SET-POINT IDENTIFICATION OF AN OZONE WATER TREATMENT PROCESS

S. NEMMICH, A. TILMATINE and N. HAMMADI

APELEC Laboratory, Djillali Liabes University of Sidi Bel-Abbes, ALGERIA. Email: atilmatine@gmail.com

***Abstract*— The efficiency of an ozone water treatment process depends on a multitude of factors, including both the command factors and the environmental conditions. The aim of the present work is to analyze the application of the response surface experimental design techniques that make use of quadratic functions for modeling the ozone water treatment process. Due to the importance of the robustness of such treatment process, the aim of this paper is to validate an experimental procedure for optimizing the treatment efficiency and for determining the set point. The experiments were carried out on a laboratory experimental bench using Venturi injection system, where three controllable factors were analyzed: the high voltage level, the oxygen flow rate and the water flow rate. Three "one-factor-at-a-time" experiments followed by a center-faces composite design were performed. Once the variation limits of the input variables are determined, a mathematical model was proposed which served to analyze the influence of the factors and their interactions. At the end, an optimal set point was identified.**

***Keywords*— Experimental modeling, ozone generation, experimental design, water treatment, set-point.**

I. INTRODUCTION

Ozone is a powerful disinfecting and oxidizing agent, which is used in a wide range of applications, such as municipal and wastewater treatment, food processing, sterilization, bio-decontamination, ozone dentistry and spinal applications. Most of the ozone for practical applications is produced by electrical discharges at atmospheric pressure. The main electrical discharge used for this purpose is a corona discharge (Pekarek, 2012; Chen and Davidson, 2003; Morent and Leys, 2005) and a dielectric barrier discharge (Masuda *et al.*, 1988; Nomoto *et al.*, 1995; Pietsch and Gibalov, 1998; Jodzis *et al.*, 2011).

Dielectric barrier discharge (DBD) is a typical high pressure gas discharge and can produce non-thermal plasma at atmospheric pressure (Tao *et al.*, 2009; Fridman *et al.*, 2009). Generally, DBD is generated by AC source of periodic sine or pulse waves, and its study and applications have been much reported over the past twenty years.

The DBD is usually generated between two metallic electrodes, of which one electrode is covered with dielectric. DBD discharges, which are sometimes called silent discharges, usually operate at frequencies between 0.05 and 500 kHz (Boonduang *et al.*, 2012). However, some ozone generator configurations generate inhomogeneous heat-up of the dielectric barrier and result in the decrease of ozone production efficiency. Experimental studies concerning ozone generator configuration such as electrode arrangements, gap lengths, dielectric materials, operating gases and electrical discharges have been widely studied to obtain the optimum discharge conditions for high yield ozone production (Fang *et al.*, 2008; Park *et al.*, 2006; Moon and Jung, 2007; Sung and Sakoda, 2005).

The difficulty of the problem is that ozone water treatment process is a multifactorial process. In such application, for instance, the list of factors influencing the outcome of the process includes the discharge gap, the length of the generator, the high voltage level, the voltage frequency, the water flow rate, the gas flow rate and so on. The problem is to find the set of values of the above-listed factors for which the outcome of the process is optimal? In this work, a cylinder-cylinder DBD ozone generator has been developed and investigated by focusing on dissolved ozone concentration.

The aim of our research was to validate an experimental procedure for optimizing the treatment process using a home-made experimental set-up, which was successfully used for modeling and optimization of several electrostatic processes (Dascalescu *et al.*, 2005; Tilmatine *et al.*, 2009; Medles *et al.*, 2007a, b). Three "one-factor-at-a-time" experiments, corresponding to three controllable factors, then a factorial composite design, were performed based on the following two-step strategy:

1) Identifying the domain of variation of the controlled variables;
2) Finding the best choice of the set point.

II. MATERIAL AND METHODS

Figure 1 show the dielectric barrier discharge reactor used in this study. The inner cylindrical electrode is an adhesive aluminum sheath inserted in a glass tube with a closed contact of their surfaces. The glass tube with a thickness of 2.5 mm, acting as a dielectric barrier, is 300 mm long and 44 mm outer diameter. The grounded cylindrical electrode is a stainless steel tube with an inner diameter of 39.0 mm and 250 mm length (Fig .2).

The laboratory experimental setup employed is described in Fig. 3 and shown in Fig. 4. The contaminated water to be treated (5) was set in motion using a water pump (6). A Venturi system (4) enables injection of ozone within the water loop and mixed water with ozone (7) is considered as treated water. The ozone is generated with the home-made ozone generator which is

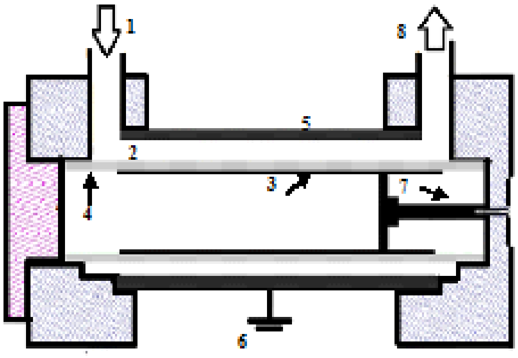

Fig. 1. Longitudinal cross section of the developed ozone generator (discharge gap: e = 1 mm). (1) Oxygen inlet. (2) Gas gap for discharge. (3) Inner cylinder electrode (aluminum adhesive). (4) Dielectric barrier (glass). (5) Outer cylindrical electrode (6) Terminal to ground. (7) High-voltage terminal. (8) Ozone outlet.

Fig. 2. Photograph of ozone generator

fed by an oxygen concentrator (NIDEK medical Nuvo lite Mark 5), of variable flow rate (1 to 5 L/min).

The high voltage power supply used in this study is of a switching type. A 220 V AC voltage was stepped down by a transformer and rectified into DC voltage of about 72 V. IGBT (Insulated Gate Bipolar Transistor) was used as electronic switch and controlled by an oscillator to generate a high frequency current. Low voltage with high frequency current was stepped up by a high voltage step up transformer, then the voltage of 1-10 kV at frequency of 0-30 kHz were obtained. In present work, the optimum frequency for discharge tube is 20 kHz.

All the experiments were carried out in stable climatic conditions of temperature (20-25 °C) and humidity (60-70 %).

The applied voltage U was measured with a digital scope (GWINSTEK GDS-3154) using a high voltage probe (Tektronix p6015A), while the current I was estimated by measuring the drop voltage across an ohmic resistance of 100 Ω. The consumed power of the ozone generator was estimated by calculating the average value of the product U*I over 2500 points. A list of values of voltage U and current I is provided by the scope and then the power is estimated by calculating the average value of the product U*I.

A. Experimental designs methodology

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

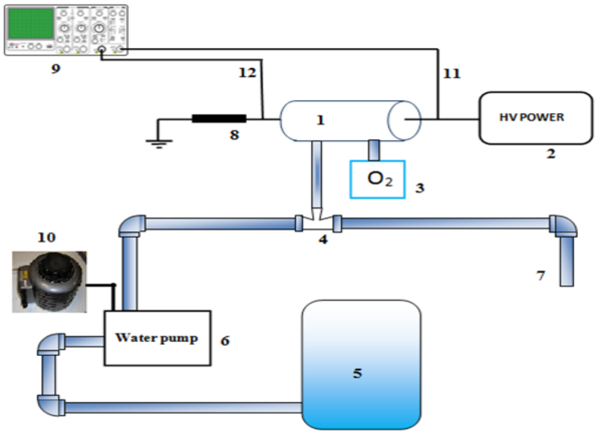

Fig. 3. Descriptive diagram of the ozone water treatment process. 1. Ozone Generator, 2. Resonant inverter high-voltage high-frequency power supply, 3. Oxygen concentrator, 4. Venturi injector, 5. Water reservoir, 6. Water pump, 7.Treated water output. 8.100 Ω resistor, 9. Digital oscilloscope, 10. Variac, 11-High voltage measurement probe. 11-Low voltage measurement probe

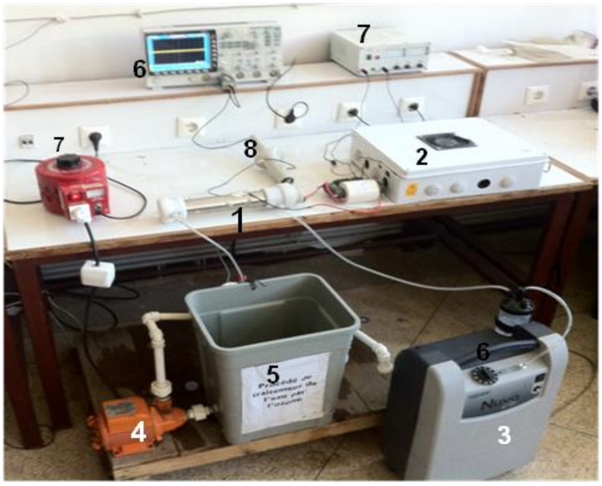

Fig. 4. The experimental set-up. 1 -Ozone Generator, 2 –high-voltage power supply, 3 - oxygen concentrator, 4-Water pump, 5 - Water to be treated, 6-Digital oscilloscope, 7-Variac, 8-high voltage probe

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 (Frigon and Mathews, 1996; Taguchi, 1987; Eriksson *et al.*, 1987).

Before starting the experiments, it is necessary to set the best and suitable design which can model the process with the most possible precision. In this paper, the Composite Centred Faces design (CCF), which gives quadratic models, was adopted. It is possible to determine a quadratic dependence between the output function to optimize (response) and the input variables u_i (i=1, …, k) (factors):

$$y = f(u_i) = c_0 + \sum c_i u_i + \sum c_{ij} u_i u_j + \sum c_{ii} u_i^2 \,. \qquad (1)$$

Knowing that Δu_i and u_{i0} are respectively the step of variation and the central value of factor i, reduced centred values of input factors may be defined by the following relation:

$$x_i = (u_i - u_{i0})/\Delta u_i \,. \qquad (2)$$

With these new variables, the output function becomes:

$$y = f(x_i) = a_0 + \sum a_i x_i + \sum a_{ij} x_i x_j + \sum a_{ii} x_i^2 \,. \qquad (3)$$

The coefficients can be calculated or estimated by a data-processing program, in such a way to have a minimum variance between the predictive mathematical model and the experimental results.

MODDE 5.0 software (Umetrics AB, Umea, Sweden) was used, which is a Windows program for the creation and the evaluation of experimental designs (MODDE 5.0, 1999). The program assists the user for interpretation of the results and prediction of the responses. It calculates the coefficients of the mathematical model and identifies best adjustments of the factors for optimizing the process. Moreover, the program calculates two significant statistical criteria which make it possible to validate or not the mathematical model, symbolized by R^2 and Q^2. The former is called the goodness of fit, and is a measure of how well the model can be made to fit the raw data; it varies between 0 and 1, where 1 indicates a perfect model and 0 no model at all. The latter is called goodness of prediction, and estimates the predictive power of the model. Like R^2, Q^2 has the upper bound 1, but its lower limit is minus infinity. For a model to pass the diagnostic test, both parameters should be high, and preferably not separated by more than 0.2–0 .3.

B. Design of ozone water treatment experiments

Design of experiments methodology is useful for screening, optimization and robustness testing. Screening experiments are designed in this paper to identify the domain of variation of the three factors, classical "one-factor-at-a-time" experiments. The optimization stage of an experimental procedure should enable the determination of factor values for which the ozone concentration is a maximum and the consumed power is a minimum.

In this paper the experiments were carried out by varying three control variables:

1. High-voltage level *U* (in kV);
2. Oxygen flow rate *OF* (in L/min);
3. Water flow rate *WF* (in L/min)

III. RESULTS AND DISCUSSION

A. Domain of variation of the control factors

The variation limits of the above control factors are defined by following "one-factor-at-a-time" experiments.

Experiment 1.1. Variable voltage *U*(4-8kV), at constant values of oxygen flow rate *OF*=5 L/min and water flow rate *WF*= 8 L/min.

Experiment 1.2. Variable Oxygen flow rate (1-5 L/min), at constant values of voltage *U*=7*kV* and water flow rate *WF* = 8 L/min.

Experiment 1.3. Variable Water flow rate *WF* (8-16 L/min) at constant values of voltage *U*=7kV and oxygen flow rate OF = 5 L/min.

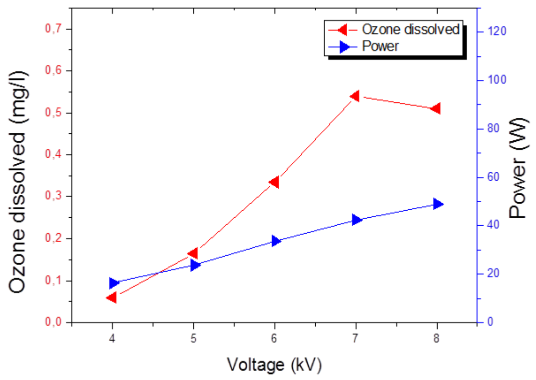

Fig. 5. Evolution of the dissolved ozone concentration and the consumed power according to the voltage (*WF*=8 L/min, *OF*=5 L/min).

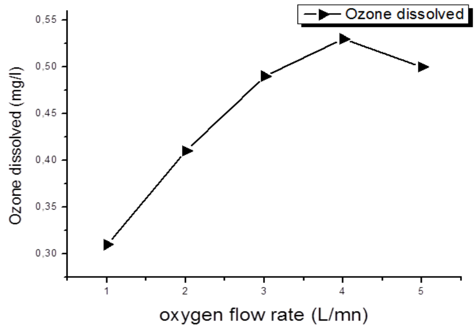

Fig. 6. Evolution of the dissolved ozone concentration according to the oxygen flow rate (*U*= 7 kV, *WF*=8 L/mn).

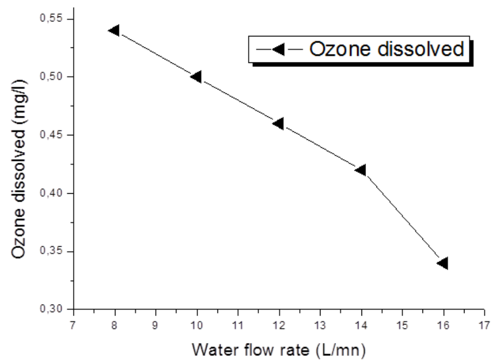

Fig. 7. Evolution of the dissolved ozone concentration according to the water flow rate (*V*=7kV, *OF*=5 L/min).

Experiments 1.1–1.3 are given, respectively, in Figs. 5-7. T he dissolved ozone concentration in water (mg/L) was considered as significant for the evaluation of the process and represented as functions of the three control factors. In addition, the variation of the consumed power as function of the voltage *U* is presented in Fig. 5.

The domain of variation of *U*, *OF* and *WF* were then defined based on results obtain in this section. Thus, the results of experiments 1.1 (Fig. 5) obtained according to the applied voltage show that U should not exceed 7 kV, otherwise the dissolved ozone concentration will decrease, while the consumed power is being increased.

When the applied voltage V is small the DBD cannot be observed since a minimum threshold voltage is required, which is equal to 4 kV in our case. However, if *V* is increased beyond a determined value (*V*= 7 kV for the case of our experimental conditions), no more ozone is generated and the extra power is transformed in overheating. Indeed, heat is considered as a by-product of ozone production because the discharge generates heat which suppresses ozone generation by converting some product ozone back to oxygen.

Therefore, the variation interval of *U* was defined between 6 and 8 kV.

The graph in Fig. 6 shows that in the conditions of Experiment 1.2, the ozone concentration increases up to 4 L/min and then decreases. At higher values of the oxygen flow rate, a more significant amount of oxygen passes throw the ozone generator without being transformed into ozone. Moreover, with increasing the flow rate the gas residence time in the discharge is decreased, thus the ozone generation rate is gradually suppressed because oxygen atoms are exhausted faster to the gas exit, without causing more production of ozone. For this factor, the variation interval was therefore chosen between OF_{min} = 3 L/min and OF_{max} = 5 L/min.

In the conditions of Experiment 1.3 (Fig. 7), as expected, the dissolved ozone concentration was continually decreasing according to the water flow rate. Consequently, the domain of variation of this factor was defined as WF_{min} = 8 L/min and WF_{max} = 16 L/min.

B. Set Point Identification

The identification of the set point U_0, OF_0 and WF_0 by using a central CCF design was performed; the two levels "max" and "min" are the limits established in previous section for each of the three control variables (U_{min}, U_{max}), (OF_{min}, OF_{max}) and (WF_{min}, WF_{max}), the central point (U_c, OF_c and WF_c) being calculated as follows:

$$U_c = \frac{(U_{\min} + U_{\max})}{2} = \frac{6+8}{2} = 7\text{kV} \cdot \quad (4)$$

$$OF_c = \frac{(OF_{\min} + OF_{\max})}{2} = \frac{3+5}{2} = 5\frac{\text{L}}{\text{min}} \cdot \quad (5)$$

$$WF_c = \frac{(WF_{\min} + WF_{\max})}{2} = \frac{8+16}{2} = 12\frac{\text{L}}{\text{min}} \cdot \quad (6)$$

The results of the experiment are given in Table 1.

The mathematical model of the response considered for optimization, which is the dissolved ozone concentration in water *OC*, was obtained with MODDE 5.0 as follows:

OC [mg/L] = 0,457 + 0,096 V* -0,014 OF - 0,038 WF* -0,02U*OF +0,02 U*WF + 0,0025 WF*OF - 0,067 U^{*2} - 0,059 WF^{*2} + 0,057 OF^{*2}

Since the statistical criteria R^2 and Q^2 were higher and close to the unit (R^2 = 99.2 %; Q^2 = 90.3%), the above model was validated and has been used for prediction and optimization analysis. The coefficients of the model are plotted in Fig. 8.

According to this model, we notice that the effect of the voltage is the more significant compared to the water flow rate effect. These comments are valid only when the factors vary in the previous defined intervals. The main factor for ozone generation is definitely the applied voltage which creates the electric discharge. Without the DBD, the oxygen molecules cannot be transformed in ozone and no water treatment is then possible. On the other hand, the water flow rate has a negative effect and should be decreased to a determined value according to the ozone output rate. With higher water flow rate, the concentration of the dissolved ozone becomes lower. On the other hand, the effect of the oxygen flow rate was found to be not significant. As explained above, there is no point to increase the oxygen flow rate beyond 4 L/min because much of it will exit from the ozone generator without being transformed in ozone. Furthermore, obtained results from the mathematical model didn't indicate any interaction between the three factors, since their effects are not significant.

According to this model, the optimum of the process (i.e., the highest amount of ozone concentration and the lowest value of the consumed power) should be obtained for *V*=6 kV, *OF* = 3 L/min and *WF* = 16 L/min. Figure 9 shows the iso-response contours obtained with the present model.

Table 1. Results of the experiment.

Test N°	Voltage U (kV)	Water flow WF (L/min)	Oxygen flow OF (L/min)	Ozone concentration OC (mg/L)
1	6	8	3	0.35
2	8	8	3	0.52
3	6	16	3	0.24
4	8	16	3	0.475
5	6	8	5	0.34
6	8	8	5	0.495
7	6	16	5	0.26
8	8	16	5	0.43
9	6	12	4	0.28
10	8	12	4	0.505
11	7	8	4	0.44
12	7	16	4	0.36
13	7	12	3	0.52
14	7	12	5	0.51
15	7	12	4	0.455
16	7	12	4	0.455
17	7	12	4	0.455

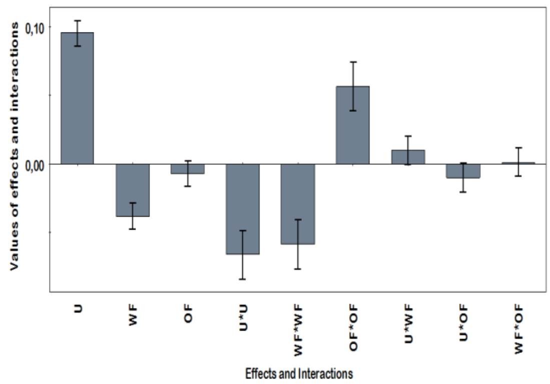

Fig. 8. Plotted coefficients of the obtained model

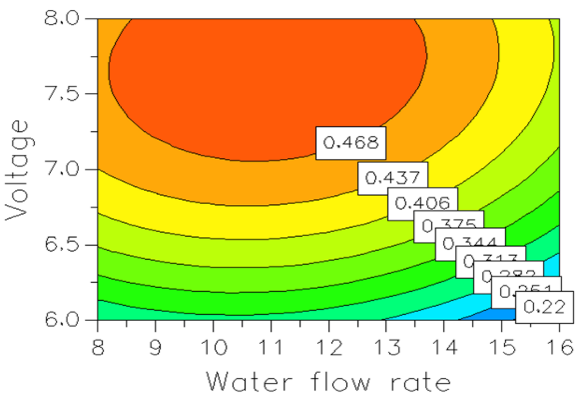

Fig. 9. Response contour plots for of ozone concentration

Contour plots of the response function (ozone concentration) was computed with MODDE, in order to make a comparison between the effects of the voltage V and the water flow rate WF for a constant oxygen flow rate *OF*= 4 L/min. As seen in Fig. 9, the effect of *V* is much higher than that of *WF*. A minimal ozone rate is obtained for higher values of water flow (14.5<*WF*<16 L/min) and lower values of voltage *V* (6.0<*V*<6.2 kV). On the other hand, the ozone production is maximal for higher values of *V* ranging between 7.2 and 8.0 kV.

III. CONCLUSION

Experimental design methodology is a powerful tool for the set point identification of an ozone water treatment process. Through the proper selection of the operating conditions, the process can be made less sensitive to variations, thus avoiding the costly eventualities of bad treatment.

Further research is needed to determine and subsequently minimize the effects of at least two other groups of factors that can cause variations in process response: 1) the geometric dimensions of the ozone generator (length and air gap discharge) and 2) uncontrolled modifications of the environmental conditions (temperature, relative humidity). They will hopefully contribute to the design of more robust process.

REFERENCES

Boonduang, S., S. Limsuwan and W.P. Kongsri, "Effect of Oxygen Pressure and Flow Rate on Electrical Characteristic and Ozone Concentration of a Cylinder-Cylinder DBD Ozone Generator," *Limsuwan Procedia Engineering*, **32**, 936–942 (2012).

Chen, J. and J.H. Davidson, "Ozone Production in the Negative DC Corona: The Dependence of Discharge Polarity," *Plasma Chem. Plasma. Process*., **23**, 501–518 (2003).

Dascalescu, L., A. Samuila, A. Mihalcioiu, S. Bente and A. Tilmatine, "Robust control of electrostatic separation processes," *IEEE Transactions on Industry Applications*, **41**, 715–720 (2005).

Eriksson, L., E. Johansson, N. Kettaneh-Wold, C. Wikström and S. Wold, *Design of Experiments: Principles and Applications*, Learnways AB, Stockholm (2000).

Fang, Z.m Y. Qiu, Y. Sun, H. Wang and K. Edmund, "Experimental study on discharge characteristics and ozone generation of dielectric barrier discharge in a cylinder-cylinder reactor and a wire-cylinder reactor," *J. Electrostatics*, **66,** 421-426 (2008).

Fridman, A., A. Chirokov and A. Gutsol, "Non-thermal atmospheric pressure discharges," *J. Phys. D: Appl. Phys*., **38,** R1–R24 (2005).

Frigon, N. and D. Mathews, *Practical Guide to Experimental Design*, 1st Edn., Wiley, New York (1996).

Jodzis, S., T. Smolinski and P. Sowka, "Ozone Synthesis Under Surface Discharges in Oxygen: Application of a Concentric Actuator," *IEEE Trans. Plasma. Sci*., **39,** 1055–1060 (2011).

Masuda, S., K. Akutsu, M. Kuroda, Y. Awatsu and Y. Shibuya, "A ceramic-based ozonizer using high-frequency discharge," *IEEE Trans. Ind. Appl*., **24**, 223–231 (1988).

Medles, K. A. Tilmatine, F. Miloua, A. Bendaoud, M. Younes, M. Rahli and L. Dascalescu, "Set point identification and robustness testing of electrostatic separation processes," *IEEE Transactions on Industry Applications*, **43,** 618-626 (2007a).

Medles, K. L. Dascalescu, A. Tilmatine, A. Bendaoud and M. Younes, "Experimental Modelling of the Electrostatic Separation of Granular Materials," *Particulate Science and Technology*, **25**, 163-171 (2007b).

MODDE 5.0. *User guide and tutorial*, Umetrics (1999).

Moon, J. and J. Jung, "Effective corona discharge and ozone generation from a wire-plate discharge system with a slit dielectric barrier," *J Electrostatics*, **65**, 660-666 (2007).

Morent, R. and C. Leys, "Ozone Generation in Air by a DC-Excited Multi-Pin-to-Plane Plasma Source," *Ozone Sci. Eng*., **27**, 239–245 (2005).

Nomoto, Y., T. Ohkubo, S. Kanazawa and T. Adachi, "Improvement of ozone yield by a silent-surface hybrid discharge ozonizer," *IEEE Trans. Ind. Appl*., **31**, 1458–1462 (1995).

Park, S., J. Moon, S. Lee and S. Shin, "Effective ozone generation utilizing a meshed-plate electrode in a dielectric-barrier discharge type ozone generator," *J. Electrostatics.*, **64.** 275-282 (2006).

Pekarek, S. "Experimental study of surface dielectric barrier discharge in air and its ozone production," *J. Phys. D: Appl. Phys.*, **45**, 075201 (2012).

Pietsch, G. and J.V. Gibalov, "Dielectric barrier discharges and ozone synthesis," *Pure Appl. Chem.*, **70**, 1169–1174 (1998).

Sung, Y. and T. Sakoda, "Optimum conditions for ozone formation in a micro dielectric barrier discharge," *Surf & Coat Tech.*, **197**, 148-153 (2005).

Taguchi, G., *System of Experimental Designs*, Kraus International Publications, New York (1987).

Tao, S., L. Kaihua, Z. Cheng, W. Jue, Z. Dongdong, Y. Ping and Z. Shichang, "Electrical characterization of dielectric barrier discharge driven by repetitive nanosecond pulses in atmospheric air," *Journal of Electrostatics*, **67**, 215–221 (2009).

Tilmatine, A., S. Bendimerad, M. Younes and L. Dascalescu, "Experimental analysis and optimisation of a free-fall triboelectric separator of granular plastic particles," *International Journal of Sustainable Engineering*, **2**, 184–191 (2009).

Received: May 5, 2014.
Accepted: August 18, 2015.
Recommended by Subject Editor: Jorge Solsona.