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SOIL AND WATER DECONTAMINATON BY EXTRACTION WITH SURFACTANTS

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ABSTRACT

Samples of soil from contaminated sites and industrial sludges and residues were treated in the laboratory by extraction with water solutions containing surfactants and other additives. The contaminants included coal tar, PCBs, hydrocarbons, chlorinated hydrocarbons, nitrotoluenes and heavy metals. Some of the samples contained highly complex mixtures of organic and heavy metal contaminants. Reduction of the contaminant levels to less than 1 ppm was typically attained by the extraction treatment. The contaminant-laden wash liquors were purified by ultrafiltration, followed by air flotation. The cost of treatment by a commercial-scale, integrated process, consisting of the extraction and wash liquor purification steps, was estimated to be in the range of $50 - $80 per ton of treated soil, depending on soil matrix properties, chemical composition of the contaminants, and other site-specific factors.

INTRODUCTION

The method of soil and water decontamination reported here involves a novel separation process (the GHEA Process), which is based on the solubilization of organic and inorganic contaminants by a water solution containing selected surfactants and other additives, followed by steps to separate the contaminants and the surfactants, respectively, from the wash solution.

The GHEA process is suitable for on-site and in-situ applications. Wastewater, as well as water associated with sludges and sediments, is treated to high purity, and may be discharged to the local waterway or sewer. The surfactant used for solubilization is fully recovered and recycled to the process. The contaminants fraction is isolated as a concentrate, which may be reclaimed or disposed of, depending on its composition. No other effluents or residues are produced by the GHEA process.

BACKGROUND

Soil washing is a method of treatment based on extraction with water, or water plus additives, such as surfactants, chelating agents, acids and alkalis. The method is largely in the developmental stage (laboratory and pilot scale) in the U.S., with some commercial projects reported in Europe.

Solubilization of organic and heavy metal compounds in water with the aid of surfactants and other additives is well known. In treatment of contaminated soil in the laboratory, it is generally possible to achieve any target level of separation by adjusting the achieve any target level of separation by adjusting the concentration of additives, increasing the wash liquid to soil ratio, repeated washings, etc. In industrial operation, however, there is a problem with disposing of the wash and rinse liquids, which contain the additives as well as the impurities. The volume of the wash and rinse liquids may be 10 - 20 times as large as the volume of the treated soil. In the case of highly toxic impurities (such as PCBs), it is essential to have a cost-effective means for purification of the wash and rinse liquids, in order to be able to discharge or reuse the water. It is also necessary to recover the surfactants for repeated use, and to isolate the impurities as a highly concentrated fraction, which is thereby amenable to reclamation or disposal at low cost, due to its correspondingly small volume.

The status of soil washing applications has been reviewed in recent publications and meetings [1,2]:

- The Heijmans process (Netherlands) extracts organics and metals from excavated soil. The organic pollutants are then separated from the wash liquor by air flotation, and the metal impurities by precipitation.

- The HWZ process (Netherlands) extracts organics and metals from excavated soil. The metals are separated from the wash liquor by precipitation, and the organics by carbon adsorption.

- The Harbauer process (W. Germany) extracts organics from excavated soil. The impurities are separated from the wash liquor by air flotation followed by carbon adsorption.

- The TAUW process (Netherlands) removes Cadmium from soil by in-situ acid leaching. The metal is separated from the leachate by on-site ion exchange.

The use of chelating agents such as EDTA to extract metals from soil has been described by Ellis et al. [3]. Reductions in the range of 22% to 100% are reported for various metals.

Extraction of metals by mineral acids is widely practiced for ore beneficiation in the mining industry. Studies on the application of hydrometallurgical techniques for separation of lead and arsenic at Superfund sites were reported by Schmidt et al. of the U.S. Bureau of Mines [2]. The extracts were treated by precipitation and air flotation. Cleanup of radionuclides (uranium, radium and thorium) by acid extraction, followed by precipitation and air flotation, is also described in an EPA technology review [4].

Soil washing may also be applied in situ, i.e., without excavation. The general design of in-situ soil flushing systems is described in a recent EPA publication [5]. The cleaning solution is sprayed on or injected into the contaminated zone. A system of extraction wells is installed downstream of the underground aquifer to collect the leachate. The leachate is pumped to an onsite facility which removes the contaminants prior to reinjection. Several in-situ projects and studies are reported:

- Soil flushing of organics with surfactants at the Volk Air Base in Camp Douglas, Wisconsin.
- Herbicides flushing with water in Sweden.
- Combined soil flushing with biodegradation for remediation of wood-preserving sites.

The technical approach of the GHEA process is based on the use of selected surfactants in water solution to extract the contaminants from the soil. The resulting wash liquor is purified by separating the surfactant/contaminant complex from the bulk of the wash water. The complex is then split into two fractions: a surfactant fraction which is recovered for repeated use, and a highly concentrated contaminants fraction which may be reclaimed, used as fuel, or treated as appropriate.

The solubilizing capability of surfactants is related to the presence of both hydrophilic and lipophilic groups within their molecular structure. This enables surfactants to interact with both polar and non-polar substances. The mechanisms of interaction between surfactant and solute molecules in water have been investigated by Shimamoto et al. [11,12,13]. Accordingly, the main form of interaction with non-polar solutes is micellar encapsulation, since the interior of a micelle provides a non-polar micro-environment. The interactions with polar and ionic species are primarily through hydrogen bonding and electrostatic forces.

The solubility of surfactants in water is a function of temperature, pH and ionic strength [6,7]. The GHEA process separates the surfactant/contaminant complex from the wash liquor by phase separation under controlled pH and temperature.

Surfactant molecules tend to concentrate and form films at gas/liquid and solid/liquid interfaces. This property has been used in large-scale beneficiation of metal ores by air flotation. Recent studies have shown that it is possible to separate metal solutes out of water by air flotation down to 0.1 ppm levels [8,9,10]. The GHEA process employs air flotation in a novel combination with ultrafiltration to achieve complete recovery of the surfactants from the wash liquors and reduction of dissolved metals to ppb levels.

Separation of organic solutes from the surfactant/contaminants complex is effected by steam stripping and extraction with organic solvents. The extract is then distilled to reclaim the solvent.

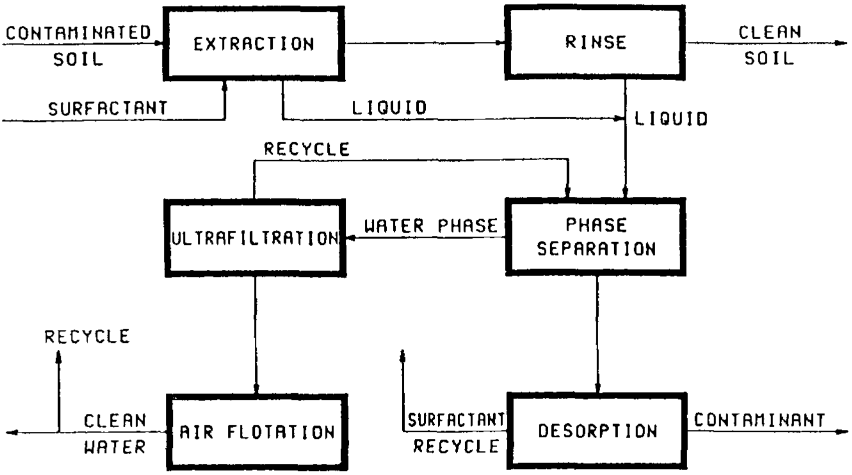

FIGURE 1: THE GHEA SOIL WASHING PROCESS

PROCESS DESCRIPTION (Figure 1)

The GHEA Process employs both ionic and nonionic surfactants of the following classes: fatty acid sulfates, alkylaryl sulfates, alkylaryl ammonium halides, alkylphenyl ethoxylates and ethylene oxide/propylene oxide block copolymers. Nonionic surfactants are effective in adsorbing both polar and non-polar organic solutes, and ionic surfactants adsorb counterion solutes.

Treatment of excavated soil begins by stirring the contaminated soil in the surfactant solution under controlled time, temperature and pH conditions. The composition and dosage of surfactants are functions of the types and levels of contaminants present in the soil. The extraction step is followed by filtration and rinsing with recycle water in order to remove all traces of the surfactants from the soil.

Following the extraction step, the wash liquid is separated into a water phase and a dense surfactant phase. The phase separation is effected under controlled temperature and pH conditions which are specific to the surfactant formulation.

The water phase is treated by a combined ultrafiltration/air flotation process which removes the residual surfactant. Metal solutes are separated out of solution in the air flotation step under controlled alkaline conditions in the range: $8 < pH < 11$. This combined process has been developed by Ghea Associates. The operation is simple and highly effective.

Separation of organics from the surfactant is effected by steam stripping and solvent extraction. The solvent is recovered by distillation and the surfactant is returned to the process.

The current status of process development is as follows:

- Treatability tests have been conducted on soil and water samples containing petroleum hydrocarbons, polynuclear aromatic compounds, volatile organic compounds, PCBs, chloro-organics, nitroaromatics and heavy metals. The results indicate exceptional separation efficiencies and a broad range of applicability to mixtures of organic and inorganic contaminants (See Table 1).

- Engineering and cost evaluations of the process indicate that the cost of treatment is in the range of $50 - 80 per ton of soil. This compares favorably with other separation processes which have been estimated to be in the range $90 to $200/ton.

- A pilot plant consisting of a 25-gallon extraction unit and downstream separators has been installed at the Hazardous Substance Management Research Center (HSMRC) laboratories in Newark, New Jersey. The pilot plant is designed for complete treatment of soils, sludges, slurries, and contaminated water, and uses equipment and systems which are directly scalable to commercial operations. The pilot plant is operated under the U.S. EPA Emerging Technologies Program.

- In-situ soil flushing tests have been conducted on undisturbed soil samples from a contaminated site in Northern New Jersey. The 6-month tests have shown that soil flushing with surfactants enhances the leaching rate by a factor of 15 to 20, relative to water flushing. The leachate is treated by the Ghea water purification process to a degree which allows recycle of the water to the soil flushing operation. The surfactant is separated from the leachate and regenerated for repeated use. A permit application has been submitted to the New Jersey DEP for construction and operation of a 25 gpm soil flushing facility on the site, based on the Ghea process.

TREATABILITY TEST DATA

Bench and pilot-plant tests have been conducted on various soil, sludge and water samples. Most of the samples were obtained from New Jersey sites, which contain petroleum hydrocarbons, aromatic compounds, PCBs, coal tar, heavy metals, and oily sludges. All the soil samples from the New Jersey sites contain fine silt and clay, typically in the 25 - 50% range. The concentration data presented below is in mg/kg for soil, and mg/liter for water.

DISCUSSION

The characteristics of the GHEA process which offer significant advantages over other treatment methods are:

- Applicability to complex mixtures of contaminants in a single integrated process. Since this is a physico-chemical method, the GHEA process efficiency is not diminished by toxicity or refractivity of the substrates. The versatility of the GHEA process stems from the ability to vary the surfactant formulation and strength as required for attaining the desired separation. In comparison, biological methods are limited to non-refractive organics. Solvent extraction methods are limited to organics. Inciner-

TABLE 1: TREATABILITY DATA

Matrix	Untreated Sample	Treated Sample	Percent Removal
Volatile Organic Compounds (VOCs):			
Soil	20.13	0.05	99.7%
Water	0.684	<0.010	>98.5%
Total Petroleum Hydrocarbons (TPH):			
Soil	13,600.	80.	99.4%
Polychlorinated Biphenyls (PCBs):			
Soil	380.00	0.57	99.8%
Trinitrotoluene in Water	180.0	<0.8	>99.5%
Coal Tar Contaminated Soil:			
Benz-a-pyrene	28.8	<0.1	>99.7%
Benz-k-fluoran	24.1	4.4	81.2%
Chrysene	48.6	<0.1	>99.8%
Benzoanthracene	37.6	<0.1	>99.7%
Pyrene	124.2	<0.1	>99.9%
Anthracene	83.6	<0.1	>99.8%
Phenanthrene	207.8	<0.1	>99.9%
Fluorene	92.7	<0.1	>99.9%
Dibenzofuran	58.3	<0.1	>99.8%
1-Me-Naphthalene	88.3	1.3	98.5%
2-Me-Naphthalene	147.3	<0.1	>99.9%
Chromium in soil	21,000.	630.	99.7%

ation is limited to combustibles. Vacuum stripping is limited to volatiles, and carbon adsorption is cost effective only for polishing low-strength water solutions of regenerable contaminants.

- Applicability to solid and water media, including slurries, sludges, and sediments. The GHEA process uses water as an extraction medium and purifies the water, as well as the soil, prior to discharge. Therefore, the presence of water in the soil, in whatever proportion, is compatible with the GHEA operation. In comparison, solvent extraction has no provision for treating contaminated water.

- Unlike the existing commercial soil washing processes, the GHEA process has a proven ability to decontaminate the silt and clay fractions of the soil efficiently and cost effectively. The key to our success in decontamination of clays is the ability to use high-strength surfactant formulations, and that is economically feasible only if the surfactant is reclaimed.

- The capital and operating costs of the GHEA process are low. The energy requirement is low in comparison with other processes, because the GHEA process does not include high-energy operations such as refrigeration, vacuum or gas compression. The capital and operating costs, estimated at $50 - 80 per ton, are lower than the estimates of $90 - 200 reported for other separation processes.

- Our engineering studies are based on a continuous-flow design for minimization of storage, handling and operating labor. The approach to engineering of a commercial treatment plant is based on shop fabricated, skid-mounted process modules which are transported to the site for assembly. This would provide maximum flexibility and cost savings in assembly, disassembly and applicability to widely different sites and site conditions.

- The surfactants used by Ghea are non-toxic and non-flammable, and are thus suitable for in-situ soil flushing.

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