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Sonic and ultrasonic removal of chemical contaminants from soil in the laboratory and on a large scale

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Abstract

Power ultrasound can be used for the rehabilitation of industrial sites or the reclamation of polluted land by the removal of chemical and biological contamination from soil. In this paper some current laboratory research and the potential for the scale-up of chemical decontamination is reviewed. Two basic mechanisms for acoustically enhanced soil cleaning have been suggested (a) an increase in the abrasion of suspended soil in slurries leading to the removal of contaminated material from the surface of particles and (b) an improvement in leaching out of more deeply entrenched materials.

1. Introduction

These days most of the developed world is aware of the problems caused by soil pollution. Gone are the days when waste could simply be buried and forgotten. Legislation is becoming tougher and so methods of both preventing and curing pollution are receiving a great deal of attention. Soil that is contaminated with chemicals presents a range of problems to the environment. These can include the destruction of ecosystems a loss in agricultural productivity, contamination of water resources and human and animal illness through direct ingestion of dust and the consumption of foods which have been grown on contaminated land. The chemical pollution itself can arise from a number of sources e.g. fall-out from incinerators or nuclear plants, residual pollution from industrial sites or the retention of herbicides or insecticides used in agriculture. There are two ways in which acoustic energy can enhance soil washing. These are predominantly mechanical and involve a combination of abrasion to remove superficial impurities and improved solvent leaching of contaminants from the interior of particles.

1.1. Surface cleaning of soil particles

Many conventional soil washing processes are based on the principle that pollutants adsorb onto very small particles “fine fractions” of the soil such as silt, clay and humic matter which themselves tend to be attached to coarser sand and gravel particles. The coarser particles make up the majority of the soil content. A primary aim in soil washing is therefore to dislodge and separate these fine components from the bulk soil. If the pollutant materials can be detached from the bulk, possibly together with some other surface contamination, a “concentrated” volume of polluted soil can be produced. This can then be treated or disposed of, and a large volume of residual soil which requires relatively little treatment and can be returned to the site as back fill.

A comparison has been made of the efficiencies of conventional and ultrasonically assisted pollutant extraction procedures using model soil samples (granular pieces of brick) that had been deliberately contaminated with copper oxide at an overall concentration of 51 ppm [1]. Analysis of the brick particles after 30 min sonication on a Vibrating Tray™ (Fig. 5) [2] revealed an average reduction in copper content to 31 ppm, a reduction of about 40%. Using a conventional mechanically shaken tray for the same time period the

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Table 1
Ultrasonic washing of brick particles

Brick remaining after washing	Particulate material < 20 mesh	Fines	Aqueous wash
Washing with conventional mechanical shaker			
746.5 g	2.9 g	0.63 g	12.6 l
(48 ppm Cu)	(310 ppm Cu)	(3200 ppm Cu)	(0.22 ppm Cu)
Total reduction in copper contamination in treated brick = 6%			
Washing with ultrasonic Vibrating Tray™			
744.7 g	3.4 g	1.89 g	13.5 l
(31 ppm Cu)	(96 ppm Cu)	(4700 ppm Cu)	(0.49 ppm Cu)
Total reduction in copper contamination in treated brick = 40%			

Initial mass of brick 750 g, copper contamination 51.4 ppm.

residual contamination was 48 ppm representing a reduction of only 6% (Table 1).

1.2. Leaching of pollutants from within particles

Any improvement in the penetration of solvent into particulate matter will result in the enhanced removal of soluble material that may be trapped inside the solid particles. This process is referred to as ultrasonic leaching and has been investigated for the decontamination of different types of soils e.g. landfills, mining spills and river sediments. Batch tests for accelerating leaching have used ultrasound for the removal of radio nucleotides and heavy metals from soils [3]. The application of ultrasound has also been found to aid precious metal recovery from waste products including industrial, municipal and mine wastes [4].

Although there is plenty of experimental evidence that ultrasound improves leaching the exact mechanism is not fully understood. Swamy and Narayana have suggested models for leaching in the absence and presence of ultrasound (Fig. 1a and b) [5]. Normal leaching takes place as the solvent front moves inward and steady state diffusion occurs through the depleted outer region and is equal to the rate of reaction within the reaction zone itself (Fig. 1a).

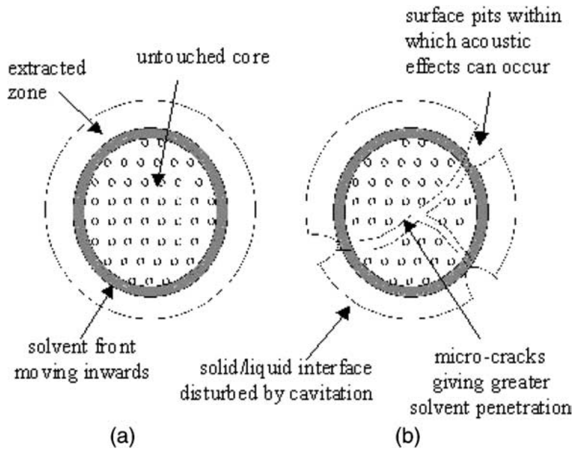

Fig. 1. Leaching of contaminants from soil particles (a) normal leaching, (b) in the presence of ultrasound.

Under the influence of ultrasound, normal leaching occurs but several additional factors contribute towards improvements in the efficiency. These include:

(i) Asymmetric cavitation bubble collapse in the vicinity of the solid surface leading to the formation of high-speed microjets targeted at the solid surface. The microjets can enhance transport rates and also increase surface area through surface pitting.
(ii) Particle fragmentation through collisions will increase surface area.
(iii) Cavitation collapse will generate shock waves which can cause particle cracking through which the leaching agent can enter the interior of particle by capillary action.
(iv) Acoustic streaming leading to the disturbance of the diffusion layer on the surface.
(v) Diffusion through pores to the reaction zone will be enhanced by the ultrasonic capillary effect.

2. Studies of ultrasonic soil washing

Soil pollution can occur through a variety of causes and the search for methods of removing it is actively pursued. A wide range of technologies is available but the main processes depend upon washing out the contamination, using bacteria to digest it, producing an impenetrable barrier to stop contaminant migration and heating. Of these options washing is the most attractive but it suffers from one difficulty—the production of large volumes of contaminated solvent rather than soil.

Australia suffers from soil pollution as do many developed countries and a group in New South Wales have begun a project in which ultrasound is used to enhance the rate of clean-up of soils [6]. Among the range of pollutants investigated were insecticides and polycyclic aromatic compounds. Laboratory studies have been made of the removal of various contaminants from soils. Most were carried out on slurries using a 12.5 mm tip diameter Misonix horn delivering approximately

170 W. Pure DDT and PCB (Aroclor 1260) were dissolved in acetone diluted with water and added to washed, fine sand (effectively pure SiO_2). This was tumbled and allowed to dry to ensure even adsorption onto the sand surface. Fifty weight% aqueous sand slurries were then made up. After insonation, the liquid was decanted and the soil was dried. The samples were then subjected to conventional extraction in order to determine the residual contaminant levels by GCMS. The results are shown in Figs. 2 and 3.

The study was extended to the decontamination of a riverine sediment. This was an industrial site polluted with PAH compounds (containing at least 15 compounds). The sediment, with an original contamination of approximately 400 ppm, was made up to 44.4 wt.% with distilled water and then sonicated as above. The results are shown in Fig. 4. These studies are in progress and results using a 4 kW ultrasonic system on a pilot plant scale will be published elsewhere.

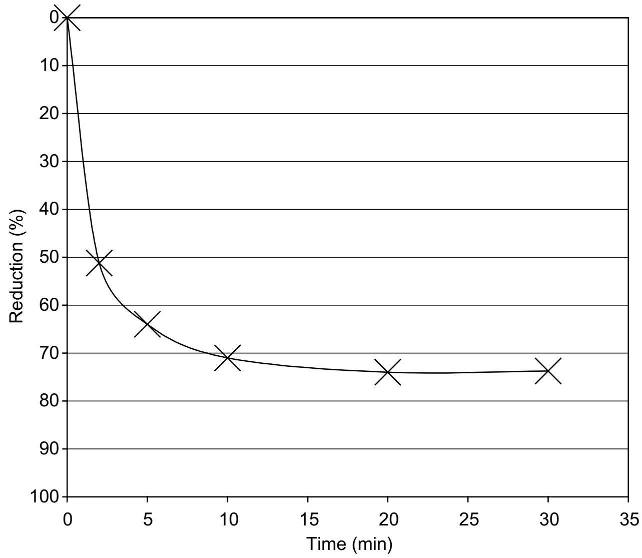

Fig. 2. Pesticide removal. (DDT ≈250 ppm in sand. Using 20 kHz 200 g contaminated "soil" in 200 g water.)

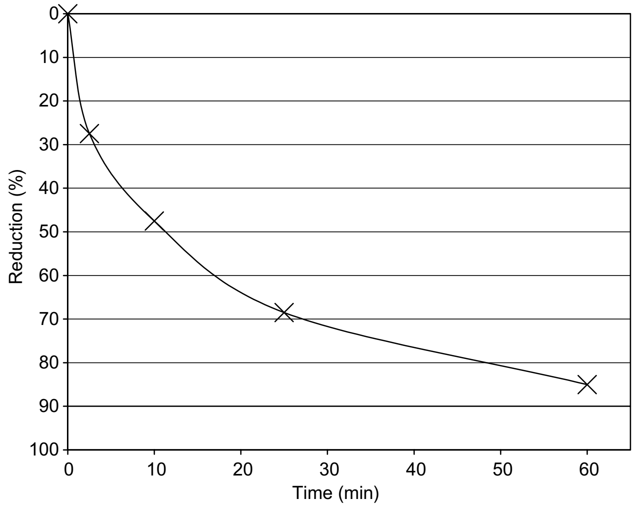

Fig. 3. Industrial pollutant PCB removal. (Aroclor 1260, a very common commercial product ≈250 ppm in sand. Using 20 kHz, 200 g contaminated "soil" in 200 g water.)

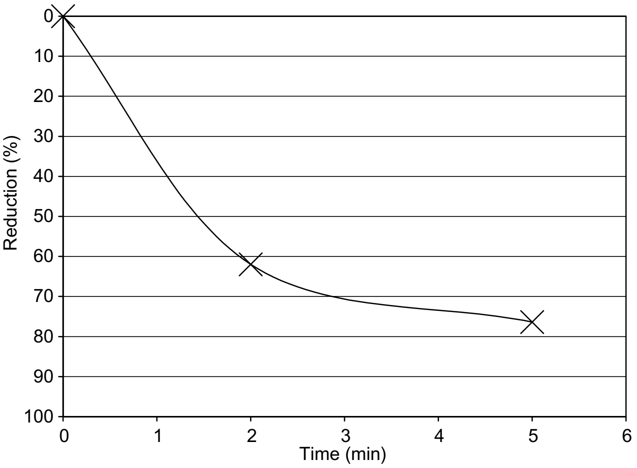

Fig. 4. Industrial pollutant PAH removal. (Samples from contaminated site ≈400 ppm. Using 20 kHz, 200 g contaminated "soil" in 200 g water.)

3. The scale-up of soil washing using acoustic energy

3.1. Using ultrasonic frequencies

We can extrapolate the laboratory results from Fig. 2 to obtain an estimate of the cost of scale-up treatment. Thus if 200 g of DDT-contaminated soil is treated for 10 min at 150 W to obtain 70% destruction of pesticide a simple scale-up would suggest that 1 kW h would treat 8 kg or we need 125 kW h of power per tonne of soil. Now, this does not represent very much power/cost compared with other technologies, especially at industrial rates (e.g. currently in Australia off-power big users in New South Wales pay 14 cents per kW h compared with 15 cents for domestic use). The question is can one scale up proportionally and this is why pilot plants are important. In fact, one might hope to obtain economies of scale.

The industrial adoption of ultrasonic soil washing depends upon the availability of scale-up systems. These do exist but generally they are not simply bigger versions of laboratory equipment although the use of a large scale Vibrating Tray™ or a multiple probe system could be considered as such. The development of flow systems is, as might be expected, the best choice for handling the large volumes of slurry which must be treated in soil washing. A selection of approaches to scale-up equipment is presented below.

This system comprises of a suspended metal trough which is subjected to ultrasonic vibrations by a

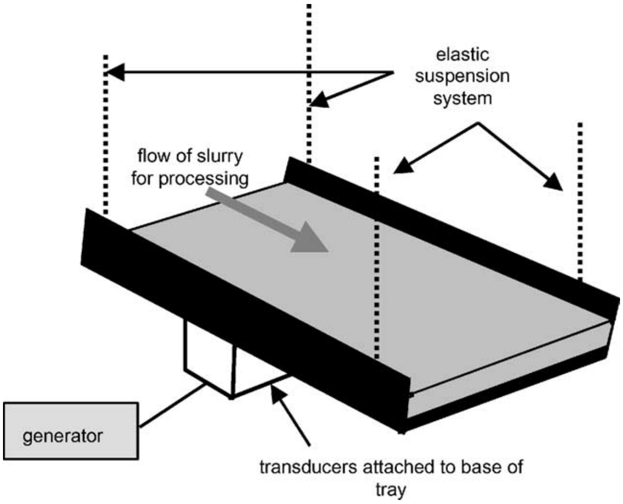

Fig. 5. Schematic representation of the vibrating tray device used for the cleaning of contaminated soils.

transducer array welded to its base. The whole system is in resonance and so any fluid or suspension that flows down the tray will be subject to ultrasonic agitation that can be harnessed for soil washing (Fig. 5).

The device was originally designed by the Lewis Corporation for the processing of coal or metal ores at rates up to 20 ton/h. The vibrating tray is generally used in open-air conditions and is clearly both large scale and extremely robust. In the particular case of coal recovery from waste tips the process involves mixing the coal "waste" with equal quantities of water and, after crude screening to remove rocks, allowing gravity to carry the mixture down the vibrating tray (20 kHz). With a residence time of only a few seconds this process yields marketable low ash coal product with clay and sand suspended in the wash water that can be allowed to settle in a pond and the water recycled for further processing.

A simple extension to laboratory studies with an ultrasonic probe is the use of an array of high power probes in a bath through which the soil slurry is passed.

Such an approach has been adopted for the high power treatment of wastewater or sewage sludge. A 30 kW unit used for sludge disintegration uses a series of probes (each 1 kW and operating at 20 kHz) are configured on one side of the flow system [7]. High-powered probes operating at 20 kHz with individual power ratings of up to 4 kW are available for this purpose. At this level of power input the transducers require cooling to avoid the overheating and consequent depolarisation which can accompany such usage.

As an alternative to high power probe inserts it is also possible to use an array of transducers slotted into a tube (Fig. 6). In such a system the metal transducer face is emitting the energy and the tube is simply providing the support and the container for the flowing liquid [8].

If transducers are fixed to the external surface of a tube then the tube itself becomes the source of ultrasonic energy. The liquid to be processed can then be passed through the tube and receive sonication directly from the ultrasonically vibrating walls. One device of this type is effectively a submersible transducer that has been developed in the shape of a rod [9]. When placed inside a tube the reactor produces steady and highly intense cavitation in a volume of 2.5 l at an intensity of 250 W/l[1] at 40 kHz. The performance intensity in the cavitation field can be increased by the additional equipment with oscillating systems on the outside of the reactor housing (Fig. 7).

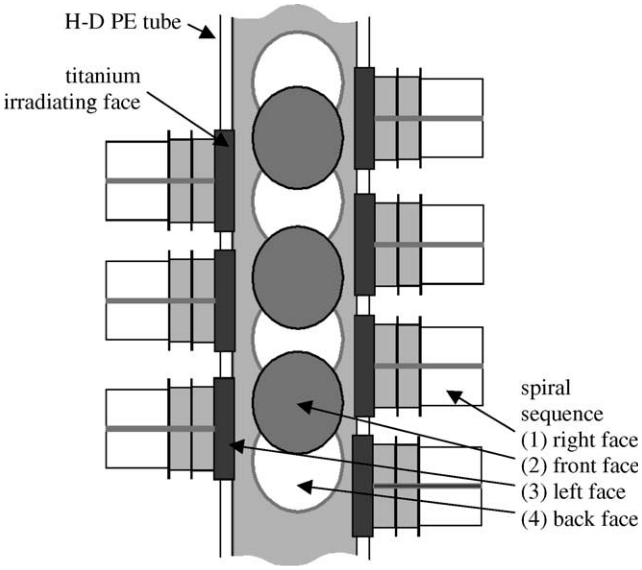

Fig. 6. Reaction tube with transducer inserts in a spiral configuration.

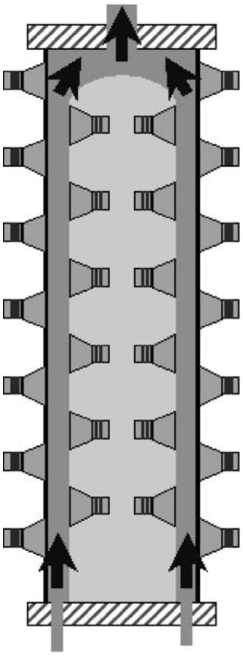

Fig. 7. Bandelin radial ultrasonic processor.

3.2. Using audible frequencies

A significantly different system has been introduced to large scale processing using audible acoustic energy. This involves generated vibrational energy through the use of resonant bending modes in a large cylindrical steel bar [10]. The bar is driven into a clover leaf type of motion by firing three powerful magnets in sequence which are located at each end of the bar. The bar is supported by air springs so that the ends and the centre are then caused to rotate at a resonance frequency depending on its size (Fig. 8). One such unit, operating at a power of 75 kW, drives a bar that is 4.1 m long and 34 cm in diameter at its resonance frequency of 100 Hz. The bar itself weighs 3 ton and produces a vibrational amplitude at each end of 6 mm—considerably larger than the amplitudes available through sonochemical processing and hence better for the dispersal of materials in liquids.

For processing applications, mixing chambers are rigidly mounted on each end of the bar. Material in the form of a liquid or slurry can then be pumped through these chambers in order to perform operations such as mixing, grinding and the destruction of hazardous waste. The bar itself has a large mass and this will determine the actual operating harmonic frequency of the system but fine tuning allows resonance to be maintained using different chambers and process conditions. For continuous mixing operations it is necessary to feed and discharge process fluids and so the connecting tube between the vibrating chamber and the (typically) rigid external piping system must be flexible, fatigue resistant and chemically inert to the materials being processed. The chambers used for any particular application will be process specific in terms of both residence time and the internal surface area and geometry.

A variety of applications of this technology have been investigated ranging from:

- relatively simple systems for soil de-agglomeration and gold ore leaching for which carbon steel and industrial hose are readily available, through
- moderately aggressive ozone–aqueous-organic mixtures requiring stainless steel and PTFE-lined tubing, to
- chemically aggressive and hazardous systems such as the nitration of benzene and the preparation of sodium dispersion in mineral oil. For these applications, carefully selected materials and double containment are required to ensure safety as well as product quality.

There are some parallels and differences between the use of audible sound frequencies for processing and the use of ultrasound for sonochemistry. Thus a comparison of the treatment of dyes in water has indicated that low frequency sonication does not appear to produce absolutely the same type of sonochemical reaction for their decomposition in water as ultrasound [11]. This is perhaps not unexpected if we extrapolate back from the high generation of radical species at 1 MHz through the lower generation at 20 kHz and then down to audible. However, this does not mean that there are no sonochemical reactions brought about by low frequency sonication. One might expect that hydrodynamic cavitation is very likely to occur in the liquid in the cell attached to the end of the vibrating bar under the extreme vibrational conditions that exist. It seems likely that the high rates of dye destruction by sonication and ozonation may well involve the sonochemically aided decomposition of ozone.

A modular plant has been developed which incorporates the vibrating bar technology for the extraction and chemical destruction of polychlorinated biphenyl (PCB) contamination from contaminated soil. High intensity mixing is provided by the 75 kW generator and this is used in three process areas: PCB extraction, PCB destruction and solvent recovery.

The test materials used in the programme were composed of a range of materials (Table 2). The sand matrix is relatively easy to treat but PCB in asphaltic and concrete matrices proved more difficult to extract.

A schematic flowsheet for the extraction and PCB destruction components of the demonstration test is presented in Fig. 9. Both the PCB content and nature of

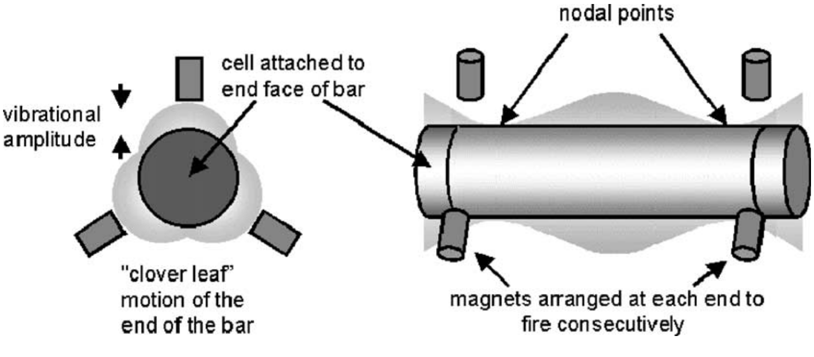

Fig. 8. SESI vibrating bar system.

Table 2
Material used in the extraction of PCB's from soils[a]

Soil components	
Major	Sand, asphalt (high PCB), concrete
Minor	Silt, clay, soil organics

[a] The material used was stored from an industrial site excavation with average PCB content 550 ppm (highly variable). It was ground for ease of processing.

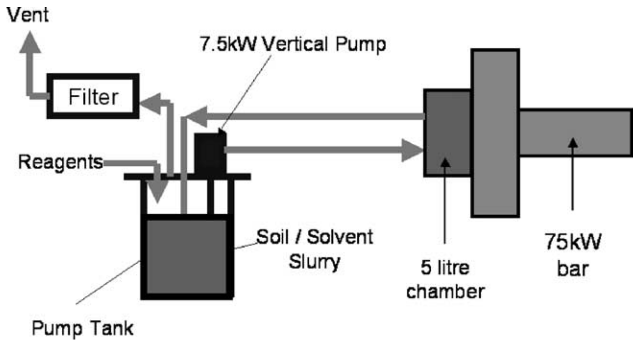

Fig. 9. Pumped test on batch system.

Table 3
Typical product PCB contents (mg/kg, i.e. ppm)

Test condition	Feed soil	Soil residue	Analysis
0.2 kW/l, 105 min	700	<2	GC–ECD
0.2 kW/l, 105 min	700	<0.4	GC–MS
2.0 kW/l, 105 min	550	<2	GC–ECD

For analysis the residual soil is extracted for 2 h in 1:1 acetone/hexane. Hexane soluble extract analyzed.

process chemicals require double containment of the flexible connections between the mixing chamber and the fixed piping system.

Results achieved are presented in Table 3. The 2 mg/kg (2 ppm) PCB level is significant since this is emerging as an international standard for unconfined disposal. It is also, for practical purposes, the limit of detection for dirty sample analysis by soxhlet extraction and gas chromatography with an electron capture detector ("GC–ECD"). Use of an ion selective mass spectrometer coupled with high-resolution GC ("GC–MS") permits a 0.4 mg/l detection limit, which was also achieved. The tendency of regulators to set disposal limits at or near analytical detection limits may not be very scientific, and it is questionable whether even GC–ECD is a practical process control tool. However, from a process operator's point of view it is important to determine in the testing phase whether process performance is sufficient to survive another tightening of the regulatory screw.

The use of the 75 kW generator for pilot testing provides assurance of process performance at commercial scale (3–4 ton of soil/hour average).

4. Conclusions

Small scale (laboratory) cleaning of soil samples has proven to be effective and some large scale trials have been shown to be promising. Now that legislation is becoming even tighter in terms of permitted residual concentration of pollutants there is a major drive towards real clean-up, as opposed to either excavating and burying somewhere else or simply putting a barrier around the perimeter of a site and laying tarmac over the top.

This is why equipment manufacturers are interested in scaling up ultrasonic soil washing, they know that there is a market out there for new clean-up technologies.

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