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Constantinos V. Chrysikopoulos
Technical University of Crete

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Eric T. Vogler and Constantinos V. Chrysikopoulos

Department of Civil and Environmental Engineering, University of California, Irvine, California, USA

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[1] The effect of acoustic waves on the transport of a conservative tracer in a water saturated column packed with glass beads was investigated. It was observed from the experimental data that the addition of acoustic waves, in the frequency range between 60 to 245 Hz, to a steady background pressure gradient, enhances solute transport compared to the base case consisting of only a background pressure gradient. Furthermore, it was found that the effective velocity of the solute is approximately inversely proportional to the frequency of the acoustic wave. *INDEX TERMS:* 1832 Hydrology: Groundwater transport; 1829 Hydrology: Groundwater hydrology; 1831 Hydrology: Groundwater quality; 1878 Hydrology: Water/energy interactions; 1894 Hydrology: Instruments and techniques

1. Introduction

[2] Various aspects of contaminant transport enhancement in porous media have been extensively explored in recent years by environmental engineers and scientists who design remediation methods for contaminated subsurface formations. For example, remediation techniques for groundwater and soil contaminated by nonaqueous phase liquids (NAPLs), often employ surfactants to reduce surface tension and enhance mobilization of NAPLs [*Mason and Kueper*, 1996; *Fortin et al.*, 1997], or employ cosolvents and humic substances to increase the solubility of slightly soluble NAPLs [*Brandes and Farley*, 1993; *Li et al.*, 1996; *Tatalovich et al.*, 2000]. Furthermore, bioremediation techniques employ bacteria that enhance contaminant degradation [*Chaudhry*, 1994]. Unfortunately, all of these remediation methods introduce additional contaminants into the subsurface. In contrast, the use of acoustic waves in groundwater remediation does not lead to further contamination of the subsurface environment. Acoustic waves have been successfully employed in enhanced oil recovery operations [*Beresnev and Johnson*, 1994] and perhaps can also be used to reduce the time and cost of groundwater remediation.

[3] The novel idea of using acoustic waves as a potential remediation method has only recently received attention. Since Biot's fundamental work [*Biot*, 1956a, 1956b] on elastic waves in saturated porous media, there have been significant developments on sound propagation in saturated porous media as well as porous media saturated by two immiscible fluids [*Santos et al.*, 1990; *Parra and Xu*, 1994; *Geerits and Kelder*, 1997]. However, to our knowledge, the possible enhancement of solute transport in water saturated porous media due to the presence of acoustic waves has not yet been investigated. This work focuses on experimental quantification of acoustic enhancement of solute transport in water saturated porous media.

2. Background

[4] Acoustic waves travel in saturated and unsaturated porous media by the propagation of small scale dilatations of the porous matrix. Because the scale of dilatation is small, the strain is reversible within the elastic limit of the porous medium, hence the term elastic waves. Early research on acoustic waves in saturated porous media was primarily focused on various aspects of wave propagation. For example, *Biot* [1956a] identified that elastic wave propagation in saturated porous media consists of a fast wave where the pore fluid and porous matrix move in phase and a slow wave where the pore fluid and porous matrix move out of phase. Because the pore fluid and porous matrix are moving out of phase in the slow wave, oscillatory flow of pore fluid with respect to the porous medium occurs.

[5] In a subsequent study, *Biot* [1956b] addressed the breakdown of Poiseuille flow at elastic wave frequencies greater than approximately 100 Hz and determined that the friction factor between the pore fluid and pore walls increases with increasing frequency of the elastic wave. In a similar study, *Qian* [1998] investigated the dynamic properties of viscosity in porous media and found that the effective viscosity increases with increasing acoustic frequency. *Zhou and Sheng* [1989] reported that the permeability of a porous medium decreases with increasing elastic wave frequency. *Cherskiy et al.* [1977] performed experiments on rock core samples and determined that the permeability increases with increasing sound intensity caused by a possible destruction of water films within the pore spaces. It should be noted that *Biot* [1956b], *Qian* [1998], and *Zhou and Sheng* [1989] did not consider a background steady flow component which was experimentally accounted for by *Cherskiy et al.* [1977].

[6] The effect of flow oscillations on solute transport in tubes has been investigated by several researchers. For example, *Watson* [1983] determined that solute dispersion effects along a pipe due to steady and oscillatory flow are additive. Oscillatory flow has also been found to influence dispersion in tubes. For example, *Aris* [1960] considered a background flow component for oscillatory flow in a tube and derived a Taylor dispersion coefficient containing terms proportional to the square of the amplitude of the pressure pulsations. However, the coefficients of these terms were found to rapidly approach zero with time. *Chatwin* [1975] found that for oscillatory flow in tubes, the axial dispersion of a solute depends on the frequency of oscillation and it is

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larger at low frequencies. Furthermore, *Jimenez and Sullivan* [1984] found that the diffusivity of a dissolved contaminant is a function of the kinematic viscosity of the interstitial fluid as well as the acoustic frequency.

[7] The effect of elastic waves, either by physical or acoustic vibration, on NAPLs (e.g., oils and solvents) in capillary tubes has also been investigated by various researchers. For example, *Kalinitchenko and Sekerj-Zenkovitch* [1998] observed, both experimentally and theoretically, that harmonic pressure oscillation leads to a steady state motion of the contact line between two immiscible fluids in a capillary. *Hilpert et al.* [1996] has shown that the meniscus between two immiscible fluids in a capillary tube can slip due to acoustic resonance.

3. Mathematical Formulation

[8] The one–dimensional transport of a non-sorbing solute or a conservative tracer in water saturated porous media under a constant hydraulic gradient can be described by the following linear, second–order partial differential equation:

$$\frac{\partial C(x,t)}{\partial t} = -U_e \frac{\partial C(x,t)}{\partial x} + D_e \frac{\partial^2 C(x,t)}{\partial x^2}, \qquad (1)$$

where C is the liquid phase solute concentration [M/L^3]; t is time [t]; U_e is the effective interstitial fluid velocity [L/t]; and D_e is the effective hydrodynamic dispersion coefficient [L^2/t]. The effective interstitial fluid velocity is defined as

$$U_e = U + U^*, \qquad (2)$$

where U is the steady state, background interstitial fluid velocity [L/t] and U^* is the additional velocity component attributed to acoustic pressure [L/t]. Similarly, the effective dispersion coefficient is defined

$$D_e = D + D^* = (U + U^*)\alpha_L + \mathcal{D}_e = U_e\alpha_L + \mathcal{D}_e, \qquad (3)$$

where $D = U\alpha_L + \mathcal{D}_e$ is the hydrodynamic dispersion coefficient [L^2/t]; α_L is the longitudinal dispersivity [L]; $\mathcal{D}_e = \mathcal{D} >/\tau^*$ is the effective molecular diffusion coefficient [L^2/t] (where $\mathcal{D}$ is the molecular diffusion coefficient [L^2/t], and $\tau^* > 1$ is the tortuosity coefficient [-]); and D^* is the additional dispersion component attributed to acoustic pressure [L^2/t]. It should be noted that the concept of effective parameters has been applied in numerous groundwater flow and solute transport studies [*Valocchi*, 1989; *Chrysikopoulos et al.*, 1990, 1992; *Kabala and Sposito*, 1991; *Chrysikopoulos*, 1995].

[9] For a finite system, the following initial and boundary conditions can be used [*Kreft and Zuber*, 1978]

$$C(x,0) = 0, \qquad (4)$$

$$-D_e \frac{\partial C(0,t)}{\partial x} + U_e C(0,t) = M_0 \delta(t), \qquad (5)$$

$$-D_e \frac{\partial C(L,t)}{\partial x} + U_e C(L,t) = U_e C_f, \qquad (6)$$

where $M_0 = M/A\theta$ is the mass injected over the cross sectional area of the column (where M is the injected mass [M], A is the cross sectional area of the porous medium [L^2], and θ is porosity [-]); δ is the Dirac delta function [1/t]; and C_f is the effluent flux concentration [M/L^3]. It should be noted that C corresponds to the in situ or resident concentration, whereas C_f is the flux concentration defined as the ratio of the solute mass flux to the volumetric fluid flux [*Kreft and Zuber*, 1978]. Initial condition (4) establishes a zero background concentration. Boundary condition (5) describes the flux influent pulse concentration. Boundary condition (6) describes the flux effluent solute concentration at the end of the packed column $x = L$. The solution to the governing equation (1) subject to conditions (4)–(6), for the effluent concentration of a one-dimensional packed column of length L is obtained by straightforward Laplace transform procedures to yield:

$$C(x,t) = \frac{xM_0}{2U_e t}\left(\frac{1}{\pi D_e t}\right)^{1/2} \exp\left[-\frac{1}{4D_e t}(x - U_e t)^2\right]. \qquad (7)$$

It should be noted that an analytical solution to a slightly different mathematical model than the one examined in this study has been previously presented in the literature [*DeSmedt and Wierenga*, 1979].

4. Experimental Design

[10] The effect of acoustic waves on solute transport in water saturated porous media was investigated in this study by injecting a bromide tracer pulse into a 30 cm long glass laboratory column with a 2.5 cm inner diameter (Kimble Kontes, New Jersey). The column was packed with 2 mm diameter glass beads (Fisher Scientific, Pennsylvania) that were retained in the column with teflon screens placed on both the influent and effluent sides of the column. The teflon column end caps were milled to accommodate 1/4 inch stainless steel fittings (Swageloc) for 3/8 inch semi–rigid plastic tubing (Fisher Scientific, Pennsylvania). Constant flow of degassed Millipore water at a rate of 1.48 ml/min was maintained through the packed column with a microprocessor pump drive (Cole Palmer Instrument Co., Illinois). Acoustic pressure was introduced into the column through the plastic tubing with a specially designed reservoir containing a pressure transducer (TST37; Clark Sythesis, Colorado). The frequency of the acoustic pressure oscillation was controlled by a frequency generator (LG Precision, California). Acoustic pressure levels were controlled by an amplifier (Lab Gruppen, Sweden) and measured with a PCB106b pressure sensor in conjunction with a signal conditioner (PCB Piezotronics, Inc., New York) and a digital multimeter (Metex, Korea). Effluent samples were collected from a dedicated needle (sampling port) within the effluent tube. A complete schematic of the experimental apparatus employed in this study is shown in Figure 1.

[11] The tracer solution was prepared by dissolving 256 mg of potassium bromide salt (KBr) (Fisher Scientific, Pennsylvania) into a liter of total solution volume to yield a final Br^- concentration of 172 mg/L. Alkali halides are the most commonly used salts for subsurface fluid tracing [*Chrysikopoulos*, 1993]. A slug of 0.6 mL of the tracer solution was instantaneously injected into the column by a side injection port midway down the column. Sample effluent volumes of 0.8 mL were collected at regular

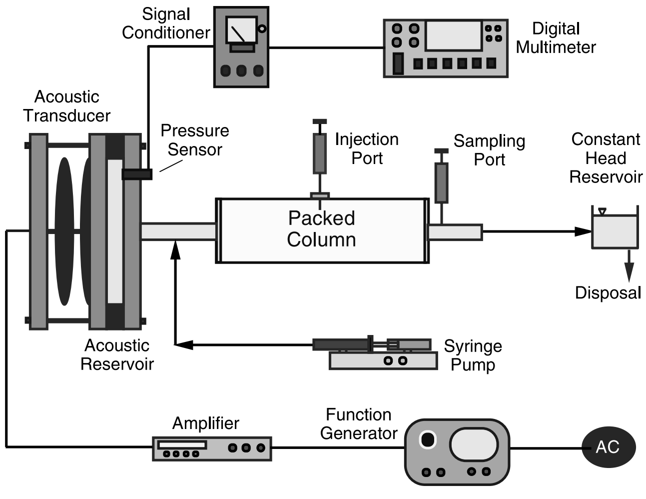

Figure 1. Experimental apparatus.

intervals using disposable 1.0 mL tuburculin plastic syringes (Becton Dickinson and Co., New Jersey). The Br^{-} concentrations of the liquid samples were determined using a Dionex DX–120 ion chromatograph (Dionex, California). The base case experiment was first conducted to determine background U, and α_L in the absence of acoustic waves (0 Hz). Subsequently, flowthrough experiments were conducted using the same procedure described for the base case, but in the presence of acoustic waves at ten different preselected acoustic frequencies.

5. Experimental Results

[12] The experimental Br^{-} breakthrough data for the base case are presented in Figure 2 (solid circles) together with

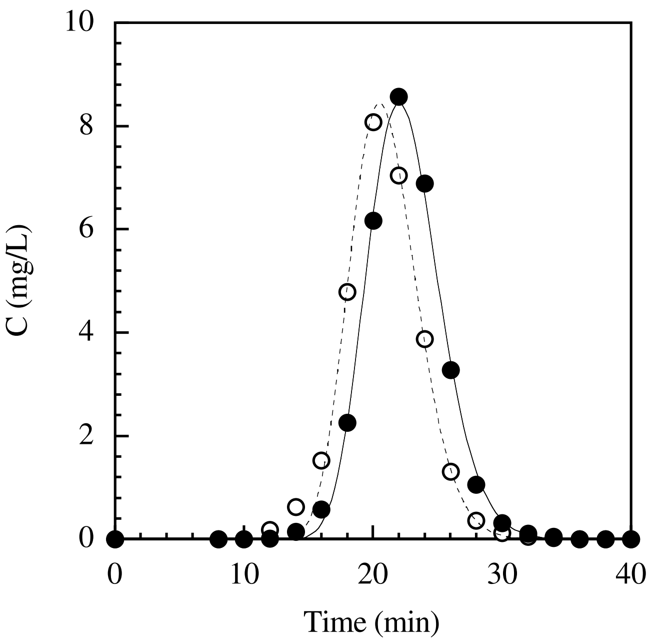

Figure 2. Tracer (Br^{-}) concentration breakthrough data (solid circles) and fitted model (solid curve) in the absence of acoustic pressure (base case) compared to breakthrough data (open circles) and fitted model (dashed curve) obtained in the presence of acoustic pressure at 60 Hz.

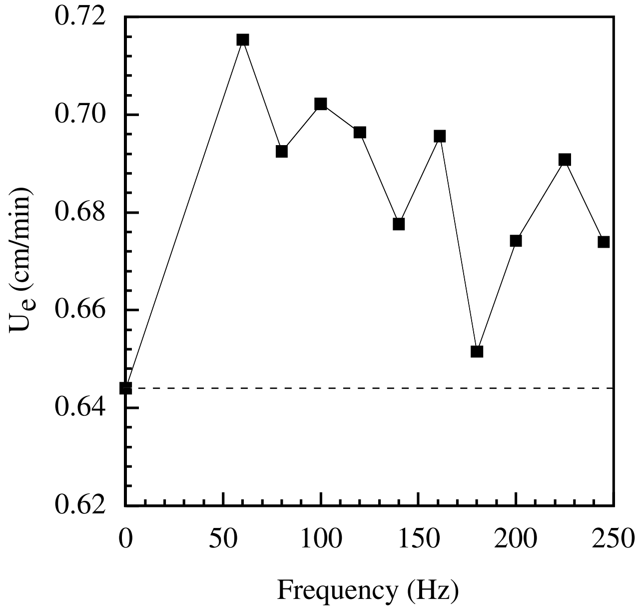

Figure 3. Experimentally determined effective velocity (solid squares) for various acoustic pressure frequencies. The dashed line represents the case of no acoustic waves present (base case).

the breakthrough data collected in the presence of acoustic waves with frequency 60 Hz (open circles). Clearly, the presence of acoustic waves leads to a faster breakthrough of the conservative tracer. The nonlinear regression subroutine *mrqmin* [*Press et al.*, 1996] was employed to estimate the dispersivity of the packed column α_L = 0.117 cm and the steady state background interstitial fluid velocity U = 0.644 cm/min, by fitting the analytical solution derived in this work, (7), to the experimental breakthrough data for the base case. The effective molecular diffusion coefficient of

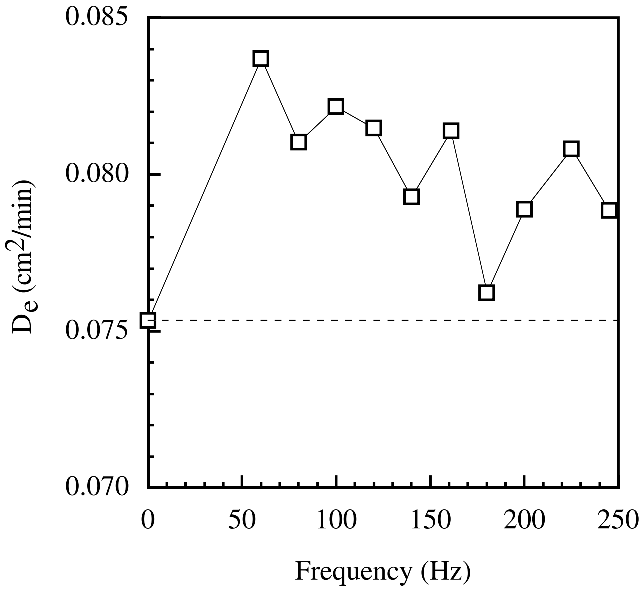

Figure 4. Effective dispersion coefficient (open squares) for various acoustic pressure frequencies. The dashed line represents the case of no acoustic waves present (base case).

Br^- used for the nonlinear regression procedure is $\mathcal{D}_e = \mathcal{D}/\tau^* = 8.62 \times 10^{-4}$ cm^2/min, or equivalently $\mathcal{D} = 1.2067 \times 10^{-3}$ cm^2/min [*Domenico and Schwartz*, 1990], and $\tau^* = 1.4$ [*de Marsily*, 1986]. Subsequently, the same fitting procedure was employed to determine the effective velocity (U_e), using a fixed value of α_L, for each of the ten different breakthrough data sets collected in this study in the presence of acoustic waves. The estimated effective velocity for the base case and the ten different frequencies ranging from 0 to 245 Hz at a constant pressure amplitude of 565 Pa are shown in Figure 3. The estimated effective dispersion coefficient for the base case and the ten different acoustic frequencies are shown in Figure 4. It should be noted that the values in Figure 4 were obtained directly from from (3) using the U_e values presented in Figure 3.

[13] Obviously, acoustic waves, particularly at low frequencies, enhance the tracer velocity and consequently enhance tracer dispersion (see Equation (3)). It should also be noted that due to experimental limitations in the low acoustic frequency range, no characteristic acoustic wave frequency was found to produce a maximum enhancement of the tracer transport. However, the enhanced interstitial fluid velocity appears to be approximately inversely proportional to the acoustic frequency.

6. Summary and Conclusions

[14] The experimental Br^- breakthrough data collected in this study indicate that acoustic waves enhance the transport of solutes in water saturated porous media. The degree of solute transport enhancement was found to be inversely proportional to the acoustic wave frequency. Due to experimental limitations, the characteristic acoustic wave frequency leading to a maximum enhancement of solute transport was not determined. However, it is assumed that this characteristic acoustic frequency, for the experimental conditions considered in this study, is within the range of the lowest frequency examined (60 Hz) and the base case (0 Hz). The experimental results suggest that further research should be undertaken to discern the governing mechanisms responsible for the enhanced transport phenomenon. The preliminary findings of this study however, may be regarded as the initial step to the development of a clean groundwater remediation method as an alternative to currently available remediation methods.

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E. T. Vogler and C. V. Chrysikopoulos, Department of Civil and Environmental Engineering, University of California, Irvine, California 92697, USA. (vogler@eng.uci.edu; costas@eng.uci.edu)