

# ATMOSPHERIC PRESSURE DIELECTRIC BARRIER DISCHARGE (DBD) IN A PLANE-TO-GRID REACTOR FOR THE REMEDIATION OF NAPL- CONTAMINATED SOILS

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## ABSTRACT

Soil pollution by non-aqueous phase liquids (NAPLs) could affect the human health through direct contact, food chain, and groundwater contamination / water cycle. Soil pollution is often caused by industrial activity and urbanization, improper disposal of chemical and industrial waste, leakages from underground storage tank rupturing, downward migration of leachates from landfills, oil spills from accidents and fuel dumping. The most common NAPLs involved in subsurface contamination include poly-aromatic hydrocarbons (PAHs), pesticides, chlorinated solvents (e.g. PCBs) and petroleum cuts (gasoline, diesel oil, kerosene) containing alkanes of high molecular mass.

For NAPL abatement, various soil remediation methods have been proposed, such as thermal desorption, bioremediation, etc. Due to some drawbacks of the conventional remediation technologies, such as the high energy demand and secondary pollution, it is of crucial importance to develop cost-effective and highly efficient methods for soil remediation. It has recently been realized that certain Advanced Oxidation Processes (AOPs) constitute promising technologies for pollution control. Non-thermal plasmas of electrical discharges are considered a highly competitive technology for the removal of organic pollutants, among the AOPs. Plasma-based technologies take the advantage of the formation of highly reactive chemical agents within the medium which is under treatment.

Non-thermal dielectric barrier discharge (DBD) plasma was examined as an advanced oxidation process (AOP) for the remediation of soils. A plane-to-grid DBD reactor operating with air at atmospheric pressure was used to remove a synthetic NAPL (i.e. a mixture of n-C<sub>10</sub>, n-C<sub>12</sub> and n-C<sub>16</sub>) from soil layers. The plasma reactor is a cross-type cylindrical custom vessel, made of 304L stainless steel and sealed with copper gaskets. It has 150 mm inner diameter, 192 mm inner height and 2 mm wall thickness. The stressed electrode is a stainless steel disc (Ø 50 mm, thickness 3mm) covered by quartz dielectric of 2 mm thickness. The grounded electrode is a stainless steel grid (Ø 50 mm) playing the role of the sample holder and allowing the free air flow through the grid/sample. The air flow is vertical at a constant rate and the electrode gap is

fixed at about 7 mm. The contaminated soil is placed uniformly on the ground electrode. The biasing voltage is provided by a custom-made AC high voltage power supply (10 kHz, 0-30 kV peak-to-peak) and its waveform is monitored on a digital oscilloscope. The concentration and composition of the residual NAPL in soil were determined with NAPL extraction in dichloromethane (DCM) and GC-FID analysis. The effect of treatment time, energy consumption, moisture content, applied voltage, and gas flow rate on the NAPL removal efficiency was investigated, the plasma active species were identified, the macroscopic gas temperature was determined, and the gaseous byproducts were analysed.

The experimental results indicated that the NAPL remediation efficiency can be as high as 99.9% after 60-120 s of plasma treatment. The energy density required to remediate completely the NAPL was about 600 J/g-soil, indicating that the DBD plasma has the potential to become a highly cost-effective technology for the remediation of NAPL-contaminated soils. Optical emission measurements and exhaust gas analysis unveiled that the plasma was rich in excited neutral and ionic nitrogen molecular species,  $\text{NO}_x$  and  $\text{O}_3$ . At the same time, the macroscopic gas temperature found to be up to about 300 °C stimulating the evaporation of NAPL molecules. By using ATR-FTIR in combination with high-throughput organic profiling analysis by GC-MS, it was revealed that alcohols and ketones with the same number of carbon atoms to that of the three alkanes ( $n\text{-C}_{10}$ ,  $n\text{-C}_{12}$ ,  $n\text{-C}_{16}$ ) were the main intermediate products of NAPL degradation (oxidation) in soil. Based on the analysis of initial NAPL/ intermediates / products in soil and exhaust gas, it is suggested that the main NAPL removal mechanism is the evaporation coupled with oxidation of pollutants in the gas and liquid phase.