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ELECTROKINETIC WAVE PHENOMENA IN FLUID-SATURATED GRANULAR MEDIA

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ABSTRACT

Electrokinetic (EK) phenomena in sediments arise from relative fluid motion in the pore space, which perturbs the electrostatic equilibrium of the double layer at the grain surface. We have developed EK techniques in the laboratory to monitor acoustic wave propagation in electrolyte-saturated, unconsolidated sediments. Our experimental results indicate that as an acoustic wave travels through electrolyte-saturated sand, it can generate electric potentials greater than 1 mV. A careful study of these potentials was performed using medium-grain sand and loose glass microspheres for a range of pore-fluid salinities and ultrasonic frequencies. Experimental results are also shown to compare well with numerical and analytical modeling based on the coupled electrokinetic-Biot theory developed by Pride (1994).

INTRODUCTION

Most underwater imaging and naval operations require accurate tools for predicting the acoustical properties (for example, the attenuation and sound-speed dispersion) of the seabed. Experimentally derived, ad hoc viscoelastic fluid and solid models commonly used in ocean acoustics are often unable to predict how frequency-dependent behavior varies with sediment type because they lack a direct connection between micro- and macroscale properties. Models based on the Biot theory of poroelasticity [1] remedy this situation. Only Biot theory predicts the existence of relative motion between the fluid and solid skeleton on the macroscale. Hence one application of our work is to the question: Are seabed sediments best described by viscoelastic fluid or solid models, or poroelastic ones? Because relative fluid motion generates an EK effect that can be measured relatively easily, techniques based on it offer a unique means of assessing how successfully Biot theory (and its extensions) model sediments.

Poroelasticity relies on a separation of length scales, and necessarily regards fluid and solid phases as *coincident* on the macroscale (that is, each macroscopic material point is composed of both fluid and solid phases), as in Figure 1. A number of techniques can be used to derive Biot theory. We have focused on volume-averaging methods, which require the acoustic wavelength l to be much larger than the linear size L of the representative elementary volume (REV) over which averaging takes place. Moreover, L ought to be much larger than the characteristic size of the pores d so that a suitable average can be taken. These constraints result in the triple inequality under which Biot theory can be applied: $l \gg L \gg d$, which limits operating frequencies to below approximately 1 MHz in medium-grain sand.

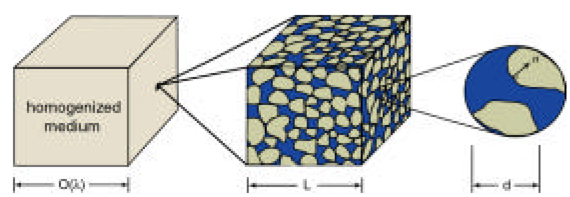

Figure 1. Hierarchical model implicit in two-scale homogenization. From left to right: (a) at the macroscopic scale, both phases are coincident when the medium is homogenized, (b) each material point consists of a volume cell made up of resolvable grains whose properties and dynamics are averaged over, and (c) the wall normal vector, n, at the solid interface.

Within each averaging volume, boundary conditions between the two microscopic phases lead naturally to interfacial coupling that acts as a source of "accumulated" viscous drag in the macroscopic equations (in the momentum equation, for example). This drag term is the origin of frequency-dependent sound speed and attenuation in ocean sediments. A similar analysis leads to additional features that account for electrokinetic coupling in the pore space.

Electrokinetic phenomena arise because an electric double layer forms near the grain surface, as shown in Figure 2. The bare surface of silicon dioxide (SiO_2, the prime constituent of glass and sand) often carries a small negative charge due to naturally de-protonated silanol groups ($Si\text{-}O^-$). When in contact with an electrolyte—say, NaCl in water—this surface charge creates an electric potential that affects the charge distribution in the surrounding fluid [2]. In the simplest case, counter ions (Na^+) in the pore fluid are attracted to and adsorbed by the negatively charged grains; they are bound chemically in an atomically thin, immobile layer. Further from the surface is a distribution of mobile counter ions in a diffuse layer. The electric potential at the interface between the immobile and diffuse layers is called the ? potential (generally negative and less than 1 mV), which is sensitive to the available binding sites at the grain surface, as well as to the electrolyte concentration and pH of the pore fluid.

A simple example of EK phenomena can be found in a fluid-filled silica capillary. An electric field (aligned parallel to the capillary wall) causes the ions in the diffuse layer to move, dragging the pore fluid along with it by way of viscous stresses. The reciprocal effect also exists: an applied pressure gradient will create both fluid flow and hence ionic convection current, which in turn, produces an electric potential. In order to study the analogous situation in poroelasticity at a macroscopic scale, a procedure must be used to determine the average acoustic and electromagnetic fields in the presence of a complex network of capillaries.

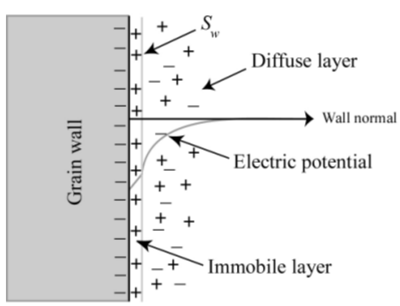

Figure 2. Electric double layer near a grain surface. Electrokinetics and fluid motion are coupled by the diffuse layer, which is free to move with the pore fluid.

Our research makes use of the coupled EK-Biot theory by Pride [3] and Haartsen and Pride [4], which is an extended form of Biot theory that couples Maxwell's equations to the fluid and solid governing equations at the microscale. We say that EK reception has occurred when an acoustic wave produces an electrokinetic voltage. While previous researchers used EK phenomena to study wave propagation and poroelasticity in porous rock and sandstones [5, 6], we designed and built a working EK reception apparatus to study coupled EK-Biot behavior in unconsolidated medium-grain sand and loose glass microspheres. The sediments had mean grain diameters of 250 and 350 microns, respectively. Solutions of NaCl in de-ionized water (with a range of conductivities) were used as saturating fluids to study the effect of conductivity on electroacoustic behavior. In each set of runs, a submersible acoustic transducer was used to transmit a 60 microsecond, 50 kHz sine wave burst within the experimental apparatus, as shown in Figure 3. Special Ag/AgCl electrodes, fixed above and below the sediment interface, were amplified 60 dB and monitored by a data acquisition PC. The entire assembly was connected to ground through a copper-mesh Faraday cage that encased the apparatus.

RESULTS

A typical set of time series for the Ag/AgCl electrodes are shown in Figures 4 and 5 for medium-grain sand. Note that the y-axis has units of microVolts. The first arrival is observed almost simultaneously at all of the electrodes in the vertical array. As our analytical and numerical modeling suggests, this arrival corresponds to a propagating electromagnetic (EM) wave that is generated when the incident acoustic pressure wave impacts the water-sediment interface. Relative fluid motion in the sediment near the interface acts as an effective current source (in the EM boundary conditions) to create EM waves that travel away from the interface at the speed of light in each medium. These wave types occur in addition to the usual mechanical waves that are transmitted into the sediment and reflected in the direction of the acoustic transducer.

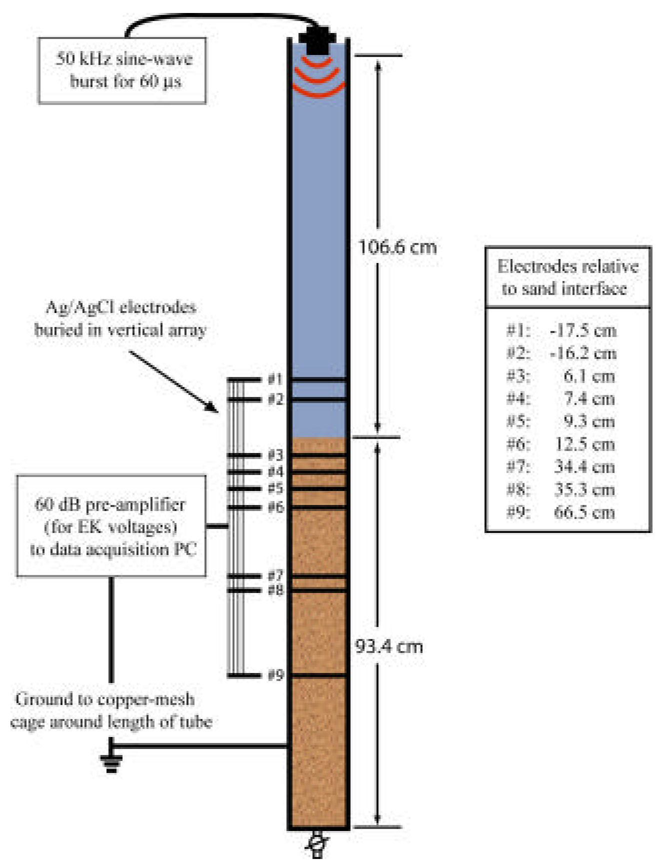

Figure 3. Electrokinetic reception apparatus. A PVC tube 7.62 cm in diameter and 2 meters long is used to hold salt water and salt water-saturated, medium-grain sand. Wave motion in the tube is monitored using special Ag/AgCl electrodes, which are positioned across the tube at a number of vertical positions.

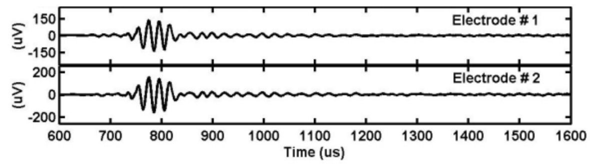

Figure 4. Electrode signals measured in water above the sediment. These time series show a single arrival, which occurs simultaneously at both electrodes and corresponds to an electric field generated as the acoustic wave impacts the water-sediment interface.

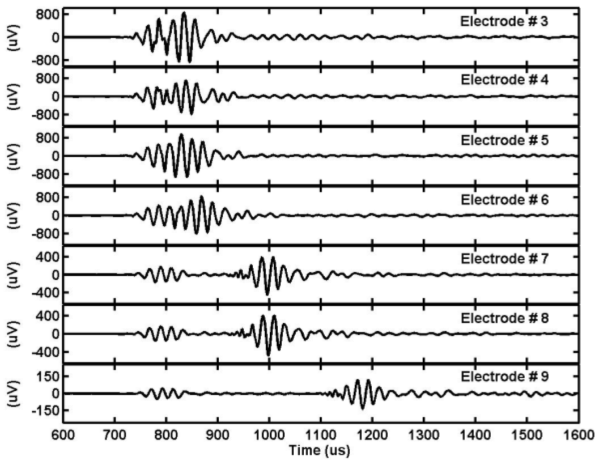

Figure 5. Electrode signals measured within the sediment. Time series from the buried electrodes depict two arrivals: The first arrival is simultaneous at all electrodes, and corresponds to an electric field generated as the acoustic wave impacts the water-sediment interface. The second arrival "moves out" in time for deeper electrodes, and corresponds to a compressional wave with a group velocity of about 1770 m/s.

There is also an electric field observed as the mechanical wave in the sediment passes each electrode. These arrivals are seen to "move out" in time for deeper electrodes. Using a chirped signal, we calculated group velocities in the sand and glass microspheres of about 1770 m/s, which are in the range of expected values for the Biot fast wave (akin to the usual compressional wave) in ocean sediments in this frequency range. We were unable to discern any higher-order modes or separately-traveling shear waves or slow waves on the electrode data. Because the second arrivals contain the dominant portion of the acoustic wave energy and travel essentially at plane-wave velocities, we treat them as plane waves in the numerical modeling.

Sand showed more variability than glass microspheres when initially saturated with weak electrolytes. Part of this phenomenon can be seen in the pore-fluid vs. bulk-sediment conductivity behavior depicted in Figure 6. We believe the variability may be due to a small fraction of clay or silt within the sand sample, which has the effect of changing the effective surface conductivity of the grains—EK-Biot theory, by itself, cannot account for this phenomenon [7, 8]. Nettelblad *et al* [9], and Wildenschild *et al.* [10] found similar behavior in saturated porous rocks and sediments with clay. We therefore used the unmodified form of EK-Biot theory derived by Pride [3] to model glass microspheres and EK-Biot theory with a constant surface conduction term (fitted from conductivity data) to model medium-grain sand.

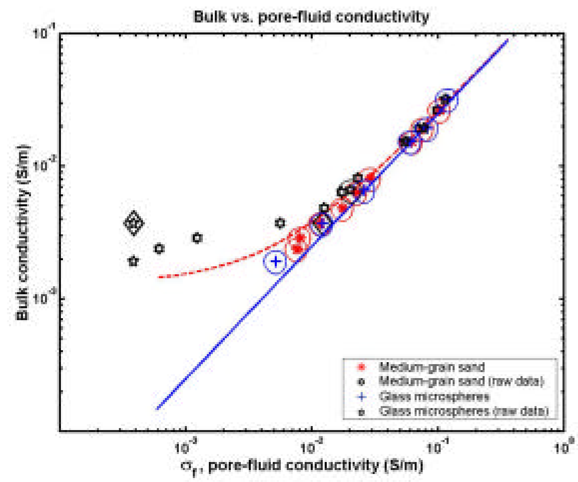

Figure 6. Bulk versus pore-fluid conductivity for medium-grain sand ("*") and loose glass microspheres ("+"). Unadjusted (raw) data points are indicated by symbols with holes in their centers. Predictions using the constant surface conduction model for sand (dashed curved) are compared to the unmodified form of EK-Biot theory for glass microspheres (solid curve). The diamond corresponds to glass saturated with pure, DI water after 16 hours of equilibration.

NUMERICAL MODELING

After solving the analytical problem of acoustic plane-wave reflection from an EK-Biot half-space, we developed predictions for the measured EK potentials that would be observed in the water and sediment over a range of frequencies, pore-fluid conductivities, and angles of incidence. (Wave motion in the apparatus pertains to normal incidence under this approximation.) Predictions also depended on separately measured, calibrated time series for the transmitted acoustic signal, as well as the mechanical and electrical properties of each medium. We used a simple inversion routine to determine Biot and electrokinetic properties of the sediments within the accepted ranges quoted in the ocean acoustics and surface chemistry literature. These values are summarized in Table 1.

The dependence of EK voltage on bulk-sediment conductivity was obtained by measuring the peak voltage at electrode #8 for runs using a range of NaCl solutions. Two datasets are compared in Figure 7: medium-grain sand ("star") and loose glass microspheres ("plus"). This data was normalized by the peak input pressure (from the transmitter) at the interface. Note that the y-axis has units of nanoVolts/Pascal, and that the x-axis has units of Siemens/meter (inverse resistivity). Predictions using the coupled EK-Biot theory (blue solid curve) for glass microspheres and EK-Biot theory with a constant surface conduction term (red dashed curve) compare well the experimental results. The constant surface conduction modification was required to match the down-turning trend in the sand data observed for small bulk conductivities.

ρ_f (kg/m^3)	ρ_s (kg/m^3)	η (kg/ms)	K_{fluid} (GPa)	K_{solid} (GPa)	
1000	2650	0.001	2.4	32.0	
K_{frame} (MPa)	G_{frame} (MPa)	ϕ	α	κ_f	κ_s
4.4x (1+0.06i)	2.9x (1+0.05i)	0.4	1.52	80	3
R_{sand} (μm)	R_{glass} (μm)	k_{sand} (10^{-12}m^2)	k_{glass} (10^{-12}m^2)	ζ (mV)	
16.0	19.0	8.0	11.0	-40	

Table 1. Mechanical and electrical properties of the water and sediment (medium-grain sand and loose glass microspheres. A common practice in poroelasticity is to use complex frame moduli to simulate inelastic damping between the grains.

SUMMARY/CONCLUSIONS

Medium-grain sand and loose glass microspheres were studied using pore-fluid conductivities that ranged between 0.0052 S/m and 0.12 S/m. Electrodes buried in the sediment measured two types of EK behavior: arrivals from an EM wave generated at the interface, which was recorded at all electrodes simultaneously, and electric potentials carried along with transmitted acoustic waves in the sediment. Electrodes above the water-sediment interface measured the EM-wave arrival in water, as well as a small disturbance of the electrode double layers caused by the incident acoustic wave.

Our central conclusions from both the data and theoretical analysis are that (1) electrokinetic disturbances recorded by the EK reception apparatus were robust and self-evident. In both medium-grain sand and glass microspheres, fast-wave potentials were often greater than 500 µV, while the EM-wave potentials were usually 100 µV in magnitude. These values correspond to efficiencies greater than 150 nV/Pa and 30 nV/Pa, at 50 kHz, respectively. (2) Our experimental data compares well to the results of Ahmed [11], who measured EK voltages generated by constant flow rates in sand and loose glass microspheres, and Pengra [3], who studied low-frequency EK behavior in consolidated porous media. (3) Both the constant surface conduction model and the unmodified form of EK-Biot theory were able to predict the trends and magnitudes of the laboratory data with good accuracy. The material parameters used to optimize the fits were reasonable and entirely within the expected ranges.

If the constant surface conduction model is accurate for clay-bearing sands and/or other geological materials—and there is experimental evidence to support this claim—then the true magnitudes of the electric potentials will deviate from those predicted by the unmodified form of EK-Biot theory, at least for weak electrolytes. In particular, caution should be exercised when using EM-wave amplitudes to infer the electrolytic properties of the saturating fluid, as is done in seismoelectric imaging of oil and gas reservoirs, for example.

With this caveat, coupled EK-Biot theory was found to be an accurate predictor of electrokinetic phenomena in granular materials. Moreover, since the ad hoc viscoelastic fluid and solid models commonly used in ocean acoustics do not predict relative fluid motion on the macroscale, they are incapable of predicting EK phenomena. The essential mechanism of relative fluid motion in poroelasticity is critical to understanding the acoustics of unconsolidated sediments.

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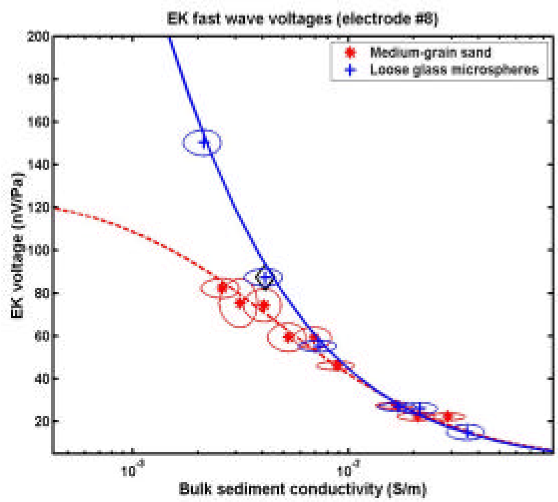

Figure 7. Peak electrode voltages, measured at electrode #8, versus bulk sediment conductivity. Data and predictions using coupled EK-Biot theory for loose glass microspheres ("plus" and solid blue curve) and EK-Biot theory with a fitted value for the surface conductivity for medium-grain sand ("star" and dashed red curve) compare extremely well.

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