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Running title: Induced polarization under non-isothermal conditions

Highlights

- Proper application of spectral induced polarization (SIP) in samples polluted by dense non-aqueous phase liquids (DNAPL)
- Temperature and saturation separately affect electrical complex resistivity
- Temperature has a considerable effect on Cole-Cole parameters in a 2D tank
- Saturation changes have a remarkable impact on phase shift
- 2D tank results on complex resistivity confirm 1D cell findings

Abstract

In this decade, electro-geophysical methods are widely used in different environmental subjects. Study on soil remediation polluted by DNAPLs has become a certain need for all countries. Geoelectrical methods show their potential to facilitate evaluating decontamination processes. Our challenge in this study was to understand how coupled temperature and saturation changes affect electro-geophysical parameters in a contaminated 2D sample. The primary objective was to evaluate the efficiency and potential of spectral induced polarization (SIP) for monitoring the recovery of dense non-aqueous phase liquids in contaminated porous media. A set of 2D tank experiments investigated the impacts of temperature and saturation changes on the electrical complex resistivity of a saturated porous medium under non-isothermal conditions. The measurements were made with a coal-tar and water fluid pair in a porous medium that has been simulated by 1 mm glass beads. We circulated hot water around the tank and used an immersion heater to heat the porous medium in the tank at different stages. The SIP technique (also called complex resistivity) was used to measure the complex electrical resistivity of a medium in the frequency domain. The experimental results for a simple drainage case were validated using numerical modeling. The complex electrical resistivity was used to obtain the saturation field before and after imbibition. For this purpose, the generalized Archie's law obtained for the same fluid pair with 1D cells (with a vertical flow) was used. Our results from electrical resistivity measurements for saturation fields are in accordance with 2D tank images and can illustrate the saturation change with pointwise resistivity measurements. The results show that saturation change has the main role in electrical resistivity variation compared to temperature (5 to 7%). We also studied the effects of temperature change on the Cole-Cole parameters and the results confirm our previous findings with the same variation trend in these parameters. The results from varying electrical complex resistivity from the 2D tank (with vertical and horizontal flow) in the laboratory conditions will help us to understand the coupled temperature and saturation effects on complex resistivity in a real polluted site case.

*Keywords

Induced polarization, modeling, pollution, resistivity, environmental

Introduction

Pollution of soil due to dense non-aqueous phase liquid (DNAPL) spilling has become a crucial issue of concern during the last decade, causing numerous geo-environmental (e.g., mitigation of DNAPLs on aquifers) problems (Habiyakare et al., 2022). In polluted sites, DNAPLs sink below the water table and cause dissolved plumes in the subsurface (Pankow and Cherry, 1996; Power *et al.*, 2014). After many years of research, soil contamination due to DNAPLs is still one of the main challenges for the environmental and geophysical communities (Kavanaugh, Suresh and Rao, 2003). DNAPLs, including coal tar (CT), contaminate soils and water widely when released in the subsurface, mainly from industrial parks. DNAPLs are part of a group of immiscible fluids that pass through the saturated and unsaturated zones due to gravity and capillary forces, via migration passages made of pores, until stopping at an impermeable stratum (Illangasekare *et al.*, 1995; Power *et al.*, 2014). Various remediation techniques can be used in these sites to depollute them. These include pump and treat, thermal, bioremediation, and chemical methods (Beyke and Fleming, 2005; Hadley and Newell, 2012). Pump-and-treat is the common and first remediation technique to remove the free product. After pumping, the residual saturation can be removed using biochemical remediation techniques.

Geophysics has shown the value of the long-term monitoring of clean-up processes (e.g., Brewster *et al.*, 1995; Wilson *et al.*, 2009). Of all the geophysical methods, electromagnetic methods and especially electrical methods are useful for studying the processes involved during clean-up and degradation of pollutants (Chambers *et al.*, 2004; Mao *et al.*, 2016; Deng *et al.*, 2020), 3D imaging of a jet fuel spill (Orozco *et al.*, 2021; Helene, Moreira and Bovi, 2020). The electro-geophysical methods, especially direct current (DC) resistivity, induced polarization (IP) techniques, and spectral induced polarization (SIP) have proved their potential in geological conditions, in saturated conditions (e.g., Weller, Nordsiek and

Debschütz, 2010; Slater *et al.*, 2014; Revil *et al.*, 2015; Kremer *et al.*, 2016b; Sparrenbom *et al.*, 2017), unsaturated conditions (e.g., Maineult, Jougnot and Revil, 2018), oil, gas, and mining industries (e.g., Friedrichs *et al.*, 1999), soil pollution and remediation monitoring (e.g., Pearce and Zuluaga, 2004; Ling and Zhang, 2017; Liao *et al.*, 2018) and civil engineering (e.g., Susanto *et al.*, 2013). In a multiphase porous medium, various combinations of different parameters such as temperature, saturation, level of compaction, and pore fluid properties can have a similar effect on the measurements (e.g., Sen and Goode, 1992; Denicol and Jing, 1998). Therefore, studying the effect of each parameter to determine their role and ideally, their related simultaneous effects on geophysical parameters is necessary.

Sadowski (1988) performed the first laboratory study on different clay samples with different pollutants and different concentrations. He found that the SIP method could be used to identify organic contaminants in clayey soils for different concentrations. Börner, Gruhne and Schön (1993) contributed to laboratory research on the effects of organic liquid contaminations on different rock types and sandstones. They demonstrated that both the real and imaginary parts of complex resistivity are affected by the properties of the contaminant filling the pores. They noted that in some cases the various contaminants could be differentiated using the imaginary part and emphasized the effect of interactions between pore fluid characteristics and pores' internal surface structure.

The existence of organic contamination can affect capacitive conduction in the IP response due to the surface reaction between contamination and soil grains (Vaudelet *et al.*, 2011). IP responses in these contaminated soils are related to the saturation (Breede *et al.*, 2012), distribution, concentration, and types of contaminants present in soil (Kemna *et al.*, 2012). Previous studies demonstrate that the presence of contaminants (Gasoline and leachate results of acidification by organic and carbonic acids) in the soil increases resistivity (Benson, Payne and Stubben, 1997; Sauck, 2000). Martinho, Almeida and Matias (2006) reported an increase in chargeability due to the presence of pure organic contaminations (gasoline, isopentane, toluene, and benzene) in clayey soil. Many studies based on the SIP method have shown an increase in amplitude and phase shift due to the existence of hydrocarbons (Vanhala, Soininen and Kukkonen, 1992; Börner *et al.*, 1993; Schmutz *et al.*, 2010). However other SIP studies reported a decrease in amplitude and phase shift in different media consisting of different pure and dissolved hydrocarbons (Weller and Börner, 1996; Vanhala, 1997; Revil, Schmutz and Batzle, 2011).

Recently, Iravani *et al.* (2020a, 2020b) evaluated how temperature and saturation changes in two DNAPLs (coal tar and chlorinated solvent) affect complex electrical resistivity and relative permittivity. They used an oven to study temperature change and a 1D drainage-imbibition system to study how saturation changed these two parameters. They carried out experiments in cylindrical cells filled with glass beads (to simulate soil/porous media) and saturated with water and CT. The relationship between electrical resistivity and relative permittivity with saturation was investigated for two different coupled immiscible fluids, water/coal-tar and canola oil/salty ethanol. They found that temperature changes these two geophysical parameters linearly for the coal tar (CT) and chlorinated solvent coupled with water. The effect of saturation change was not linear, and they did state that the generalized Archie's law could fit their experimental data well. Colombano *et al.* (2020) investigated the relationship between resistivity and saturation for chlorinated solvent. They used a drainage-imbibition protocol to monitor electrical resistivity and permittivity. Their objective was to estimate residual saturation indirectly. They used the complex refractive index model (CRIM) and generalized Archie's law to model their permittivity and resistivity data, respectively. They reported that estimated electrical resistivity presented less correlation with the experimental data compared to permittivity. They concluded that temperature and saturation affect the electrical resistivity and relative permittivity of the medium. Colombano *et al.*

(2021) investigated the imbibition of a mixture of chlorinated solvents by thermal and chemical enhancements (Colombano *et al.*, 2021). They found that the temperature has no effect on DNAPL recovery that could be linked to the actual properties of the oils. However, coal tar is more viscous than most chlorinated solvents. Moreover, the dynamic viscosity of coal tar decreases significantly with temperature (Philippe *et al.*, 2020). To the best of our knowledge, no other work is reported on electrical resistivity measurements for coal tar.

Blondel *et al.* (2014) compared lab and field results in clayey and clayey-silt soils. They quantified a resistivity increase, normalized chargeability, and decreased resistivity in measurements with oil not degraded. When it is partially degraded they saw no changes in the phase-lag and chargeability parameters. Gourry, Jeannot and Proust (2001) also showed that under certain conditions, oil pollution like this can increase chargeability because of high resistivity and it is rather related to wettability. This statement was not confirmed by Blondel *et al.* (2014).

Orlando and Palladini (2019) carried out a series of laboratory tests to study how resistivity and permittivity vary in a 2D cell in a multiphase (two-phase) system. The contamination they used was HFE-7100 (hydrofluoroether) in saturated (with pure, dissolved, and gaseous phases) grained glass bead (GB) porous media. They used geophysical measurements and physico-chemical constraints to monitor these parameters over a long period. Different geophysical techniques were used to propose empirical models to monitor water saturation in a multiphase porous media contaminated by DNAPL. Photographs were used to confirm the geophysical data. Low frequency IP can detect the pure HFE-7100 phase. In another study, Orlando and Renzi (2015) studied how electrical resistivity varies in ground due to the existence of DNAPL in a laboratory study. Like Orlando and Palladini (2019), they used a GB-packed medium contaminated with HFE-7100 in a saturated condition. They found that different DNAPL phases have different impacts on the physico-chemical characteristics of the contaminated medium. In their study, free and dissolved DNAPL caused a change in electrical resistivity. They concluded that electrical resistivity coupled with the GPR technique can be used for characterizing the contaminated sites because neither technique could be sensitive enough for a long time.

It is known that the presence of organic contaminants modifies the impedance of the fluid-grain interface through clay polymerization (Olhoeft, 1985; Vanhala, 1997). The presence of organic pollutants as free phase changes the distribution of the conductive aqueous phase in the pore space (like air can do in unsaturated media). As SIP response changes with varying water/air saturation (e.g., Ulrich and Slater, 2004; Titov et al., 2004; Binley et al., 2005, Maineult et al. 2018), free-phase hydrocarbons should also produce a measurable SIP effect. A few studies have been conducted on complex resistivity laboratory measurements either on artificially contaminated soil or rock samples or on field samples from contaminated sites. Even though the results differ in the observed effects, they proved that the SIP response of soils and rocks is measurably influenced by hydrocarbon contamination. However, a detailed understanding of how contamination affects SIP response is still lacking. Many studies did not pay sufficient attention to controlling the nature or distribution of contaminants in the porous media, and on assessing independently the effects of dissolved, adsorbed, and free-phase, leading sometimes to what appear to be contradictory results. Here, we assumed that the SIP response of the contaminant obeys a law similar to what Vaudelet et al. (2011) observed above a plume of chlorinated solvents, i.e., a Cole-Cole like behavior.

The study of geophysical parameters during remediation by the thermal enhanced method is influenced by two environmental parameters: temperature due to soil heating (Iravani *et al.*, 2020a, Philippe *et al.*, 2020) and saturation changes (Iravani *et al.*, 2020b; Colombano *et al.*, 2020) due to contamination pumping. In previous studies, we separately investigated the impact of temperature and saturation on electrical resistivity in contaminated saturated porous media with CT (Iravani *et al.*, 2020a, 2020b).

In this study, we investigated the coupling of transient heat transfer in multiphase flow with saturation variation in a 2D tank. No work to date has studied transient temperature transfer and saturation changes at the same time in 2D due to its complexity. Heating in the 2D tank mimics the soil heating in the field by increasing the temperature of pollutants to decrease their viscosity. These phenomena increase pollutant mobility and so increase the pumping rate of contamination in saturated soil during depollution. Heating the samples also changes the geo-electrical parameters. The goal of the current study is to determine the saturation field in the 2D tank from resistivity measurements. All these experimental studies have been done to implement the SIP for monitoring the real remediation process of DNAPLs in the field.

It is worth noting that one of the main challenges in the monitoring of the subsurface with geophysical methods is that there are few methods (e.g., boreholes) for verifying the recorded geophysical response of the medium to validate the remediation process (Spies, 1996; Keys, 2017).

Theoretical background

Spectral-induced polarization mechanisms

SIP is an extension of the IP method with alternating current (AC) in the frequency domain. SIP is used to study the ratio and phase shift between voltage and current of the subsurface or a sample. Generally, electrical polarization in the range of studied frequencies (i.e., from 0.01 to 10,000 Hz) has four main mechanisms including electrode polarization, membrane polarization, Maxwell-Wagner polarization, the polarization of the diffuse layer (electrical double layer, EDL) including the Stern layer polarization (e.g., Kemna *et al.*, 2012).

This study does not separately investigate the different mechanisms involved to characterize the effect of CT saturation and temperature changes.

Electrical complex resistivity

The electrical resistivity (ρ) in a homogeneous sample can be obtained by multiplying resistance (R) and a geometric factor (K) related to the position of the electrodes (for current injection and voltage measurement). The complex electrical resistivity reads (e.g., Guéguen and Palciauskas, 1994; Ghorbani, 2007; Glover, 2015):

$$\rho_{(f)} = \rho'_{(f)} - i\rho''_{(f)} = |\rho_{(f)}| exp(i\varphi) \quad (1)$$

where ρ' and ρ" are, respectively, the real and the imaginary parts of complex resistivity and $|\rho_{(f)}|$ and φ are the amplitude and the phase shift of the resistivity response (for low phase, below a few hundred of mrads, can be simplified as $\varphi \approx \rho''_{(f)}/\rho'_{(f)}$. These equations can be written in terms of the electrical conductivity, $\sigma_{(f)}$ ($S.m^{-1}$) which is the inverse of the electrical resistivity.

A very first relationship to study the effect of temperature on electrical resistivity was presented by Dakhnov (1962) as:

$$\rho_w = \frac{\rho_{w_0}}{1+\alpha(T-T_0)} \quad (2)$$

where ρ_w is the resistivity of water at temperature T and ρ_{w0} is the resistivity of water at the initial temperature T_0, and α is the temperature coefficient of resistivity (see Iravani et al, 2020a, for instance). We used equation (2) to obtain α value for our experiments using thermocouples, with a reference temperature of T_0=20°C, where T is the temperature measured by thermocouples, and ρ_{w0} is the averaged measured resistivity at 20°C. This linear relationship was defined for water but can be used for DNAPL mixtures (Iravani *et al.*, 2020a).

In the literature, no universal model has been presented with the consideration of the physico-chemical properties of each phase in a multiphase system in regard to the frequency changes. The lack of a developed comprehensive model on this subject steers researchers to fit the experimental data of complex resistivity with the phenomenological models. One of the most used phenomenological models is presented by Cole and Cole (1941) and was derived from the Debye (1929) model, which was firstly developed for relative permittivity (Pelton *et al.* 1978).

Pelton *et al.* (1978) proposed a model for SIP spectra that exchanged relative permittivity with electrical complex resistivity for practical applications that describes

$$\rho(\omega) = \rho_0 \left[1 - m(1 - \frac{1}{1+(i\omega\tau)^c})\right] \tag{3}$$

where ρ_0 is the low-frequency resistivity, c is the so-called Cole-Cole exponent, *m* is the chargeability, and τ is the mean relaxation time (*m* and c can be in the range of 0 and 1). If c equals 1 this equation will be similar to the Debye model. For this study, the Cole-Cole model was chosen because it has been demonstrated to be a good model to represent empirically the polarization of single grains or pores (e.g., Leroy *et al.* 2008; Niu & Revil 2016).

In this study for saturation prediction, the generalized Archie's law, which is developed by Glover (2010) based on the classical Archie's law (Archie, 1942), was used. It is defined as below

Archie's law $$\sigma_{mixed} = \sigma_{(f)}\Phi^m \tag{4}$$

and

Generalized Archie's law $$\sigma_{mixed} = \sum_{i=1}^{n} \sigma_i \Phi_i^{m_i} \tag{5}$$

where $\sigma_{(f)}$ (S/m) is the conductivity of the liquid inside the pores, Φ is the porosity, *i* is the indication of each phase and m is the cementation exponent.

In this study, we validate mixing models and calibration curves obtained from the previous study in cells and columns (Iravani *et al.*, 2020a, 2020b) using a 2D two-phase flow and comparing the results with imaging for validation. In a previous study, Iravani *et al*. (2020a) showed both resistivity and phase are related to the temperature variation in 1D columns. They found a linear relationship between temperature and resistivity. By using the Cole-Cole model they also found that temperature affects the frequency domain. Iravani et al. (2020b) studied the saturation impact on the magnitude and phase of electrical complex resistivity. They reported the generalized Archie's law was applicable to study the multiphase medium contaminated by CT. They found that saturation change has a direct impact on electrical resistivity.

Material and methods

A laboratory-scale experiment was carried out to study temperature and saturation effects on complex electrical resistivity during the drainage and imbibition of DNAPLs in a 2D tank. The experiment was done in a saturated porous medium with an immiscible liquid pair (CT and water). In the following, we describe the essential points, experimental setup, and procedures used for this experiment.

2D tank setup

The 2D tank has dimensions of 30 cm × 50 cm × 7 cm (height, width, depth). A schematic form of the front, back, and top of the tank is shown in Figure 1. The body of the 2D tank is made of a solvent resistant material called polyvinylidene fluoride (PVDF). The front wall of the tank with a thickness of 0.8 cm is made of a transparent glass rectangle through which pictures are taken at each stage.

A stainless-steel heating element was inserted inside the left-side of the tank to produce the non-isothermal condition. Fourteen type K thermocouples (green circles) record temperatures in the tank. The thermocouples were connected to a Campbell CR-1000 acquisition system. The temperatures were measured with sampling intervals of 30 s. For the

SIP measurements (Figure 1c), black and blue circles represent 30 current and 30 potential electrodes, respectively, 5 cm apart from each other. The electrical AC is injected between two stainless steel current electrodes (diameter=65 mm) made of nickel-cobalt alloy (MP35N) and the response voltage is measured between two handmade non-polarizable potential electrodes ($Cu/CuSO_4$) with a diameter of 1 cm. Non-polarizable electrodes are used in SIP studies to measure the voltage response of an injected current in a medium (e.g., Kremer *et al.* 2016a; Abdulsamad *et al.*, 2016). In non-polarizable electrodes, current can freely pass without polarization. Among common electrode solutions, $Cu/CuSO_4$ was chosen based on the method proposed by Maineult, Bernabé and Ackerer (2004). The solution was made using a mixture of 72.75 g of milli-Q water (ultrapure water), 26 g of $CuSO_4$, and 1.25 g of gelatin in 100 g of solution. A heating shaker was used to mix the solution for one hour at 80°C.

The non-polarizable electrodes were placed outside the samples to avoid inflow interference that in turn can interfere with electrical current signals (Maineult *et al.*, 2004). A porous ceramic (Ameteksi scientific instruments) was used in the interface of non-polarizable electrodes and the saturated porous media. Each non-polarizable electrode was connected to the SIP-lab IV (manufactured by Radic Research) with a shielded cable to avoid contamination of the data due to electromagnetic coupling (Maineult *et al.*, 2004; Bagotsky, 2005).

Glass beads (GB) made of silica with a diameter of 1 mm, forming a pack with a porosity of 40% ± 2%, were used as a porous medium. Different GB sizes were tested and this size was chosen to simulate a porous medium in the lab with close physical properties to field soil (e.g., the grains size and the porosity of the medium). The tap water was degassed using an ultrasonic tank (VWR Ultrasonic Cleaner - USC500D). For obtaining temperature variation effects in a non-isothermal condition, an immersion heater heats from the left side of the tank, and heat transfers to the right side. In non-isothermal conditions, there is a possibility of studying the temperature and saturation changes at the same time based on how temperature varies across the different halves of the tank and how saturation varies during imbibition from the bottom middle of the tank.

Water is injected using five pipes located at the bottom of the tank with the possibility of cutting fluid currents by using clips and clamps. The pipes are numbered and connected to a peristaltic pump (Watson Marlow 530U). The tank is filled step by step by first filling 5 cm bottom layer of well spread GB and then water was injected until the porous medium was fully saturated. This procedure should be carried out with great patience to avoid any air bubbles being captured inside the porous network. These bubbles, especially when they are connected to electrodes or thermocouples, can cause significant measurement errors. This operation is repeated step by step until reaching the top of the 2D tank and when all electrodes were buried by GB and water (almost 27.5 cm). A camera is placed on a fixed tripod in front of the 2D tank. Photos were taken during increasing CT levels and imbibition at regular time intervals of three and one minutes, respectively.

Previous experiments using imaging, time-domain reflectometry (TDR), and mass balance analysis showed a 2D flow behavior (Colombano *et al.*, 2021). However, the character of the electric field can be in 3D, and therefore, our numerical model uses a 3D geometry (with a small thickness) to simulate electrical fields.

Experimental and measurement procedure

Twenty-seven SIP measurements of an equatorial dipole-dipole (EDD) array were performed to resolve the variations of the electrical resistivity within the 2D tank. The conventional location of measurement for an EDD array is in the middle of the rectangles in Figure 1c. The exact place of measurements with the EDD array is illustrated in Figure 2 with blue circles and the number of measurements. Before and after each experiment, measurements were made with tap water to examine the performance and accuracy of electrodes and calculate the geometric factors (K). The geometric factor for water is easy to obtain because the resistivity of water is homogeneous and can be measured by a portable conductivity meter and the resistance by SIP lab. Then we calculated the K factor from resistivity over resistance. To validate the work, the measured geometric factors should be close to each other. According to our experience, the non-similarity of the geometrical factors was mainly related to non-polarizable electrodes or their surroundings. A blank test (with water only) was done for at least 24 hours to have enough data to increase measurement accuracy. The measurement readings may have serious errors, because of air trapping inside the polarizable electrodes. With a simple test with only water, these deficit electrodes can be replaced. In the next step, the experimental geometric factors (K) must be compared with ones that will be estimated by a numerical model for all 27 SIP measurements.

After creating a stable porous medium saturated with water, CT was injected from the bottom of the 2D tank (using the same points of entry where water was injected). For each step of increasing DNAPL content, measurements were made until the DNAPL has been reached to the middle of the 2D tank at 15 cm. This procedure was repeated three times to obtain a horizontal interface between CT and water in the middle of the tank. In the experimental process, obtaining a perfect flat boundary with CT was not easy due to fingering and dissimilar distribution in the different parts of the 2D tank. After stabilizing the experimental setup with the portion of half GB+water and the other half GB+CT, SIP measurements were made before heating, in the frequency range of 0.183 Hz to 20 kHz. Resistivity and temperature were measured at all levels in the 2D tank.

After heating the tank from the left side and diffusing the heat to the right side of the tank, our experimental protocol was done for drainage and imbibition of water where SIP measurements were conducted at each step. The heating time takes almost 12 hours to have a stable temperature in each thermocouple (measurement zone) of the tank. When the recorded temperature was stable, the pumping of CT from the bottom middle of the tank was started.

Due to the rapid imbibition, it was not possible to measure more than two frequencies during this dynamic stage of the experiments. Therefore, during the imbibition period, the SIP spectra were recorded in two frequencies (1.46 Hz and 2.92 Hz), close to the ones used commonly in the field scale. The resistivity calculations were, therefore, based on a single-frequency approach.

Iravani *et al.* (2020a) reported the α values (equation (2)) for the same liquids (water and CT) with and without GB in 1D cells and columns. From their comparison between the α value of water with other studies, normally the α value should be almost the same. We calculated this value again using our 2D tank experimental data to be sure it was the same in this new 2D configuration. In three levels of measurements, the top level is filled with GB+water and the bottom level with GB+CT. Figure 3 is the linear regressions of the resistivity data with the temperature where the y-axis is the ratio of the resistivity at reference temperature (T_0=20°C) and resistivity at T (ρ_0/ρ) minus 1, and the x-axis is the temperature difference ($T - T_0$). The slopes of the linear regression of the 9 measurements at the top with GB+water and the bottom GB+CT are presented in Figure 3a and Figure 3b. The obtained regression is in accordance with the previous finding by Iravani *et al.* 2020a,b. Hayashi (2004) studied the linear relationship between electrical conductivity and temperature that was usually used in previous studies on water with different salinity and chemical substances. Regardless of the compositional and structural differences, the relationship between these two parameters of all different water samples was linear. He suggested being more cautious if temperature values are dissimilar to the reference temperature of 25 °C. The α values for water-saturated GB (0.052) and CT saturated GB (0.075) are the same as what we found in columns (Iravani *et al.*, 2020a) but the coefficients of determination are less (GB+water 13.09% and GB+CT 3.28%) than columns for both.

According to Iravani et al. (2020b), the final equations for correcting resistivity for both multiphase systems are:

$$\rho_{(GB+water)}= \rho_{0(GB+water)}/(1 + 0.052(T - 20) \quad (6)$$

$$\rho_{\,(GB+CT)}= \rho_{0(GB+CT)}/(1 + 0.075(T - 20) \quad (7)$$

After validating α values in two different geometries and despite the complexity of CT components, these two equations can be used in our 2D tank for temperature correction of electrical resistivity in the multiphase system of GB+CT and GB+water. The porous medium in the tank was subjected to heating with an immersion heater. The temperature fields (back view) in the tank before and after imbibition are shown in Figure 4. We used Surfer software to interpolate the data. Surfer interpolated the temperature and the corrected resistivity in the range of 110 to 100,000 **Ω**.m with the Kriging method. In this study, for all data fitted with Surfer, the same griding methods as Kriging were used with the experimental variograms fitted with linear component type and no trend removal. The temperature diffuses faster in the water at the top due to its higher thermal conductivity. The temperature close to the heater is almost 50°C and due to heat loss, it decreases across the tank to room temperature (20°C).

Experimental electrical resistivity value was measured and corrected with temperature for three testing EDD array levels. In the scatter plots of Figure 4, each point corresponds to a mean value of a square using an EDD array. Before imbibition, the first level of EDD at the bottom of the tank had almost the same resistivity. The decreasing resistivity in the middle part of the tank after imbibition suits the CT saturation decreasing and replacing DNAPL with water in this part. The points in the boundary of the tank were considered as same as their neighbor points for interpolation. Our study of temperature and saturation effects has been done separately for the condition that both factors want to be considered. After correcting the data for the temperature effect, the impact of saturation changes could be determined. The next step was pumping CT out from the middle bottom of the tank. Performing the imbibition step was to simulate the thermally enhanced pump and treat technology for soil remediation. The objective was to monitor how water and CT saturation vary inside the 2D tank using the electrical resistivity measurements.

Validation of the electrical resistivity measurements

Before starting the main drainage-imbibition experiment, to validate the performance of the electrodes, the first experiment was carried out by increasing the CT level in the tank at the normal room temperature (20°C). In the following, this experiment's data will be compared to the results obtained from a numerical model. After filling the half bottom of the tank with GB+CT, the resistivity was measured for 24 hours, then the temperature of the 2D tank was increased to 30 and 40°C by circulating hot water at a given temperature around the 2D tank to keep the temperature constant in all 2D tank parts. Compared to heating elements, heating with this method can result in temperature homogeneity throughout the whole tank.

For the main experiment, a laboratory protocol of a multiphase system was defined in the 2D tank that will be briefly described in the following. The 2D tank was filled with GB and two different liquids, water as a conductive fluid and CT with low conductivity. At the starting point, the tank was filled with GB and water layer by layer (Figure 5a). Figure 5b is the tank filled with GB and water, ready for injecting CT from the 5 points at the bottom of the tank (Figure 5c). It is necessary to slowly inject CT to have the relatively horizontal CT levels (Figure 5d). Despite all these considerations, in one case (Figure 5e) when a bump shape CT surface appears, slight injecting from the sides of the tank can balance the surface of CT in the tank.

We used an open-source modeling tool BERT (Boundless Electrical Resistivity Tomography) developed by Günther and Rücker (2015) to model this simple drainage experiment. The possibility of modeling closed geometries as laboratory cells and tanks encouraged us to use this modeling code. The theory of developing this code based on the finite element method has been expanded from the previous study done by Rücker, Günther and Spitzer (2006). In the other studies, code was developed for arbitrary electrode shapes (Rücker and Günther, 2011) and long electrodes (Ronczka, Rücker and Günther, 2015).

The 2D tank, by using three series of EDD arrays in different levels, was modeled by BERT. The fundamental physical theory for electrical resistivity used in BERT software is based on Ohm's law (Günther et al., 2006). Assuming that the electric fields are conservative and the magnetic effects are negligible, the mathematical equations can be written as follow

$$\mathbf{J} = \sigma\mathbf{E};\ \mathbf{E} = -\boldsymbol{\nabla}V, \tag{8}$$

where $\mathbf{J}$ ($A.m^{-2}$) is the current density, $\mathbf{E}$ ($V.m^{-1}$) is the electric field intensity, σ ($S.m^{-1}$) is the electrical conductivity (the reciprocal of resistivity ρ), and V (V) is the electric potential (primary variable). The initial condition was ρ(t=0)=30 Ohm.m that corresponds to the electrical resistivity of glass beads saturated by water. Neumann boundary condition (no-current flow boundary condition) was imposed on all 2D tank borders. The number of irregularly structured meshes was 1036942 elements.

In the other experiment in the 2D tank, increasing CT level is also simulated centimeter by centimeter from the bottom of the 2D tank up to 18.0 cm from the 2D tank bottom. Before these steps, we simulate a homogeneous tank filled with only GB and water.

After injecting CT, the resistivity values were recorded for these different levels, until reaching the top of the middle EDD level (17.5 cm). This data allows us to study how resistivity varies in the experiment and the model according to an increase in CT level.

Results and discussions

Correction of data with temperature

In remediating polluted sites with the thermal enhanced method, the effect of temperature on the electrical resistivity should be corrected before studying the effect of saturation change on geophysical parameters. At the beginning of the experiment, the mean temperature in the tank was 20°C. The temperature varied between 19.1 and 22.8°C. The resistivity was corrected with temperature by using the relation proposed by Dakhnov (1962) and Keller and Frischknecht (1966) to have a geophysical parameter that is only controlled by saturation.

Electrical resistivity fields

The electrical resistivity variation was used to predict the water/CT saturation change. The evolution of logarithmic apparent and inverted electrical resistivity (to facilitate the comparison before and after temperature correction) during the imbibition test is shown in Figure 6. The objective is to show how temperature changes can impact the calculation of resistivity. Iravani et al. (2020a) presented how the temperature affects the resistivity value in a multiphase sample. To study the effect of saturation change due to pumping, it is needed to deduct the effect of the heating. Figure 6a,b show the log of resistivity before temperature correction, and Figure 6c,d show the same results with temperature correction. This is the starting point of our study on the saturation effect on resistivity values in the 2D tank. Figure 6 is cut 10 cm from the left side of the tank to eliminate the influence of the metallic heater on the resistivity of the medium.

Validation of the setup configuration and electrical resistivity measurements using BERT modeling

In modeling a physical phenomenon, the main constraint for a modeler is dealing with boundary conditions (Stroud, 2006). The first boundary problems are the Neumann boundary conditions (Broggini et al., 2017). We need a dipolar current injection to avoid overflow from the boundaries of the 2D tank (Günther and Rücker, 2013). Using a reference electrode inside the tank could solve this problem. This reference electrode should be placed slightly away from the current and potential electrodes inside the tank. The second problem that arises for Neumann domains is the non-uniqueness of the solution of the partial differential equation due to a lack of calibration (Günther and Rücker, 2013). We used a calibration electrode to overcome this problem, which must be placed at the boundaries of the tank. The potential at this point (electrode) must be zero. The method used in the modeling was moving both electrodes (reference and calibration electrodes) in the tank and checking if the results obtained were consistent or not (Günther and Rücker, 2013).

To validate our experimental measurements with numerical modeling, we completed two steps: In the first step, the ratio of geometrical factors K obtained from the experiment with water on the 2D tank and model were compared in Figure 7. In both the experiment and model, the K varies around 0.45 to 0.5 for the EDD arrays with ±20% error. We noticed that in points close to the metal grid on both sides of the tank (e.g., points 23, 24, and 25) the difference in K factors obtained from the model and experiment are more significant because, in the model, the effects of metal grids have not been considered.

In Figure 8, after increasing the level from 2 cm (entering the influence zone of EDD measurements) resistivity was increased both in the experiment and model. Since the influence zone of all measurements begins at 2.5 cm. In this figure, the model simulation results properly coincide with the experimental data (Pearson's correlation coefficient, R=0.981). This validates the setup configuration and shows that the electrodes measure the resistivity with good precision. In the transaction part at a height of 3 and 4 cm, an overestimation was seen compared to the experimental resistivity data. This overestimation could be due to the effect of increasing CT and its direct effect on the resistivity value predicted by the model.

Phase variation during imbibition

During the imbibition period, both amplitude and phase shift of complex resistivity measured in the middle of the tank were affected by temperature and saturation changes. Figure 9 shows how phase shift varied with time in the pumping influence zone in the middle of the tank for two different frequencies (*f*=1.46 and 2.92 Hz). Nine measuring zones (we call them points in the following) in the same vertical positions were chosen from three levels of the EDD array in the middle of the tank.

Points 12, 15, and 18 are at the top, consisting of the water-saturated zone (GB+water). The results showed that the phase shift is close to zero in these three points. These results confirm previous findings on 1D cells and columns for the water-saturated cases (Iravani *et al.*, 2020b). Points 11, 14, 17, and 10, 13, 16 are at the middle and bottom of the tank with an EDD array, respectively. During the imbibition (pumping process), CT was pumping from the middle bottom of the tank, and therefore, water replaced CT forming the cone of depression. As shown in Figure 9, imbibition increased and then decreased phase shift at points 11, 14, and 17 (at the middle of the tank), but it was constant at points 10, 13, and 16 (at the bottom of the tank). This variation occurred sooner at point 14 than at point 11, which corresponds with our expectation because imbibition firstly affected the middle points, then the side points because of the cone of depression. Besides, the phase variations at points 14 and 17 happened at the same time. Drops in phase shift at the end of imbibition happened later for points 11 and 17 compared to 14 due to the delay of the imbibition effect from the middle of the tank reaching the sides. After imbibition (dominating water saturation), the phase shift dropped to a value close to zero. An increase in phase was observed in all 3 points in the middle of the tank, which can show the complex behavior of the spectra and phase due to the instantaneous change of water/CT saturation. However, we observed a weak response to imbibition tests for points 10, 13, and 16 situated at the bottom of the tank because they were not influenced by imbibition.

After dominating water in each block of the measurement, the recorded values became stable and close to zero. Comparing results of the phase change between Figure 9a and Figure 9b shows that small changes in frequency have a large effect on phase shift in layers containing DNAPL. The higher frequency, the higher the phase shift.

Predicting saturation in 2D tank

In a previous study, Iravani *et al.* (2020b) found a relation between saturation state and the amplitude and phase shift of the SIP spectra for CT+water, to have a calibration relationship for further uses in the 2D tank (current study) and even thereafter in field studies as below (Iravani *et al.*, 2020b):

$$\sigma_{mixed} = 0.0345\theta_w{}^{1.8} + 10^{-3}(0.4 - \theta_w)^{5.2} + 10^{-14}(0.6)^{2.34} \tag{9}$$

σ_{mixed} was obtained from the inverse relationship with the measured resistivity of the multiphase system in each zone. The only variable in equation (9) is water content (θ_w), which can be present as water saturation after using this equation ($S_w = \theta_w/\Phi$).

Figure 10a,b, and Fig 10c,d show the saturation status and front image of the multiphase system before and after imbibition in the 2D tank, respectively. In Figure 10c,d, the strong reduction of DNAPL saturation in the middle of the 2D tank is due to imbibition. The complex nature of CT that sticks into the 2D tank glass wall surface did not allow image analysis to obtain the saturation. Therefore, we could not make a quantitative comparison of the saturation using these images.

Figure 10 shows resistivity measurements in different influence zones with interpolation that can give us the saturation field. In Figure 10c, we expected more saturation changes before and after the pumping at the left side of the tank compared to its right side. The heating from the left side has a direct effect on the viscosity of CT and consequently, the pumping rate, but the DNAPL/water interface is not horizontal and we could not quantify and make this assumption. As observed in Figure 6d and Figure 10c, the effect of imbibition seems to be well observed particularly in both sides compared to the middle.

The CT saturation was not exactly 100% even before starting imbibition because some percentage of water was captured in the pore spaces before injecting CT. It seems the Archie model does not have enough precision (the effect of surface conductivity should not be neglected) in very high resistivity (high CT saturation) and that monitoring in a small 2D tank does not allow us to capture the cone of depression with high precision for imbibition.

Effect of temperature and saturation on Cole-Cole parameters

In the non-isothermal condition before starting imbibition when the bottom half of the tank was filled with GB+CT, the complex resistivity was measured in the frequency domain (e.g., under electrical impedance conditions) before and after heating. The tank was heated from the left side by a heater and heat was dispersed from the left side of the tank to the right. The most influenced part is close to the heater. There the temperature changed from 20 to 50°C, and that change affects two EDD measurements at the far-left side of the tank (No. 1 and 4). Figure 11 shows how the four Pelton parameters vary at 20 and 50°C. The experimental data were fitted to the Pelton model by using a Matlab code by Maineult *et al.* (2017). The fitting curves of the experimental data and model are presented in Figure 12.

We have already studied how these parameters vary with temperature in cells and columns (Iravani *et al.*, 2020a). Studying these parameters in the 2D tank showed that ρ_0 and log (τ) were decreasing with temperature increases. Raising the temperature from 20 to 50°C slightly increased c but both m and c were so close to 1 that it can be considered as a constant value. The fact that c is close to 1 means that the SIP response obeys a Debye model, and the fact that m is close to 1 means that the medium is highly chargeable because of poor dispersion of the spectra. These variations in results were the same for both measurements (1 and 4). Our findings on how the four parameters changed in the 2D tank confirm the previous findings on cells and columns with this difference: m was 0.89 and in the 2D tank for both measurements was almost 1.

Conclusions

The main goal of this study was to move one step closer to execute the usage of electro-geophysical methods to monitor the saturation changes in soils contaminated by a DNAPL (like CT) under non-isothermal conditions. In particular, applications of the SIP method in contaminated porous media were examined experimentally in the laboratory by using a 2D sample tank. Glass beads of 1 mm diameter were chosen because of similar physical properties to field soil. This allows to model as much as possible actual field conditions in the laboratory.

After preparing the non-isothermal condition by heating from the left side of the tank and correcting the temperature effect, we studied how the electrical complex resistivity varies during imbibition. Changes in the electrical complex resistivity magnitude were measured and the saturation field was obtained from the resistivity data. From the electrical resistivity measurements, we predicted the changes of the saturation field before and after imbibition accordingly. However, the proposed electrical resistivity monitoring did not produce the cone of depression during imbibition with high precision. This may be related to CT's very high resistivity and the small laboratory setup. The phase changes in 9 points in the middle of the tank in all three levels showed that the phase significantly decreases after increasing water saturation (imbibition period).

It is concluded that the temperature (5 to 7%) and saturation change due to replacing CT with water affects measured electrical complex resistivity. The effect of saturation change is much more considerable compared to the temperature. In the coupling of temperature and saturation variation after correcting resistivity with temperature (with a linear relation), the saturation effect will be the only variable in the real case with remediation surveys under the thermal-enhanced technique.

The effect of saturation change by injecting CT from the bottom of the tank was evaluated by BERT as a numerical modeling tool. The results show that the measurements with EDD arrays have been properly fitted by the BERT model. This validates our setup configuration and confirms that our electrodes functioned properly. The goal of our study was not to present comprehensive mechanistic modeling of the results but to present the experiment and the results that show the effect of heating and pumping on the SIP response, and its interest for field measurements. Developing a mechanistic model is a step beyond, the subject of future research.

The reported results showed that the proposed generalized Archie's law from the previous study could predict the saturation. However, during pumping at the bottom of the tank, the proposed coefficient did not perfectly reflect the saturation change. Finding the proper coefficients of generalized Archie's law can lead to precise saturation estimation in both high and low resistivity. These results at the bottom level during imbibition could be due to placing the current electrodes in the part that is saturated with CT. Our previous finding in 1D cells supports also the idea that in a case where the current electrodes are located in a saturated DNAPL part, SIP reports very high resistivity although the potential electrodes are still placed in the water-saturated part.

How Cole-Cole parameters change before and after heating were evaluated for 20 and 50°C before and after heating. The results of this study show that Cole-Cole parameter variations are in accordance with our previous findings.

We believe that the most outstanding part of our findings is the development of knowledge on how to monitor imbibition processes where both temperature and saturation change together in a 2D system. Validating our results in electro-geophysical field studies would advance their reliability in various environmental conditions. Our findings might be used for soils with similar physical properties.

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FIGURE LEGENDS

Figure 1 Schematic layout of the a) front, b) top and, c) back of the 2D tank with the description of each element. In the front view of the tank, a double glazed transparent glass is used as a wall to better control the temperature and imaging purpose. Isothermal system includes a system of heating all around the tank using hot water. A stainless-steel heating element was inserted inside the left-side of the tank to produce the non-isothermal condition. In sub-figure c) the red square showed an EDD measurement. There are 27 EDD measurements in three different levels.

Figure 2 Equatorial dipole-dipole arrays from the front side of the tank. 10 current (metal) electrodes and 10 potential (non-polarized) electrodes in each level can record 9 EDD measurements. In three levels of measurements, 27 measurements can be recorded. Using other arrays (e.g., Wenner Beta) is also possible between two levels.

Figure 3 Linear regression to find the α values for a) GB+water in the top level and b) GB+CT for the bottom level of EDD array. The obtained α values for GB+water and GB+CT are in accordance with the results of the previous study on 1D columns (Iravani et al., 2020a).

Figure 4 Back view of the temperature field in the tank under non-isothermal conditions a) before, and b) after imbibition (the plus (+) signs are the positions of the thermocouples and red dot is the pumping point at the middle bottom of the tank). A significant temperature (54 and 18 °C) difference between the right and the left side of the tank is due to using a heater at the right side of the tank (in the back view).

Figure 5 Different steps of experimenting on the 2D tank. a) filling the tank with GB and water, b) tank packed with GB and fully saturated by water, c) injecting CT from 5 points at the bottom of the tank (for the places of injecting points, please see Figure 1a), d) increasing step by step the level of the CT in the tank, e) possible bump shape of the CT surface – the flat interface is ideal, f) almost half of the tank filled with GB+CT. This stage is the starting point of the imbibition experiment.

Figure 6 Log of the resistivity field in the tank a) before imbibition, b) after imbibition without temperature correction, and c) before and d) after imbibition with temperature correction, e) inverted log resistivity before temperature correction, and f) inverted log resistivity after temperature correction (the circle (●) and blue arrow signs are respectively the positions of the EDD measurements and drainage points).

Figure 7 Experimental and model ratio of K factor for 27 measurements of electrical resistivity with EDD array. Point values closer to 1 show a better model prediction of the experimental data. Points far from the walls have better results because of the metallic grid used in the side walls.

Figure 8 Variation of resistivity as a function of CT height for EDD array at the bottom level of the tank. These are apparent resistivity obtained from the average values of the 9 measurements at the bottom level of the 2D tank (measurements No: 1, 4, 7, 10, 13, 16, 19, 22, and 25; see Figure 2).

Figure 9 Variation of phase shift during imbibition period at the middle of the tank in 9 points (12, 15, and 18 at the top level, 11, 14, and 17 at the middle level, and 10, 13, and 16 at the bottom level) for the frequencies of a) 1.46 Hz and b) 2.92 Hz.

Figure 10 a) The measured CT saturation field before imbibition, b) front image of the 2D tank before imbibition, c) the measured CT saturation field after imbibition, and d) front image of the 2D tank after imbibition. The black lines in “a” and “c” are the counters between two areas (GB+water and GB+CT).

Figure 11 Effect of temperature on Cole-Cole parameters (20 and 50°C) close to the heater. As it was shown in Figure 5, just points close to the heater have recorded the temperature around 50 °C.

Figure 12 Fitting curves of experimental data and model for a) point 1 at 20°C, b) point 1 at 50°C, c) point 4 at 20°C, and d) point 4 at 50°C. For more information about choosing the presented points, see Figure 11 description).

Data Availability Statements

The datasets generated and analyzed during the current study are available from the corresponding author on reasonable request.

Declaration of interests

☒ The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

☐The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: