Acoustically Enhanced Remediation of Contaminated Soils and Ground Water

Volume I

Topical Report

October 1995

RECEIVED
APR 24 1997
OSTI

Work Performed Under Contract No.: DE-AR21-94MC30360

For

U.S. Department of Energy
Office of Environmental Management
Office of Technology Development
1000 Independence Avenue
Washington, DC 20585

U.S. Department of Energy
Office of Fossil Energy
Morgantown Energy Technology Center
P.O. Box 880
Morgantown, West Virginia 26507-0880

MASTER

By

Weiss Associates
5500 Shellmound Street
Emeryville, California 94608-2411

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED **HH**

Disclaimer

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

**Portions of this document may be illegible
in electronic image products. Images are
produced from the best available original
document.**

CONTENTS

	Page
1. EXECUTIVE SUMMARY	12
1.1 Background	12
1.1.1 Phase I Program	13
1.2 Literature Review	13
1.3 Laboratory Test Apparatus	14
1.3.1 Data Acquisition System	14
1.4 Sample Materials and Surrogates	14
1.5 Laboratory Test Matrix	15
1.6 Laboratory Test Results and Analysis	16
1.6.1 Air Permeability Testing	16
1.6.2 Hydraulic Conductivity Testing	16
1.6.3 NAPL-Extraction - Purging with Water Testing	17
1.6.3.1 Fluid Flow Results	17
1.6.3.2 Strain Amplitude Results	18
1.6.3.3 Optimization Results	18
1.7 AER Technology Performance	18
1.7.1 Field Deployment Strategy	18
1.7.2 Contaminant Modeling	19
1.7.2.1 Oklahoma Sand	20
1.7.2.2 Porcelain Clay	20
1.8 Phase II Recommendations	21
2. INTRODUCTION	23
2.1 Objectives	23
2.2 Phase I Program Overview	23
2.3 The Phase I Team	24
2.4 Literature Review	24
2.4.1 Effects Of Acoustic Excitation On Fluid Flow	25
2.4.2 Non-Linear Soil Behavior	29
2.4.3 Laboratory Measurement Techniques	29
2.4.4 Acoustic Sources	30

3. LABORATORY TEST APPARATUS	35
3.1 Test Cell	35
3.2 Test Sample Instrumentation	38
3.2.1 Dynamic Pressure Transducers	38
3.2.2 Accelerometers	38
3.2.3 Thermocouples	38
3.2.4 Pressure Transducers	38
3.3 Experimental Control	39
3.3.1 Signal Generator And Power Amplifier	39
3.3.2 Pressure Sources And Regulators	39
3.3.3 Measuring Purging Agent Flow	39
3.3.4 Specialized Flow Checks	40
3.4 Data Acquisition and Archiving System	40
3.4.1 Overview	40
3.4.2 Data Acquisition System	41
3.4.3 Data Archiving	43
3.4.4 Data Analysis	44
4. SAMPLE MATERIALS AND SURROGATES	45
4.1 Sample Materials	45
4.1.1 Oklahoma Sand	45
4.1.2 Ajax Sand	45
4.1.3 Artificial Silt	45
4.1.4 Porcelain Clay	46
4.2 Laser Particle Size and Mineralogical Analyses	46
4.2.1 Laser Particle Size Analyses	46
4.2.2 Mineralogical Analyses	46
4.3 Surrogate Contaminants	47
4.4 Sample Preparation and Handling	47
4.4.1 Oklahoma And Ajax Sands	47
4.4.2 Artificial Silt	48
4.4.3 Porcelain Clay	48
5. LABORATORY TEST MATRIX	49
5.1 Summary of Laboratory Experiments	49

5.2 Test Procedures	50
5.2.1 Air Permeability Testing	50
5.2.2 Hydraulic Conductivity Testing	51
5.2.2.1 Oklahoma Sand	51
5.2.2.2 Ajax Sand	51
5.2.2.3 Artificial Silt	52
5.2.2.4 Porcelain Clay	52
5.2.3 NAPL Extraction - Purging With Water	53
 6. LABORATORY RESULTS AND ANALYSIS	 55
6.1 Air Permeability	55
6.1.1 Oklahoma Sand	55
6.1.2 Ajax Sand	56
6.1.3 Summary	56
6.2 Hydraulic Conductivity	58
6.2.1 Oklahoma Sand	58
6.2.2 Ajax Sand	58
6.2.3 Artificial Silt	59
6.2.4 Porcelain Clay	59
6.2.5 Summary	60
6.3 NAPL Extraction - Purging with Water	65
6.3.1 Background	65
6.3.2 Tests in Oklahoma Sand	68
6.3.2.1 Water - Canola Oil, Test Set No. 1	68
6.3.2.2 Water - Canola Oil, Test Set No. 2	69
6.3.2.3 Water - Soltrol 220	70
6.3.3 Tests In Ajax Sand	80
6.3.4 Summary	80
6.4 Laboratory Strain Amplitudes	82
6.5 Frequency Sweep Analyses of Non-Linear Soil Behavior	82
6.6 Optimization Testing	83
 7. ACOUSTIC REMEDIATION TECHNOLOGY PERFORMANCE	 85
7.1 Assessment of Acoustical Sources	85
7.2 Field Deployment Strategy	85
7.2.1 Single Source	89
7.2.2 Broadband Sources	89

7.2.3 Phased Arrays	92
7.2.3.1 Localized Intensification With Phased Arrays	92
7.2.3.2 Full Volume Sweeps	94
7.2.3.3 Other Array Geometries	97
7.2.3.4 Summary	98
7.3 Contaminant Modeling	99
7.3.1 Oklahoma Sand	99
7.3.1.1 Model Assumptions and Calibration	99
7.3.1.2 The Remediation Process	103
7.3.1.3 Summary	113
7.3.2 Porcelain Clay	113
7.3.2.1 Summary	113
8. RECOMMENDATIONS FOR PHASE II	115
8.1 Phase 1 Summary of Results	115
8.2 Phase II Program	115
9. REFERENCES	117

FIGURES

Figure 1. Potential Effects of Acoustic Excitation Fields on the Solid Matrix, DOE Contract No. DE-AR21-94MC30360, Phase I	26
Figure 2. Potential Effects of Acoustic Excitation Fields on the Pore Fluids, DOE Contract No. DE-AR21-94MC30360, Phase I	27
Figure 3. Comparison of Normalized Modulus Reduction Relationships for Sands (modified after Iwasaki et al., 1978; taken from Sun, Golesorkhi, and Seed, 1988), DOE Contract No. DE-AR21-94MC30360, Phase I.	32
Figure 4. Damping Ratios for Sands (taken from Seed, Wong, Idriss, and Tokimatsu, 1994), DOE Contract No. DE-AR21-94MC30360, Phase I	33
Figure 5. Normalized Modulus Reduction Relationships for Clays with Different Plasticity Indices (taken from Sun, Golesorkhi, and Seed, 1988), DOE Contract No. DE-AR21-94MC30360, Phase I	34
Figure 6. Schematic Cross-Sectional View of the Laboratory Test Cell, DOE Contract No. DE-AR21-94MC30360, Phase I	36
Figure 7. Photograph of the Laboratory Test Cell, DOE Contract No. DE-AR21-94MC30360, Phase I	37
Figure 8. Schematic of the Data Acquisition System, DOE Contract No. DE-AR21-94MC30360, Phase I	41
Figure 9. Front Panel of the Data Acquisition System Virtual Instrument, DOE Contract No. DE-AR21-94MC30360, Phase I	42
Figure 10. Section of the Programming Code Behind the Data Acquisition System Virtual Instrument, DOE Contract No. DE-AR21-94MC30360, Phase I	43
Figure 11. Main Tables of the Data Acquisition System Database, DOE Contract No. DE-AR21-94MC30360, Phase I	44
Figure 12. Air Permeability of Oklahoma Sand Excitated Results, DOE Contract No. DE-AR21-94MC30360, Phase I	57

Figure 13. Cumulative Flow Volume versus Time for the First Porcelain Clay Test, DOE Contract No. DE-AR21-94MC30360, Phase I	62
Figure 14. Cumulative Flow Volume versus Time for the Second Porcelain Clay Test, DOE Contract No. DE-AR21-94MC30360, Phase I	63
Figure 15. Hydraulic Conductivity of the Second Porcelain Clay Test, DOE Contract No. DE-AR21-94MC30360, Phase I	64
Figure 16. Idealized Water-DNAPL Relative Permeability Plot (taken from Schuille, 1988), DOE Contract No. DE-AR21-94MC30360, Phase I	66
Figure 17. Oklahoma Sand - NAPL Surrogate (Canola Oil) Pump and Treat Simulation 1, DOE Contract No. DE-AR21-94MC30360, Phase I	71
Figure 18. Oklahoma Sand - Partial Water Saturation Versus Time, DOE Contract No. DE-AR21-94MC30360, Phase I	72
Figure 19. Oklahoma Sand - Water - NAPL Surrogate Relative Permeability, DOE Contract No. DE-AR21-94MC30360, Phase I	73
Figure 20. Oklahoma Sand NAPL - Surrogate - Water Relative Permeability, DOE Contract No. DE-AR21-94MC30360, Phase I	74
Figure 21. Semi-Logarithmic Extrapolation Technique Example, DOE Contract No. DE-AR21-94MC30360, Phase I	77
Figure 22. Oklahoma Sand - NAPL Surrogate (Canola Oil) Pump and Treat Laboratory Test No. 2, Baseline Data Shown in Gray and Excited Data Shown in Black, DOE Contract No. DE-AR21-94MC30360, Phase I	78
Figure 23. Oklahoma Sand - NAPL Surrogate (Soltrol 220) Pump and Treat Laboratory, Baseline Data Shown in Gray and Excited Data Shown in Black, DOE Contract No. DE-AR21-94MC30360, Phase I	79
Figure 24. Ajax Sand - NAPL Surrogate (Canola Oil) Pump and Treat Laboratory, Baseline Data Shown in Gray and Excited Data Shown in Black, DOE Contract No. DE-AR21-94MC30360, Phase I	81
Figure 25. Plot of Time Versus Frequency for Dry Oklahoma Sand, DOE Contract No. DE-AR21-94MC30360, Phase I	83
Figure 26. Trade Space of Strain and Field Intensity Compared Against Experimental Test Regime, DOE Contract No. DE-AR21-94MC30360, Phase I	87

Figure 27. Intensity Versus Color Coding Used in Source Intensity Analyses, DOE Contract No. DE-AR21-94MC30360, Phase I	88
Figure 28. Intensity Versus Location for a Single Source, DOE Contract No. DE-AR21-94MC30360, Phase I	89
Figure 29. Intensity Versus Location Produced by Two Sources that Incoherently Sum, DOE Contract No. DE-AR21-94MC30360, Phase I	90
Figure 30. Intensity Versus Location Produced by Four Sources that Incoherently Sum, DOE Contract No. DE-AR21-94MC30360, Phase I	91
Figure 31. Intensity Versus Location Produced by Eight Sources that Incoherently Sum, DOE Contract No. DE-AR21-94MC30360, Phase I	91
Figure 32. Intensity Versus Location Produced by Two Sources that Coherently Sum for Maximum at Center of Array, DOE Contract No. DE-AR21-94MC30360, Phase I	93
Figure 33. Intensity Versus Location Produced by Four Sources that Coherently Sum for Maximum at Center of Array, DOE Contract No. DE-AR21-94MC30360, Phase I	93
Figure 34. Intensity Versus Location Produced by Eight Sources that Coherently Sum for Maximum at Center of Array, DOE Contract No. DE-AR21-94MC30360, Phase I	94
Figure 35. Maximum Intensity Versus Location Produced by Sweeping Two Sources Across the Volume, DOE Contract No. DE-AR21- 94MC30360, Phase I	95
Figure 36. Maximum Intensity Versus Location Produced by Sweeping Four Sources Across the Volume, DOE Contract No. DE-AR21- 94MC30360, Phase I	95
Figure 37. Maximum Intensity Versus Location Produced by Sweeping Eight Sources Across the Volume, DOE Contract No. DE-AR21- 94MC30360, Phase I	96
Figure 38. Maximum Intensity Versus Location from and Array of 32 Phase Controlled Sources of 1 kW Intensity Each, DOE Contract No. DE-AR21-94MC30360, Phase I	96

Figure 39. Maximum Intensity Versus Location in an Eight Element Circular Array Plus on Additional Element in the Center, DOE Contract No. DE-AR21-94MC30360, Phase I	97
Figure 40. Maximum Intensity Versus Location by a Linearly Spaced Phased Array, DOE Contract No. DE-AR21-94MC30360, Phase I	98
Figure 41. Laboratory Test Cell Model Domain, DOE Contract No. DE-AR21-94MC30360 Phase I	101
Figure 42. Oklahoma Sand - NAPL Surrogate (Canola Oil) - Water Model of Laboratory Results, DOE Contract No. DE-AR21-94MC30360, Phase I	102
Figure 43. Stimulated Cumulative NAPL Surrogate (Canola Oil) Recovery, DOE Contract No. DE-AR21-94MC30360, Phase I	104
Figure 44. Oklahoma Sand - NAPL Surrogate (Soltrol 220) - Water Model of Laboratory Results, DOE Contract No. DE-AR21-94MC30360, Phase I	105
Figure 45. Stimulated Cumulative NAPL Surrogate (Soltrol 220) Recovery, DOE Contract No. DE-AR21-94MC30360, Phase I	106
Figure 46. Simulated NAPL - Ground Water Domain in Modeling the Effects of Acoustically Enhanced Remediation, DOE Contract No. DE-AR21-94MC30360, Phase I	108
Figure 47. NUFT Modeling Results of Oklahoma Sand - Water - NAPL Surrogate (Canola Oil) System, DOE Contract No. DE-AR21-94MC30360, Phase I	109
Figure 48. NUFT Modeling Results of Oklahoma Sand - Water - NAPL Surrogate (Soltrol 220), DOE Contract No. DE-AR21-94MC30360, Phase I	110
Figure 49. NUFT Modeling Results of Oklahoma Sand - Water - NAPL Surrogate (Canola Oil) System, (Generation 2 - field conditions), DOE Contract No. DE-AR21-94MC30360, Phase I	111
Figure 50. NUFT Modeling Results of Oklahoma Sand - Water - NAPL Surrogate (Soltrol 220) System, (Generation 2 - field conditions), DOE Contract No. DE-AR21-94MC30360, Phase I	112

TABLES

Table 1. Test Series Summary, DOE Contract No. DE-AR21-94MC30360, Phase I	49
Table 2. NAPL Saturations by Various Methods for the Various Tests Conducted, DOE Contract No. DE-AR21-94MC30360, Phase I.	76
Table 3. Available and Research Acoustic Source Technologies, DOE Contract No. DE-AR21-94MC30360, Phase I	86

APPENDICES

Appendix A.	Analytical Reports	Volume II
Appendix B.	Chronological Log of Experiments	Volume II
Appendix C.	Laboratory Data Analysis and QA/QC	Volume II

1. EXECUTIVE SUMMARY

The Phase I laboratory bench-scale investigation results have shown that acoustically enhanced remediation (AER) technology can significantly accelerate the ground water remediation of non-aqueous phase liquids (NAPLs) in unconsolidated soils. The testing also determined some of the acoustic parameters which maximize fluid and contaminant extraction rates. A technology merit and trade analysis identified the conditions under which AER could be successfully deployed in the field, and an analysis of existing acoustical sources and varying methods for their deployment found that AER technology can be successfully deployed in-situ. Current estimates of deployability indicate that a NAPL plume 150 ft in diameter can be readily remediated.

The development of an AER technology addresses the U.S. Department of Energy's (DOE's) need for innovative and improved in-situ remediation methods to (1) expedite clean up of contaminants, (2) expedite clean up of contaminated low permeability soils, (3) reduce clean up time and cost, and (4) diminish human health and ecological risk at contaminated sites.

This program focused on unconsolidated soils because of the large number of remediation sites located in this type of hydrogeologic setting throughout the nation. It also focused on NAPLs and low permeability soil because of the inherent difficulty in the remediation of NAPLs and the significant time and cost impact caused by contaminated low permeability soils.

The objective of the AER program is to develop a commercial field viable technology. Phase I, the proof-of-principle testing, was the first step. Development of a successful AER technology will dramatically advance the science and art of extracting contaminants from the subsurface.

This overall program is recommended for Phase II which will address the technology scaling requirements for a field scale test. Presented below is a summary of each section of this report.

1.1 Background

Weiss Associates has been contracted to the DOE Morgantown Energy Technology Center (METC), Contract No. DE-AR21-94MC30360, to perform the following integrated three phase program that will investigate the use of acoustic excitation fields¹ (AEFs) to enhance the remediation of low permeability unconsolidated soil and ground water under both saturated and unsaturated conditions.

¹ Acoustic excitation fields are defined as a region in space subjected to acoustic excitation from an acoustical source.

- Phase I - Laboratory Scale Parametric Investigation
- Phase II - Technology Scaling Investigation
- Phase III - Large Scale Field Tests

Phase I is a laboratory proof of concept investigation designed to characterize the effects of AEFs on fluid and contaminant extraction rates and to investigate the viability of deploying AER technology in the field. Phase II is a technology scaling study designed to bridge the gap between the Phase I results conducted on samples which are on the order of inches and the data required for field scale testing which is on the order of hundreds of feet. Phase III will consist of field scale testing at a non-industrialized, non-contaminated site to test and debug the technology, and at an industrialized and contaminated site to validate the technology. This report summarizes the work conducted during Phase I.

1.1.1 Phase I Program

A series of one-dimensional (1-D) laboratory tests were conducted to examine the influence of AEFs on the fluid (water and/or air) and NAPL contaminant extraction rates in unconsolidated soil samples approximately 1.5" diameter and 3.5" long. These data were then used in a computer model to predict the in-situ effects of AER. Additionally, a Technology Merit and Trade Study was conducted to determine the engineering viability of deploying the technology in the field. Phase I was to examine both unsaturated and saturated zone conditions but time and budget constraints resulted in a focus on saturated conditions only.

1.2 Literature Review

A literature review was conducted to (1) identify prior work related to this study, (2) develop an awareness of ongoing research and development efforts, and (3) collect acoustic properties information for use in engineering analyses and modeling. To date, some laboratory testing of the use AEFs primarily in the ultrasonic range (above the range of human hearing, about 20 kilohertz [kHz] or greater), has been conducted for both enhanced oil production and environmental remediation. These results are in general, compared and contrasted to the results obtained in this Phase I program. The literature review did not identify any readily obtainable field scale testing results of the technology. However, it did provide some insight into the possible mechanisms controlling AER, measurement techniques, acoustical sources, and the mathematical modeling of their emissions. Figures 1 and 2 summarize our hypothesized potential effects of AEFs on the solid matrix and pore fluid, respectively. These acoustically generated mechanisms all act towards rendering the remediation process faster and more cost-effective. Quantification of these individuals mechanisms, however, was beyond the scope of this investigation.

1.3 Laboratory Test Apparatus

Figure 6 shows a cross-section of the laboratory test apparatus used to evaluate the effects of AEFs on contaminated soil and ground water. An electro-mechanical shaker, Ling Dynamic Systems Model 411, 44 pound peak force, DC to 9 kHz, was used to excite vibrations in a test soil sample. The sample was placed in a thin, heat-shrink mylar jacket between two end caps that are fitted with 120 mesh screens for fluid flow measurements. The shaker and sample assembly are enclosed in a sealed chamber that is pressurized to simulate the stress on the soil at different depths in the subsurface. The wall of the chamber is a Plexiglas cylinder to allow continuous monitoring of the condition of the sample and the flow of fluids in the inlet and outlet lines.

The test apparatus has been designed to perform a 1-D simulation of the soil vapor extraction or NAPL ground water pump and treat (P&T) remediation processes. It has been used in this Phase I program to develop a basic understanding of acoustically enhanced NAPL ground water P&T.

1.3.1 Data Acquisition System

A data acquisition system (DAS) was developed to acquire data from the various sensors and instruments in the test apparatus. It is mainly comprised mainly of data acquisition hardware, sensors, and a graphical software interface that controls the instrumentation and enables one to easily view, acquire, and save the acquired data. The DAS hardware consists of a digitizing board which captures the majority of the data, a general purpose interface bus (GPIB) controller card that enables communication with GPIB (IEEE-488) instruments, and the sensors themselves. The software interface was written using LabVIEW, a commercial programming environment specifically designed for laboratory data acquisition and instrument control.

1.4 Sample Materials and Surrogates

Four commercially-available soils were used in this testing program (1) a well-sorted, fine homogeneous sand, referred to as Oklahoma sand, (2) a poorly sorted heterogeneous sand with 9% silt and clay, referred to as Ajax sand, (3) an artificial silt, and (4) a porcelain clay. Two NAPL-surrogates were used during different portions of the NAPL-extraction tests with the majority of testing being performed using a vegetable oil (Crisco canola oil) dyed red with a small amount of Sudan IV dye. The density of the canola oil is 0.9182 g/mL at 70°F, making it less dense than water and by definition a lighter than water NAPL or LNAPL. However, its viscosity, 65 cPoise at 70°F, is significantly higher than for many fuel hydrocarbons and chlorinated hydrocarbons. The other NAPL surrogate used was Soltrol 220, also dyed with Sudan IV dye. Soltrol 220 was used to address questions raised on whether canola oil may behave differently than most LNAPLs because of its greater viscosity or possibly different surface chemistry. Soltrol 220 has a viscosity of 4.83 cPoise at 73°F, and is also a LNAPL.

Use of NAPL-surrogates for laboratory investigations is a well-established practice. Canola oil and Soltrol 220 were chosen as because of their relatively non-hazardous and non-toxic properties. While both of these surrogates are LNAPLs, the Soltrol 220 will provide a first approximation for remediation of fluids that are denser than water or DNAPLs. This is founded on Wilson et al. (1990) where Soltrol 130 was the most commonly used fluid in their laboratory investigation of residual liquid organics. Based on this Cohen and Mercer (1993, p. 4-19) in their work on evaluating DNAPLs recommended Soltrol 130 as an ideal fluid for determining residual saturation for a porous media. The densities and viscosities of Soltrol 130 and 220 are comparable for the purposes of this investigation.

1.5 Laboratory Test Matrix

A summary of the laboratory test matrix performed is presented in Table 1. In these tests, we endeavored to determine the optimal acoustic parameters to maximize the fluid and contaminant extraction rates for a wide range of soil types under generally saturated conditions.

Each test included two set of conditions (1) baseline without acoustic excitation, and (2) excited with acoustic excitation either at a single frequency or at a sequence of frequencies between 100 Hz and 10 kHz. Measurements were generally made at three different power levels under the excited condition. The NAPL-extraction tests simulated a ground water-NAPL P&T operation, and the laboratory testing approach was based on Stevens (1996).

Tests were generally run using five-minute duty cycle (i.e. five minutes on at a particular frequency and five minutes off), to provide information on soil hysteresis effects and how long the effects of acoustic excitation persist in a particular soil. In general, the air permeability and hydraulic conductivity tests for the two types of sand were conducted at two confining pressures (30 and 10 psi) to simulate two stress conditions in the shallow subsurface, the NAPL-extraction tests were conducted at a nominal 30 psi confining pressure, and the hydraulic conductivity analyses of porcelain clay were conducted at 23 psi.

Nearly identical laboratory test soil samples of the Oklahoma sand and porcelain clay were produced, but there was greater variability between the laboratory test soil samples of the Ajax sand and artificial silt. Since there inevitably will be some variability among the same soil-type samples, the test program focused on the relative changes in fluid flow rates in the same sample under baseline and excited conditions for the air permeability and hydraulic conductivity analyses. For the NAPL-extraction tests, it was necessary to use multiple soil samples and each test was repeated up to six times to verify that the results are statistically significant and not due to sample variability. The soil samples of Oklahoma sand were sufficiently uniform that successful comparisons between samples was possible, but this proved difficult with the Ajax sand samples due to its intrinsic variability.

1.6 Laboratory Test Results and Analysis

1.6.1 Air Permeability Testing

Oklahoma and Ajax sand air permeability testing indicated that acoustic excitation has no significant effect on the air permeability of these highly-permeable sands under the range of conditions tested. Two likely explanations for these observed results are (1) the air is very compressible so there can be no buildup of anomalous pore pressures, and (2) the flow of air is essentially unaffected by microscopic deformation of the pore throats and channels.

These tests were conducted to determine if acoustic excitation could affect air permeability and thus vadose zone remediation processes. The null results pertain only to the air permeability of the grain framework. Further testing is required in partially saturated samples containing a volatile or semi-volatile fluid, to determine if AEFs can increase the contaminant extraction rate.

1.6.2 Hydraulic Conductivity Testing

Hydraulic conductivity (K_h) testing in the Oklahoma and Ajax sand samples essentially provided null results. However, it is widely recognized in laboratory-scale soil dynamics testing (see Section 2.2.3) that the observed soil behavior depends strongly on whether any induced excess pore-water pressure is allowed to dissipate. Thus, it appears that the most likely explanation for these observed null results is that since the Phase I soil samples were always in a drained condition (see Section 6.2.5), no buildup of excess pore-water pressure could occur. In the field, the drainage conditions will be closer to undrained case and this could have a major positive effect on the soil-fluid transport properties under acoustic excitation. Further testing is required to verify this hypothesis.

The K_h testing of Ajax sand decreased during the course of the tests apparently due to fines mobilization and progressive plugging of the pore channels. However, the mobilization and production of fines will be useful for remediation since contaminants are generally sorbed on the fine-grained fraction of the aquifer (see Section 6.2.2). Additional testing is required to optimize the production of fines while minimizing the tendency to plug pores.

The K_h testing of the porcelain clay samples provided the most favorable results in this Phase I program. With acoustic excitation, a two- to four-fold increase in K_h was induced along with a possible permanent change in the intrinsic permeability of the clay, post-excitation (i.e., the K_h of the clay may have increased after being subjected to acoustic excitation).

The very low intrinsic permeability of the clay and the aforementioned acoustic excitation result provide insight into the possible operative mechanisms for how acoustic excitation may effect the soil matrix. It is suspected that the increase in K_h is probably related to (1) the generation of excess pore-water pressure during dynamic loading (see Figure 1), (2) particle

agitation of the soil matrix (see Figure 1), and (3) some combination of these two mechanisms. Perhaps dynamic excitation of the double layer may also have a still unknown effect.

The potential for a permanent change in the K_h of low permeability soil as exemplified by the porcelain clay (see Figure 15) suggests that there may be a long-term increase in the K_h of aquatards after acoustic excitation. This result further suggests that it may be possible to implement AER technology using less-expensive, intermittent acoustic excitation.

1.6.3 NAPL-Extraction - Purging with Water Testing

1.6.3.1 Fluid Flow Results

A summary of the NAPL saturations measured or calculated for the various tests conducted under this Phase I program is presented in Table 2. Three types of NAPL fractional pore volume (or saturation) data are reported (1) the empirical laboratory P&T test results based on measured produced fluid volumes, (2) the semi-logarithmic extrapolation to a common pore volume throughput (100 pore volumes) using the Jones and Roszelle (1978) method, and (3) independent analyses of the residual NAPL fractional pore volume by the Dean Stark solvent extraction method (American Petroleum Institute Recommended Practices 40 by Smith et al., 1960; Cohen and Mercer, 1993) which measures the actual fluid volume remaining in the pores.

Acoustic excitation enhanced the recovery of both canola oil and Soltrol 220 from samples of Oklahoma sand. The canola oil excited condition results showed relative NAPL-extraction improvements above baseline conditions ranging from 6% to 26% for the empirical laboratory data, and 13% to 41% for the Jones and Roszelle (1978) extrapolation. The Soltrol 220 empirical laboratory baseline results show that the NAPL recovery was not as dramatic as with the canola oil, reflecting that a standard P&T is more effective when the NAPL/water viscosity ratio is closer to unity. However, the Jones and Roszelle (1978) extrapolation of the Soltrol 220 data indicate that its recovery was actually higher than that of canola oil.

The apparent disagreement between the Dean-Stark analyses and the empirical laboratory and the semi-logarithmic extrapolation data for the canola oil is paradoxical. Its resolution is beyond the scope of this investigation, however.

Acoustic excitation did not appear to enhance the recovery of canola oil recovery from samples of Ajax sand until these laboratory soil samples were subjected to sequential frequency excitation and an increased power level. Acoustic excitation of the Ajax sand at a single frequency only showed about 2% and 4% relative NAPL-extraction improvement over baseline conditions for the laboratory empirical and Jones and Roszelle (1978) extrapolation results, respectively. However, excitation at a sequence of six frequencies, and at an increased power level and at a sequence of eight frequencies with increased power showed a 15% and 28% relative NAPL-extraction improvement over baseline conditions, respectively, for the laboratory empirical results, and 39% and 74% relative NAPL-extraction improvement over baseline conditions, respectively, for the Jones and Roszelle (1978) method. Due time constraints in the test program, only one

laboratory test of the Ajax sand (31.05.95, see Section 6.3.3) was conducted under the latter test conditions, but the trend towards higher NAPL recovery appears clear.

The increased NAPL recovery in Ajax sand with high-amplitude, multiple-frequency sequential excitation may offer a key to successful field application of this technology. To a considerable extent, the null results with Ajax sand empirical laboratory data at a single frequency (only 2% observed and 4% extrapolated improvement) may just reflect the difficulty in obtaining similar samples of the poorly sorted material.

1.6.3.2 Strain Amplitude Results

An obvious question in evaluating AER viability is whether the acoustical levels required to develop the beneficial effects described above can be generated in-situ with available acoustic source technologies. To address this issue, the strain amplitudes² developed in the laboratory experiments were estimated and compared with strain amplitudes obtainable from available source technologies. These analyses show that the beneficial effects of acoustic excitation are observed in experiments where the strain amplitudes are between approximately 10^{-5} and 10^{-4} . As discussed in Section 7.2, these strain amplitudes are within the range that can be developed in-situ with existing source technology, especially if the sources are deployed using a phased array. During field tests, the induced strain amplitudes will be monitored using an array of geophones positioned in shallow boreholes.

1.6.3.3 Optimization Results

The optimization testing portion of the laboratory program primarily pursued two encouraging approaches (1) increasing the acoustic power input to the sample, and (2) subjecting the sample to a sequence of frequencies that were chosen because they showed strong resonances or interesting nonlinear soil behavior. Both of these approaches shows positive results and provided insight into potential keys for AER technology field deployment.

1.7 AER Technology Performance

1.7.1 Field Deployment Strategy

While a range of technologies exist for radiating large amounts of acoustical power into the subsurface (see Section 7.1), the complex geometry and geohydrology of contaminant plumes make large volume acoustical excitation desirable. However, instead of deploying a very large number of sources in many wells to treat the volume of a contaminated plume, it is preferable to use a distributed array of sources which:

² The strain amplitude is a dimensionless quantity representing the fractional change in dimension in response to an applied stress.

1. by controlling the phase of the sources, high-power acoustical energy can be directed to various parts of the plume; and
2. by modifying the phase of the sources, high intensity acoustic energy can be focused and swept throughout the contaminant volume.

The key engineering question is what levels of acoustic intensity and strain amplitude can be generated in-situ using realistic sources. Figure 26 shows an engineering trade space of acoustical frequency and intensity in comparison with the test regime of our laboratory experiments. The Phase I laboratory experiments indicate that the beneficial effects of acoustic excitation are observed when strain amplitudes are between approximately 10^{-5} and 10^{-4} .

For acoustic field deployment, the use of a single source, a broadband source, and phased arrays were evaluated. For a contaminant plume of size, a single source AER system would probably not be effective. Broadband sources can cover much larger volumes than a single source, but they will most likely be too expensive to generate the required acoustic intensities. Phased arrays, on the other hand, hold much promise for cost-effective field deployment of AER technology.

The basic concept of a phased array is that with multiple sources with identical frequencies and controlling the phases of the individual sources, constructive interference can be achieved at a given location to give a region of high acoustic intensity. The phase control of the sinusoidal waveforms from multiple sources is what constitutes the "phased array". Both circular and linear phased arrays geometries were considered for the case of acoustical sources at the ground-air interface. Extending the distribution of these sources to positions within wellbores will further enhance the ability of phase control to produce intensities beyond those shown herein.

The main effect of a phased array is that the intensity of the acoustical field is now proportional to (1) the power of the individual sources, (2) the square of the number of sources, and (3) inversely proportional to the spatial dimension of the array. By varying the relative phases of the different sources, the region of maximum intensification can be swept across the treatment volume. For example, with an eight element array and 10 kW sources, acoustic intensities over 1000 W/m^2 (strains about 10^{-5}) can readily be generated.

The challenge for cost-effective deployment of AER technology appears to be a favorable source configuration, and this will depend on the shape of the contaminant plume and the geohydrological setting. Engineering considerations in deploying a phased array in an heterogeneous geohydrological environment will be addressed in Phase II and Phase III of this program.

1.7.2 Contaminant Modeling

Both the Oklahoma sand and porcelain clay empirical laboratory test data were used to quantify the contaminant flow and transport benefits of AER technology by using a numerical

model for the Oklahoma sand and an analytical expression for the porcelain clay. Both approaches are described below.

1.7.2.1 Oklahoma Sand

A public domain computer model, NUFT (Nonisothermal Unsaturated-Saturated Flow and Transport), was used to simulate P&T remediation of a NAPL spill in ground water using the baseline and excited NAPL recovery rates measured in Oklahoma sand. Two generations of modeling were performed. The first generation of modeling attempted to duplicate the Phase I laboratory fluid flow conditions for field dimensions. These results (1) are unrealistic but representative of an extreme case for both baseline and excited conditions, and (2) were used as a bridge between laboratory and more realistic field flow conditions, the second generation of modeling. All models were calibrated using the laboratory data, and then the calibrated model was used to simulate NAPL recovery by a single pumping well. A 2-dimensional, cylindrical model domain was used to simulate NAPL recovery under field conditions (Figure 46). A single remediation well along the cylinder axis was screened over the entire thickness of the aquifer.

For the first generation of modeling and using the empirical laboratory data results (i.e., the most conservative data) for the canola oil - ground water - NAPL system, remediation was simulated for a maximum duration of 3 years (1080 days). The model results (Figure 47) show that after 1000 days of P&T, acoustic excitation increases the NAPL recovery from 40% to 58%, for an applied head difference of 52.49 ft (16 m). For the Soltrol 220 - ground water-NAPL remediation under excited conditions yields an increase in NAPL recovery from 75% to 90% after 100 days of P&T under a comparable applied head difference (Figure 48). Depending on the viscosity of the NAPL and the time frame of the simulation, the model proved to be sensitive to the hydraulic gradient created by the remediation well. For canola oil, lowering the applied drawdown to 5 m reduced the NAPL recovery by about 5% after 1000 days (Figure 47). For Soltrol 220, the drawdown dependent decrease in NAPL recovery was about 3% for the baseline and 1% for the excited conditions after 100 days of P&T (Figure 48). However, because the maximum possible recovery rate for a NAPL is determined by its residual saturation, it can be expected that with increasing P&T duration, recovery rates will be more independent of the applied drawdown.

The second generation of modeling used more realistic field dimensions and a total head difference of only 2 m (6.6 ft), Figure 46. The cumulative NAPL recovery for canola oil and Soltrol 220 for these more realistic field conditions are shown in Figure 49 and Figure 50, respectively. The total percentage of NAPL recovered is almost identical to that in the first generation, but the time frame for the same recovery increases. For canola oil, time to maximum recovery increased by a factor of 3, while for the less viscous Soltrol 220, the effect is much less pronounced.

1.7.2.2 Porcelain Clay

A zero-dimensional analytical model of aquifer cleanup was applied to the hydraulic conductivity results obtained for porcelain clay. This model assumes that the aquifer can be represented by a single, well-mixed domain of fixed volume, V , initially at a uniform ground water contaminant concentration, $c = c_0$, with recharge water having a contaminant concentration of

zero entering at a fixed flow rate Q . The model simulates P&T remediation where the contaminant is extracted from the domain at the prevailing concentration, c , at a pumping rate equal to Q . By inspection of the equations it was found that if the flow rate is doubled, then the remediation time is decreased by a factor of two.

Thus, the K_h results for clay (Section 6.2.4) are significant because if the K_h of fine-grained materials, represented by clay, can be increased by a factor of two with excitation, then the time to reach a remediation goal in this type of soil will be cut in half. Similarly, if a four-fold increase in K_h can be achieved, as evidenced in Figure 15, then the remediation time required to reach cleanup can be decreased to a quarter of what it would be without excitation.

In many remediation scenarios, the long time required to reach cleanup is controlled by the time required for diffusion of contaminants from fine-grained, low permeability sediments into coarse-grained high permeability sediments controls. If AER can significantly increase the K_h of low permeability materials, then there will be a significant reduction in environmental remediation costs. Note these favorable results pertain only to the hydraulic behavior of the system, i.e., increasing water flux through the system. Acoustic excitation should also have an unknown favorable effect on contaminant mobility that could further reduce the remediation time. This later hypothesis requires further testing.

1.8 Phase II Recommendations

Phase II is a technology scaling study designed to bridge the gap from the generally 1-D work conducted in Phase I to the 3-D understanding and engineering necessary for a field scale test of AER technology. To have a field deployable system for Phase III, the Phase II portion of this AER program must determine:

1. how an AEF interacts with a geohydrochemical environment specifically in regards to heterogeneities at all scales; and
2. how to engineer the deployment of phased array system.

To achieve these objectives we recommend that Phase II focus on identifying the characteristics of a field deployable AER technology system based on 2-D laboratory extraction testing at a scale of about 5 cubic feet, under AEFs generated by the phased arrays, and 2-D and 3-D modeling of (1) the fluid flow and transport behavior under baseline and excited conditions, and (2) the acoustical responses generated in the soil. This Phase II activity will use an actual DNAPL during testing because this project has been placed in DOE's DNAPL program. The outlined Phase II approach will allow for:

1. evaluating the coupling, propagation, and control of AEFs in the subsurface;
2. determining optimum well field and/or surface array configurations;
3. evaluating technology performance, safety, and cost; and

4. validation at the scale of testing computer models to predict the coupling, propagation, and control of acoustic energy, and the resultant physical and chemical effects in the subsurface.

In addition to the aforementioned program, a test site will be selected for the Phase III large field test.

2. INTRODUCTION

2.1 Objectives

Weiss Associates has been contracted by the United States Department of Energy (DOE) Morgantown Energy Technology Center (METC) under Contract No. DE-AR21-94MC30360 to perform an integrated three phase program that will investigate the systematics of using AEFs to enhance the remediation of low permeability unconsolidated soils and ground water under both saturated and unsaturated conditions. The three phases of the program are:

- Phase I - Laboratory Scale Parametric Investigation
- Phase II - Technology Scaling Study
- Phase III - Large Scale Field Tests

This research and development effort addresses the need for innovative and improved in-situ remediation methods to (1) expedite the clean-up of contaminated, low-permeability soil and ground water, (2) reduce the cost of cleanup, and (3) diminish the human health and ecological risks associated with contaminated sites.

Phase I is a laboratory proof of concept investigation designed to characterize the effects of AEFs on fluid and contaminant extraction rates from unconsolidated soils and to investigate the viability of deploying AER technology in the field. Phase II is a technology scaling study designed to bridge the Phase I laboratory testing conducted on samples which are on the order of inches and the data required for field scale testing which is on the order of hundreds of feet. Phase III will consist of field scale testing at a non-industrialized, non-contaminated site to test and debug the technology, and field test at an industrialized and contaminated site to validate the technology. This report summarizes the work conducted during Phase I.

2.2 Phase I Program Overview

There have been many observations to indicate the potential of acoustically enhanced soil and ground water remediation. However, many of the previous data consist of single point measurements. The laboratory scale parametric investigation was designed as a series of one-dimensional (D) laboratory experiments to:

1. verify that AEF technology can enhance the in-situ remediation of contaminated soil and ground water; and

2. evaluate the relationships between the fluid and contaminant extraction rates, the frequency and power density of the AEF, and induced physio-chemical changes for a variety of soil types at different pressures and saturation conditions.

During Phase I, a series of 1-D tests examined the influence of AEFs on the fluid and contaminant extraction rates from laboratory-size, unconsolidated soil samples approximately 1.5" diameter and 3.5" long. These data were then used in a computer model to predict the effects of the acoustically enhanced soil remediation (AER) process.

The Phase I program was focused on NAPL remediation in unconsolidated low permeability sediments under both saturated and unsaturated conditions. During the Phase I testing, only the water and NAPL effluent were measured. Not investigated were the effect of AEFs on the dissolved component, but this is a recommended Phase II activity. Given the inherent difficulties in testing low permeability soil, the Phase I test program evaluated changes induced by AEFs on (1) NAPL-recovery in permeable soil, and (2) hydraulic conductivity in low permeability soil.

Note that Weiss Associates submitted to METC in May 1995, a "No-Cost Contract Extension" request, because the Phase I program was behind schedule. To place in the program on schedule and within budget, elimination of the unsaturated zone testing (e.g., soil vapor extraction tests) was recommended along with more focus on the NAPL-ground water extraction and optimization testing because very encouraging results were being obtained.

2.3 The Phase I Team

The Phase I work has been conducted by a team comprised of personnel from Weiss Associates and Scientific Applications & Research Associates, (SARA) Inc. with contributions from Dr. J. K. Mitchell of the Department of Civil Engineering at Virginia Polytechnic Institute, and Ms. Dorothy Bishop of Lawrence Livermore National Laboratory.

2.4 Literature Review

A literature review was conducted to (1) identify prior work related to this study, (2) develop an awareness of ongoing research and development efforts, and (3) collect soil acoustic properties information for use in modeling. The relevant information and contacts identified during this search were incorporated in the design and conduct of the laboratory tests.

During the initial literature search conducted during the project proposal effort several pertinent papers were located. During the current review, we obtained references cited in several of these papers, located copies of papers from the former Soviet Union that were cited by Beresnev and Johnson (1994), and arranged to have the essential parts of these papers translated.

A comprehensive electronic search of major databases was conducted. Keyword searches were performed on several major data bases including the National Technology Information

Service, Searchable Physics Information Notes, the U.S. Army Waterways Experiment Station database, Earth Science Data Center, Compendix, Alternative Treatment Technology Information Center, Cleanup Information Bulletin Board System, Gopher Internet searches and the University of California text and periodical catalogs.

Abstracts for over 200 relevant papers were identified during this process. After review of the abstracts, relevant papers were acquired and a brief summary of each paper was entered into a tabular database. During the course of this search we identified papers that contain useful information on the effects of acoustic excitation, non-linear soil behavior, potential acoustic sources and analytic modeling. In particular, the U.S. Army Waterways Experiment Station library has provided useful data on soil characteristics. We also identified researchers at the University of Houston and Kansas State University who have been working on ultrasonic (above the range of human hearing, approximately 20 kilohertz (kHz) or greater) enhancement of contaminant extraction. The acoustic literature review also found several references on sources and mathematical modeling of acoustical source emissions.

To date, some laboratory testing of the use AEFs primarily in the ultrasonic range, has been conducted for both enhanced oil production and environmental remediation. These results are in general, compared and contrasted to the results obtained in this Phase I program. The literature review did not identify any readily obtainable field scale testing results of the technology. The literature review provided several hypothesized potential effects of AEFs on the solid matrix and pore fluid (Figure 1 and Figure 2, respectively). These acoustically generated mechanisms all act towards rendering the remediation process faster and expectedly more cost-effective. Where applicable, the results obtained in Phase I are referenced to these effects. However, quantification of these effects was beyond the scope of this investigation. A summary of the key papers is presented in the following sections.

2.4.1 Effects Of Acoustic Excitation On Fluid Flow

Duhon (1964) performed a pioneering laboratory study on the effect of acoustic excitation on the water flood recovery of oil. Sandstone cores were saturated with diesel oil, core test fluid or SAE 10 oil, and then flushed the cores with water at a constant injection pressure. The procedure was then repeated while the core was subjected to an ultrasonic excitation at frequencies of 1, 3.1 and 5.5 MegaHertz (Mhz). The tests were stopped shortly after water breakthrough when the cumulative produced fluid was near 65 percent (%) of the pore volume. The oil recovery was increased by 6.4 to 14.7% during these acoustic excitation floods, with the maximum effect observed with the lowest frequency and the lower viscosity oil. No significant temperature increase was observed in the effluent fluid, and initial oil saturation did not appear to effect the overall oil recovery.

Figure 1. Potential Effects of Acoustic Excitation Fields on the Solid Matrix, DOE Contract No. DE-AR21-94MC30360, Phase I

Figure 2. Potential Effects of Acoustic Excitation Fields on the Pore Fluids, DOE Contract No. DE-AR21-94MC30360, Phase I

Cleveland and Garg (1993) studied the effect of ultrasonic excitation on the removal of fines from soil. Contaminants preferentially adsorb to fine particles, and extraction of fines could significantly enhance subsurface contaminant extraction. Dr. Cleveland reported (oral commun., 1994) that the high frequencies, 20 kHz and the ultrasonic transducer used in his study were not very effective, and that the power required to treat in-situ soil would probably be excessive.

Beresnev and Johnson (1994) performed a detailed review of numerous studies regarding the effect of acoustic waves on fluid recovery in rock over a wide range of frequencies ranging from earthquakes waves below 1 Hz to laboratory tests at frequencies above 1 Mhz. The authors concluded that significant evidence exists to suggest that acoustic excitation can influence oil recovery, but that additional work is required to determine the causes and optimal application of the effect. This study also discusses numerous observations of changes in recovery in existing wells following seismic excitation from a variety of sources such as earthquakes and overland traffic (e.g., trains). The paper includes several tables summarizing the type and results of the various studies reviewed, and includes many studies performed in the former Soviet Union. As part of the current investigation approximately twenty potentially important papers were identified, located, and translated into English.

Reddi et al. (1993, 1994) used a 20 kHz, 500 Watt ultrasonic horn to acoustically excite mixtures of sand and clay at various densities in cylindrical tanks where the sediment is at atmospheric pressure. An increase in hydraulic conductivity of up to several orders of magnitude was observed in all tests with a maximum increase noted after 8 to 10 minutes of excitation. However, they recognized that during the hydraulic conductivity measurements, there may have been leakages at the soil-wall and soil-vibrator (ultrasonic horn) interfaces. Grain size distribution analyses of the soil indicate that particles smaller than 0.04 mm were mobilized by the ultrasonic field, and that particles in the 0.04 mm to 1 mm range were fractured.

Reddi and Challa (1994) studied the effect of AEFs on the production of NAPL ganglia. Sand samples containing trapped NAPL ganglia were shaken on a 60 Hz shaker table at displacement amplitudes of 0.25 to 0.5 mm for 15 to 20 minutes. Between 15 and 88% of the trapped NAPL was released during vibration, and pumping water through the sample while shaking appeared to increase NAPL recovery. Theoretical calculations were used to calculate the size of the trapped NAPL ganglia, and to evaluate the effect of sample density and the initial conditions.

Chemical Engineering (May 1995) reports that De Ruiter Milieutechnologie B. V. (Halfweg, Netherlands) and the Technical University of Delft, Netherlands have developed an AER technology that reportedly mobilizes hydrocarbons entrapped in a soil by breaking up oil droplets that are held in the pores by capillary forces. This report indicates that because the natural frequencies of oil drops are size dependent, the transmitter's frequency is varied over an undisclosed sweep rate. Reported results indicate that 71% of diesel fuel was removed from an unspecified soil in five days with acoustic excitation, compared to 40% in 11 days without excitation. It appears from this brief report that the results are from laboratory tests since Chemical Engineering indicates that De Ruiter plans to conduct field trials at the end of 1996.

2.4.2 Non-Linear Soil Behavior

The earthquake engineering community has a large body of research on the non-linear behavior of soils and soil dynamic properties (e.g., Seed et al., 1984; and Sun et al., 1988). It is now generally recognized that at strain amplitudes below approximately 10^{-6} , the soil's response to vibrations is linear and anelastic with relatively low values of damping. Figure 3 and Figure 4 illustrate that, in a wide variety of sands, the soil response starts to be non-linear at strains near 10^{-5} and with increasing strain amplitudes there is a progressive reduction in the shear rigidity (or modulus) and increase in the damping. The same non-linear behavior has been observed in a wide variety rocks (Gordon and Davis, 1968; Winkler et al., 1979).

Sun et al. (1988) investigated the dynamic moduli and damping ratios for cohesive, clay soils and found that with increasing plasticity, as measured by the plasticity index (P.I.), the onset of the non-linear behavior shifts from the onset strain levels in sands to higher strain amplitudes (Figure 5). The porcelain clay used in this study has a moderately low P. I of 18 and would be classified in Figure 5 as a Type C2 clay soil.

The liquefaction of sands during earthquakes is caused by the build-up of excess pore fluid pressure in saturated sands during undrained cyclic loading. Yokel et al. (1980) proposed that there is threshold cyclic shear strain near 10^{-4} that is sufficient to begin generating excess pore fluid pressure during undrained cyclic loading. The threshold strain level appears to be relatively insensitive to the relative density of the sand and the confining pressure. Based on the current understanding of non-linear soil behavior, it appears that many of the reported time-dependent, fluid-recovery effects observed with acoustic excitation are related to the build-up and dissipation of excess pore fluid pressure.

2.4.3 Laboratory Measurement Techniques

Stevens (1966), based on theoretical work by Lee (1963), developed the basic approach used to apply acoustic excitation to soil samples in this Phase I investigation. Stevens used this technique to measure the complex Young's (and later shear) modulus of unfrozen and frozen soils as functions of the soil bulk density, strain amplitude and confining pressure. The focus of Phase I, however, was on measuring the effects of acoustic excitation on soil-fluid transport properties, rather than on measuring the elastic moduli of soils. Figure 19 in Stevens (1966) was used in this investigation to estimate the Young's modulus of Oklahoma sand and thus the strain amplitudes in the Phase I soil samples.

It is widely recognized (e.g., Scott, 1963, pp. 325-331) that in laboratory-scale soil mechanics or soil dynamics testing the observed soil behavior depends strongly on whether any induced excess pore-water pressure is allowed to dissipate. In soil mechanics parlance, if any induced excess pore-water pressure can dissipate on a time scale shorter than the soil deformation process then the sample is said to be drained. However, if the excess pore-water pressure cannot dissipate, either because the pore fluid effluent lines are closed or the sample has very low permeability that hinders drainage, then the sample response is said to be undrained.

With highly permeable sands such as Oklahoma sand, the soil will be in a drained condition whenever the pore fluid effluent lines are left open, as in the hydraulic conductivity measurements made during this Phase I investigation. In contrast, the hydraulic conductivity of porcelain clay is sufficiently low that excess pore-water pressure can build-up during dynamic loading even when the pore fluid lines are open. It is important to recognize that different drainage conditions may exist in a laboratory sample than are present in-situ due to unavoidable differences in the soil volumes and the lengths of the drainage paths.

2.4.4 *Acoustic Sources*

A review of the acoustics literature revealed a number of references on acoustical sources and the mathematical modeling of source emissions. A brief summary of these findings is presented below.

Omnes (1990) provides a brief review on different downhole sources as well as several requirements for these sources. These requirements include a discussion on the amount of energy needed, the bandwidth of the acoustic source, and the necessary well dimensions, including the mechanical limits of the well.

Mahrer and Zook (1993) report the use of a 1.2 kJoule (kJ) electrodeless seismic source or sparker operating at 300 Hz to 1.2 kHz, and a summary of a test performed at a Texaco test site in Humble, Texas.

Fisher et al. (1993) report the use of a small explosives in a borehole for cavitation induced seismic activity. The explosives used in these test had energies of 30 kJ to 170 kJ.

Paulsson (1994), in cooperation with a number of major international oil companies, has developed a high power seismic source and demonstrated successful long range cross-well and single-well imaging using a downhole hydraulic seismic vibrator. The source operates in the frequency range of 10 Hz to 1 kHz with a power output of a few kilowatts.

Yu and Liu (1992) have developed a model representing a cylindrical acoustic source and its application in borehole acoustics. Similarly, Zhang et al. (1993) have developed a model for multipole sources in a borehole surrounded by a transversely isotropic elastic solid.

U.S. Navy acoustical literature was reviewed and found to contain a good summary of acoustical sources developed for the Navy. This information includes descriptions of sources, performance characteristics, and availability.

The following references indicate areas of acoustic research which show potential as either downhole or surface sources.

Dering et al. (1993) developed high power acoustic beam sources in the infrasound frequency range. They documented analyses and experiments of linear and nonlinear propagation, and conducted analyses and experiments with unique beam forming concepts.

Dering et al. (1994) developed and tested two high power audio range acoustic sources. These sources included a combustion driven siren and a flame vortex resonator. This work also included the development of an antenna for use with the source.

Sollee et al. (1995) developed, tested, and assessed a combustion driven thermo-acoustic resonator, including a multi-element phased array. Propagation under various conditions was also studied.

Figure 3. Comparison of Normalized Modulus Reduction Relationships for Sands (modified after Iwasaki et al., 1978; taken from Sun, Golesorkhi, and Seed, 1988), DOE Contract No. DE-AR21-94MC30360, Phase I.

Figure 4. Damping Ratios for Sands (taken from Seed, Wong, Idriss, and Tokimatsu, 1994), DOE Contract No. DE-AR21-94MC30360, Phase I

Figure 5. Normalized Modulus Reduction Relationships for Clays with Different Plasticity Indices (taken from Sun, Golesorkhi, and Seed, 1988), DOE Contract No. DE-AR21-94MC30360, Phase I

3. LABORATORY TEST APPARATUS

3.1 Test Cell

Figures 6 and 7 show a cross-section and a photograph of the laboratory test apparatus, respectively. An electromechanical shaker, Ling Dynamic Systems Model 411, 44 pound peak force, DC to 9 kHz, is used to excite vibrations in a test soil sample, 1.50" diameter and about 3.50" long. The sample is placed in a thin, heat-shrink mylar jacket (0.005" wall thickness) between two end caps that are fitted with 120 mesh screens for fluid flow measurements. The shaker and sample assembly are enclosed in a sealed chamber that is pressurized to simulate the stress on the soil at different depths in the subsurface. The wall of the chamber is a Plexiglas cylinder so the condition of the sample and the flow of fluids in the inlet and outlet lines can be continuously monitored.

The test apparatus has been designed to perform a 1-D simulation of the soil vapor extraction or NAPL ground water P&T remediation processes. It has been used in this Phase I program to develop a basic understanding of acoustically enhanced NAPL ground water P&T.

As the test sample and top end cap are accelerated by the shaker, the inertial loading of the test sample due to its own mass and the mass of the top end cap generates a variable strain (or fractional change in length) in the sample. This approach (i.e., forced oscillations with a lightly-loaded end condition) is an established technique in experimental mechanics (see Section 2.4.3). While the strain amplitudes in the sample depend strongly on the complicated resonance modes of the three-element composite oscillator (the lower end cap, sample and upper end cap), using this approach, strain amplitudes above 10^{-4} at frequencies below 1 kHz were achieved.

The following electrical and pressure feedthroughs are needed to operate the test apparatus inside the pressurized chamber:

1. an electrical feedthrough supplying power to the shaker;
2. a pair of feedthroughs supplying either the gas or liquid permeant to the soil sample;
3. four electrical feedthroughs for operating the accelerometers and pressure transducers; and
4. three thermocouples for monitoring the temperature of the sample.

The test chamber is made primarily of acrylic plastic and is pressurized up to 100 psi. To address any concerns regarding pressure containment, a Lexan safety enclosure was fabricated to contain any pieces of acrylic plastic in case of failure. A pressure release valve (set at 125 psi) was

mounted on the top plate of the test chamber to prevent the buildup of high pressures that could lead to a failure of the pressure housing. Also, an analog pressure gauge was mounted on the top of the test chamber for use in setting the cell pressure and to provide a readout that is independent of the data acquisition system (DAS).

During the tests with acoustic excitation, the noise levels in the laboratory were well below Cal/OSHA guidelines but were still annoying and not conducive to regular conversation or office work. Hearing protection was provided for the person conducting the tests, for other staff working in the laboratory, and for visitors.

Figure 6. Schematic Cross-Sectional View of the Laboratory Test Cell, DOE Contract No. DE-AR21-94MC30360, Phase I

Figure 7. Photograph of the Laboratory Test Cell, DOE Contract No. DE-AR21-94MC30360, Phase I

3.2 Test Sample Instrumentation

3.2.1 *Dynamic Pressure Transducers*

There is a dynamic pressure transducer (PCB Piezotronics Model #SH1121A21) in direct contact with the soil at each end of the soil sample. The dynamic pressure transducers are used to directly measure the applied stress at each end of the soil sample and to identify frequencies where the soil response is strongly nonlinear. Dynamic stress measurements and an estimate of Young's modulus from an empirical correlation (see figure 19 in Stevens, 1966) were used to estimate the strain amplitude, or fractional change in length, in the soil sample.

The dynamic pressure transducers are extremely rugged and reliable, and despite some corrosion on the surface of one transducer, did not require any special attention or repairs during the testing program.

3.2.2 *Accelerometers*

A small accelerometer (PCB Piezotronics Model #338A01) was mounted at each of the end caps to measure the vibrations (i.e., acceleration, velocity, and displacement) at the top and bottom of the sample. These sensors were used to identify sample resonances and nonlinear soil behavior.

On two occasions, the top accelerometer was returned to PCB Piezotronics for repair of the electrical connector. The connector on this small accelerometer is fragile and may have failed prematurely either due to repetitive handling or the vibrations during our experiment. The accelerometer was recalibrated after each repair with essentially no change in the calibration.

3.2.3 *Thermocouples*

Thermocouples were used to measure surface temperatures along the length of the soil sample, at the top of the shaker, and in the reservoir of fluid permeant (either water, canola oil or Soltrol 220).

Thermocouples are extremely reliable and provide accurate readings up to the point where there is physical interruption of the thermocouple junction. The thermocouples were regularly examined for any signs of corrosion but never required replacement.

3.2.4 *Pressure Transducers*

Semiconductor, strain-gage pressure transducers (Data Instruments XPRO series) were used to measure the quasi-static gas and liquid pressures at various points in the test apparatus. One transducer (0 to 100 psi) was used to continuously monitor the cell confining pressure, while

two additional transducers with different sensitivities (0 to 100 psi and 0 to 15 psi) were used to monitor the fluid inlet pressure during the flow experiments.

3.3 Experimental Control

The laboratory experiments were conducted using a combination of manual and computer-based controls. Pressure settings were set manually using precision regulators and then usually left at a constant value throughout an experiment. The vibration frequency and amplitude were changed either manually or under computer control.

3.3.1 *Signal Generator And Power Amplifier*

A Stanford Research Systems Model DS345 programmable signal generator was used to generate the input waveform either manually or computer control using the IEEE-488 interface. The input waveform was usually sinusoidal at a constant frequency that was chosen because the sample was near resonance, under maximum periodic extension and compression, or the material response was markedly nonlinear. Selected tests were performed by repeatedly cycling through a set of four to six discrete frequencies of interest, usually remaining at each frequency for 15 to 20 seconds so the cycle of frequencies was completed within one to two minutes. The signal output from the signal generator was input to a 600 Watt audio frequency power amplifier. The output from the power amplifier was used to drive the Ling Dynamic Systems Model 411 shaker which has a well-matched eight ohm input impedance.

3.3.2 *Pressure Sources And Regulators*

The various fluid and gas pressures used in these experiments were generated using two cylinders of compressed nitrogen with associated regulators, a fluid reservoir that could be pressurized using compressed nitrogen, and a simple apparatus that can deliver large volumes of water at a nearly constant head. One of the gas cylinders supplied confining pressure to the test chamber for simulating a range of subsurface depths. The other nitrogen cylinder was used to supply the required gas flow for air permeability analyses and, on occasion, to pressurize the pore fluid reservoir for hydraulic conductivity analyses of low permeability clay samples. All of the NAPL-extraction tests were performed using the constant head water reservoir and a reservoir of NAPL.

3.3.3 *Measuring Purging Agent Flow*

During air permeability measurements, the air flow through the sample (up to 1 liter/minute) was measured using a Sierra Top Track Model 821 air mass flow meter. This meter was placed on the inlet side of the sample and proved to be extremely stable and reliable.

For single-phase, liquid permeability measurements, the flow through the sample was measured either by continuously weighing the produced fluid using a digital balance connected to the data acquisition system, or by collecting the effluent liquid in a graduated cylinder or a small inner diameter tube.

For the NAPL ground water extraction tests, a small quantity of hydrophobic Sudan IV dye was added to the canola oil and Soltrol 220 permeants. This enabled us to readily distinguish between the NAPL-surrogates and water. The displaced volumes of water and oil were collected in a series of 10 mL and 100 mL polypropylene graduated cylinders. The cylinders were allowed to stand overnight, so the two phases could totally separate, before measuring the volumes of NAPL and water.

The polypropylene material is preferentially NAPL-wet so the NAPL tends to form a ring around the inside of the graduated cylinder. A set of secondary standards was prepared with known quantities of NAPL so the displaced volumes of NAPL and water could be estimated by visual comparison and interpolation. The resulting uncertainty in the volumetric measurements was approximately 0.1 mL. After completing a series of NAPL-extraction tests, the soil samples were sent to PTS Laboratories in Santa Fe Springs, CA for analysis of the residual NAPL saturation by the Dean Stark toluene extraction (American Petroleum Institute Recommended Practice RP40).

3.3.4 Specialized Flow Checks

Food coloring was added to the water permeant during shakedown testing of the laboratory apparatus and later hydraulic conductivity analyses of the porcelain clay. After testing, the samples were cut in half and confirmed visually that there was a uniform distribution of the permeant within the sample. These simple checks showed (1) uniformity of the Oklahoma sand samples, (2) no fluid bypass using the heat-shrink mylar jackets for the testing of permeable sands, and (3) a soft latex rubber jacket under 20 psi confining pressure eliminates any fluid bypass for hydraulic conductivity analyses of clay.

3.4 Data Acquisition and Archiving System

3.4.1 Overview

The DAS was developed to acquire data from the various sensors and instruments in the experiment. It is mainly comprised of acquisition hardware, sensors, and a graphical software interface that controls the instrumentation and enables one to easily view, acquire, and save the acquired data. The DAS hardware consists of a digitizing board which captures the majority of the data, a general purpose interface bus (GPIB) controller card that enables communication with GPIB (IEEE-488) instruments, and the sensors themselves. The software interface was written using LabVIEW, a commercial programming environment specifically designed for laboratory data acquisition and instrument control. LabVIEW is a graphical program development application

with extensive libraries of functions for data acquisition, instrument control (GPIB and serial), data analysis, data presentation, and data storage.

To store and organize the vast amounts of data acquired by the DAS, a custom database was developed and used. The database stores all the vital information for each experiment, including the acquired waveforms. One can easily search and organize the experimental data by a variety of key fields.

3.4.2 Data Acquisition System

The center of the DAS is a PC computer which runs the LabVIEW software, and contains the digitizing board and GPIB controller. The majority of the sensors are tied into the computer via the digitizing board. A schematic of the entire DAS is shown in Figure 8. The function and importance of each instrument has been documented in Sections 3.1 and 3.2.

The heart of the DAS system is the high-resolution digitizing board (National Instruments AT-MIO-16X). The board takes analog inputs from up to 16 channels and converts them at 16-bit resolution to digital signals which the computer can read. The board is capable of a sustained sampling rate of 100 kSamples/sec.

The interface between the digitizing board and computer was written using LabVIEW. Programs written in LabVIEW are termed virtual instruments (VIs) due to their close emulation in appearance and function to actual instruments.

A library of LabVIEW virtual instruments were developed to interface the various sensors and instruments to the digitizing board and computer. The main data acquisition VI the user interacts with is shown in Figure 9.

Figure 8. Schematic of the Data Acquisition System, DOE Contract No. DE-AR21-94MC30360, Phase I

Figure 9. Front Panel of the Data Acquisition System Virtual Instrument, DOE Contract No. DE-AR21-94MC30360, Phase I

This VI allows the user to control all aspects of the DAS including which channels are sampled, the sampling rate of each channel, the gain applied to each channel, the length of the acquisition, and whether to save the data. The data can also be viewed on the screen in real time in actual engineering units. Some quantities such as root mean square (RMS) values and permeability are also calculated in real time as the acquisition proceeds. GPIB instruments, such as the function generator, and serial instruments, such as the mass balance, can also be controlled with this VI.

Behind the front panel of the DAS VI is a sophisticated programming code which handles the sequencing of the data acquisition, the data display, the data file creation, and user interaction (see Figure 10).

The programming is divided into a series of subroutines, called SubVIs, each handling a particular task. All the routines are located in a LabVIEW library file named "SARA DOE.LLB".

3.4.3 Data Archiving

There are various data files saved during data acquisition. Each data file basically stores the waveform of the data acquired, information as to the setup of the digitizing hardware, as well as general information such as the date and time of the acquisition. The files have different filenames based upon the type of data acquired (high speed data, low speed data, etc.).

To organize and store the vast amount of data acquired, a structured query language (SQL) database application was written using PowerBuilder, a database development software. The database reads the LabVIEW data files and stores the information into appropriate tables and fields in the database (see Figure 11). Storing the data in a database allows data to be viewed, arranged, searched, and organized in an intuitive and logical manner.

All the information contained within the original LabVIEW datafile, including the waveform itself, is stored in the database. In fact, once the data is stored within the database, the original LabVIEW data files can be deleted or archived. The LabVIEW data files can be recreated at any time from the data in the database using the export routines in the database.

Figure 10. Section of the Programming Code Behind the Data Acquisition System Virtual Instrument, DOE Contract No. DE-AR21-94MC30360, Phase I

Figure 11. Main Tables of the Data Acquisition System Database, DOE Contract No. DE-AR21-94MC30360, Phase I

3.4.4 Data Analysis

LabVIEW provides a host of routines to help analyze the data acquired. A series of LabVIEW programs were written to read and analyze the data files generated by the DAS VI. These programs are located in a LabVIEW library entitled "POSTPRSS.LLB".

4. SAMPLE MATERIALS AND SURROGATES

4.1 Sample Materials

The following commercially-available materials were used for this testing program: Oklahoma 90 sand, Ajax all-purpose sand, Aardvark porcelain clay, and a silt-sized, crushed potassium feldspar. These materials were used because (1) they are already relatively well-characterized, (2) they are readily available for additional testing, and (3) other investigators could easily obtain the same materials to verify our results.

4.1.1 *Oklahoma Sand*

Oklahoma 90 sand is a natural, aeolian, highly-sorted, pure-quartz fine sand with a mean grain size of 160 microns. Individual sand grains are well-rounded. Oklahoma sand is free of clay minerals and contains only trace amounts of silt-sized quartz.

4.1.2 *Ajax Sand*

Ajax all-purpose sand (Ajax Cement Co., Inc., Atwood, CA) is a poorly-sorted sand with grains up to 5 mm in diameter. To prepare samples of this material, the as-received material was shifted through a sieve with screen openings near 2 mm. The sieved sand has a bimodal particle size distribution and contains about 9% total silt and clay.

4.1.3 *Artificial Silt*

Clean silts are relatively uncommon in nature. For this testing program, samples from the coarsest portion of the crushed Custer feldspar (Pacer Corporation, Custer, SD) that is used to manufacture Aardvark porcelain clay were used. The laser particle size analyses (described below) showed that Custer feldspar contains approximately 20% clay-size particles. For these experiments, the as-received material was washed through a 400 mesh sieve to remove the finer particles. The remaining particles have diameters between 38 and 45 microns, and are classified as a coarse silt.

4.1.4 Porcelain Clay

Aardvark porcelain clay (Aardvark Clay and Supplies, Santa Ana, CA) contains approximately 25% silt-sized crushed quartz, 25% silt-sized crushed feldspar, 25% kaolin and 25% "ball" clay from Kentucky. This prepared material is a realistic analog for many silty clay soils. It contains no air bubbles and is essentially isotropic.

A sample of Aardvark porcelain clay was submitted to a commercial soil testing laboratory (Earth Tech, Huntington Beach, CA) for testing. Earth Tech measured the clay's hydraulic conductivity (ASTM D5084), water content (ASTM D2216), and Atterberg limits (ASTM D4318). The results (Appendix A) show that Aardvark porcelain clay has a hydraulic conductivity for a clay ($1.9\text{E-}8$ cm/sec at 4.8 psi effective confining pressure). The moisture content is 25.9% by dry weight. The Atterberg limits are widely used in soil mechanics to classify clay soils. Aardvark porcelain clay is classified as a lean clay (classification CL) with a liquid limit of 34, plastic limit of 16, and P.I. of 18.

4.2 Laser Particle Size and Mineralogical Analyses

4.2.1 Laser Particle Size Analyses

Samples of the Oklahoma 90 sand, Aardvark porcelain clay, Custer feldspar (as received) and Ajax all-purpose sand (after sieving through a 2 mm sieve) were sent to Core Petrophysics, Inc. in Houston, TX for laser particle size analysis. For Ajax sand, it was necessary to perform a combined sieve and laser particle size analysis since the laser particle size analyzer cannot measure grains larger than 0.5 mm. The particle size analyses are included in Appendix A.

4.2.2 Mineralogical Analyses

Samples of the crushed feldspar and Aardvark porcelain clay were sent to Sandia National Laboratories in Albuquerque for X-ray diffraction (XRD) analyses to determine the composition of Aardvark porcelain clay, and whether there is mineralogical evidence for 20% clay in Custer feldspar.

The XRD analyses (Appendix A) show that the Custer feldspar is essentially pure K-feldspar (microcline) with only a trace of illite. There is no evidence of kaolinite or other clay minerals. The crushing process generates approximately 20% clay-size particles of microcline, apparently due to this mineral's excellent cleavage. The XRD analyses also show that Aardvark porcelain clay contains only kaolinite, quartz and K-feldspar. The "ball" clay is another form of kaolinite with a different particle size or crystallinity.

4.3 Surrogate Contaminants

Two non-aqueous phase liquids (NAPLs) were used during the testing program. Most of the NAPL-extraction tests were performed using a vegetable oil (Crisco canola oil) dyed red with a small amount of Sudan IV dye. A sample of the canola oil was sent to Core Laboratories in Long Beach, CA for analysis of the oil density (ASTM D4052) and viscosity (ASTM D445). The density of the canola oil is 0.9182 g/mL at 70°F, making it less dense than water and by definition a lighter than water NAPL or LNAPL. However, its viscosity, 65 cPoise at 70°F, is significantly higher than for many fuel hydrocarbons and chlorinated hydrocarbons.

Questions were raised whether the canola oil may behave differently than fuel hydrocarbons, due either to its greater viscosity or surface chemistry. To answer these questions, additional NAPL-extraction tests in Oklahoma sand using Soltrol 220 were performed. Soltrol 220 is a refined isoparaffinic solvent manufactured by Phillips Petroleum Company. Its fundamental physical properties are well known from previous studies. At 73°F, the oil density is 0.789 g/mL, the viscosity is 4.83 cPoise (Illangasekare et al., 1994), the surface tension with water is 42 dynes/cm, and the surface tension with air is 27 dynes/cm (Szlag and Illangasekare, 1992). Soltrol 220 is also an LNAPL.

Canola oil and Soltrol 220 were selected as NAPL-surrogates because of they are relatively non-hazardous and non-toxic. While both of these surrogates are LNAPLs, the Soltrol 220 will provide a first approximation for remediation of fluids that are denser than water or DNAPLs. This is founded on Wilson et al. (1990) where Soltrol 130 was the most commonly used fluid in their laboratory investigation of residual liquid organics. Based on this Cohen and Mercer (1993, p. 4-19) in their work on evaluating DNAPLs recommended Soltrol 130 as an ideal fluid for determining residual saturation for a porous media. The densities and viscosities of Soltrol 130 and 220 are comparable for the purposes of this investigation.

4.4 Sample Preparation and Handling

4.4.1 Oklahoma And Ajax Sands

Oklahoma and Ajax sand samples were prepared using a slow, dry pluviation method (described below). This procedure is not an ASTM method, but the same approach is widely used in soil dynamics testing laboratories to prepare densely-packed samples with a narrow range of porosities.

Samples of Oklahoma sand were prepared with remarkable uniformity. The mean porosity of the Oklahoma samples is 31.8% with a range of plus or minus 0.2%, while the hydraulic conductivity values vary by only 6%. The samples of Ajax sand were much more variable, apparently due to the intrinsic heterogeneity of this soil material. The sample porosities vary between 32.7 and 33.7%, while the hydraulic conductivity values vary by about 20%.

The sand and silt samples are placed in a double jacket, consisting of an inner heat-shrinkable, thin-walled mylar jacket (0.005" thick) and an outer latex rubber jacket (0.012" thick). The mylar jacket enables one to make ideal cylindrical samples, while the outer rubber jacket is used to make the pressure seal.

To eliminate the possible effects of changes in relative humidity, bulk quantities of sand were stored in a sealed plastic containers. The small quantity of sand was then placed in a glass separatory funnel, and the sand was allowed to "rain" slowly through a grounded brass funnel into the mylar jacket. The brass funnel was positioned about 10 cm above sediment surface and continuously moved by hand in an irregular, circular motion so as to fill the jacket in a uniform manner. The surface of the sand was leveled before inserting the upper end cap. A single 120 mesh stainless steel screen is mounted on the face of each end cap to prevent the loss of sand grains. The outer latex rubber jacket is then applied using a vacuum sleeve expander.

For the hydraulic conductivity analyses, all samples were initially vacuum-saturated to eliminate the known artifacts caused by incomplete saturation. Aardvark porcelain clay is manufactured so that it contains no air bubbles. For the NAPL-extraction tests, as described more fully in Section 5.2.3, the samples were initially vacuum-saturated with water and then flooded with NAPL until no more water was produced. Consequently, in these tests, the smaller pores and mineral surfaces are preferentially water-wet as they would be in the subsurface.

4.4.2 Artificial Silt

Silt samples were prepared by mixing the dry, sieved feldspar powder with water and pouring the "mud" into a thin-walled mylar jacket. Excess water was allowed to free drain from the sample through the lower end cap. In this initial state, the silt is thixotropic and has a porosity over 50%. By gently squeezing the jacket, it was possible to momentarily liquefy the sample and to progressively push the top end cap into the mylar jacket. The resulting silt material is water-saturated and has a porosity near 43.5%. The samples were finally consolidated under 20 psi confining pressure for two hours before testing.

4.4.3 Porcelain Clay

Clay samples were prepared from blocks of clay by hand-trimming using a manual soil lathe. After trimming to the desired diameter, the clay samples were picked up using a split shell, trimmed to length, positioned between the upper and lower end caps and finally jacketed using a single latex rubber membrane. Earth Tech's hydraulic conductivity analyses, at a net effective confining pressure of only 4.8 psi, show that a soft latex rubber jacket under confining pressure effectively eliminates any fluid bypass.

5. LABORATORY TEST MATRIX

5.1 Summary of Laboratory Experiments

Table 1 shows the matrix of tests conducted on four different soil types (1) Oklahoma sand, a pure-quartz, well-sorted fine sand, (2) Ajax all-purpose sand, a poorly-sorted sand with 9% silt and clay, (3) an artificial silt, and (4) a porcelain clay, a good analog for many silty clays.

Table 1. Test Series Summary, DOE Contract No. DE-AR21-94MC30360, Phase I

Soil Type	Hydraulic Conductivity	Air Permeability	NAPL Extraction Purging Canola Oil with Water	NAPL Extraction Purging Soltrol 220 with Water
Oklahoma Sand	X	X	X	X
Ajax Sand	X	X	X	
Silt	X			
Clay	X			

In these tests, we endeavored to determine the optimal acoustic parameters to maximize the fluid and contaminant extraction rates for a wide range of soil types usually under saturated conditions. The laboratory testing measurement techniques were based on Stevens (1966), see Section 2.4.3.

Each test, or set of experiments in the case of NAPL-extraction tests, included baseline conditions without acoustic excitation, and then excited conditions with acoustic excitation either at a single frequency or at a sequence of frequencies between 100 Hz and 10 kHz. Measurements were generally made at three different power levels in addition to the baseline condition. The NAPL-extraction tests simulate a ground water-NAPL pump and treat operation.

Tests were generally run using five-minute duty cycle, i.e. five minutes on at a particular frequency and five minutes off, to provide information on soil hysteresis effects and how long the effects of acoustic excitation persist in a particular soil. In general, the air permeability and hydraulic conductivity analyses for the two types of sand were conducted at two confining pressures (30 and 10 psi) to simulate the stress conditions in the shallow subsurface, the NAPL-extraction tests were conducted at a nominal 30 psi confining pressure, and the hydraulic conductivity analyses of porcelain clay were conducted at 23 psi.

Modest time-dependent reductions in hydraulic conductivity with acoustic excitation were observed during some of the Phase I testing, presumably due to sample consolidation. However,

the test equipment utilized did not allow for a measure of porosity. The air permeability analyses were not Klinkenberg-corrected since (1) the intent of this portion of the test program was to measure soil properties relevant to soil vapor extraction in the vadose zone, rather than using air permeability analyses to estimate hydraulic conductivity values in the saturated zone, and (2) the Phase I testing was only conducted at two confining pressures which are not sufficient for the correction.

Nearly identical samples of the Oklahoma sand and porcelain clay materials were produced for testing, but there was greater variability between the samples of Ajax sand and the artificial silt. Since there inevitably will be some variability among same soil-type samples, for the air permeability and hydraulic conductivity analyses, the test program focused on the relative changes in fluid flow rates in the same sample under baseline and excited conditions using a five-minute duty cycle. For the NAPL-extraction tests, it was necessary to use multiple samples and each test was repeated up to six times to verify that the results are statistically significant and not due to sample variability. The samples of Oklahoma sand were sufficiently uniform that successful comparisons between samples were possible, but this proved to be impossible with the Ajax sand samples due to their intrinsic variability.

Appendix B is a chronological log of the experiments conducted during this program and provides specific information on how individual tests were conducted, including the pressures, frequencies and acoustic power levels. Any particular problems are noted. The following test procedures are generalized to convey the overall view of the Phase I test conduct.

5.2 Test Procedures

5.2.1 Air Permeability Testing

The following test procedures that were used to measure the air permeability of Oklahoma sand. Acoustic data was collected at three power levels at confining pressures of 10 and 30 psi.

1. Performed a system calibration by flowing air through a Plexiglas tube with the same length and diameter as the samples.
2. Excited the sample at a low strain level near 10^{-5} , at sequence of different frequencies (100 Hz, 300 Hz, 1 kHz, 3 kHz, 10 kHz and two resonant frequencies) while continuously measuring the gas mass flow and air permeability. A duty cycle of five minutes on and five minutes off between frequencies was used to check for time-dependent, sample hysteresis effects.
3. Increased the strain level to an intermediate value near 3×10^{-5} and repeated the same sequence of frequencies (using the same duty cycle) while continuously measuring the air permeability.

4. Increased the strain level to a high level above 10^{-4} and measured the air permeability using the same sequence of frequencies and duty cycle.
5. Excited the sample with high strain, repetitive impulsive waveforms. The signal generator was used to generate a variety of unipolar pulse trains. Continuously measured the air permeability as the excitation was turned on and off using a five minute duty cycle.

Simplified procedures were used to measure the air permeability of Ajax sand at a single confining pressure of 30 psi. Following baseline analyses, data were collected over a full range of frequencies (100 Hz, 300 Hz, 750 Hz, 1 kHz, 2 kHz, 5500 and 10 kHz) using an intermediate power level that gave strain amplitudes near 3×10^{-5} .

5.2.2 Hydraulic Conductivity Testing

5.2.2.1 Oklahoma Sand

The following test procedures that were used to measure the hydraulic conductivity of Oklahoma sand. Acoustic data was collected at three power levels at confining pressures of 10 and 30 psi. For water at room conditions, a hydraulic conductivity of $1.0\text{E-}3$ cm/sec corresponds approximately to a water permeability of $1.0\text{E-}12$ meters squared in SI units, which for practical purposes is equivalent to 1 Darcy in petroleum engineering units.

1. Performed a system calibration by flowing water through a Plexiglas tube that has the same length and diameter as the samples.
2. Excited the sample at a low strain level near 10^{-5} , at a sequence of three different frequencies (100 Hz, 1 kHz and 10 kHz). A digital balance was used to continuously record the throughput of water as the sample was excited. The average flow rate was calculated by best-fitting a line to the data over a five minute interval. The same duty cycle (five minutes on and five minutes off between frequencies) was followed to check for time-dependent, sample hysteresis effects.
3. Increased the strain level to intermediate values near 3×10^{-5} and measured the hydraulic conductivity using the same sequence of frequencies and duty cycle.
4. Increased the strain level to a high level above 10^{-4} and measured the hydraulic conductivity using the same sequence of frequencies and duty cycle.

5.2.2.2 Ajax Sand

The following test procedures that were used to measure the hydraulic conductivity of Ajax sand. Acoustic data was collected at three power levels at confining pressures of 10 and 30 psi.

1. Performed a system calibration by flowing water through a Plexiglas tube that has the same length and diameter as the samples.
2. Excited the sample at an intermediate strain level near 3×10^{-5} at a sequence of three frequencies (100 Hz, 1 kHz and 10 kHz) while continuously monitoring the throughput of water. The same duty cycle (five minutes on and five minutes off between frequencies) was followed to check for time-dependent, sample hysteresis effects. During acoustic excitation muddy water was produced from the sample, but without excitation the effluent water was essentially clear.
3. Increased the strain level to a high level above 10^{-4} . Monitored the effluent water and measured the hydraulic conductivity using the same sequence of frequencies and duty cycle. With more intense acoustic excitation, the effluent water became muddier and the hydraulic conductivity started to drop. Reversing the flow direction was attempted but the test was abandoned prior to developing definitive insight into potential optimization strategies.

5.2.2.3 Artificial Silt

Simplified test procedures were used to measure the hydraulic conductivity of the artificial silt.

1. Performed a system calibration by flowing water through a Plexiglas tube that has the same length and diameter as the samples
2. Collected baseline data at 20 psi confining pressure where the effluent water was essentially clear.
3. Collected data using a single excitation frequency near 665 Hz. Operating at a near maximum power level, the throughput of water was continuously measured using the digital balance as the sample was excited for approximately ten minutes. With acoustic excitation, milky water was produced from the sample and the hydraulic conductivity decreased by about 35%. The test was continued following roughly the same duty cycle (five minutes on and five minutes off) to check for time-dependent, sample hysteresis effects.

5.2.2.4 Porcelain Clay

The following simplified test procedures were used to measure the hydraulic conductivity of the porcelain clay.

1. Performed a system calibration by flowing water through a Plexiglas tube that has the same length and diameter as the samples
2. The clay samples were allowed to equilibrate overnight with the confining pressure regulated at 23 psi, the pore fluid inlet pressure was 12 psi and the pore fluid outlet pressure was one atmosphere. The inlet reservoir was pressurized using compressed

nitrogen and the fluid output from the sample was collected in a small diameter (0.062 inch inside diameter) plastic tubing.

3. The following morning, the output fluid level was marked and then measured at regular 30 minute intervals using a centimeter rule. The fluid level in the tube rose about 2 cm during each 30 minute period, and the fluid level could be easily read to the nearest 0.05 cm. Using this approach, the baseline (without acoustic excitation) hydraulic conductivity was measured over a four to six hour period.
4. Acoustic excitation was then applied continuously at a sequence of frequencies (e.g., 360 Hz, 3.2 kHz, 860 Hz, 10.2 kHz, 1160 Hz and 3.7 kHz, with 20 seconds at each frequency) at a high power level while the hydraulic conductivity was measured. The fluid throughput increased so the fluid level was measured at regular 20 minute intervals over a two to three hour period.

The analyses were repeated using a new sample and a similar sequence of frequencies but increased the acoustic power to the maximum level possible. For this test, the temperature of the sample was monitored and a layer of oil was placed in the inlet reservoir to keep air from going into solution in the water. A shear failure was observed near the base of the sample immediately after turning on the excitation. The flow rate steadily increased and excitation was continued for two hours until the sample temperature reached 95° F. The excitation was ceased and the flow rate was monitored for another four hours, observing a time-dependent, sample hysteresis effect.

5.2.3 NAPL Extraction - Purging With Water

A majority of the NAPL-extraction tests were performed using a canola oil NAPL surrogate. Samples were prepared following the procedures described in the previous section. The sample dimensions and the total mass of the dry sample assembly were recorded. The sample was then placed in the test apparatus under confining pressure and vacuum saturated with water. After saturating the sample, the water flow rate was measured over a one-minute period. The sample was then removed from the pressure chamber and weighed. The sample was returned to the pressure chamber and flooded with NAPL until no more water was produced. The displaced fluids were collected in a graduated cylinder and the volume of displaced water was measured. The final NAPL flow rate was recorded. The sample was then removed from the pressure chamber and weighed again.

The sample was replaced in the test cell, and water was allowed to flow from the water reservoir through the sample, from the bottom to the top, under an essentially constant head near 150 cm of water. The flow rates were high enough to satisfy the Rapoport and Leas (1953) condition and still give a reasonable capillary number near 10^{-5} so the injected water does not tend to breakup NAPL ganglia.

The progress of the water flood was monitored visually and the displaced fluids were collected in a series of graduated cylinders. For the first part of the test, where the flow rates are low, the displaced NAPL and water were collected in a series of ten 10 mL graduated cylinders at

one-minute intervals. After water breakthrough, where the flow rate increases, the displaced fluids were collected in a series of ten 100 mL graduated cylinders at five-minute intervals. The flooding process typically continued for one hour which was long enough so that there was negligible incremental recovery of NAPL (i.e., the sample was near irreducible oil saturation). The graduated cylinders were allowed to stand overnight before the measuring the volumes of NAPL and water. Following the test, the sample was sent to a commercial laboratory for an independent analysis of the residual NAPL saturation.

Identical procedures were followed using separate samples to evaluate how acoustic excitation affects the NAPL extraction rate. As with the hydraulic conductivity analyses of clay, the acoustic excitation was applied continuously during the course of the final water flood.

Most of the experiments were conducted using a single, sample-specific excitation frequency near 1100 Hz where the accelerometers show that the sample is under maximum periodic extension and compression. At the end of the testing program, a few additional samples were analyzed using a sequence of up to six excitation frequencies.

6. LABORATORY RESULTS AND ANALYSIS

6.1 Air Permeability

Air permeability, k_a , measurements were conducted on (1) well-sorted Oklahoma sand and (2) poorly-sorted Ajax sand. The air permeability measurements were made under baseline and excited conditions using both sinusoidal and impulsive excitations. These measurements were not Klinkenberg-corrected for reasons cited in Section 5.1.

6.1.1 Oklahoma Sand

More than 29,000 individual air permeability measurements were made under computer control during 14 tests on a dense, random pack of Oklahoma sand. The testing indicated (see combined excited test results in Figure 12) that the mean values of k_a for the various tests all ranged from approximately 7.0 to 8.0 μm^2 (7.0 to 8.0 darcies) and were unaffected by acoustic excitation under the range of conditions tested.

Tests 1 - 5 evaluated the effects of the acoustic excitation amplitude and frequency on the air permeability. Each test was conducted at a sequence of frequencies, that is 0 Hz (or baseline), 100 Hz, 0 Hz, 300 Hz, 0 Hz, 1 kHz, 0 Hz, 3 kHz, 0 Hz, 1 kHz, and 0 Hz, and at three different power levels. The mean values of k_a ranged from 6.96 to 7.75 μm^2 , with standard deviations of 0.34 - 0.50 μm^2 .

During Test 6, the sample was analyzed under alternating baseline conditions and resonant frequencies of 930 and 3730 Hz. This test also gave a null result.

Tests 7 - 10 repeated the analyses in Tests 1 - 5 at 10 psi confining pressure. The mean k_a values ranged from 6.92 to 8.20 μm^2 , with standard deviations of 0.32 - 0.51 μm^2 . The results at 10 psi are not as tightly clustered as the analyses at 30 psi but the mean values of k_a are still within two standard deviations of the other analyses.

Test 11 repeated the analyses in Test 6 at a lower 10 psi confining pressure. This test consisted of 200 k_a measurements at sequence of frequencies: 708 Hz, 0 Hz, 708 Hz, 3275 Hz, and 0 Hz. The resonant frequencies were lower than in Test 6 because the sand has a lower modulus at the lower confining pressure. This test also gave a null result.

Tests 12 - 14 evaluated the effects of impulsive acoustic excitation. The excitation waveform consisted of one-half of a 1 kHz square wave where the pulse width of the "box car" was a variable percentage (0, 5, 0, 6.3, 0, 8.3, 0, 12.5, 0, 25, 0, 50, and 0%) of the full duty

cycle time. The analyses were repeated for three power levels. The tests were conducted at a 100 psi confining pressure. The mean k_a values ranged from 7.19 to 7.85 μm^2 , with standard deviations of 0.34 - 0.68 μm^2 . These tests using impulsive excitation also gave a null result since the mean k_a values are within two standard deviations of each other.

6.1.2 *Ajax Sand*

Two different tests were performed using Ajax sand, with a total of 4,100 individual k_a measurements.

Test 1 averaged 150 k_a measurements at a sequence of excitation frequencies (0 Hz, 100 Hz, 300 Hz, 750 Hz, 1 kHz, 2 kHz, 5.5 kHz, 10 kHz, 5.5 kHz, 2 kHz, 1 kHz, 750 Hz, 300 Hz, 100 Hz, 820 Hz, and 0 Hz) using an intermediate acoustic power level. The confining pressure was 30 psi. The mean k_a values ranged from 3.00 to 3.43 μm^2 , with standard deviation of 0.10 - 0.14 μm^2 . While the range of k_a values was just over three standard deviations (3 s.d.), the maximum difference in k_a between successive frequencies, i.e., between baseline and excited conditions, was only 0.19 μm^2 or slightly more than 1 s.d.

Test 2 evaluated the reliability of using fewer repeat measurements at each excitation frequency. In this case, 100 k_a measurements were taken at each of the signal frequencies used in Test 1, with the same confining pressure, air flow rate, and excitation signal amplitudes used in Test 1. The mean k_a values range from 1.97 to 2.28 μm^2 , with standard deviations of 0.04 - 0.06 μm^2 .

The Ajax sand tests essentially yielded null results. However, the air permeability did decline over the course of the 4,100 measurements, suggesting that there was some incremental consolidation of the sample as a result of the acoustic excitation. With the test apparatus used in this Phase I investigation, the suggested reduction in porosity could not be directly measured.

6.1.3 *Summary*

Null results are a very frustrating laboratory outcome. Nevertheless, these results strongly suggest that acoustic excitation has no significant effect on the air permeability of highly-permeable sands under the range of conditions tested. Two likely explanations are (1) the air is very compressible so there can be no buildup of anomalous pore pressures and (2) the flow of air is essentially unaffected by microscopic deformation of the pore throats and channels.

The air permeability tests were conducted to determine if acoustic excitation could affect air permeability and thus vadose zone remediation processes. It needs to be emphasized that these null results pertain only to the air permeability of the grain framework. Further testing should be conducted with a variety of soil types, partially saturated with volatile to semi-volatile solvents, to determine if any of the pore fluids effects hypothesized in Figure 2 are operative.

Figure 12. Air Permeability of Oklahoma Sand Excitated Results, DOE Contract No. DE-AR21-94MC30360, Phase I

6.2 Hydraulic Conductivity

Hydraulic conductivity, K_h , measurements were conducted on (1) well-sorted Oklahoma sand, (2) poorly-sorted Ajax sand, (3) artificial silt, and (4) porcelain clay. These tests were generally conducted under alternating baseline and excited conditions using sinusoidal excitation at a sequence of frequencies and acoustic power levels.

6.2.1 Oklahoma Sand

Under computer control, 3,750 K_h measurements were made on four samples of Oklahoma sand. Each test averaged 150 K_h measurements at a sequence of frequencies (0 Hz, 100 Hz, 1 kHz, and 10 kHz) using three acoustic power levels and two confining pressures (30 and 10 psi). The mean K_h values ranged from 4.93 to 7.65 E-3 cm/sec, with standard deviations of 0.13 - 0.79E-3 cm/sec.

There was a moderate and steady decline in the K_h of Oklahoma sand during the course of these measurements, suggesting some incremental consolidation of the unconsolidated grain pack as a result of the acoustic excitation. However, with the test apparatus used the corresponding porosity measurement could not be made.

These results stand in contrast to those reported by Reddi et al. (1993, 1994). They used a 20 kHz, 500 Watt ultrasonic horn operating in a tank of sand at atmospheric pressure (see Section 2.2.1) and they observed an increase in hydraulic conductivity of up to several orders of magnitude with acoustic excitation. They reported, however, possible fluid bypass along the cell wall and ultrasonic horn during their excited measurements.

6.2.2 Ajax Sand

Under computer control, 3,600 K_h measurements were made on four samples of Ajax sand. Each test averaged 150 K_h measurements at a sequence frequencies (0 Hz, 100 Hz, 1 Hz, and 10 kHz) using two acoustic power levels and two confining pressures (30 and 10 psi). The mean K_h values ranged from 3.11 to 6.06E-3 cm/sec, with standard deviations of 0.10 - 0.27E-3 cm/sec.

The K_h decreased during the course of the 3,600 measurements apparently due to progressive plugging of the pore channels with mobilized fine-grain particles. At high acoustic power levels, large amounts of muddy water were produced from the sample, while during baseline testing the effluent water was clear. This finding is consistent with

Reddi et al. (1993, 1994) except that their work was done with the sediment at atmospheric pressure.

Mobilization of fines is one of the mechanisms postulated in Figure 1. This will be advantageous for remediation since the contaminants are generally sorbed on the fine-grained

fraction of the aquifer. Additional testing is needed to optimize the production of fines from an aquifer while minimizing the adverse side effects of mobile fines plugging the pores.

6.2.3 Artificial Silt

The artificial silt had a surprisingly high baseline K_h of $1.01\text{E-}3$ cm/sec (approximately 1 darcy). By sieving, a sample with only coarse silt-size particles was prepared, and it turned out that this material had a K_h that probably is not representative of natural silt materials. The permeability is so high because the silt grains are very well sorted and the sample has a porosity near 43.5%.

During the baseline measurements, the effluent water was essentially clear. With excitation, as in the Ajax sand, milky water was produced from the sample and an initial 34% reduction in the K_h to $0.66\text{E-}3$ cm/sec occurred. This conductivity reduction is attributed to the combined effects of particle migration and excitation-induced consolidation. The test continued for another hour following a rough five minutes on and five minutes off duty cycle to check for time-dependent, sample hysteresis effects. There was a suggestion that the K_h was increasing by about 10% with acoustic excitation. However, due to limited time, this promising trend could not be investigated further.

A silt sample was prepared with hydraulic properties similar to a well-sorted very fine sand. With such a high permeability, the soil material was being tested under essentially drained conditions. If there was in fact a 10% increase in K_h with excitation, then this may indicate that excitation was inducing a slight increase in the pore-water pressure. This suggests that it may be possible to reduce the tendency for pore plugging with fines migration by varying the acoustic power level or sequence of excitation frequencies.

6.2.4 Porcelain Clay

The K_h of two samples of a porcelain clay were measured under baseline and excited conditions. A sample of the same material was also sent to Earth Tech's geotechnical laboratory, where $K_h = 1.9 \times 10^{-8}$ cm/sec was measured under similar stress conditions. Note that the porcelain clay is comprised of at least 75% non-expansive minerals.

Figure 13 shows the cumulative water flow under baseline and excited conditions for our initial porcelain clay measurement. The baseline value of $K_h = 2.30 \times 10^{-8}$ cm/sec is comparable to Earth Tech's analysis. The excited value of $K_h = 4.58 \times 10^{-8}$ cm/sec represents a nearly two-fold increase. The excited data were obtained with the power amplifier at its highest gain setting, and with the function generator output at 1 V peak to peak (pp).

Figure 14 shows the cumulative flow behavior during the second set of porcelain clay measurements. The baseline $K_h = 1.35 \times 10^{-8}$ cm/sec is somewhat lower than the values measured by Earth Tech and in our initial test (Figure 13). For this sample, the function generator output

was increased to 2 V pp to give the maximum possible acoustic power. With the increased acoustic power, a shear failure developed near the base of the test sample immediately after turning on the acoustic excitation. After two hours of excitation, the sample temperature increased from 71 to 95 degrees F, due to the combined effects of heating from the shaker and anelastic attenuation, and the water flow rate increased nearly six-fold.

Figure 15 shows the time-dependent K_h variations corresponding to the results in Figure 14. The excited hydraulic conductivities peaked at a value of $K_h = 5.5 \cdot 10^{-8}$ cm/sec, which corresponds to over a four-fold increase in the intrinsic permeability. The significance of this conductivity increase is discussed in Section 7.3.2. The flow rate increased even more due to the temperature-dependent reduction (about 30 percent) in the viscosity of water. This figure also shows how K_h declined with time after acoustic excitation to a value ($K_h = 1.9 \cdot 10^{-8}$ cm/sec) that is higher than the baseline value for this test but within the range of the other baseline analyses. These data are suggestive (but not conclusive) that the post-excitation intrinsic permeability of the clay was increased. Additional testing is required to investigate this effect.

6.2.5 Summary

The hydraulic conductivity analyses for the Oklahoma and Ajax sand samples gave essentially null results. It is widely recognized in laboratory-scale soil dynamics testing (see Section 2.2.3) that the observed soil behavior depends strongly on whether any induced excess pore-water pressure is allowed to dissipate. The most likely explanation for the null results is that the soil sample was always in a drained condition for these analyses, because the pore fluid lines were left open during the permeability measurements, and there could be no buildup of excess pore-water pressure. The drainage conditions in-situ are expected to be closer to undrained and this could have a major positive effect on the soil-fluid transport properties.

The hydraulic conductivity of Ajax sand decreased during the course of the tests apparently due to fines mobilization and progressive plugging of the pore channels. However, the mobilization and production of fines may be useful for remediation since the contaminants are generally sorbed on the fine-grained fraction of the aquifer (see Section 6.2.2). Additional testing will be required to optimize the production of fines while minimizing the tendency to plug pores.

The hydraulic conductivity analyses of the porcelain clay samples gave the most favorable results in these laboratory tests, i.e., a two- to four-fold increase in the hydraulic conductivity with acoustic excitation. The clay has very low permeability and these results provide insight into the possible operative mechanisms for acoustic excitation effects on the soil matrix. The increase in K_h is probably related to the generation of excess pore-water pressure during dynamic loading. Particle agitation may be another soil matrix effect observed in these clay tests (Figure 1), and possibly dynamic excitation of the double layer may also have a still unknown effect.

The results in Figure 15 suggest that there may be a long-term increase in the hydraulic conductivity of aquitards after acoustic excitation. The time-dependent decay of K_h after excitation also suggests that it may be possible to implement acoustically enhanced remediation using less-expensive, intermittent acoustic excitation.

In summary, the results in Figures 13, 14, and 15 strongly suggest that acoustically enhanced remediation is viable, and some insights into the operative mechanisms have been developed.

Figure 13. Cumulative Flow Volume versus Time for the First Porcelain Clay Test, DOE Contract No. DE-AR21-94MC30360, Phase I

Figure 14. Cumulative Flow Volume versus Time for the Second Porcelain Clay Test, DOE Contract No. DE-AR21-94MC30360, Phase I

Figure 15. Hydraulic Conductivity of the Second Porcelain Clay Test, DOE Contract No. DE-AR21-94MC30360, Phase I

6.3 NAPL Extraction - Purging with Water

Ground water-NAPL extraction tests were conducted on (1) well-sorted Oklahoma sand, and (2) poorly-sorted, Ajax sand to evaluate the effectiveness of AER of NAPL-contaminated soil and ground water.

6.3.1 Background

Multiphase fluid flow in a porous medium is a function of the fluid relative permeability in that porous medium and relative permeability depends on saturation history. Therefore, NAPL-extraction measurements are non-reversible and are much more time consuming than either air permeability or hydraulic conductivity measurements. Each measurement sequence is, in effect, a destructive measurement and cannot be repeated using the same sample. Consequently, current NAPL extraction results are based on much smaller data bases than either the k_a and k_w results. Evaluation of the AER technology was accomplished by comparing measurements on multiple baseline and excited samples. Given this fact, there is an immediate source of data noise from our inability to make identical multiple samples. The data analysis and interpretation must accommodate this uncertainty.

The results of the laboratory NAPL extraction measurements are better understood in light of the laboratory test apparatus (Figure 6) and the laboratory test procedures (Section 5.2.3).

The data collection and analysis approach is similar to that used for the extended simulated P&T and relative permeability measurements that are common in the petroleum industry (Salathiel, 1973). The environmental equivalent of an extended waterflood is a ground water P&T of a NAPL spill. Thus, the terms extended simulated P&T and NAPL-ground water P&T are used interchangeably herein. The samples are initially vacuum saturated with water. The sample pore volume, V_p , and porosity, ϕ , are estimated using two different techniques (1) dry sample mass, m_d , sample volume, V_T , and matrix grain density, ρ_g ; and (2) m_d , water wet mass, m_{100} , and water density, ρ_w . Comparison of these two different estimates allows a qualitative evaluation of the data quality. The hydraulic conductivity, K_h , is determined from water flow rate q_w , sample dimensions, and water pressure head, h .

The water-saturated sample is then flooded with the NAPL until no more water is produced. The initial NAPL (fractional pore) volume, V_{Ni} , and irreducible water saturation, S_{wir} , are estimated using two different techniques (1) volume of the displaced water, V_{wd} , end cap plumbing volume, V_e , and the pore volume, V_p ; and (2) mass of the water-saturated sample assembly, M_{Tw} , mass of the sample assembly at $S_w = S_{wir}$, M_{Swir} , V_e , V_p , water density, ρ_w , and NAPL density, ρ_N . Comparison of these two different estimates allows a qualitative evaluation of the data quality. The NAPL flow rate, q_N , is measured to give an estimate of NAPL permeability at S_{wir} .

The NAPL flood is followed by a simulated P&T, during which the produced volumes of water and NAPL are recorded as functions of time. The simulated P&T is conducted either under baseline or excited conditions. The NAPL relative permeability, k_{rN} , is estimated from the NAPL flow rate, q_N , and maximum NAPL flow rate, q_{Nmx} . The water relative permeability, k_{rw} , is estimated from the water flow rate, q_w , and maximum Water flow rate, q_{wmx} .

Flow and recovery of multiple-phase fluids through porous materials can be best understood in term of the relative permeabilities of the wetting and non-wetting phases k_{rw} and k_{rnw} , respectively (Acre et al., 1992; Buckley and Leverett, 1942; Cohen and Mercer, 1993; Conrad et al., 1992; Osoba et al., 1951; Rapoport and Leas, 1953; and Schuille, 1988). In its simplest form, the relative permeability of a given fluid in a multi-phase fluid filled porous medium is the ratio of its effective permeability, in the presence of one or more other immiscible fluids, to its value if it were the only fluid in the porous material. The relative permeability will vary as a function of the fractional or partial saturation of the pore space with that fluid, S_f , and will depend on the matrix wettability, the interfacial tension and viscosity contrasts between the immiscible fluids, and the capillary forces at the pore throat entries (Acre et al., 1992; Cohen and Mercer, 1993; and Mackay and Cherry, 1989). While it is possible to estimate the relative permeability from the above data using empirical correlations, it is much more accurate and straightforward to conduct NAPL and/or simulated P&T measurements and to evaluate k_{rN} and k_{rw} directly from the laboratory data.

Figure 16 is an idealized water/DNAPL relative permeability plot from Schuille (1988). The NAPL relative permeability, k_{rN} , starts out at 1.0, for very high S_N , goes through a "stretched S-shaped response" over an intermediate region of two-phase flow, and asymptotically approaches 0 for very low values of S_N . For two immiscible fluids (e.g., NAPL and water), the fractional water saturation $S_w = 1 - S_N$. Like the NAPL relative permeability relation, the water relative permeability, k_{rw} , has a similar dependence on S_w .

The saturation extremes of Figure 16 are regions of near single phase fluid flow. For S_w less than the irreducible water saturation, S_{wir} , the NAPL is the only fluid to flow. For S_N less than the residual NAPL saturation, S_{Nr} , water is the only fluid to flow. For laboratory evaluation of traditional P&T and enhanced NAPL recovery technologies, it is desirable to evaluate S_{Nr} since this will determine the maximum possible, free product NAPL recovery. Once S_{Nr} is reached, any further reduction in S_N can only be achieved as NAPL goes into aqueous solution (an extremely slow process) or by modifying the physical conditions of the aquifer. AER is one technology that may modify the physical conditions in the aquifer.

The NAPL extraction tests were short term laboratory experiments designed to evaluate the efficiency of AER for NAPL contaminated soil and ground water. This was accomplished by comparing NAPL recovery for baseline and excited conditions for the simulated P&T of NAPL saturated random pack cores initially at S_{wir} . The NAPL extraction test results yielded NAPL and water recovery as a function of time.

Figure 16. Idealized Water-DNAPL Relative Permeability Plot (taken from Schwille, 1988), DOE Contract No. DE-AR21-94MC30360, Phase I

The initial laboratory data displays were fluid (NAPL, water, and total fluid) recovery vs time. This display described the laboratory protocol but offered little insight into potential field performance. A better measure of potential field performance is to plot cumulative NAPL recovery vs cumulative simulated P&T, which was also done. This display provided a qualitative visual display of background vs excited simulated P&T efficiencies.

Three measures of NAPL recovery efficiencies were utilized to evaluate the NAPL recovery efficiency (1) estimated fractional pore volume (FPV) NAPL remaining at the end of the simulated P&T, (2) FPV NAPL recovered by Dean Stark extraction from the measurement sample after the simulated P&T, and (3) back extrapolated 100 PV simulated P&T S_{Nr} estimate using the Jones and Roszelle (1978) method.

The estimated FPV remaining at the end of the simulated P&T is obtained by mass balance calculations on the laboratory test data. The details of the calculation of this estimate are described in Appendix C.

The Dean Stark S_N is based on the NAPL recovered by Dean Stark extraction of the measurement sample after the simulated P&T tests are complete. Dean Stark extraction is a standard heat and solvent technique used to clean measurement cores prior to petrophysical measurements in the petroleum industry (Smith, et. al., 1960).

The semi-logarithmic back extrapolation provides an estimate of flood S_{Nr} for the short term laboratory tests by plotting the inverse of cumulative simulated P&T (PV^{-1}) vs remaining NAPL and back extrapolating to $PV^{-1} = 0$. Because of the short term duration (1 - 2 hr) of the simulated P&Ts and the asymptotic approach to residual saturation, S_{Nr} , this NAPL saturation condition was never really reached during the NAPL extraction tests. The current study modified this technique, using a semi-logarithmic display and back extrapolating to $PV^{-1} = 0.01$ (100 PV of simulated P&T).

The inability to prepare identical random packs resulted in samples with variations in hydraulic conductivity and relative permeability. The ultimate result of these variations was non-equal simulated P&T at constant measurement times. In addition to the cumulative NAPL recovery vs cumulative simulated P&T displays, the ratio of cumulative NAPL recovery to cumulative simulated P&T was also computed as a measure of simulated P&T efficiency.

Appendix C presents a detailed discussion of our laboratory data analysis and QA/QC process.

6.3.2 Tests in Oklahoma Sand

Two sets of NAPL-extraction tests were conducted in Oklahoma sand using canola oil, and a follow-up set of tests was run using Soltrol 220. The three sets of tests are described in the following sections.

6.3.2.1 Water - Canola Oil, Test Set No. 1

Initially, a total of 16 NAPL-extraction tests were performed using the canola oil NAPL surrogate (seven baseline and a total of nine excited tests; six tests at frequencies from 1100 to 1130 Hz, two tests at 914 and 918 Hz and one test at 1060 Hz).

Figure 17 is a compilation of the results for the six baseline and six excited tests at 1100 to 1130 Hz. This figure shows that on average there is a clear separation between the baseline and excited test results and that with excitation there was more NAPL recovered per pore volume of the simulated P&T. Figure 18 plots the mean values of S_w (with error bars) versus time for the same baseline and excited tests. There is a statistically significant difference between the mean S_w values for the baseline and excited tests, with a separation of about 8 saturation units at the conclusion of the tests. This corresponds to a 26% reduction in the final NAPL saturation in the

Oklahoma sand with acoustic excitation. Table 2 summarizes the NAPL saturation results for the different test groups, as analyzed by various methods.

Figures 19 and 20 are composite NAPL surrogate (canola oil) and water relative permeability plots for the baseline and excited tests in Figure 17, respectively. Both the excited k_{rN} and k_{rw} data are shifted significantly toward higher S_w (lower S_N) from the baseline data, indicating improved NAPL recovery with excitation. The transition regions of two-phase flow in Figures 19 and 20 are steeper than the similar portions of the generalized relative permeability plot, shown in Figure 16 because of the very high Oklahoma Sand permeability ($3.5 \times 10^{-3} < K_h < 6.8 \times 10^{-3}$ cm/sec). Low permeability sediments have more extended (in terms of saturation range) transition region than high permeability sediments. The high apparent S_{wir} and S_{Nr} values on the Oklahoma Sand relative permeability plots are due to the relatively high viscosity of the NAPL used (65 cpoise at 70° F, for canola oil).

The cumulative NAPL recovery plots (Figure 17), S_w vs. treatment time plots (Figure 18) and relative permeability plots (Figures 19 and 20) all provide some indication of residual NAPL saturation, S_{Nr} . However, the asymptotic nature of these curves makes it difficult to precisely quantify S_{Nr} . An alternative method for estimating S_{Nr} (Jones and Roszelle, 1978) is to plot S_N vs $1/(\text{pore volume simulated P\&T})$ on a semi-logarithmic grid and then extrapolate the results to a desired common time. Figure 21 shows an example of the semi-logarithmic extrapolation technique applied to a baseline experiment with canola oil.

Table 2 summarizes the back extrapolated estimates of S_{Nr} for the twelve tests in Figure 17. Semi-logarithmic back extrapolation gave a mean baseline S_{Nr} estimate of 0.263 after 100 PV of P&T, versus an excited S_{Nr} estimate of 0.183 after 100 PV. This corresponds to a 30% reduction in S_{Nr} with an excited P&T operation.

The NAPL recovery results from this study are comparable to the excited results obtained by Duhon (1964), and are slightly lower than the excited results obtained by Reddi and Challa (1994), which may be explained by the higher viscosity of the canola oil used in this study, see Section 2.4.1. It is hypothesized that acoustic excitation is helping to overcome the capillary forces and surface tension (Figure 2) which hold the NAPL within the pore space.

Fourteen of the samples in Figure 17 were sent to PTS Laboratories in Santa Fe Springs, CA for independent analyses of the residual NAPL saturation by solvent extraction (the Dean Stark technique see Smith et al. 1960, and Cohen and Mercer, 1993). The results of these analyses (Table 2 and Appendix A) found no significant difference between the values of S_{Nr} for the baseline and excited samples. Review of these data and the sample handling protocol suggested that trimming the ends from the samples prior to submitting the samples to PTS Laboratories may have introduced systematic errors into the subsequent analyses.

6.3.2.2 Water - Canola Oil, Test Set No. 2

Due to the apparent discrepancy between the test results in Figure 17 and the results of the extraction analyses (Table 2 and Appendix A), three additional baseline and three additional excited NAPL-extraction tests were conducted using canola oil. Figure 22 is a summary plot of

the NAPL recovery data as a function of water throughput for this second set of tests. Two of the excited tests show improved NAPL recovery as compared to the baseline tests. However, the third excited test (12.06.95b) was as inefficient as the least efficient baseline test (12.06.95a). Review of the 12.06.95b data sheets revealed no obvious data problems. The empirical laboratory data indicate a 6% improvement in NAPL extraction over baseline conditions.

The results from Test Sets No. 1 and No. 2 were reviewed to see if there were any systematic differences between the two data sets. The only significant finding found from this review was that the second set of baseline data (07.06.95a, 12.06.95a, 13.06.95a) had significantly higher NAPL recoveries than the earlier baseline data. Review of the recorded laboratory conditions provided no systematic reason for this.

Table 2 summarizes the back extrapolated S_{Nr} estimates for the test results in Figure 21. Semi-logarithmic back extrapolation gave a mean baseline S_{Nr} estimate of 0.170 after 100 PV of P&T, and an excited P&T S_{Nr} estimate of 0.148 after 100 PV, for a 13% reduction in S_{Nr} .

The independent, Dean-Stark extraction results (Table 2 and Appendix A) show that the three baseline samples have essentially the same residual NAPL saturations near 16%, while two of the excited samples have significantly lower residual NAPL saturations near 11 and 6%, respectively. These data correspond to 32 and 63% reductions in the residual NAPL saturation as compared to baseline conditions. Further resolution of questions raised by Test Sets No. 1 and No. 2 will require additional testing that is beyond the scope of this Phase I investigation.

6.3.2.3 Water - Soltrol 220

Canola oil was chosen as a NAPL surrogate because, as a non-toxic, non-hazardous vegetable oil, it posed no special handling and/or disposal problems. While its viscosity of 68 cp at 68°F is considerably lower than many crude oils and refinery residuals, its viscosity is considerably higher than most fuel and chlorinated hydrocarbons. It has a viscosity that is that is comparable to creosote and coal tar compounds (Cohen and Mercer, 1993) and higher API gravity crude oils.

Questions were raised as to whether canola oil may behave significantly differently than fuel or chlorinated hydrocarbons due to either its greater viscosity or its surface chemistry. To address these questions, additional NAPL-surrogate extraction tests in Oklahoma sand using Soltrol 220 were conducted. Soltrol 220 is a relatively non-hazardous, non-toxic refined petroleum hydrocarbon that has been used in previous studies (Illangasekare et al., 1994). Wilson et al. (1990) used a very comparable fluid, Soltrol 130. The physical properties of Soltrol 220 are given in Section 4.3.

Figure 17. Oklahoma Sand - NAPL Surrogate (Canola Oil) Pump and Treat Simulation 1, DOE Contract No. DE-AR21-94MC30360, Phase I

Figure 18. Oklahoma Sand - Partial Water Saturation Versus Time, DOE Contract No. DE-AR21-94MC30360, Phase I

Figure 19. Oklahoma Sand - Water - NAPL Surrogate Relative Permeability, DOE Contract No. DE-AR21-94MC30360, Phase I

Figure 20. Oklahoma Sand NAPL - Surrogate - Water Relative Permeability, DOE Contract No. DE-AR21-94MC30360, Phase I

A total of ten NAPL-extraction tests were conducted using Soltrol 220 (five baseline tests, two excited sets at a single frequency of 1100 or 1130 Hz, and three excited tests with continuous excitation using a sequence of frequencies). Figure 23 is a summary plot of the NAPL recovery data for the ten tests as a function of water throughput. There is only a small difference between the baseline and excited results, but the average excited data show about 19% improvement over average baseline conditions (Table 2). Semi-logarithmic back extrapolation gave a mean baseline S_{Nr} estimate of 0.060 after 100 PV of P&T, and an excited S_{Nr} estimate of 0.043 after 100 PV, for a 41% improvement in residual NAPL saturation over baseline (Table 2).

There is also a discrepancy between the above results from the NAPL-extraction tests and the Dean-Stark extraction analyses (see Table 2). The Dean-Stark analyses found a mean baseline S_{Nr} estimate of 0.196, and a mean excited S_{Nr} estimate of 0.149, for a 24% reduction in S_{Nr} . Resolution of this discrepancy would require additional testing that is beyond the scope of this Phase I investigation.

Acoustically Enhanced Remediation of Contaminated Soil and Ground Water

Table 2. NAPL Saturations by Various Methods for the Various Tests Conducted, DOE Contract No. DE-AR21-94MC30360, Phase I.

Tests Conducted	Laboratory Tests			Semi-Logarithmic Extrapolation ²			Dean Stark Extraction ⁵		
	Measured Residual NAPL Saturation ^{1,3}			NAPL Saturation ³ after 100 PV ⁴ Pump & Treat					
	Baseline (FPV ⁹)	Excited (FPV)	% Improvement ¹⁰	Baseline (FPV)	Excited (FPV)	% Improvement	Baseline (FPV)	Excited (FPV)	% Improvement
Oklahoma Sand/Canola Oil/Water, Test No. 1	0.2746	0.2042	25.64	0.2633	0.1833	30.38	0.1780	0.1740	2.25
Oklahoma Sand/Canola Oil/Water, Test No. 2	0.2015	0.1888	6.30	0.17	0.1483	12.76	0.1623	0.1109	31.67
Oklahoma Sand/Soltrol 220/Water	0.0656	0.0529	19.36	0.068	0.04	41.18	0.1963	0.1490	24.10
Ajax Sand/Canola Oil/Water ⁶	0.1766	0.1732	1.93	0.1521	0.1467	3.55			
Ajax Sand/Canola Oil/Water ⁷	0.1766	0.1502	14.95	0.1521	0.0933	38.66			
Ajax Sand/Canola Oil/Water(31.05.95 test) ⁸	0.1766	0.1264	28.43	0.1521	0.04	73.70			

1 Empirical laboratory results for a simulated pump and treat of a NAPL-contaminated ground water

2 Using Jones and Roszelle (1978) Method

3 Average values from multiple tests

4 Pore volume throughput of water

5 Modified ASTM solvent extraction method a standard laboratory technique for measuring immiscible fluid saturation. in the petroleum industry. Measured NAPL content was divided by the measured porosity of the sample to obtain FPV (see footnote 9).

6 Tests conducted at a single frequency

7 Tests conducted with a sequence of six frequencies

8 Tests conducted with a sequence of eight frequencies and with an increased power level

9 Fractional pore volume occupied by NAPL

10 Acoustically excited NAPL recovery relative to baseline (non-excited) recovery

Figure 21. Semi-Logarithmic Extrapolation Technique Example, DOE Contract No. DE-AR21-94MC30360, Phase I

Figure 22. Oklahoma Sand - NAPL Surrogate (Canola Oil) Pump and Treat Laboratory Test No. 2, Baseline Data Shown in Gray and Excited Data Shown in Black, DOE Contract No. DE-AR21-94MC30360, Phase I

Figure 23. Oklahoma Sand - NAPL Surrogate (Soltrol 220) Pump and Treat Laboratory, Baseline Data Shown in Gray and Excited Data Shown in Black, DOE Contract No. DE-AR21-94MC30360, Phase I

6.3.3 Tests In Ajax Sand

A total of 16 NAPL-extraction tests were conducted in Ajax sand using canola oil (seven baseline tests, six excited tests at frequencies between 775 Hz and 10.0 kHz, and three excited tests using a sequence of frequencies). Figure 24 is a summary plot of the NAPL recovery data as a function of the water throughput. The excited results using a single excitation frequency did not separate from the baseline data. This null result is attributed to the greater variability between multiple Ajax sand samples, the intrinsic heterogeneity of the samples' physical and hydraulic properties, and possible effects of fingering.

Two of the sequential excitation tests were among the most effective of the NAPL recovery results, but were still not separated from the baseline data. The third sequentially excited data set (31.05.95) separated from both the baseline data sets and the other excited data sets by approximately 5%. These data was obtained with a very high power level and using a sequence of eight frequencies, instead of a single frequency or only four frequencies for the other sequential excitation tests.

The empirical laboratory test excited results show about a 2% improvement over baseline conditions for the single frequency excitation tests, a 19% improvement over baseline using a sequence of six frequencies, and a 28% improvement over baseline when the power level was increased and a sequence of eight frequencies were used (Table 2). The semi-logarithmic back extrapolation (Table 2) provides similar results. For the same conditions just described, the semi-logarithmic extrapolation indicates that the excited NAPL saturations after a 100 PV simulated P&T are 4, 39, and 74%. Dean Stark extractions were not conducted on these samples.

6.3.4 Summary

Acoustic excitation enhanced the recovery of both canola oil and Soltrol 220 from samples of Oklahoma sand. The empirical laboratory test results show an increased NAPL recovery with Soltrol 220 that was not as dramatic as with the canola oil, reflecting that a standard simulated P&T (i.e., unassisted P&T) is more effective when the NAPL/water viscosity ratio is closer to 1.0 (Craig, 1971).

Acoustic excitation did not appear to enhance the recovery of canola oil recovery from samples of Ajax sand until a very high acoustic power level was used with a sequence of eight excitation frequencies. To a considerable extent, the null results with Ajax sand probably reflect the difficulty in obtaining similar samples of the poorly sorted material. The increased NAPL recovery with high-amplitude, multiple-frequency sequential excitation may offer a key to successful field application of this technology.

The apparent disagreement between the Dean-Stark analyses and the empirical laboratory data remains a paradox. Its resolution is beyond the scope of this investigation.

Figure 24. Ajax Sand - NAPL Surrogate (Canola Oil) Pump and Treat Laboratory, Baseline Data Shown in Gray and Excited Data Shown in Black, DOE Contract No. DE-AR21-94MC30360, Phase I

6.4 Laboratory Strain Amplitudes

The laboratory experiments show that acoustic excitation can increase the hydraulic conductivity of low permeability soils and improve the effectiveness and efficiency of P&T operations. An obvious question is whether the acoustical levels required to develop the beneficial effects can be generated in-situ with available acoustic source technologies (discussed in Section 7.1). To address this issue, the strain amplitudes developed in the laboratory experiments were estimated and compared them with available source technologies.

For the strain amplitude analyses, a frequency sweep was applied to a sample of Oklahoma sand at 30 psi and measured the local acceleration and dynamic pressure at both ends of the sample. The accelerometer readings were converted to the average strain over the length of the sample by taking the difference between the accelerometer readings, dividing by the angular frequency, and converting the resulting voltage to a displacement based on the accelerometer calibrations. In a similar manner, readings from the dynamic pressure transducers were used to estimate the average stress and thus the strain amplitude, assuming that the sand has a Young's modulus of 0.4 GPa (figure 8 in Stevens, 1966).

These results of analyses show that the beneficial effects of acoustic excitation are observed in experiments where the strain amplitudes are between approximately 10^{-5} and 10^{-4} . As discussed in Section 7.2, these strain amplitudes are within the range that can be developed in-situ with existing source technology, especially if the sources are deployed using a phased array. During field tests, the induced strain amplitudes will be monitored using an array of geophones positioned in shallow boreholes.

6.5 Frequency Sweep Analyses of Non-Linear Soil Behavior

Frequency sweep analyses were performed on samples of Oklahoma and Ajax sand to identify frequency ranges where nonlinear soil behavior is occurring within the samples. These regions of nonlinear soil behavior are of interest since it has been proposed that nonlinearities can generate high strain levels in the soil material, and thus lead to more efficient remediation.

The frequency sweep used for these analyses was a linear scan from 100 Hz to 11 kHz in 2.5 seconds. The outputs from the four dynamic sensors (two accelerometers and two pressure transducers) were recorded in pairs through a Stanford Research Systems model SR650 dual-channel Bessel filter to prevent aliasing. The filtered signals were recorded using a LabVIEW virtual instrument with a sampling rate of 30 kHz, saved on disk and later analyzed using MatLab routines.

Figure 25 shows example data from a frequency sweep analysis on a sample of dry Oklahoma sand. The data are presented as a waterfall plot that graphs the Fourier transform of the difference between the top and bottom accelerometers as a function of frequency. The main, bright line that starts at the origin and proceeds to the top of the graph, represents the fundamental driving frequency applied to the sample.

Figure 25. Plot of Time Versus Frequency for Dry Oklahoma Sand, DOE Contract No. DE-AR21-94MC30360, Phase I

The two less bright lines below the fundamental, which also start at the origin, are the second and third harmonic frequency components. There are several bright spots along these lines that indicate areas of strong nonlinear soil behavior in the frequency ranges of 1 to 2 kHz, 3 to 5 kHz and 6 to 7 kHz. Below the third harmonic line, there are still weaker line segments that are the fourth and fifth harmonics of the fundamental frequency. The existence of these harmonics provides direct evidence of nonlinear soil behavior, and these types of plots can be used to identify frequency ranges that may be optimal for soil remediation. This approach was used during optimization testing to help identify frequencies with interesting nonlinear soil behavior.

6.6 Optimization Testing

The optimization testing portion of the testing program primarily pursued two encouraging approaches (1) increasing the acoustic power input to the sample, and (2) subjecting the sample to a sequence of frequencies that were chosen because they showed strong resonances or interesting nonlinear soil behavior. The key results from the optimization testing are:

1. Increasing the acoustic power during an Ajax sand water permeability test produced large amounts of muddy water (Section 6.2.2) but progressively plugged the sample. During the hydraulic conductivity testing of silt fines were also mobilized as they were in the case for Ajax sand, but the resulting permeability decrease in the silt sample was apparently reversed by subjecting the soil sample to periodic excitation. This approach may be a means to reverse the permeability reduction effect. Additional testing is needed to determine whether there are conditions that will maximize fines production without significantly reducing the permeability.
2. Increasing the acoustic power to the maximum level possible during the second clay water permeability test increased the hydraulic conductivity of the clay by a factor of 4 (Section 6.2.3).
3. Increasing the acoustic power during the Ajax sand NAPL-extraction testing was one of the factors that resulted in a clear, discernible separation between baseline and excited results.
4. Subjecting the Ajax sand to a sequence of eight frequencies may have been one of the factors that resulted in a clear, discernible separation between the baseline and excited results. It has also been reported in Chemical Engineering (see Section 2.4.1) that multiple frequency excitation can increase the effectiveness of AER.

7. ACOUSTIC REMEDIATION TECHNOLOGY PERFORMANCE

7.1 Assessment of Acoustical Sources

The results of our laboratory studies show that the beneficial effects of acoustic excitation are observed in experiments where the strain amplitudes are on the order of approximately 10^{-5} to 10^{-4} . It will be shown in the next section that by using a number of low to medium power sources in a phased array concept, it will be possible to generate similar beneficial strain levels in-situ for acoustically enhanced remediation.

There are a number of available acoustic sources with suitable output power levels. Ultimately, the selection will be made based on a number of factors including the frequency range, ease of frequency and phase control, output power levels, size (especially for downhole sources), cost, and other operational considerations. Table 3 summarizes the different source categories and some of their relevant characteristics.

7.2 Field Deployment Strategy

While a range of technologies exist for radiating large amounts of acoustical power into the subsurface (see Section 7.1), the complex geometry and geohydrology of contaminant plumes make large volume acoustical excitation desirable. One might imagine that it would be necessary to install a large number of excitation wells throughout the plume. However, due to the high cost of installing wells, it is preferable to instead use a distributed array of sources. With a distributed array of sources, and:

1. by controlling the phase of the sources, high-power acoustical energy can be directed to various parts of the plume; and
2. by modifying the phase of the sources, high intensity acoustic energy can be focused and swept throughout the contaminant volume.

In the following discussion, comparisons are provided between the AEFs that can be produced using a single source, with multiple sources that combine incoherently, and various phased array approaches which combine coherently.

In evaluating the effectiveness of acoustical source technologies, it is necessary to have an *a priori* indication of what levels of acoustical field intensity or strain amplitude are needed to have a significant effect on the remediation process. The results of our laboratory experiments indicate (see Section 6.4) that the beneficial effects of acoustic excitation are observed in experiments where the strain amplitudes are between approximately 10^{-5} and 10^{-4} .

Table 3. Available and Research Acoustic Source Technologies, DOE Contract No. DE-AR21-94MC30360, Phase I

SOURCE TECHNOLOGY	DESCRIPTION	AVERAGE RADIATED POWER LEVELS	FREQUENCY BAND (kHz)	DEMONSTRATED IN SEISMIC APPLICATIONS	COST (k\$/kW)	BENEFITS & ISSUES
Compressed Air Driven	Piston and orbiter configurations	Kilowatt	0.01 to 10	Yes	1-10	<ul style="list-style-type: none"> • Simple technology • Low frequency roll off • Complicated adaptation to phased arrays
Hydraulically Driven	Piston and orbiter configurations	Kilowatt to tens of kilowatts	0.01 to 10	Yes	1-10	<ul style="list-style-type: none"> • Low frequency roll off • Complicated adaptation to phased arrays
Electro-mechanical	Solenoid vibrators and rotary orbiter configurations	Kilowatts	0.01 to 10	Yes	1-10	<ul style="list-style-type: none"> • Low frequency roll off • Adaptable to phased array operation
Piezoelectric Transducers	Oscillator based on piezoelectric contractions	Tens of watts per transducer, large arrays possible	0.1 to 1000	Yes	10-100	<ul style="list-style-type: none"> • Strong low frequency roll off • Readily adaptable to phased array operation
Flexensional Transducers	Electrically-driven, flexural mode piezoelectric oscillators	Hundreds of kilowatts	0.1 to 5	No	10	<ul style="list-style-type: none"> • Adaptable to phased array applications • Efficient coupling to soil • May potentially exploit beneficial nonlinear vibration effects
Large Column Flame Resonator	Thermoacoustic oscillations, combustion instability within remediation well column.	Potentially very large (megawatts)	0.01 to 5	No	0.1 to 10, Very low, can potentially burn remediation by-product vapors to produce oscillations	<ul style="list-style-type: none"> • High frequency roll off. • Complications from low efficiency of coupling energy from air column to soil. • Developmental technology
Combustion Driven Pulser	Low-frequency source, repetitive combustion wave detonation events.	Potentially very large (megawatts)	0.1 to 0.1	No	10	<ul style="list-style-type: none"> • Can draw from seismic applications of explosives. • High frequency operation is limited. • Developmental technology
Water Siren	Oscillations generated by periodically interrupted water jet.	Potentially very large (megawatts)	0.01 to 0.1	No	Unknown	<ul style="list-style-type: none"> • Water filled well minimizes impedance losses. • Developmental technology.

Figure 26 shows an engineering trade space of acoustical frequency and intensity in comparison with the test regime of our laboratory experiments. Several basic engineering factors restrict the portion of this trade space where AER should be feasible.

1. strain levels that are too low, roughly below 10^{-6} , will probably not induce significant variations in permeability, relative permeability, and other controlling remediation parameters;
2. strain levels that are too high, roughly above 10^{-3} , can lead to soil liquefaction that should be avoided at most remediation sites;
3. frequencies that are too high, roughly above $3 \cdot 10^4$ Hz, will not propagate very far in the subsurface; and
4. sources above 1 MW will not be available in the near future, and ultimately are probably unaffordable.

This highly simplified analysis leaves a roughly triangular region of likely AER operation.

Figure 26. Trade Space of Strain and Field Intensity Compared Against Experimental Test Regime, DOE Contract No. DE-AR21-94MC30360, Phase I

The key engineering question is what levels of acoustic intensity and strain amplitude can be generated in-situ using realistic sources. In the following sections, analyzed are the acoustic intensities that can be developed using different source geometries, and will describe the engineering trade-offs that will affect the implementation of AER. In these analysis, considered are only sources operating at the ground surface. There will be analogous results for buried sources, and optimal beam forming/phase steering in 3-dimensions may require multiple sources operating in a well.

Figure 27 shows a variable gray scale for interpreting the results in the following sections. The gray scale covers a range that runs from 1000 W/m^2 to 0.1 W/m^2 , or strains of 10^{-5} to 10^{-7} , respectively. Based on our laboratory results, it appears that acoustic intensities or strain levels near the upper end of this range are needed for implementation of AER.

Figure 27. Intensity Versus Color Coding Used in Source Intensity Analyses, DOE Contract No. DE-AR21-94MC30360, Phase I

7.2.1 Single Source

Figure 28 shows the acoustical intensity as a function of location along the air-ground interface with a single 10 kW source located at the center of the grid (50m, 50m location). In this case the intensity drops away as the inverse of the square of distance from the source, so at 50 m from the source, the intensity is less than 1 W/m^2 , a level well below that required to cause significant strain in the soil.

With a single source, the intensity at a given range is proportional to the power of the device. Thus, increasing the source to a megawatt would still only lead to an intensity of about 600 W/m^2 at 50 meters, or a strain on the order of 10^{-6} . A single source may be useful in the case of smaller contaminant treatment zones. For example, at 5 m from a 10 kW source, the intensity would be approximately 600 W/m^2 , which may be viable for the treatment at some site conditions.

Figure 28. Intensity Versus Location for a Single Source, DOE Contract No. DE-AR21-94MC30360, Phase I

7.2.2 Broadband Sources

With broadband wave sources, (i.e. multi-frequency) there are incoherent summation of intensities. This is analogous to the summation of light intensities from a set of light bulbs where there is no apparent interference between the sources. Similar effects occur with multiple acoustical sources if they drift away from their primary drive frequency or if there is not strict phase control. Figures 29 to 31 show the total intensity versus position for 2, 4, and 8 multielement arrays, respectively, centered about the (x,y) location of (50 m, 50 m). Each acoustical source has a power output of 10 kW.

Even with the 8 element array (Figure 31) the intensity at the center of the array is only about 10 W/m^2 which is insufficient for AER. The intensity will scale linearly with the number of acoustical sources, and as the inverse square of the spatial dimension of the array. While multiple sources can cover much larger volumes than a single source, it quickly becomes very expensive using multiple, incoherent sources to generate the required acoustic intensities over an extended volume.

Figure 29. Intensity Versus Location Produced by Two Sources that Incoherently Sum, DOE Contact No. DE-AR21-94MC30360, Phase I

Figure 30. Intensity Versus Location Produced by Four Sources that Incoherently Sum, DOE Contact No. DE-AR21-94MC30360, Phase I

Figure 31. Intensity Versus Location Produced by Eight Sources that Incoherently Sum, DOE Contact No. DE-AR21-94MC30360, Phase I

7.2.3 Phased Arrays

The basic concept of a phased array is that, if one has multiple sources with identical frequencies and can adjust their phases, then the constructive interference can be controlled at a given location to give a region of high acoustic intensity. The phase control of the sinusoidal waveforms from multiple sources is what constitutes the "phased array".

Illustrated below are the AEFs that can be produced using various phased array geometries. Considered are both the "circular" (described above) and linear arrays. Note that based on the contaminant spill geometry, altering the phased array layout can assist in maximizing acoustical intensities within the geohydrological volume of interest. With sufficient characterization of the contaminant plume geohydrological setting, nonlinear array analyses can be used to select locations for the elements of multielement acoustical array.

Note that the analysis presented herein is restricted to the case of acoustical sources at the ground-air interface. Extending the distribution of these sources to subsurface wellbore locations will further enhance the ability of phase control to produce intensities beyond those shown here. As part of the Phase II program, these additional geometries will be examined, plus the effects of acoustic scattering and soil heterogeneities, so that the tools required to assess AER for a given treatment site will exist.

7.2.3.1 Localized Intensification With Phased Arrays

By maintaining the source frequencies at a single value and controlling the phases of the individual sources, constructive interference effects can lead to locally enhanced acoustical excitation fields. In this section, presented are the results for the local AEF enhancement that can be produced by a phased array. Figures 32 to 34 show the results for a phased array of 10 kW sources with a total of 2, 4, and 8 elements that coherently sum, respectively. The main effect of a phased array is that the intensity of the acoustical field is now proportional to the square of the number of elements. For the case of two elements, the increase in intensity at the center of the array is a factor of two higher than in Figure 29 in Section 7.2.2, even though the sources have the same power output. With eight elements, the increase in intensity is even greater with the center intensity being a factor eight times higher than in Figure 31, or 80 W/m^2 .

This is a fairly significant acoustical effect. Clearly, the optimal strategy for field deployment of AER is to increase the number of sources and to use a phased array approach to develop higher localized intensities. In the case of downhole sources, there will of course be trade-offs associated with the cost of installing additional wells. Therefore, an alternative approach is to create a three-dimensional array by installing multiple sources in a smaller number of wells. With a properly designed array, the acoustic intensity will still increase as the square of the number of sources. For graphs, with eight wells and eight sources per well, an effective acoustical excitation factor of 4,000 on a single source can be achieved.

Figure 32. Intensity Versus Location Produced by Two Sources that Coherently Sum for Maximum at Center of Array, DOE Contract No. DE-AR21-94MC30360, Phase I

Figure 33. Intensity Versus Location Produced by Four Sources that Coherently Sum for Maximum at Center of Array, DOE Contract No. DE-AR21-94MC30360, Phase I

Figure 34. Intensity Versus Location Produced by Eight Sources that Coherently Sum for Maximum at Center of Array, DOE Contract No. DE-AR21-94MC30360, Phase I

7.2.3.2 Full Volume Sweeps

By varying the relative phases of the different sources, the region of maximum intensification can be swept across the treatment volume. Figures 35 to 37 show the maximum intensity that can be temporarily generated at any location by sweeping the zone of constructive interference across the volume. For example, with an eight element array (Figure 37), intensities approaching 100 W/m^2 can be established across most of the volume. This approach would be particularly useful if, as in the second porcelain clay experiment (Figure 14), the beneficial effects of acoustic excitation persist for some time after excitation.

To further illustrate the importance of increasing the number of sources, Figure 38 shows the maximum intensity that can be temporarily generated at any location with 32 sources each having an intensity of only 1 kW . A fairly high intensity over 100 W/m^2 can be produced across the central volume. Increasing the power of each source would further increase the local intensity. For example, with 10 kW sources, acoustic intensities over 1 kW/m^2 could be generated across the central volume.

Figure 35. Maximum Intensity Versus Location Produced by Sweeping Two Sources Across the Volume, DOE Contract No. DE-AR21-94MC30360, Phase I

Figure 36. Maximum Intensity Versus Location Produced by Sweeping Four Sources Across the Volume, DOE Contract No. DE-AR21-94MC30360, Phase I

Figure 37. Maximum Intensity Versus Location Produced by Sweeping Eight Sources Across the Volume, DOE Contract No. DE-AR21-94MC30360, Phase I

Figure 38. Maximum Intensity Versus Location from an Array of 32 Phase Controlled Sources of 1 kW Intensity Each, DOE Contract No. DE-AR21-94MC30360, Phase I

7.2.3.3 Other Array Geometries

Other array geometries can be used to enhance the maximum acoustical field based on the shape of the contaminant plume and the geohydrological setting. Figure 39 shows the effect of adding another source at the center of an eight element array in Figure 37. The added center source increases the intensity in the ring-shaped region between the nine sources by about a factor of two.

Figure 40 shows the maximum intensity that can be generated at each location using a linear array of eight sources. The acoustic intensity along the center of the array is increased by a factor of two, but the fall off in the transverse direction is faster than with either the eight- or nine-element circular array (Figure 37 or 39). A linear or curvilinear array may work better at certain locations depending on the shape of subsurface contamination and the geohydrology.

This above results suggest that for any given remediation site there may be favorable source array geometries that will not require a large number of wells for source emplacement. The source optimization process can be accomplished using some nonlinear analysis techniques, and these techniques will likely be developed as part of the Phase II program.

Figure 39. Maximum Intensity Versus Location in an Eight Element Circular Array Plus on Additional Element in the Center, DOE Contract No. DE-AR21-94MC30360, Phase I

Figure 40. Maximum Intensity Versus Location by a Linearly Spaced Phased Array, DOE Contract No. DE-AR21-94MC30360, Phase I

7.2.3.4 Summary

Phased arrays provide flexibility in the production of high intensity acoustical fields in a given treatment area. Phased arrays can be much more effective than using single sources or multiple sources that are incoherently combined, since with phased arrays it is possible to control the localized intensification of acoustic energy. With phased arrays, the maximum local intensity is:

1. proportional to the power of the individual sources;
2. proportional to the square of the number of sources; and
3. inversely proportional to the spatial dimension of the array.

It follows directly from the first two relations that, in general, it will be better to divide the total source effort into a larger number of relatively low power sources, rather than using a small number of high power sources.

The ability to direct the acoustic energy depends on the spatial dimensionality of the source. For full three-dimension control, it will be necessary to layout the sources in a three-dimensional configuration. The challenge will be to identify favorable source array configurations, depending on the shape of the contaminant plume and the geohydrological setting, that will not require a large number of wells for source emplacement.

7.3 Contaminant Modeling

Both the Oklahoma sand and porcelain clay Phase I test data were used to quantify the contaminant flow and transport benefits of AER technology. This was accomplished by using a numerical model for the Oklahoma sand and an analytical expression for the porcelain clay. Both approaches are described below.

7.3.1 Oklahoma Sand

A public domain computer model, NUFT (Nonisothermal Unsaturated-Saturated Flow and Transport), was used to simulate P&T remediation of a NAPL spill in ground water using the baseline and excited NAPL recovery rates measured in Oklahoma sand. The model used a single pumping well to simulate NAPL recovery, and it was calibrated using soil properties derived from the Phase I laboratory data. The model results describe the advantages of AER technology.

NUFT, a numerical multiphase, multicomponent, multidimensional model (Nitao, 1995), is based on conceptual and mathematical models of multiphase flow and contaminant transport summarized by Bear and Nitao (1993). It was designed to simulate two- and three-dimensional multiphase (air, water or NAPL) flow and contaminant transport in the vadose zone and groundwater. Applications include the simulation of remediation systems such as soil vapor extraction systems, aqueous ground water P&T systems, NAPL ground water P&T systems, wastewater injection, and steam stripping.

The NUFT code uses a finite-difference discretization scheme using either Cartesian or cylindrical grid systems in one-, two-, or three-dimensions. Boundary conditions and source or sink terms can be specified as functions of time to simulate time-dependent phenomena such as intermittent pumping and cyclical recharge patterns. NUFT was verified and benchmarked against a number of analytical and numerical models by Lee et al. (1993). In general, excellent agreement was found between NUFT results and the benchmark solutions.

7.3.1.1 Model Assumptions and Calibration

Based on the available data and the scope of this investigation, the following assumptions were made for this modeling effort:

1. one- and two-dimensional computational grids were used for the model calibration and field simulations, respectively;
2. ground water flow is fully-saturated, horizontal, neglecting any vertical flow components;
3. soil is homogeneous and isotropic;
4. isothermal conditions at 73°F exist;

5. advective movement of water and NAPL was simulated using the Bucket-Leverett assumption³ (Bear and Bachmat, 1991); and
6. negligible dissolution of the NAPL in water occurs.

The model was calibrated using the two data sets of relative permeability measurements for the canola oil - water and Soltrol 220 - water systems in Oklahoma sand. The simulated NAPL saturations and recovered NAPL pore volumes were compared to measured values to evaluate the quality of the model simulations.

Figure 41 shows the model domain simulation of the laboratory measurements on a cylinder with a diameter of 1.5 in (3.81 cm) and a length of 3.5 in (8.89 cm). The cylinder was divided vertically into 20 model cells with a uniform thickness of 0.175 in (0.445 cm). The model domain was filled with a homogeneous, isotropic porous medium with 7 darcy permeability and a porosity of 0.32. These are the average physical properties of Oklahoma sand measured during the laboratory tests. The simulator model baseline and excited relative permeabilities for each phase as a function of phase saturation were those measured in the laboratory tests. The only differences between the baseline and excited simulator models are the use of different relative permeability - phase saturation relationships. The viscosities and densities of canola oil, Soltrol 220 and water were taken as 65 cp and 0.918 g/cm³, 4.83 cp and 0.789 g/cm³, and 1 cp and 1.00 g/cm³, respectively. Water was allowed to enter the model domain at the bottom, with water and NAPL outflow at the top. A constant pressure differential of 1.5E3 Pa was specified between water inflow and outflow to simulate the pressure head (150 cm H₂O) that was used in the laboratory tests.

Figure 42 shows the NAPL saturation profile in the simulated 1-D laboratory test cell for the canola oil - water system after 60 minutes. The model results show baseline residual NAPL saturations along the length of the laboratory test cell that range between 27% and 32%, while with excitation the residual saturations decrease to values between 22% and 27%. The average residual NAPL saturations from the empirical laboratory baseline and excited tests are 0.27 and 0.21, respectively (Table 2). The model results are in general agreement with the average results from laboratory tests.

³ This assumption neglects the effects of capillary pressure changes when they are much smaller than the changes in hydrostatic pressure in the model domain.

Figure 41. Laboratory Test Cell Model Domain, DOE Contract No. DE-AR21-94MC30360 Phase I

Figure 42. Oklahoma Sand - NAPL Surrogate (Canola Oil) - Water Model of Laboratory Results, DOE Contract No. DE-AR21-94MC30360, Phase I

Figure 43 shows the simulated cumulative removal of NAPL versus the throughput of water for the Canola oil - water system. The baseline NAPL recovery plateaus at about 58% of the total pore volume, while the excited NAPL recovery reaches an asymptotic level at about 63% of the pore volume. The shape of the recovery curve and the difference between the baseline and excited conditions correspond well with the Phase I laboratory results, but the simulated NAPL recoveries are lower than those measured in our laboratory tests. Possible refinements in the model to better match the laboratory results are beyond the scope of this contract.

Figure 44 shows the NAPL saturation profile in the simulated laboratory test cell for the Soltrol 220 - water system after 10 minutes. The model results show baseline residual NAPL saturations along the length of the laboratory test cell that range between 10% and 9%, while with excitation the residual saturations decrease to values between 5% and 4%. The model results approximate the values measured in the laboratory tests and estimated by semi-logarithmic back extrapolation (see Table 2).

Figure 45 shows the simulated cumulative removal of NAPL versus the throughput of water for the Soltrol 220 - water system. The baseline NAPL recovery plateaus at about 65% of the total pore volume, while the excited NAPL recovery reaches an asymptotic level at about 67% of the total pore volume. The predicted 2% difference between baseline and excited conditions is close to the values measured in the laboratory tests and estimated by semi-logarithmic back extrapolation (see Table 2). Based on these comparisons of laboratory and modeling results, it is concluded that the simulation adequately represents the physical properties and processes of the laboratory experiment. These soil properties are used in the following section to simulate a field-scale remediation.

7.3.1.2 The Remediation Process

A 2-dimensional, cylindrical model domain was used to simulate NAPL recovery. Only the water and NAPL phases were modeled, air flow was ignored. Accordingly, the simulated aquifer was idealized as a confined aquifer 16 m below the piezometric surface. Because the model was calibrated using laboratory data, the initial or first generation simulations attempted to duplicate laboratory fluid flow conditions for field dimensions. These results are unrealistic but they do represent an extreme case (for both baseline and excited conditions) and were used as a bridge between laboratory and field flow conditions. In a second generation of simulator models, more realistic field conditions were used.

Figure 43. Stimulated Cumulative NAPL Surrogate (Canola Oil) Recovery, DOE Contract No. DE-AR21-94MC30360, Phase I

Figure 44. Oklahoma Sand - NAPL Surrogate (Soltrol 220) - Water Model of Laboratory Results, DOE Contract No. DE-AR21-94MC30360, Phase I

Figure 45. Stimulated Cumulative NAPL Surrogate (Soltrol 220) Recovery, DOE Contract No. DE-AR21-94MC30360, Phase I

The first generation simulations were an analogue of a confined aquifer 16 m below the piezometric surface to match the Phase I laboratory test conditions for confining pressure and liquid saturation conditions. The computational grid was comprised of 20 horizontal cylinder-shells and 10 vertical layers. Grid spacing was 0.5 m (1.64 ft) both horizontally and vertically. This resulted in a cylinder with a diameter of 20 m (65.62 ft) and a length of 5 m (16.4 ft). Most of the model domain was initially saturated with water. To simulate aquifer contamination, a 5 m (16.4 ft) wide disc-shaped accumulation of NAPL at an initial saturation of 60% was centered around a single remediation well placed along the cylinder axis and screened over the entire thickness of the aquifer. To ensure horizontal (1-D, radial), flow, the pressure difference between the remediation well and the outer boundary of the model domain was set to 1.569E5 Pa. This represents the maximum head difference of 16 m (52.49 ft) that can be obtained using the dimensions described above. Both the pressure difference and the background pressure by far exceed expected values for average field applications, but were chosen to approximate the laboratory conditions under which the model was calibrated. To evaluate the sensitivity of the model results to the pressure gradient, a smaller drawdown of 5 m was applied in the second simulation for this first generation case.

Using the relationship between NAPL saturation and relative permeability as determined in the Phase I laboratory tests, NAPL remediation was simulated for a maximum duration of 1080 days for canola oil and 100 days for Soltrol 220. The model results in Figure 47 and Figure 48 show the cumulative NAPL recovery expressed as a percentage of the initial NAPL in-place. For canola oil and an applied head difference of 16 m (52.49 ft), acoustic excitation increased NAPL recovery from 40% to 58% (Figure 47). For Soltrol 220, ground water - NAPL remediation under excited conditions yields an increase in NAPL recovery from 75% to 90% (Figure 48).

Depending on the viscosity of the NAPL and the time frame of the simulation, the system proved to be sensitive to the hydraulic gradient created by the remediation well. For canola oil (viscosity = 65 cp), lowering the applied drawdown to 5 m reduced NAPL recovery by approximately 5% after 1000 days (Figure 47). For Soltrol 220 (viscosity = 4.83 cp), the drawdown dependent decrease in NAPL recovery was approximately 3% for the baseline and 1% for the excited conditions after 100 days of P&T. However, because the maximum possible recovery rate for a NAPL is determined by its residual saturation, it can be expected that with increasing the duration of P&T, recovery rates will be more independent of the applied drawdown.

In the second generation simulator modeling, more realistic field conditions were evaluated. The horizontal dimension of the computational grid was enlarged to a total diameter of 100 m (Figure 46). Between the remediation well in the center of the cylinder and the outer model boundary, a total head difference of 2 m (6.6 ft) was specified. All other model parameters were identical to those used in the first generation simulations described above.

The second generation model results in Figure 49 and Figure 50 show the cumulative NAPL recovery for canola oil and Soltrol 220 under quasi-field conditions. Note that the total percentage of NAPL recovered is almost the same as for the previous simulations under laboratory conditions. However, the time frame of the recovery changes. For canola oil, time to maximum recovery increased by a factor of 3, 1000 days to 3000 days. For the less viscous Soltrol 220, the effect is much less pronounced.

Figure 46. Simulated NAPL - Ground Water Domain in Modeling the Effects of Acoustically Enhanced Remediation, DOE Contract No. DE-AR21-94MC30360, Phase I

Figure 47. NUFT Modeling Results of Oklahoma Sand - Water - NAPL Surrogate (Canola Oil) System, DOE Contract No. DE-AR21-94MC30360, Phase I

Figure 48. NUFT Modeling Results of Oklahoma Sand - Water - NAPL Surrogate (Soltrol 220), DOE Contract No. DE-AR21-94MC30360, Phase I

Figure 49. NUFT Modeling Results of Oklahoma Sand - Water - NAPL Surrogate (Canola Oil) System, (Generation 2 - field conditions), DOE Contract No. DE-AR21-94MC30360, Phase I

Figure 50. NUFT Modeling Results of Oklahoma Sand - Water - NAPL Surrogate (Soltrol 220) System, (Generation 2 - field conditions), DOE Contract No. DE-AR21-94MC30360, Phase I

7.3.1.3 Summary

The modeling results in Figures 47 through 50 show significant improvement in both the effectiveness and efficiency of NAPL recovery. For a given NAPL-water viscosity ratio, the ultimate recovery improvement is independent of hydraulic gradient. The speed of which a given recovery is achieved, however, is dependent on both the viscosity contrast and the hydraulic gradient.

7.3.2 Porcelain Clay

A zero-dimensional analytical model of aquifer cleanup was applied to the hydraulic conductivity results obtained for porcelain clay. The approach followed that of Isherwood et al. (1990). This model assumes that the aquifer can be represented by a single, well-mixed domain of fixed volume, V , initially at a uniform ground water contaminant concentration, $c = c_o$, with recharge water having a contaminant concentration of zero entering at a fixed flow rate Q . This model simulates P&T remediation where the contaminant is extracted from the domain at the prevailing concentration, c , at a pumping rate equal to Q . The analytical solution for this model is:

$$c = c_o e^{-kt} \quad (\text{Equation 1})$$

where k is a constant described below, t is the time to reach a remediation value, and

$$k = -Q/Vn \quad (\text{Equation 2})$$

where n is the effective porosity. Rewriting equations 1 and 2

$$\ln(c/c_o)Vn = Qt \quad (\text{Equation 3})$$

Since Q is directly proportional to hydraulic conductivity (i.e., Darcy's law), it can be seen by inspection that if the flow rate is doubled, then the remediation time is decreased by a factor of two.

The hydraulic conductivity results for clay (Section 6.2.4) are significant because if the hydraulic conductivity of fine-grained materials, represented here by clay, can be increased by a factor of two with excitation, then the time to reach a remediation goal will be cut in half. Similarly, if a four-fold increase in hydraulic conductivity can be achieved, as evidenced in Figure 15, then the remediation time required to reach cleanup can be decreased to a quarter of what it would be without excitation.

7.3.2.1 Summary

In many remediation scenarios, the long time required to reach cleanup is controlled by the time required for diffusion of contaminants from fine-grained, low permeability sediments into coarse-grained high permeability sediments. If AER can significantly increase the hydraulic conductivity of low permeability materials, then there will be a significant reduction in environmental remediation costs. Note these favorable results pertain only to the hydraulic

behavior of the system, i.e., increasing water flux through the system. Acoustic excitation may also have an unknown favorable effect on contaminant mobility that could further reduce the remediation time.

8. RECOMMENDATIONS FOR PHASE II

8.1 Phase I Summary of Results

The Phase I program has successfully demonstrated that AEFs can enhance the remediation of contaminated soils and ground water, fluid flow modeling has shown that the acoustically enhanced process can enhance remediation, and engineering analyses indicates that an acoustically phased array configuration could generate the required acoustic energy to remediate a site.

A number of unresolved issues developed as a result of the Phase I investigation and these are described fully in the text. We will attempt to address some of these critical issues within the scope of the Phase II portion of the program.

8.2 Phase II Program

Phase II is a technology scaling study designed to bridge the gap from the generally 1-D work conducted in Phase I to the 3-D understanding and engineering necessary for a field scale test of AER technology. To have a field deployable system for Phase III, we need to determine in Phase II:

1. how an AEF interacts with a geohydrochemical environment specifically in regards to heterogeneities at all scales; and
2. how to engineer the deployment of phased array system.

To achieve these objectives we recommend that that the Phase II focus on identifying the characteristics of a field deployable AER technology system based on 2-D laboratory extraction testing at a scale of about 5 cubic feet, under AEFs generated by the phased arrays, and 2-D and 3-D modeling of (1) the fluid flow and transport behavior under baseline and excited conditions, and (2) the acoustical responses generated in the soil. This Phase II activity will use an actual DNAPL during testing. The outlined Phase II approach will allow for:

1. evaluating the coupling, propagation, and control of AEFs in the subsurface;
2. determining optimum well field and/or surface array configurations;
3. evaluating technology performance, safety, and cost; and
4. validation at the scale of testing computer models to predict the coupling, propagation, and control of acoustic energy, and the resultant physical and chemical effects in the subsurface.

In addition to the aforementioned program, a test site will be selected for the Phase III large scale field test. The results obtained in Phase I fully support initiation of Phase II.

9. REFERENCES

- Acre, S. D., R. R. Ross, M. R. Sclaf, E. R. Graber, J. W. Mercer and R. M. Cohen, eds., 1992. Dense Non-Aqueous Phase Liquids - A Workshop Summary, U.S. Environmental Protection Agency, EPA/600/R-92/030.
- Bear, J. and Y. Bachmat, 1991. Theory and Applications of Transport in Porous Media - Introduction to Modeling of Transport Phenomena in Porous Media, Kluwer Academic Publishers, 553 pp.
- Bear, J., E. Nichols, J. Ziagos and A. Kulshrestha, 1994. Effect of Contaminant Diffusion into and out of Low-Permeability Zones, Lawrence Livermore National Laboratory, UCRL-ID-115626.
- Bear, J. and J. J. Nitao, 1993. Infiltration and Contaminant Transport in the Vadose Zone (Non-isothermal), Part 1: Conceptual and Mathematical Models, Lawrence Livermore National Laboratory, UCRL-JC-111742, part 1.
- Beresnev, I. A. and P. A. Johnson, 1994. Elastic Wave Stimulation of Oil Production: a review of methods and results, Geophysics, vol. 59, pp. 1000-1017.
- Buckley, S. E. and M. C. Leverett, 1942. Mechanism of Fluid Displacement in Sands, Trans. AIME, vol. 146, pp. 107-116.
- Chemical Engineering, 1995. A Sound Way to Recover Oil From Soil, Cimentator Section, May, p.23.
- Cleveland, T. G. and S. Garg, 1993. Field Demonstration of Acoustically Enhanced Soil Washing System For In-Situ Treatment of Low-Permeability Soils, Abstract in Proceedings of the Gulf Coast Hazardous Substance Research Center's 1993 Symposium on Emerging Technologies: Metals, Oxidation, and Separation.
- Cohen, R. M. and J. W. Mercer, 1993. DNAPL Site Evaluation, C. K. Smoley, Boca Raton, Florida.
- Conrad, S. H., J. L. Wilson, W. R. Mason and W. J. Peplinski, 1992. Visualization of Residual Organic Liquid Trapped in Aquifers, Water Resources Research, vol. 28, pp 467-478.
- Craig, F. F., 1971. The Reservoir Engineering Aspects of Waterflooding, Monograph, vol. 3, Society of Petroleum Engineers of AIME, Dallas.

- Dering, J. P., J. L. Sollee, and T. M. Rynne, 1994. Non-Lethal Weapons for Helicopter Use: Phase I Final Report, SARA TN 94-013 (Secret), Prepared for U.S. Army AVRDEC, Contract # DAAJ02-C-0019.
- Dering, J. P., T. M. Rynne and J. L. Sollee, 1993, HPAB Incapacitation Weapon: Phase I Investigation, SARA TN 93-007, prepared for Army ARDEC.
- Duhon, R., 1964. An Investigation of the Effect of Ultrasonic Energy on the Flow of Fluids in Porous Media, Ph.D. Dissertation, University of Oklahoma, 162 pp.
- Fisher, R., I. M. Mason and C. C., Mosher, 1993. Cavitation-Induced Seismic Events Caused by Small Explosions in Boreholes, Geophysics, vol. 58, pp. 1565-1572.
- Gordon, R. B. and L. A. Davis, 1968. Velocity and Attenuation of Seismic Waves in Imperfectly Elastic Rock, Journal of Geophysical Research, vol. 73, pp. 3917-3935.
- Illangasekare, T., D. Znidarcic, G. Walser and J. Weaver, 1994. An Experimental Evaluation of Two Sharp Front Models for Vadose Zone Non-Aqueous Phase Liquid Transport, U.S. Environmental Protection Agency, EPA/600/R-94/197.
- Isherwood, W.F., C.H. Hall, and M.D. Dresen, 1990. CERCLA Feasibility Study for the LLNL Site, Lawrence Livermore Laboratory, UCRL-AR-104040.
- Jones, S. C. and W. O. Roszelle, 1978. Graphical Techniques for Determining Relative Permeability from Displacement Experiments, Journal of Petroleum Technology, vol. 15, pp. 807-817.
- Lee, K. H., A. Kulshrestha and J. J. Nitao, 1993. Interim Report on Verification and Benchmark Testing of the NUFT Computer Code, Lawrence Livermore National Laboratory, UCRL-ID-113521.
- Lee, T. M., 1963. Method of Determining Dynamic Properties of Viscoelastic Solids Employing Forced Vibration, Journal of Applied Physics, vol. 34, pp. 1524-1529.
- Mackay, D. M. and J. A. Cherry, 1989. Groundwater Contamination: Pump-and-Treat Remediation, Environmental Science and Technology, vol. 23, pp. 630-636.
- Mahrer, K. D. and B. J. Zook, 1993. A Field Test of an Electrodeless Arc Discharge, Borehole Seismic Source, Geophysics, vol. 58, pp. 1558-1564.
- Nitao, J., 1995. Reference Manual for the NUFT Flow and Transport Code, Lawrence Livermore National Laboratory, UCRL-ID-113520, in preparation.
- Omnes, G., 1990. Experimental Study of the Coupled Cord Downhole Seismic Source, 60th Annual International Meeting, Society of Exploration Geophysicists, pp. 148-155.

- Osoba, J. S., J. G. Richardson, J. K. Kerver, J. A. Hafford and P. M. Blair, 1951. Laboratory Measurements of Relative Permeability, Petroleum Transactions of AIME, T.P. 3020, vol. 192, pp. 45-56.
- Paulsson, B. N. P., 1994. Development of a Downhole Hydraulic 3-Component Seismic Source for Cross-well Seismic, Reverse VSP and Single-well Seismic Applications, unpublished technical briefing.
- Rapoport, L. A. and W. J. Leas, 1953. Properties Of Linear Waterfloods, Transactions of AIME, vol. 198, pp. 139-148.
- Reddi, L., S. Berliner and K. Y. Lee, 1993. Feasibility of Ultrasonic Enhancement of Flow in Clayey Sands, Journal of Environmental Engineering, vol. 119, pp. 746-752.
- Reddi, L. and S. Challa, 1994. Vibratory Mobilization of Immiscible Liquid Ganglia in Sands. Journal of Environmental Engineering, vol. 120, pp. 1170-1190.
- Reddi, L., A. Hadim, R. N. Prabhushankar and F. Shah, 1994. Vibratory Extraction of Clay Fines From Subsurface, Journal of Environmental Engineering, vol. 120, pp. 1544-1558.
- Salathiel, R. A., 1973. Oil Recovery by Surface Film Drainage in Mixed-Wettability Rocks, Journal of Petroleum Technology, vol. 25, pp. 1216-1224.
- Schwille, F., 1988. Dense Chlorinated Solvents in Porous and Fractured Media: Model Experiments (English translation), Lewis Publishers, Ann Arbor, Michigan.
- Scott, R. F., 1963. Principles of Soil Mechanics, Addison-Wesley, 550 pp.
- Seed, H. B., R. T. Wong, I. M. Idriss and K. Tokimatsu, 1984. Moduli and Damping Factors for Dynamic Analyses of Cohesionless Soils, University of California Berkeley Earthquake Engineering Research Center, UCB/EERC-84/14.
- Sollee, J. L., T. M. Rynne and J. P. Dering, 1995. Selective Area/Facility Denial Using High Power Acoustic Beam Technology: Phase I Final Report, SARA TN-003, prepared for U.S. Army ARPA, Contract # DAAH-01-94-C-R242.
- Stevens, H. W., 1966. Measurement of the Complex Moduli and Damping of Soils Under Dynamic Loads, U.S. Army Cold Regions Research and Engineering Laboratory, Technical Report 173.
- Sun, J. I., R. Goleorkhi and H. B. Seed, 1988. Dynamic Moduli and Damping Ratios for Cohesive Soils, University of California Berkeley Earthquake Engineering Research Center, UCB/EERC-88/15.
- Szlag, D. C. and T. H. Illangasekare, 1992. Experimental Investigation in Modeling of Immiscible Liquid Entrapment in Porous Media, Paper in Proceedings of Conference on Hazardous Waste Research.

- Wilson, J. L., S. H. Conrad, W. R. Mason, W. Peplinski and E. Hagen, 1990. Laboratory Investigation of Residual Liquid Organics, U.S. Environmental Protection Agency, EPA/600/6-90/004.
- Winkler, K., A. Nur and M. Gladwin, 1979. Friction and Seismic Attenuation in Rocks, *Nature*, vol. 277, pp. 528-531.
- Yokel, F. Y., R. Dobry, D. J. Powell and R. S. Ladd, 1980. Liquefaction of Sands During Earthquakes: The Cyclic Strain Approach, Paper in Proceedings of The International Symposium on Soils Under Cyclic and Transient Loading, Swansea, January 7-11, 1980.
- Yu, S. and Y. Liu, 1992. Form Factor of a Cylindrical Sonic Source and Application in Borehole Acoustics, *Journal of the Acoustical Society of America*, vol. 93, pp. 2592-2598.
- Zhang, B., H. Dong and K. Wang, 1994, Multipole Sources in a Fluid Filled Borehole Surrounded by a Transversely Isotropic Elastic Solid, *Journal of the Acoustical Society of America*, vol. 96, pp. 2546-2555