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Non-Invasive Characterisation of NAPL Contaminated Land by Spectral Induced Polarisation – A Literature Review

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Contents

1 Introduction

The BGS has undertaken a programme of research in the use of SIP tomography for non-aqueous phase liquid (NAPL) contaminated land characterisation. This research has been funded under the joint NERC/ERPSRC URGENT Programme (Grant No. GR/M34010), and has been carried out in partnership with the University of Birmingham. The primary objectives of the BGS components of the work were to undertake:

- synthetic modelling studies;
- model tank/column experiments using controlled releases of NAPLs to correlate SIP spectra and images against known geometries, media properties, flow paths and hydraulic parameters;
- field-testing at industrial sites to validate laboratory and interpretative criteria.

The aim of this review is to provide the context in which this research was carried out. The review includes an introduction to problem of NAPL contamination and transport, the benefits of applying geophysics to the NAPL contamination problems, a description of the SIP technique, and a summary of previous studies involving the use of SIP to the detect and monitor NAPL contamination.

2 The Problem of NAPL Contamination

2.1 SOURCES OF CONTAMINATION

Groundwater contamination by NAPLs has been increasingly recognised as a serious environmental problem. NAPLs have been widely used by a range of industries for many decades, and have been introduced into the subsurface by spills, uncontrolled releases and leaking storage and disposals facilities (ERL, 1987; Barry, 1991; Pankow et al, 1996). NAPLs are typically divided into two categories on the basis of their density relative to that of water; dense NAPLs (DNAPL) have a density greater than water, whilst light NAPLs (LNAPL) are less dense than water. The most common LNAPLs are petroleum products, and petroleum additives such as the slightly soluble BTEX compounds (benzene, toluene, ethyl benzene and xylenes) (Lucius et al., 1992) and the more soluble methyl tertiary-butyl ether (MTBE) and alcohols. Common DNAPL contaminants include solvents, coal tars, PCB oils and pesticides (Cohen and Mercer, 1993); however, chlorinated solvents are the most prevalent DNAPL contaminants.

2.2 SUMMARY OF NAPL MIGRATION AND FATE

Upon release NAPLs migrate downwards under gravity through the unsaturated zone, leaving behind them residual liquid, entrained in the pore space by capillary forces and surface tension. Given a sufficient volume of liquid the NAPL will reach the saturated zone; at this point an LNAPL will spread out laterally over the capillary zone. DNAPLs, however, continue to migrate downwards into the saturated zone until an impermeable layer is reached, at which point pooling and lateral spreading may occur. Dissolution of NAPL in contact with flowing groundwater can lead to the production of solute plumes, whilst volatilisation of free product within the vadose zone can produce vapour plumes also leading to an increase in the extent of contamination. NAPLs are typically characterised by low solubilities and are resistant to biodegradation and natural attenuation; they are, however, soluble enough to cause levels of contamination far greater than those permissible in drinking water. Consequently, even small amounts of NAPL can present a long-term source of contamination. Reviews covering NAPL transport, fate, characterization and remediation are provided by Mercer and Cohen (1990), Huling and Weaver (1991), Cohen and Mercer (1993), Newell et al. (1995) and Gallo et al. (1998).

2.3 HYDROGEOLOGICAL RESEARCH IN NAPL TRANSPORT AND FATE

Numerous laboratory experiments (Chevalier and Peterson, 1999) have been undertaken in an attempt to understand how NAPLs move through the subsurface. Some of the first studies carried out into DNAPL movement were carried out by Schwille (1988) and Keuper et al. (1989); these experiments involved the visual observation of dye stained NAPL movement through a range of media. Subsequent studies have also used visual observation of DNAPL flow in porous (Illangasekare et al., 1995; Smith and Zhang, 2001) and fractured (Kueper and McWhorter, 1991; Longino and Kueper, 1999) media to investigate NAPL behaviour, remediation (Conrad et al., 2002), and to assist in the development of numerical models describing multiphase flow (Oostrom et al., 1999; Trantham and Durnford, 1999). Similar experiments have been conducted to study LNAPL flow, both to understand flow and entrapment mechanisms (Pantazidou and Sitar, 1993; Van Geel and Sykes, 1994a; Catalan and Dullien, 1995) and to contribute to the production of numerical models (Van Geel and Sykes, 1994b; Oostrom et al. 1997; Schroth et al., 1998).

Though much of the research into NAPL transport has comprised laboratory studies using tanks and columns, research into the NAPL behaviour, natural attenuation and remediation has also been carried out at field sites using controlled releases (Poulsen and Kueper, 1992; Kueper et al., 1993; Broholm et al., 1999) or accidental spills (Ostendorf et al., 1993; Sehayek et al., 1999; Lee et al., 1999).

3 The Role of Geophysics

Geophysical techniques, and in particular electrical methods, have been successfully used to monitor hydraulic experiments in the laboratory. Studies by Archie (1942) defined the link between pore fluid resistivity, gas and NAPL saturation and the formation resistivity. This work had provided the basis for the use of electrical techniques to detect and monitor aqueous and nonaqueous groundwater contaminants. Examples of the geophysical monitoring of fluid movement in the laboratory are provided by Binley et al. (1995), Binley et al. (1996), Gheith and Schwartz (1998), Slater et al. (2000), Depountis et al. (2001) and Slater et al. (2002). These studies have highlighted the benefits of non-invasive monitoring techniques that can allow plume geometry and solute concentrations to be determined. A number of these studies have employed time-lapse differential imaging in order to isolate the effects of pore fluid changes over time from heterogeneities related to the physical properties of the porous media.

The characterisation of NAPL contaminated field sites and source zones are essential for the successful design of remedial strategies. Conventional site investigation relies in part upon intrusive methods, such as boreholes and cone penetration tests. These techniques provide information of the subsurface only at discrete intervals and have the potential to worsen contamination by opening NAPL migration pathways through impermeable and low permeability zones. Intrusive techniques can also result in the migration of vapours and contaminated materials to the surface, potentially causing toxic chemical, fire or explosion hazards. Alongside intrusive techniques geophysical methods, such as ground penetrating radar, electrical methods, seismic reflection and electromagnetic surveying, are now beginning to be applied to NAPL contamination problems (ITRC, 2000), both to assist in site characterisation (e.g. Chambers et al., 1999; Ogilvy et al. 1999; Chambers, 2001, Ogilvy et al., 2002) and to monitor NAPL contamination (Olhoeft, 1987; Ward et al., 1992; Greenhouse et al., 1993; Brewster et al., 1995; Daily et al., 1995; Sauck et al., 1998; Atekwana et al., 2000; Sauck, 2000;

Sneddon et al., 2000) and remedial processes (e.g. Daily and Ramirez, 1995; Schima et al., 1996; Newmark et al., 1998). Geophysical techniques can have the advantages of being relatively inexpensive, reducing the need for intrusive investigations, and providing spatially continuous information regarding subsurface structure.

A number of studies employing SIP to detect and monitor NAPL both in the laboratory and the field will be considered in Section 5.

4 The SIP Method

4.1 BACKGROUND AND APPLICATIONS

Electrical impedance is a complex quantity consisting of a magnitude and a phase component. Impedance magnitude is the amplitude of the frequency dependent resistance, whilst the impedance phase provides is a measure of induced polarization (IP). Frequency domain SIP involves the measurement of impedance over a range of frequencies; the resulting measured phase and amplitude spectra are dependent on the physical and chemical properties of the material. In geophysics SIP measurements are typically made in the mHz to kHz frequency range. This range is often restricted even further, e.g. <100 Hz, in order to reduce the inductive coupling effects, which are more pronounced at higher frequencies. The time-domain SIP response can also be determined by measuring transient voltages at time intervals after the applied current is switched off (Johnson, 1984). The SIP method is also referred to as complex resistivity. Overviews of the SIP method are provided by Sumner (1976) and Telford et al. (1990).

SIP was initially applied to the characterisation of metallic mineral ore bodies (Pelton et al., 1978; Vanhala and Peltoniemi, 1992). The technique has also been used to characterise sedimentary rocks in terms of their surface area and other hydrogeological properties (Borner and Schon, 1991; Ruffet et al., 1991; Kulenkampff and Yaramanci, 1993; Vanhala and Soininen, 1995; Borner et al., 1996; Weller and Borner, 1996; Boadu and Seabrook, 2000). Other uses of SIP have included the detection of buried metallic objects such as waste (Weller et al., 1999), UXO (Daily et al., 2000), and pipelines (Parra, 1984). Similar techniques are also used for biomedical imaging (Boone et al., 1997; Cheney et al., 1999), and industrial process tomography (Williams and Beck, 1995; Beck et al., 1998). However, its use in detecting and discriminating between NAPLs is now the focus of considerable research efforts (Vinegar and Waxman, 1984a; Olhoeft, 1985a; Olhoeft, 1985b; Olhoeft, 1986; Walther et al., 1986; Sadowski, 1988; King and Olhoeft, 1989; Vanhala et al., 1992; Borner et al., 1993; Vanhala, 1997).

4.2 MECHANISMS OF IP

The two main mechanisms of IP are metallic and membrane polarization. Metallic polarization (Angoran and Madden, 1977) occurs when a voltage is applied across an electrolyte filled pore space blocked by a conducting mineral grain. The transfer from electrolytic conductance to electrical conductance impedes the current flow, and the electrolyte/mineral interface becomes polarized. As applied voltage is removed a transient decay voltage is produced as the ions diffuse back to their original positions. Membrane polarization (Vacquier et al., 1957) occurs when the movement of ions within a rock mass are restricted, due to blockages at pore throats caused by layers of loosely bound ions on the mineral grain surfaces. As with metallic polarization, a residual voltage is produced due to diffusion-controlled relaxation of the ions when the applied current is removed. Membrane polarization is particularly associated with the presence of clay minerals (Vacquier et al., 1957; Marshall and Madden, 1959; Klein and Sill, 1982; Vinegar and Waxman, 1984b), and is commonly related to cation exchange capacity (Vinegar and Waxman, 1984a; Borner et al., 1996), specific surface area (Borner and Schon, 1991; Borner et al., 1996) and porosity (Ruffet et al., 1991; Boadu and Seabrook, 2000) of rock and sediment. Pore fluid saturation (Kulenkampff and Yaramanci, 1993; Ulrich and Slater, 2002) and pore fluid chemistry has also been shown to exert a major control on the IP response, with increasing electrolyte conductivity general resulting in a decrease in the measured IP effect for sediments and disseminated metalliferous mineral bodies (Keevil and Ward, 1962; Vinegar and Waxman, 1984a; Lesmes and Frye, 2001).

An additional mechanism effecting SIP measurements are clay-organic reactions. Studies by Olhoeft (1985b) and King and Olhoeft (1989) demonstrated that clay-organic reactions occurring within clay rich sediment and rock contaminated with a range of NAPLs exerted a strong influence on the measured phase spectra of the samples. Other lesser factors that have been observed to influence the SIP response include electrokinetic, thermoelectric and electromagnetic effects (Marshall and Madden, 1959).

4.3 SIP MODELS

Several models have been developed to describe the electrical frequency dependence displayed by earth materials. The Cole-Cole model (Pelton et al., 1978; Brown, 1985; Lesmes et al., 2000; Xiang et al., 2001), which was originally applied to the characterization of metalliferous ore bodies, is perhaps the most common and widely used. Cole-Cole models (single and multiple) have been shown to fit the measured spectra from a broad range of other materials including many uncontaminated (Ruffet et al., 1991; Vanhala and Peltoniemi, 1992; Boadu and Seabrook, 2000) and contaminated (Piggott et al., 2000) rocks and soils, and artificial targets (Kemna et al., 2000). Other formulations include the Cole-Davidson (Pelton et al., 1978), constant-phase (Borner et al., 1996) and Dias (Dias, 2000) models.

4.4 NUMERICAL INVERSION

The inversion of SIP data for electrical imaging has been carried out on a ‘frequency by frequency’ basis using DC resistivity algorithms (e.g. Oldenburg and Li, 1994) where apparent chargeability is obtained by employing a linear approximation (Siegel, 1959), or by algorithms that fully account for the complex nature of impedance (Weller et al., 1996; Ramirez et al., 1999; Weller et al., 1999; Yang et al., 2000). Inversion methodologies in which Cole-Cole parameters are recovered from SIP measurements have also been developed for both time (Yuval and Oldenburg, 1997) and frequency domain (Routh et al., 1998; Loke et al., 2001) data. However, synthetic modelling studies by Chambers et al. (2001) indicated that Cole-Cole parameter distributions are difficult to recover from measured SIP data, as they are essentially second order effects. Cole-Cole parameters are therefore likely to be even more difficult to recover from field data, which are subject to greater noise influences.

4.5 SIP IMAGING

SIP imaging involves the collection of data sets comprising multiple electrical measurements (amplitude and phase) using multi-electrode arrays. Collection of data sets for electrical imaging or tomography generally requires the use of computer controlled automated measurement systems and multi-electrode arrays, due to the typically large number of measurements made during the course of a survey. These data are processed using inversion algorithms (Section 4.4) to produce tomograms of subsurface electrical property distributions. Examples of SIP imaging procedures are given by Ramirez et al. (1999), Daily et al. (2000) and Kemna et al. (2000).

5 The Application of SIP to the Investigation of NAPL Contaminated Land

5.1 LABORATORY SAMPLE MEASUREMENTS

5.1.1 Apparatus

Laboratory sample holders for SIP measurements are constructed from non-polarising materials such as glass, silicone and plastics (Vinegar and Waxman, 1984a; Vanhala and Soininen, 1995; Boadu and Seabrook, 2000). Four electrodes are generally used, two current and two potential, and a measuring instrument with a high input impedance for potential measurements in the in order to minimise the polarization of the measurement electrodes (Olhoeft, 1985b; Vanhala and Soininen, 1995). Vinegar and Waxman (1984a) demonstrated that the polarizing properties of the current electrodes are not an important consideration, as they have no effect on the measured polarization. Measurement electrode composition has however been demonstrated to effect measurement quality (Vanhala and Soininen, 1995). Non-polarising silver-silver chloride (Vinegar and Waxman, 1984a; Vanhala and Soininen, 1995) and copper-copper sulphate (Boadu and Seabrook, 2000) electrodes have been used, but are difficult to clean and are easily contaminated. Testing has demonstrated that measurement quality using stainless steel and aluminium (Vanhala and Soininen, 1995) potential electrodes can be poor. Platinum potential electrodes are perhaps the most commonly used (Vinegar and Waxman, 1984a; Olhoeft, 1985b; Vanhala and Soininen, 1995; Borner et al., 1993; Vanhala, 1997), as they are easy to clean, and are less susceptible to electrode polarization than the other metallic electrodes tested. Studies by Chambers (2001) have demonstrated that electrode size is also a crucial factor effecting measured IP response; excessive miniaturisation of electrodes was shown to result in very poor measurement quality.

5.1.2 Sample Analysis

A number of other workers have considered the effects of NAPL contamination on SIP characteristics of laboratory samples. Vinegar and Waxman (1984a) used complex resistivity measurements to determine NAPL saturation in shaly sands. Olhoeft (1985b) described the SIP response of a number of clay containing rock and soil samples contaminated with a range of organic liquids. Olhoeft (1986) presented a studying illustrating the effects of organic waste on the properties of a montmorillonite soil. King and Olhoeft (1989) described the differences between the response of uncontaminated montmorillonite and toluene contaminated montmorillonite. Vanhala et al. (1992) studied the effects of toluene, ethylene glycol and heptane on glacial till samples. Borner et al. (1993) studied the effects of adding hexane, dichloromethane, benzene, methanol and mixed oils to a range of clays including bentonite and kaolin. Vanhala and Soininen (1995) considered the effects of adding heptane to bentonitic till samples. Vanhala (1997) described the effect of waste and motor oil, and maturation time on the properties of sand and till. Piggott et al. (2000) considered the effect of the iso-paraffin SOLTROL-100 on Ottawa Sand, using relatively high frequency (1 kHz to 1MHz) SIP measurements. Scott and Barker (2001) presented a studying illustrating the effect of motor oil contamination to the SIP response of clean Triassic Sandstone samples from the UK.

5.2 TOMOGRAPHIC IMAGING

Few studies have demonstrated the application of SIP technology to the tomographic imaging of NAPL. The examples of tomography that do exist in the literature all utilise a 'frequency by frequency' inversion methodology to calculate electrical models in terms of amplitude and phase.

Cho (1996) describes a laboratory tank experiment in which tetrachloroethylene (PCE) migration through sand was monitored using electrical impedance tomography. A hydrophobic dye was used to stain the PCE to allow visual confirmation of DNAPL distribution. Measurements were made using carbon electrodes.

Weller et al. (1996) monitored the migration of NAPL through sediment using the SIP technique. The experiments were carried out in a large laboratory tank, which contained zones of fine, medium, and coarse sand, and gravel respectively. Platinum electrodes arranged in a plane along the side, base and top of the tank. The DNAPL trichloroethene (TCE) was introduced into the top of the tank. The progress of the TCE was monitored by repeat measurements using a fixed dipole-dipole configuration, to produce breakthrough curves in terms of phase and amplitude. Measurements were carried out at a low frequency, between 1 and 4 Hz. Tomographic images were generated before and after infiltration. Breakthrough curves were also generated from a xylene infiltration experiment, in unsaturated conditions. Further inverted tomographic images were generated in attempt to identify residual xylene distributions resulting from fluctuations in the water table.

Daily et al. (1998) described a field experiment in which the movement of a controlled release of dyed PCE through a large sand filled tank was monitored using electrical impedance tomography. Time lapse images generated from data collected using frequencies of 1, 64, 256, 512 and 1024 Hz were used to identify NAPL induced changes in the subsurface. Lead electrodes were used for data collection.

6 Concluding Remarks

NAPL contamination is a serious and widespread problem and the focus of much research in the fields of both hydrogeology and geophysics. The strength of geophysical methods lies primarily in the potential for the collection of non-invasive and spatial information regarding subsurface contamination. Electrical techniques, including SIP, have shown particular promise in relation to the detection and monitoring of NAPL contamination; such contamination has been demonstrated to alter the electrical properties of rocks and soils as a result of the highly resistive nature of most NAPLs, and by mechanisms such as clay organic reactions. Though much research effort has gone into the study of the SIP response of 1D laboratory samples the full potential of SIP imaging to monitor laboratory experiments and field contamination has yet to be realised.

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