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Briefing: In situ decontamination of sediments using ozone nanobubbles and ultrasound

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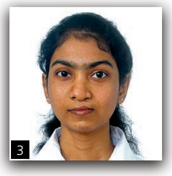

This paper describes innovative research on cleaning contaminated sediments. It uses three emerging technologies, namely, ultrasound, ozone and nanobubbles, to provide a cost-effective and environmentally sustainable on-site treatment technology with a lower total cost over a shorter time span. The ultrasound energy provides agitation and soil decontamination. The ozone reacts with desorbed contaminants to remove them from the river. The nanobubbles help in the dissolution of ozone gas in water. Once the treatment is completed, any remaining dissolved ozone will break into oxygen and will help to revitalise microbes and the ecosystem. The initial bench-scale test results are promising, and the study will be continued in order to identify and optimise parameters that will influence the removal efficiency of contaminated sediments.

Introduction

Over 50% of the global population lives in urban centres and this number is supposed to increase over the next decade. Megacities around the world are usually amalgamated to major rivers, which are polluted by chemicals released from major industries within megacities. River sediments have become one of the major transporters of heavy metals in the aquatic environment. Contaminated sediments are a significant environmental problem that impairs the uses of many water bodies, often contributing to fish consumption advisories. Hence, it essential to remediate contaminated sediments. Most remediation methods include dredging the contaminated sediments to be treated at off-site treatment plants. Off-site treatment plants usually solidify/stabilise contaminated sediments or are utilised in the production of construction materials for beneficial use. However, such off-site treatment is quite expensive. Hence, the common practice of remediation is dredging and disposal in a controlled landfill. There are risks involved with resuspension of the contaminants during dredging, and disposal in landfills is associated with long-term liability and difficulty locating disposal sites. In situ remediation technologies can prevent the risks involved with the resuspension of the contaminants during dredging and difficulties associated with disposal of contaminated sediments. This paper describes an innovative in situ sediment decontamination technology with ultrasound and ozone nanobubbles. Three emerging technologies, namely, ultrasound, ozone treatment and nanobubbles, are combined in this research to address a major problem: in situ decontamination of sediments.

Ultrasound technology

The purpose of the ultrasound energy is to provide agitation that will maintain the sediments in a suspended state, detach contaminants formulated on the surface of sediments and release them to the bulk solution. The ultrasound energy will also generate bulk motion to enhance uniform application of the ozone to all contaminated particles and to facilitate desorption of contaminants from sediments. The application and use of sound waves with a frequency higher than 20 kHz is defined as ultrasound technology, which is used in medical technology, cleaning technology and non-destructive testing using a wide range of frequencies. Ultrasound is usually generated by piezoelectric crystals with frequencies ranging from 20 kHz to 100 MHz, where an electrical signal matching the natural frequency of the crystal is applied to generate ultrasound. Ultrasonic cleaning works by providing shear forces to remove the material adhering to a surface. This shear force is developed by cavitation. Ultrasound causes high-energy acoustic cavitation – that is, the formation of microscopic vapour bubbles in the low-pressure (rarefied) part of the ultrasonic wave. These bubbles collapse in the compression part of the wave, creating very minute but high-energy movements of the solvent that result in localised high shear forces during cavitational collapse, causing intense heating of bubbles. These localised hot spots have temperatures of roughly 5000°C, pressures of 500 atm and a lifetime of a few microseconds (Suslick, 1990). Applications to chemical reactions exist in both liquids (homogeneous) and liquid–solid mixtures. Application of ultrasound energy to a soil slurry such as contaminated sediments causes acoustic cavitation. Shock waves from cavitation in liquid–solid slurries produce highvelocity interparticle collisions, the impact of which is sufficient to desorb the contaminants from soil. In soil decontamination application, the contaminants (organics and inorganics) are usually attached to the clay and silt fractions. The application of ultrasound energy results in generating localised high shear forces to separate and desorb organic and inorganic compounds and heavy metals from sediments or soils, thereby increasing the mobility of contaminants in the soil water suspension. Increasing the mobility of contaminants will allow the remediation process to oxidise directly or to leach the contaminants by using surfactants. Ultrasound and surfactants were used to remove the polycyclic aromatic hydrocarbons (PAHs) in soil heavily contaminated with coal tar, achieving more than 90% contaminant removal efficiency (Meegoda *et al*., 1995). Acoustic cavitation due to ultrasound energy coupled with vacuum pressure was used to facilitate the removal of chromium (Meegoda and Perera, 2001) and PAHs from sediments (Meegoda and Veerawat, 2002).

Oxidation using ozone

The roles of ozone are to degrade desorbed organic contaminants in the sediment into intermediate products that are soluble in the aqueous phase and also to oxidise the desorbed heavy metals from sediments into soluble ions for enhanced removal by filtration. Ozone is an unstable gas and readily reacts with organic and inorganic substances. It is a highly reactive and powerful oxidant that has been used in the chemical industry as an oxidising agent and is also used extensively in the treatment of drinking water. Upon release of its oxidising potential, ozone reverts back to oxygen, from which it was generated. Application of ozone does not leave a chemical residual, and under ambient conditions, it has a half-life of 10–20 min, whereas half-life can be extended to a month if supplied as nanobubbles. Ozone is usually generated electrically on demand and cannot be stored for later use. Ozone is generated by irradiation of an air stream with ultraviolet light at a wavelength of 185 nm or by passing dry air or oxygen through a corona discharge generator. Interest in the use of ozone to remediate contaminated soils has been rising over years, particularly in application on non-volatile organic compounds that are not removed by conventional soil venting. Ozone can be utilised in both gaseous and aqueous forms. When utilised in the aqueous phase, ozone can be applied to the soil in a similar manner to that used in soil washing. Another benefit of using ozone is that, after a short period of time, ozone that has not reacted reverts back to atmospheric oxygen and therefore no toxic residues of the oxidant remain in the soil. Ozone is reported to be expedient for the degradation of PAHs in soils.

Nanobubbles

The ozone gas is to be delivered as nanobubbles to increase the ozone gas dissolution in water and to enhance the ozone gas stability in the liquid phase. Nanobubbles are nanoscopic gaseous (typically air) cavities in aqueous solutions that have the ability to change the normal characteristics of water. Ordinary bubbles have a diameter ranging from 1 μm and larger and are in equilibrium due to capillary and buoyant forces. If not in equilibrium, these bubbles quickly rise to the surface of a liquid and collapse, whereas nanobubbles remain in water. Nanobubbles which have a diameter of <100 nm can remain in liquids for an extended period of time. The stability of nanobubbles is supported by the electrically charged liquid–gas interface, which creates repulsion to prevent bubble coalescence, and by the high dissolved gas concentration in the water by maintaining a small concentration gradient between the interface and the bulk liquid (Ushikubo *et al*., 2010). The zeta potential measured in the water after the introduction of oxygen micro- and nanobubbles was in the range from −45 to −34 mV and from −20 to −17 mV in water bubbled with air, indicating the presence of stable electrical charge on the bubble surface (Ushikubo *et al*., 2010). Some advantages of the micro- and nanobubbles are their high specific area (surface area per volume) and the high stagnation time in the liquid phase, which increase the gas dissolution.

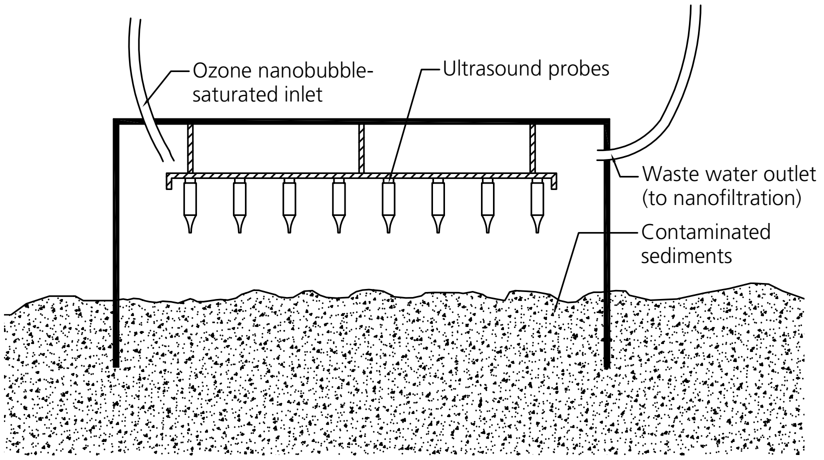

Figure 1. Draft design of the field application chamber

Table 1. Initial trials with lower water temperature at the end of the experiment

Ultrasound (frequency 20 kHz)		Temperature: °C		Efficiency: %
Sonication time: min	Ultrasound power: W	Pre	Post	
10	900	11·0	15·0	80·41
24	900	12·0	16·8	90·02
36	1050	13·2	17·0	92·99

Proposed technology

The proposed technology using ultrasound and ozone nanobubbles will be integrated inside a fully contained system as shown in Figure 1, and once that section of the river sediment is treated, the treatment chamber is moved to an adjoining section of the river in a grid. The proposed field implementation will be using a chamber of size 10 feet × 10 feet × 5 feet (3·05 m × 3·05 m × 1·52 m) ($L \times W \times H$), made of stainless steel containing ultrasound transducers spaced at 1 foot (0·305 m) intervals in a grid. It will be lowered to the sediment bed from a barge. The chamber will be allowed to sink into the sediment to a depth of about 2–3 feet (0·610–0·914 m) due to its weight, and then ozone nanobubble-mixed water will be supplied to the system as depicted in Figure 1. At the beginning, water trapped in the chamber above the sediment will be circulated to saturate that with the ozone nanobubbles, after which the ultrasound transducer rack is lowered to apply ultrasound energy to agitate and desorb the contaminants from the sediment while using ozone nanobubble-saturated water to oxidise the pollutants. During the ultrasound application, river sediments will be mixed with water-containing ozone nanobubbles to form a slurry. After ultrasound treatment, sediments are allowed to settle and the generated waste water above the settled sediments inside the chamber will be extracted and treated through a portable waste water treatment facility on a barge or on-site by using nanofiltration/precipitation to capture heavy metals and other residual chemicals before recirculating the waste water back into the containment chamber. This will ensure the capture of heavy metals and other residual chemicals prior to the release of water back to the river.

Experimental results

Ultrasound (1500 W (Blue Wave Ultrasonics Inc., model IM-1620, 240 V, maximum power 1500 W, frequency 20 kHz) and ozone nanobubbles (model T Series, Welsbach Ozone System Corporation, USA; model BT-50FR nanobubble nozzle, Riverforest Corporation, USA) were applied to the beaker containing contaminated soil and water. The soil/water ratio was kept at 4%. The temperature, ozone concentration and sonication time were selected as key variables. The interaction between ultrasound and water generated heat, raising the temperature. High temperatures resulted in low removal efficiencies. Hence, the ultrasound application was broken into 2 min intervals and lower initial temperatures were selected. Table 1 shows that the process is successful with low operating temperatures ranging from 8 to 15°C. The application of sonic power in discrete intervals allowed the system to saturate and ready for the next sonication.

Summary and conclusions

A new in situ remediation technology using ultrasound and ozone nanobubbles was proposed. An extensive experimental programme is under way to validate the above technology. The preliminary results of this study indicated promising data from using ozone nanobubbles and ultrasound to remove PAHs from sediments. The preliminary research also showed high removal efficiencies of PAHs from the contaminated sediments at lower temperatures and high ultrasound power levels. With the encouraging preliminary results, the proposed technology can be used in the winter months when the river is minimally used for recreation purposes. This study will be continued in order to identify and optimise parameters that will influence the removal efficiency of contaminated sediments.

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REFERENCES

Meegoda JN and Perera R (2001) Ultrasound to decontaminate metals in dredged sediments. *Journal of Hazardous Materials* **85(1–2)**: 73–89.

Meegoda JN and Veerawat K (2002) Ultrasound to decontaminate organics in dredged sediments. *Soil and Sediment Contamination* **11(1)**: 91–116.

Meegoda JN, Ho W, Bhattacharjee M *et al.* (1995) Ultrasound enhanced soil washing. *27th Mid-Atlantic Industrial & Hazardous Waste Treatment Conference, Leigh University, Bethlehem, PA, USA*, pp. 470–479.

Suslick KS (1990) Sonochemistry. *Science* **247(4949)**: 1439–1445.

Ushikubo FY, Furukawa T, Nakagawa R *et al.* (2010) Evidence of the existence and the stability of nanobubbles in water. *Colloids Surfaces A: Physicochemical and Engineering Aspects* **361(1–3)**: 31–37, http://dx.doi.org/10.1016/j.colsurfa.2010.03.005.