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Pulsed Corona Discharge, a New and Effective Technique for Water and Air Treatment

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ABSTRACT: New technologies to solve environmental problems are in demand nowadays. Existing methods cannot always be used because they are too expensive, have unwanted byproducts or simply don't work. A new approach is to create active species in situ so transport losses can be avoided. Pulsed corona discharges is such a method. Its principle is that high energy electrons are created during the propagation phase of the streamer. These electrons dissociate molecules and create radicals such as O, OH, N_2 (A) and indirectly HO2, O3 and others. All these radicals start chemical reactions which cause, mainly, oxidation of impurities present in the gas or water. There in an attempt in this paper discuss the pulsed corona discharge as an effective technique in water, gas and air treatment based on some related researches which investigate various aspects of water and gas cleaning using this technique. So the methodology used in this paper is descriptive, library and analytical. The required materials and information for this research have been complied using related scientific papers.

Keywords: pulsed corona discharge, water treatment, ozone generation, Fenton reaction, air pollution control

INTRODUCTION

International and national regulations are becoming increasingly severe concerning the quality of the environment and the amount of pollutants discharged into wastewater streams form different process plants. Many of these pollutants are of hazardous or toxic nature posing serious problems to the environment. Thus it becomes necessary from both the legal and ecological points of view to control carefully wastewater effluents and to develop new techniques to treat the pollutants (Lukes, 2001).

In addition, the increasing concern about the environmental problems facing the world such as acid rain, ozone depletion, global warming and smog has determined the interest toward developing new and more effective gas cleaning technologies (Mizuno *et al.,* 1995).

High power pulsed corona is a promising type of high-voltage discharge that can be well controlled and that has interesting environmental applications. With pulsed corona discharges energy is deposited in a gas or in a liquid in a highly concentrated form.

The research on corona discharge already has a considerable history. Practical applications receive more and more interest in recent years, mostly for environmental applications such as gas water cleaning. Large-scale applications necessitate the use of pulsed corona discharges. In order to be competitive in the large market of cleaning systems the pulsed power supplies must be very efficient, reliable and compact (Briels *et al.,* 2007).

Operating over a wide temperature and pressure range pulsed corona generates electrons, free radicals, excited molecules and UV radiation. Direct bond cleavage or interaction through radicals can break down various hazardous organic pollutants. The corona discharge greatly enhanced the absorption of various gases (SO_2, NH_3, H_2S, Cl_2, CO_2) due to the decrease in gas-phase mass-transfer resistance. Nonhazardous fragments or compounds that can be further treated using conventional techniques are the result (H_2O, CO_2, NO_2, HCI). Also, pulsed corona discharge technique have been applied to oil products removal dependent on treatment parameters (Malik, 2003, Bisht *et al.,* 2014, Kornev *et al.,* 2014).

Pulsed corona in water leads to the formation of high local electric fields, electrons eaq-, Ho and OHo radicals and the repetitive formation of shock waves. In gases the energized electrons produce radicals through dissociative electron attachment or through electron impact dissociation '.

Areas considered for corona applications are related to various industrial emissions and (cleaning) processes: emissions of VOC and NO, flue gas, off-gases and soil vapor extractions, auto (and diesel) emissions, incinerators, cleaning of bio gases, high-temperature gas processes, industrial water pollutants, process water, drinking water, sterilization and disinfection, food pasteurization.

Advantages of pulsed corona treatment are: simultaneous removal of' several pollutants, high destruction efficiency, no demands on temperature and pressure, not vulnerable, no damage from high loads, widely applicable, simply installed, compact, little service, can be small scale, no additives (van Heesch *et al.,* 1997).

The rest of this paper is organized as follows. Section 2 provides a general description of the pulsed corona discharge technique. In Section 3, we describe a research conducted on high-power pulsed corona for treatment of pollutants in heterogeneous media. A new technique of phenol degradation using pulsed streamer corona discharge in combination with the Fenton reaction is described in section 4. In section 5, we review a study on generation of ozone by pulsed corona discharge over water surface in hybrid gas-liquid electrical discharge reactor. Section 6 provides the results of a research on analysis of drinking water after treatment by corona discharge. In section 7, air pollution control using pulsed corona discharge is presented, and a brief conclusion in Section 8 concludes the paper.

A. General description of the pulsed corona discharge technique

The electrical discharges, particularly pulsed corona discharges (PCDs) seem to be the most promising technique water and gas cleaning because of the following advantages.

-PCD can effectively oxidize a wide variety of toxic organic compounds in air and in water.

-Pulsed electric fields can kill a wide range of harmful microbial contaminants in water.

-They involve simple technology and relatively lower installation cost.

The PCD technique has become the subject of a number of studies in the recent past and it is being tested on an industrial scale.

In a PCD reactor, high voltage pulses of sharp rise time and short duration are applied across the electrodes, which accelerate (energize) free electrons. The energized electrons ultimately collide with and ionize, dissociate or excite the ambient molecules thus producing more free electrons and finally an electron avalanche called a streamer. Since the mean free path is longer in gas phase than in liquid phase, the streamers initiate in pre-existing micro-bubbles or in the micro-bubbles produced by local heating of the liquid. Dissociation of the ambient molecules produces free radicals, e.g. H_2O dissociates into H• and OH•. Bubbling oxygen gas in the discharge gap of the reactor results in the production of O• by dissociation of O_2 and it also boosts the rate of production of Ho and OH•. Interaction of the free radicals, such as Ho, OH•, O•, etc. with each other and with ambient molecules also results in the production of O_3 and H_2O_2. These species, i.e. high-energy electrons, H•, OH•, O•, O_3, H_2O_2, neutral molecules in an excited state, and ionic species produced by the corona discharges, are collectively called 'chemically active species'.

The chemically active species, particularly OH•, attack and decompose toxic organic compounds in water by a mechanism quite similar to that taking place in an atmospheric water droplet. UV-radiation, shock waves and supercritical water conditions are also produced by the electrical discharges in water, which make the environment more suitable for the decomposition of toxic organic compounds. The capability of PCDs to decompose aqueous pollutants has been demonstrated by the decomposition of phenol and some other organic compounds, and by decolourization of organic dye in water.

A major drawback of PCD technique is the relatively higher energy consumption as compared with other competitive processes. Research in the recent past shows that the energy efficiency can be improved by operating the corona discharge in the gas phase close to water surfaces rather than in the water and by changing the discharge mode from corona to spark discharges. Similarly, energy efficiency was found to improve by the addition of hydrogen peroxide, activated carbon or ozone in the discharge gap of the reactor (Malik, 2003, Malik *et al.,* 2001, Malik *et al.,* 2002).

B. High-power pulsed corona for treatment of pollutants in heterogeneous media

A technical overview of a European project on pulsed corona (PC) treatment of polluted streams has been studied in (Pokryvailo *et al.,* 2006) which concentrates on increase of chemical efficiency and development of reliable and affordable pulsed-power systems having a commercial potential. Versatile high-power systems that are capable of cleaning both aqueous and gaseous streams in heterogeneous media, in either a corona above water reactor or an aerosol reactor (Pokryvailo *et al.,* 2004) have been developed. Both reactors are capable of high-phenol removal yields from aqueous streams and increase the biochemical oxygen demand/chemical oxygen demand ratio for several nonbiodegradable wastewaters to such degree that further biodegradation becomes possible. The best result obtained with four CAW modules is 33% conversion with a yield of 200 g/kWh. The aerosol reactor has achieved 99% conversion at 18 g/kWh. These yields can be increased by the addition of H_2O_2, Fe_2SO_4, or NaOH, depending on the individual application.

It has been demonstrated that the reactors can increase the BOD/COD ratio (biochemical oxygen demand (BOD)/chemical oxygen demand (COD) ratio) in some cases of nonbiodegradable wastewater to such degree that further biodegradation becomes possible. However, the process was not successful on all wastes. A complication that was found is the formation of nitrogen oxide. Using oxygen in the reactors instead of air can solve this problem.

With gaseous streams polluted by toluene, styrene, and malodorous constituents, the following has been demonstrated:

1) The PC combined with a catalyst achieves the reduction rates of toluene that are higher than 99%.

2) The corona-discharge reactor in combination with the cold catalyst showed very high odor-removal efficiency.

3) No adsorption in the catalysts was observed.

From a technical point of view, the system scale-up appears to be straightforward. A first step of scale-up is demonstrated, and the cleaning efficiency even improves due to better mixing and shorter treatment times. The estimated price of water treatment in systems that were scaled up to 50 m^3/h is 2 C/m^3. Incineration of chemical waste costs up to approximately 500 C/m^3, so the PC water-treatment technology for low pollutant concentrations may become highly competitive.

Reliable operation of all-solid-state compact nanosecond pulsers has been demonstrated. The pulsers make use of magnetic compression techniques, and owing to a fast primary switch, only one compression stage suffices for the forming of narrow HV pulses. For the second phase of the project, a high-power pulser was designed. It generates 60-kV 100-ns wide pulses across a 100-Ω load. The rise time is 15 ns, and the pulse energy is 4.0 J at an 82% efficiency with a resistive load. Witha plasma load, the efficiency is 62%. The PRF that is limited to 500 Hz by the current capability of the freewheeling diode may be doubled with more powerful diodes. Then, the power level will reach 4 kW to accommodate the requirements of a number of applications; this is realizable with forced air-cooling. With water-cooling, the average power may be further increased .

C. Phenol degradation in water by pulsed streamer corona discharge and Fenton reaction

A number of studies have utilized pulsed discharges generated either in the water bulk or in the gas phase over the water surface or inside gas bubbles introduced into the water bulk, to produce strongly oxidative species, such as hydroxyl radicals (OH), O atoms, ozone (O_3) and hydrogen peroxide (H_2O_2) with a goal of the degradation of aromatic compounds present in the water (Panorel 2013). The aromatic organic compounds present in the water are oxidized by these active species to simpler compounds, which further can be easily removed from the water using conventional bio-filters (Elsawah *et al.,* 2013, Christos *et al.,* 2008). It has been reported that addition of iron salts to the water processed by the pulsed streamer corona discharge enhanced degradation of several organic compounds. This was due to the known Fenton reaction which have been widely used to the degradation of a wide range of organic compounds including herbicides, pesticides, dyes, phenol and chlorinated phenols. They have been utilized in combination with not only pulsed streamer corona discharge but also UV/TiO_2 photocatalysts and ultrasounds.

In the presence of iron salts in the water, hydrogenperoxide, the most abundantly produced oxidant by the streamer corona discharge, is converted to hydroxylradical, which is much stronger oxidant than H_2O_2, through the Fenton reaction :

$$Fe^{2+} + H_2O_2 \rightarrow Fe^{3+} + OH^- + OH. \qquad (1)$$

In most investigations concerning the application of the pulsed streamer corona discharges in water for organic compound decomposition, the deionised water with additives regulating conductivity was used (Kušic *et al.,* 2005, Chen *et al.*, 2004, Dors *et al.,* 2007).

Since the studies on the pulsed streamer corona discharges in water are aimed at their application to the drinking water and wastewater purification in the tap water used in the experiment. In this research, results of the investigation of iron ions influence on phenol oxidation in both the distilled and tap water by pulsed positive streamer corona discharge are presented in fact, a novelty of this work is presenting an influence of the pulsed streamer corona discharge in combination with the Fenton reaction on phenol degradation in the tap water.

In results of the investigation of iron ions influence on phenol oxidation in both the distilled and tap water by pulsed positive streamer corona discharge are presented. The results of the investigations showed that:

-The Fenton reaction enhanced significantly phenol degradation rate (from 42% to 78%) only in the distilled water of low conductivity.

-In the tap water, which has relatively high conductivity (600μS/cm), the Fenton reaction was weak and had to be initiated by lowering the initial pH from alkaline (7.6) to acidic (4.1).

- Enhanced phenol degradation to organic acids resulted in lower pH (in the water of a low conductivity pH reached value optimal for the Fenton reaction, i.e. pH = 4.1).

-The presence of iron ions in the water did not influence the phenol degradation products, which are dihydroxyphenols (primary products) and organic acids (secondary products).

In general, It can be said that the Fenton reaction combined with the pulsed streamer corona discharge is not effective in the purification of drinking water. Thus, other chemical activators or other types of discharges are needed for this purpose.

D. Generation of ozone by pulsed corona discharge over water surface in hybrid gas-liquid electrical discharge reactor

Advanced oxidation water treatment processes that utilize highly reactive radicals such as the hydroxyl radical are of increasing interest for the degradation of organic compounds in contaminated and polluted waters. Various combinations of ozone, hydrogen peroxide, and UV light have been studied as means to produce hydroxyl radicals in water and to effectively degrade many organic compounds (Wei *et al.,* 2014, Sretenovic *et al.,* 2012).

Ozone has typically been produced by electric discharge plasma reactors placed upstream of the water treatment process, and the gas phase ozone thus produced has been spared into the liquid alone or in combinations with the addition of hydrogen peroxide and UV light.

In attempts to bring the ozone generation step closer to the ozone utilization step and to develop plasma reactors that may be suitable for direct water treatment a number of different types of plasma reactors utilizing electrical discharges have been studied. For example, AC, DC or pulsed electrical discharges have been generated in a variety of electrode geometries either directly in the liquid phase or in the gas phase in close proximity to the liquid surface. Electrical discharges generated directly in the liquid were demonstrated to initiate a variety of physical and chemical effects in water including the high electric field, intense ultraviolet radiation, overpressure shock waves and, of particular importance, formation of various reactive chemical species such as radicals (H·, O·, OH·) and molecular species (H_2O_2, H_2, O_2, O_3). Production of these chemical species has also been reported for electrical discharges generated above the liquid water surface. These reactive species and physical conditions in turn have been shown to be effective at degrading a variety of organic compounds and in the destruction and inactivation of microorganisms in water (Locke *et al.,* 2006).

Hybrid electrical discharge reactors utilize both the gas phase non-thermal plasma formed above the water surface and direct liquid phase corona-like discharge in the water. The main advantage of these reactors is the production of the same chemical species and physical factors as initiated by the individual gas and liquid phase discharges. Some experiments have demonstrated the formation of ozone in the gas phase and H_2O_2 and OH radicals in the liquid phase in these reactors (Lukes, *et al.,* 2004). The combined action of these reactive species, as well as possible reaction processes occurring at the gas-liquid interface, can lead to enhancement of the overall efficiency of the electrical discharge process for the removal of pollutants from water

In order to optimize the power delivered into the two discharges generated in the gas and the liquid phases and to effectively tune the power supply to the hybrid discharge reactor we have constructed a gas-liquid discharge reactor with separately charged electrodes in the gas and liquid phases using two pulse power supplies, which allow us an independent control of the gas and liquid phase discharges. In (Lukes *et al.,* 2007). the formation of ozone by the gas phase discharge over the water surface in this hybrid discharge reactor has been investigated. The effects of discharge gap spacing, electrical power applied to the reactor and gas phase composition (oxygen and O_2/Ar, O_2/N_2 mixtures) on ozone production has been determined.

In this research, it has been found that, although the efficiency of ozone production was significantly enhanced using high voltage of shorter pulse width, the obtained ozone yields (~10-20 g/kWh) are still much lower than yields reported for ozone production using corona discharges in dry oxygen or air (~50-150 g/kWh). It is apparent that water vapor formed through the vaporization of water surface by the gas phase discharge is one of the most important reasons of this state. However, the formation of OH radicals in water vapor or at gas-liquid phase interface is also desired since they significantly contribute in degradation of organic compounds dissolved in water. Thus, a compromise between production of ozone in high concentrations and with high energy efficiency, and production of OH· radicals by the gas phase discharge generated above aqueous solution has to be considered in further development of hybrid gas-liquid discharge reactors. It is also emphasized that rather than development of a new type of ozone generator the final objective is the most efficient utilization of chemically active species (O_3, OH· and O radicals, and other species) formed by the gas phase discharge in the removal of pollutants from water.

E. Drinking water analysis after treatment by corona discharge

An electrical discharge technique (pulsed discharge in water) has been used in varies applications such as sterilization, treatment of the wastewater and switching (Buogo *et al.,* 1998, Lu, *et al.,* 2002, Gu *et al.,* 2002). The high voltage electrode (copper rod) was connected to PFN pulse generator placed above the surface of the water while the spherical electrode was connected to the ground submerged in water vessel. The rotating spark gap switch was designed and constructed to get pulse repetition rate of 25 Hz. The water samples (that were taken from Egyptian atomic energy water station before and after chemical treatment using chlorine) were divided equally into two samples: one unexposed control sample and tested sample, which was exposed to a number of pulses of corona discharge treatment system.

Table 1: Concentrations of trace metals of the water samples (tested and control).

Trace metal	Value (control)	Value (tested)
Copper	< 0.01 mg/l.	0.6 mg/l
Iron	0.03 mg/l.	60 mg/l
Zinc	0.1 mg/l	< 1.0 mg/l
Manganese	<0.01 mg/l.	0.03 mg/l
Magnesium	14.4 mg/l.	19.2 mg/
Calcium	28 mg/l.	32 mg/l

At the end of exposure time, the water samples collected were analyzed (bacteriological and chemical assessment in water). As regarding the chemical analysis, the concentrations of trace metals ions in the drinking water samples (control and tested) are presented in Table 1. The average physical and chemical properties of the drinking water samples including pH, conductivity, sulphate, chloride, total hardness, total dissoloved solids (TDS), total alkalinity, nitrite, nitrate and chloride from these samples where given in Table 2. As regards the Bacteriological Analysis, it determines the presence or absence of certain bacteria in a sample of water. By determining this, it can determined whether or not the water supply, from which the sample was collected, is safe to drink. The Provincial Laboratory of Public Health now performs 2 standard tests on water samples submitted for bacteriological analysis.

The control sample that was taken from the station of water was sent directly to plasma laboratory without any treatment. It was found that it contains +190 coliform bacilli/100 mL so it is not in conformity with World Health Organization (WHO) guidelines. The sample after plasma treatment is free from any fecal and plant pollution and it is in conformity with World Health Organization (WHO) guidelines (El-Aragi, 2005).

Table 2: Average physical and chemical properties of the samples (control and tested).

	Value (control)	**Value (tested)**
pH	7.54	5.75
conductivity	331 μS/cm	282 μS/cm
sulphate	45 mg/l	< 5 mg/l
chloride	30 mg/l	60 mg/
Total hardness	130 mg/l	160 mg/l
Total Dissolved Solids	215 mg/l	207 mg/l
Alkalinity	110 mg/l	30 mg/l
NO_2 nitrogen (Nitrite)	< 0.005 mg/l.	< 0.1 mg/l
NO_3 nitrogen (Nitrate)	0.04 mg/l	40 mg/l

F. Air pollution control by pulsed corona discharge

Air pollution caused by gas emission of pollutants produced by a variety of sources including coal, oil and gas burning electric power generating plants, motor vehicles, diesel engine exhausts, paper mills and steel and chemical production plants must be substantially reduced as mandated by recent national legislation and international agreements. These pollutants are major causes of acid rain, planet warming, smog and also increasingly are being thought of as detrimental to human health, to vegetation growth and to fresh water lakes (Hackam and Akiyama, 2000).

Several researches have been conducted on removal of pollutants from air using pulsed corona discharge. In (Mok and Ham, 1998), conversion of NO to NO_2 in air by a pulsed corona discharge process was studied in which a kinetic model was proposed to describe the behavior of pulsed corona discharge process applied to the removal of nitrogen oxides. The proposed model in the mentioned research takes into account radical production at every pulse and NO removal by the radicals. Some experimental results of repetitive nanosecond-pulse discharge with different duration and voltage in point-plane gaps in open air also in helium and nitrogen were reported. The results indicated that the discharge has various modes such as corona, diffuse and spark. Moreover, the combined removal of SO_2, NOx and fly ash form simulated flue gas using pulsed streamer corona.

In fact, a pulse-energized electron reactor (PEER) was developed for the combined removal of SO_2, NOx and fly ash form effluent gases. The performance of the wire -to-cylinder

PEER used in the mentioned research was rates as satisfactory and effective (Shao *et al.*, 2013, Clements, *et al.*, 1989).

CONCLUSION

Corona discharges have already been in use for gas treatment for a long time. The most important applications are ozone formation, dust precipitation and the conversion of environmentally harmful species, such as NOx, SO_2 and many hydrocarbons. The mechanism in the case of these chemical conversions is almost always oxidation. Reduction can in principle be realized in a discharge but only at high energy cost and under conditions that are very exceptional in practice.

New oxidation techniques for water cleaning are also under development. These include treatment with ozone, supercritical water, UV photons, electron beams, x-rays, ultrasound and electrical discharges in several forms. The electrical discharges, particularly pulsed corona discharges (PCDs) seem to be the most promising because of the following advantages.

-PCD can effectively oxidize a wide variety of toxic organic compounds in air and in water.

- Pulsed electric fields can kill a wide range of harmful microbial contaminants in water.

-They involve simple technology and relatively lower installation cost.

The PCD technique has become the subject of a number of studies in the recent past and it is being tested on an industrial scale. For these reasons, we discussed pulsed corona discharge as a promising technique for water and gas cleaning, and reviewed some of its applications bases on the results of several researches conducted in this regard.

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