REMEDIATION OF PLUTONIUM-CONTAMINATED SOILS

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ABSTRACT

This paper reports on work done in 2004 to determine the effectiveness of paramagnetic separation to remove plutonium from soils from the Aldermaston (UK) site. The results showed that plutonium can be effectively concentrated in soils using magnetic separation. The work also attempted to enhance the separation process through alternative pre-treatment approaches, none of which were sufficiently effective to warrant continued study. These approaches were (1) the use of calgon to break clay bonds; (2) thermal oxidation to oxidize organically-complexed plutonium; and (3) ultrasonic vibration to break physical bonds between plutonium oxide and soils particles.

SUMMARY

An experiment was undertaken for AWE plc to determine the effectiveness of paramagnetic separation for removal of plutonium from soils from the Aldermaston site. The work, done by the Geosciences Advisory Unit of Southampton University and Terra Verde Services, Ltd., was performed in a hot laboratory at Southampton University.

The objective was to demonstrate that the soils could be treated to below 0.4 Bq/gm of plutonium and hence be eligible for free release.

The results of the work were as follows:

- A significant fraction of the AWE soils could be free release (ie, < 0.4 Bq/gm [0.01 nCi/gm] of plutonium) by size partitioning below 75 μm.
- Of the soils treated by paramagnetic separation, approximately 10% of the treated soil mass was separated by the magnetic process. This is in contrast to previous work at the Nevada Test Site using this technology which found that a significant fraction of the soil was captured in the magnetic separation process.[1]

[1] Information on previous work can be found in: "Evaluation of Remediation Methods for Plutonium Contaminated Soil" by S Hoeffner et al, Clemson Environmental Technology Laboratory, 2001; "Evaluation of Technologies for Volume Reduction of Plutonium-Contaminated Soils from the Nevada Test Site" by Papelis, et. al., DOE/NV/10845-57, 1996; and "Magnetic Separation for Environmental Remediation" by A. R. Schake et al, Los Alamos National Laboratory, LA-UR-94-3373, 1994.

- Paramagnetic separation was effective in concentrating plutonium in soil samples. The plutonium concentration in the magnetic fraction was found to be up to 8 times higher than in the feedstock.
- As a result of this work, engineering concepts are being developed which would enable more than 95% of the AWE soils to be treated for free release.

INTRODUCTION

AWE plc established a contract with the Geosciences Advisory Unit of Southampton University and Terra Verde Services, Ltd. to conduct hot tests on soils containing up to 1.9 Bq/gm of plutonium. The objectives of the project were as follows:

1. Using contaminated soils provided by AWE, test the effectiveness of size partitioning, paramagnetic separation, ultrasonic pretreatment, thermal pretreatment and washing using alternative reagents to determine their effectiveness for cleaning the soils.
2. Based on the results, develop engineering approaches which would enable at least 90% of the soils to be decontaminated to below 0.4 Bq/gm (0.01 nCi/gm).

The work was undertaken at the Oceanography Centre of Southampton University during April and May 2004. The results of the work are summarised in this paper.

Soils Characterisation

The work focused on three different soil types, labelled Batch 1, Batch 2 and Batch 3. The size distribution for these soils is shown in Figure 1. The plutonium distribution as a function of grain size is shown in Figure 2.

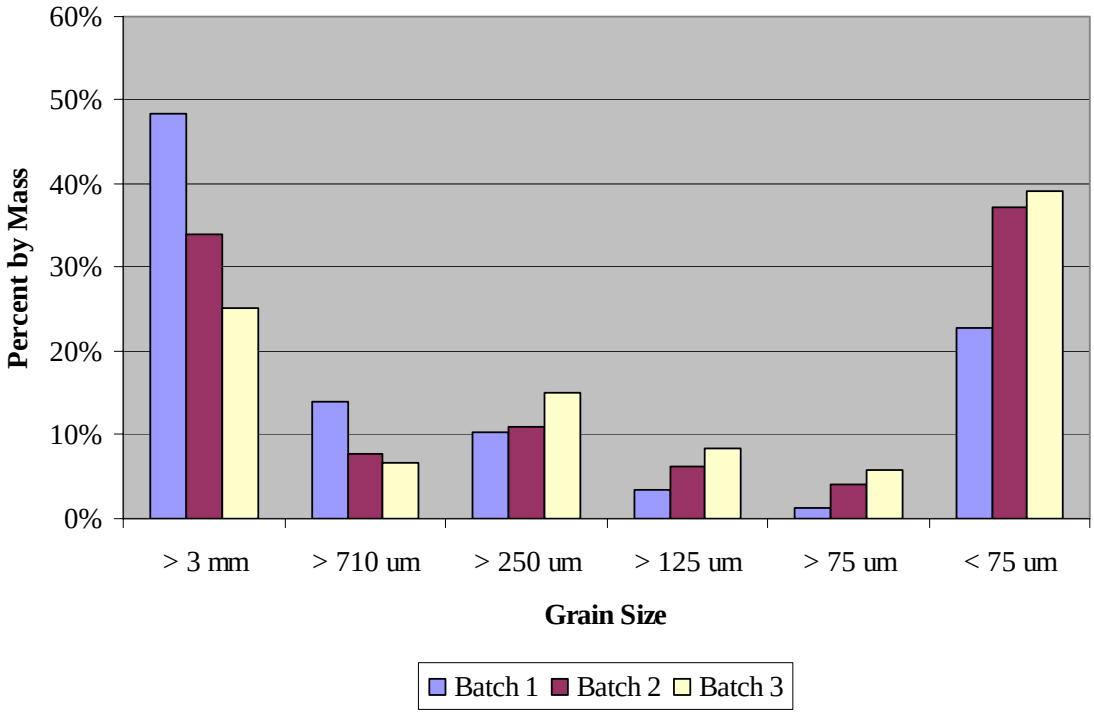

Fig. 1. Size distribution of AWE soils

Batches 1 and 2 were soils typically found in the Aldermaston region of England. They contain a significant amount of silt and clay. Batch 3 samples were taken from a river area and exhibited a wide variation in clay, sand and silt content. In addition, these samples contained a high fraction of organic matter.

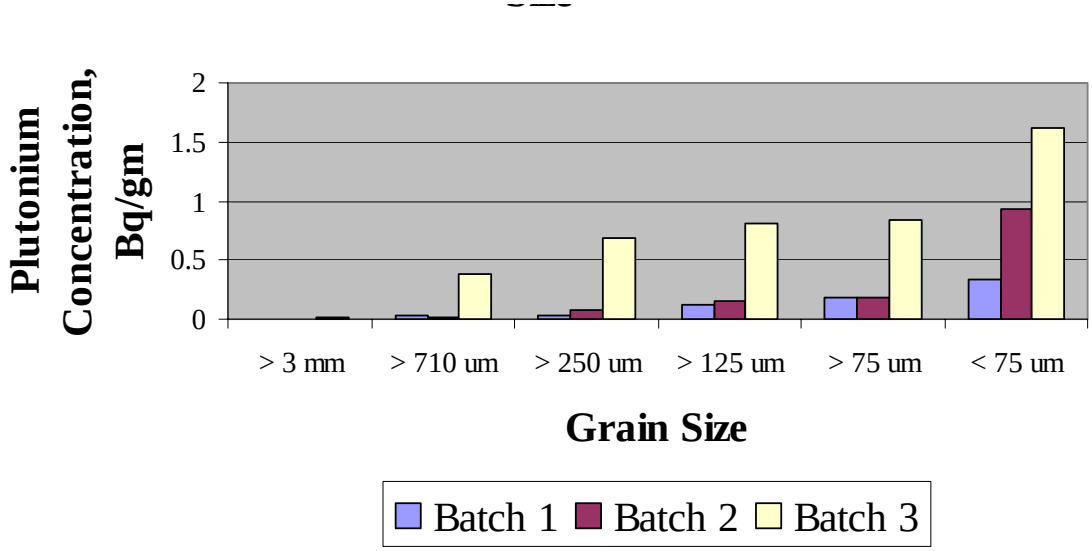

Fig.2. Plutonium concentration as a function of grain size

Magnetic Separation of Plutonium

Plutonium oxide is paramagnetic. Figure 3 shows that paramagnetic species have a linear magnetic response with varying magnetic field strength. It is this property which enables such species to be concentrated using very high magnetic fields generated by superconducting magnets.

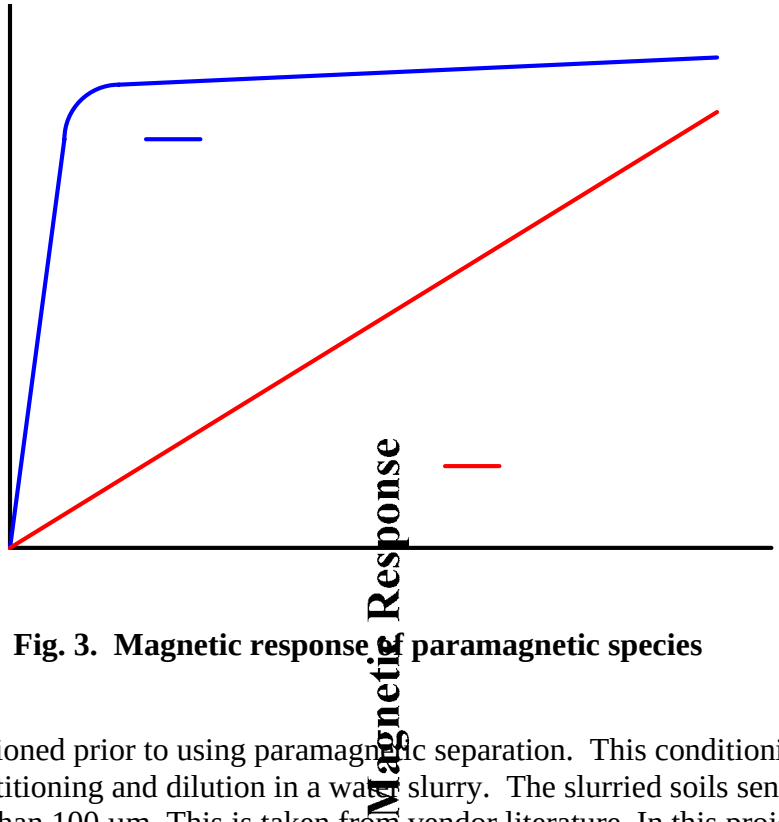

Fig. 3. Magnetic response of paramagnetic species

Soils must be conditioned prior to using paramagnetic separation. This conditioning involves, at a minimum, size partitioning and dilution in a water slurry. The slurried soils sent to the magnet should be no larger than 100 µm. This is taken from vendor literature. In this project, a 75 µm cut was used. Figure 4 shows the simplified flow diagram.

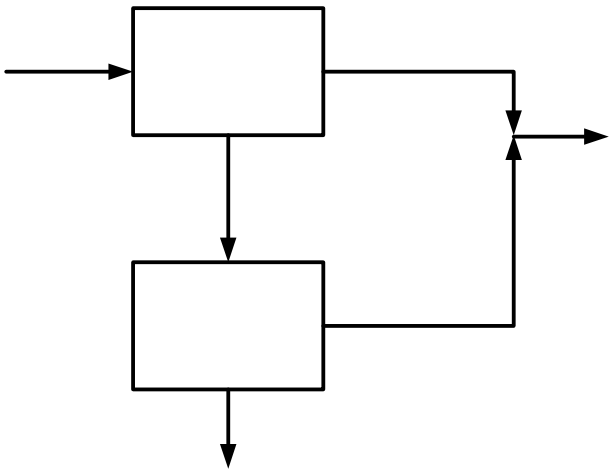

Fig. 4. Simplified flow diagram

The results of the magnetic separation are shown in Table I.

Table I. Magnetic Separation Results; 5 Tesla Magnetic Field

Test	Initial Mass (g)	Magnetic Mass (g)	Percent Magnetic	Feed[2] Pu Concentration	Magnetic Fraction Pu Concentration
B1 – a	494	40	8%	1.0	1.0
B1 – b	450	74	16%	1.0	1.5
B1 – c	391	28	7%	1.0	1.6
B1 – d	450	44	10%	1.0	2.4
B2 – a	396	23	6%	1.0	1.4
B2 – b	449	30	7%	1.0	2.5
B2 – c	476	53	11%	1.0	7.7
B2 – d	462	30	6%	1.0	4.1
B3 – a	328	14	4%	1.0	1.6
B3 – b	178	31	17%	1.0	1.8
B3 – c	349	34	10%	1.0	1.0
B3 – d	260	22	8%	1.0	2.0

Alternative Pretreatments

Three alternative pre-treatment approaches were used to determine if plutonium separation efficiency could be enhanced.

- Sodium HMP (Calgon): Sodium HMP was used to pretreat some samples. This compound is known to break the bonds of some clay particles. It was used to determine if the plutonium particles were physically bound in the clay particles. Visually, sodium HMP caused the < 75 μm clay particles to disperse. However, no benefit was observed for the magnetic separation.
- Thermal Pretreatment: It was suspected that some of the plutonium had formed organic compounds which are not paramagnetic. Samples were roasted in an oxygen environment to oxidize the plutonium. While this may have been accomplished, the AWE soils combined to form ceramic blocks. To continue would have required subsequent grinding which was judged to be inappropriate for economic and health and safety reasons. Hence the effort was discontinued.
- Ultrasonic Vibration: A slurried sample was subjected to ultrasonic vibration to determine if plutonium oxide particles could be dislodged from larger soil particles, hence making them available for magnetic separation. This experiment yielded no significant gains in separation efficiency and was discontinued.

All of these tests were done on samples of the Batch 1 and Batch 2 soils.

[2] Plutonium concentrations are reported here relative to a feed concentration of 1.0.