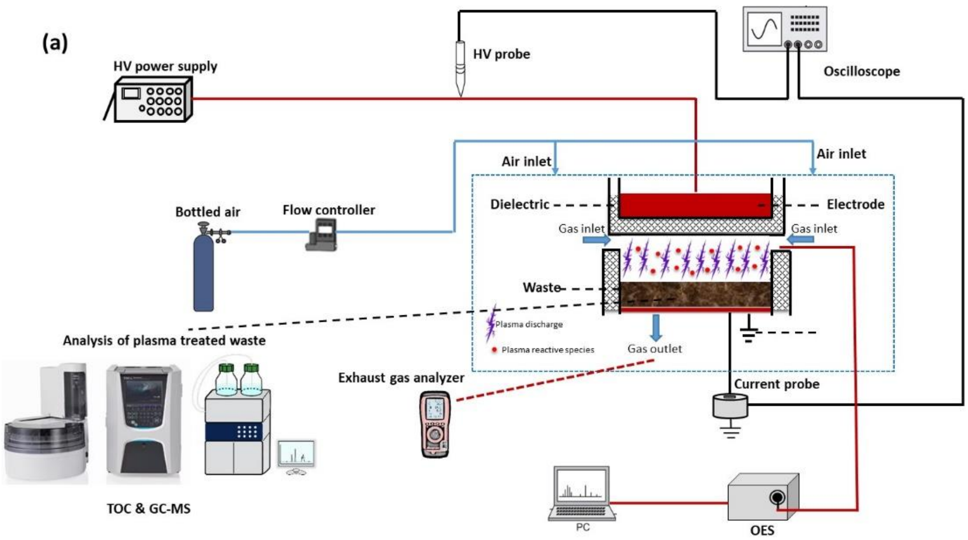
Assessing the efficiency of dielectric barrier discharge (DBD) plasma as an advanced oxidation process for the remediation of oil-drilling cuttings

Dielectric barrier discharge (DBD) plasma was investigated as a non-thermal process (NTP) for the treatment of oil drilling cuttings (Fig.1.,b). Several plasma conditions were investigated in a plane-to-grid DBD reactor whereas a combination of analytical methods (GC-FID, GC-MS/MS) was used to shed light on the oxidation processes. In particular, the effect of treatment time, applied voltage, energy efficiency, and air flow rate on the total organic carbon (TOC) removal was thoroughly investigated. In addition, the performance of the DBD was determined by measuring the residual total petroleum hydrocarbons (TPH) and polycyclic aromatic hydrocarbons (PAHs) concentration in drilling cuttings after plasma treatment along with the identification of oxidation byproducts in solid and gas phase. The findings of this study indicate that NTP process could be considered an interesting alternative for the treatment of solid wastes originated from the oil industry.



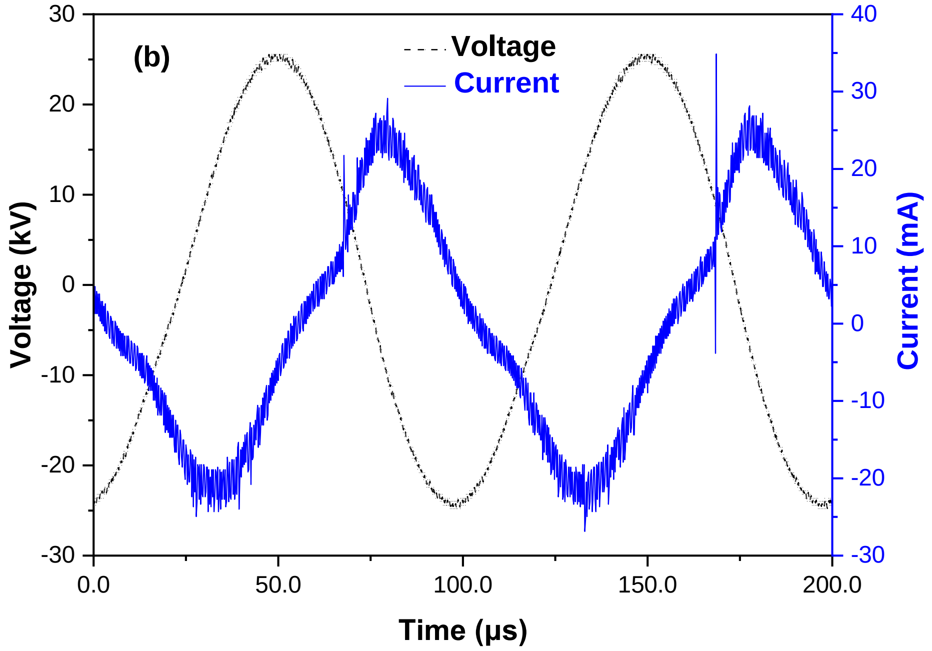


Figure 1. (a) Schematic diagram of the NTP system used to treat the oil-based drilling cuttings by DBD and characterize the plasma and (b) typical voltage-current waveforms during waste treatment in the DBD reactor.

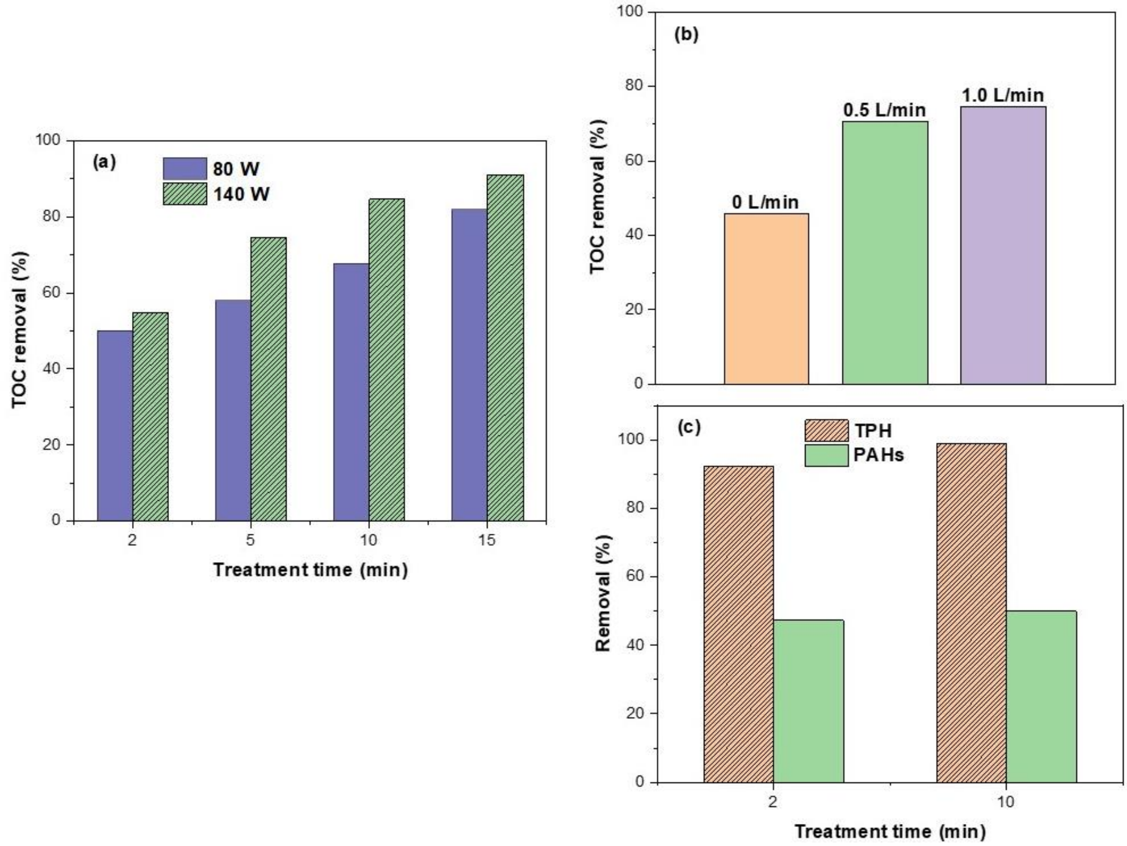


Figure 2. (a) TOC removal as a function of the plasma treatment time for different DBD power (air flow rate: 1 L min^{-1}), (b) TOC removal at various air flow rates (DBD

power: 140 W, treatment time: 5 min) and (c) The removal efficiency of the total petroleum hydrocarbons (TPH) and polycyclic aromatic hydrocarbons (PAHs) after 2 and 10 min of NTP treatment (DBD power: 80 W, air flow rate: 1 L min^{-1}).

At the lower DBD power (80 W), a significant percentage (~50%) of the initial organic carbon content was removed after only 2 min of plasma treatment, which gradually increased as the treatment time increased, reaching a value of 81.8% after 15 min of treatment. At a higher DBD power (140 W), the TOC removal efficiency increased, compared to that achieved at 80 W, due to the higher energy supplied to the DBD, resulting in enhanced electron density and RONS concentration. After 2 min of treatment, the TOC removal efficiency was 54.8%, and became very high (~91.1%) after 15 min of treatment, which is one of the highest values that have ever been reported in the literature for solid wastes (Fig.2a).

Without air injection in the DBD reactor, the TOC was reduced by 45.8%, indicating a significant transformation of the organic content present in oil drilling cuttings to CO_2 even under zero air flow rate. Under air flow conditions, the TOC removal was increased compared to no flow conditions, being 70.7% and 74.6% for gas flow rate 0.5 and 1 L/min, respectively (Fig.2b). In other words, the TOC removal efficiency increased by ~63% with the air flow increasing from 0 to 1L/min. The TOC removal efficiency has the tendency to increase with the gas flow rate increasing, and this can be justified as follows: the existence of a carrier gas in the DBD reactor can change the discharge characteristics resulting in enhanced concentrations of the oxidative plasma species.

The very significant reduction of TOC from oil drilling cuttings, due to plasma treatment, is further supported by measuring the concentration of the total petroleum hydrocarbons (TPH) and polycyclic aromatic hydrocarbons (PAHs). TPH and PAHs were measured under constant DBD power of 80 W, after 2 and 10 min of NTP treatment (Fig.2c). The reduction of TPH was respectable, reaching at 92.1% and 98.9% after 2 and 10 min of treatment, respectively while the corresponding reduction of PAHs was 47.2% and 51%. These results highlight the capacity of the NTP process to degrade very fast the high molecular weight hydrocarbons of the heavily contaminated oil drilling cuttings.

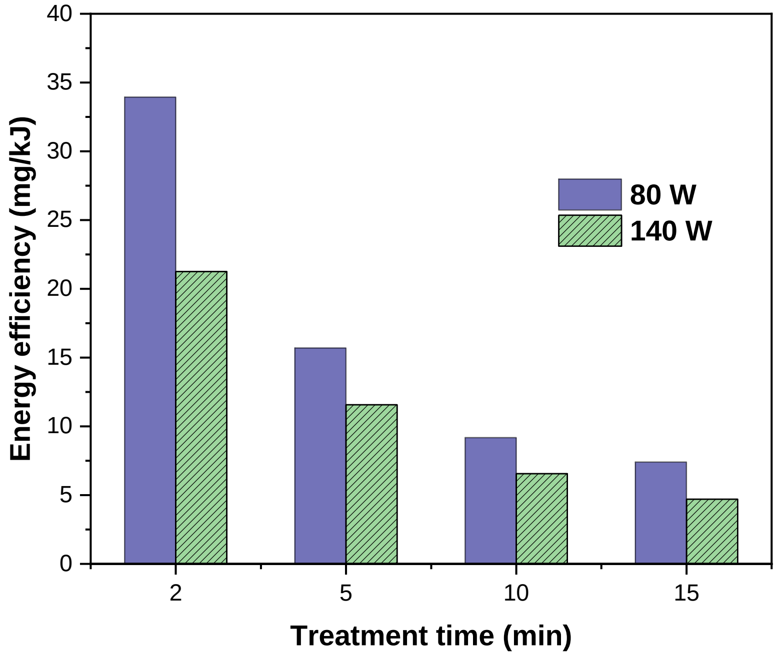


Figure 3. The energy efficiency of the process as a function of treatment time for 80 and 140 W DBD power (air flow rate: 1 L min^{-1}).

At short treatment time (i.e. 2min), where the TOC removal efficiency reached at 50% and 54.8% for power consumption of 80 and 140 W, the energy efficiency was 33.9 mg/kJ and 21.2 mg/kJ, respectively. Even at long treatment times (i.e. 15 min), where an impressive TOC removal was achieved, the energy efficiency was noticeable, being 7.4 mg/kJ and 4.7 mg/kJ for DBD power 80 and 140 W, respectively. In other words, a high energy efficiency was obtained ranging from ~5 to ~35 mg/kJ (Fig.3) for TOC removal efficiencies ranging from ~50 to ~91% (Fig.2a). Notwithstanding that in the complex petroleum wastes and oil drilling cuttings, this very high percentage of TOC removal achieved over very short treatment time, and associated with the aforementioned energy efficiency could be considered advantageous compared to other studies.

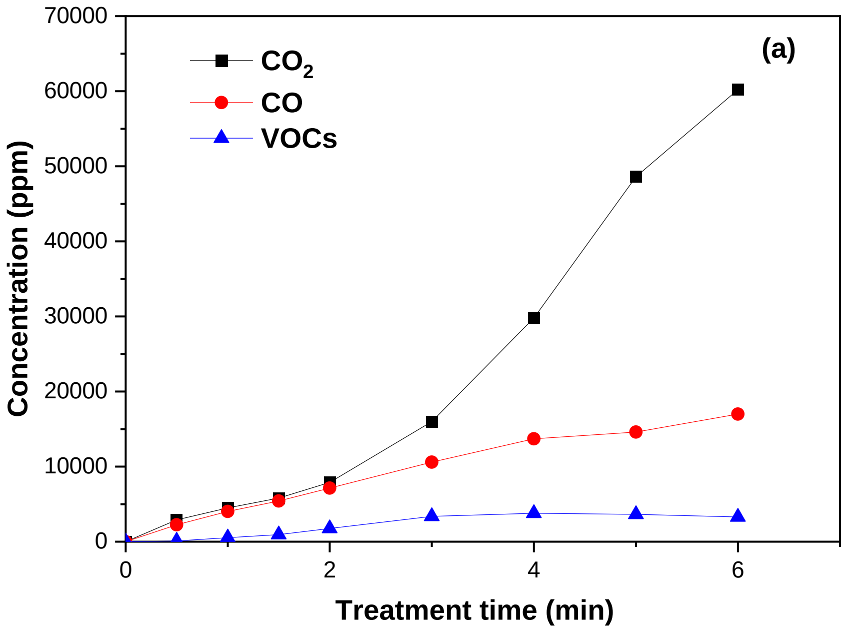


Figure 4. Transient evolution of the CO_2, CO and VOCs concentration in the exhaust gases of DBD reactor detected during a period of 6 min

The transient concentration of CO_2, CO and VOCs is illustrated in Fig. 4. Apparently, the organic pollutants removed by DBD plasma were converted primarily to CO_2, and secondarily to CO, while the concentration of VOCs released was quite low.

Considering that the process optimization at the lab-scale is the critical step before the implementation of the NTP process at larger scale, the results of this study can act as the steppingstone for further research. Our next steps include the investigation of several plasma and operational parameters in the current lab-scale DBD reactor and the design and construction of a pilot system consisting of multiple DBD electrodes able to treat a remarkable amount of solid waste. By keeping the lab-scale DBD reactor parameters (electrode gap, sample thickness, etc.) constant and using additional DBD electrodes, it is expected that a larger mass of waste will be effectively treated accompanied by an analogous increase in energy consumption, thus maintaining at least the energy efficiency that was achieved for the present lab-scale DBD reactor. The number of DBD electrodes will be adjusted based on the maximum power of the available HV power supply. To treat even larger amounts of solid waste, parallel operation of the aforementioned pilot-scale reactors may be required.