Thermal and Non-Thermal Plasma for Destruction of Pollutants

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Wastewater remediation has become a global concern; thus, the development of innovative advanced oxidation processes (AOPs) for wastewater treatment is still a major challenge. Advanced oxidation processes (AOPs) are innovative tools that involve an introduction of energy (e.g., chemical, electrical and radiative) into the reaction zone to generate highly reactive species, especially hydroxyl radicals (HO•). In this sense, plasma technology is included among AOPs and considered as a promising remediation technology for water remediation. Developed since 1987 [1], the technique is environmentally friendly, simple and effective for decomposing toxic organic compounds in raw water or wastewater [2 - 4]. In view of these facts, applications of atmospheric pressure electrical plasma technologies for water treatment attract the increasing interest and emerge as technological opportunities for the plasma community [5].

Plasma is considered the fourth state of the matter, it is a partially or fully ionized gas and it contains ions and free electrons (electrical gas) [5, 6]. Plasma systems are traditionally defined into two major categories, i.e., thermal and non-thermal, in terms of electronic density or temperature [7].

Plasma-based water treatment (PWT) involves the generation of HO• radicals 'in situ' and from the water itself; therefore, unlike most AOPs and conventional processes, little or no chemical additives are required. Additionally, plasma offers a broader range of chemical and physical treatment mechanisms.

In plasma generation technologies, power source is one of the primary determinants, and various types, including corona discharges (DC), pulsed corona, dielectric-barrier discharge (or silent discharge, DBDs), gliding arc discharge (GAD), RF-driven atmospheric-pressure plasma jet, electron beam and microwave discharges can be used [6,8-15]. Alternate current (AC), direct current (DC) and pulse electrical discharge can be disposed into a wide range of electrode configurations. Fig. 1 shows a schematic basic dielectric-barrier discharge configuration (adapted from [9]).
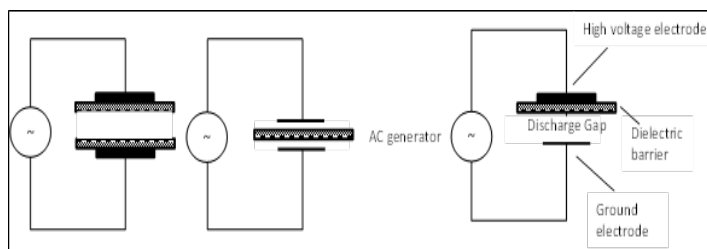

Figure 1: Basic dielectric-barrier discharge configurations (adapted from [9]).

For pulsed electrical discharge, the reactor is driven by a pulsed electric generator able to create very sharp high voltage pulse with a nanosecond or microsecond range duration time. Thus, in this discharge process, only free electrons gain high energy with producing energetic electrons leading to non-thermal plasma generation [6]. For example, pulsed electrical power can be applied for several discharge types such as CD, DBD and GAD, due to its high ionization efficiency and low energy consumption. The electrical generators are normally DC rather than AC for arc water plasma discharge because of its less flicker generation and noise, a more stable operation and a minimum of two electrodes, etc. [17].

The gliding arc discharge (GAD) is considered as an innovative technology exhibiting a dual character of thermal and non-thermal plasma and a typical reactor, consisting of insulating cover, "knife-edge" divergent electrodes, high voltage power supply, the nozzle and impedance [16]. In gliding arc discharge processes, high voltage is introduced between two or more thin "knife-edge" divergent electrodes and the narrowest point is electrical breakdown resulting in arc discharge when the electric field reaches approximately 3 kV mm^{-1} in air. As a high velocity gas flows through the nozzle, the length of the arc (actually a thermal plasma) and the temperature of the ionized gas increase.

It is advisable to apply lower-energy corona and glow discharge

processes for treating organic contaminants with low concentrations. For high concentrations of organic compounds in the liquid phase, DC arc plasma or gliding arc plasma may be more effective. Hybrid gas–liquid reactor and aerosol reactor are especially effective for degrading gas and liquid phase pollutants simultaneously. As for pulsed or DC pulseless corona discharge, they can readily be applied to treat the water of low electrical conductivity because of a higher efficiency of radical formation. DC glow discharge maybe the first-rank option for high conductivity water remediation [6].

Thermal plasmas are widely applicable for the destruction of noxious compounds, either in the form of complex mixtures like mixed liquid wastes, in the form of contaminated soil or sludge, or in the form of mixed liquid and solid waste. Thermal plasma is produced through arc discharges, torches or radiofrequency and it is sustained by introducing sufficiently high electrical energy to allow plasma constituents to be in thermal equilibrium. Thus, a high flux of heat is created, which can be used in processing even the most recalcitrant wastes via thermal incineration processes [17]. Fig. 2 shows some thermal plasma sources for environmental applications [18].
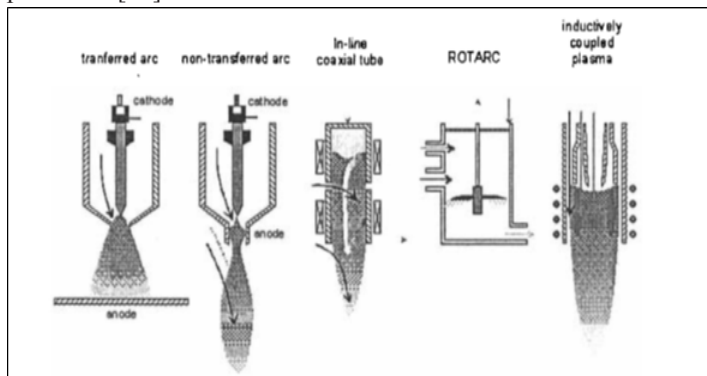

Figure 2. Some thermal plasma sources for environmental applications (from [18], with permission).

Thermal plasma has been applied for contaminated solution treatment due to its high energy densities and temperatures, oxidation or reduction atmosphere and rapid quenching rate (10^5-10^6 K s^{-1}), which can produce non-equilibrium chemical compositions [6]. Because it does not sustain a local thermodynamic equilibrium, it offers high selectivity and energy efficiency for chemical reactions. The widely utilized thermal plasma generation reactor is a torch configuration. The contaminated solution is usually directly introduced into the torch as plasma forming gas. When the arc is ignited, the aqueous solution can be evaporated spontaneously to provide the generation of 100%-water plasma due to enormous heat from an anode. Simultaneously, the anode is cooled by the water evaporation; therefore, no cooling-controlled units are required. In the highly reactive plasma zone, there is a large fraction of electrons, ions and excited molecules together with the high temperatures. On the basis of temperature distribution along the plasma jet, the arc plasma can be divided into three thermal regions, namely, arc region (over 10,000 K), plasma flame region (about 6000 K), and downstream region (lower than 1000 K). Thus, when wastewaters are injected into the arc discharge plasma zone, they are heated very rapidly by the plasma. The volatile matter is released and cracked giving rise to hydrogen and light

hydrocarbons such as methane and acetylene via thermal plasma pyrolysis process. Furthermore, the optical emission spectrum indicates that arc discharge can dissociate the water molecules at high temperatures into HO•, H•, and O• radicals. These active species are directly or indirectly beneficial for the oxidation of a target pollutant. Therefore, this process provides an alternative method for pollutant treatment by plasma technology. Using thermal plasmas, organic compounds can be destroyed with high efficiency, metals can be recycled, and heavy metals and low level radioactive materials can be vitrified in a non-leachable slag. The major disadvantage of this plasma process is the use of high electrical energy. However, it may be economically viable for refractory and high concentration toxic organic wastewater treatment.

In contrast, non-thermal plasma is obtained using less power (usually DC, DBDs, GAD, glow discharge and spark discharge), characterized by an energetic electron temperature much higher than that of the bulk-gas molecules [6]. Similarly to thermal plasma, it does not express a local thermodynamic equilibrium, and therefore, it also offers high selectivity and energy efficiency in plasma chemical reactions. The non-thermal plasmas used for environmental applications are mainly high-pressure discharges, such as DBDs, pulsed corona discharges and dielectric packed bed reactors as well as electron beam sustained plasmas. An increasing number of investigations are devoted to the decomposition of nitrogen and sulfur oxides in flue gases and of volatile organic compounds (VOCs) emanating from various industrial processes. Many hazardous organic compounds are readily attacked by exciting species, free radicals, electrons, ions and/or UV photons generated in DBDs. Moreover, investigations are going on to use DBDs for the generation of H_2 and elemental sulfur from H_2S and for the conversion of the greenhouse gases CO_2 and CH_4 to syngas or liquid fuels. The removal of hazardous organic pollutants from waste water is also a growing issue in environmental research. In particular, pulsed corona discharges, DBDs and contact glow discharge electrolysis techniques are being studied for the purpose of cleaning water. A well-established means of water purification is by ozone treatment. The ozone synthesis typically takes place in electrical discharges. However, electrical discharges in aerated water are also possible, and they produce some other strongly oxidizing agents, such as HO•, H•, O•, H_2O_2, etc. Therefore, instead of 'ex situ' electrical discharges for ozone production, the 'in situ' electrical discharges in water may provide a means to utilize most of these chemically active species for water purification. Moreover, the strong electric fields of electric discharges are also lethal to several kinds of microorganisms in water. Furthermore, the electrical discharges in water may also produce UV radiation, which helps in the destruction of pollutants. Besides these high pressure discharges, reduced pressure discharges, such as SWDs, have also proven their suitability for environmental applications, e.g. for the transformation of VOCs detrimental to health, and molecules utilized in the micro-electronics industry, such as SF_6 and C_2F_6, which strongly contribute to the greenhouse effect. Chlorinated hydrocarbon pollutants such as trichloroethylene can be treated in a SWD in oxygen or air-water mixture and converted into to light gases, primarily CO_2, CO, HCl and Cl_2.

Based on the plasma-phase distribution, electrical discharges with liquids can be subdivided into three main groups, i.e., electrical discharges above the liquid surface, direct electrical liquid dis-

charges and discharges in bubbles/vapor in liquids [4]. For electrical discharges above the liquid surface, the plasma generation and gas phase breakdown above the liquid surface are rather similar to the gas electrical discharge. However, similarly to electrical discharges above the liquid surface with the liquid as an electrode, the existence of the liquid surface affects the physical and chemical aspects of the processes occurring in the gas-liquid interface. The reason is that the discharge current is transported through the water electrode by ions, which have a much smaller mobility than electrons in metals. Additionally, water has a smaller secondary electron emission coefficient and is easier to deform and evaporate than most metals. On the other hand, bulk-phase reactions in the liquid can in turn be influenced by the plasma formed at the gas–liquid interface. Strongly oxidative reagents (HO•, O•, O_3, etc.) generated in the gas-liquid interface can diffuse into water and initiate chemical reactions. The mechanism for direct discharge in liquid is complicated since water as a discharge medium is much denser than gases and does not express the long periodic range of the atomic and molecular moieties as in most solids. In electrical liquid discharge, the current is transferred by slow ions in water that is strongly affected by liquid conductivity. Moreover, the purity, such as dissolved gases which form micro-bubbles in the liquid, plays a significant role in the breakdown process. Electrical discharges in bubbles/vapor in water are completely surrounded by the liquid which serves as an electrode. Normally, electrical discharges with external gas bubbling or in the vapor phase at metal electrode surface can be categorized into this group. The basic chemistry of this type is generally similar to that in gas-phase discharge with liquid as an electrode. However, electrical discharges in bubbles own a large contact surface area of gas and liquid, increasing the efficiency for gaseous phase species diffusing into the liquid and require lower energy to generate the plasma than directly in the liquid. Finally, these electrical discharges have certain common chemical reaction mechanisms and physical phenomena, such as the formation of molecular and radical species and UV light generation. Besides producing reactive chemical species (HO•, O•, H•, HO_2•, O_2, O_3, H_2O_2, H_2), the plasma channels formed directly in water can reach temperatures of up to several thousands of Kelvin and thermally degrade molecules, emit UV and visible light, and generate shockwaves capable of inducing cavitation [4, 19, 20]. While these features make plasma attractive as a stand-alone technology, they also open up the possibility of combining plasma with other AOPs. For example, the generation of UV light allows plasma to be effectively integrated with photocatalysis [21 - 23].

Non-thermal plasmas (NTPs) are good technologies for hazardous chemical destruction, pollution control, and surface decontamination [8]. Different techniques are able to generate plasmas in non-thermal form - sometimes called non-equilibrium or 'cold' plasmas - from an electrical discharge or bombardment of gas with an electron beam of high energy; they contain different electron, ion, and neutral species, where the electrons have the highest temperature (1-10 eV). DC, pulsed corona, DBDs, RF-driven atmospheric-pressure plasma jet, electron beam, microwave discharges, and corona discharges are used [10-15]. In non-thermal plasma, the energetic electrons can collide with background molecules (N_2, O_2, H_2O, etc.) producing secondary electrons, photons, ions and radicals [24]. Thus, those plasma are good sources of highly reductive and oxidative reactive species (depending on the content of

the solution) such as reactive oxygen species (ROS), O(^{3}P), HO•, N, H, NH, CH, O_3, O_2($^1\Delta$), H_2O_2, the own energetic plasma electrons (~ few eV), etc.; the gas temperature is near ambient (~ 300 K). Nitrogen-based reactive species (RNS) such as NO_2•, NO radicals, NO_3^-, NO^-, and its derivatives formed with water, including nitrites, nitrates and peroxynitrites can be also be formed. All these species have at the same times biocidal effects (e.g., on *Escherichia coli* bacteria) in water. The plasma technologies also have interest for their potential use in biological and medical applications.

Among the various chemical species produced by plasma at the gas–liquid environment, HO•, O•, O_3 and H_2O_2 are the main ROS generally accepted to play the dominant role in non-thermal atmospheric-pressure plasma systems. Gas-phase discharges in contact with liquid, and at the gas–liquid environments, generate various chemical effects in the liquid. Depending on the type of discharge, its energy, and the chemical composition of the surrounding environment (of both gas and liquid phases), various types of plasma-chemical reactions can be initiated and several primary and secondary species can be formed by these discharges in the gas and at the gas–liquid interface, which can penetrate or dissolve into the liquid and initiate chemical and biocidal processes therein [25]. The presence of these species allows using the method in multiple applications: removal of gases from gas streams (e.g., industrial exhausts and incinerators containing SO_x and NO_x), decomposition or fragmentation of aliphatic (methane, propane, propene, butane [14, 26]), destruction of polyhalogenated aliphatic hydrocarbons into smaller, more-easily combustible ones. NTPs can also be used to 'activate or 'crack' hydrocarbon fuels, promoting the combustion of fuels, treatment of aromatic hydrocarbons (toluene, pyrene, and substituted aromatics [27]), gases of solids and groundwaters contaminated with VOCs (with previous transfer to the vapor phase by pumping and application of heat and fluidization in an inert gas), cleaning of contaminated surfaces, etc. [5, 28], halocarbons in soil and water or emitted as stack gases, nitrogen oxides (particularly nitric oxide, NO) in flue and engine-exhaust gases, cleaning of VOCs, degradation of aldehydes [29]. NTPs have also been applied to combustion because it can increase the combustion efficiency and improve the combustion stability for the reduction of pollutant emissions [30-32]. Combustion engine exhaust treatment for automobiles and power plants is an important issue and many articles are dedicated to NO_x (NO and NO_2) and SO_2 removal, together with treatments for diesel and gasoline engine exhausts [15]. The method is excellent for destruction of carbohydrates. A large range of solvents and pollutants can be effectively removed: alkanes, alkenes, alcohols, aldehydes, ketones, and aromatic or halogenated compounds [33]. Surface decontamination applications include radionuclides [34] and chemical and biological warfare (CBW) agents [35].

Because the electrical discharge plasma technology leads to various physical and chemical effects, such as formation of oxidizing species (HO•, H•, O•) and molecules (H_2O_2, O_3, etc.), shockwave, ultraviolet light and electrohydraulic cavitation, plasma degradation process is generally regarded as a combined process of some other AOPs including ozonation, UV photolysis and pyrolysis, etc. and has the advantages of no demands on temperature and pressure, insensitive to contaminants and environmental friendliness [6]. Over the past few years, electrical discharge has been applied

successfully on many aspects, such as analysis of heavy metals in solution, synthesis of nanoparticles, and removal of organic pollutants. Two types of treatment processes can be employed to apply plasma methods to degradation of pollutants: (a) indirect plasma, represented by ozone generators whereby the plasma is used to generate the oxidant, which is then delivered to a separate reactor for water treatment; (b) direct plasma, which contacts directly the liquid phase containing the contaminants [5, 36].

Figure 3 shows the most important chemical processes induced by air plasma in water [25].
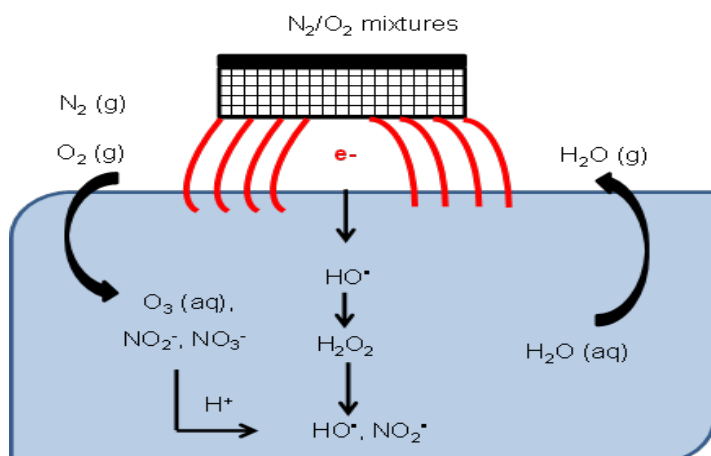

Figure 3: Chemical processes induced by the air plasma in water, adapted from [25].

The composition of the carrier gas, particularly the oxygen content and the relative humidity are important factors [25]. NTPs can affect flame stability and, in this way, potentially reduce the emission of pollutants. Operating an NTP reactor at atmospheric pressure provides much greater processing rates and throughput than low-pressure reactors. Atmospheric-pressure NTPs can be easily created by electrical discharges in a variety of gases (including air).

In contrast with incineration, plasma processes present little interaction with the environment, and the process can be easily coupled with a biological treatment. It should be mentioned that mineralization is generally incomplete because of the formation of carboxylic acids, alcohols, aldehydes, or ketones of low molecular weight, which represents a disadvantage. Acetic and propionic acids are very resistant and require catalysts for their destruction, and the same occurs with halogenated aromatic compounds (1,2-dihalobenzenes, PCBs, perchlorophenols). The low solubility of O_2 in water generates mass transfer problems, which can also limit the efficiency. The main disadvantage is the necessity of employing very expensive construction materials [5].

The technique does not generate toxic by-products, such as dioxins or furans, it operates at pressures and temperatures close to room,

it does not require fuel, and it can simultaneously remove hazardous organics and emissions of the type SO_x/NO_x [28]. It does not require catalysts.

Although the PWT technology has not reached yet sufficient level of development to be used in practice, some studies have been conducted to identify and characterize design parameters and physical phenomena that influence treatment efficiency [2,19]. Malik [2] calculated and compared relative energy yields of about 27 major types of plasma reactors, revealing dramatic differences in the energy yields, up to five orders of magnitude. The most efficient were pulse powered reactors, in which plasma is formed in gas phase and the waste solution sprayed into it. Additionally, Stratton et al. [19] established that the chemical reactions responsible for the degradation of organic solutes can be more accurately represented, mathematically, as heterogeneous reactions, rather than homogeneous reactions A general design principle, which prescribes maximizing contact between the plasma and the treated solution, was proposed, and the authors applied this design principle to create seven different ''contact-oriented'' reactors. The removal efficiencies attained in this work compared quite favorably with those achieved by other AOPs for the degradation of Rhodamine B, investigating how selected reactor design parameters affect the removal efficiency of the dye, using the resulting relationships to originate a more universal guideline for developing new reactors.

As an example of plasma applications, VOCs destruction in DBD and packed-bed reactors has been intensively investigated during the last ten years, because these discharges can be produced with very simple and low cost generators [9].

On the other hand, glow discharge plasma in aqueous solution (GDP) is a novel electrical process in which the plasma is sustained between a metal electrode and an electrolytic solution [37-39]. Using a pointed anode in contact with the surface of an electrolytic solution, a sheath of vapor is formed due to Joule heating. If the applied voltage is sufficiently high, the vapor breaks down and GDP forms. During GDP, various chemically active species such as HO•, HO•, and H_2O_2 are formed in high yields in the solution. It has been found that phenols, aniline, nitrobenzene, and dyes, etc., can be mineralized by using GDP. As reported in [37], reducing species are also produced in GDP. For example, an efficient reduction of hexavalent chromium (Cr(VI)) to the non-toxic trivalent state (Cr(III)) induced by GDP generated between a pointed platinum anode and the surface of an aqueous solution has been achieved [38]. Experimental results show that Cr(VI) could be smoothly reduced to trivalent state Cr(III) by using GDP with 500 V as the optimum operating voltage. The rate of Cr(VI) reduction was enhanced by either decreasing the solution pH or adding radical scavengers to the solution. At initial pH 2.0, 100 mg/L of Cr(VI) was completely reduced within 10 min of GDP treatment in the presence of 100 mg/L phenol. Fig. 3 shows the schematic diagram of the GDP reactor for Cr(VI) reduction [38].
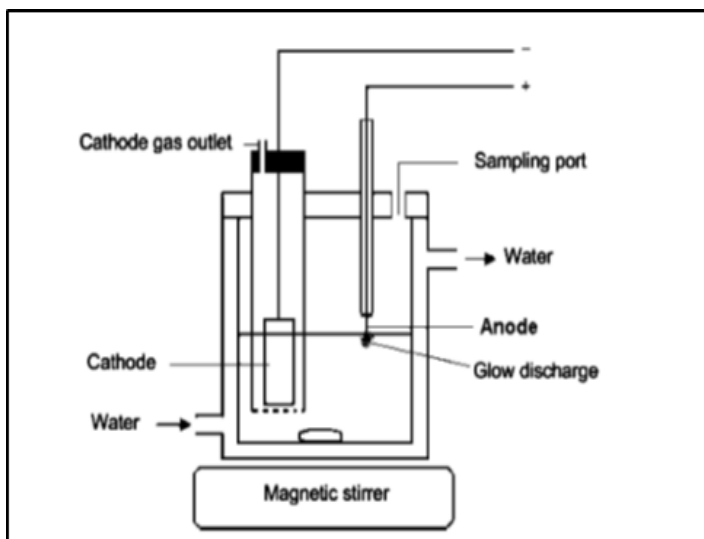
Figure 3: Schematic diagram of the GDP reactor for Cr(VI) reduction (from [38], with permission).

On the other hand, ultrasound is another technology that has been also used for water treatment, cleaning and degassing, extraction of biologically active compounds, food processing, advanced oxidation processes, synthesis of nanostructured materials, and redox reactions of actinide ions [5, 40, 41]. The usual technology spans frequencies in the 20 kHz–1 GHz range, but the sonochemical effects are usually observed in the more narrow range of 20 kHz-2 MHz. The ultrasound of higher frequencies is used for medical and material diagnostics. Sonochemistry does not arise from a direct action of ultrasonic waves on molecules, but rather from acoustic cavitation, i.e., cavitation is a set of consecutive events: nucleation, growth and violent collapse of microbubbles in liquids submitted to ultrasonic vibrations. There is a general consensus that the chemical and physical effects of power ultrasound are related to extremely rapid implosion of the cavitation bubbles occurring at the final stage of collapse. Acoustic cavitation does not only lead to chemical transformation, but also to light emission, known as sonoluminiscence (SL). Regarding sonoluminiscence (SL), two different types of can be distinguished: light emission from a cloud of bubbles (multibubble sonoluminiscence (MBSL)) and light emission from a single cavitation bubble trapped in a standing acoustic wave of relatively weak acoustic pressure (single bubble sonoluminiscence (SBSL)). The most recent spectroscopic studies on SBSL and MBSL reveal that the origin of extreme intrabubble conditions is related to non-equilibrium plasma formed inside the collapsing bubbles. In degassed water, the SBSL is enough to be visible to the naked eye. During MBSL in water saturated with noble gases such as Ar at low ultrasonic frequency, weakly excited plasma with vibrational distribution is formed. At high-frequency ultrasound, the plasma inside the collapsing bubbles exhibits behavior typical for strong vibrational excitation. Plasma formation during SBSL was observed in concentrated H_2SO_4 preequilibrated with Ar. The light emission spectra exhibit the lines from excited Ar atoms and ionized oxygen O_2^+; the formation of this last species is inconsistent with any thermal process. Furthermore, the SBSL spectra in H_2SO_4 show emission lines from Xe^+, Kr^+, and Ar^+ (with energies ranging from 26.0 eV to 34.2 eV), in full agreement with plasma hypothesis. The photons and the "hot" particles generated by cavitation bubbles enable the excitation of nonvolatile species in solutions increasing their chemical reactivity. However, the gas temperature obtained at the same conditions is only about 1 eV. Such a discrepancy cannot be understood by adiabatic heating during bubble collapse, but it can be explained by the formation of non-equilibrium plasma with an electron temperature higher than the gas temperature. This means that each cavitation bubble can be considered as a plasma chemical microreactor providing highly energetic processes at almost room temperature of the bulk solution. The photons and the "hot" particles produced inside the bubble enable the excitation of nonvolatile species in solutions, thus increasing their chemical reactivity. This quantum phenomenon produces a plasma far from the thermodynamic equilibrium with vibrational temperature of HO• much higher than the intrabubble gas temperature.

Conclusions

As repeated several times in the paper, use of plasma technologies are considered as promising remediation methods for water remediation. The techniques are environmentally friendly, simple and effective for decomposing toxic organic compounds or transforming inorganic pollutants into less noxious species, either in raw water or in wastewater. Reactions can be performed under atmospheric conditions, no additives are needed and the technologies emerge as opportunities for the plasma community to continue research and development on these issues.

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