DEVELOPMENT OF AN ACTINIDE MAGNETIC SEPARATION SYSTEM FOR ENHANCEMENT OF THE TRUclean SYSTEM

Harry E. Plumblee, Jr.
Consultant

Ronald F. May, Thomas L. Tolt and Terry K. Wenstrand
Lockheed Environmental Systems and Technologies Company

F. Coyne Prenger, Walter F. Stewart and Dallas D. Hill
Mechanical and Electronic Engineering Division
Los Alamos National Laboratory

ABSTRACT

In a Cooperative Research and Development Agreement between Lockheed and Los Alamos National Laboratory, the participants are developing field-capable, pilot-scale hardware to transfer high gradient magnetic separation technology into production applications for decontaminating soils containing actinide compounds. In addition to applying magnetic separation to soil decontamination, the paper illustrates how this technology would be integrated into Lockheed's TRUclean soil remediation process. After summarizing a separator specification study, the paper discusses the design of a reciprocating canister system that will enable high duty cycle operation of the high gradient magnetic separator.

INTRODUCTION

Decontamination of soil containing heavy metals, radioactive materials, and hazardous organics is a subject of great interest, and many firms are using and/or developing remediation approaches for highly contaminated sites. These approaches include both physical and chemical methods, most of which have been developed during the past century for a variety of other uses. This paper deals with issues associated with physical separation methods. Many physical separation unit processes have been used, or are proposed for use, in separating contaminants from soil. Most of these are based on separation by particle size and specific gravity, and include screening, spiral classifiers, and centrifugal separators.

Lockheed Environmental Systems and Technologies Company (LESAT) has, as one line of business, the remediation of soil contaminated with radioactive and/or hazardous organic waste. As part of the remediation process, LESAT employs the TRUclean process, acquired when Lockheed purchased AWC, Inc., of Las Vegas, NV. The original patented TRUclean process is based on gravimetric separation using a mineral jig. This process was used for many years in, for instance, gold mining. Since acquisition of AWC, LESAT has greatly expanded the TRUclean system to encompass many other unit processes, including grinding mills, attrition scrubbers, vibrating screens, trommel screens, riffle sluices, centrifugal separators, multi-gravity separators, spiral classifiers, shaker tables, and super decanters. The TRUclean process also includes chemical separation methods, such as chemical leaching and countercurrent ion exchange. The size and location of individual unit processes is customized for each remediation job, based on soil and contaminant characteristics. This custom soil washing system is achieved by testing and optimizing a preliminary design at the LESAT Soils Treatability Laboratory. This Laboratory includes a pilot-scale version of each unit process used by LESAT.

In many situations, the highest concentration of contaminants is found in the size fractions of soil $\leq 50\ \mu m$ and, often, $\leq 10\ \mu m$. When this occurs the physical separation devices mentioned above lose effectiveness, and, in fact, useful decontamination may not be achieved without resorting to more expensive chemical methods. As an example, LESAT is currently involved in a remediation project for a site that has been contaminated with depleted uranium fragments and particles. At this site, 20-30% of the soil and contaminant mix is in the fine particle size range or slimes (particle sizes $<25\ \mu m$). Separation of uranium from these slimes is not possible by standard physical separation means. However, by utilizing the magnetic susceptibility of impurities, such as uranium compounds, physical separations can be achieved. This paper discusses a program to transfer magnetic separation technology out of the laboratory and other industries to the radioactive and mixed-waste remediation industry.

MAGNETIC SEPARATION BACKGROUND

High Gradient Magnetic Separation (HGMS) applies a strong magnetic field across a canister containing a loosely packed ferromagnetic matrix to selectively remove feebly magnetic particulates from multicomponent suspensions. The feed stream passes through a high magnetic field interlaced with high field gradients (created by the presence of the matrix). The magnetic force acting on solid particles is proportional to the product of the magnetic field, the field gradient, and the difference in magnetic susceptibility of the particles and the host fluid. Depending on whether the susceptibility difference between a particulate and water is positive (all ferro- and paramagnetic materials and some diamagnetic materials) or negative (many diamagnetic materials), a particle is attracted to different points on the surface of the matrix elements. Particle motions within the separator are governed by the combination of inertial, frictional, gravitational, magnetic and interparticle forces. In most instances, the absolute value of the magnetic force acting on diamagnetic materials (e.g., quartz, calcite, and alumina) is less than that acting on ferro-and paramagnetic materials (e.g., all iron and most uranium and plutonium inorganic compounds). By an appropriate balance of these forces, the ferro- and paramagnetic particles can be selectively captured on the surface of the matrix elements.

Only three common metals (iron, nickel and cobalt), many of their alloys, and a few number of compounds, e.g., ferrites, are ferromagnetic. Many compounds formed by the transition, rare earth, and actinide elements are characteristically paramagnetic, i.e., those materials having positive magnetic susceptibility as a result of unpaired electrons which can align in a *magnetic* field. HGMS has been used for many years in the mineral benefication industry. Iron ores, bauxite, and kaolinite clay are a few of the minerals which have been purified on an industrial scale using high gradient magnetic separators. In these cases the minerals collected in the separator include weakly magnetic oxidized taconite iron ore; ilmenite and the iron oxides, hematite and goethite, in bauxite; and iron-stained titanium oxide impurities in kaolinite clay. Recommended reviews of the use of HGMS (also called WHIMS or wet high intensity magnetic separation), as applied to mineral benefication, can be found in Oberteuffer (1) and Iannicelli and Murray (2).

At present, the greatest use of HGMS is in the kaolin industry. Kaolinite mined in the southeastern United States contains ferrous titanium oxides of $\leq 1\,\mu m$ particle size, which have a volumetric magnetic susceptibility of roughly 30×10^{-6} (SI). Kaolinite mined in Great Britain is purified by HGMS removal of 5 μm mica particles, to reduce iron content, and the removal of muscovite (which contains 1-2% Fe_2O_3), to reduce alkali content, as reported by Clark (3). These weakly magnetic impurities are removed from aqueous kaolinite slurries using both electromagnetic and superconducting magnet separators with bore diameters up to 2.1 and 3.0 m. Process rates of 36-55 metric ton/h of kaolinite are regularly achieved using 20 to 30 wt% slurries. The treated product has a significantly increased brightness for use in the paper industry after the impurity content is reduced by $\geq$50% with HGMS.

The application of magnetic separation to actinide removal from solid plutonium processing wastes was investigated at Los Alamos National Laboratory (LANL) by Avens and his colleagues (4,5) using two devices: a rare earth permanent magnet roll separator and an open gradient magnetic separator. Dry feed samples of graphite and thermite type reduction sand and slag of nominally $\geq 50\,\mu m$ were treated to remove plutonium residues from about 2 wt% in the feed to the 0.1-0.5 wt% range. The plutonium levels were low enough in the treated wastes such that they could be considered for direct discard without further processing.

Electrostatic forces prevent the application of conventional dry magnetic separation techniques to wastes with particle sizes $\leq 50\,\mu m$. In a slurry, surface chemistry can be carefully controlled to eliminate, or greatly reduce, interparticle forces. Wet magnetic separation of actinide contaminated soils should enable higher separation efficiencies to be met for plutonium and uranium compounds, even in the micron and submicron size range.

JOINT PROGRAM WITH LOS ALAMOS NATIONAL LABORATORY

Lockheed and LANL entered into a Cooperative Research and Development Agreement (CRADA) in February 1992 to help develop, and produce applications of high gradient magnetic separation technology. As part of the agreement, DOE/OTD will provide funding to the Los Alamos team, while Lockheed will provide the funding for its team members. The primary objective of the CRADA is to develop the necessary technology to field an HGMS system that will yield economically acceptable reductions in contaminated soil volume. The scope of this work involves developing a database of HGMS results, using both contaminant surrogates and actinide compounds mixed into water and into slurries containing selected well-characterized soil components and clean soils from contaminated sites. This database will be used to refine a separation performance model that relates various material and operating parameters to separation effectiveness. This model, and the results of the tests performed to date, were used to establish the design for the pilot-scale contaminant separation system discussed in this paper.

SUMMARY OF CRADA RESULTS TO DATE

The experimental program at LANL is established in three phases, further details of which are described in a companion paper by Avens, et al. (6). In the first two phases, experiments were performed using paramagnetic compounds such as CuO (which has a volume magnetic susceptibility of 240×10^{-6} approximately 2/3 that of PuO_2) as surrogates for actinide compounds. Small concentrations of these surrogates were mixed into aqueous slurries containing varying amounts of clay and silica solids. These tests were designed to measure the separator performance under a wide range of operating and feed conditions. Separation efficiencies in tests that used a slurry consisting of 4 μm mean diameter CuO at a concentration of 400 ppm in water with an appropriate surfactant passed once through an ultrafine steel wool at magnetic fields $\geq$2 T (tesla) were generally $\geq$98%. When a micron-size illite-beidellite clay was added to the CuO/water mixture at a concentration of 10 wt%, the first-pass separation efficiency for the copper oxide was approximately 97% in a test at an applied field of 8 T.

CHARACTERISTICS OF ELECTROMAGNETIC AND SUPERCONDUCTING MAGNETIC SEPARATORS

Wet high gradient magnetic separator systems have been under development since the 1930's. The designs have basically drifted toward the use of two types of magnets, electromagnetic or superconducting, and a flow-through separator canister filled with either magnetic stainless steel spheres, expanded metal screens, grooved plates or steel wool. An electromagnet is capable of producing a field up to 2 T. Superconducting magnets are routinely designed to produce fields of 8 T, and even greater field strengths can be achieved with attendant significant increases in the initial cost of the equipment. Large magnets, both electromagnetic and superconducting, are in common use, including several production solenoid-type superconducting magnets of 2.1 and 3.0 m bore diameter operating at 2 T in the kaolin industry.

A superconducting magnet operating at fields greater than 2 T will allow greater separation efficiencies through the linear contribution of applied field to the magnetic force if the contaminant of concern saturates magnetically at an applied field greater than the operating field of the magnet. Indeed, most paramagnetic minerals at room temperature exhibit a linear increase in magnetization, with an increase in field at fields well above 2 T. Consequently, the separation efficiency of the mineral loparite with a size distribution $< 10\,\mu m$ continues to increase nearly linearly with field in a superconducting separator as reported by Cheremnykh (7).

A superconducting magnet designed to produce a 7 T field costs approximately 50-100% more than a superconducting magnet designed to operate at 2 T (Stekly (8)). At a constant operating field, the primary cost of operating a superconducting magnet is the replacement or reliquefaction of the helium that is lost to boil-off because of heat transfer into the cryostat dewar from the environment. The He boil-off rate may be of the order of 0.2 to 1.0 liter/h. At a cost of approximately \$5/liter, LHe is quite inexpensive. However, He boil off increases dramatically as a result of joule heating in the nonsuperconducting electrical leads when the magnet is being charged or discharged. Therefore, it is desirable to leave the magnet at a constant field and to translate the matrix out of the high field to facilitate flushing of the captured contaminants out of the matrix. By contrast, electromagnets can be powered up and down repeatedly without additional power costs because these magnets require a constant supply of electrical power to operate at a desired field.

One approach to the operation of a superconducting magnet separator is to utilize a reciprocating canister system. One canister is positioned within the magnetic field, while the other is backflushed at a position outside of the magnet (9, 10, 11, 12). When the active canister reaches its capacity, it is shuttled out of the field, while the other is moved into the field. The dead time, or the time when the separator is not collecting contaminants, can be greatly reduced using this mode of operation. Sometimes it is necessary to first rinse the matrix with clean water (while operating at a high field strength) to rinse out nonmagnetic trapped soil particles before doing a high-velocity backflush at a low field to remove the captured contaminants. In this case, the separator downtime is reduced to the rinse time plus the time for canister translation. In the kaolin industry, Riley and Hocking (13) estimate that this type of dual-canister arrangement can be operated at a duty cycle of 50-60%.

APPROACH TO ESTABLISHING REQUIREMENTS AND SPECIFICATIONS FOR A PILOT-SCALE MAGNETIC SEPARATOR SYSTEM

A separator system specification study was performed to develop the design requirements for a pilot scale magnetic separator. This study was based on the comparison of a number of operational or functional requirements of the prototype system developed, in part, on experimental input from the first two phases of the CRADA work performed with LANL. These operational requirements were placed into two categories: mandatory and desired. A decision optimization technique, or trade study, that included considerations of system flexibility was then used to select an operating system.

Before selecting an HGMS system, the operational requirement criteria were defined. These criteria consist of the mandatory requirements that all design alternatives must meet and the desired requirements upon which the alternatives are scored as to determine their relative capability for satisfying these requirements. The selected mandatory requirements are listed in Table I.

A list of desired system requirements was compiled. A number of these requirements were essentially refinements of the *mandatory* requirements discussed above, and others were related to cost. Weighting factors that reflected the relative importance of each requirement in relation to all others were assigned to each of these desired requirements. Table II shows selected *desired* requirements along with their assigned weight factor (10 being the most important, 1 the least).

Design alternatives were identified considering both electro- and superconducting magnet-based systems and various canister system designs. The canister designs included those with either primarily radial or axial flow in the matrix. The alternative magnet designs and sizes were selected on the basis of current commercial availability or demonstrated ability to deliver custom designs on schedule. In this process, thirteen combinations of magnet and canister configurations were identified and evaluated, but an evaluation of each of these against the mandatory requirements eliminated seven configurations. The remaining six magnet/canister type configurations met every mandatory requirement. The radial canister systems were determined to be questionable for the backwash volume criterion. The six passing configurations included four electromagnet systems and two superconducting magnet systems, each of which was next evaluated against the desired characteristics. The alternative that had the best score for a given desired requirement was given a value of ten. The rest were ranked accordingly in sequentially decreasing numerical order. Total scores were determined by the summation of individual requirements.

TABLE I
Mandatory Criteria for Pilot-Scale HGMS Separator

- the ability to process solid particles $\leq 75\ \mu m$ in slurries 5-20 wt% solids
- a slurry processing capability of $\geq$11.4 L/min (3 gal/min)
- an operating duty cycle > 75%, where duty cycle is defined as the percentage of operating time at which the feed slurry is flowing through the matrix for magnetic collection of contaminants (or > 50-60% if a rinse period of the matrix at the operating field is necessary prior to the matrix backflush outside of the field)
- A volume of the contaminant waste stream $\leq$10% of the volume of the initial soil; a magnetic field that has rapid ramp up/down, or a canister system capable of shuttling in and out of the field without magnet shutdown
- a magnetic bore with a 40 cm minimum axial length in which the field is $\geq$1/2 of the peak field
- the magnet and operational support equipment proved to be rugged for field operations
- a separator that will interface with slurry preprocessing equipment, [i.e., the TRUclean pilot-scale system (0.2 m3 {1/4 yr^3} soil-processed/hr)]
- capital and overall operating costs that are less than that of competitive processes, such as chemical leaching.

Adverse consequences were considered to account for the probability of failure in the selection of a proposed separator system. The adverse consequences considered in the analysis were the inability to accommodate higher throughput rates, the inability to maximize slurry residence time in the matrix, and the inability to process slurries of high solids content while maintaining high duty cycles of operation. The overall result of this analysis, considering mandatory and desired requirements and adverse consequences, was the selection of an 8 T superconducting magnet system with a 15.2 cm (6 in.) warm-bore diameter operated with a reciprocating dual canister system. A diagram of the selected system type is shown in Fig. 1. An RFQ package was submitted to selected vendors for bid, and these bids have been received for evaluation.

SEPARATOR CANISTER DESIGN AND EVALUATION

A dual canister system has been designed for use in pilot-scale tests. This system was designed for use with the a 15.2 cm diameter, vertically oriented, warm-bore superconducting magnet discussed above. The canister system shown in Fig. 2 consists of two identical aluminum canisters, adjustable in length. These canisters are designed to hold ferromagnetic matrices and are joined end to end with an aluminum tube connector, which is also adjustable in length. Each canister holds an annulus-shaped, loosely packed ferromagnetic matrix, either 10.1 or 30.5 cm long (1.5 or 4.4 L volume, respectively). The matrix is contained between a 14 cm i.d. housing and a 3.3 cm o.d. aluminum pipe that extends along the canister axis. The central pipe terminates with an open end, a short distance in front of a cusp, that is machined into an aluminum end cap to deliver the slurry into the matrix. This pipe extends beyond the length of the canister system, well outside the magnet. The slurry exits the canister after passing through the matrix through a tee-fitting, which is joined to a second pipe extending outside of the magnet. This configuration allows one to pump the slurry through the matrix in one direction when the canister is positioned in the magnetic field, then to reverse the flow to back-flush the contaminants out of the matrix when the canister is out of the magnetic field. The second canister at the opposite end of the system has the same configuration, but is reversed so that its inlet and outlet connecting pipes extend outside the opposite end of the magnet bore.

To eliminate the creation of large stresses in the magnet coil when the canister train is raised and lowered inside the bore (in some instances this can lead to a rapid transition of the magnet from the superconducting to nonsuperconducting state) it is necessary to provide a uniform axial distribution of ferromagnetic alloy along the length of the canister train. Thin magnetic stainless steel sleeves are positioned at the ends of the canisters where the ferromagnetic matrix is absent. The aluminum tube, which acts as a connector of the two

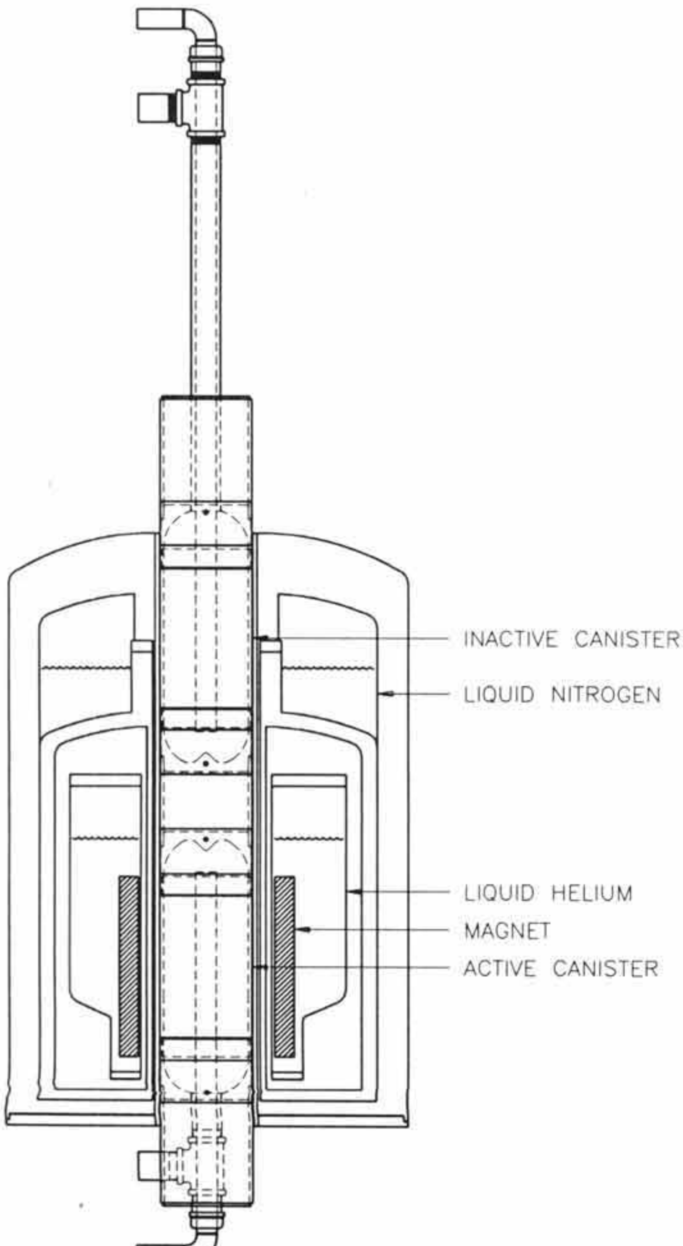

Fig. 1. Schematic of dual canister system with 12" long matrix compartment: installed in a 6: diameter warm bore superconducting magnet.

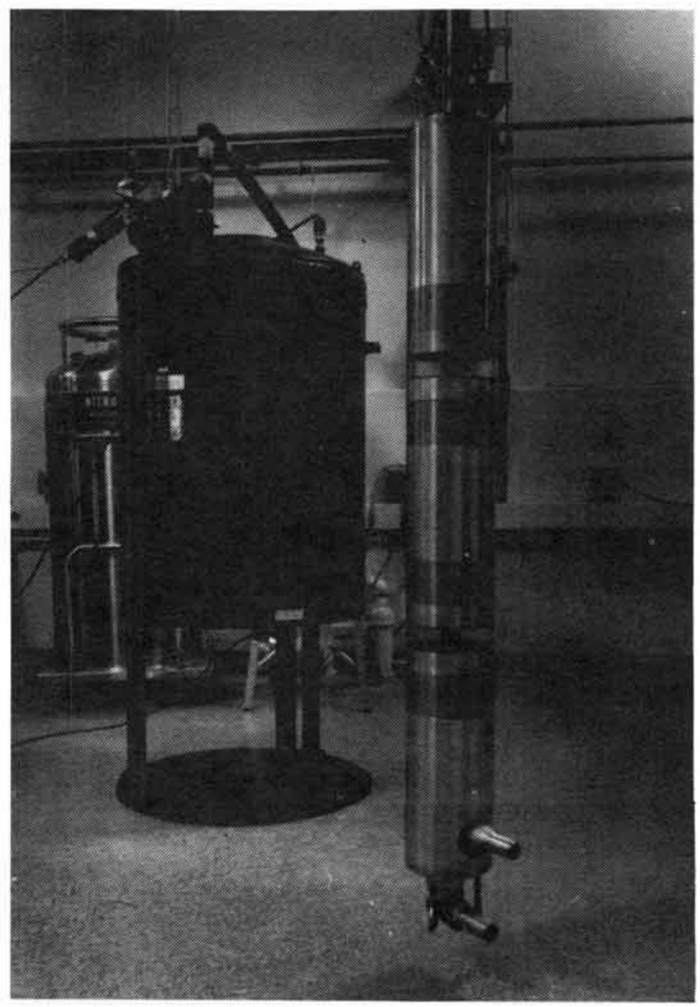

Fig. 2. Dual canister train with 10 cm long Plexiglass matrix housings shown with 8 T, 15.2 cm diameter warm bore superconducting magnet.

TABLE II
Desired Criteria for HGMS Separator System

Weight Factor	Requirement Description
10	Minimize capital costs
10	Minimize power costs (incl. costs of cryogenic fluids, if applicable)
9	Ensure that the axial field is ≥2 T for a distance of 50 cm
8	Minimize time needed for power shutdown and ramp up
7	Ensure system's ability to accommodate dual canister operation
6	Ensure that matrix is rugged enough to handle high flow rates and pressures countered during backflush period
6	Maximize system availability and reliability
5	Ensure that maximum magnetic field strength is ≥5 T
4	Provide instrumentation for radiation verification of effluent stream
2	Ensure that holding or surge tank circuit will enable recycle passes at reduced solids content in slurry

independent canisters, is filled with a dummy ferromagnetic matrix at the same packing density as that of the active matrices.

A series of tests has begun in which slurries are prepared from clean soils obtained from various DOE sites. These tests are being done to evaluate flow behavior in the dual-canister system without an applied magnetic field. The susceptibility of different types of matrices and the susceptibility of void fraction to clogging will be determined using pressure drop measurements. In addition, characteristics of the backflush operation, including fluid flow velocity and volume and air sparging, are under study. From these studies, we plan to develop techniques that can be used to minimize the volume of the collected contaminant waste.

FUTURE PLANS

As the CRADA experimental program is concluded in FY 1994, several tests will be conducted with the pilot-scale separator system, including magnetic separation tests, using cold soils from locations near radioactively contaminated soils that have been spiked with various actinide oxides. These tests will be followed with magnetic separation tests at the LESAT laboratory in Las Vegas using radioactively contaminated soils from several locations. The tests will be conducted using the magnet system discussed herein, following the acquisition and checkout of the magnet. The longer term goal is to use the pilot scale magnetic separator system in field tests, in conjunction with other components of the TRUclean system. These tests will be scheduled with the appropriate agencies, following validation data from the laboratory tests discussed above.

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