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Sonication Effect on NAPL Extraction from Soils

초음파를 이용한 흙에서의 NAPL의 추출에 관한 연구

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요 지

초음파를 이용한 지반 세척의 효율을 높이기 위한 방법이 최근에 개발되었지만 그에 대한 연구는 아직 초기 단계인 실정이다. 본 연구는 초음파가 지반 정화에 미치는 영향에 대하여 실내 실험을 통해 고찰하였으며 특히 초음파 에너지의 강도, 시료의 종류 및 밀도, 그리고 수두의 변화가 지반 세척에 미치는 영향에 대하여 연구를 수행하였다. 상기의 연구 결과 지반 세척시 초음파가 세척 능력을 상당히 향상시키는 것으로 나타났으며 초음파에너지가 강할수록, 흐름 속도가 느릴수록 초음파의 효과가 크게 나타났다.

Abstract

Use of ultrasonic waves to enhance the effectiveness of soil flushing method is a new in-situ remediation technique. However, there has not been an analytical method that can be used to evaluate the effectiveness of ultrasonic wave under different conditions. This study was undertaken to investigate the degree of enhancement in contaminant extraction due to ultrasonic waves for different levels of ultrasonic power, soil type, soil density, and hydraulic gradient. The study was conducted in the laboratory using a specially designed and fabricated test device. The test soils were a Ottawa sand, a fine aggregate, and a natural soil, and the surrogate contaminant was a Crisco Vegetable Oil. The test results indicated that sonication can enhance pollutant removal considerably. Increasing sonication power will increase pollutant extraction. The faster thè flow rate is, the smaller the degree of enhancement will be. The pollutants in dense soils are more difficult to be removed than in loose soils. However, the effect of soil density on pollutant removal enhancement due to sonication appears to be less significant compared with the other factors.

Keywords : Soil flushing method, Ground contamination, In-Situ remediation, Ultrasound, Stress waves, NAPL, Contaminant, Extraction

1. Introduction

Petroleum hydrocarbons are commonly found in the grounds of urban and suburban areas due to the possible leakage of gasoline, motor oils, and diesel fuel from underground storage tanks. The polluted ground needs to be cleaned in order to avoid potential hydrocarbon contamination of ground water aquifer. Currently, there are different remediation methods, e.g., replacement, pump-and-treat, vapor extraction, and flushing methods. However, a method that can be effective and also economical for a broad range of field conditions is not yet available.

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For the development of an effective ground remediation method, there has been considerable research focusing on the technique of enhancing soil flushing method. Although there are data showing that ultrasonic waves are capable of removing non-aqueous phase liquid (NAPL) hydrocarbons from soils, a methodology for evaluating the effectiveness of sonication is yet to be developed. It is with this objective in mind that this study was undertaken.

2. Current State of Knowledge

Available information about sonication effects on extraction of NAPL hydrocarbon from porous media is limited and piecemeal. Berliner et al. (1984, 1987) reported that high intensity ultrasonic waves can disperse particulate suspensions and enhance contaminants extraction. Pogosyan et al. (1989) also demonstrated that stress waves can drastically increase the gravitational phase separation of hydrocarbon from water. According to Nikolayevskiy (1989), there exists a threshold sonication level, below which the hydrocarbons are immobile due to their presence as isolated droplets in the pore. Ultrasonic waves can create a stream or cluster from these droplets resulting in a greater mobility of the hydrocarbons.

Others, Simkin and Surguchev (1991) and Simkin (1993) attributed the increased production of oil (hydrocarbons) to a decrease for water and an increase for oil in the relative phase permeability due to stress waves. For soils with low permeability, Cleveland and Garg (1993) reported that ultrasonic excitation can suspend fine particles to which the contaminants are strongly sorbed. The fine particles subsequently can then be removed by flushing water through the soil. Also, Reddi and Challa (1994), and Reddi and Wu (1996) presented that ultrasonic waves can increase not only the mobility of NAPL ganglia but the porosity of the soil as well, resulting in a decrease in viscosity and buoyant pressure.

Very few studies are available on the effectiveness of using acoustic waves to enhance contaminant extraction in soil flushing method. Ellen et al. (1995) reported a 30% increase in contaminant extraction due to acoustic excitation. They hypothesized that acoustic waves could overcome the capillary force on contaminants in a soil by alternating over- and under-pressures which produce pulling and pushing action to contaminant droplets. Thus, large contaminant droplets can be broken into smaller droplets. These smaller droplets can be flushed out more easily. Another study by Iovenitti et al. (1995) reported a 6% to 26% improvement in contaminant extraction. With this test result, they presented a fairly comprehensive discussion of possible mechanisms responsible for the enhanced contaminant extraction. They summarized the effects of acoustic excitation in two aspects -- on porous grain framework and on pore fluids. The effects on grain framework include (i) vibrational alignment or reordering of soil particles to decrease impedance in flow direction, (ii) temporary increase in soil porosity due to particle agitation, (iii) disintegration of pore-blocking material, and (iv) cavitation in fine-grained soils resulting in porosity/permeability increase. The effects on pore fluid involve (i) an increase in fluid temperature, volume, and pore pressure due to increased kinetic energy, (ii) a decrease in fluid viscosity, (iii) disintegration and mobilization of sorbed contaminants due to increased molecular movement, and (iv) lower surface tension resulting in coalescence of stationary contaminant droplets and flow.

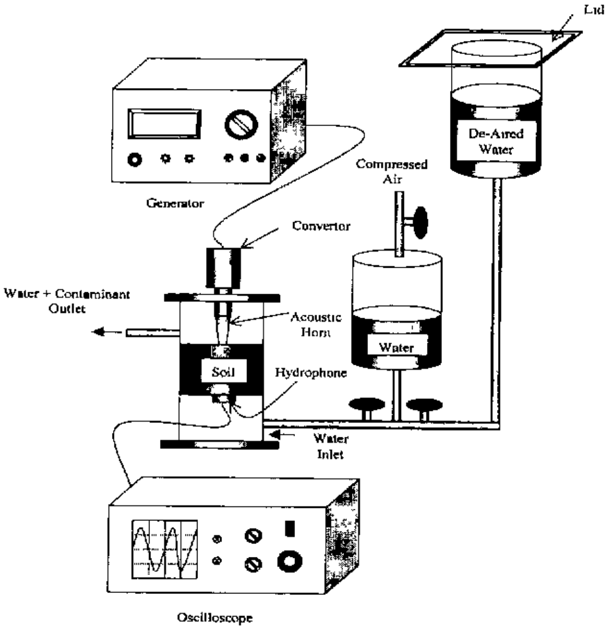

Fig.1 Test setup for soil flushing test

The available information reviewed above shows that ultrasonic waves can increase percolation rate and facilitate removal of entrapped contaminants. The degree of enhancement varies with many factors, e.g. soil type, soil density, flow rate, temperature, waves frequency, energy level, etc. Since nearly every available study focused only on limited specific conditions without a systematic investigation for a broad spectrum of the various influencing factors, a methodology is not yet available for evaluating the effectiveness of ultrasonic waves. Such a methodology is essential in the practical application of using stress waves to enhance the effectiveness of soil flushing.

3. Laboratory Investigation

3.1 Test Equipment

The laboratory testing involved one-dimensional soil flushing and soil characteristics determinations. The soil flushing test was conducted using a specially designed and fabricated test equipment which was shown schematically in Fig. 1. As shown, the test setup was composed of two parts--a test chamber with water supply and flow regulators and an ultrasonic processor.

The test chamber was made of a Plexiglas cylinder having an inside diameter of 7.3 cm with a height of 30 cm. The bottom and top of the cylinder were sealed with aluminum caps. These caps were fitted with o-rings for complete seal and were securely fastened to the cylinder with screws. The bottom cap was provided with an inlet connecting to a bellofram which was connected to both the test chamber and de-aired water reservoir to apply the needed water pressure for flow through the test soil specimen. The de-aired water reservoir was connected to the water tap.

The ultrasonic processor is composed of a generator, a converter, an acoustic horn, and a flat tip as shown in Fig. 1. The generator (or power supply) converts conventional 60 Hz AC at 120V to 20 kHz electrical energy at approximately 1,000V. The high-frequency electrical energy is fed to the converter to transform the energy to mechanical vibration. The vibrator is tuned to vibrate at 20 kHz. The acoustic horn and flat tip amplify the longitudinal vibration of the converter.

The ultrasonic energy applied to the soil specimen was monitored using a hydrophone which was mounted at the bottom of the soil specimen. The hydrophone was connected to GoldStar OS-9020G 20MHz oscilloscope to monitor the output voltage from the receiver (hydrophone) throughout the experiments.

3.2 Test Soil and Contaminant

The test soils were Ottawa sand, a fine aggregate, and a natural soil. Some physical properties of the test soils are shown in Table 1. As shown in Fig. 2, the particle size of Ottawa Sand ranged between 0.18 mm and 0.8 mm with $D_{10} \approx 0.27$ mm and $D_{50} \approx 0.45$ mm. The fine aggregate material was poorly graded having a maximum particle size of 10.0 mm with $D_{10} \approx 0.54$ mm and $D_{50} \approx 1.4$ mm. Natural soil was well graded having a maximum particle size of 10.0 mm with $D_{10} \approx 0.25$ mm and $D_{50} \approx 1.1$ mm.

The test surrogate contaminant was Crisco Pure Vegetable Oil. The density of Crisco Pure Vegetable Oil was 0.9182 g/ml at 20°C, and the viscosity was 67cP at 20°C. The solubility and volatility are very low and negligible. These characteristics were provided by the Crisco Company.

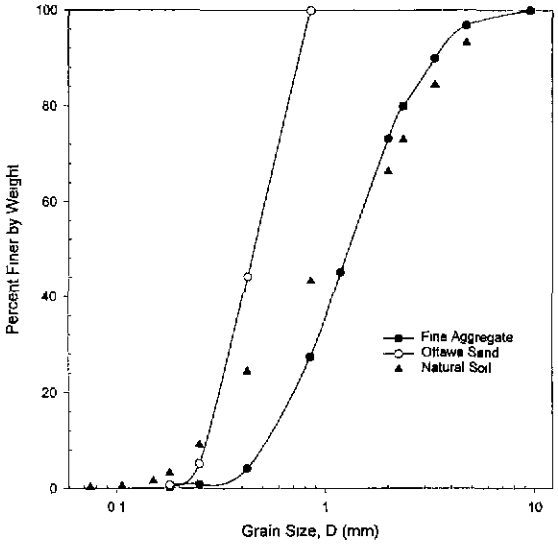

Fig. 2 Particle size distribution of test soils

3.3 Test Procedures

The soil flushing tests were conducted for two conditions--without ultrasonic waves and with ultrasonic waves at 20 kHz frequency. The test specimens were prepared from the mixture of air dried soil sample and Crisco Pure Vegetable Oil. The test specimen had a diameter of 7.3 cm and a height of 5.0 cm. Two void ratios (e), three levels of hydraulic gradient (i), and two levels of ultrasonic energy (W) were tested.

The major steps involved in the experiments are as follows. A pre-weighed soil (330.6g for Ottawa Sand and 337.1g for fine aggregate and natural soil) was mixed thoroughly with a desired amount (70% saturation of the porosity) of Crisco Pure Vegetable Oil. The mixture of soil and oil was carefully placed into the test chamber. At both ends of the soil specimen are placed #100 mesh screens to retain soil particles while allowing water to flow through the soil specimen. The soil specimen was then saturated with water; the water level was maintained at the top of the soil specimen. The soil specimen was then subjected to ultrasonic waves at 20 kHz frequency. Under the action of ultrasonic waves, the clean water was allowed to flow upward through the soil specimen under a specified hydraulic gradient. The effluent was collected in a 500 ml Polypropylene cylinder. The effluent in the cylinder was allowed to stand overnight for gravitational segregation of oil from water. The volumes of the separated water and oil are then measured.

4. Experimental Results and Discussions

For the test specimen of Ottawa Sand prepared at a void ratio of 0.67 and subjected to a hydraulic gradient of 1.6, the percent of contaminant removal is plotted against water flow volume in Fig. 3 for both without and with sonication at 50W and 100W power. The figure shows that the percent of contaminant removal increases with water flow volume at a decreasing rate and reaches a constant after 25 PV (pore volume), and that sonication increases contaminant removal considerably. It is also seen that the percent contaminant removal is much greater for 100W than 50W power. According to the figure, the maximum amount of contaminant removal is approximately 50, 55, and 62% for flushing without and with sonication at 50W and 100W, respectively. Thus, there is approximately 50% residual amount of contaminant remained inside the soil regardless of the duration of flushing without sonication. With sonication, the residual amount can be reduced to approximately 45 and 38% for sonication power of 50W and 100W, respectively.

Fig. 4 presents sonication power effect on contaminant removal after 5 PV water flow volume for two levels of void ratio and three levels of hydraulic gradient. For a constant void ratio and hydraulic gradient, the percent contaminant removal increases with increasing sonication power to a maximum around 100W then decreases. The drop in contaminant removal beyond about 100W can be attributed to cavitation effect because cavitation was observed at 140W power. When cavitation takes place, the swarm of minute air bubbles may impede upward contaminant movement resulting in a drop in contaminant removal. A similar cavitation effect on oil flow through sandstone was reported by Fairbanks and Chen (1971). They observed cavitation at 100W sonication power. The lower sonication power for their case was probably because

Table 1 Physical properties of test soils

Parameter	Ottawa Sand	Fine Aggregate	Natural soil
C_u (Uniformity Coefficient)	1.85	3.21	12 9
D_{10}, mm (Effective Grain Size)	0 27	0.54	0.25
e_{min} (Min Void Ratio)	0 46	0.46	0.59
e_{max}(Max. Void Ratio)	0.67	0.80	0 90
G_s(Specific Gravity)	2.65	2.69	2.69
Unified Classification	SP	SP	SW

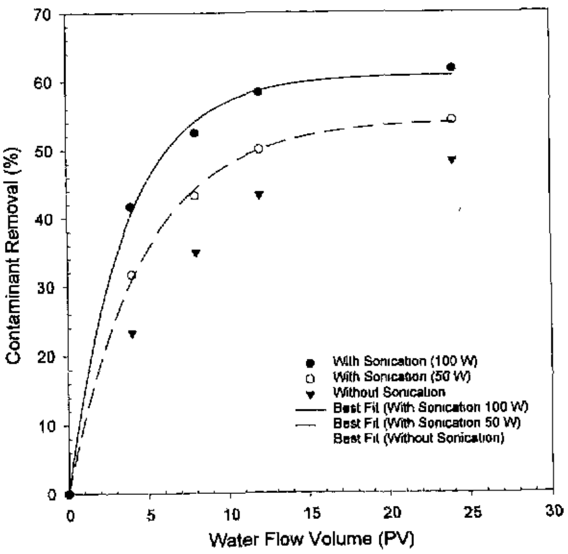

Fig. 3 Percent contaminant removal vs. water flow volume for e=0.67, i=1.6, with and without sonication

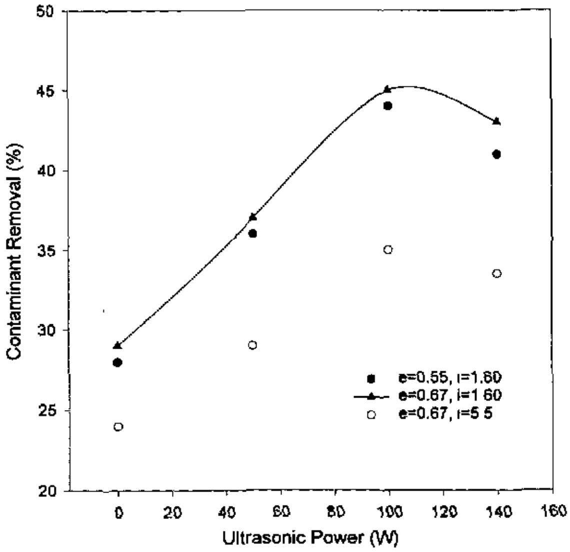

Fig. 4 Ultrasonic power (W) vs. contaminants removal (%) (Ottawa sand)

of their smaller test samples which had a diameter of 7.3 cm and a height of 0.5 ~ 2.0 cm.

The effect of void ratio on contaminant removal is demonstrated in Fig. 4. It is seen that the contaminant in soils with a higher void ratio can be removed more easily than in soils with a lower void ratio. The test results indicate, as would be expected, that contaminants in loose soils can be extracted more easily than dense soils. With sonication, the effect of soil density on contaminant removal seems to less than the case without sonication.

Another important factor affecting contaminant removal is the flow rate. The flow rate depends on the hydraulic gradient applied to the system; the higher the hydraulic gradient is, the greater the flow rate will be. Fig. 5 shows that increasing flow rate decreases contaminant removal. It is seen that the percent contaminant removal decreases as the discharge velocity of flushing water increases, and that the relation appears to be independent of soil type and void ratio. Thus, for the same amount of flushing water, slow flushing water can remove contaminants more easily than fast flushing water regardless of soil type and density. Possible explanations for the observed flow rate effects are as follows:

Before the contaminant can be flushed out of the soil, the soil/contaminant bond must be broken first. Inside the soil mass, the contaminant is either trapped within the pore space formed by interlocked soil particles or adsorbed on the surface of individual particles or both. Regardless of the nature of the bond, the breakdown of the contaminant/soil bond is a time-dependent process. For slower flushing, the percolating water has longer time to interact with soil /contaminant system. As a result, the slow flushing water is more efficient to remove contaminant than the fast flushing as observed from the experiment.

5. Summary and Conclusions

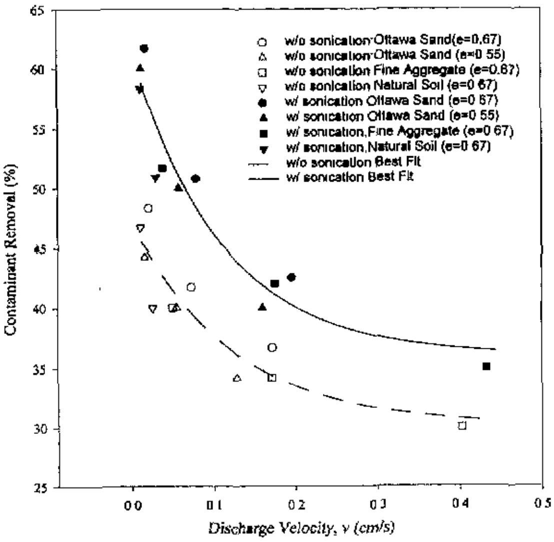

Fig. 5 Contaminant removal (%) vs. discharge velocity

Non-aqueous phase liquid (NAPL) hydrocarbons such as gasoline, motor oils, and diesel fuels are major ground pollutants in urban and suburban areas. Polluted grounds can be remediated in-situ using the soil flushing method. With an application of ultrasonic waves during flushing, the soil flushing technique can become an effective method for in-situ remediation of polluted grounds. There has been information showing that ultrasonic waves can enhance contaminant extraction. However, an analytical method which can be used to evaluate the effectiveness of ultrasound under different conditions is not yet available. This study was undertaken to investigate the degree of enhancement in contaminant extraction due to ultrasonic waves.

The study was conducted in the laboratory using a specially designed and fabricated test device. The test soil was a sand and the surrogate contaminant was a Crisco Vegetable Oil. The test results indicated that sonication can enhance pollutant removal considerably, and that the degree of enhancement depends on a number of factors such as sonication power, water flow rate, and soil density. Increasing sonication power will increase pollutant extraction. The faster the flow rate is, the smaller the degree of enhancement will be. The pollutants in dense soils are more difficult to be removed than in loose soils. However, the effect of soil density on pollutant removal enhancement due to sonication appears to be less significant compared with the other factors.

Based on the results of this study, it is concluded that there are quite a few factors that can influence sonication effect on contaminant removal in a complex manner. This study has investigated only some of the more important factors. Thus, the database needs to be expanded. Accordingly, to achieve the final goal of developing an analytical method for evaluation of sonication effect on contaminant removal, the current study is being continued.

Acknowledgement

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