Acoustic monitoring of sediment-pore fluid interaction

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Abstract. Acoustic and chemical measurements were made on water-saturated synthetic sediments undergoing diagenesis from uncompacted glass beads into a cemented glass bead, sandstone-like material. The sediment-water interaction was investigated as a function of time by monitoring the acoustic properties, water electrical conductivity, pH, and ion concentration. A reduction in the transmitted acoustic wave amplitude was correlated with a decrease in the total number of ions in the pore water. The lowest acoustic wave amplitude occurred when the ionic strength in the pore water had reached a minimum value. Calculation of the electrostatic potential showed that the force of repulsion between beads was strongest when the acoustic wave amplitude was at its minimum and the double layer electrostatic interaction potential between the beads was at a maximum value. At longer times the acoustic wave velocity of the transmitted wave increased as the cementation progressed. These results suggest that the chemical interaction between the pore fluid and grain alters the strength or stiffness of grain-to-grain contacts, which in turn alters the elastic modulus of the sediment frame. The important implications of this work are the potential use of acoustic or seismic techniques to monitor ion exchange between pore-fluids and sediments in-situ.

Introduction

Sediments in nature are subjected to chemical weathering that results in the dissolution and precipitation of minerals within pores and at grain contacts. If seismic methods are to be used to monitor contaminant transport or remediation, it is important to understand how sediment-water interactions affect acoustic wave propagation. Cementation of grains from precipitation will alter the strength and specific stiffness of grain contacts. Several investigators have studied the effect of grain contact models on seismic wave propagation through granular material. Murphy and Winkler (1984) observed that the absorption of vapor on internal surfaces of sandstone resulted in a decrease in surface energy and suggested from theoretical calculations that this would reduce the frame modulus. Dvorkin & Nur (1991) and Dvorkin (1994) found that the stiffness of a poorly consolidated system of grains does not depend on confining pressure, but on the stiffness and size of the cement layer between the grains. Nihei (1992) used a non-welded contact theory (Schoenberg, 1980; Pyrak-Nolte et. al., 1990) to examine wave propagation through poorly consolidated rocks. He found that seismic wave attenuation was a function of the number of contacts (i. e. grain size), the specific stiffness of the grain contact and the specific viscosity of the grain contact

(i.e. a dissipative mechanism). In this paper we present the results of a laboratory investigation on the effect of sediment-water interaction on acoustic wave propagation through synthetic sediments. Two water-saturated, homogenous, synthetic sediments with a mean grain size of approximately 70 microns were studied by making chemical and acoustical measurements over an extended period (450 hours to 1000 hours) to monitor changes in electrical conductivity, pH, pore water chemical composition, grain composition and acoustic velocities and amplitudes. Sediment-water interaction produced significant exchanges of Si, Na and Ca between the pore fluid and the grains, indicating dissolution and precipitation of oxides. The compressional wave amplitude increased and decreased depending on the exchange of ions between sediment and water. These data suggest that the chemical interaction between the pore fluid and grain alters the diffuse double layers and affects the strength or stiffness of grain-to-grain contacts, which in turn will alter the elastic modulus of the sediment frame.

Experimental set-up and procedure

In the experiments, fully-saturated unconsolidated synthetic sediments F1 and F2, were created from spherical soda-lime glass beads saturated with de-ionized water. Sample F1 was composed of beads that ranged in size from 53 to 150 microns with a mean size of 70 microns. Sample F2 was composed of beads that ranged in size from 63 to 72 microns with a mean diameter of 67 microns. The compositions of the beads for each sample was determined using an inductively coupled plasma mass spectrometer (ICP-MS) and are listed in Table 1.

For acoustic measurements, the beads were packed in an optical cuvette with a sample dimension of 60 mm by 60 mm with a sample height of 54.5 mm and 55.4 mm for samples F1 and F2, respectively. Acoustic measurements were made using a set of spherically-focused water-coupled piezoelectric transducers with the center frequency of 0.5 MHz to send and receive the signal. To make the acoustic measurements, the sample is placed in a water tank and the transducers are placed at normal incident to the sediment-water interface. Data are collected in one millimeter intervals along a centered scan line 20 mm in length and 20 mm from the edge of the cuvette. At each point, a waveform is recorded and stored on the computer. This method[4] [5] [8] yields a data set composed of one spatial and one temporal dimension.

To perform measurements of electrical conductivity and pH, the beads were packed in beakers with a bead to water weight

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Table 1. Percent chemical Composition Of Bead Samples F1 and F2

	F1 (%)	F2 (%)
SiO_2	68.570	71.065
Na_2O	16.007	14.483
MgO	4.3671	3.7006
Al_2O_3	0.4388	0.4388
CaO	10.036	10.009
Fe_2O_3	0.5813	0.3038

ratio of 0.35 for F1 beads and 0.30 for the F2 beads to simulate the bead/water packing used in the acoustic measurements. Conductivity and pH measurements were repeated four times for the F2 beads (Samples labeled HF2-1, HF2-2, HF2-3, and HF2-4) for a period of a 1000 hours and a single set of measurements were made for the F1 beads (sample labeled HF1-1) for a period of 450 hours.

The ion-concentration of the pore fluid was determined using (ICP-MS) on samples of pore fluid from HF2-1 and HF2-3 over a period of 464 hours. For each measurement, 5 milliliters of pore water were collected from the samples and 5 milliliters of deionized water were added to maintain the same water volume in the beaker. Each pore fluid sample was diluted 100 times with 2% HNO_3 to analyzed the Mg, Fe, Ca, and Al content, and was diluted 1000 times to analyze the Na and Si concentration.

Experimental results and analysis

Acoustic wave attenuation is obtained by measuring the change in the peak-to-peak (P-P) amplitude of a transmitted wave, i.e. the total voltage between the first and second antinodes. An average P-P amplitude was calculated by averaging the measured P-P amplitude for the 20 waveforms collected along the scan line. The change in P-P amplitude was calculated relative to the first measurement. For both samples, the change in P-P amplitude (Figure 1) is non-monotonic in time. The data from both samples exhibited a minimum around 80 hours. Prior to 80 hours, the compressional wave amplitude decreased. After the observed minimum amplitude at 80 hours, the P-P amplitude increased monotonically. The magnitude of the minimum P-P amplitude differed between the F1 and F2 samples. F1 sediments exhibited a decrease in P-P amplitude of 60%, while F2 sediments showed a 15% decrease.

In the initial 30 hours, sample F1 and F2 showed a increase in velocity of 0.5% and 1.7% respectively, and then increased very slowly (Figure 2) up to 100 hours and 400 hours, respectively. The initial increase in velocity is related to the compaction rate of the sediments. During the first 30 hours, all of the samples exhibited compaction of 0.6mm out of 55mm sample depth caused by the weight of the beads (Figure 3). After this time, the compaction length appeared to reach a fixed value. The compaction amount that occurred in the first 30 hours is equivalent to a reduction in porosity of 1%. This reduction in porosity altered the effective modulus of the sediments, and thus
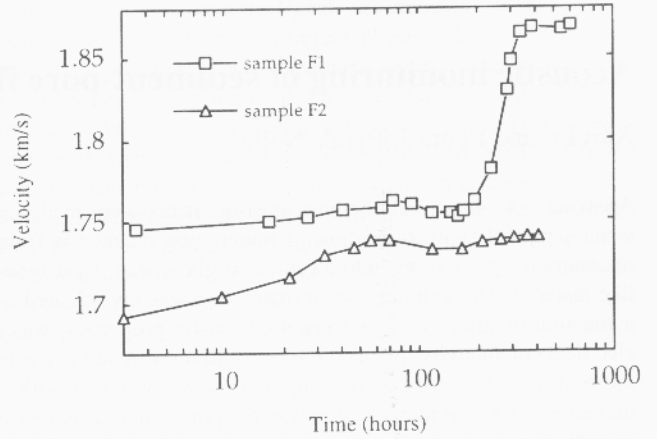

Figure 2. The acoustic wave velocity variation as a function of time for F1 and F2 samples.

affected the velocity. For F1 beads, the wave propagation velocity was observed to increase again after 200 hours. This indicated that cementation occurred at the grain contacts from precipitation of silica oxides out of the pore water. The cement increased the particle contact area and contact stiffness which reduced the relative movement between grains.   These data suggest that the acoustic wave velocity is a sensitive indicator of the compaction and cementation. However, the velocity data did not show any variation around 80 hours when the wave propagation amplitude displayed a minimum value.

Physico-chemical measurements were made to monitor the chemical interaction between the sediment and the pore-water. Electrical conductivity measurements provide information on the ion concentration in the water (Figure 4). During the first 3 hours, the conductivity of the pore water was relatively high (up to 1950 microsiemens for F1 beads and around 450 microsiemens for F2 beads) because of the release of surface alkalis from the beads into the water. After 3 hours, the conductivity decreased sharply to only 45% -55% of its initial value indicating that some surface precipitation had occurred. From 80 hours to 200 hours, the conductivity was relatively constant, that is ions were being released from and precipitated onto the beads. After 200 hours, the conductivity increases and the dissolution of the beads dominates. The pore water from the F1 beads exhibited a higher
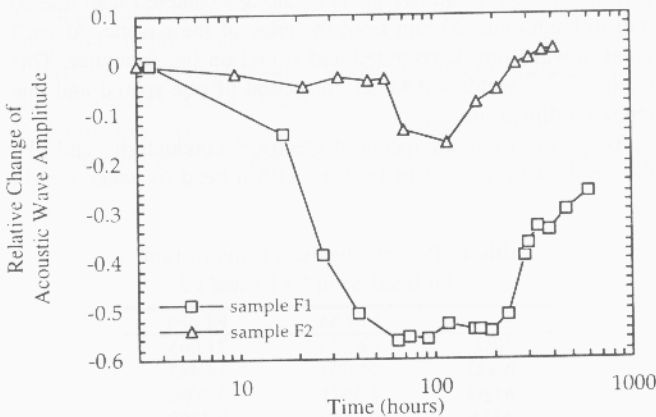

Figure 1. Relative change in the Peak-Peak acoustic wave amplitude as a function of time for F1 and F2 samples.
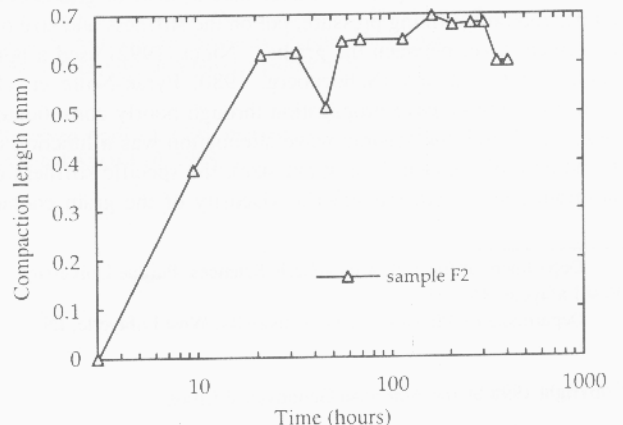

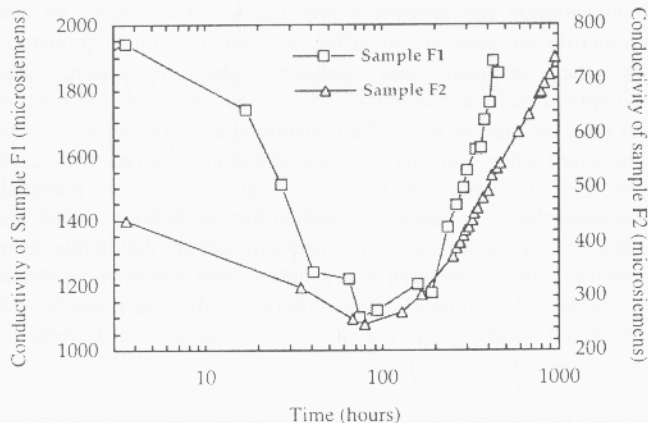Figure 3. The compaction length as a function of time for F2 sample.

Figure 4. Pore water electrical conductivity as a function of time for F1 and F2 samples.

conductivity than pore water from F2 bead samples, which suggests that the F1 beads may have a higher solubility than the F2 beads. This is supported by the chemical composition data of the F1 and F2 beads listed in Table1. The F1 beads have a slightly lower SiO_2 content and slightly higher levels of Na_2O, CaO and MgO compared to the F2 beads. These alkaline elements are easier to release into water than silicon. The higher solubility of the F1 beads compared to F2 beads may have affected the magnitude of the P-P amplitude change. Initially, the pH of the pore water is high (11-12), and then decreased over time because the formation of surface metal-complexation and metal-hydroxide consumed the OH⁻ in the pore water and caused a reduction in surface energy. After 300 hours the pH stabilized, resulting in the maintenance of a constant negative surface charge. To support the conductivity and pH data interpretation, ICP-MS analyses were performed on the pore water samples of F2 beads to determine the ion species and the ion concentrations in the pore water as a function of time. From the ICP-MS analysis it was determined that the beads initially released Na^+ Ca^{2+} and Si^{4+} into the water. At 80 hours, the Ca^{2+} and Al^{3+} concentration in the water decreased which implies that some minerals formed by these ions were precipitated. After 80 hours, the release of Si^{4+} and Na^+ ions accelerated, which implies that the

rate of dissolution of the glass beads increase with time. The ICP-MS data for the pore water show that two stages of intergrain precipitation occurred. The first stage was rapid with the metal ions precipitating onto the beads. The amount of precipitation was very limited which can be calculated from the concentration of the ions that precipitated from the water. The second surface stage is slow and lasts longer, resulting in the precipitation of silica cement from the solution.

In the first stage of surface precipitation, when the beads are not cemented together, we hypothesize that the electrostatic interaction forces between the beads changes with time which results in the decrease and increase of the acoustic wave amplitude. The electrostatic interaction forces between two glass beads in solution includes the Van Der Waals force and electrical-double layer electrostatic force. Between similar materials the Van Der Waals force is always attractive. According to Lifshitz theory[3], the Van Der Waals interaction potential (W_v) between two surfaces can be calculated from:

$$W_V = -\frac{A}{12\pi D} \quad [3] \tag{1}$$

Where W_v is the interaction potential per area (Jm^2), where a negative sign indicates attraction; A is the Hamaker constant, which is about 10^{20} J; and D is the distance between two surfaces. The double-layer electrostatic force is caused by the surface electric charge of the glass beads. Based on the surface chemistry, the glass is negatively charged on the surface when glass is in a solution with a pH greater than $pH_{pz}(2\sim3)$[12] when the surface net charge equals zero. From our water-bead system, the pH value was always greater than 9.61, indicating that the surfaces of the glass beads were always negatively charged. The negative surface charge is balanced by an equal but oppositely charged region of counterions, some of which are bound (Stern layer)[12], usually transiently, while other ions form a cloud of ions in rapid thermal motion close to the surface, known as the diffuse electric-double layer. The similarly charged surfaces repel each other electrostatically in solution. The electrostatic repulsive potential (W_e) between two surfaces is calculated from:
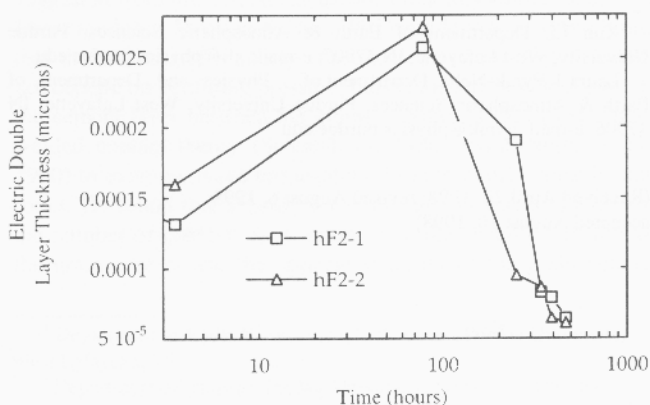

Figure 5. The electric double layer thickness of the glass beads as a function of time. The largest layer thickness occurred at 80 hours for both samples. The increase in the electric double layer thickness weakens the contact between the beads which will affect the mechanical behavior of the grain contacts.
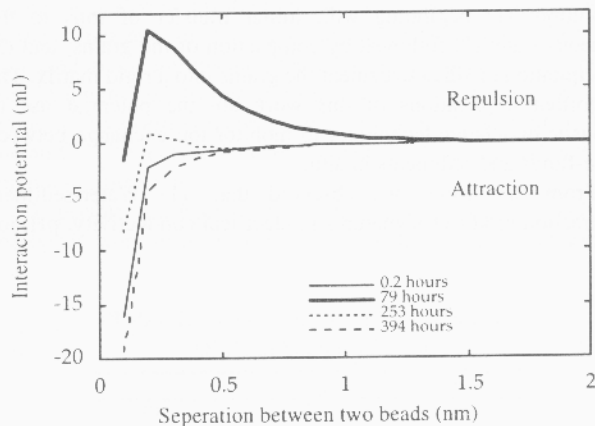

Figure 6. Interaction potential versus seperation profiles between two beads for the sediments at different time. Constant surface potential of 100 mv is assumed. At 80 hours, the surfaces repel strongly, a big energy barrier exists at about 0.2 nm.

$$W_e = \frac{64KT\rho_\infty\gamma^2}{\kappa}\exp(-\kappa D) \qquad (2)$$

$$\gamma = \tanh(\frac{Ze\varphi_0}{4KT})$$

$$\kappa = \sum_i (\frac{\rho_x e^2 Z^2}{\varepsilon\varepsilon_0 KT})^{0.5}$$

Where $1/\kappa$ is the Debye length (or "electrical double layer thickness"); K is Boltzmann's constant (1.381×10^{23} JK^{-1}); T is the temperature (293 K); ρ is the number density of the ions in the solution (m^{-3}) ; D is the separation of two surfaces (m); Z is the ion valency; e is electronic charge (-1.602×10^{19} C); φ_0 is the surface potential; ε is the dielectric constant of water (80 at 293K); and ε_0 is the permittivity of free space (C^{2}J^{-1}m^{-1}). Because the pH value of the pore water was always greater than 9, which is far above pH$_{pzc}$ (2~3), this suggests that the surface of glass is highly charged, and that the surface potential can be assumed to be constant with time. The total electrostatic interaction potential between two beads is calculated from

$$W = W_e + W_v \qquad (3)$$

Figure 5 shows that, the electrical double layer thickness increased during the initial 80 hours as the ion concentration in the pore water decreased. After 80 hours, the double layer thickness decreased as the ion strength increased in the pore water. From the calculated interaction potential (Figure 6), it is clear that the electrostatic interaction potential is strongly repulsive at 80 hours when the acoustic amplitude reached a minimum. The double-layer electrostatic force potential overcame the Van Der Waals attractive potential for separations less than 1nm and formed a relatively high energy barrier at about 0.2 nm. As a result, the glass beads were repelled. The increase in repulsive force between beads may have weakened the contacts between the beads and reduced the contact stiffness. A reduction in contact stiffness would result in a decrease in acoustic wave amplitude.

Summary

The combined acoustic and chemical analyses of synthetic sediments enabled tracking of several different stages in the rock evolution: (1) beginning with initial transfer of ions to the aqueous state; (2) followed by compaction of the grains; and (3) precipitation of silica to cement the grains into a rigid matrix. The important implications of this work are the potential use of acoustic or seismic techniques to monitor ion exchange between pore-fluids and sediments in-situ.

From this work, we observed that: (1) Water-sediment interaction produces signatures in electrical conductivity, pH, ion concentration and acoustic signal. (2) Acoustic amplitudes and velocities are sensitive to different physico-chemical properties of the sediment and sediment-water interaction. The compressional wave amplitudes increased and decreased relative to the exchange of ions. The transmitted acoustic wave velocity increased when compaction or cementation occurred. (3) Based on the calculation of the electrostatic interaction potential between the glass beads, it is shown that at 80 hours, when the total ions in the pore water reached a minimum, the double layer repulsive force exhibited a maximum. This force of repulsion weakened the contact stiffness between the glass beads and produced a reduction in the contact stiffness and the decrease of wave amplitude.

Reference

Dvorkin, J., Mavko, G., and et al., The Effect of Cementation on the Elastic Properties of Granular Material, Mechanics of Materials, 12, 207-217, 1991.

Dvorkin, J., Effective Properties of Cemented Granular Materials, Mechanics of Materials, 18 (4), 351-366, 1994.

Israelachvili,J, Intermolecular & Surface Forces, Academic Press Inc., San Diego, 1992.

Li, X., Acoustic Monitoring of Sediment-Water Interaction, M.A. Thesis. University of Notre Dame, 79pp, Nov. 1997.

Mullenbach, B. L., Acoustic Imaging of Sediments, M.A. Thesis. University of Notre Dame, 140pp, July, 1996.

Murphy III, F. M., Winkler, K. W., and et al., Frame Modulus Reduction in Sedimentary Rocks: The Effect of Adsorption on Grain Contacts, Geophysical Research Letters, 1(9), 805-808, 1984.

Nihei, K. T., Micromechanics of Seismic Wave Propagation in Granular Rocks, PHD Thesis, University of California at Berkeley, 167pp, Dec. 1992.

Nagy, P. B., Bonner, B. P. and Adler, L., Geophysical Research Letters, 22, 1053-1056, 1995.

Pride, S. R. and Morgan, F. D., Electrokinetic Dissipation Induced by Seismic Waves, Geophysics, 56(7), 914-925, 1991.

Pyrak-Nolte, L. J., L.R. Myer, and N.G.W. Cook, Transmission of seismic waves across single natural fractures, J.Geophys.Res., 95 (B6), 8617-8638, 1990.

Schoenberg, M. Elastic wave behavior across linear slip interfaces, J. Acoust. Soc. Am. 68(5), 1516-1521, 1980.

Werner, S., Chemistry of The Solid-Water Interface, NewYork, Wiley, 1992.

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