*Desalination and Water Treatment*
www.deswater.com

doi: 10.5004/dwt.2018.22114

109 (2018) 337–343
March

# Degradation of acetaminophen in water by using dielectric barrier discharge

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Received 7 December 2016; Accepted 26 February 2018

## ABSTRACT

Acetaminophen is the most commonly used drug worldwide for relieving pain and fever. Consequently, it has been detected in groundwater in different areas of the world. In this study, acetaminophen removal from water using a dielectric barrier discharge (DBD) system, operated in the open air, was investigated. Our results show that treating water for only 0.5 min at 1, 5, 25, 50, 75 and 100 mg $L^{-1}$ acetaminophen concentration resulted in the removal of 98.85%, 99%, 89.16%, 42.84%, 33.86% and 32.87% of the initial acetaminophen amount, respectively. These removal rates increased significantly when the treatment duration was extended to 2 min, reaching 100%, 99.40%, 99.66%, 90.62%, 85.89% and 77.09% of the initial amount of acetaminophen at similar concentrations, respectively. The initial pH of the solution causes a negligible variation in the degradation of the acetaminophen; therefore, it was not found as an important parameter. The pH of the solution decreases during the plasma treatment. The final pH of different solution was always within 15%, regardless of the initial concentration. Optical emission spectroscopy of the DBD during operation revealed the prominent reactive species and possible reaction pathways. The acetaminophen removal efficiency of the DBD system used in this study for water treatment is higher than that reported for other systems employed in previous studies. Moreover, as the discharge is generated here in the open air, it is very cost-effective. Further studies are recommended to investigate DBD operated in open air for the removal of other organic pollutants from water.

*Keywords:* Paracetamol; Water treatment; Decontamination; Plasma

## 1. Introduction

Acetaminophen ((*N*-(4-hydroxyphenyl)acetamide), $C_8H_9NO_2$, Fig. 1), also known as paracetamol, is a common and widely used drug for relieving minor pains and fever. Its consumption in some areas is estimated at approximately 25–50 tons per million individuals [1]. Due to its release to the environment, acetaminophen is among the most frequently detected drugs in groundwater worldwide [2,3]. Lopez et al. [4] reported the occurrence of acetaminophen in 27% of groundwater samples collected from 494 different sites in France during 2011, at concentrations below the threshold of toxicological concern of 0.1 μg $L^{-1}$. The occurrence of acetaminophen in surface water at low

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concentrations (0.001–0.003 μg $L^{-1}$) was reported in Yamaska river in Canada during the summer of 2005 [5]. Moreover, Gottschall et al. [6] reported the presence of acetaminophen at a low concentration of 0.013 μg $L^{-1}$ in tile water after application of municipal dewatered biosolids to an agricultural land in Ottawa (Canada), while it was not detected in groundwater at the same experimental site. However, as a part of a similar study, acetaminophen was detected at the concentration of 0.146 ± 0.089 μg $L^{-1}$ in the runoff samples after 36 d of agricultural land treatment with biosolids [7]. The primary sources of acetaminophen and other drugs in sewage water are possibly excretion of ingested drugs, disposal of unused or expired tablets and the release of effluents from drug manufacturing facilities [8]. Use of dewatered municipal biosolids in agricultural lands is permitted in different countries. While such practice enhances the soil physicochemical properties and provides nutrients to plants, it is a way for some persistent pharmaceuticals to be released in the environment with the municipal biosolids, thus reaching the groundwater [9,10]. A recent study showed that the application of dewatered municipal biosolids to soil sub-surface has a significant effect on the dissipation rate of some biodegradable compounds; this practice is recommended to accelerate the degradation of some molecules and reduce their potential environmental risk [11]. Since detection of minute quantities of pharmaceuticals in the groundwater has become possible, the awareness about their presence and effects has increased [12]. The outcomes and the environmental impacts of the pharmaceutical compounds have also been studied. Authors of extant studies in this field have detected acetaminophen in sewage effluents in Europe and the United States, at concentrations ranging from 6 to 10 μg $L^{-1}$. In treated wastewater effluents, acetaminophen concentrations as high as 200 μg $L^{-1}$ have been reported [13]. It was present at 71 ng $L^{-1}$ in drinking water samples in France during 2011 [3].

Even if its ingestion does not affect human health, it can have an adverse impact on the aquatic life [14]. Yet, despite these risks, we have yet to find a technically and economically viable solution that would eliminate this highly potent interference of pharmaceuticals with ecological systems.

While the technical viability of multiple methods has been investigated, their economic impacts are rarely studied [15]. Some noteworthy methods include advanced oxidation processes (AOPs) for the degradation and mineralization of acetaminophen (by oxidizing acetaminophen in aqueous solutions) such as $H_2O_2$/UV [16], anodic oxidation [17], ozonation [8,16], photo-Fenton [15], fluidized-bed Fenton [18] and photocatalytic oxidation [19]. AOPs are very promising due to their safety and reliability in removing acetaminophen residues from the water. In AOPs, acetamide and hydroquinone were consistently among the identified aromatic products, which suggests the primary OH-radical attack on the aromatic ring [20,21]. Continued oxidation of the aromatic products formed during initial oxidation of acetaminophen will generate carboxylic acids (e.g., malic, malonic, hydroxyacetic, formic, oxamic, oxalic), which will be finally mineralized to $CO_2$ and $H_2O$ [22]. Among the existing AOP techniques, non-thermal plasma methods such as pulsed corona discharge (PCD) are particularly favorable for commercial application due to the simple yet efficient means of generating in situ OH-radicals and ozone. Ono and Oda [23] established that OH-radicals are formed due to the collision of water molecules with electrons or reaction with O, O(1 D) or H atoms. The concentration of micro-discharges on the surface of the treated water [24] implies formation of radicals, thus involving short-living oxidants.

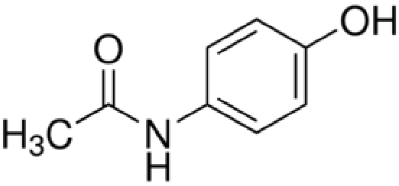

Fig. 1. Structure of acetaminophen.

Different types of plasma systems have been used for pharmaceuticals degradation. However, only PCD systems have been applied specifically to acetaminophen [25]. Usually, in such systems, a particular gas such as oxygen is used [26]. However, this makes the process expensive and impractical. In this study, we have investigated the effect of open-air dielectric barrier discharge (DBD). The objective of this research was to test the efficiency of open-air DBD in the degradation of acetaminophen in comparison with the previous plasma systems. Acetaminophen concentration in the solution was similar to that employed the previous studies to ensure a reliable comparison of energy efficiencies. The emission from the plasma was analyzed when the variable concentration of acetaminophen was used in the solution. The effects of pH adjustment and treatment time were also evaluated.

## 2. Materials and methods

### 2.1. Chemicals

Acetaminophen (4′-hydroxyacetanilide, 4-acetamidophenol, *N*-(4-hydroxyphenyl)acetamide, *N*-acetyl-4-aminophenol, APAP, paracetamol) was purchased from Sigma-Aldrich (Jeddah, Saudi Arabia).

### 2.2. Experimental details

A lab-scale DBD system (Fig. 2), developed by Nanjing Suman Company (Jiangsu Sheng, China), was used for the

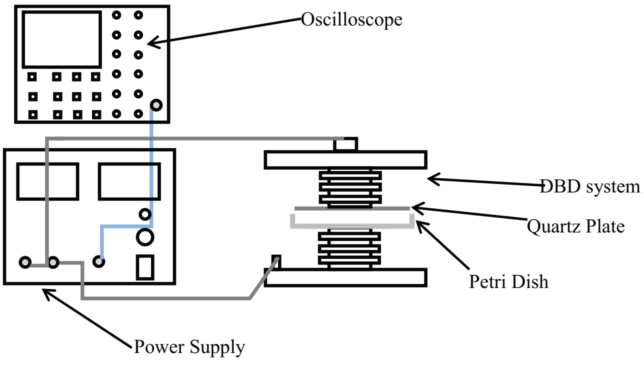

Fig. 2. Schematic diagram of the lab-scale dielectric barrier discharge (DBD) system used for the water treatment.

experiment. The system consists of stainless steel electrodes arranged in a parallel plate configuration. During the measurements, 10 mL of the solution was kept in a glass petri dish on the bottom electrode, while the top electrode was covered with a 1-mm thick quartz plate. The gap between the two dielectrics (quartz plate and petri dish) was ~7 mm, while the distance between the solution surface and the quartz plate was ~ 5 mm. The plasma was generated with a high voltage (kHz) power supply. The voltage was measured with Testec HVP-15HF High voltage probe and charge across a 0.47 µF capacitor on a Lecroy WaveSurfer 24x-A Oscilloscope. Finally, the plasma power was calculated using the standard Lissajous figure method [25]. All the measurements were performed at 40 W power and 10 kHz frequency. The energy density (ED) was calculated as $(P \times T)/V$; where $P$ is the plasma power, $T$ is the treatment time and $V$ is the volume of the solution in the reactor. After the plasma treatment, pH, the quantity of pharmaceutical and total organic carbon (TOC) within the treated solution were measured.

### 2.3. Analysis

Acetaminophen standards were prepared in Milli-Q water and were stored at 4°C until required for experiments. Samples were analyzed by ultra-performance liquid chromatography UPLC (Ultimate 3000, Thermo Scientific, Jeddah, Saudi Arabia) with ultraviolet detection (UPLC-UV). Thermo Scientific Acclaim™ 120 A EC-C8 (4.6 mm × 150 mm, 5 µm pore size, Jeddah, Saudi Arabia) was used, and the UV detector (Ultimate 3000, Thermo Scientific) was set at 243 nm. The injection volume was set at 10 µL by using auto-sampler (Ultimate 3000, Thermo Scientific). The mobile phase consisted of methanol and water in the 1:4 ratio. The column temperature was set at 30°C, the flow rate was 1 mL $min^{-1}$, and acetaminophen retention time was 4.2 min. The calibration curve was linear at six calibration standards of 0.05, 1, 5, 25, 50 and 75 mg $L^{-1}$. Degradation products were determined by liquid chromatography-mass spectrometry LC-MS instrument (LCQ fleet ion trap, Thermo Scientific). Samples were directly infused to ion trap MS without chromatographic separation using 500 µL syringe at a flow rate of 10 µL $min^{-1}$. Analysts in the samples were ionized using positive ESI mode. The I spray voltage was 5 kV, while the capillary temperature and voltage was 275°C and 10 V, respectively. Samples were screened for compounds using full-scan mode for *m/z* in the 100–500 range.

## 3. Results and discussion

### 3.1. Degradation of acetaminophen in water with time at different concentrations

The removal rate of acetaminophen from water using 40 W DBD plasma treatment in the ambient air increased significantly with time. Specifically, acetaminophen concentration decreased from 100 mg $L^{-1}$ (initial concentration) to 64.2 mg $L^{-1}$ after 0.5 min, after which it decreased gradually to reach 4.3 mg $L^{-1}$ after 4 min (Fig. 3). This decline in the concentration is due to acetaminophen degradation with time until its complete mineralization. The removal efficiency of acetaminophen from water was significantly higher at lower concentrations when treated at two different durations (0.5 and 2 min). This higher efficiency at lower concentration might be attributed to the fast dissipation of acetaminophen from less saturated solutions (Fig. 4). The acetaminophen removal rates when water was treated for only 0.5 min at 1, 5, 25, 50 and 75 mg $L^{-1}$ concentrations were 98.85%, 89.16%, 69.16%, 42.84% and 33.86% of the initial amount, respectively. These rates increased significantly when the treatment time was increased to 2 min, to reach 100%, 99.40%, 99.66%, 90.62% and 85.89% of the initial acetaminophen amount for the previously mentioned concentrations, respectively (Fig. 4). Degradation products were determined by LC-MS as shown in Fig. 5. The mass fragmentation of acetaminophen resulted in three fragment peaks, along with the molecular ion peak at *m/z* 151.8 (Fig. 5). The first fragment was observed at *m/z* 118.8 due to the removal of one –OH and one $-CH_3$

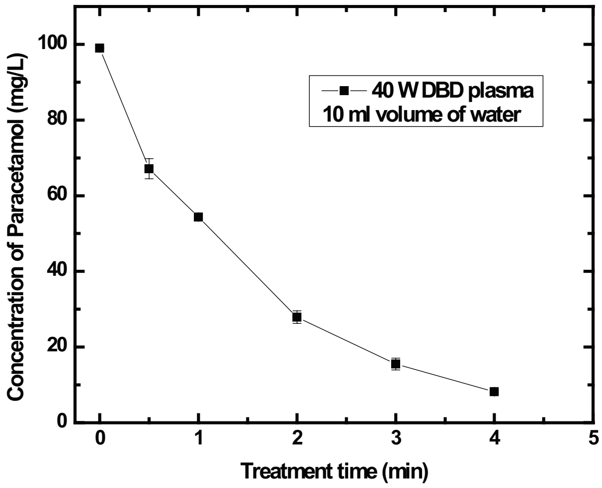

Fig. 3. Removal of acetaminophen from water as a function of time, using a 40 W DBD plasma operated in ambient air.

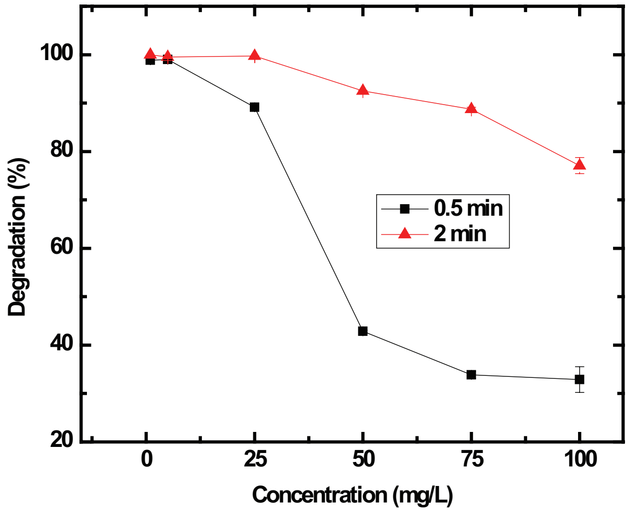

Fig. 4. Degradation rate of acetaminophen from water at various concentrations (1, 25, 50, 75 and 100 mg $L^{-1}$) treated for 0.5 and 2 min with DBD.

group, giving rise to the peak at *m/z* 118.8. This fragment was further broken into two fragments at *m/z* 99.9 and *m/z* 76.8 (Fig. 5). The first fragment at *m/z* 99.9 was observed due to the cleavage of the benzene ring to produce an unsaturated ketone. The second fragment was due to the removal of –NHC=O group from the molecule, resulting in a benzene ring with high electron density as free radical (*m/z* 76.8). The relatively fast acetaminophen dissipation (about 100% in 2 min) in the system employed as a part of this study indicated no toxicity changes following treatment, in contrast to the treatment by chlor(am)ination. Ding et al. [27] reported the formation of toxic disinfection carbonaceous and nitrogenous byproducts, such as trihalomethanes, haloacetonitriles and haloacetamides, during chlor(am)ination of acetaminophen in aqueous solution.

The acetaminophen removal rate close to 100% of the initial amount from water treated for 2 min for the concentrations in the 1–25 mg $L^{-1}$ range is very promising, given the efficient energy use by the DBD (9.6E05 J $L^{-1}$). The short treatment time and cost-effectiveness (no gas utilized in the system), along with the low detected acetaminophen concentrations in groundwater, the influents and effluents of wastewater treatment plants (0.001–30 μg $L^{-1}$) [2,28,29] make these results very useful. Mai et al. [30] reported the acetaminophen removal rate of 87.54% of the initial amount added in water using a nitrifying trickling filter system. Wang et al. [31] reported a higher removal rate of this compound at 1 mg $L^{-1}$ concentration in water by ferrate (VI), noting that it reached 99.6% of the initial amount after 60 min. In our study, at 1 mg $L^{-1}$ concentration, the 98.85% and 100% removal rate was achieved after 0.5 and 2 min, respectively (Fig. 3). The total organic carbon removal rates from aqueous solutions in all experiments were negligible (data not shown). No data is reported in pertinent literature regarding the use of DBD in acetaminophen removal from water. Liu et al. [32] used a similar plasma system to remove iopromide, an organic compound used as a contrast medium, from water using nitrogen as carrier gas, at an ED of 1.5E05 J $L^{-1}$ to reach an efficient removal of 98.8% of the initial acetaminophen amount after 10 min of treatment. This removal rate was less than 40% at 2 min of treatment, keeping the ED constant. On the other hand, when the same plasma DBD system was used ex situ for the removal of antiepileptic drug carbamazepine from aqueous solution, the degradation rate was 10% of the initial amount after 3 min. However, the degradation rate was only 81% of the initial value after 60 min of treatment with the in situ discharge [33].

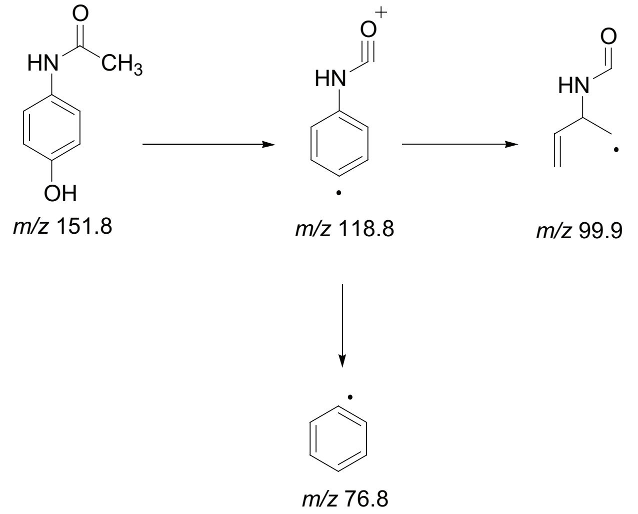

Fig. 5. Mass fragmentation pattern of acetaminophen.

### 3.2. Effect of pH on degradation of acetaminophen in water

pH value of the aqueous solution might affect the dissipation pathways of pharmaceuticals. In this study, we investigated acetaminophen degradation at three pH values (4, 7 and 10) as a function of treatment time (0.5, 1, 2, 3 and 4 min) in 100 mg $L^{-1}$ aqueous solution. Our results showed that the initial pH value of the acetaminophen aqueous solution had no significant effect on its degradation. The removal rates of acetaminophen from water at pH 4 were 32.78%, 49.74%, 73.28%, 92.81% and 99.68% after 0.5, 1, 2, 3 and 4 min of treatment with DBD, respectively. These values were close to the rates obtained at pH 7 (32.87%, 45.68%, 72.09%, 88.50% and 95.82%, respectively) using the same treatment time. The pH value of treated aqueous solution decreased with the plasma treatment time from 7 to 2.5 after 0.5 min of treatment, after which the pH value decreased gradually to reach 1.5 after 4 min (Fig. 6). On the other hand, at pH 10, the acetaminophen removal rate was as low as 24.13% after 0.5 min of treatment. Later, the rates were close to those obtained at pH 4 and 7, resulting in a removal efficiency of 44.88%, 84.4%, 90.1% and 96.2% of the initial acetaminophen amount after 1, 2, 3 and 4 min of treatment with DBD (Fig. 7). Results shown in Fig. 8 indicate that the pH of the treated solution decreased as the treatment time increased at all investigated concentrations (1, 5, 25, 75 and 100 mg $L^{-1}$). Our results are in accord with those reported by Salehi and Moussavi [34], who noted that acetaminophen removal using adsorption onto $NH_4Cl$-induced activated carbon was not pH

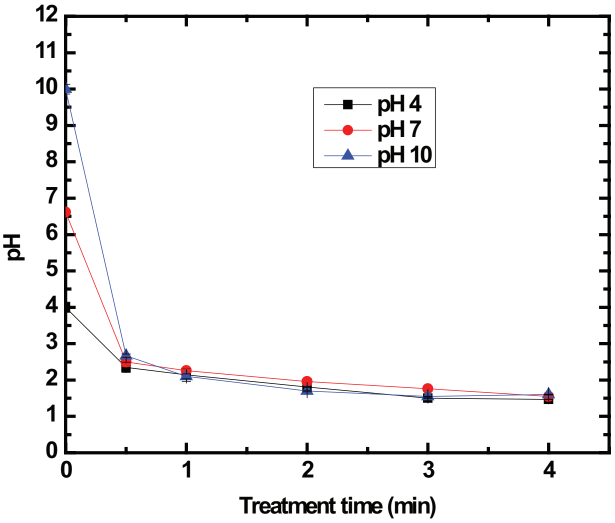

Fig. 6. Effect of ambient-air DBD treatment of an aqueous solution of acetaminophen (100 mg $L^{-1}$) at different pH values (4, 7 and 10) with 40 W DBD on pH.

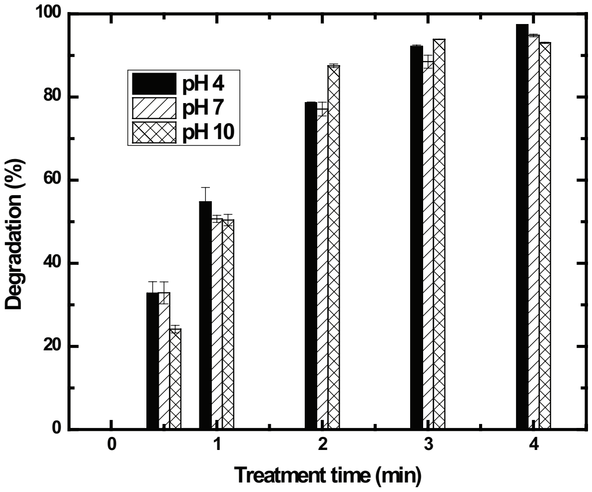

Fig. 7. Degradation rate of acetaminophen from water at various pH values (4, 7 and 10) with DBD operated in ambient air as a function of time.

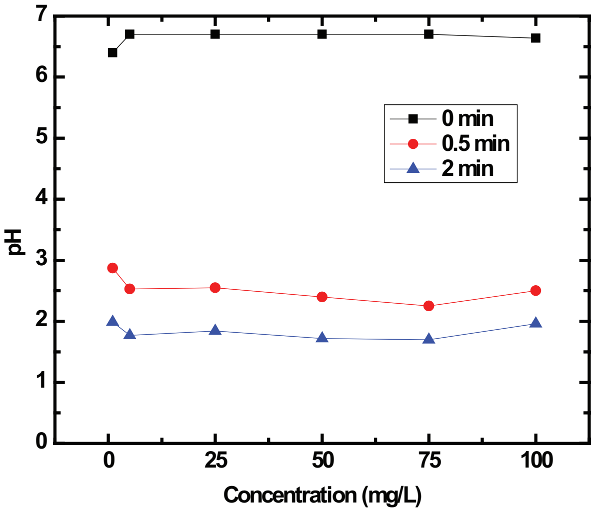

Fig. 8. Effect of plasma treatment of an acetaminophen aqueous solution on pH at different concentrations (0, 25, 50, 75 and 100 mg L$^{-1}$) treated with DBD.

dependent. In contrast, results reported by Panorel et al. [25] showed that acetaminophen degradation in a PCD improved at higher pH values. This could be due to the enhanced reactivity of dissociated phenolic acetaminophen toward oxidation. This incongurance among the findings may be attributable to the effect of different removal systems used in the aforementioned studies. Furthermore, the pH value of the acetaminophen aqueous solution during PCD treatment decreased from 6 to 3 after 30 min of treatment [35]. Su et al. [35] showed that the removal of acetaminophen from water in an aerated Fenton reactor was the highest at pH 3, noting a removal rate of 99% after 40 min of treatment. Overall, even though our results showed no effect of pH on the removal of acetaminophen from aqueous solution, the efficiency of DBD system used remained higher than that of the systems employed in previous studies [29,34,35].

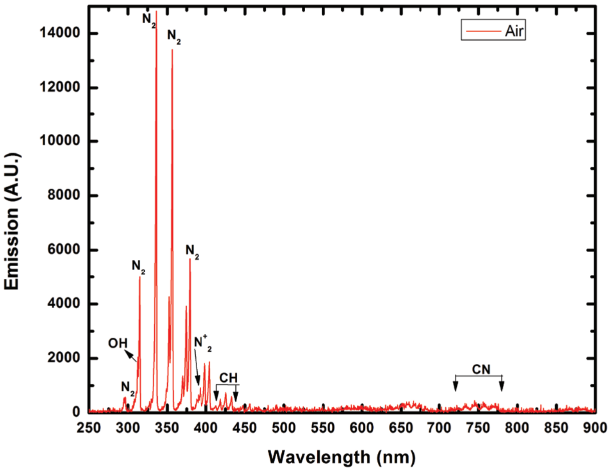

Fig. 9. Optical emission spectrum obtained during the DBD treatment of an aqueous solution of acetaminophen.

### 3.3. Reactive species

The optical emission spectrum obtained during the DBD treatment of 100 mg L$^{-1}$ acetaminophen solution is presented in Fig. 9. It shows the dominant reactive species produced in the discharge and can also provide information on the reaction products from the interaction pollutants. Fig. 9 shows significant emission from OH and $N_2$ radicals. These nitrogen and hydroxyl radicals are generated by the plasma and play a significant role in pollutant degradation. There is a weak emission from CH and CN bands, corresponding to the reaction products resulting from the interaction between the dominant radicals and the carbon-based pollutants in the water.

### 3.4. Energy efficiency

The ED of 9.6E05 J L$^{-1}$ was used in this study to reach the most significant removal rate (100%) of acetaminophen from water in open-air conditions. Even though Wang et al. [31] reported a better energy efficiency of a DBD system at 1.5E05 J L$^{-1}$ in the removal of iopromide from water, our system is more cost-effective because it is operated in the ambient air without the need for any expensive gases. However, the ED might be variable depending on the DBD system operating conditions.

## 4. Conclusions

In this study, the removal of acetaminophen from water using a DBD system was investigated. The system operated in ambient air and achieved high energy efficiency, and the acetaminophen removal rate from water ranged from 86% to 100% of the initial amount at different concentrations for a treatment time of 2 min. The removal efficiency and time were higher than those reported for other systems in the pertinent literature. The initial pH of the solution causes a variation in the degradation of the acetaminophen within 8%, which is not significant. Also, the variation does not follow a certain trend; therefore, it cannot be considered as an important parameter. The plasma treatment results in the decrease of the pH of the solution. Despite very different initial pH of the solution, the pH changed to 2.5 ± 0.5 within just 0.5 min of the plasma treatment. Further plasma treatment of up to 3.5 min just decreased the pH further to only 1.5. The final pH of different solution was always within 15%, regardless of the initial concentration.

The optical emission spectroscopy of the system revealed the prominent nitrogen and hydroxyl reactive species, which degrade the pollutant. This short treatment time and open-air operation make the system affordable and very useful for large-scale operation. This DBD system might also be useful for the removal of other organic pollutants from water.

## Acknowledgments

Authors would like to thank the funding agency, of this study which is the General Directorate for Research Grants (GDRG), at King Abdulaziz City for Science and Technology (KACST), Saudi Arabia; under grant number MS 35-15.

Authors would also like to extend special thanks to Dr. Mohammed Oteef and Dr. Mukul Sharma (Jazan University) for their assistance in sample analysis. Authors further express their gratitude to Professor Abdullah Basahi (CERS, Jazan University) and Dr. Galib Souadi (Physics Department, Jazan University) for their support and use of facilities.

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