Non-Thermal Plasma Applied to Acetone-Polluted Water: a Possibility to Decontaminate and to Obtain High Energetic Gases

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Abstract-**The present work depicts results acquired from a chemical model and validated by a set of experiments on the treatment of acetone-polluted water by a non-thermal plasma technique. Results reveal the formation of hydrogen and carbon monoxide, a combination of gases better known as syngas, which were then clarified with a chemical kinetic model. From this model, the influence of hydroxyl radicals is highlighted by extraordinary reactivity during the acetone treatment and its conversion to syngas ($CO+H_2$). This study proposes a low energy density non-thermal plasma technique to depollute wastewater containing different levels of acetone.**

Keywords- Acetone Polluted Water; Non-thermal Plasma; Energetic Gases Production

I. INTRODUCTION

Derived from various processes such as the manufacture of basic chemicals, plastics, motor vehicle parts, fuel, meat products, and electronic equipment, wastewater is characterized by contamination with volatile organic compounds (VOCs). These compounds are listed in several countries as priority pollutants due to their harmful effects on ecosystems and human health [1, 2]. Due to the significant mobility of VOCs they can be ubiquitous; their presence has been detected in air and sediment of the Antarctic, in vegetation near Mount Everest, in marine water and in the surface sediments of lakes [3-5].

Removal of organic pollutants from water can be done with traditional treatment processes such as carbon adsorption and biological degradation; acetone, for example, is relatively easy to biodegrade [6, 7]. However, polluted groundwater also contains organic compounds that are difficult to biodegrade; furthermore, the acetone concentrations can be very low, at which point the biomass remains unstable and its treatment becomes problematic [8, 9]. Based on these observations, several studies have been undertaken regarding organic pollutant removal, such as the application of pervaporation in VOC removal from water [10]; in this technique, pervaporation membranes separate water and vapour respectively from the feed and permeated side [11]. Extraction of acetone has been realized [12], and phenol extraction is achievable when the pervaporation process is operated in conjunction with a decanter module [13].

Another type of technology used to treat VOC-polluted water is the use of an advanced oxidation processes (AOP); this technique maximizes the oxidizing potential of the hydroxyl radical (•OH), known to be more reactive than ozone (O_3) and hydrogen peroxide (H_2O_2) [14, 15]. There exist several treatments based on AOP, some of which employ chemical oxidizers such as O_3 and H_2O_2 assisted by UV lamps to generate •OH radicals [16] or a classic old reactive system, the Fenton process, in which •OH radicals are formed by adding H_2O_2 to Fe^{2+} salts [17, 18].

Non-thermal plasma techniques could be considered an AOP, due to the formation of oxidizing species •OH, O• and H•, molecules (H_2O_2, O_3) and UV light, combining them in a unique process from those described above [19]. For this reason, non-thermal plasma has been successfully applied to treat wastewater polluted with phenols [20, 21], with textile dyes [22-24], with the pesticide endosulfan [25], with toxic heavy metal ions such as Ni(II) [26]. Non-thermal plasma techniques have also been applied to the treatment of more complex molecules, such as estradiol or other chemicals derived from the pharmaceutical industry [27].

A previous study thoroughly examined the application of plasma technology to wastewater remediation [27]; from this review, it was concluded that plasma processes are attractive when applied during pre-treatment to convert persistent compounds into biodegradable intermediates, which can then be treated by a biological oxidation process at a lower cost. Several experimental plasma devices have also been described in terms of their removal and energy efficiencies, highlighting the advantages of DC arc plasma to treat phenols [28].

In the present work we will present preliminary results concerning the acetone treatment and the subsequent formation of syngas by a gliding arc discharge that could be roughly considered as non-thermal plasma.

II. BASIC PRINCIPLES AND CHEMICAL KINETIC MODEL

A gliding discharge arc can be produced by two divergent electrodes with a gas flowing between them. Where the electrodes are closest, the arc moves toward the diverging section until a critical length value is attained; at this point the discharge restarts. The formation of radicals occurs in the diverging section; the plasma is considered to be non-thermal or lacking in thermodynamic equilibrium.

A non-thermal plasma is a gas ionized by electrical discharges; it is characterized by electrons with high kinetic energy or high electronic temperatures that collide with background molecules (H_2O, N_2, O_2) to produce radicals, ions, photons and secondary electrons.

The model used in this study takes into account the radical formation from the collision of energetic electrons with molecules of water and air; the newly formed radicals then interact to convert acetone into H_2 and CO.

In plasmas, a large number of equations occur; here we detail 29 equations with respect to their importance on acetone removal and syngas production. The rate coefficients for radical formation and reactions with acetone are shown in Table 1 [28-44].

TABLE 1 CHEMICAL REACTIONS CONSIDERED IN THE KINETIC MODEL OF ACETONE REMOVAL

Reaction	Rate coefficient ($cm^3mol^{-1}s^{-1}$)
$e^- + O_2 \xrightarrow{k_1} O\bullet + O\bullet + e^-$	2×10^{-11}
$e^- + O_2 \xrightarrow{k_2} O\bullet + O(^1D) + e^-$	1×10^{-8}
$e^- + N_2 \xrightarrow{k_3} N\bullet + N\bullet + e^-$	6.5×10^{-9}
$e^- + O_2 \xrightarrow{k_4} O_2^-$	2.8×10^{-13}
$e^- + O_2 \xrightarrow{k_5} O^- + O\bullet$	6.5×10^{-11}
$e^- + O_2 \xrightarrow{k_6} O_2(a^1\Delta g) + e^-$	1.3×10^{-9}
$e^- + O_2 \xrightarrow{k_7} O_2^+ + 2e^-$	8×10^{-10}
$e^- + N_2 \xrightarrow{k_8} e^- + N_2(A)$	4.5×10^{-10}
$e^- + CO_2 \xrightarrow{k_9} CO + O^-$	2.5×10^{-12}
$e^- + H_2O \xrightarrow{k_{10}} \bullet OH + H^- + e^-$	3.3×10^{-10}
$e^- + H_2O \xrightarrow{k_{11}} H^- + OH^-$	9×10^{-11}
$O(^1D) + H_2O \xrightarrow{k_{12}} \bullet OH + \bullet OH$	2.2×10^{-10}
$O(^1D) + N_2 \xrightarrow{k_{13}} O\bullet + N_2$	2.6×10^{-11}
$O(^1D) + O_2 \xrightarrow{k_{14}} O\bullet + O_2$	6.3×10^{-12}
$O(^1D) + H_2O \xrightarrow{k_{15}} O\bullet + H_2O$	1.2×10^{-11}
$O(^1D) + O_2 \xrightarrow{k_{16}} O\bullet + O_2(a^1\Delta g)$	3.4×10^{-11}
$N_2(A) + O_2 \xrightarrow{k_{17}} N_2 + O\bullet + O\bullet$	2.5×10^{-12}
$N_2(A) + N_2 \xrightarrow{k_{18}} 2N_2$	3×10^{-16}
$(CH_3)_2CO + O\bullet \xrightarrow{k19} CH_3COCH_2\bullet + \bullet OH$	7.93×10^{-16}
$(CH_3)_2CO + \bullet OH \xrightarrow{k20} CH_3COCH_2\bullet + H_2O$	0.23
$(CH_3)_2CO + \bullet OH \xrightarrow{k21} CH_3COOH + \bullet CH_3$	2.71×10^{-17}
$CH_3COCH_2\bullet + O_2 \xrightarrow{k22} CH_3CH_2COOO$	1.45×10^{-12}
$CH_3COCH_2\bullet + O_2 \xrightarrow{k23} CH_3COCH_2OO\bullet$	1.28×10^{-12}
$CH_3COCH_2\bullet + O_2 \xrightarrow{k24} CH_3OOCOCH_3$	9.80×10^{-13}
$\bullet CH_3 + O\bullet \xrightarrow{k25} CH_3O\bullet$	1.54×10^{10}
$\bullet CH_3 + O\bullet \xrightarrow{k26} CO + H_2 + H\bullet$	0.40×10^{-0}
$CH_3O\bullet + O\bullet \xrightarrow{k27} \bullet CH_3 + O_2$	2.51×10^{-11}
$CH_3O\bullet + O\bullet \xrightarrow{k28} CH_2O + \bullet OH$	1.0×10^{-11}
$CH_3O\bullet + \bullet OH \xrightarrow{k29} CH_2O + H_2O$	3.01×10^{-11}

From the equations reported in Table 1, a chemical mechanism is proposed [46] and schematized in Fig. 1. Fig. 1 visualizes the importance of oxygen and water vapour in the formation of key radicals $\bullet OH$ and $O\bullet$, responsible for syngas formation and acetone removal.
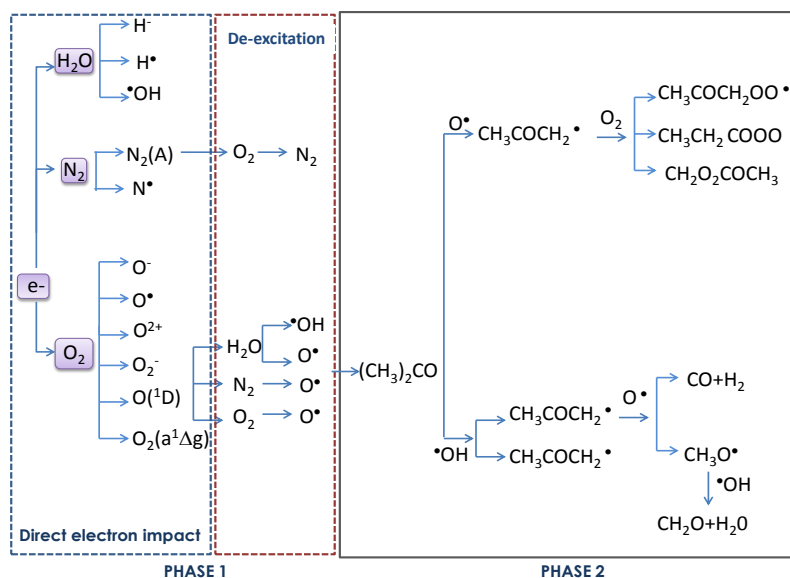
Fig. 1 Chemical mechanism in plasma, the radical formation is accomplished in phase 1;

the H_2-CO production and acetone conversion are observed in phase 2

A temporal distribution of chemical species could be accomplished with a mass balance applied to the equations listed in Table 1; the obtained system of ordinary differential equations can be solved with Matlab software. Figs. 2 and 3 report the evolution of species as a function of time. As seen in Fig. 2, radicals, ions and excited species are formed at approximately 10^{-8} seconds. The concentration of excited oxygen, $O(^1D)$, rapidly diminishes; this is explained by its interaction with H_2, N_2 and H_2O, subsequently forming $O\bullet$, $\bullet OH$ and N radicals as schematized in Fig. 1. These radicals have the higher concentrations in the plasma.
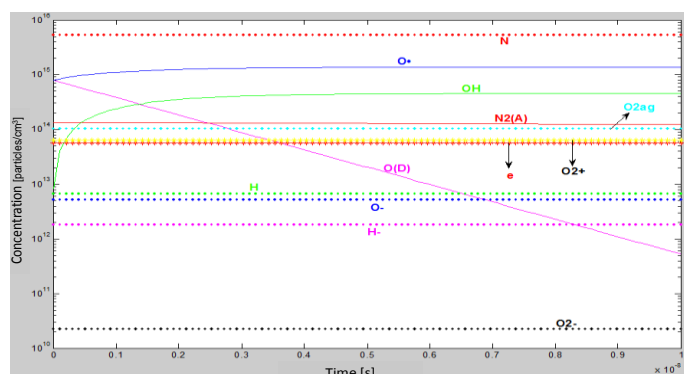

Fig. 2 Radical production

Fig. 3 demonstrates the influence of plasma in 200ppm of acetone dissolved in water (the nitrogen and oxygen are provided by ambient air). In this specific case, radical concentrations are low and the formation of H_2 and CO is insignificant.
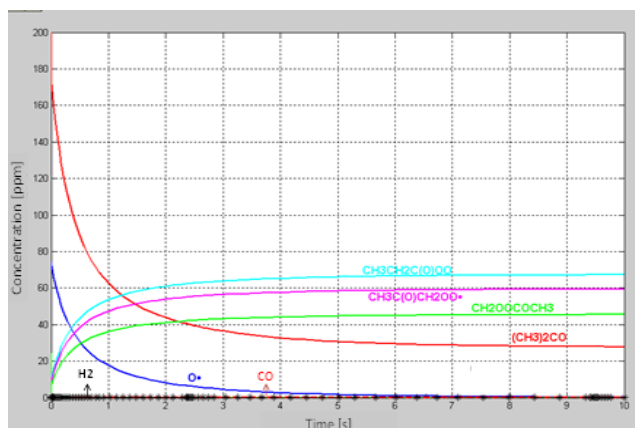

Fig. 3 Acetone, $(CH_3)_2CO$, removal

The acetone removal could be enhanced by augmenting the O• and •OH concentrations. For example, by augmenting O• concentration to 100ppm, almost a total acetone removal can be achieved. In this case, the influence of •OH is not as significant as that of the O• radical, as observed in Fig. 4. To augment O• radicals, a supplementary injection of air to the process could be realized.
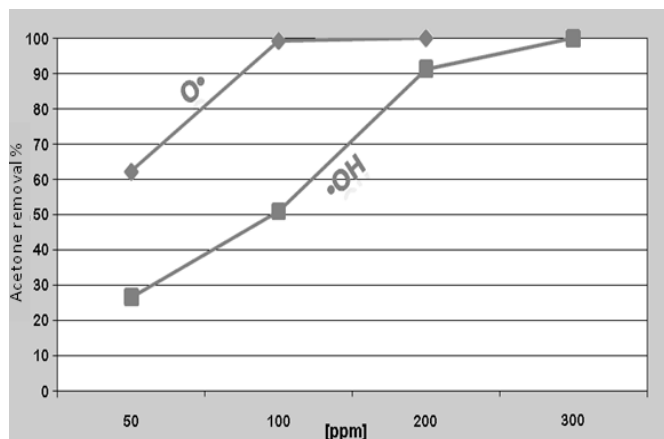

Fig. 4 Influence of O• and •OH radicals on acetone removal

III. EXPERIMENTAL SETUP AND EXPERIMENTAL FINDINGS

A non-thermal plasma reactor was used to treat acetone dissolved in water; it consists of three tungsten diverging electrodes. As demonstrated in Fig. 5, the plasma forms at the closest point between the three electrodes; the plasma arc moves towards the diverging section, pushed by the gas flow and by electromagnetic force created by the arc current. The discharge length grows until a critical length value is attained, determined by the power supply limit. This configuration is called a gliding arc discharge; it combines thermal plasma (thermodynamic equilibrium) with non-thermal plasma (out of thermodynamic equilibrium). In non-thermal plasma, the kinetic energy of electrons is superior to that of heavy compounds (e.g. ions, molecules, radicals, etc.) whereas in thermal plasma, all compounds have the same kinetic energy and/or temperature. In the non-thermal plasma region, radicals are constantly formed and acetone is decomposed into energetic species as explained in the previous sections.
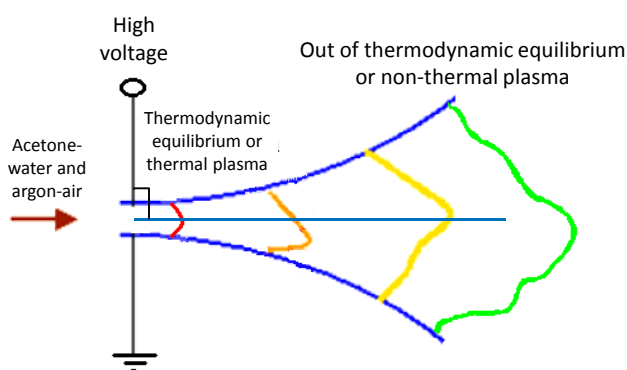

Fig. 5 Non-thermal plasma

The reactor used consists of a quartz cylinder with three 90W electrodes. The circuit supply is comprised of three half bridge inverters with a resonant circuit load constituted by step-up transformers 1:20 for TR1, TR2 and TR3, in series with capacitors C1=C2=C3=110nF. High voltages can be obtained (5kVpp) in the plasma discharge.

The acetone-water is added by direct injection with an argon-air flow. The process of injection is continuous at a feed rate of 5 ml per second. A mass spectrometer is used to identify formulated compounds before and after acetone treatment. An optical emission spectroscopy (OES) study was also conducted in order to observe the atomic and molecular compounds formed in plasma. The experimental setup is depicted in Fig. 6.
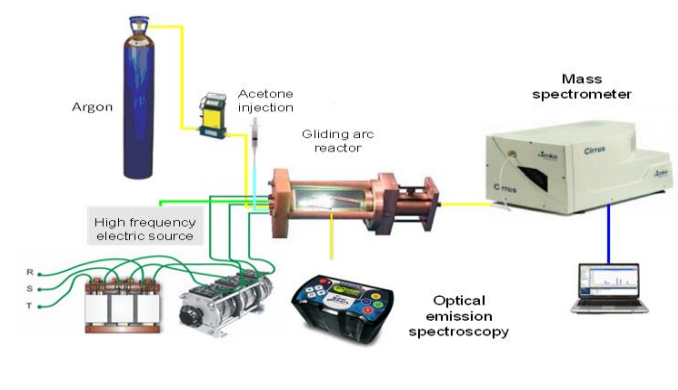
Fig. 6 Experimental setup

Plasma containing acetone, as depicted by Fig. 7(a), has a larger volume and a better distribution along the electrodes than argon plasma, shown in Fig. 7(b). This is possibly due to an increase of electrons in the presence of acetone, augmenting the electrical conductivity of the plasma.

Fig. 7 (a) Plasma formed with only argon; (b) plasma formed with acetone and argon

OES was applied to identify molecules as C_2 or atomic compounds (argon or hydrogen) in plasma. As can be seen in Figure 8(b), when acetone is introduced in the argon plasma, C_2 bands appear between 450 and 550nm [47]. The C_2 could be a precursor to carbonaceous products formed in a plasma process, such as carbon nanotubes [48]; however, in this case, only amorphous carbon was observed. From this study, the formation of H_2 is also corroborated with the presence of an H line situated at a wavelength of 656.3nm [49].
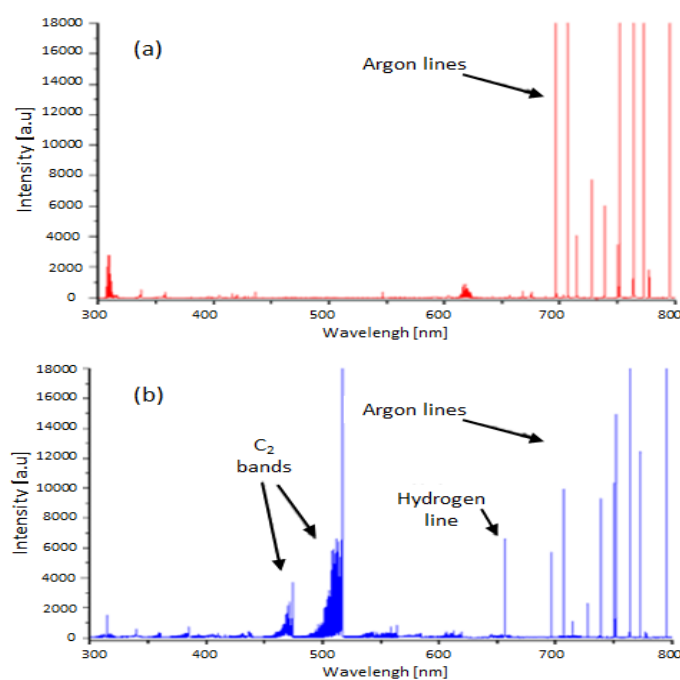

Fig. 8 OES Study. (a) Argon plasma; (b) argon-acetone plasma

Mass spectra of argon-air plasma (without acetone-water) and argon-air acetone-water, are respectively reported in Figs.

9(a) and 9(b). According to the NIST library [50] some characteristic lines of acetone are reported at 43, 58 and 15 uma; no acetone lines can be observed in Fig. 9(b).

The removal efficiency cannot be strictly calculated from these results; however, 90% acetone removal is concluded with respect to equipment precision and potential experimental error. Under this consideration, the energy efficiency of the plasma reactor attains 6×10^{-4} mol/J as compared to a DC water plasma torch, for which energy efficiencies are approximately 1.7×10^{-7} mol/J [51].

A slight increase in H_2 synthesis is also observed, the intensity relation of H_2/N_2 increasing during the treatment of acetone compared to that observed in argon-air plasma (see Fig. 9b). CO production cannot be confirmed, because its characteristic lines could be overlapped with these of N_2 on mass spectra.
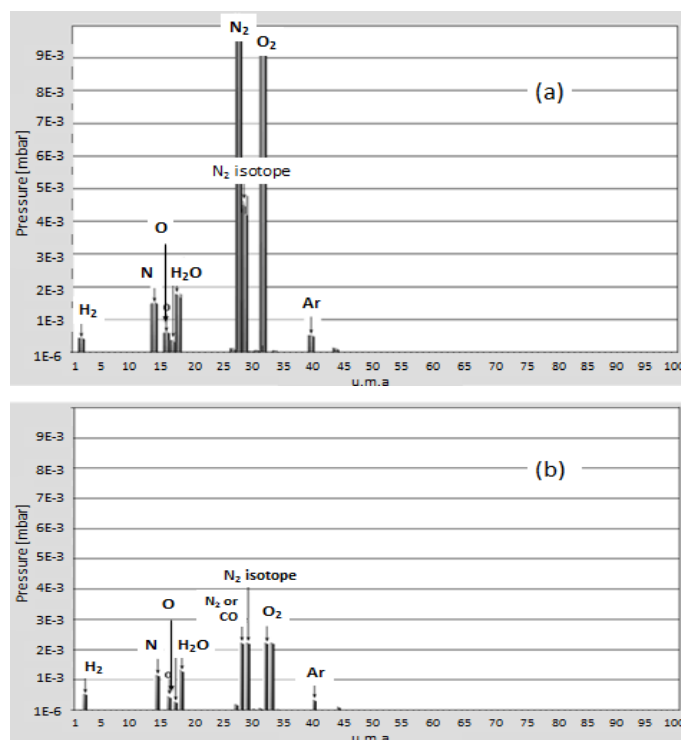

Fig. 9 Mass spectra of (a) air-argon plasma and (b) acetone-water-air-argon plasma

IV. CONCLUSIONS

The role of O• radical on acetone treatment was highlighted in the analysed chemical mechanism; experimentally, this is accounted for by air injection to the plasma reactor. Preliminary results indicate that gliding arc could be considered a promising technique for the continuous treatment of acetone-polluted substances at a considerably low energetic value. In order to further quantify the removal efficiency, subsequent experiments should realize the characterization of products should with gas chromatography.

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