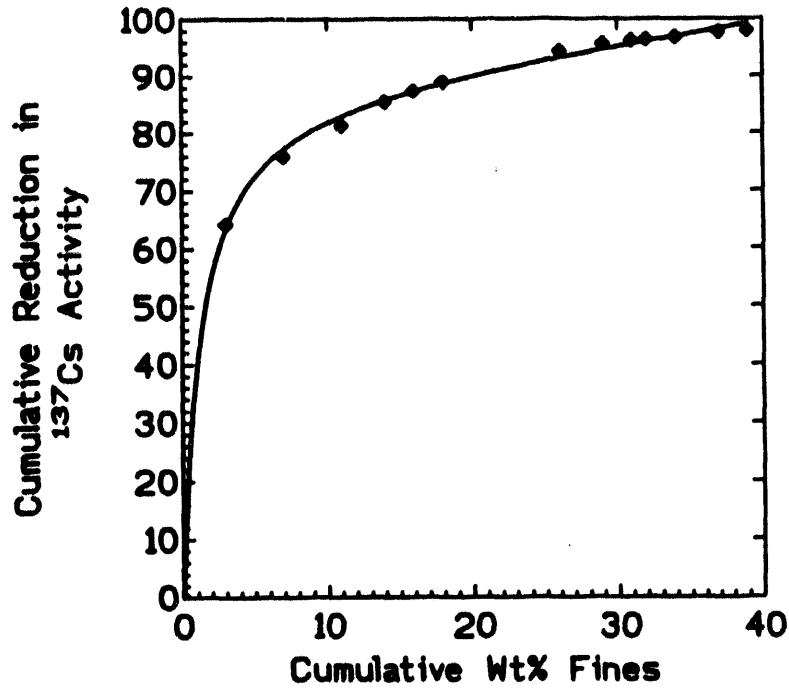
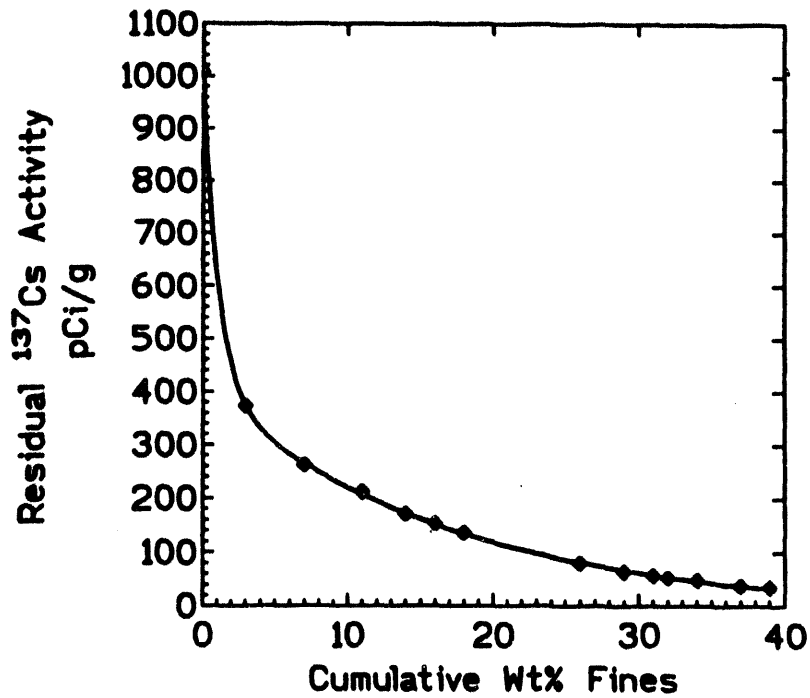


Figure. 7-4. Residual ^{137}Cs Activity of the Gravel Fragment as a Function of Cumulative Mass of Fines.



approximate assumption that the gravel consisted of an equivalent spherical particle. The equivalent diameter (d) of gravel particles in a grinding load (W) was computed as:

$$V = W/n\rho \quad (7-4)$$

Where V is the volume of each gravel particle, n is the number of gravel particles in each grinding load, and ρ is the particle density of soil material. The equivalent diameter for each gravel particle is calculated as,

$$d = [6W/\pi n\rho]^{1/3} \quad (7-5)$$

The thickness of the material ground off during each successive grinding period was calculated as one half of difference between successive computed equivalent diameters.

The computed cumulative thickness removed during the prolonged surface grinding correlated with both the cumulative percent reduction, and the residual ^{137}Cs activity (Figure 7-5, 7-6). The relationship indicated that almost two-thirds of the total activity in this gravel particle resided in the first 0.2-mm of the surface. The data indicated that a major fraction (about 85%) of the total ^{137}Cs activity can be mobilized by grinding off the first millimeter of the gravel surface. These computations suggested that only a minor fraction of ^{137}Cs has penetrated beyond the first three millimeter of the gravel surface matrix. This preliminary experiment indicated that it was possible to remove a major fraction of the contaminant by grinding the first few millimeters of the gravel surfaces.

The data from autogenous surface grinding experiments conducted with the CB processor are listed in Table 7-2. The results showed that the rate of removal of ^{137}Cs activity from the gravel particles depended on the liquid medium that was present during the surface grinding process. When surface grinding was conducted in water medium, about 75% of the total ^{137}Cs activity was mobilized from the gravel fractions with the formation of about 13% fines (Figure 7-7). By contrast, the surface grinding conducted in an electrolyte medium appeared to mobilize 20 to 35% less activity while generating similar amounts of fines as water medium experiments. This apparent retarding effect of electrolyte on contaminant mobilization seemed anomalous because attrition scrubbing of sand fractions conducted in electrolyte resulted in consistently enhanced mobilization of radionuclide contaminants (DOE-RL 1993a). One possible explanation for this anomaly is that the mobilized Cs in the presence of electrolyte may form neutral and/or anionic Cs-citrate soluble complexes that may be diffusing deeper into the gravel matrix. Alternatively, the difference between ^{137}Cs removal in the presence of water and in electrolyte may be a perceived anomaly resulting from small data sets.

Residual ^{137}Cs activities in the gravel fractions also showed functional relationships with the cumulative fines generated (Figure 7-8). At the end of grinding experiments, gravels ground in water medium contained significantly lower ^{137}Cs activities (10 to 35 pCi/g) than gravels ground in electrolyte (146 to 211 pCi/g) even though the similar mass percentage of fines were generated in both sets of tests.

Table 7-5. Cumulative Reduction in ¹³⁷Cs Activity of the Gravel Fragment as a Function of Cumulative Grinding Thickness.

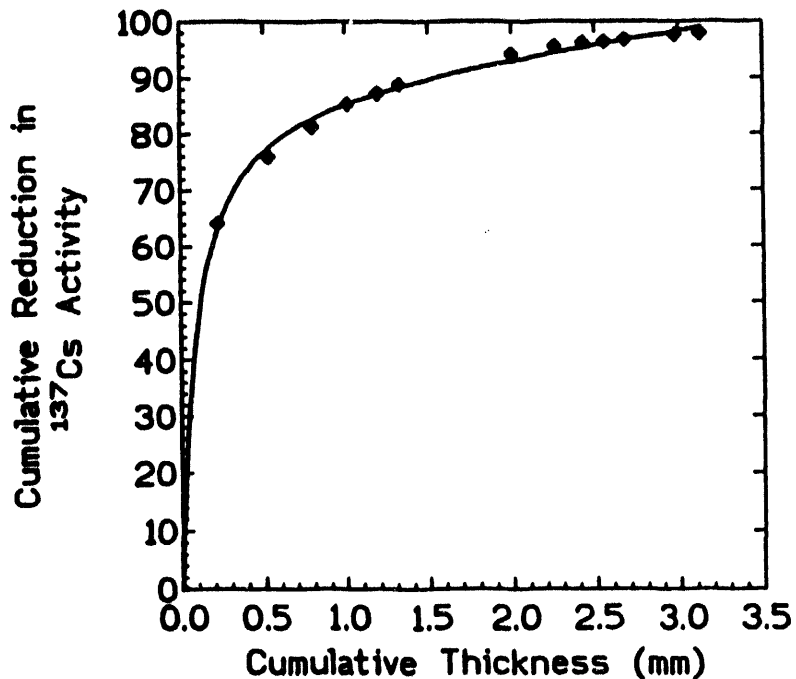


Table 7-6. Residual ¹³⁷Cs Activity of the Gravel Fragment as a Function of Cumulative Grinding Thickness.

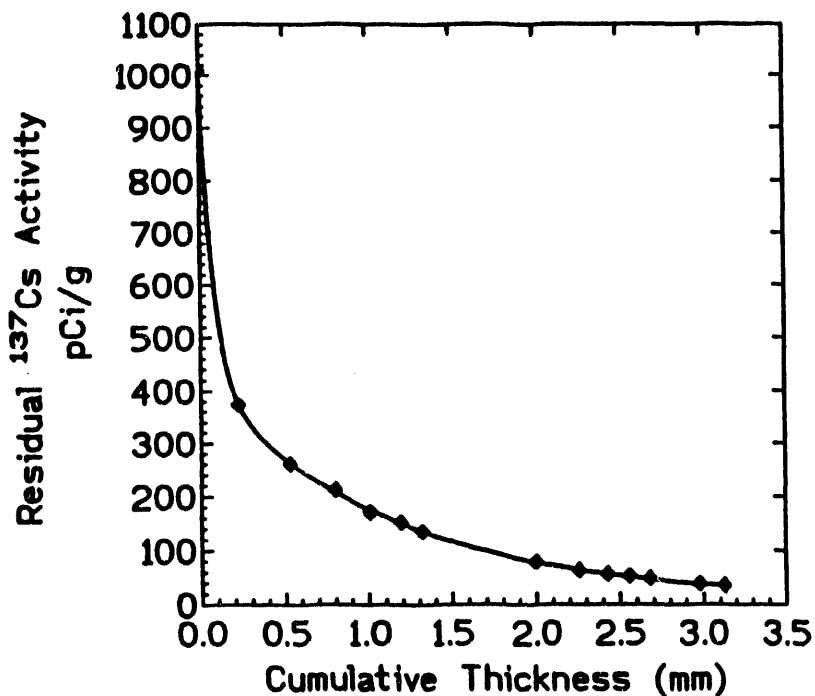


Table 7-2. Autogenous Surface Grinding Data for Gravel Fractions from 116-F-4 Pluto Crib Soil.

Cumulative Grinding Time (min)	Total Residual ¹³⁷ Cs Activity (pCi)	Cumulative % Reduction in Total Activity	¹³⁷ Cs Activity per Unit Mass (pCi/g)	Cumulative Wt % Fines
Experiment I (Water Medium)				
0	150449	0	186	0
20	39407	74	49	14.2
40	30304	80	27	19.4
60	19129	87	19	25.3
80	9635	94	10	31.4
Experiment II (Water Medium)				
0	525274	0	266	0
40	114070	78	66	12.7
50	45752	91	28	18.4
70	17466	97	12	25.7
Experiment III (Water Medium)				
0	824455	0	391	0
20	212412	74	120	16
40	102557	88	70	30.5
60	46515	94	35	36.9
Experiment IV (Electrolyte Medium)				
0	545874	0	403	0
20	274965	50	247	18.0
40	212106	61	205	23.7
60	164253	70	172	29.5
80	128635	76	146	34.7
Experiment V (Electrolyte Medium)				
0	701212	0	343	0
60	405897	42	243	18.3
80	330873	53	211	22.5

Figure 7-7. Relationship Between Reduction in ¹³⁷Cs Activity and the Mass of Fines Generated During Autogenous Surface Grinding.

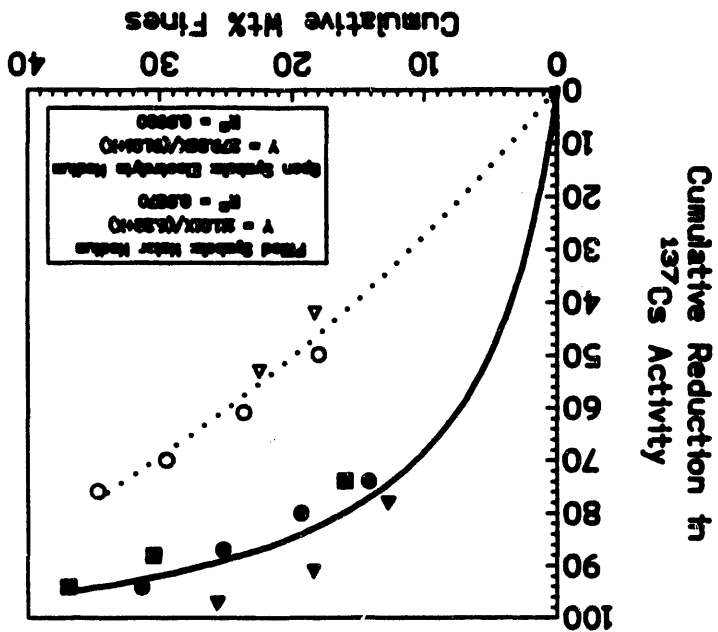
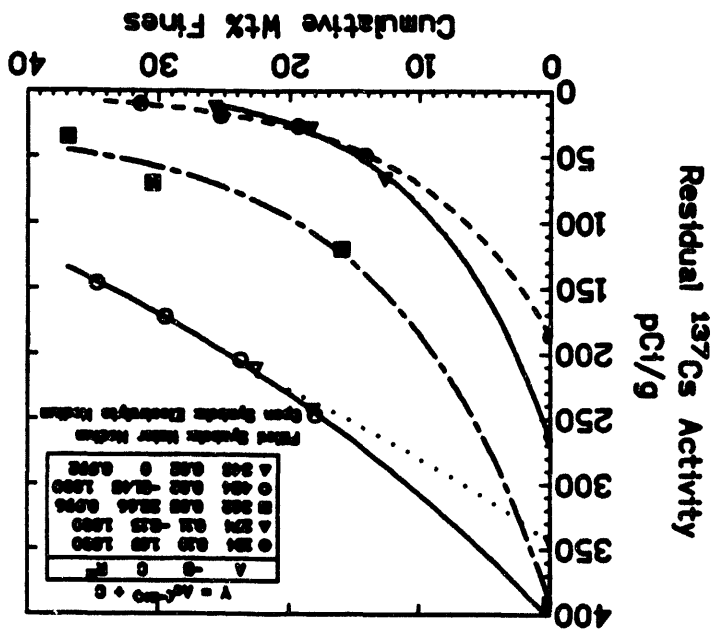


Figure 7-8. Relationship between the Residual ¹³⁷Cs Activity of Gravel Fractions and the Cumulative Mass of Fines.



The cumulative percent reduction, and the residual ^{137}Cs activities were also correlated with the computed cumulative thickness removed during grinding (Figure 7-9, 7-10). Again these relationships showed that major fractions (about 74% to 78%) of the total ^{137}Cs activity associated with the gravelly material in the 116-F-4 soil can be mobilized by grinding off the first millimeter of the surface in water medium. The relationships also showed that to mobilize comparable fractions of contaminant in the electrolyte medium, it was necessary to grind off almost 2.5-mm thickness of gravel surface.

Residual activities per unit mass in the gravel fractions appear to decline exponentially with cumulative thickness of the material removed by grinding (Figure 7-10). This grinding model appears to be valid for gravel material containing initial ^{137}Cs activities ranging from about 190 to 403 pCi/g. For gravel material containing lower activities (190 to 270 pCi/g), removal of about 1.5 to 2-mm layer through grinding in a water medium would result in residual activity at or below the TPG for ^{137}Cs . For higher activity gravel fractions (about 400 pCi/g of ^{137}Cs) to meet the TPG, approximately an additional millimeter layer needs to be removed by wet (water medium) surface grinding. These autogenous surface grinding tests showed that use of an electrolyte appears to be inimical to effective contaminant mobilization from gravel fractions. These data however need to be confirmed by conducting additional surface grinding experiments, perhaps also using other complex-forming electrolytes to unravel the unexpected trend. Based on these CB processor tests, it was apparent that most the effective removal of ^{137}Cs contamination from gravel fractions of 116-F-4 can be achieved when surface grinding is conducted in a water medium.

The gravel fractions from 116-F-4 soil comprising 80% of the mass contain ^{137}Cs activities ranging from 186 to 337 pCi/g (Table 5-1). The results from these autogenous surface grinding experiments showed that these gravel fractions can be treated effectively to meet the TPG goal. The effective CB grinding generates on average about 24% by weight of contaminated fines (on the whole soil basis) that needs to be disposed. The residual gravel fractions (about 56% of the total mass) would contain residual ^{137}Cs activities that meet the TPG goal.

Figure 7-9. Correlation Between the Reduction in ¹³⁷Cs Activity and the Cumulative Grinding Thickness for Gravel Fractions

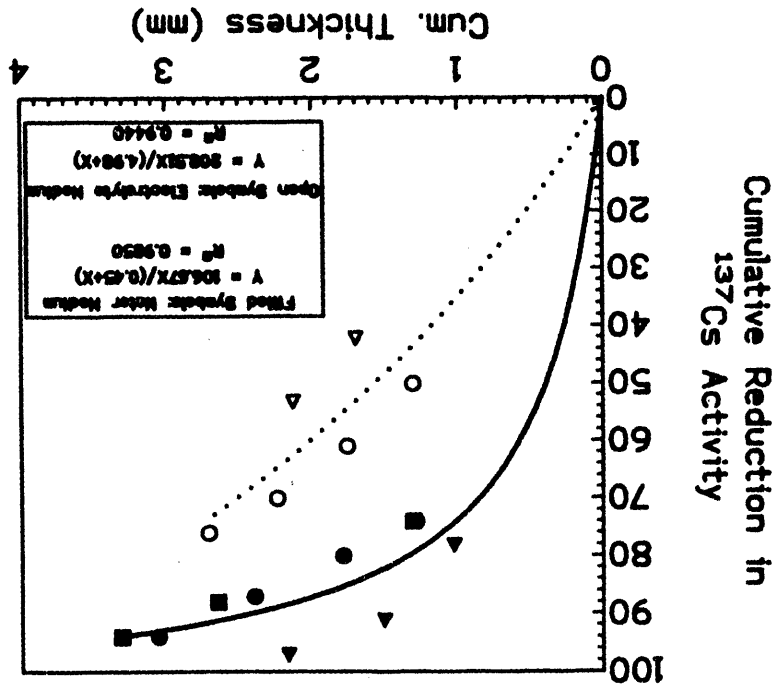
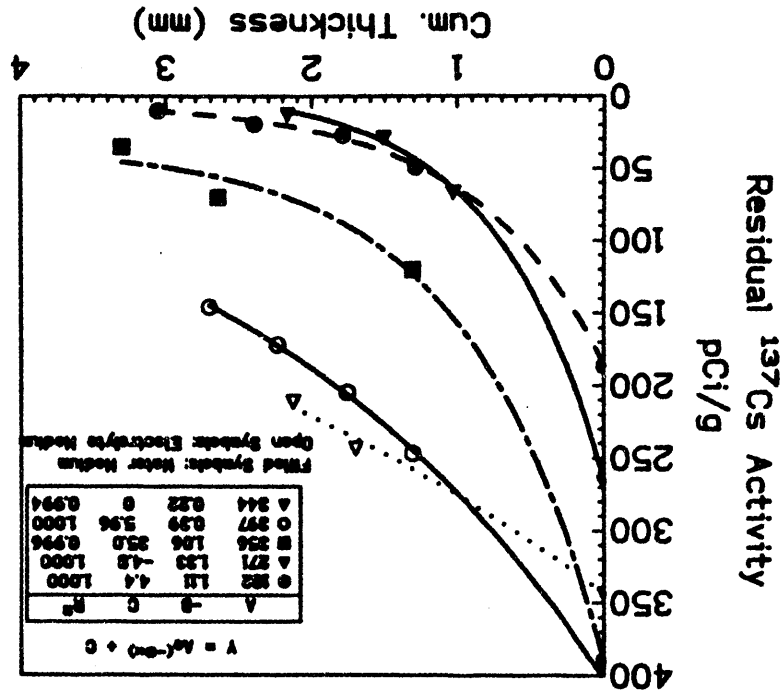


Figure 7-10. Residual ¹³⁷Cs Activity of Gravel Fractions as a Function of the Cumulative Grinding Thickness.



8.0 CHEMICAL EXTRACTION

8.1 OBJECTIVE

The objective of using chemical extraction was to evaluate the feasibility of removing contaminants from 116-F-4 Pluto Crib soil using chemical solutions. This operation could be used if physical treatment techniques such as attrition-scrubbing and autogenous surface grinding do not remove contaminants to required levels or as a replacement to soil separation in the event that chemical extraction of the entire soil volume is more economical. Depending on the soil and the levels of contamination, chemical extraction may also be an option for treating the fines generated during the physical treatment processes.

The chemical extraction of solids to selectively remove elements of interest is a proven technique used in the metallurgical and chemical processing industries for many years. The success of this technique generally lies in the proper selection of extractants (chemicals) and in understanding the kinetics of the reactions of concern. With this information, the proper selection of equipment to perform the extraction can be made and further scale-up studies can be conducted.

To process large quantities of soils, two main processing methods are available. The first choice is to use a stirred vat where contact of the leachant and soil can be easily controlled. The equipment for this process is relatively simple and can be scaled to handle very large volumes. The process can be operated in a continuous mode if a number of vats are used in series or operated in a batch mode with a single vat. The second possibility for leaching the soil is to add the extractant at the start of the soil-washing process (e.g., in the trommel) and to allow the reactions to take place while the physical separation is being performed. The advantage of this method is that very little equipment besides the soil-washing system is needed. However, the solids-to-liquid ratios and contact time in the trommel may not be optimal for the chemical extraction process to be effective.

As part of a previous soil-washing study, a number of chemical extractants were tested both at ambient and elevated temperatures for their effectiveness to remove ^{60}Co , ^{137}Cs , and ^{152}Eu from 116-D-1B soil from 100 Area of Hanford (DOE-RL 1993b). The extractants that were tested included acetic, citric, hydrochloric acids (that are typically used in chemical extractions of soils); three widely used reductive extractants (a neutrally buffered solution containing sodium citrate and sodium bicarbonate with sodium dithionite added as a reductant); a reductive acid extractant consisting of a mixture of acetic acid and hydroxylamine hydrochloride, and an extractant (consisting of a mixture of ammonium citrate and citric acid with sodium dithionite added as a reductant). Two additional extracts that were specifically formulated (but are currently being evaluated for patents) were also tested as part of this previous study. Also, the effects of parameters such as extract composition, contact time, and temperature were investigated to optimize the extractive process for the 116-D-1B (Batch III) soil.

The data obtained from these extraction experiments (DOE-RL 1993b) showed that the most effective extractant for removal of contaminant radionuclides such as ^{137}Cs from the 100 Area Hanford soils was a proprietary extractant (Extractant II) containing non-toxic, biodegradable organic compounds. Based on this information, all chemical extraction tests on sand-fractions (0.25- to 2-mm size fraction) of 116-F-4 soil were conducted with Extractant II.

8.2 EQUIPMENT AND PROCEDURES

The extraction tests were performed using a 1-L-capacity stirred vat system immersed in a temperature-controlled water bath. Tests were conducted at 96 °C by contacting 400 g of Extractant II with 100 g of 2- to 0.25-mm-sized fractions (sand fraction) of 116-F-4 soil for a set period, 6 hr. Extractions were conducted on sand fractions from wet sieving and the 0.25- to 2-mm fraction that had been attrition scrubbed in two stages with electrolyte. Following chemical extraction, solids were separated and wet sieved through a 0.25-mm sieve using deionized distilled water and oven-dried ($>0.25\text{-mm}$) at 105 ± 5 °C. The oven-dried solids were analyzed for the residual ^{137}Cs activity. The concentrations of extraction solutions were 0.25, 0.5, and 1 formality. A one formal solution contained one formula weight of the compound dissolved in one liter of solution.

8.3 RESULTS AND DISCUSSION

The extraction data (Table 8-1) showed that the lowest concentration (0.25 formal) Extractant II removed about 72% of the ^{137}Cs activity. Increasing the extractant concentration to one-half formal concentration released about 79% of bound ^{137}Cs with residual activity of 57 pCi/g in treated sand fraction. These extractions generated between 13% to 15% fines ($<0.25\text{-mm}$ fraction). Extractions conducted with a unit formal solution released approximately 84% of the initial ^{137}Cs activity resulting in residual activity of about 43 pCi/g. The data showed that in this soil, the fractional release of bound ^{137}Cs from the 2- to 0.25 mm size fraction material was a nonlinear function of the concentration of extractant (Figure 8-1). This functional relationship indicated that increasing extractant concentrations resulted in proportionately less incremental mobilization ^{137}Cs from the sand fraction of this soil. Such nonlinear relationship suggested that some fraction of ^{137}Cs in this soil was present in recalcitrant forms.

Chemical extraction of two-stage attrition-scrubbed sand fraction indicated that using 0.25 formal extractant II, ^{137}Cs activity could be reduced by 65% with concomitant generation of 7% fines. Extraction conducted at one-half formal concentration removed 76% of activity resulting in a residual ^{137}Cs activity (27 pCi/g) below the TPG of 30 pCi/g. These experiments implied that incrementally smaller amount of ^{137}Cs would be mobilized with extraction concentrations in excess of one-half formality. Thus the chemical costs would start to increase faster than the increase in ^{137}Cs removal.

These chemical extraction tests indicated that it was possible to reduce the residual ¹³⁷Cs activity in the sand fraction (2- to 0.25-mm-size fraction) of 116-F-4 soil first with two-stage attrition scrubbing with electrolyte followed by chemically extracting with one-half formal concentration of Extractant II. This combination of physical and chemical treatment results in an overall 87% reduction of ¹³⁷Cs activity and generation of about 18% fines (about 10% during attrition scrubbing and 9% during chemical extraction). The residual sand-size material contained 27 pCi/g of ¹³⁷Cs activity. The extraction test results provided a useful basis for selecting a combination of soil pretreatment and an extractant concentration to achieve different levels of residual ¹³⁷Cs in the sand fraction of 116-F-4 soil.

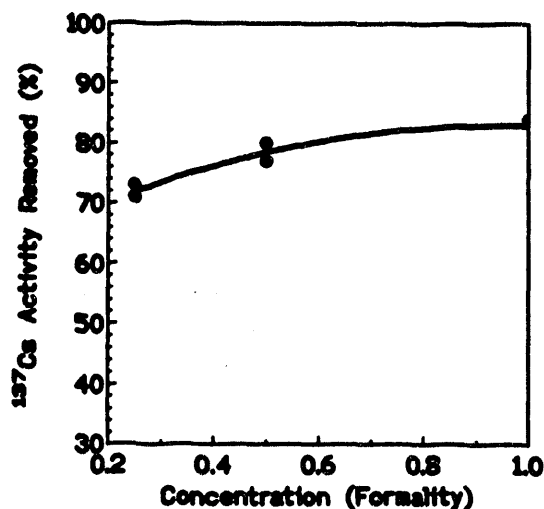
Table 8-1. Chemical Extraction Data for 116-F-4 Pluto Crib Soil (2- to 0.25-mm-Sized Fraction)^a.

Replicate #	Formal Conc.	¹³⁷ Cs (Final Activity) pCi/g	Reduction in activity %	Wt. % Fines
1	0.25	70	73	14.8
2	0.25	77	71	13.6
1	0.50	52	80	13.6
2	0.50	61	77	13.0
1	1.00	43	84	9.2
2	1.00	43	84	7.0
1	0.25	40	65 ^b	7.0
1	0.50	27	76 ^b	9.0

^aInitial ¹³⁷Cs activity was 260 pCi/g.

^bInitial activity of ¹³⁷Cs after two-stage attrition scrubbing with electrolyte was 114 pCi/g.

Figure 8-1. Relationship Between ¹³⁷Cs Activity (%) Removal and the Extractant Concentration.



9.0 WASTE WATER TREATMENT

9.1 OBJECTIVE

The overall cost and feasibility of using soil-washing techniques for the treatment of radionuclide-contaminated soils depend partly on the ability to treat the liquid wastes generated. The liquid waste streams may include water from operations used in the physical separation process (e.g., grizzly, trommel, screens, hydrocyclone), spent chemical solutions from chemical extraction operations (leachates for recycle or disposal), or water or electrolytes from attrition scrubbing operations.

One of the goals for developing an optimum soil-washing flow sheet is to minimize waste stream volume. Achievement of this goal requires that much of the water and chemical solutions be recycled. The concentrations of suspended solids in recycled water and solutions should be reduced to levels low enough to prevent detrimental impact on processing equipment such as spray nozzles and pumps. Typically, the suspended solid load in waste streams is reduced through settling and, if necessary, through flocculation. Additional processing of the streams may also be necessary to remove dissolved contaminant buildup in the recycle and bleed streams.

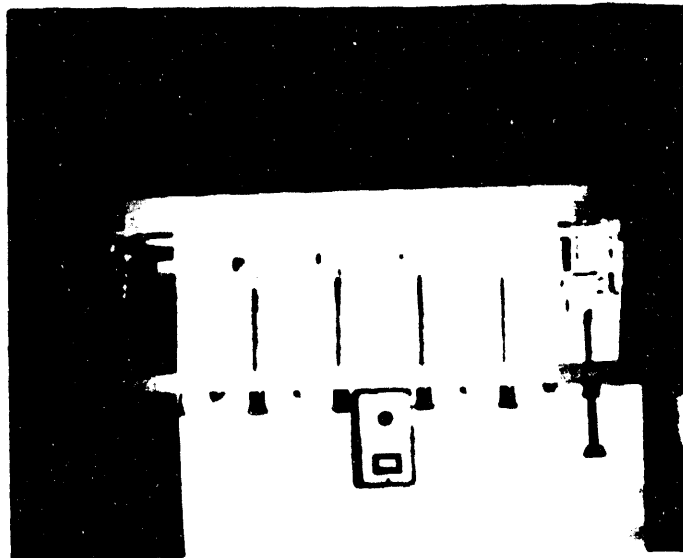
The objective of the water tests was to test the feasibility of removing suspended soil particles from liquid streams generated during bench-scale soil-washing tests conducted on 100-F-4 Pluto Crib soil. The treated waste stream may be recycled, evaporated, or may be discharged into soil hence, purge water criteria (WHC, 1988, Section 8.0) was used as a basis of comparing the quality of treated water. The waste streams included wash water generated during wet sieving, autogenous surface grinding, and electrolyte-laden wash water from two-stage attrition scrubbing. A set of flocculation tests was conducted on each of these waste streams. Measured parameters included turbidity, pH, conductivity, and activities of selected radionuclides remaining in clarified liquids.

9.2 EQUIPMENT AND PROCEDURES

The flocculation tests were conducted with a six-station Phipps and Bird batch reactor unit equipped with stainless-steel stir paddles (Figure 9-1). Turbidity measurements were conducted using a Hach Ratio/XR Turbidity meter calibrated with formazine standards. The waste solutions used in these tests were generated during soil-washing feasibility experiments conducted on 116-F-4 soil. These aqueous streams consisted of wash water generated from wet screening of <2-mm soil material through a 0.25-mm (60-mesh) sieve, electrolyte-laden wash solution resulting from two-stage attrition scrubbing of 2- to 0.25-mm fraction, and wash water resulting from autogenous surface grinding of gravel fractions. All waste solutions were thoroughly stirred and allowed to settle for 15 min to remove larger particles.

Six 250-mL aliquots were then removed from each of the waste solutions and placed in 500-mL beakers.

Figure 9-1. Phipps and Bird Batch Reactor Unit.



Several polyelectrolytic flocculation agents were used in these experiments. These polyelectrolytes consist of long-chain compounds with various types of ionic and nonionic groups and are used extensively in water purification and wastewater treatment. These polymeric compounds bring about flocculation by adsorbing and promoting bridging between suspended particles. One of the advantages of these polymeric flocculating agents is that they are effective over a broad pH range, whereas conventional flocculents such as ferric chloride or alum are effective over a relatively narrow pH range. Because of their effectiveness, a polymeric agent [CATFLOC-L, (a registered trademark of Calgon Corporation)] was used to treat actinide-containing pond waters from Rocky Flats (Triay et al. 1993). CATFLOC-L and a high-molecular weight anionic polymer [POL-E-Z-692, (a registered trademark of Calgon Corporation)] have been previously used to effectively treat wash waters from bench-scale soil washing tests on 116-D-1B trench soil (DOE-RL 1993b).

Preliminary flocculation tests were conducted on 116-F-4 washwaters using polymeric agents CATFLOC-L and POL-E-Z-692. However, the results showed that these polymers were not effective in significantly reducing the turbidity of wash waters from 116-F-4 soil. Therefore, a different set of polymeric agents were used to conduct the flocculation tests. After establishing a broad range of concentrations that effectively flocculated the wash waters, optimized tests were conducted to further narrow the concentrations of flocculents. These tests were conducted by allowing a initial 15-min settling period to remove the settleable solids and decanting the suspension. Next, the suspension was transferred to 500 ml beakers that were placed under the stirring paddles of the Phipps and Bird Testing Apparatus. After adding and mixing appropriate flocculents, the suspensions were allowed to

settle for 10 to 30 min before the supernatant was drawn off for measuring turbidity, pH, and conductivity. Aliquots were also removed to measure ^{137}Cs activities and concentrations of regulated constituents.

9.3 RESULTS AND DISCUSSION

The flocculation tests showed that a combination of AQUAFLOC 460 (a registered trademark of GRACE Dearborn) and AQUAFLOC 456C (a registered trademark of GRACE Dearborn) were effective flocculents for wash water generated from wet sieving (Table 9-1). After 10 min of settling, turbidity values as low as 18 nephelometric turbidity units (NTUs) were achieved for wash waters after addition of 50 mg/L of each flocculent. This represented about 96% reduction from initial turbidity of 500 NTU. Settling for 30 min did not significantly improve the turbidity values observed after 10 min of settling. The pH of the waste water was not significantly altered by the addition of flocculents. The conductivity values showed that the control and clarified wash waters contained concentrations of dissolved constituents that are typical of potable waters. The ^{137}Cs activity in the best clarified supernatant was measured at 174 pCi/L (same order of magnitude as the proposed MCL of 119 pCi/L, Federal Register 40 CFR, 141, 142) which was more than an order of magnitude lower than the purge water criterion of 2000 pCi/L. The activities of ^{60}Co and ^{152}Eu were both less than 300 pCi/L in this supernatant (purge water criterion for ^{60}Co is 1000 pCi/L and no criterion exists for ^{152}Eu). This optimized test showed that by adding polyelectrolytes AQUAFLOC 460 and 456C at concentrations of 50 mg/L each to the wash water it was possible to remove the bulk of the suspended solids and ^{137}Cs activity from these wash waters.

Significant reductions in turbidity (99.8%) were also observed when the electrolyte-laden waste water from two-stage attrition scrubbing was treated with a combination of three flocculents (Table 9-2). Turbidity values as low as 18 NTU were observed when the wash water with initial turbidity of 9540 NTU was treated with an optimum combination of 100, 50, and 150 mg/L of polyelectrolytes AQUAFLOC 2404, DEARTEK 2401 (both registered trademarks of GRACE Dearborn), and AQUAFLOC 460, respectively. Such reductions in turbidity (99.8%) indicate that almost all of the suspended particles can be flocculated in 10 to 30 min of settling after treatment. Use of these flocculents did not affect the pH of the treated waste water. The conductivities of control and treated wash waters ranged between 2800 to 2900 $\mu\text{S}/\text{cm}$ indicating relatively low concentrations of dissolved constituents. The activities of radionuclides ^{60}Co and ^{152}Eu in the best clarified supernatants were very low (< 300 pCi/L). However, the activity of ^{137}Cs in these AQUAFLOC-clarified supernatants (about 5600 pCi/L) exceed the purge water criterion of 2000 pCi/L. Therefore, the treated waste water from two-stage scrubbing with electrolyte needs additional treatment, such as ion exchange, to remove dissolved ^{137}Cs . These tests showed that it is possible to flocculate a major fraction of colloidal particles from the highly turbid waste solutions that result from

two-stage scrubbing (with electrolyte) of the sand fraction of 116-F-4 soil.

The wash waters resulting from autogenous surface grinding had very high concentrations of suspended particles (turbidity after 25 min settling ranged from 10640 to 35140 NTU). Using an optimum combination of flocculents (275 mg/L of DEARTEK 2401, and 40 mg/L of AQUAFLOC 456C) turbidities as low as 10 NTU (99.97% reduction) were achieved after 10 min of settling (Table 9-3). Addition of these flocculents did not significantly alter the pH of the wash water. The wash water before and after treatment had very low conductivity values indicating that the concentration of dissolved constituents in this waste water was similar to that of potable waters. The activities of all three radionuclides (^{137}Cs , ^{60}Co and ^{152}Eu) in the best clarified supernatant were 52, <100, and <300 pCi/L respectively. These levels of activities are more than an order of magnitude less than the respective purge water criteria and even below the proposed MCLs of 119, 218, and 841 pCi/L respectively. Clearly, these treated waste waters meet both the proposed MCL and the purge water criteria.

Wet sieving was conducted with water whereas, attrition scrubbing was conducted with an electrolyte. The more effective autogenous surface grinding tests were conducted with only water. This difference process water was reflected in both waste water characteristics and treatability. For instance, untreated waste waters from wet sieving and autogenous surface grinding had significantly lower turbidities, conductivities, and soluble ^{137}Cs activities as compared to the electrolyte-containing wash water from attrition scrubbing. Following treatment, the best supernatant from attrition scrubbing contained an order of magnitude, or more, higher dissolved ^{137}Cs activity than found in the best supernatants of the other two waste water types. It appears that among the three treated waste streams only the attrition scrubbing waste water needs to be treated further for dissolved ^{137}Cs removal.

These flocculation tests showed that it is possible to remove almost all of the suspended solids (96% to 99.97%) from the liquid waste streams generated from soil-washing of 116-F-4 soil using additions of commercially available polyelectrolytes. The activities of all radionuclides (^{137}Cs , ^{60}Co , and ^{152}Eu) in all treated waste streams (except ^{137}Cs in treated electrolyte containing waste waters from two-stage attrition scrubbing) were well below the purgewater criteria. These tests showed that additional factors need to be examined that include the effects of recycling effluent on the build-up of soluble contaminants and macroions and the use of ion-exchange or precipitation processes for radionuclide removal from attrition-scrubbing waste waters.

Table 9-1. Flocculation Data on Wash water from Wet Sieving 116-F-4 Pluto Crib Soil

Flocculent Conc. mg/l AQUAFLOC 460 + 456 C	Turbidity (10 min) NTU	Turbidity (30 min) NTU	pH	Conductivity (μ S/cm)	¹³⁷ Cs Activity in Supernatant (pCi/L)	⁶⁰ Co Activity in Supernatant (pCi/L)	¹⁵² Eu Activity in Supernatant (pCi/L)
Control	500	450	8.9	143	9340	< 300	< 200
30 + 50	40	40	7.9	140	--	--	--
35 + 50	35	36	8.9	171	--	--	--
40 + 50	23	24	9	192	318	< 300	< 200
45 + 50	19	19	8.9	197	202	< 300	< 200
50 + 50	18	18	9	185	174	< 300	< 200
55 + 50	34	33	9	122	237	< 300	< 200
60 + 50	52	52	9	131	--	--	--
75 + 50	47	44	8.9	125	--	--	--
100+ 50	236	224	8.8	157	--	--	--
50 + 40	26	--	9	128	--	--	--
50 + 45	21	--	9	183	--	--	--
50+ 50	18	--	--	--	--	--	--
50 + 55	27	--	8.9	184	--	--	--
50 + 60	25	--	8.9	133	--	--	--

Initial Turbidity: 4500 -5000 NTU (15 min settling)

Typical Conductivities: Distilled water: 4 μ S/cm, drinking water: 50 - 1500 μ S/cm.

Table 9-2. Flocculation Data on Wash waters from Attrition Scrubbing of Sand Fractions from 116-F-4 Pluto Crib Soil.

Flocculent Conc. (mg/l) AQUAFLOC 2404 + DEARTEK 2401 + AQUAFLOC 460	Turbidity (10 min) NTU	Turbidity (30 min) NTU	pH	Conductivity (μ S/cm)	¹³⁷ Cs Activity in Supernatant (pCi/L)	⁶⁰ Co Activity in Supernatant (pCi/L)	¹⁵² Eu Activity in Supernatant (pCi/L)
Control	--	9540	3.4	2840	23410	<200	<200
50 + 50 + 50	--	550	3.3	2910	--	--	--
100 + 50 + 50	--	450	3.4	2850	--	--	--
150 + 50 + 50	--	420	3.7	2710	--	--	--
200 + 50 + 50	--	390	3.8	2650	--	--	--
100 + 100 + 50	--	290	3.5	2770	--	--	--
100 + 150 + 50	--	240	3.5	2720	--	--	--
100 + 200 + 50	--	170	3.5	2750	--	--	--
100 + 50 + 100	--	54	3.5	2790	--	--	--
100 + 100 + 100	--	67	3.6	2680	--	--	--
100 + 50 + 150	--	18	3.5	2830	5682	<200	<200
100 + 100 + 150	--	19	3.6	2780	5550	<200	<200
100 + 150 + 150	--	20	3.5	2830	5725	<200	<200
100 + 100 + 200	--	23	3.6	2770	--	--	--
Control	7850	5160	--	--	--	--	--
100 + 50 + 150	25	22	--	--	--	--	--
100 + 100 + 100	20	17	--	--	--	--	--

Table 9-3. Flocculation Data on Wash waters from Autogenous Surface Grinding of Gravel Fractions from 116-F-4 Pluto Crib Soil.

Flocculent Conc. DEARTEK 2401 + AQUAFLOC 456-C (mg/l)	Turbidity (10 min) NTU	Turbidity (30 min) NTU	pH (10 min)	Conductivity (μ S/cm)	¹³⁷ Cs Activity in Supernatant (pCi/L)	⁶⁰ Co Activity in Supernatan t (pCi/L)	¹⁵² Eu Activity in Supernatant (pCi/L)
Control	10640	8460	7.8	143	--	--	--
40 + 50	270	260	7.7	142	--	--	--
50 + 50	170	135	7.7	108	--	--	--
100 + 50	150	148	7.8	110	--	--	--
200 + 50	95	86	7.7	146	--	--	--
200 + 100	174	--	--	--	--	--	--
Control	35140	25140	9.3	101	9538	<100	<300
200 + 50	1000	843	9	112	--	--	--
225 + 50	79	81	9	115	--	--	--
250 + 50	55	52	9	110	--	--	--
275 + 50	47	49	9	111	--	--	--
300 + 50	79	78	8.6	114	--	--	--
275 + 40*	10	9	--	--	52	<100	<300
275 + 50*	12	14	8.6	--	140	<100	<300
275 + 60*	12	12	8.9	117	--	--	--
275 + 70*	16	16	8.9	126	--	--	--

Sequence of addition were 2401 with 1 min stirring followed by 456-C addition with 1 min stirring except for samples marked with asterisk the stirring was conducted for 2 min after adding 456-C.

10.0 DISCUSSION AND RECOMMENDATIONS

10.1 CONTAMINANT RADIONUCLIDE IN 116-F-4 PLUTO CRIB SOIL

The sediment characterization data indicated that the principal contaminant in the 116-F-4 Pluto Crib bulk soil was ^{137}Cs . Cesium is an alkali element, and its chemical properties are similar to other elements of this group (Li, Na, K, Rb, and Na). All alkali elements are univalent cations, and their adsorption preference on mineral surfaces increases with increasing ionic radii ($\text{Li} < \text{Na} < \text{K} < \text{Rb} < \text{Cs}$). Because of larger ionic radius and less degree of hydration, Cs adsorbs with higher affinity than other alkali cations. Cesium ions are known to adsorb specifically on wedge sites of micas where they can substitute for the potassium ions that commonly occupy these interlayer sites. These specifically adsorbed cesium ions are hard to displace by any other cations except those with comparable ionic radii and hydration status. Ammonium ions, due to their ionic radii being similar to cesium, can effectively compete with cesium for the highly specific wedge sites. The data obtained on other 100 Area soils confirmed that attrition scrubbing of soil with ammonium ion-containing electrolyte released more cesium than that released when scrubbing was conducted with deionized water (DOE-RL 1993b). This observation is also supported by the enhanced removal of cesium from 116-F-4 soil achieved by the acid-ammonium acetate extraction as compared to the other extractive steps conducted as part of the sequential extraction process used in the sediment characterization phase of this study. Because the 100 Area soils are known to contain mica minerals, any treatment scheme (attrition scrubbing or chemical extraction) should include chemical components to mobilize as much of the cesium as possible that is specifically adsorbed on micaceous wedge sites that are present in the bulk rock matrices. Other radionuclides, ^{60}Co , $^{152,154}\text{Eu}$, ^{90}Sr , and Pu and regulated metals were present at relatively low concentrations relative to their TPGs.

10.2 RESULTS OF THE TREATABILITY TESTS

The sample of 116-F-4 Pluto Crib bulk soil contained ^{137}Cs as the principal radionuclide contaminant which was present in all size fractions at levels (> 186 pCi/g) well above the TPG. The bulk of the soil mass was present in gravel (78.7%) and sand-size fractions (12.9%). Therefore, to achieve significant mass recovery, these two size fractions need to be treated to reduce the burden of ^{137}Cs in this soil.

The treatability test that was conducted on gravel fractions consisted of autogenous surface grinding. Surface grinding tests were conducted on water-washed gravel fractions using a CB processor. The data showed that surface grinding in water medium was effective in reducing the residual activity of gravel fractions to about 19 pCi/g while generating about 24% (bulk soil basis) of contaminated fines.

The tests conducted on sand fraction (2 - 0.25-mm) of the 116-F-4 soil indicated that a two- or three-stage attrition scrubbing with an electrolyte resulted in residual ^{137}Cs activities that ranged from 75 to 114 pCi/g. However, a combination of attrition scrubbing in two-

stages followed by chemical extraction mobilized sufficient quantities of ^{137}Cs and brought down the residual activity to about 27 pCi/g.

Each of these effective treatment processes has a number of advantages and disadvantages. The advantages of two-stage attrition scrubbing with electrolyte include a grinding process that is conducted at ambient temperature and the scrubbing that is conducted at high pulp densities (about 79%); thus requiring only a small amount of electrolyte, and the processing time is relatively short (15 to 30 min). The disadvantages of this process include (1) the generation of contaminated fines that need to be disposed of, (2) the need to remove fines between each scrubbing stage to prevent reduction in scrubbing efficiency, and (3) process wash water with high content of suspended solids (turbidities between 5100 to 9600 NTU) and residual electrolyte that need treatment before recycling.

The principal advantage of the chemical extraction process is that the process can be optimized to remove cesium to the level required to just meet the TPG; therefore, the chemical costs can be reduced for specific circumstances. Second, the chemical extraction is a single-step treatment process that does not require any intermediate processing step. The principal disadvantages are that the extractive process is conducted above ambient temperatures (80 to 96 °C); the extractant solubilizes cesium so that the process effluent will likely require precipitation, solvent extraction, and/or ion exchange treatment (DOE-RL 1993b); and chemical extraction processes are generally more costly than physical treatment processes such as size-fractionation and attrition scrubbing.

The advantages of autogenous surface grinding are that the gravel particles grind each other without additional abrasive or extractants, and second, gravel fractions containing initial ^{137}Cs activities up to about 400 pCi/g can be ground effectively to reach very low residual activities (10 to 35 pCi/g). The principal disadvantages are that the fines generated during grinding may amount to as much as a third of the treated mass, and the wash waters from grinding treatment may reach very high turbidities (8500 to 25000 NTU) requiring treatment before recycling. It should also be noted that because of a limited number of experiments, factors that influence autogenous surface grinding such as consistency, uniformity of grinding, and energy requirements were not evaluated. These additional data would be needed to evaluate in detail the scale-up factors for conducting pilot or field-scale autogenous surface grinding.

These laboratory-scale test data agree with published data that a number of factors such as the physical, chemical, and mineralogical properties, and the type and level of radionuclide contamination control the choice and effectiveness of the various soil-treatment schemes. Previous data (DOE-RL 1993b) and the results of this study indicate that by choosing appropriate soil-washing schemes it is possible to treat different types of radionuclide-contaminated soils. As a final step, however, these successful soil-washing schemes need to be analyzed for their cost effectiveness as compared to other remediation alternatives.

10.3 SOIL WASHING PROCESS OPTIONS FOR 116-F-4 SOIL

As a result of these treatability tests, three physical soil-washing options were identified on the basis of particle-size distribution and levels of contamination in each particle size, the results of autogenous surface grinding, attrition scrubbing, and chemical extraction of sand fractions (2- to 0.25-mm size fraction) of 116-F-4 Pluto Crib soil. These processing options are listed in Tables 10-1 and 10-2. These tabulations show the mass and contaminant distribution in four size fractions; namely >2-mm (+10 mesh), 2- to 0.25-mm (-10 +60 mesh), 0.25- to 0.074-mm (-60 +200 mesh), and <0.074-mm (-200 mesh). Calculated average radionuclide activities in cumulative coarser and finer size fractions are also tabulated.

Without wet-sieving, all of the excavated material would need to be disposed irrespective of where the size-cut is made (Table 10-1). Further, wet-sieving data showed that ^{137}Cs activities in each of the size fractions exceeded the TPG by at least an order of magnitude. Therefore, wet sieving itself was not a process option.

The first processing option consists of wet-sieving followed by autogenous surface grinding of gravel-size (> 13.5-mm) material. Mass and activity calculations for this option showed that about 55% of the bulk soil can be recovered with an average ^{137}Cs activity of 19 pCi/g. The fines generated during grinding (about 24%) and the untreated mass of soil (roughly 21%) need to be disposed. This treatment option removes about 97% of the ^{137}Cs content from the bulk soil.

The second processing option includes autogenous surface grinding of gravels and two-stage attrition scrubbing (with electrolyte) followed by chemical extraction to treat the sand fraction (0.25- to 2-mm). The treated sand (about 11% of the mass) would contain residual activity of 27 pCi/g of ^{137}Cs . This option results in about 66% treated soil mass with an average ^{137}Cs activity of 20 pCi/g. About one third of the total soil mass (about 26% fines generated from treatment processes, and 8% untreated material) needs to be disposed. This option results in about 98% reduction in ^{137}Cs content in the bulk soil.

Among the two soil-washing process options for the 116-F-4 soil, the second option results in 66% recovery of potential backfill material. Only an additional 11% of treated soil is recovered in this option over the first process option, and this requires extensive treatment of the sand fraction by two-stage attrition scrubbing with an electrolyte followed by chemical extraction. Cost/benefits analyses would be needed to justify the additional treatment required in option 2.

10.4 RECOMMENDATIONS

The soil-washing treatability data collected during this study can provide a basis for designing a pilot-scale soil-washing test at the 116-F-4 Pluto Crib. However, a number of important questions need to be addressed further before, or in parallel with, the pilot-scale tests. One issue needing attention is the potential mobilization of regulated trace metals and

Table 10-1. Wet-Sieving Process (No Treatment) for 116-F-4 Soil.

Size Fraction	Size (mm)	Particle-Size Distribution (Wt%)			Average ¹³⁷ Cs Activities (pCi/g)			No Processing Option		Wet-Sieving Option											
		Wt. in Fraction	Cum. in Fraction and Finer	Cum Wt. Larger	In Fraction	In Fraction and Finer	In Coarser Fractions	Wt%	¹³⁷ Cs Activity pCi/g	10M Cut (2-mm)		60M Cut (0.25-mm)		200M Cut (0.074-mm)							
										Wt%	¹³⁷ Cs Activity pCi/g	Wt%	¹³⁷ Cs Activity pCi/g	Wt%	¹³⁷ Cs Activity pCi/g						
-200M	0.074	2.3	2.3	97.7	2620	2620	280	100 (ERDF)	331	19	556	6.1	1268	2.3	2620						
-40+200M	0.25	3.8	6.1	93.9	409	1268	271							81	279	93.9	271	97.3	280		
-10+60	2.00	12.9	19.0	81.0	219	556	279														
+10M	> 2.00	81.0	100.0	0.0	279	331															
Total		100.0																			

Table 10-2. Autogenous Surface Grinding and Attrition Scrubbing Options for 116-F-4 Soil.

Autogenous Surface Grinding w/Water (> 13.5-mm Fraction)	Wt %	¹³⁷ Cs Activity pCi/g	Autogenous Surface Grinding w/Water (> 13.5-mm Fraction) and Two-Stage Attrition Scrubbing with Electrolyte and Chemical Extraction (2- to 0.25-mm Fraction)	Wt %	¹³⁷ Cs Activity pCi/g
Contamination Reduction: 97% Fines generated + Untreated: 23.6% + 21.3%			Contamination Reduction : 96% Fines Generated + Untreated: 26% + 8.4%		
Autogenous Surface Grinding Feed (> 13.5-mm)	78.7	278	Autogenous Surface Grinding Feed (> 13.5 -mm)	78.7	278
			Two-Stage Att. Scrubbing & Chem. Extraction Feed (2- to 0.25-mm Fraction)	12.9	219
Fines generated from Autogenous Surface Grinding	23.6	883	Fines generated from Autogenous Surface Grinding	23.6	883
			Fines generated from Att. Scrubbing and Chem. Extraction	2.3	1104
To ERDF			TO ERDF		
-10M (<2-mm)	19.0	556	-60M (<0.25-mm)	6.1	1268
+10M-13.5-mm	2.3	291	+10M-13.5mm	2.3	291
From Autogenous Surface Grinding	23.6	883	From Autogenous Surface Grinding	23.6	883
			From Two-Stage Attrition Scrubbing w/Electrolyte and Chemical Extraction	2.3	1104
Total to ERDF	44.9	714	Total To ERDF	34.3	927
To BACKFILL			To BACKFILL		
> 13.5-mm from Autogenous Surface Grinding	55.1	19	> 13.5-mm Fraction from Autogenous Surface Grinding	55.1	19
			2- to 0.25-mm Fraction from Two-Stage Attrition Scrubbing & Chemical Extraction	10.6	27
Total to BACKFILL	55.1	19	Total to BACKFILL	65.7	20

radionuclides from the treated backfill material. The washing treatments, especially the chemical processes, may perturb the normally tightly bound trace constituents and make them more susceptible for long-term release. Therefore, extraction tests using the TCLP should be conducted on the treated backfill material during the pilot-scale tests.

The second issue relates to the data from laboratory-scale testing of autogenous surface grinding. Factors that need additional evaluation before pilot- or field-scale tests include consistency and uniformity of surface grinding, and retention times and energy requirements for pilot-scale rod or ball mills or other types of surface grinding units.

Another factor that should be examined concurrent with or prior to pilot-scale testing is the recyclability of aqueous waste streams such as wash water from particle classification operations and the very dilute electrolyte solution from attrition-scrubbing processes. It is necessary, therefore, to establish the number of times the treated liquid waste streams can be recycled before the buildup of contaminants and macroions in reused solutions prevents further recycling. These studies should include a bench-scale study of waste water and electrolyte recycling, the rate of increase in concentrations of all major cations and anions, and the contaminant radionuclide (^{137}Cs) in solution and in suspension during the recycling process. This study should also include treatability tests on bleed-off waste streams for removal of suspended solids and soluble radionuclides through flocculation, precipitation, and ion exchange. These tests are necessary because the cost effectiveness of soil-washing operations also depends on the recyclability of liquid waste streams. Such recycling and water treatment bench-scale tests are being performed on effluent from soil-washing tests on 116-D-1B (Batch III) trench soils (DOE-RL 1993b) in the fourth quarter of FY94. Further testing using 116-F-4 Pluto Crib soil may be required after results of the 116-D-1B wash water recycling tests are evaluated.

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Appendix A

Soil Washing Test Methods and Data Quality

SOIL WASHING TEST METHODS

The test plan (100 Area Soil Washing Treatability Test Plan, DOE, 1992) was developed to examine the soil washing treatability of 100 Area soils. This test plan was based on the general information regarding the types and concentrations of contaminants expected to be present in the 116-C-1 and 116-D-1B trenches. Based on the test plan, a detailed set of procedures were developed to conduct the characterization and a set of bench-scale tests were developed to incorporate any additional data that were obtained on these soils. For instance, during sampling of trench 116-C-1, it was found that the soil was coarse-textured with significant fraction of the material present as gravels. This information was incorporated into the test procedure by including autogenous surface grinding as one of the methods of treating gravel fractions. Autogenous surface grinding procedure using a centrifugal barrel processor was utilized to treat the gravel fraction of 116-F-4 Pluto Crib soil.

During the bench scale-tests, the test methods used closely followed the methods outlined in the test plan and the procedures with two exceptions. First, the test procedures included the Linear Density Gradient (LDG) method as a means of establishing specific contaminant-mineral associations. However, during the tests, it was found that the same type of information could be obtained by a combination of autoradiography and scanning electron microscopy with energy dispersive x-ray analyses (SEM-EDS). Therefore, these alternate methods were used to obtain necessary data. Second, during bench-scale tests, to assess the effectiveness of Cs removal static leaching at ambient and at high (96 C) temperatures was used on gravel fractions from 116-C-1 (Batch II). Based on the results obtained from these tests and the chemical extraction tests, it was concluded that heap leaching (usually conducted at ambient temperature for extended time periods) experiments would not offer any improvements in Cs removal performance. Therefore, heap leaching tests were not conducted on 116-F-4 soil.

