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OZONE GENERATION USING A DIELECTRIC BARRIER DISCHARGE REACTOR FOR INCREASING FOOD SHELF LIFE

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Abstract: *Nowadays great attention has been paid to the DBD discharge on atmospheric pressure because of its wide application perspective in many industrial fields. This discharge is a little specific kind because one electrode is covered by a dielectric material, thereby preventing the discharge to move towards electrical breakdown. One of its most important applications is the production of ozone (o3) for air treatment specially dedicated to the food industry to extend storage life of food products. The aim of this paper is to study and identify the set point of electrical parameters of DBD discharge for ozone generation using the Response Surface Modeling (RSM) method; the optimal values obtained are then applied in a system of conservation of food products. The experiments were carried out on a laboratory experimental bench. Obtained results pointed out that the investigated parameters of DBD have significant effect not only on ozone production, but also for prolonging shelf life of food products.*

Key words: *Dielectric barrier discharge, Ozone, Food shelflife, Optimization.*

1. Introduction

The industrial and transport sectors are responsible for a large part of polluting emissions into the atmosphere, both locally and globally. In 1992, many countries adhered to the Kyoto Protocol on climate change to reduce all of these emissions. These adherences have been translated at European level by the implementation of new directives imposing authorized emission limits for many pollutants and the streghtening of already existing standards [4], [8-10], [14]. These standards require improvement of treatment techniques. The conventional methods such as the selective catalytic reduction method and the lime-gypsum method still remain difficult to treat exhaust gases energy efficiently and inexpensively. Their energy efficiency and their initial and running costs are still a downside for the developing nations [7], [16, 17].

Recently, new methods appear as

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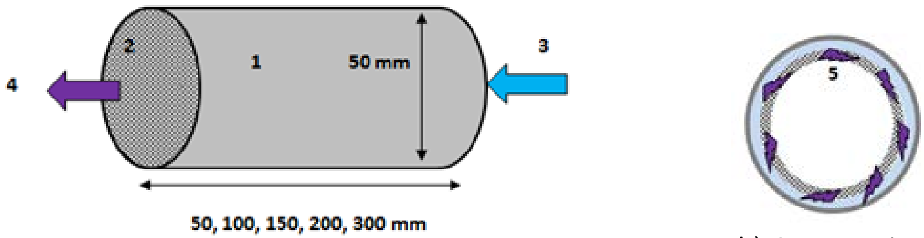
alternative solutions such as plasma discharge [1], [3], [18,]. The device is based on the dielectric barrier discharge. It is capable of producing conditions that enable various chemical reactions to occur [6], [11].

The main component produced by the dielectric barrier discharge is ozone (O_3) which is a powerful oxidizer and has a much higher disinfection potential than chlorine and other disinfectants. Ozone finds its application mainly in water treatment and air purification. The dielectric barrier discharge (DBD) method has proved to be the best method to produce ozone [12], [19]. Dried air or oxygen is forced to pass through a 1-2 mm gap.

Indeed, the production of ozone by a dielectric barrier discharge (DBD) depends on several factors. In such applications, the list of factors influencing the process includes the level of the applied voltage, the signal frequency and the duration of the dielectric barrier discharge (DBD) application [15]. Thus, it's not simple to determine with precision the optimal values of the process factors. An experimental procedure relying on the response surface modelling method for optimizing the ozone generation process was employed using a home-made experimental set-up, comprising a DBD reactor device. Three "one-factor-at-a-time" experiments, corresponding to three controllable factors, followed by a factorial design were performed based on a two steps strategy: fixing the variation domain of the input variables and searching the optimum set point. The obtained values of such factors were used to show that the ozone generated by a dielectric barrier discharge can be considered an advantageous solution for air disinfection and storage of food in the food industry.

2. Materials and Methods

Surface DBD reactors of cylindrical shape were achieved. The dielectric barrier consists of a glass tube having a diameter of 50 mm, different lengths of 50, 100, 200, 300 mm and a thickness of 2 mm. The electrodes are a fine strip of adhesive aluminum such as an internal electrode bonded directly on the outer surface of the glass tube with a 2 mm thickness. The external metal electrode, a stainless steel mesh was placed inside the glass tube in contact with it (Figure 1).



a) *Longitudinal section* b) *Cross section*

Fig.1. *Schematic representation of an ozone generator with surface DBD*
1) Internal electrode; 2) external electrode; 3) oxygen input; 4) ozone output; 5) discharge area

When a high voltage is applied to these electrodes, a bluish color plasma is formed on the surface of the tube which is in contact with the mesh electrode and is distributed homogeneously along the tube (Figure 2).

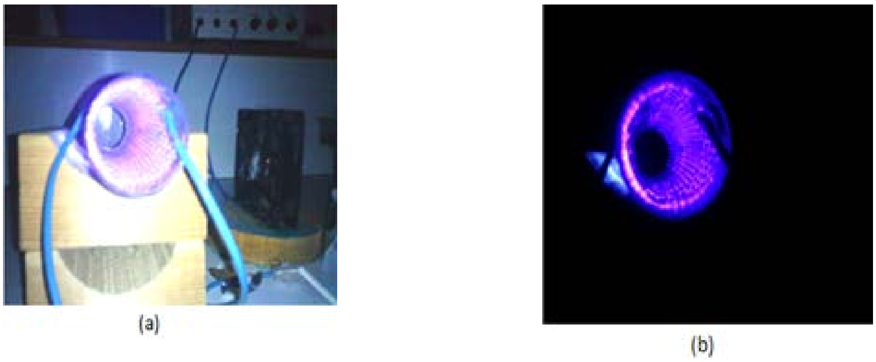


Fig. 2. *Discharge inside the DBD reactor*
a) *Day photo* b) *Night photo*

In operation, the plasma or the discharge extends over the interior surface of the cylinder between the grid and the glass, as shown in Figure 2.The power supply used comprises two main elements, a power amplifier (TREK 30 kV/20A) and an electrical signal generator (Toellenertoe 7301) which delivers alternating signals with an adjustable frequency range from 0 to 5.5 kHz, to power the ozone generator.

The ozone generator or DBD reactor was supplied by an oxygen concentrator (NIDEK medical Nuvo Lite Mark), with a constant flow of 1 l / min.

The ozone concentration produced by the reactor is measured using the ozone monitor (Ozone-solution-106H). The voltage V delivered by the power amplifier applied to the ozone generator is measured using a digital PICOSCOPE, while the current I was measured using a hall effect current sensor (Pearson electronic). The signals (current and voltage) are stored and transferred to a computer (Figure 3).

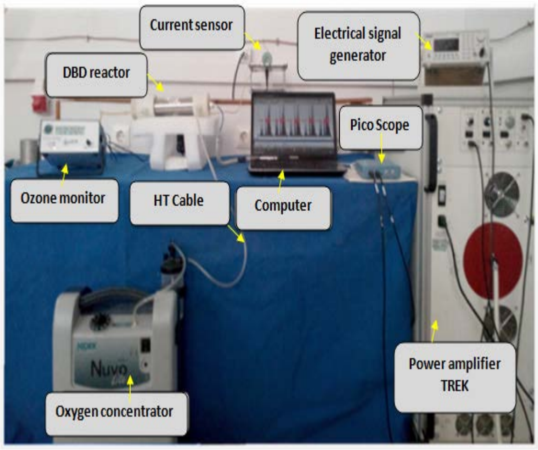


Fig. 3. *The experimental set-up*

3. Results and Discussion

For all the experiments carried out in this section one factor varied while the values of the other two factors remained constant.

Thus, Figure 4 represents the variation of the DBD reactor efficiency, in terms of ozone generation (mg/l), according to the applied voltage (V). The latter varied from 1 to 12 kV at constant values of f = 300 Hz, L = 150 mm and T = 20 seconds.

The obtained results show that the surface DBD starts quickly with low tension and the ozone generation rate increases with the applied voltage. Moreover, at a given applied voltage (V= 10 kV), the ozone generation rate reaches a higher value and then decreases (Figure 4).

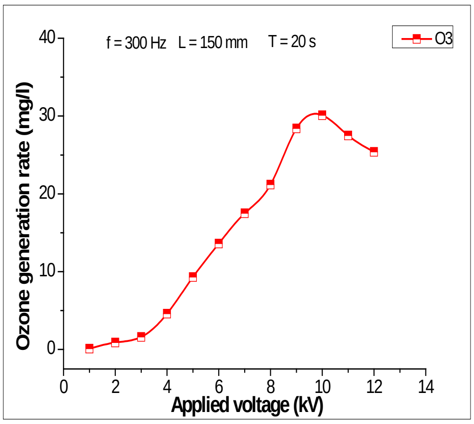


Fig. 4. *Ozone generation rate according the applied voltage*

The high voltage applied breaks the oxygen molecule which passes through the DBD discharge zone into two oxygen atoms. These two atoms are subsequently attached to two new oxygen molecules to form an ozone molecule. This ozone formation process increases with the increase in the applied voltage resulting in the increase of the generated ozone rate. The increase in voltage also generates breakdowns in the DBD reactor which cancels the voltage thus causing the ozone formation process to stop.

In the same way, in Figure 5, the variation of ozone generation by the DBD reactor (mg/l) as function of power supply frequency (f) is represented. The power supply frequency varied from 50 Hz to 1100 Hz for the sinusoidal signal using the function generator for an applied voltage of 5 kV, DBD length of 150 mm and a total duration of the DBD application of 20 seconds.

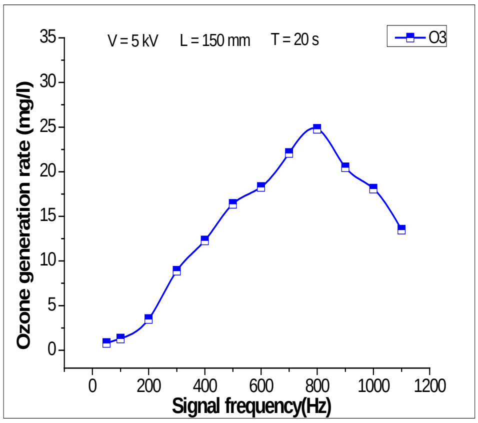


Fig. 5. *Ozone generation rate according to power frequency signal*

The increase in power supply frequency causes a rapid application of the voltage, which translates into a high speed of the ozone formation process. This effect stops from a certain limit value of the frequency, equal to 800 Hz in our case. Beyond this value, the signal is no longer effective (pulse regime) and the rate of ozone generation decreases (Figure 5).

Figure 6 shows the ozone generation rate according the length of the DBD reactor (L). In this case, the DBD length varied from 50 mm to 300 mm at constant values of an applied voltage of 5 kV, a power supply frequency of 500 Hz and a total duration of DBD application of 20 seconds.

When the DBD length is small the generation of ozone is low, the oxygen that passes through the discharge zone does not have enough time to completely transform into ozone. In addition, the length increases, the generation of ozone increases and beyond a certain value of the length of the DBD, the rate of generated ozone decreases because the generated ozone is still in the space of the discharge which then is then facilitated by the effect of the high voltage of its re-transformation into oxygen (Figure 6).

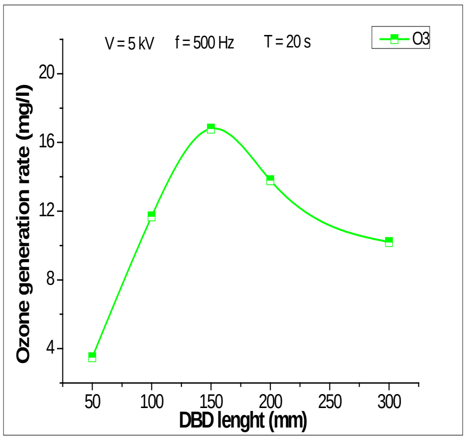


Fig. 6. *Ozone generation rate according to reactor DBD length*

Figure 7 shows the ozone generation rate according the total duration of DBD reactor application (T). Indeed, the total duration of DBD application varied from 10 to 120 seconds at constant values of an applied voltage of 5 kV, a power supply frequency of 300 Hz and a reactor DBD length of 150 mm.

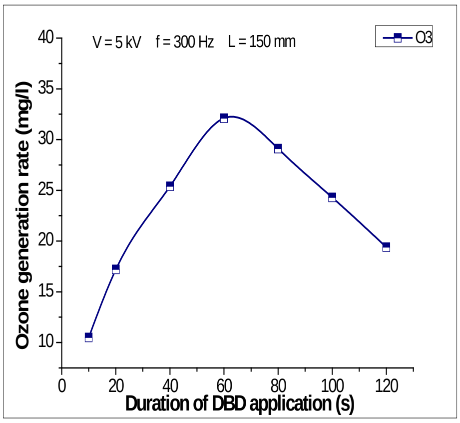


Fig. 7. O*zone generation rate according to the total duration of DBD reactor application*

In the same manner, the ozone generation rate increases with the increase of the total duration of DBD application up to a limit value and then decreases. The decrease in ozone level is explained by the heating of the DBD reactor. This rise in temperature breaks the ozone molecules. This process requires a cooling system.

3.1. Experimental Designs Methodology

The methodology of the experimental designs makes it possible to determine the number of experiments to be achieved according to a well defined objective, to study several factors simultaneously, to reduce dispersion related to measurements, to appreciate the effects of coupling between factors and finally to evaluate the respective influence of the factors and their interactions.

The Composite Centred Faces design (CCF), which gives quadratic models, was adopted. A quadratic dependence is established between the output function to optimize (response) and the input variables [2], [5].

MODDE 5.0 software (Umetrics AB, Umea, Sweden) was used, which is a Windows program for the creation and the evaluation of experimental designs [13].

The three following factors are studied:

1. Applied Voltage V (kV);
2. Power signal frequency f (Hz);
3. Reactor DBD length L (mm);
4. Total duration of DBD reactor application T (s).

Indeed, the results that had been obtained in the previous section served for the definition of the domain of variation of V, f, L and T. Thus, V_{min} = 9 kV and V_{max} = 11 kV were retained as the limit values for applied voltage.

Similarly, for power signal frequency, the domain of variation was chosen as f_{min} = 700 Hz and f_{max} = 900 Hz. Indeed, we opted for the reactor DBD lenght as L_{min} = 100 mm and L_{max} = 200 mm as limits of variation domain of L.

For the total duration of DBD application the domain of study was defined as T_{min} = 40 seconds and T_{max} = 80 seconds.

The results of all the experiments are given in Table 1 and Figures 8 - 9 for the domain of variation of V, f, L and T previously defined to indentify a mathematical model using the MODDE 5.0 software.

According to all of the experiments, the modeling software MODDE 5.0 gave us a mathematical model of ozone generation rate (mg/l) using a surface dielectric barrier discharge. This mathematical model is very satisfactory because the coefficients R2 and Q2 are very close to 1 (R^2 = 0982, Q^2 = 0.914). MODDE 5.0 also gives the effect of each parameter on the ozone generation rate (Figure 8).

Table 1

Results of ozone generation experience according to the variation of the factors' values

Exp. no.	V (kV)	f (Hz)	L (mm)	T (s)	O3 (mg/l)
1	9	700	100	40	31
2	11	700	100	40	24,3
3	9	900	100	40	19,6
4	11	900	100	40	15
5	9	700	200	40	25,8
6	11	700	200	40	8,7
7	9	900	200	40	10,9
8	11	900	200	40	1,5
9	9	700	100	80	13,3
10	11	700	100	80	9,1
11	9	900	100	80	10,1
12	11	900	100	80	12,6
13	9	700	200	80	13,6
14	11	700	200	80	1,2
15	9	900	200	80	11,3
16	11	900	200	80	4,7
17	9	800	150	60	25,1
18	11	800	150	60	18,4
19	10	700	150	60	22,5
20	10	900	150	60	20,4
21	10	800	100	60	18,2
22	10	800	200	60	15,1
23	10	800	150	40	20,2
24	10	800	150	80	16,3
25	10	800	150	60	22,1
26	10	800	150	60	22,1
27	10	800	150	60	22,1

According to the validated model, the applied voltage (V), DBD length (L) and total duration of DBD application (T) are the most important and influential factors in the rate of generated ozone. However, the ozone generation rate using a surface DBD should be higher by using an appropriate value of each parameter.

Moreover, except for the interaction between the DBD length and the total duration of DBD application, other interactions are less significant.

In addition, the greatest value of ozone generation rate (O_3) should be obtained for the applied voltage of V_0 = 9 kV, power supply frequency f_0 = 700 Hz, DBD length L_0 = 134, 5 mm and a total duration of DBD application T_0 = 40 seconds (Figure 9).

The mathematical model of ozone generation rate obtained:

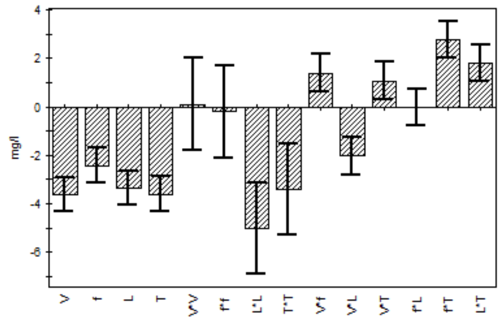


Fig. 8. *Values of the influence of each factor and their interactions*

$$O3 = 21.80 - 3.62V^{*} - 2.41f^{*} - 3.35L^{*} - 3.6T^{*} - 5L^{*2} - 3.4T^{2*} + 1.39V^{*}f^{*} - 2.03V^{*}L^{*} + 1.06V^{*}T^{*} + 2.76f^{*}T^{*} + 1.79L^{*}T^{*} \quad (1)$$

	Factor	Role	Value	Low Limit	High Limit
1	Applied Voltage	Free		9	11
2	PS-Frequency	Free		700	900
3	DBD Lenght	Free		100	200
4	DBD Duration	Free		40	80

Iteration: 5001 Iteration slider:

	1	2	3	4	5	6	7
	Applied Voltage	PS-Frequency	DBD Lenght	DBD Duration	Ozone genration Rate	iter	log(D)
1	9	700,001	134,461	40,0005	33,6229	5000	1,9729
2	9	700	134,494	40,0018	33,6228	5000	1,9729
3	9	700	134,763	40,0001	33,6229	5001	1,9729
4	9	700,001	134,576	40,0017	33,6229	5001	1,9729
5	9	700	134,547	40,0002	33,6229	5001	1,9729
6	9	700,003	134,9	40,0002	33,6225	5001	1,9729
7	9	700,007	134,603	40,0005	33,6226	5000	1,9729
8	9	700,001	134,26	40,0016	33,6227	5000	1,9729

Fig. 9. *Subroutine of MODDE.05 representing the set point*

3.2. Application of the Reactor for Food Storage

The DBD reactor powered by the optimal values defined in the previous section was then used as an ozone generator for the disinfection of air in order to extend the shelf life of food products. The laboratory setup used is described in Figure 10. The apples to be stocked are put inside a glass enclosure; the oxygen concentrator allows the injection of ozone inside the enclosure every day.

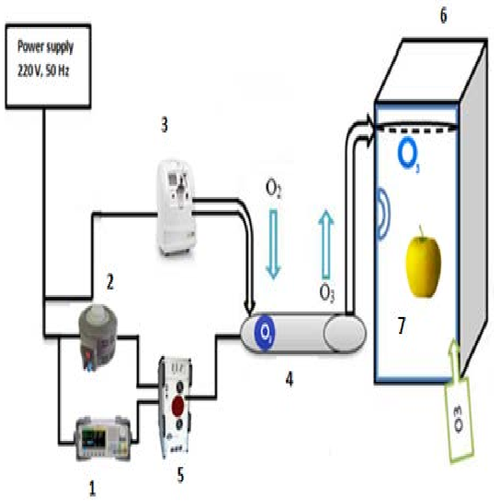


Fig. 10. *Schematic description of the disinfection process:*
1. Function generating; 2. Auto-transformer;3. Oxygen concentrator; 4. Ozone generator; 5. TREK; 6. Glass enclosure; 7. Food to be processed

A similar enclosure closed and placed in open air without treatment contains the same food products (apples). The results are expressed in terms of weight loss and visual aspect for the two enclosures (Figures 11 and 12). Weight loss in % was calculated by:

$$WL(\%) = \frac{W_i - W_f}{W_i}. \qquad (2)$$

where Wi is the initial weight of apple (g) and Wf is the final weight of apple (g).

In the photos, we notice that the products located in the open-air enclosure had lost their qualities and their weights after a few days as compared to the products placed in the ozone-disinfected enclosure. This is further proof of the effectiveness of ozone in preserving and slowing down the ripening process of agrifood products.

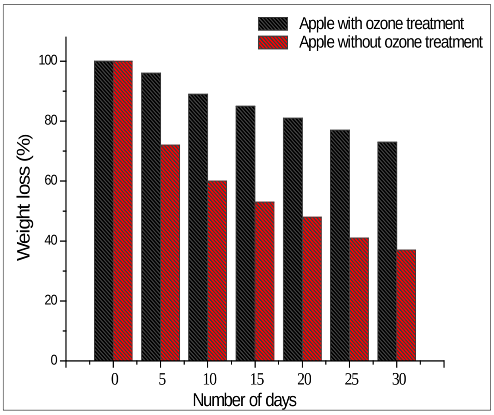


Fig. 11. *Evolution of apple weight according to shelf life*

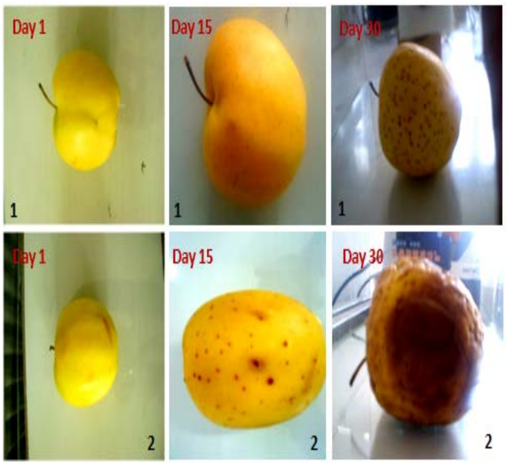


Fig. 12. *Evolution according to the visual aspect of apples:*
1) Treated sample; 2) Control sample

4. Conclusion

Ozone is the most powerful oxidant and disinfectant used for air purification. It does not produce any undesirable derivatives and turns back into oxygen. However, it has a short life.

The best way to generate ozone is to use a dielectric barrier discharge, particularly the surface discharge. This kind of discharge avoids the occurence of electric arcs and can be operate with low voltage which gives high yields. The dielectric barrier discharge is a multifactorial process; optimization is therefore an important step which has allowed us to provide appropriate values of the parameters of the DBD to increase the performance of this system.

Finally, the application of ozone in the food industry seems to be very beneficial. In the future, a combination of ozone with cold storage will be an alternative solution for several sectors.

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