

Numerical and Experimental Investigation of Steam Stripping of Lnapl-Contaminated Soils

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ABSTRACT

Steam injection is an example of thermal remediation technology which involves the injection of steam into the sub-surface and simultaneous recovery of fluids from extraction wells. The injected steam heats the soil and created a steam zone that expands from the injection wells as more steam is injected. The main objective of the research work was to carry out numerical modeling and laboratory experiments in order to evaluate the effectiveness of steam injection for remediation of soils contaminated with Light Non Aqueous Phase Liquids (LNAPLs). MATLAB (Matrix Laboratory) package was used for the numerical modeling while the experimental aspect involved the injection of dry steam produced from a boiler into a sand column which contain a saturated mixture of LNAPL contaminant (i.e. Kerosene) and sample of sand (two samples of sand were used; fine and coarse grain sand). As steam was introduced into the column, the viscosity of the contaminant reduces due to increase in temperature which makes the contaminant to flow and was recovered at the base of the set-up over time. It was discovered that increase in pressure and the soil grain size increases the LNAPL flow rate and reduces the time of treatment. Also the sand grain size affect the recovery rate and efficiency as well as the time of treatment for possible clean up to be achieved. Steam injection was found to be about 80-85% efficient in the removal of LNAPL contaminants from contaminated soil. The result has therefore demonstrated the effectiveness of steam injection process as a thermal soil remediation technique.

Keywords: Contaminated soil, Effectiveness, Kerosene, LNAPL, Steam Injection, time of treatment,

I. Introduction

Industrial activities during the last five to six decades have created a large number of sites where chemical spills have contaminated the subsurface. Many sites in which industrial activities take place in Nigeria are contaminated due to accidental surface spills, intentional dumping at disposal sites, or leakages from underground storage tanks and landfills. These contaminants include chlorinated solvents, gasoline or fuel, coal tar and creosote. These contaminants are almost immiscible with water and will often be present as non aqueous phase liquids (NAPL). Non Aqueous Phase Liquids (NAPLs) are hazardous organic liquids such as dry cleaning fluids, fuel oil, and gasoline that do not dissolve in water, of which there are two classes: light NAPLs (LNAPL) and Dense NAPLs (DNAPLs). LNAPL is one of a group of organic substances such as gasoline, fuel oil and petroleum chemicals (mainly benzene, toluene, xylene and benzene derivatives) that are relatively insoluble in water and are less dense than water. LNAPLs such as oil, tend to spread across the surface of the water table and form a layer on top of the water table. Dense NAPLs (DNAPLs), such as the common solvent trichloroethylene, other DNAPLs include

coal tars, which contain PAHs (Polycyclic Aromatic Hydrocarbons), and transformer oil, which may include mixtures of PCBs (Polychlorinated Biphenyls). DNAPL is one of a group of organic substances that are relatively insoluble in water and are denser than water. DNAPLs tend to sink vertically through sand and gravel aquifers to the underlying layer. A significant portion of contaminated soil and groundwater sites contain NAPLs, and they are particularly hard to remove from the water supply. NAPLs are always associated with human activities and cause severe environmental and health hazards. As a result of the risk of human exposure to these chemicals, it maybe necessary to remediate a contaminated site. This is most often done by excavating the contaminated soil and treating it off site. However, at some site the volume of contaminated soil might be too large or there might be buildings that make it impossible to excavate, therefore remediation has to take place in-situ without disturbing the buildings in the environment. In-situ remediation technique seeks to treat the contamination without removing the soil vapor extraction in the unsaturated zone. Unfortunately, these technologies have shown to be very inefficient at NAPL sites. The mass transfer rate

from the heterogeneously distributed NAPL becomes diffusion-limited and large volume needed to be flushed to achieve clean-up. At heavily contaminated sites, the clean-up maybe in order of decades when applying these technologies. Steam injection is the most commonly used thermal technology during which the contaminated soil is heated and this strongly affects the physical-chemical properties of the contaminants in most cases to the benefit of the recovery process.

Some of the knowledge and technique developed in petroleum engineering for enhanced oil recovery by steam injection are useful to the problem of steam stripping for remediation of LNAPL contaminated soils. Steam injection also termed steam enhanced extraction was initially developed by the petroleum industry for enhancing oil recovery and was more recently been adapted to remediate soil and aquifers [1] [2]. It has been applied in unsaturated as well as saturated zone [3] and is generally more efficient in porous media such as sand [4]. However, there is a distinct difference between enhanced oil recovery and ground water remediation. In enhanced oil recovery, the objective is to remove the maximum amount of oil from the reservoir for a long as it is economically feasible. Small amount of oil left in the formation are usually ignored. In contrast, the purpose of remediation efforts is to remove as much of the contaminants as possible until possible clean up level are achieved.

Steam injection is rather new and very promising technology for remediating subsurface hydrocarbon contamination convective technologies like pump-and-treat and soil vapour extraction have shown to be inadequate in many cases, in particular for highly heterogeneous conditions. Very often these techniques have failed to reach the desired clean up level [5] and therefore new technologies are needed to remediate the large number of contaminated sites. Several one dimensional laboratory experiments have shown that steam can efficiently remove volatile or semi-volatile contaminants in any concentration from both saturated and unsaturated porous media [6]. Two dimensional studies have shown that also heterogeneous porous media can be remediated by steam injection [7],[8]. Full scale operations have supported these results suggesting that the technique ensures rapid and satisfactory clean up of even very complicated contaminants [9],[10].

Steam injection is an example of thermal technology which involves the injection of steam into the sub-surface and simultaneous recovery of fluids from extraction wells. The injected steam heats the soil and created a steam zone that expands from the injection wells as more steam is injected. If the steam temperature is high enough to heat the contaminants up to its boiling point the contaminants will vaporize

and will be recovered as vapor, on the other hand if the boiling point is not reached, increase in the temperature of the soil reduces the viscosity of the contaminant and in so doing make the contaminants to flow in the phase and it is recovered as liquid contaminant from the soil. Early exploratory experiments with steam injection for soil remediation were carried out in the Netherlands by [11]. [12] performed laboratory experiments to study fundamental aspect and to demonstrate the feasibility of steam injection as an in-situ remediation technique. They found in some cases that only one pore volume of fluid had to be displaced by steam injection to achieve cleanup standards. Simplified measurements of recovery efficiency of kerosene in one-dimensional experiments to vacuum assisted steam stripping were conducted by [13], who also performed observations of two dimensional steam front movements.

Several mechanisms are responsible for LNAPL recovery by steam injection ([14],[15],[16],[17]) primary mechanisms include evaporation in steam zone, vaporization in the hot water zone, and large pressure gradients at the condensation front that result in increase in capillary number. The effectiveness of steam injection as a remediation technology technique depends on the ability to enhance these mechanisms. During steam injection, high volatile components having high vapor pressure and a boiling point below the steam condensation temperature vaporized as the steam front approaches. The LNAPL vapor is subjected to local gaseous-phase mass transfer mechanisms, which in a steam injection process dominated by the large convective flux of the steam, and the LNAPL is carried along as hydrocarbon component in the gas phase. Thus any generated vapors are advected toward the cooler region, where condensation of both the steam and the vaporous contaminant occurs. A bank of liquid distillate develops ahead of the condensation front, if contaminant is completely vaporized at a temperature less than the steam condensation temperature, complete removal of the LNAPL is possible. The LNAPL components remaining in the steam zone that do not completely vaporize (semi volatile components) evaporate at an enhanced rate due to increase temperature and increased liquid phase molar fraction [18]. In the oil recovery literature, the term steam distillation is commonly used to describe the two different phenomena, vaporization and evaporation. Hence, the primary recovery mechanism for LNAPLs is steam distillation. The third major mechanism is due to high pressure gradients that occur in the steam zone close to the condensation front and that facilitate displacement of LNAPL ganglia into the condensation zone, where they are transported by the liquid bank. Furthermore, the steam-water

thermodynamics equilibrium constant at the pore level in conjunction with the high steam water interfacial tension, produces an additional pressure increment at the upstream end of a ganglion extending through the stream condensation zone.

The main aim of the research work was to carry out numerical modeling and laboratory experiments in order to evaluate the effectiveness and efficiency of steam injection for ex-situ remediation of soil contaminated with Light Non Aqueous Phase Liquids (LNAPLs). The effect of various parameters were investigated on LNAPL recovery efficiency. These parameters include (1) Steam injection pressure, (2) Soil-grain-size distribution, and (3) time of treatment

II. Model Formulation

The following equations were formulated by

[19]

i. Permeability determination

$$k_o = \frac{Q\mu_o L}{A\Delta P} \dots \dots \dots (1)$$

Where

- k_o – permeability (Darcy)
- μ_o – viscosity (Cp)
- L – length of cell (m)
- A – cross sectional area (m²)
- Δp – pressure drop

ii. Hydraulic conductivity

$$K = \frac{k_o \rho g}{\mu_o} \dots \dots \dots (2)$$

- where K – hydraulic conductivity ($\frac{m}{s}$)
- k_o – permeability (m² = 1.01 × 10¹² darcy)
- ρ – fluid density (kg/m³)
- μ_o – fluid viscosity (pa)
- g – gravitational constant (9.8 m/s²)

iii. Critical flow rate determination

$$q_{oc} = \frac{\pi k_o h_o^2 g \Delta \rho}{\mu_o \ln \left(\frac{r_2}{r_1} \right)} \dots \dots \dots (3)$$

where

- q_{oc} – the critical flow rate
- k_o – LNAPL permeability
- h_o – the depth of the LNAPL pool
- $\Delta \rho$ – density difference between the LNAPL and the overlying ground water
- μ_o – the viscosity of the LNAPL
- r_1 – the radius of the well bore
- r_2 – the radius of the pool

The model predicts a particularly strong influence of the depth of the LNAPL pool on the critical LNAPL flow rate.

iv. Porosity determination

$$\phi = \frac{V_v}{V_b} \dots \dots \dots (4)$$

where

- ϕ – porosity
- V_v – volume of void space
- V_b – bulk volume (cross sectional area × length)
- v. Energy, E, needed to heat a volume, V, of soil to a temperature, T.**

$$E = V(T - T_i)(C_r \rho_r (1 - \phi) + \rho_w C_w S_w \phi) = V(T - T_i) \bar{\rho} \bar{c} \dots \dots \dots (5)$$

Where

- T_i – the initial temperature
- C_r – the heat capacity of the soil grains
- ρ_r – the density of the soil grains
- C_w – the heat capacity of water
- ρ_w – the density of water
- ϕ – the porosity
- S_w – the water saturation in the heated zone
- $\bar{\rho} \bar{c}$ – overall heat capacity

vi. The energy released when injecting steam into a porous media with the temperature T_i

$$E = m_s (H + T - T_i) C_w \dots \dots \dots (6)$$

Where

- m_s – the mass of steam
- H – enthalpy of vapourisation of water
- T – the steam temperature

vii. Yield = $\frac{\text{initial oil-produced oil}}{\text{initial oil}} \times 100\% \dots \dots \dots (7)$

- Initial oil i.e. quantity of oil mixed with the sand
- Produced oil i.e. quantity of oil recovered from the sand

viii. % LNAPL recovery efficiency = $\frac{\text{Total volume of LNAPL recovered}}{\text{initial volume of LNAPL in the soil}} \dots \dots \dots (8)$

ix. Steam flow rate = $\frac{\text{recovered volume of LNAPL}}{\text{time}} \dots \dots \dots (9)$

3.1 Numerical Simulation

Using MATLAB, we solve technical computing problems faster than with traditional programming languages such as C, C++, and FORTRAN. MATLAB is used to code the above listed governing equations for numerical simulation, a graphic user interface (GUI) is design for easy input of data's and output of results. Necessary relationships have been plotted on a graph then compared with the simulated values.

3.2 Experimental Approach

Laboratory-scale column experiments were designed to evaluate the mobilization and recovery of LNAPLs by steam flooding. Initial phases of the experimental approach included soil preparation and packing and evaluation of soil properties.

The experiment was carried out in the Fluid Mechanics/Thermodynamics Laboratory of Ladoke Akintola University of Technology, Ogbomosho, Oyo State in Nigeria.

In the present work, the selected soils are characterized by their grain-size distribution. The test soil types are poorly graded soils of uniform grain size and well-graded soils in which several grain sizes was mixed to obtain different grain size distribution slopes. The soil was carefully placed into the column to avoid any segregation, density variation, or channeling within the column. At both ends of the column, a layer of gravel was included for better filtration and to ensure uniform axial flow conditions. The direct method was used to measure the porosity. The volume of the void space (V_v) was determined by volumetric analysis following water flooding and bulk volume (V_b) inside the column was calculated from measurement of the cross sectional area and length of the soil column. The porosity was then determined simply as $n=V_v/V_b$.

Steam injection experiment was conducted in an instrumented 1-dimensional soil column- the column was 50cm long and 20cm in diameter and was placed uniformly with a soil of selected grain-size

distribution. Air-free saturated steam was passed from the steam boiler to the core face in insulated lines, steam traps was used to remove any condensate form in the lines. During steam injection, column effluent was passed through a condenser and was collected in a graduated cylinder in which LNAPL separated from water, allowing separated measurement of LNAPL and water quantities. Steam, at a selected pressure (and corresponding temperature) was then injected beyond the point of steam break through. The amount of LNAPL removed and axial temperature profile in the column was monitored periodically. Steam injection was stopped when the condensed liquid is free of LNAPL. The final LNAPL residual saturation and the corresponding LNAPL recovery efficiency was determined by mass balance. The residual Saturation is define as the fraction of void space occupied by LNAPL that could not be recovered by steam injection in additional pore volumes. The LNAPL recovery efficiency will be calculated as:

% LNAPL recovery efficiency = ratio of total volume of LNAPL recover to initial volume of LNAPL in the soil.

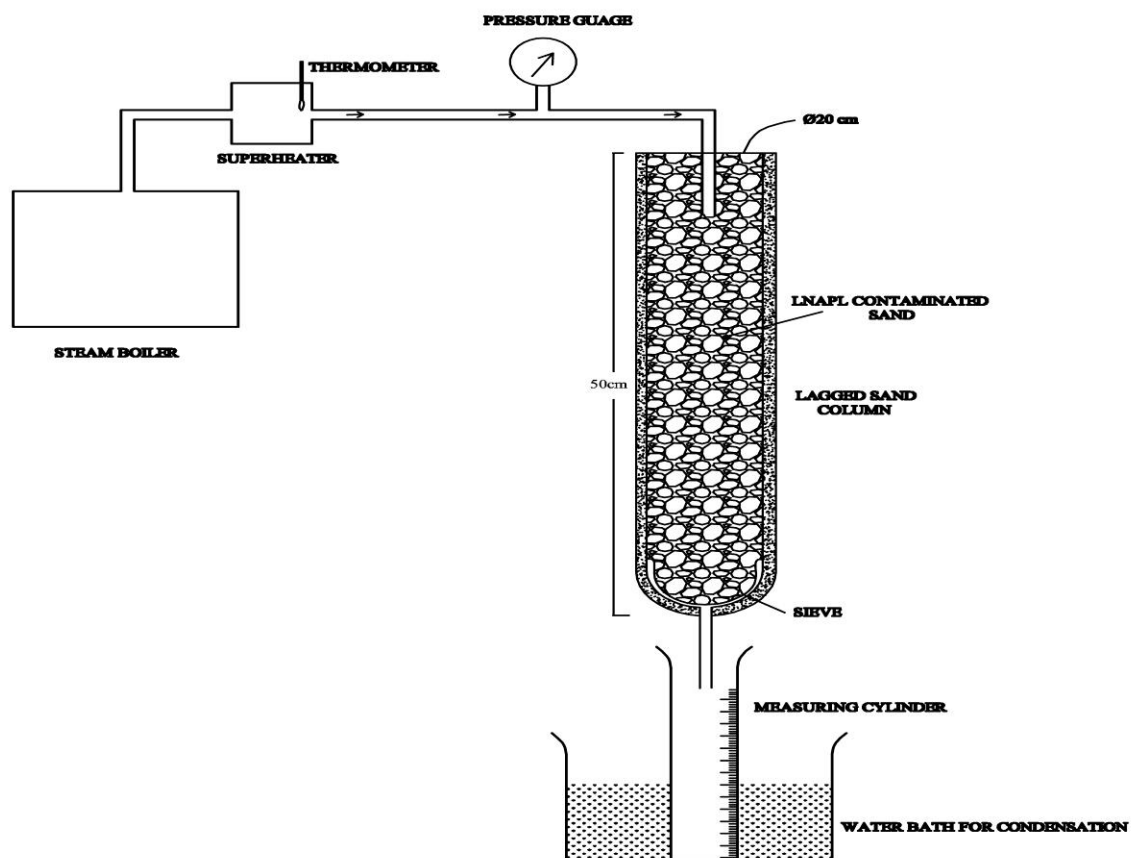


Fig 1. Experimental Set-Up for Steam Injection Process

III. Results and Discussion

The experimental data were compared with numerical simulation and the graphs of the experimental data were plotted and compared with the simulation graph. Higher steam injection pressure results in higher steam flow rate. The table 1 and 2 below show the effect of steam injection pressure on

LNAPL flow rate using fine and coarse grain sand. The variation of steam flow rate with steam injection is illustrated for the selected soil-grain size and it is shown that steam flow rate is directly proportional to steam injection pressure.

Table 1: Steam Injection Pressure and LNAPL Flow Rate for Fine grain sand

Sand grain size, D (mm)	Steam injection pressure, bar	LNAPL Flow rate, cm ³ /min
0.31	0.4	7.6385
0.31	0.8	16.5539
0.31	1.2	28.7464
0.31	1.6	45.2158
0.31	2.0	65.9621

Table 2: Steam Injection Pressure and LNAPL Flow Rate for Coarse grain sand

Sand grain size, D (mm)	Steam injection pressure, bar	LNAPL Flow rate, cm ³ /min
1.20	0.4	19.1385
1.20	0.8	28.0539
1.20	1.2	42.1464
1.20	1.6	60.8158
1.20	2.0	89.1375

Effect of Steam Injection Pressure on LNAPL Recovery Efficiency

Experiments were conducted with different steam injection pressure and the volume of kerosene recovered was recorded at interval of 20 minute. At the beginning of the steam injection, the pressure was maintained at 0.5bar and the efficiency of kerosene recovery was calculated by dividing the total volume of kerosene recovered by the initial volume added to the soil. As the steam injection pressure was increased, the efficiency of kerosene recovery increased and reached maximum at 1.5bar. The experiments were carried out with the same concentration of kerosene in the soil and the result showed that the kerosene recovery efficiency increases with increase in steam injection pressure. The graph 2 shows the LNAPL recovery efficiency against time of treatment at pressure of 0.5bar, 1.0bar and 1.5bar. Steam injection pressure has a significant effect on LNAPL recovery. An increase in steam injection pressure yields faster LNAPL recovery and requires small amount of steam (number of pore volumes to achieve minimum LNAPL residual saturation).

Effect of Soil Grain Size on LNAPL Recovery Efficiency

Experiments were conducted with two types of sand having a uniform grain size and contaminated with kerosene, the sands used were fine and coarse grain sand. The kerosene recovery efficiency in the coarse sand during the experiment was plotted against time of treatment and compared with the numerical simulation (graph 3 and 4). At the beginning of steam injection the recovery efficiency was higher for coarse grain sand compare with that of fine grain sand at the same interval of time, due to larger pore space in coarse sand which increases its permeability.

Effect of Steam Injection Pressure on steam LNAPL Flow rate

In order to investigate the effect of steam injection pressure on flow rate of kerosene, the experiments were conducted with steam injection pressure varied from 0.4bar to 2bar with interval of 0.2bar and the corresponding kerosene flow rate was determined for each steam injection pressure. The

experiments were conducted with coarse-grain and fine-grain sand.

Because the removal rate is proportional to the steam injection pressure [20], [21], and relatively larger amount of contaminant was recovered at higher steam injection pressure. Maximum kerosene flow rate occur at pressure of 2.0bar.

Temperature Profile

The temperature profile in the sand column was obtained for the experiment. Dry steam was injected at temperature of 160°C and pressure of 1.75bar for 300 minutes. This experiment was conducted with kerosene-contaminated sand sample having a mean grain size of 0.31 mm. The column

base temperature distribution is displayed in Figure 4.6 for the experiment. As steam is introduced into the sand column, heat is been absorbed by the upper layer of the sand which make the temperature at the base of the sand column to be maintained at a constant value (room temperature) for a specific period of time before the thermometer at the base of the sand column sense an increase in temperature of the sand column. The temperature of the sand column increases with time as steam was continuously supplied into the sand column. The temperature profile was then generated with the temperature recorded at the base of the sand column against the time of treatment.

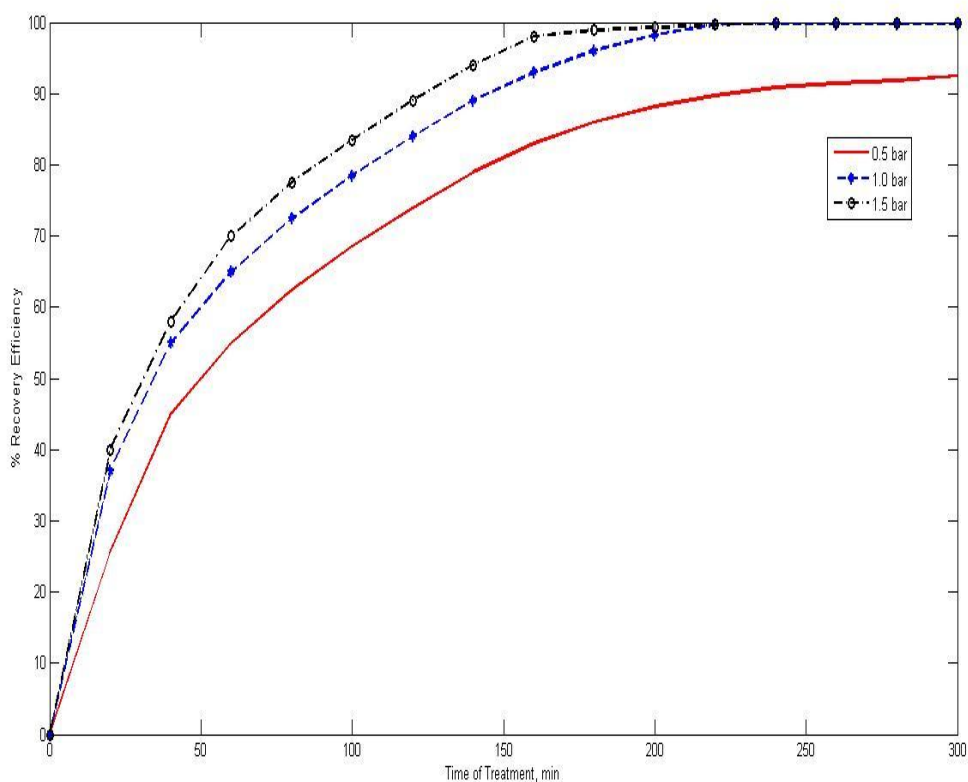


Fig 2. Effect of Steam Injection Pressure on LNAPL Recovery Efficiency

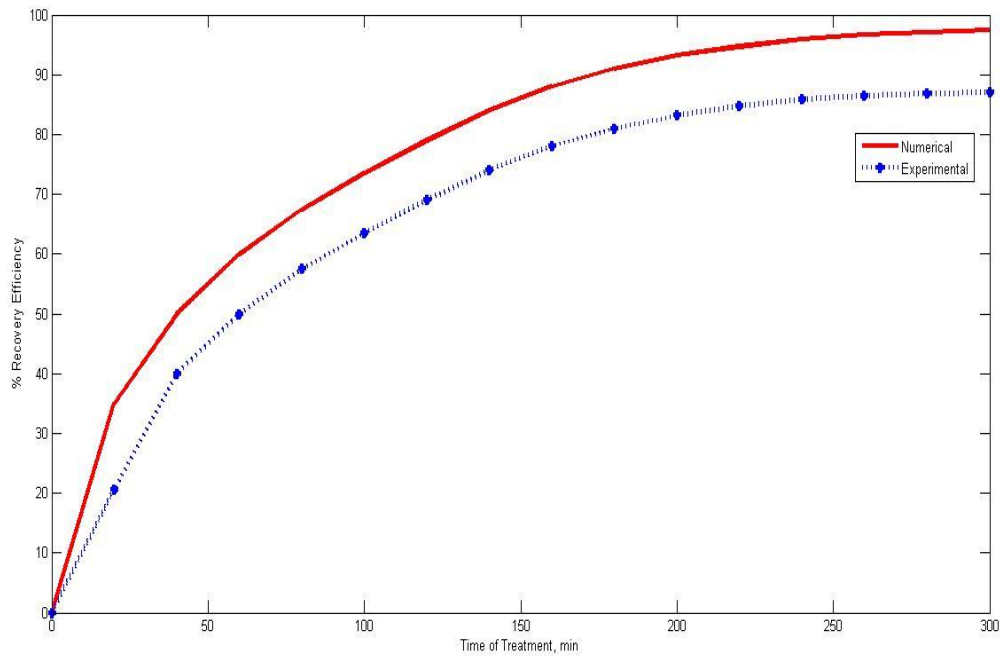


Fig 3. Graph of recovery efficiency against time for coarse grain sand

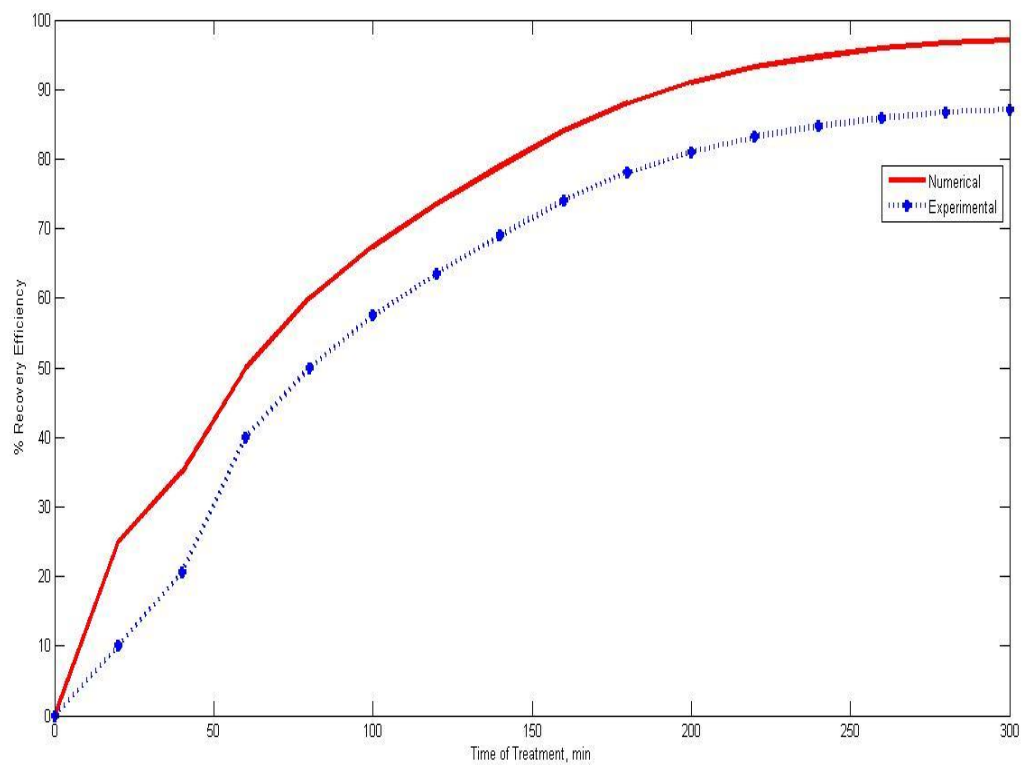


Fig 4. Graph of Recovery Efficiency against Time for Fine Grain Sand

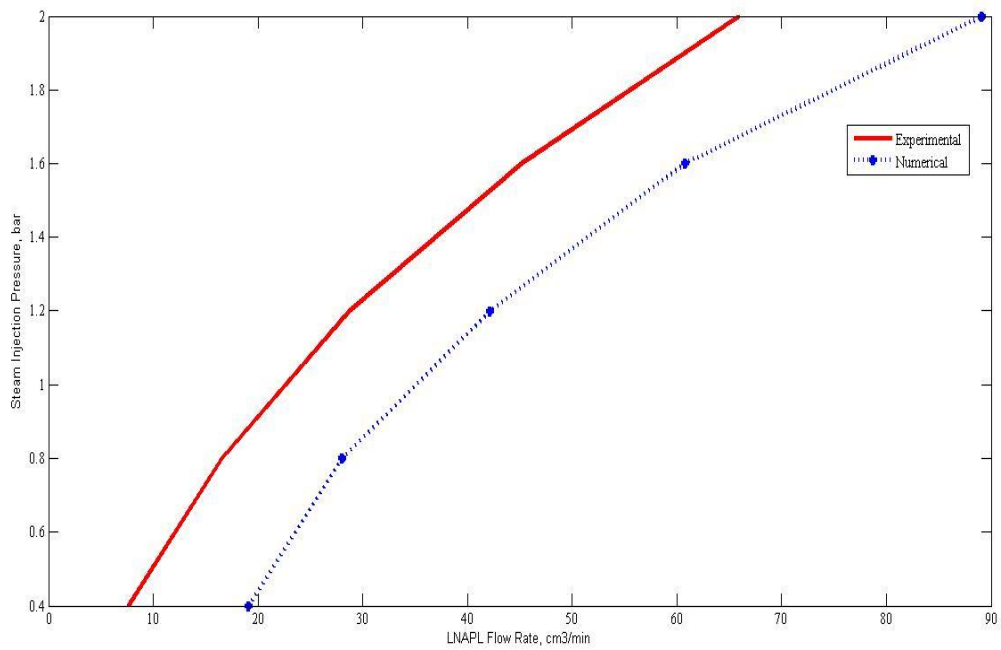


Fig 5. Effect of Steam Injection Pressure on Kerosene Flow Rate for Coarse Grain Sand

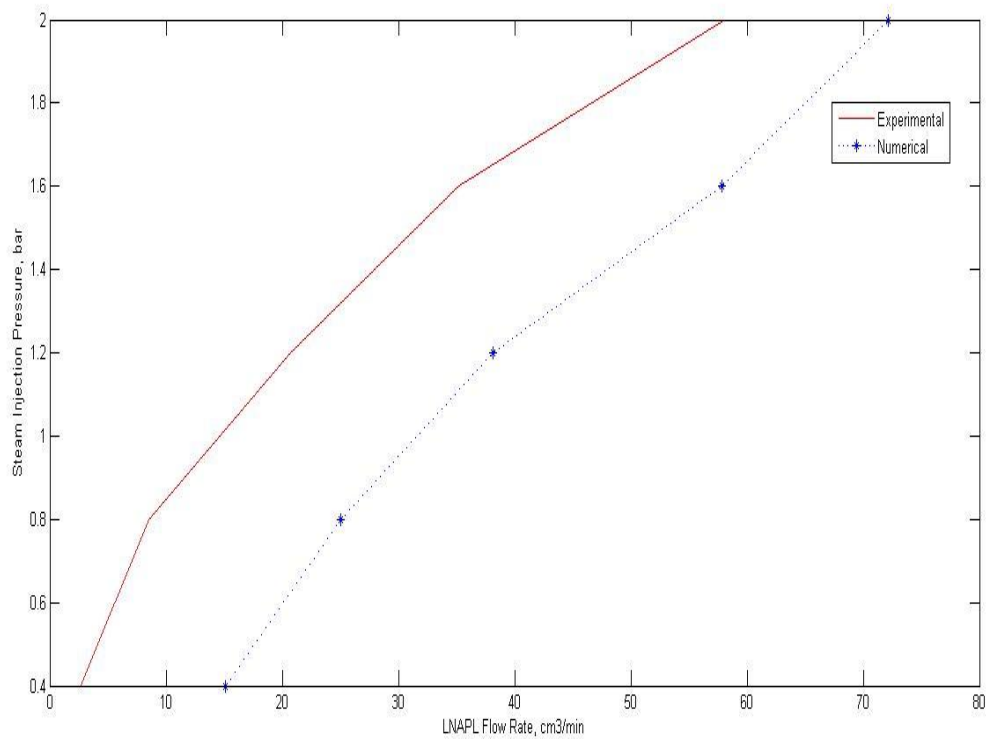


Fig 6. Effect of Steam Injection Pressure on Kerosene Flow Rate for Fine Grain Sand

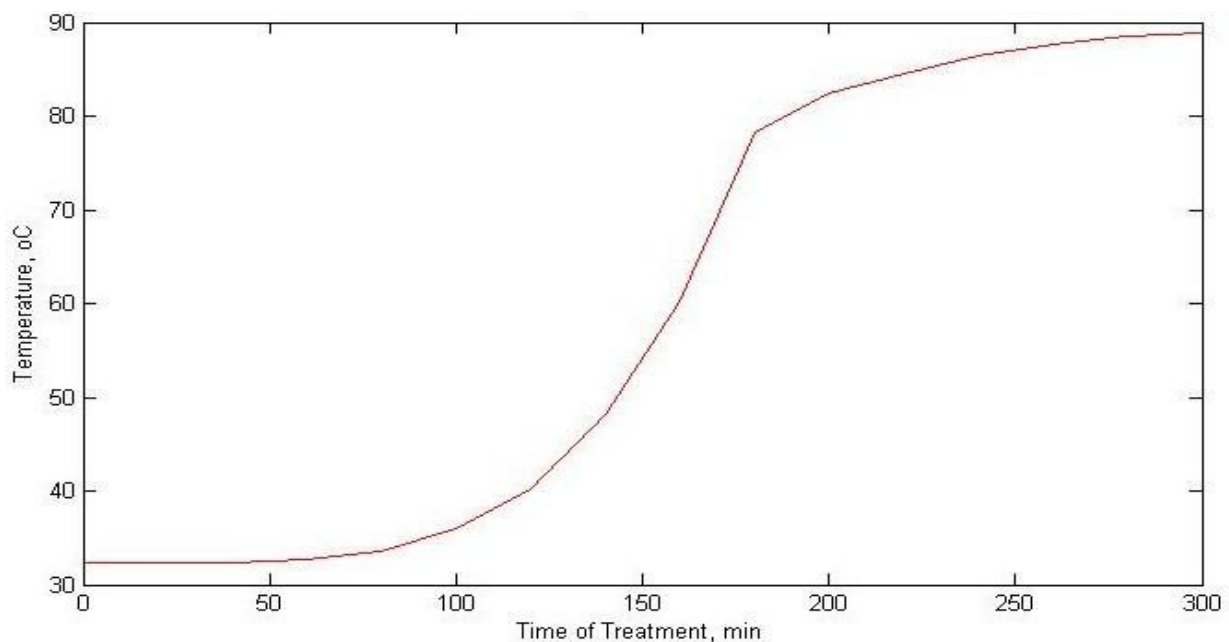


Fig 7. Temperature Profile Diagram

IV. Summary and Conclusion

Experimental and numerical studies were performed to evaluate the effectiveness of steam injection for remediation of LNAPL-contaminated soils. Experiments were conducted in one dimensional sand column contaminated with kerosene, and parametric studies were performed to evaluate the effect of soil grain size, steam injection pressure, and steam inlet temperature on kerosene recovery efficiency, time of treatment and flow rate. The following were concluded from the results of the research work:

1. Steam injection process was found suitable and effective for thermal soil remediation.
2. The higher the steam injection pressure the higher the rate of flow of LNAPL.
3. The higher the steam injection pressure the lower the time of treatment.
4. Lesser time is needed for treatment in coarse grain soil compare to fine grain soil at the same injection pressure due to higher permeability in coarse grain soil
5. In relating the numerical modeling and the experimental investigation, numerical modeling was found to be more accurate than experimental investigation due to losses and human error in the cause of carrying out the experiment.

V. Acknowledgment

The authors wish to thank Engr. O.S. Olaoye and Mr. T. Adeyemo of Mechanical Engineering Department, LAUTECH, Ogbomoso, Oyo State for release of steam boiler used for this work.

References

- [1] E. Davis, How Heat Can Enhance in-situ Soil and Aquifer Remediation: Important Chemical Properties and Guidance on Choosing the Appropriate Technique. Robert S. Kerr Environmental Research Laboratory, EPA/540/S-97/502, 1997.
- [2] K. S. Udell and L. D. Stewart, UCB-SEEHRL. 89-2. In: University of California, Berkeley, CA, USA, U.S. Environmental Protection Agency. 2004. In-situ thermal Treatment of Chlorinated Solvents-Fundamentals and Field Applications, EPA 542-R-04-01057, 1989a.
- [3] L. A. Smith and R. E. Hincsee, In-situ Thermal Technologies for Site Remediation, Ft. Lewis Publishers, Boca Raton, FL, USA, 1993.
- [4] G. Heron, S. Carroll and S. G. D. Nielsen, Full-Scale Removal of DNAPL Constituents Using Steam Enhanced Extraction and Electrical Resistance Heating, Ground-Water Monit. Remediat., 2005, 25(4): 92-107.
- [5] Danish EPA, Natur og Miljøpolitisk Redegørelse, Miljø-og Energiministeriet. In Danish, 1999.
- [6] J. R., Hunt, N. Sitarand and K. S. Udell, Non aqueous Phase Liquid transport and cleanup.1. Analysis of mechanisms. Water Resour. Res. 24, 1988a, 1259-1269.
- [7] M.T. Itamura, Removal of Chlorinated hydrocarbons from homogeneous and heterogeneous porous media using

- steam.UMI Dissertation Services, Ann Arbor MI, 1996.
- [8] H.Y. She and B.E. Sheep, Removal of perchloroethylene from a layered soil system by steam flushing, *Ground Water Monitoring and Remediation*, (19), 1999, 70-77.
- [9] R.Newmark, and various authurs, Dynamic underground stripping project. Lawrence Livermore National Laboratory, UCRL-ID-116964-V-1-4, 1994.
- [10] R. L. Newmark, R. D. Aines, K. G. Knauss, R. N. Leif, M. Chiarappa, B. Hudson, C. Carraigan, A. Tompson, J. Richards, C. Eaker, R. Weidner and T. Sciarotta, In-situ destruction of contaminants via hydrous pyrolysis/oxidation: *Visalia field test*. Lawrence ,Livermore Nitional Laboratory, UCRL-ID-132671 ,1998.
- [11] B. Hilberts, In-situ steam stripping. In: *Contaminated Soil*, Proc First Int. TNO Conf., (Assink, J.W. and Van Den Brink, Eds.)Utrecht, The Netherlands, November 11-15. 1985, pp. 680-687.
- [12] J. R.Hunt, N. Sitar and K. S. Udell , Non aqueous Phase Liquid transport and cleanup.2. Experimental studies. *Water Resour. Res.* 24, 1988b, 1259-1269.
- [13] A. E., Jr., Lord, R. M. Koener and V. P. Murphy, Laboratory studies of vacuum-assisted steam stripping of organic contaminants from soil. In: Proc. 14th Annu. EPA Conf. on Land Disposal, Remedial Action, and Treatment of Hazardous Waste, Cincinnati, OH. 1988, pp. 65-92.
- [14] B.T. Willman, V.V. Valleroy and G.W. Rauberg, Laboratory studies of oil recovery by steam injection. *SPE Reservoir Eng.* 1961, 95-104.
- [15] L. D. Stewart and K. S. Udell, Mechanisms of residual oil displacement by steam injection. *SPE Reservoir Eng.* 1988, pp 1233-1242.
- [16] K. S. Udell and L. D. Stewart, Mechanisms of in-situ remediation of soil and groundwater contamination by combined steam injection and vacuum extraction. In: *Symp. On Thermal Treatment of Radioactive and Hazardous Waste*, AICHE Annual Meeting, San Francisco, CA. 1989b.
- [17] R. W. Falta, K. Pruess, I. Javandel, and P. A. Witherspoon, Numerical modeling of steam injection for removal of non aqueous phase liquids from the subsurface. II. Code validation and application. *Water Resour. Res.* 28, 1992a, 433-449.
- [18] K. S. Udell and L. D. Stewart, Mechanisms of in-situ remediation of soil and groundwater contamination by combined steam injection and vacuum extraction. In: *Symp. On Thermal Treatment of Radioactive and Hazardous Waste*, AICHE Annual Meeting, San Francisco, CA. 1989c.
- [19] R. W. Falta, K. Pruess, I. Javandel and P. A. Witherspoon, Numerical modeling of steam injection for removal of non aqueous phase liquids from the subsurface. II. Code validation and application. *Water Resour. Res.* 28, 1992b, 451-465.
- [20] W.L. Crow, E.P. Anderson and E.M. Minugh, Subsurface venting of vapours emanating from hydrocarbon product on groundwater. *Groundwater Manage*, 1987, Res. 51-57.
- [21] A.L. Baehr, G.E. Hoag and M.C. Marley, Removing volatile contaminants from the unsaturated zone by inducing adjective air-phase transport. *J.Contam.Hydrol.*, 1989, 1-26.