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Study the Effect of Dielectric Barrier Discharge (DBD) Plasma on the Decomposition of Volatile Organic Compounds

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Abstract

Recently, research has focused on non-thermal plasma (NTP) technologies as a way to remove volatile organic compounds from the air stream, due to its distinctive qualities, which include a quick reaction at room temperature. In this work, the properties of the plasma generated by the dielectric barrier discharge (DBD) system and by a glass insulator were studied. Plasma was generated at different voltages (3, 4, 6, 7, 8 Kv) with a fixed distance between the electrodes of 5 mm, and a constant argon gas flow rate of 2.5 L/min. DBD plasma emission spectra were recorded for each voltage. The Boltzmann plot method was used to calculate the electron temperature in the plasma (T_{ρ}) , and the Stark expansion method was used to calculate the electron density (n_e) . The decomposition of organic compounds (cyclohexane) was also studied using DBD plasma. The results showed that the potential difference between the two electrodes has a clear effect on the plasma parameters, as the temperature of the electrons (T_e) and the density of electrons (n_e) increase with the increase in the potential difference between the two electrodes. The DBD plasma system proved to be a good way to decompose volatile organic compounds, as the results proved the emission of hydrogen gas as one of the dissociation products of cyclohexane.

1. Introduction

The Dielectric Barrier Discharge (DBD) is an electrical gas discharge that uses a high applied voltage in the gap between the electrodes with a dielectric between them [1]. The most popular dielectric barrier materials are glass, quartz, ceramics, epoxy, and enamels. Teflon plates, silicon rubber, plastic sheets, and other materials can also be used. DBD has been successfully used to improve the wettability, printability, and adhesion of polymer surfaces as well as to reduce pollution. It can enhance surface processing at atmospheric pressure while maintaining plasma chemistry and ambient pressure. The most important characteristic of DBD is that, compared to other techniques like low-pressure discharges, fast-pulsed high-pressure discharges, or electron beam injection, it is significantly simpler to maintain a non-thermal equilibrium plasma state. Another key advantage of atmospheric pressure DBD is its versatility with regard to geometrical configuration, operational parameters, and operating medium [2]. Non-thermal discharges are caused by the dielectric acting as a barrier, restricting current flow, and preventing spark generation. When used at atmospheric pressure, this arrangement can be used to treat delicate substrates like living textures without causing heat damage. When a DBD is used at high pressures, the discharge splits into numerous micro discharges that occur over the discharge zone or in the discharge gap at equally spaced intervals [3]. Surface discharge (SD) is the discharge that takes place on the dielectric surface, whereas volume discharge

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DBD plasma, VOC, cyclohexane, Non-thermal plasma, Chlorine.

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Received: Aug. 14, 2022 Accepted: Oct. 12, 2022 Published: Dec.01,2022 (VD) fills the space between the electrodes. The intensity of the light on the dielectric surface is caused by the discharge that is produced [4]. There are many uses for DBD plasma; one of these is its use in the decomposition of compounds. Non-thermal plasma (NTP) is a standout option for the removal of Volatile Organic Compounds (VOCs) from polluted air [5].

VOCs generally arise from three origins: building materials and furniture, people activities and outdoor sources [6]. Small-scale urban businesses such as dry cleaning stores, printing shops, painting sites, and hair and beauty salons contain significant total Volatile Organic Compounds levels depending on the business activity performed [7-9].

The building of a DBD plasma system, Optical Emission Spectroscopy (OES) and its operation at atmospheric pressure are described in this paper. One of method for diagnosing plasma is OES. For years, plasma properties such as electron temperature, Debye length, Debye number, electron density, and plasma frequency have been determined using OES. The Boltzmann plot method was used to calculate the electron temperature of plasma [10-13]:

$$\ln\left[\frac{\lambda_{ji}I_{ji}}{hcA_{jig_j}}\right] = -\frac{1}{kT}\left(E_j\right) + \ln\left|\frac{N}{U(T)}\right|$$
(1)

where: I_{ji} is the relative emission line density between energy levels i and j. While g_j is the degeneracy or statistical weight of the upper level emitted from the transition phase λ_{ji} , is the wavelength (in nm), E_j is the excitation energy (in eV) for level i. A_{ji} is the possibility of automatic transmission of radiation from level i to the lower-level j, N refers to the densities of the population of the state, k is Boltzmann constant.

The following formula is used to compute Debye's length (λ_D) [13-15]:

$$\lambda_{D} = \left[\frac{\varepsilon_{0 \ k_{\rm B} \ T_{e}}}{n_{e \ e^{2}}}\right]^{\frac{1}{2}} \cong 7.43 \times 10^{2} \ \left(\frac{T_{e \ (e \ V)}}{n_{e}}\right)^{\frac{1}{2}}$$
(2)

where: n_e is the electron density, ε_o is the permittivity of free space, k_B is Boltzmann constant, n_e is the density of electrons, e is the electron charge, T_e is the plasma temperature. Plasma frequency can be given as [12]:

$$\omega_{pe} = \sqrt{\frac{n_e e^2}{m_e \varepsilon_0}} \tag{3}$$

where: ε_0 is the permittivity of free space, m_e is the mass of the electron and e is the charge of an electron.

Debye number (N_D) is given by the following formula [11]:

$$N_D = \frac{4}{3} \pi \lambda_D^3 n_e \tag{4}$$

The aim of this work is to study the effect of DBD plasma on the decomposition of volatile organic compounds.

2. Experimental Part

The plasma generation chamber contains two brass rigid cylinders, each 40 mm high and 25 mm in diameter, about 5 mm apart. Insulating material of glass of 1 mm thickness is placed between the two electrodes. The plasma generation chamber

(made of Teflon) is cylindrical of 50 mm diameter, with an inlet for argon gas and another for its exit. The plasma is ignited using an alternating electric current source of variable frequency that supplies a high voltage in the range of 0 to 20 kV. A cone-shaped glass flask containing cyclohexane, a chlorine glass flask, and an electric heater are also used in the system. Fig. 1 shows the experimental arrangement.



Figure 1: DBD System for Organic Compound Decomposition.

The working steps are as follows:

First step: The plasma was generated using a glass barrier (of 1 mm thickness) at variable voltages (3, 4, 6, 7, 8 kV), the distance between the electrodes was fixed at 5 mm and a constant flow rate of argon gas 2.5 L/min was maintained. The generated plasma was diagnosed using an optical spectrometer for each voltage value to study the effect of its change on the plasma parameters.

Second step: The conical flask was filled with 60 mL cyclohexane, and 10 mL chlorine was placed in the glass beaker. Cyclohexane was heated with an electric heater until cyclohexane vapor was formed; then, argon gas was passed at a flow rate of 2.5 L/min through a tube connected to the cyclohexane flask to obtain a mixture of argon and cyclohexane gas. This mixture was passed through to the DBD plasma system and exited through the tube connected to the chlorine liquid beaker. The pH of the chlorine liquid in the beaker was measured.

Third step: Plasma was generated at a constant voltage of 4 kV with a fixed distance between the electrodes of 5 mm, and with an argon gas flow rate of 2.5 L/min. The generated plasma was diagnosed using an optical spectrometer method adopted to detect the products of the volatile compound and obtain the spectrum of the plasma generated during the passage of cyclohexane vapor.

3. Results and Discussion

Fig. 2 exhibits the emissions spectra of the produced plasma using different values of voltages to study the effect of the applied voltages on the produced spectra.

From the figure the resulted spectra located at (696.54, 706.72, 714.7, 727.29, 738.4, 750.39, 763.51, 772.38, 794.82, 801.48, 811.53, 826.45, 842.46, 852.14, 866.79, 912.3, 922.45, 965.78, and 978.45). The figure indicates that the intensity of the peaks increases with the raising of the applied voltage. With the increase of the potential difference, the acceleration of the electrons increases, and thus collisions increases, which leads to an increase in the intensity of the peaks due to the increase in ionization and irritation in the atoms of the medium. It was noted that the maximum intensity was with the 8 kV, and the minimum at 3 kV.



Figure 2: The optical emission spectra of DBD plasma generated using glass barrier at different voltages of 3,4,6,7 and 8 kV.

Fig. 3 shows the Lorentzian fitting with different applied voltages in the range of (3-8 kV). The fittings located the maximum intensity at 763.4 nm for the different voltages. The intensity improved with increasing the applied voltage; maximum intensity of 0.045 was recorded at 8 kV, and the lowest one at 3 kV.

Fig. 4 shows the relation of $\ln \left[\frac{\lambda_{ji}I_{ji}}{hcA_{jig_j}} \right]$ versus the highest energy level (Ej) using a dielectric barrier discharge barrier the values of start with 3kV to 8kV. The figures include the statistical coefficient (R^2) also the fitting equation for each voltage. The results of the figure indicate the variation in the (R^2) values to be in the range (0.9658 – 0.947 eV).

When the potential difference between the two electrodes increases, the temperature of the electron increases, as shown in Fig. 5. This increase is because the electrons are exposed to a high potential difference, accelerated more and hence their kinetic energy and temperature increases. Although the increase of kinetic energy of the electrons leads to an increase in the collisions and thus an increase in the electron density, the energy gained by the electrons as a result of acceleration is much greater than the energy lost due to the collisions. This shows that an increase in the voltage between the two electrodes leads to an increase in the temperature of the electrons this result is agreement with [1]. Table 1 shows the values of electron temperature (T_e), electron density (n_e), Debye length (λ_D) and plasma frequency (f_p) that were calculated for the plasma produced at different voltages.



Figure 3: The Lorentzian fitting representing intensity versus wavelength for different voltages with the glass barrier.



Figure 4: Boltzmann plots for produced plasma dielectric barrier discharge.



Figure 5: The electron density and electron temperature for the produced plasma using different voltages in the range (3-8 kV).

Voltages(kV)	T _e (eV)	FWHM (nm)	$n_e \times 10^{17} (cm^{-3})$	f _p (Hz) ×10 ¹²	$\lambda_D \times 10^{-6} (cm)$
3.0	0.995	0.950	6.419	7.195	0.925
4.0	1.287	1.000	6.757	7.382	1.026
6.0	1.400	1.100	7.432	7.742	1.020
7.0	1.457	1.200	8.108	8.086	0.996
8.0	1.593	1.200	8.108	8.086	1.042

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Fig. 6 shows the change in pH as a function of time. The acidity of the beaker contents increases with time due to the emission of hydrogen gas, one of the products of the dissolution of the cyclohexane compound, as it combines with chlorine to form hydrochloric acid.

Fig.7 shows the emission spectrum of DBD plasma in the presence of cyclohexane vapor. The spectrum has a clear peak of carbon at a wavelength of 939.852 nm and two peaks associated with CO_2 at 441.3 nm and 493.2 nm wavelengths. This results agree with [9].



Figure 6: The change in pH value as a function of time.



Figure 7: The emission spectrum of DBD plasma in the presence of cyclohexane vapor.

4. Conclusions

The potential difference between the two electrodes of the DBD system has a clear effect on the plasma parameters. The results showed that the electrons temperature increased with the increase of the potential difference because when the electrons are exposed to a high voltage difference, they accelerate more and their kinetic energy increases and thus the temperature of the electron increase; while the electron density increases with the increase in the voltage due to the increase in the kinetic energy of the electrons causing an increase in collisions. Thus, the electron density increases. DBD plasma system is a good method to break down harmful volatile organic compounds (VOCs). We recommended studying the effect of changing the type of gas used on the efficiency of the system in decomposition volatile organic compounds.

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Conflict of interest

Authors declared that they have no conflict of interest.

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دراسة تأثير بلازما تفريغ الحاجز العازل (DBD) على تحلل المركبات العضوية المتطايرة

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الخلاصة

في السنوات الأخيرة ركزت الأبحاث على تقنيات البلازما الغير حرارية (NTP) كطريقة لإزالة المركبات العضوية المتطايرة من مجرى الهواء. لما لمها من صفات مميزة والتي تشمل التفاعل السريع في درجة حرارة الغرفة. في هذا العمل تمت دراسة خصائص البلازما النتيجة عن نظام تفريغ الحاجز العازل (DBD) وباستخدام عازل زجاجي. تم توليد البلازما بجهد متغير (3,4,6,7,8) كيلو فولت وبمسافة ثابتة بين الأقطاب الكهربائية 5ملم وبمعدل تدفق ثابت لغاز الاركون (2.5) لتر/ دقيقة. تم تسجيل اطياف السريع في درجة حرارة الغرفة. في هذا العمل تمت دراسة خصائص البلازما النتيجة عن نظام تفريغ الحاجز العازل (DBD) وباستخدام عازل زجاجي. تم توليد البلازما بجهد متغير (3,4,6,7,8) كيلو فولت وبمسافة ثابتة بين الأقطاب الكهربائية 5ملم وبمعدل تدفق ثابت لغاز الاركون (2.5) لتر/ دقيقة. تم تسجيل اطياف انبعاث البلازما DBD بجهد متغير. ((n) في البلازما واستخدم طريقة البلازما والمتخدم طريقة (الهكسان الحقي) باستخدام بلازما DBD. المركبات العضوية (الهكسان الحقي) باستخدام بلازما واستخدم طريقة البلازما DBD بجهد متغير. ((n) في البلازما DBD بحهد معنو. ((n) على الميرة الهرت ((n) عنور ((n) عنور (n)) والمكنرون ((n)) والمتخدم طريقة البلازما والمركبات العضوية (الهكسان الحقي) باستخدام بلازما والمندم طريقة المالات العضوية (الهكسان الحقي) باستخدام بلازما والمرت على معاملات البلازما، حيث تزداد درجة حرارة الالكترونات ((n) والكثرونات ((n)) وكافة الالكترونات (n) مع زيادة فرق الجهد بين القطبين، وأثبت نظام البلازما، حيث تزداد درجة حرارة الالكترونات ((n)) وكافة الالكترونات ((n)) مع زيادة فرق الجهد بين القطبين، وأثبت نظام البلازما، حيث تزداد درجة حرارة المركبات العضوية الالكترونات ((n)) مع زيادة فرق الجهد بين القطبين، وأثبت نظام البلازما، حيث تزداد درجة حرارة المركبات العضوية الملايرة) ويا الم المركبات العضوية المركبات العضوي المرية المركبات العضوية المركبات العضوية المكترونات ((n)) مع زيادة فرق الجهد بين القطبين، وأثبت نظام البلازما DBD انه طريقة جيدة تبدال معنوية المعنوين ألم مرك مع على معاملي الحلقي ما المرية مع معاملة البلازما، حيث تزداد درجة حرارة الالكترونات ((n)) مع زيادة فرق الحموية معنوية المركبات العضوية العضوي العضوي المرية) مع ملولة مم معلي مرك ما مع مع مع مي مام ما مروية مع معام مما مرك مما