Laboratory investigation of seismic effects of nanoparticle dispersions in saturated granular media

Étude en laboratoire des effets sismiques des dispersions de nanoparticules dans les milieux granulaires

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ABSTRACT: Nanomaterials used in industrial applications and consumer products are widespread, thereby increasing the likelihood of unintended environmental release. The fate and transport of nanoparticles in the environment and their effects on the environment and human health are not well understood. This research investigates the potential to use seismic methods for such fate and transport studies. A test cell using piezoceramic bender elements was constructed to investigate how nanoparticles dispersed in the pore fluid of a saturated glass bead medium affect seismic wave propagation. Test cell design addresses optimal seismic wave propagation, uniformity and repeatability of the placement of the granular media and uniformity of fluid flow. Time histories were produced from two tests optimized for shear wave propagation. The first (baseline) test used deonized (DI) water. This test demonstrated the need to stabilize the sample before making measurements by first flushing several liters of liquid through the system. The second test, conducted on a new sample, used a solution of 0.05% nano Zinc Oxide (nZnO) in DI water after first flushing with DI water. Comparison of results between the two tests shows only weak repeatability between test specimens. Despite this, results of the second test still indicate a significant change in response in the presence of nZnO, particularly in signal amplitude. Studies are ongoing to increase experimental reliability and sensitivity, and to more closely approximate expected field conditions.

RÉSUMÉ : Les nanomatériaux utilisés dans les applications industrielles et produits consommation sont très répandus ; il est donc probable que ces substances se retrouvent disséminées dans l'environnement. Le destin et le transport des nanoparticules dans l'environnement et leurs effets sur l'environnement méritent un étude approfondie. Cet article étudie la possibilité d'utiliser des méthodes sismiques pour étudier ces effets. Une cellule a été construite pour voir comment les nanoparticules dispersées dans le fluide interstitiel du verre change la propagation d'ondes. La conception de la cellule d'essai permet d'étudier la propagation des ondes sismiques, le positionnement des milieux granulaires et l'uniformité de l'écoulement du fluide. Les temps de parcours de la propagation des ondes sismiques dans une dilution d'oxyde de zinc nano 0,05 % (NZnO) dans une matrice de billes de verre sont présentés et comparés à ligne de base. Nous avons trouvé une légère réduction de la vitesse de cisaillement et de compression en présence de NZnO par rapport aux valeurs initiales. Nous proposons des études plus complexes qui se rapprocheraient des conditions dans la nature.

KEYWORDS: nanoparticles, seismic, fate and transport, piezoceramic, bender elements.

1 INTRODUCTION

The use of nanoparticles in industrial applications and consumer products has become widespread and continues to grow. As applications of nanoparticles increase, so does the likelihood of unintended environmental release, including the possibility of a large-scale spill event. The fate and transport of nanoparticles dispersed in the environment are largely unknown (Conlon, 2009; Klaine et. al., 2008), and their effects on the environment and human health are also not well understood. Consequently, methods are needed for detecting, characterizing, and monitoring subsurface transport of nanoparticles. The capability of electrical geophysical methods has shown some promise in the spectral induced polarization (SIP) response to select nanoparticles in saturated sand laboratory columns (Joyce et. al., 2012). This paper investigates the seismic response to nanoparticles in a similar laboratory setting, in order to complement the SIP results and evaluate another geophysical method.

We have developed a test cell that uses piezoceramic bender elements to investigate how nanoparticles dispersed in the pore fluid of a glass bead matrix can affect seismic wave propagation characteristics. To minimize chemical interactions between the granular medium and the nanoparticle solution and to provide uniform grain morphology, non-reactive glass beads are used for the granular medium. Seismic wave characteristics (spectral content, travel time, signal amplitude) can be scrutinized for distinguishing characteristics. Results may suggest whether seismic methods are suitable for nanoparticle fate and transport studies.

This paper reports on the test cell design and development of experimental procedures. Some preliminary travel time and signal amplitude results using 0.05% nZnO solution are included.

2 TEST CELL DESIGN AND TESTING PROTOCOLS

The test cell design is based on preliminary work by Rajabdeen et al. (2012). The sample or experimental treatment housing is a translucent 15.2-cm inner-diameter PVC cylinder with custom end caps that are fitted with piezoceramic bender elements (Figure 1). The bender elements serve as seismic transmitter and receiver. The elements used in this study are two-piezo layer transducers made with PSI-5A4E piezoceramic (a Lead Zirconate Titanate (PZT) piezoceramic), parallel-poled, nickel electrodes and using brass center reinforcement (Piezo Systems Inc.).

The bender elements are 12.7 mm square and 0.5 mm thick. Elements are potted in vinyl caps using epoxy which are placed inside small-diameter PVC tubes that pierce and affix to the centers of the end caps (Figure 1). This configuration is intended to be robust while also creating impedance traps to encourage transmission of seismic energy through the sample rather than through the test apparatus. The cantilever length (protrusion into the sample) is 4 mm, approximately one-third the length of the element. A short cantilever length reduces dependency of the system resonance frequency on the sample matrix properties (Lee and Santamarina, 2005). The bender elements are installed in-plane such that the S-wave energy propagates along a direct, straight-line path while the strongest P-wave arrival at the receiver would be reflected from the cylinder walls (Figure 1). The test cell design integrates the same considerations of signal attenuation and near-field effects that have been applied to bender-element testing in oedometers (e.g., Dyvik and Olsen (1991), Zeng and Ni (1998), Lee and Santamarina (2005), Lee et al. (2007)). The bender element tip-to-tip separation is 6.2 cm, yielding a ratio of test specimen diameter to sensor separation of approximately 2.45. The sample height is 15.8 cm. Figure 2 contains a view of the test system including key ancillary equipment.

Glass beads used for the granular testing matrix are 0.5 mm in diameter. Consideration is given to optimize uniformity and repeatability of the placement of the glass beads, in order to minimize unwanted acoustic impedance contrasts in the medium, to discourage preferential fluid flow pathways, and to facilitate comparisons among tests. The oven-dried glass beads are vibrated into place using a vibratory table operating at 60 Hz for 20 minutes with a surcharge load of 28 kg. The surcharge mass was selected based on the methodology laid out by ASTM D4253 - 00(2006) Standard Test Methods for Maximum Index Density and Unit Weight of Soils Using a Vibratory Table. The surcharge mass is kept in place throughout testing. It approximates a vertical overburden stress of 15 kPa.

The glass beads are flooded with de-ionized (DI) water which is introduced through upward flow distributed across the cross-section of the experimental column. The water is plumed into the bottom of the test chamber to reduce the amount of entrapped air and is plumed under gravity flow with total head not exceeding 0.3 meters. The fluid-flow gradient is kept low to discourage entrainment of air bubbles and approximate laminar flow. The fluid passes through a set of baffles and a perforated disc before entering the sample (Figure 1). This process is intended to encourage a uniform wetting front and discourage fingering and the formation of preferential flow pathways. A somewhat loose-fitting top cap allows the cell to be completely flooded and allows the overburden stress to act directly on the specimen. Excess water exits the test apparatus through a port above the cap. Satisfactory dispersion of liquid through the glass-bead-filled test chamber was demonstrated using colored dye (Figure 3). The capacity of the fluid-delivery system including test cell is approximately 1.7 liters.

Data are collected with a Data Physics (Data Physics, Inc.) dynamic signal analyzer. Single sine pulses are created using a function generator. The sampling interval is 9.301 microseconds. The signal is not filtered during data capture. The reported result is an average from 350 pulses, which are repeated at 0.7-second intervals.

3 BASELINE TESTING

Through resonance testing and experimental trials, optimum frequencies to test for shear and compression were determined to be 1 kHz and 8 kHz, respectively. The compression measurements have not yet been resolved satisfactorily because of complications with electrical crosstalk between source and receiver and are not presented here. Results of baseline testing for shear (using DI water, with no experimental treatment) are shown in Figure 4. Four datasets are collected. The first dataset is collected after initial inundation of the sample. Repeat collections occur after 4, 8 and 12 additional liters of DI water are flushed through the sample. The time history from the initial measurement is dramatically different from those collected later. Results demonstrate that up to four liters of fluid need to be flushed through the system before the response stabilizes. The flushing process is likely to improve the signal by expelling entrained air.

The zero time in Figure 4 corresponds to source initiation. Based on first arrival by visual interpretation, the shear wave velocity of the specimen is approximately 170 m/s. Considering the timing for the first troughs in the signals, velocity estimates among the three measurements (after flushing 4, 8 and 12 liters of DI water) would vary by approximately +/- 3 %. Amplitudes at the first trough vary by +/- 17%.

4 NANOPARTICLE EFFECTS

The test sequence was repeated on a new sample, in which a solution of 0.05% (by weight) nZnO solution was introduced. The nanoparticle treatment solution is made through sonication to uniformly disperse the nanoparticles in DI water. Eight liters of clean DI water were flushed through the test chamber first, followed by 4 liters of the nanoparticle solution.

Results are shown in Figure 5. Again, the dataset conducted before flushing was not representative of results after flushing. The time histories collected after flushing but before introducing the nZnO treatment would ideally be the same as those collected in the baseline test. However, comparison with Fig. 4 demonstrates that the tests are not closely repeatable between samples, despite careful efforts to replicate test conditions. For example, consider the difference in signal amplitude between the two figures. This is an unfortunate discrepancy that requires further investigation. From Figure 5, considering the time histories gathered after flushing 4 and 8 liters of DI water, the shear wave velocity is approximately 150 m/s, which represents a decrease of 12 % with respect to baseline (Figure 4). Also, for the later test (Figure 5), results do not stabilize as fluid flushing occurs to the extent seen in baseline testing. Repeating analyses of the timing and amplitude of the first trough, velocity estimates among the measurements after flushing 4 and 8 liters would vary by approximately +/- 9 % and amplitudes by +/- 3 %. With respect to the baseline test, the percentage variabilities are higher for velocity and lower for amplitude.

Despite the variability observed with DI water, the trial conducted after introducing the nZnO experimental treatment (labeled in Figure 5 as “12”) shows distinctly different results from those collected earlier on the same test specimen. Signal amplitude is dramatically increased, and timing is delayed. To effectively quantify differences, cross-correlation of traces, signal matching or coda wave analysis might be applied (Lee and Santamarina 2005, Dai et al. 2012).

The significant increase in signal amplitude might be due to agglomeration of nZnO onto the glass bead matrix (Klaine et. al., 2008) which would enhance contacts in the skeletal structure of the glass bead matrix, thereby reducing signal attenuation. The smaller decrease in velocity is not as readily explained. One hypothesis (which is yet untested) is that the flushing process allows sufficient displacement of the glass beads to permit introduction of nanoparticles between contacts. This possibility seems unlikely given the slow influent flow rate and the amplitude of the surcharge pressure.

5 CONCLUSIONS

A laboratory apparatus has been developed to study seismic response to the presence of nanoparticles dispersed in the pore fluid of saturated granular media. Uniform placement of the solid sample matrix is encouraged through the use of a vibratory table and a surcharge load. A perforated plate and baffle system for introduction of fluid at the bottom of the test cell encourage laminar flow and uniform fluid distribution which is demonstrated using dye. Baseline testing in clean water using sine pulses at 1 kHz, chosen to optimize transmission of shear energy, demonstrated that by first flushing several liters of fluid through the sample, repeated measurements would yield similar results. Testing a new sample demonstrated that close repeatability of results is not assured between samples.

The measurements revealed sensitivity of the nanoparticle treatment to shear wave energy propagation. After flushing a sample with a treatment containing 0.05% nZnO, measurements revealed a dramatic increase in signal amplitude and a slight increase in travel time. Changes might be due to agglomeration of nanoparticles on the granular matrix which enhances grain-to-grain contacts, possibly coupled with insertion of nanoparticles between grains which reduce system stiffness.

Further experimentation is required to assess if the observed responses are indeed trends. Once accomplished, the research will expand to increase experimental sensitivity and broaden scope, and to more closely approximate conditions in nature.

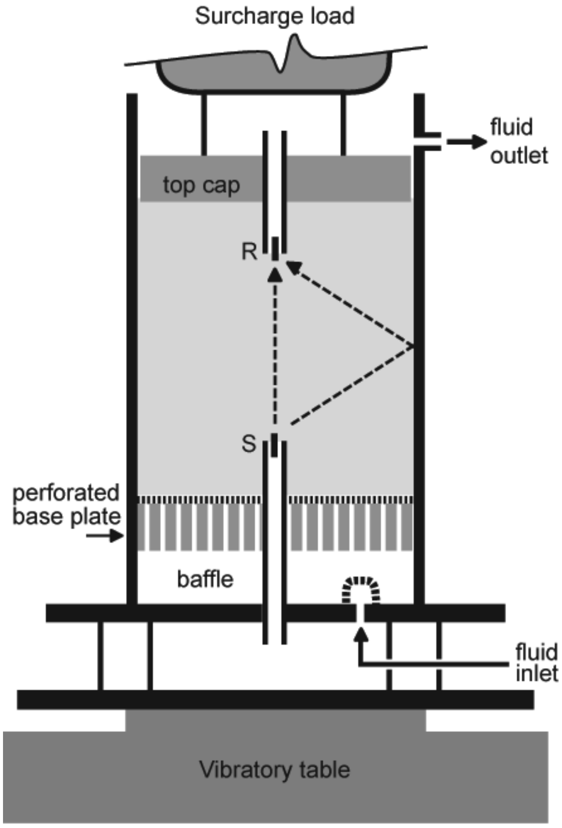

Figure 1. Schematic cross section of test cell. S, R: source and receiver bender elements, respectively. Direct-transmission S-wave path and reflected P-wave path are shown.

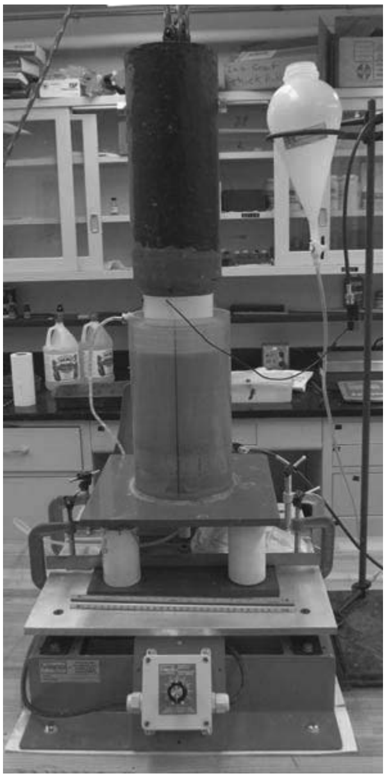

Figure 2. Photo of test cell and ancillary apparatus. Test cell is in center, situated on vibratory table. Surcharge mass is suspended from above. Reservoir for gravity feed of fluid appears as white container in the upper right of the photo.

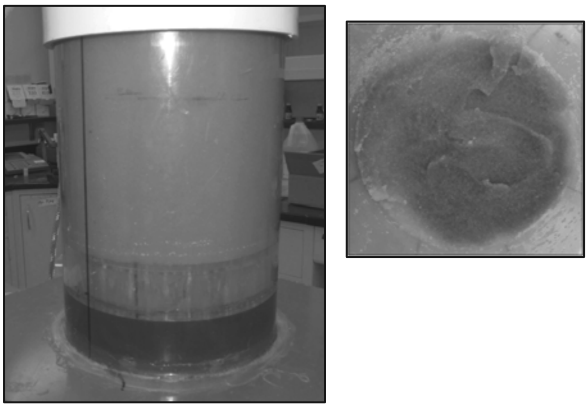

Figure 3. Dye test demonstrates uniformity of distribution of influent. Left: View of column from side. Fluid-filled baffle zone is indicated by dark shade at bottom; just above is perforated base plate. Dye appears to be distributed evenly throughout the sample. Right: View from above of glass beads inside test cell during disassembly.

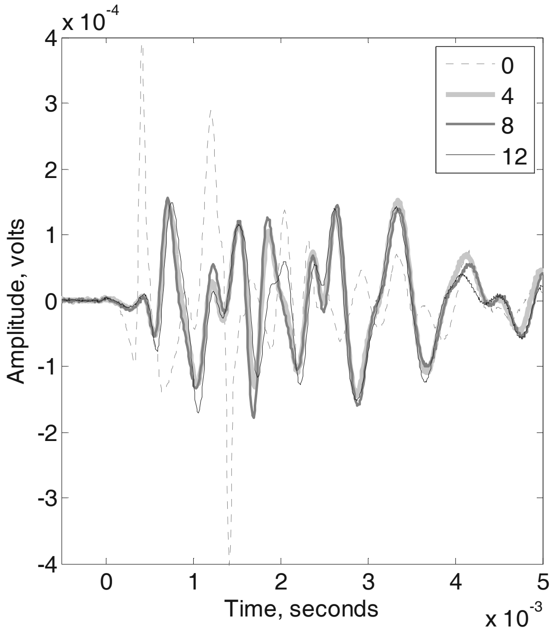

Figure 4. Results of first test: baseline. Legend refers to volume of DI water flushed through the specimen before testing, in liters.

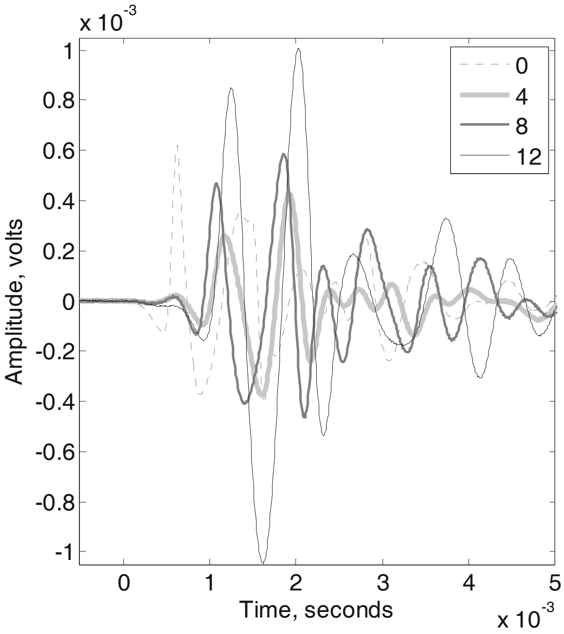

Figure 5. Results of second test. Legend refers to volume of fluid flushed through the specimen before testing, in liters. First eight liters were DI water; final four liters contained experimental treatment (i.e., trial marked "12" describes effect of experimental treatment). Note difference in vertical scale with respect to Figure 4.

6 ACKNOWLEDGMENTS

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