

# Characterization of Ozone Distribution in Distilled Water and Coconut Water Produced Using a Double Dielectric Barrier Discharge Machine

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## ABSTRACT

Ozone produced using plasma technology can act as an antimicrobial agent that could be applied in a sterilization process. A Double Dielectric Barrier Discharge (DDBD) machine has ability to produce ozone in sufficient amount for microbial inactivation. The objective of this study is to characterize the ozone distribution expressed as dissolved ozone in distilled water and coconut water produced using a DDBD machine. The information can be useful for industries to design a commercial sterilization process. The results shows that an oxygen flow rate of 0.2 L/min produces the highest ozone concentration, i.e. 3440 mg/L. In addition, the capacity of the machine is relatively similar to all off oxygen flow rate, i.e., 41.28-43.2 g/hour. The oxygen flow rate of 0.2 L/min produces the highest dissolved ozone concentration, i.e. 0.42 mg/L distilled water. This oxygen flow rate is followed by its best kinetic model based on its linear portion during dissolved ozone penetration. A zero order model could describe this penetration process well, including its best  $k$  value of 0.0292 mg/L per minute with the highest dissolved ozone concentration among other oxygen flow rates. Besides, distilled water could represent dissolved ozone penetration in coconut water. Ozone gas and dissolved ozone concentration have possitive corellation with  $R$ -square value of 0.8934.

**Keywords:** Coconut Water; Double Dielectric Barrier Discharge; Oxygen; Ozone.

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## Introduction

Plasma technology finds wide applications in agriculture, medical, wastewater treatment, and industry (Hernández-Torres et al., 2021). One of the known methods for generating plasma involves using high voltage electrodes between gas or open air, known as corona discharge (Saber et al., 2022). While air is typically an insulator and cannot deliver electron, the appropriate distance between

the electrodes leads to the ionization of gas molecules, creating a conductor (Nur et al., 2017). The high electric field generated by the electrodes results in the movement of electrons from gas molecules (Sanito et al., 2022), particularly the highly electronegative ones, to the active electrode or cathode (Okyere et al., 2022). This electron movement leads to collisions, molecular excitation, electron capture, dissociation and ionization of the molecule (Nur et al., 2017),

resulting in the production of species molecules or atoms known as plasma (Okyere et al., 2022). Ozone is one example of plasma generated by dissociation of oxygen gas molecule into single oxygen atoms, which then react with other oxygen gas molecule to create ozone gas (O<sub>3</sub>) (M. Li et al., 2018).

Naturally, ozone is generated in the earth atmosphere and has very reactive property (Liu et al., 2019). Nevertheless, it has beneficial functions due to its oxidation characteristic, and therefore, can be used as antimicrobial agents and applied in wastewater treatment. In larger scale, ozone could be implemented in various industries, such as food industries, medical care, aquaculture business, wastewater treatment, and textile industries waste treatment (Epelle et al., 2022). Ozone has also been used in drinking water industry as novel technology (Laflamme et al., 2020). Naik et al., (2020) have stated that non-thermal technologies such as high-pressure processing, pulsed electric fields, ultraviolet, and ozone can be used as alternative technologies in processing coconut water without reducing taste quality and nutrients. Comparison of ozone with high-pressure processing is that high-pressure processing requires very high pressure, up to 1000 MPa, to inactivate bacterial spores (Naveena & Nagaraju, 2020). This very high pressure certainly requires costly maintenance costs. In processing with ultraviolet and pulsed electric fields, it has a low lethal effect on bacterial spores (X. Li & Farid, 2016).

As a high mineral drink, coconut water could be used as complementary drink other than mineral water. The sugar compound in coconut water could establish the sweet taste and improve the specific flavor together with its natural compound (Mahayothee et al., 2016). The fructose, glucose, and sucrose content in coconut water is around 1,4%, 1,36%, and 0,06% respectively (Kumar et al., 2021). Concentration of Fe, Ni, Cu, Cd, Cr, Zn, Pb dan Se in coconut water is 0,3-1,5; 7,77-21,2; 0-0,71; 0-0,9; 0-0,2; 0,9-17,3; 0,1-0,9; 0-0,9; and 0-0,7 mg/L, respectively (Islam et al., 2021). Potassium was informed as the highest

mineral content in coconut water which is around 2163 µg/mL (Raj CT et al., 2023). Alchoubassi et al., (2021) explained that the trace elements in coconut water could support the metabolism process in human body.

Ozone plasma is suitable for coconut water treatment because the main compounds is water. Moisture content of coconut water is around 94% (Coulibaly et al., 2023). Rajashri et al., (2020) had applied ozone plasma to treat coconut water. They evaluated the effect of ozone treatment towards polyphenol and endogenous enzyme degradation, continued by the shelf-life evaluation. Recently, Cheong et al., (2021) measured the effectiveness of ozone generated by dielectric barrier discharge plasma reactor against multidrug-resistant bacteria and *Clostridium difficile* spore. They reported 2 log reduction in stainless steel, fabric and wood media. In addition, they found 1-2 log reduction in glass and plastic for vancomycin-resistant *Enterococcus faecium* and carbapenem-resistant *Acinetobacter baumannii* by 500 ppm ozonation within fifteen minutes. However, *Clostridium difficile* spore was the most resistant as compared to the other evaluated bacteria. Naik et al., (2020) furthermore reported that ozone has high potential to be an alternative technology in replacing thermal process (Prades et al., 2012) for coconut processing technology. Acting as antimicrobial agent, ozone could inhibit the growth of microbe. The mechanism of microbe inactivation is caused by the degradation of both vegetative and spore membranes. Ozonation conducted by Wood et al., (2020) could reduce more than 6 log for both *Bacillus subtilis* and *Bacillus anthracis* spores. Moreover, Torlak & Isik (2018) reported that ozonation could reduce 2,3-4 log *Paenibacillus larvae* spore on different media. This spore reduction capability indicates the high potential of ozone to be applied in commercial sterilization process. Additionally, ozone can naturally be degraded and separated into oxygen gas in high water content media which does not

produce hazardous byproduct (Shezi et al., 2020). Ozone has been asserted as Generally Recognized as Safe (GRAS) in 1997 and it could be used in liquid food treatment (Porto et al., 2020).

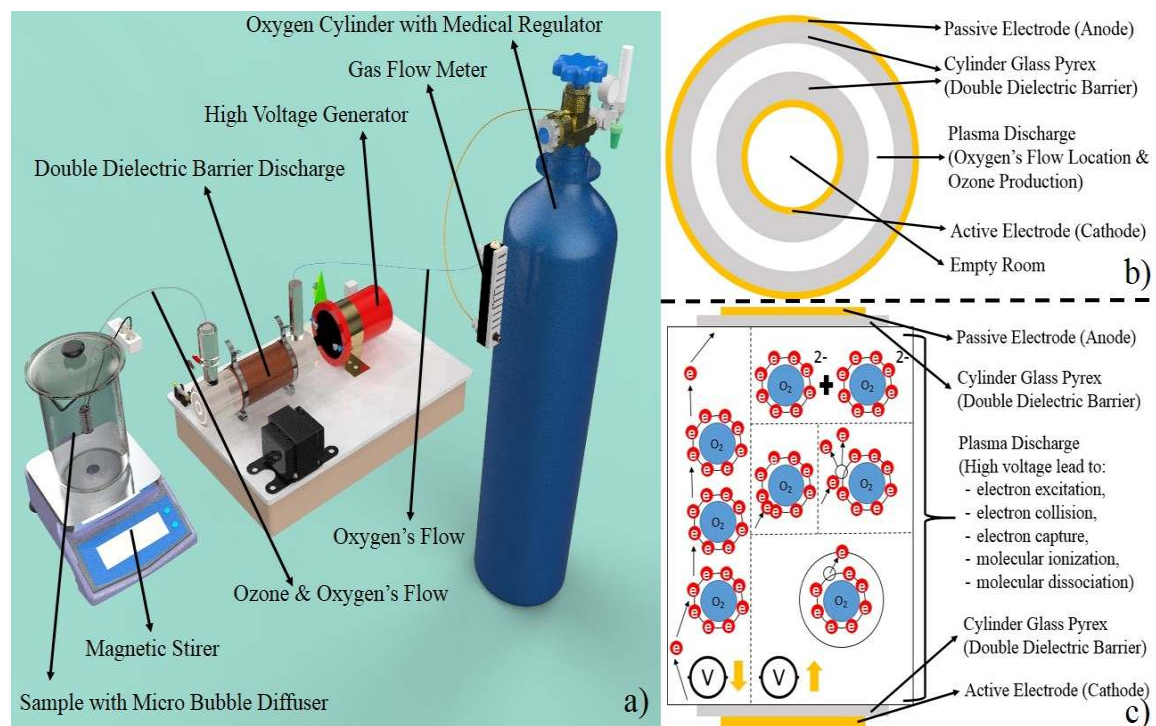
In order to effectively implement commercial sterility through sterilization processes, it is crucial to understand the characteristics of each machine being used, particularly the ozone generator under examination in this study. A key aspect of this understanding is the characterization of the distribution of dissolved ozone in the sample during the ozonation process. It is essential to identify the concentration of dissolved ozone in the sample as a function of ozonation time, as this data can provide valuable insights into the most efficient methods for ozonation. Additionally, this data can help determine the come-up time required for the machine to be ready for the ozonation process.

In this particular study, the double dielectric barrier discharge (DDBD) method is used to generate ozone by assembling a glass on each electrode. Ozone is produced between two glass surfaces, effectively creating a double dielectric setup in the machine. The aim of this study is to characterize the distribution of ozone in terms of dissolved ozone concentration in both distilled water and coconut water obtained from a DDBD machine.

## Materials and Methods

### Materials

Fresh green coconuts (*Cocos nucifera*) were procured from a local market in Banyumanik, Semarang District, Central Java Province, Indonesia. The ozone test kit reagent used in the study was obtained from Merck, while the ozone measurement instrument utilized was Spectroquant Move DC, also from Merck.



**Figure 1. DDBD Machine for Ozone Production: (a) Component Arranging Ozone Production, (b) Schematic of DDBD from Front View, (c) DDBD from Side View including Possibility for Ozone Formation**

### *Coconut Water Sample Preparation*

After removing the skin and mesocarp (fiber) layer of the green coconut fruits with a sterile knife, coconut water was extracted and filtered using Whatman 42 filter paper before being used in the study. The filtered coconut water was used immediately. As a control, distilled water was used, as it is the standard and ideal for measuring dissolved ozone concentration due to its lack of compounds that may affect the ozone concentration measurements

### *Designing of DDBD Machine*

The machine used in the study consisted of two glass cylinders that formed a double dielectric barrier. The cylinders had a length of 16.5 cm and were arranged in an inside-outside configuration, with the smaller 2 cm diameter cylinder placed inside the larger 4 cm diameter cylinder. The distance between the cylinders formed a 0.5 cm room that allowed for the passage of oxygen and the production of ozone. The outside of the smaller cylinder was coated with a copper plate that acted as the active electrode. The outside of the larger glass cylinder was also covered by a copper plate, which acted as the negative electrode. The ozone production system used pure oxygen that flowed from a container through a gas regulator and flowmeter into the DDBD chamber, as shown in Figure 1a. The DDBD chamber was equipped with a micro bubble diffuser that allowed for the flow of ozone and oxygen into the sample. The filtered coconut water and distilled water were used as samples in the study.

### *Preparation of Ozone Generator and Ozone Production*

The process began by flowing oxygen gas from a container through a medical oxygen regulator and a gas flowmeter into the reactor chamber. The flow rate of oxygen gas was adjusted to 0.2, 0.4, 0.6, and 0.8 L/min. A micro bubble gas diffuser, connected to the reactor chamber by a rubber tube, was placed at the base of a 1 L

sample container. The samples were then agitated using a magnetic stirrer at 1000 rpm. Once the gas was flowing, a high electrical voltage of 500 volts was applied to two electrodes in the reactor chamber. This voltage caused the oxygen gas to separate into single oxygen atoms, which then reacted with other oxygen gas molecules to produce ozone. The current electricity used on high electrical voltage (500 volts) is alternating current. Alternating current is suitable for ozone generation by high electrical voltage method (Mikeš et al., 2023). During ozonation process, the liquid product sample was adjusted by room temperature around 26°C.

The illustration of ozone gas production is presented in Figure 1c. The low voltage applied in both of electrodes affected the gas to have possibility for electron moving. By this character change, the electron from cathode will distribute and flow through valence electron of the gas to the anode side (Mouele et al., 2021). If the voltage between both of electrodes is increased, it will create strong electric field around the electrodes including the gas between them. Oxygen gas molecule around it will form an ozone gas molecule due to some electron activities.

### *Characterization of DDBD Machine*

The dissolved ozone concentration in the sample was measured at intervals of five minutes for a total of 60 minutes. After a certain amount of time, it is expected that the concentration of dissolved ozone will begin to stabilize. The data obtained from measuring dissolved ozone concentration was plotted in zero-order (Equation 1) and first-order kinetic (Equation 2) models. The model with the highest R-square value was chosen as the best fit for the plotted data.

$$[C_t] = k * t + [C_0] \quad (1)$$

$$\ln[C_t] = k * t + \ln[C_0] \quad (2)$$



### *Determination of Ozone Gas and Capacity of Machine for Ozone Production*

To determine the capacity of the ozone generator machine for ozone production, a measurement of ozone gas was conducted using a liter of 0.2 M potassium iodide and 100 mL of 0.4 M sodium thiosulfate. 50 mL of the potassium iodide solution was added to an Erlenmeyer flask, and the rubber tube from the machine was placed directly into the solution. The machine was then turned on for 2 minutes of ozonation, during which the presence of ozone gas would cause the solution to turn yellow (Yulianto et al., 2019). The solution was then titrated with sodium thiosulfate 0.4 M until it became clear or uncolored, and the ozone gas concentration was calculated using Equation 3, where  $O_3$  represents the

ozone gas concentration (g/L),  $V_t$  is the volume of sodium thiosulfate (mL),  $N_t$  is the normality of sodium thiosulfate (mol/L), and  $V_g$  is the volume of oxygen (L). The performance of the ozone generator machine was evaluated based on the amount of ozone gas measured from the chamber.

$$O_3 \left( \frac{g}{L} \right) = \frac{24000 * V_t * N_t}{V_g} \quad (3)$$

The ozone generation capacity of the DDBD machine was determined by converting the concentration of ozone gas from mg/L to g ozone/hour for each oxygen flow rate (L/min). This calculation was performed using Equation 4:

$$\text{Machine Capacity} \left( \frac{g}{\text{hour}} \right) = \left( \text{oxygen's flow} \left( \frac{L}{\text{min}} \right) \times O_3 \left( \frac{g}{L} \right) \right) \times 60 \quad (4)$$

### *Determination of Dissolved Ozone Concentration*

The concentration of dissolved ozone in the liquid sample was determined using a Spectroquant tube analyzer (Ateia et al., 2018). Ozone test kit reagents were added to 10 mL of the liquid sample in the tube. Firstly, 2 drops of  $O_3$ -1 reagent were added, and the tube was shaken for 10 seconds. Then, a couple of little spoon dosages of  $O_3$ -2 reagent were added from the cap of the packaging, and the tube was shaken for 10 minutes. The reaction of ozone and reagent resulted in a pink color in the test tube, which was observed within a minute. Aquadest was used for a blank reagent. The Spectroquant Move DC ozone analyzer was calibrated using the blank reagent before measuring the dissolved ozone concentration in mg/L from the sample. The distribution of dissolved ozone concentration during the ozonation process by the DDBD machine was measured for 60 minutes at oxygen flow rates of 0.2, 0.4, 0.6, and 0.8 L/min for both distilled and coconut water samples.

## **Results and Discussion**

### *Ozone Gas Production and Capacity of Machine for Ozone Production*

What is meant by ozone gas is specifically defined as the ozone gas which is produced directly from the DDBD chamber. Mechanism of the machine for ozone production is begun by flowing pure oxygen gas through DDBD chamber. In this chamber, the high electrical voltage using alternating current is conducted. The active and ground electrodes are set between oxygen track covered by a double dielectric materials. Electron discharge from high electrical voltage hits the electron in oxygen gas molecule. This activity will make a oxygen gas dissociation. A single oxygen atom will react with other oxygen gas molecule to form a ozone molecule. This ozone gas is ready to be diffused into liquid sample through micro bubble diffuser. A DDBD system is one of novel technology for plasma generator producing ozone (Hu et al., 2023). The ozone gas concentration from DDBD machine in this study could be seen in Table 1. The data shown in Table 1 indicates that the higher oxygen flow rate

passing through double dielectric barrier chamber gradually reduces the ozone gas concentration. It is clearly recognized that the ozone gas concentration are 3440, 1760, 1173.33, and 900 (mg/L) for each oxygen flow rate 0.2, 0.4, 0.6, and 0.8 (L/min), respectively. This data has similar trend with the experiment conducted by Restiwijaya et al. (2019) and Chasanah et al. (2019). They also produced the high ozone gas concentration in low oxygen flow rate input. However, their result showed that the increase of oxygen flow rate input will decrease ozone gas concentration. It reveals that ozone gas production is affected by the residence time in the location between two electrodes (Chasanah et al., 2019). Nonetheless, oxygen flow rate influences ozone gas concentration which is formed in chamber

from DDBD machine. The lower rate for oxygen passing through double dielectric chamber will give longer opportunity of time for oxygen gas dissociation and form an ozone gas molecule directly. It is caused by a longer time of oxygen molecule passing the chamber which gives long time of that molecule to be hit by high kinetic energy electrons from high voltage electrodes. In reverse condition, the very fast oxygen passing through double dielectric chamber results in the low opportunity times for ozone production due to the rapid oxygen movement from double dielectric chamber. It has been evaluated by Nur et al., (2017) that the lower gas flow input in ozone reactor chamber leads to the higher ozone gas concentration.

**Table 1. Ozone Gas Concentration and Capacity of Machine for Ozone Production**

| Oxygen Flow Rate (L/min) | Ozone Gas Concentration (mg/L) | Capacity of Machine for Ozone Production (g Ozone/hour) |
|--------------------------|--------------------------------|---|
| 0.2                      | 3440                           | 41.28   |
| 0.4                      | 1760                           | 42.24   |
| 0.6                      | 1173.33                        | 42.24   |
| 0.8                      | 900                            | 43.2  |

On the other hand, the capacity of machine to produce ozone gas is reasonably similar in the system built in this experiment. By the use of similar voltage in the entire oxygen flow rate, the result of the machine capacity is quite similar to certain gram ozone gas produced per hour. It means that the capacity of machine for ozone production is relatively constant at 42 g ozone/hour between 0.2-0.8 L/min oxygen flow rate. This results are comparable to the experiment conducted by Chasanah et al., (2019), which had similar capacity of machine in producing ozone (g/hour) in various oxygen flow rate inputs. However, this minor difference is motivated by the flow rate of oxygen passing in the ozone reactor chamber. As a result of the higher rate of

oxygen input per minute, it slightly enhances the ozone product (g) in every hour. Essentially, it is only the probability of ozone production influenced between the ozone residence time within ozone reactor and the amount of oxygen supplied in the ozone reactor chamber. According to the actual calculation data for the gram of ozone produced in an hour, the higher residence time of oxygen gas in the ozone chamber entails slightly lower capacity in contrast to the higher oxygen supply in ozone chamber as L/min. Although the ozone concentration is very high, the oxygen flow rate is considered to be low. The possibility of added treatment in the ozone machine system for large scale food/beverage processing is the installation

of continuous processing system. The liquid form of coconut water is suitable to be flown through a tube. Adopting the similar concept with Ultra High Temperature (UHT) processing, the coconut water liquid should be flown in a tube that is penetrated by ozone gas into the liquid. The dissolved ozone concentration should be held for certain time based on the time that is needed to inactivate 12 decimal logs of *Clostridium botulinum* spores. This process has to be evaluated for the necessary of dissolved ozone concentration for 12 decimal logs inactivation of that bacterial spores.

#### *Distribution of Dissolved Ozone Concentration during Ozonation Process by DDBD Machine*

Despite the fact that the machine could produce much of ozone gas molecule, it is very important to tell how much ozone is distributed to all of the product's side. Micro bubble diffuser reduced bubble size approximately into the micrometer size (Zhang et al., 2013). Following the agitation process, the micro bubble was destructed and collapsed, continued by dissolving and distributing of ozone molecule into the liquid sample's side (Zhang et al., 2013). Referring to thermal process, this distribution was analogized into the point in product which receives heat by the longest time. Dissolved ozone concentration was essential for validation or create equivalence of ozonation process for commercial sterility business. Dissolved ozone concentration become a standard point to determine the effectiveness of ozonation process to inactivate microbes or other purposes.

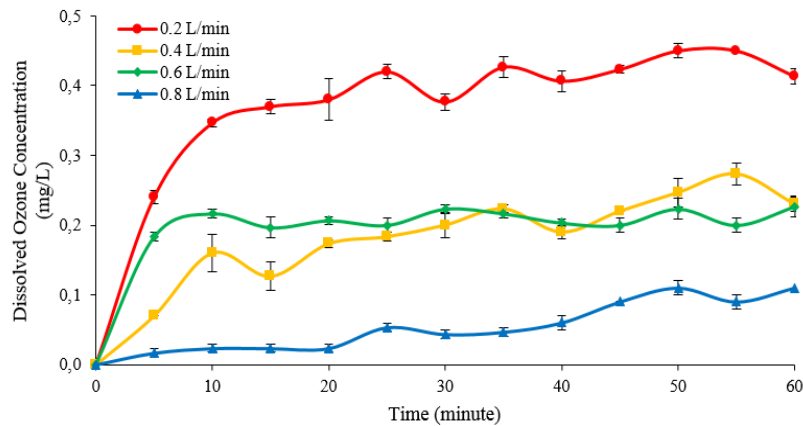
The measurement of the distribution of dissolved ozone concentration during ozonation process by DDBD machine is principal to identify, because if there is planning to design a sterilization process, the information about the ozone received in the sample needs to be seen. Especially in commercial sterility, its effectiveness is highly dependent on the amount of ozone received by sample. It is expected that the greater the amount of the dissolved ozone

presented in the sample, the more ozone molecules are capable to inactivate microbes. Furthermore, the amount of ozone gas contained in the unit volume of carrier air varies greatly depending on the machine capability in producing ozone. It likewise indicates that each ozone generator machine has its own ozone production capability. Therefore, it is necessary to characterize the DDBD machine as ozone generator machine for ozone production to the time it is dissolved into sample. The characterization of the ozone generator machine is carried out by measuring the dissolved ozone concentration in sample as a function of the ozonation time at certain oxygen gas flow rate.

The amount of dissolved ozone concentration at different oxygen flow rate as function of time is presented in Figure 2. This Figure shows that the difference of oxygen flow rate produces clear different dissolved ozone concentration. Contrariwise, Figure 3 shows the unclear dissolved ozone concentration at different oxygen flow rate. It could happen because coconut water contains interfering compound reacting with ozone kit test reagent, such as sugar, trace element, and minerals. As informed previously by Kumar (2021), coconut water contains sugar i.e fructose, glucose, and sucrose. Trace element including many of minerals are available in coconut water (Islam et al., 2021). Based on the experiment in this study, it could be known that the ozone test kit reagent which is measured by Spectroquant Move DC instrument does not work well for determining of dissolved ozone concentration in coconut water product. It could be happened because of the impurities of coconut water sample. That reagent is only appropriate for pure water. According to the data on dissolved ozone concentration in both distilled water and coconut water, they are actually still in the range of 0.1-0.5 mg/L. They also tend to be similar as to the unavailability of ozone condition at 0 minute and the saturation condition in term of dissolved ozone concentration. Based on

that consideration, distilled water could be used to interpret the ozone dissolution in coconut water. Furthermore, the tendency

for ozone penetration from the beginning of ozonation process until it reaches the initial point of saturation has been evaluated.



**Figure 2. Dissolved Ozone Concentration in Distilled Water as affected by Different Oxygen Flow Rate (L/min)**

The dissolved ozone concentration at 0.2 L/min oxygen flow in distilled water and coconut water can be viewed in Figure 2 and Figure 3. The initial dissolved ozone concentration of both distilled water and coconut water are different by the amount of 0 and 0.11 mg/L, respectively. While the dissolved ozone concentration in distilled water reaches its saturation on approximately 25 minutes after the beginning of ozonation process, the case of coconut water turns out to come up from 5 minute to 60 minutes. Comparing both samples in the 0.2 L/min oxygen flow, it can be implied that the concentration of dissolved ozone in distilled water is higher than that in coconut water. Data showing 0.4 L/min oxygen flow seems different compared to 0.2 L/min oxygen flow. According to the flow condition, coconut water has higher dissolved ozone concentration either at the beginning time or during the ozonation process, although there is a point in 10 minutes which shows higher dissolved ozone concentration in distilled water. To be underlined, the initial dissolved ozone concentration in coconut water is 0.1 mg/L, while it is in the distilled water is 0 mg/L. Nevertheless, both samples present similar tendency of ozone penetration. They start to grow from the beginning

of ozonation process, and reach the saturation point around 35 minutes.

Besides, oxygen flow rate of 0.6 L/min gives a pretty similar trend for dissolved ozone concentration between distilled water and coconut water. Both samples have similar line and point to achieve the saturation condition. Approximately at 10 minutes, both samples reach the saturation. However, the coconut water sample has the second growth line by the very low slope. It makes the higher dissolved ozone concentration in coconut water requires 60 minutes, compared to the distilled water. Although coconut water sample has higher dissolved ozone concentration at the initial time, overall, the dissolved ozone concentration in distilled water is higher than coconut water. It occurs at 5 to 50 minutes. Moreover, it also could be seen that both lines of samples are tangent during 40 to 50 minutes.

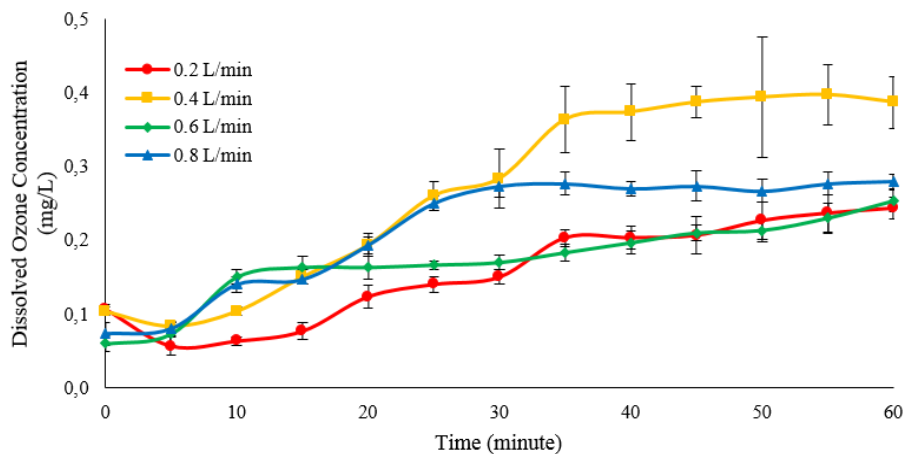
The dissolved ozone concentration on 0.8 L/min oxygen flow could be seen in Figure 2 and Figure 3 which respectively represents distilled water and coconut water. Both of them show higher dissolved ozone concentration from the starting point of ozonation process until its sixtieth minutes.



Figure 2 and Figure 3 show the clear different concentration value between distilled water and coconut water at 0.8 L/min. Furthermore, both distilled water and coconut water show that the saturation point takes place around 25-30 minutes. Nevertheless, it can be predicted that there is a growth of dissolved ozone concentration in distilled water from 30-60 minutes by a small slope.

Referring to Figure 2, it imagined about come-up time of machine from the

beginning of machine operation until when it achieved the saturation condition of dissolved ozone concentration in sample. In this case, there is no issue about the homogeneity of dissolved ozone concentration in sample. As it had been confirmed that there is no different dissolved ozone concentration in every part of the liquid sample, it can be concluded that the sample for analysis in any part of the sample is possible to be taken.



**Figure 3. Dissolved Ozone Concentration in Coconut Water as affected by Different Oxygen Flow Rate (L/min)**

#### *Characterization of Ozone Distribution by DDBD Machine*

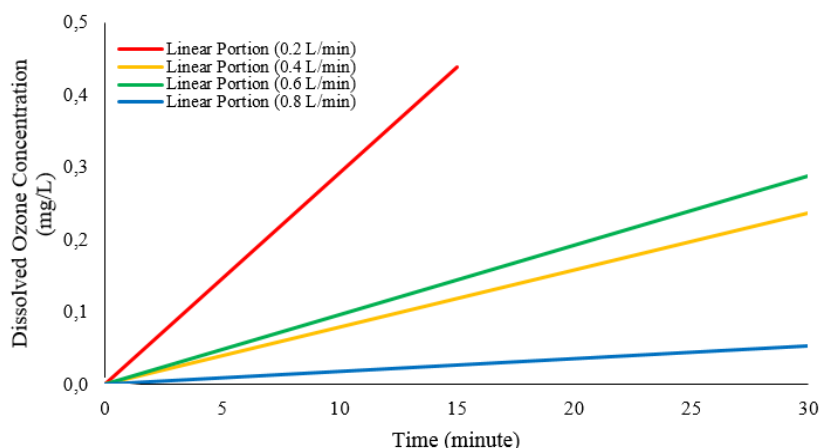
However, the characterization of DDBD machine for ozone production is evaluated from the distribution data of dissolved ozone concentration in both samples during ozonation process. The character of this machine is studied focusing on the capability of machine and its system for ozone production, including to dissolve the ozone into the liquid sample, i.e., distilled water and coconut water. This point of view could be described from the linear portion of the distribution data of dissolved ozone concentration in both samples during ozonation process. All of the ozonation process is started from penetrating step to gaining the saturation condition of ozone gas as ozone generator machine character (Chasanah et al., 2019). The exclusivity of the distribution data of dissolved ozone concentration in both samples during ozonation process

lies in the differences of dissolved ozone concentration data between both samples. For the oxygen flow rates of 0.2 and 0.6 L/min, the lower dissolve ozone concentration on coconut water presents. Oppositely, the oxygen flow of 0.4 and 0.8 L/min perform higher dissolved ozone concentration on coconut water. In fact, all of the data should have similar data on each oxygen flow rate. Owing to the fact that the highest compound in coconut water is water, it is likely to be safe and more appropriate for ozone treatment application.

However, it still has different data result for dissolved ozone concentration compared to distilled water as the standard for dissolving ozone along with the determination by ozone test kit reagent. By these differences in data result, it indicates that there are some chemical compounds interrupting the ozone test kit reagent for ozone determination. This is different from

distilled water that contains only H<sub>2</sub>O which does not react with ozone test kit reagent. Additionally, the reagent only works by the presence of ozone in the sample due to the ozonation process inside. There are a couple of possibilities for explaining the difference in dissolved ozone concentration in coconut water. First, the ozone produced by machine will react with natural coconut compound to form specific compounds that can subsequently react with ozone test kit reagent. Moreover, it is detected as ozone by Spectroquant Move DC instrument. Second, the ozone test kit reagent reacts immediately by the natural compound of coconut water and it is quantified as ozone

by Spectroquant Move DC instrument. On other side, all of the findings for each oxygen flow rate show similar trend to reach the saturation point. They have similar growth trend of dissolved ozone concentration since the beginning of ozonation process. By the similar growth and saturation point trends, the machine and its system could penetrate the ozone to dissolve into the distilled water that as well represents the penetration of ozone to dissolve into the coconut water. Moreover, the value of dissolved ozone concentration in distilled water represents the value of dissolved ozone concentration in coconut water.



**Figure 4. Penetration of Dissolved Ozone in Distilled Water by Zero Order Model**

To characterize the DDBD machine during ozone production and dissolve into the liquid sample, the linear portion of the entire data from distilled water sample are plotted into zero and first kinetic models. The evaluation can be seen through the slope resulted from both of kinetic models. In addition, the plotting of dissolved ozone concentration on linear portion by zero order kinetic model could be seen in Figure 4. From the data shown in the figure, it is explained that the flow rate of oxygen supply by 0.2 L/min results the highest k value, i.e., 0.0292 mg/L per minute. The k value indicates the coefficient value of dissolved ozone concentration (mg/L) rise within the change of time by one minute of ozonation

process. The k value of 0.2 L/min oxygen flow rate is followed by 0.6, 0.4, and 0.8 L/min to the lower k value. Essentially, this k value can also interpret the time to reach saturation of dissolved ozone concentration in the sample. The highest k value means the faster condition based on the oxygen flow rate to reach the saturation of dissolved ozone concentration in the sample. Besides, the highest k value on 0.2 L/min oxygen flow rate can be explained by the retention time of oxygen gas while passing through DDBD reactor column. The higher retention time of oxygen gas which passes through DDBD reactor column will result in the higher amount of oxygen gas dissociated into single oxygen atom (Zain et al.,

2019). This single oxygen atom will react with other oxygen gas to form ozone.

The  $k$  value of 0.4 L/min is to some extent lower than that of 0.6 L/min oxygen flow rates which are 0.0038 and 0.0040 mg/L per minute, respectively. In the case of 0.6 L/min oxygen flow, it gives the faster oxygen movement than that of 0.4 L/min which affects the lower retention time of oxygen gas in DDBD column. Either lower retention time or faster oxygen movement in DDBD column is able to reach the faster saturation of dissolved ozone concentration but in lower dissolved ozone concentration itself. Particularly, retention time affects the possibilities of ozone production regarding the amount of dissolved ozone in sample and  $k$  value as the time to reach saturation condition for dissolved ozone amount in the sample. However, the  $k$  value trend is relatively different between what is shown by 0.4 and 0.6 L/min oxygen flow rates and 0.2 and 0.8 L/min oxygen flow rates. In Figure 4, it is illustrated that between 0.2 and 0.8 L/min oxygen flow rates, the clearly difference on  $k$  value appears. It means that the relatively far-off gap between 0.2 and 0.8 L/min oxygen flow rates result the clearer difference on both the amount of dissolved ozone in sample and the  $k$  value as the time to reach saturation condition in favor of dissolved ozone amount in sample. Nevertheless, the 0.4 and 0.6 L/min oxygen flow rates appear to have insignificant difference in term of the retention time of oxygen gas.

The transition of oxygen movement speed and the transition of oxygen gas molecule dissociation opportunity occurred here. The quick oxygen gas movement initiates the low oxygen gas molecule dissociation (Maftuhah et al., 2020). This condition results in shorter time to reach the saturation condition of dissolved ozone concentration but in low amount in the sample. It is attested by the  $k$  value from 0.6 L/min oxygen flow rates. It shows shorter time in reaching the saturation condition of dissolved ozone concentration, yet in low amount of dissolved ozone in the sample. The transition in 0.6 L/min oxygen flow rate

has the tendency of 0.2 L/min in terms of time to reach saturation, high  $k$  value. The transition in 0.6 L/min also has the tendency of 0.8 L/min oxygen flow rate in terms of low amount of dissolved ozone concentration compared to 0.4 L/min oxygen flow rate.

It is different from the 0.4 L/min oxygen flow rates which has lower  $k$  value compared to the 0.6 one. The lower oxygen gas moving in 0.4 L/min oxygen flow rates entails the higher retention time of oxygen gas in DDBD column. This higher retention time results more oxygen gas molecule dissociated into a couple of single oxygen atoms that are able to react with other oxygen gas molecule to form an ozone molecule. Such condition reinforces the higher dissolved ozone concentration of 0.4 L/min oxygen flow rates at the saturation condition compared to 0.6 L/min oxygen flow rate. That reason supports the transition character of 0.4 L/min oxygen flow rate to have a tendency with 0.2 L/min oxygen flow rate that has highest dissolved ozone concentration at saturation condition. The transition in term of come up time, the 0.4 L/min oxygen flow rate has 0.8 L/min oxygen flow rate which has lowest  $k$  value.

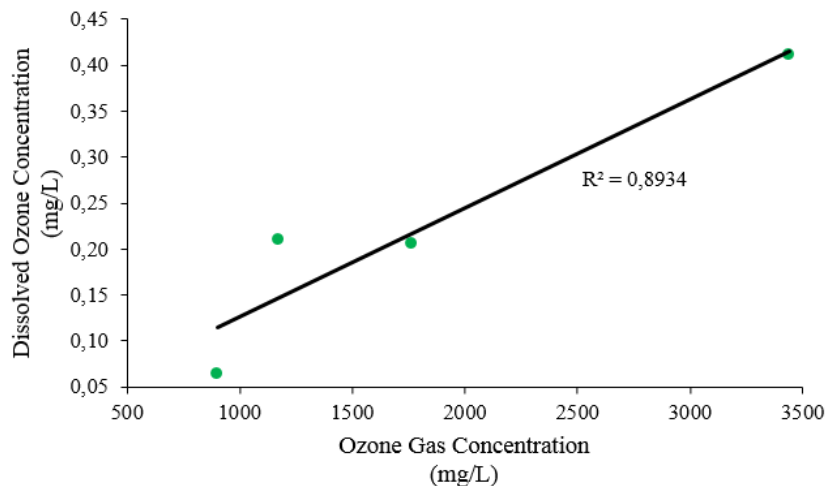
The 0.8 L/min oxygen flow rates are the lowest for both of  $k$  value and dissolved ozone concentration during ozonation process. As well, the retention time of oxygen gas in the DDBD column for every oxygen flow rate has been notified above. The 0.8 L/min oxygen flow rates possess the fastest oxygen movement compared to those of all oxygen flow rates. Subsequently, it results in the measly oxygen gas dissociated in DDBD column (Restiwijaya et al., 2019). This brings about the low dissolved ozone concentration in liquid sample including the  $k$  value reduction.

Characterization of DDBD machine for ozone production dissolved in liquid sample plotted in first order kinetic model demonstrates low performance. The line trend of all oxygen flow rates shows slightly akin tendency to the line trend plotted in zero order kinetic model. However, from

the comparison of R-squared value between zero and first order kinetic models, the zero one has higher R-squared value for all oxygen flow rates compared to the first order kinetic model. The evaluation of the best kinetic model can be approached by the highest R-squared value (Kebede et al., 2015). It implies that the best kinetic model which visualizes the ozone production and dissolving in liquid sample processes by DDBD machine follows zero order kinetic model. Further business as the use of dissolved ozone concentration determination by DDBD machine could implement the zero order kinetic model. The high slope (k) value represents the faster ozone penetration into the sample. By this characterization, there will be provided information about oxygen flow rate for ozone generator machine operation, come up time to achieve dissolved ozone stability in sample, and the concentration of dissolved ozone in the sample when it starts to reach stability.

### *Correlation Between Ozone Gas and Dissolved Ozone Concentrations*

In the beginning, it has been clearly defined that the position between ozone gas and dissolved ozone concentrations are separated. Ozone gas concentration refers to ozone gas molecule which is generated directly from the ozone reactor chamber. Ozone molecule is established from the one single oxygen atom and a oxygen molecule due to a high energy electrons moving. Meanwhile, ozone gas concentration could be interpreted as a number of ozone in default form. The sensitivity of potassium iodide to react with ozone as oxidator compound generates the change of potassium iodide solution from defined clear into yellow (Verinda et al., 2022). The large number of ozone molecule shows more intense yellow in colour. Based on the method of ozone gas concentration determination, dipping the rubber tube into the potassium iodide solution should raise the strong yellow colour.



**Figure 5. Correlation Between Ozone Gas Concentration and Dissolved Ozone Concentrations**

The separated part of dissolved ozone concentration is attributable to the presence of dissolution technology. The difficulty faced by ozone to dissolve in the liquid media leads to the reduction of the ozone gas bubble size in the liquid media. To be underlined, microbubble diffuser acting as

bubble size reducer is the separated technology from ozone reactor. While ozone reactor is used to generate ozone molecule in gas form, microbubble diffuser is applied to produce the very small size of ozone gas. The dissolution of ozone gas after in small size bubble is completed by

the agitation in liquid media. Consequently, agitation will give a force in all of liquid side. This condition possibly disturbs the small size of ozone bubble, thus make it to dissolve in the liquid media. The other difference of ozone gas and dissolved ozone concentrations is their determination condition. Ozone gas concentration is determined in potassium iodide solution with ozone injection continuously, whereas dissolved ozone concentration is determined by no ozone injection. It takes 10 mL of the ozonated liquid, separated from ozone injection system, and it is continued by addition of reagents for determination in Spectroquant Move DC analyzer (Verinda et al., 2022).

What is meant by ideal condition for relation of ozone gas and dissolved ozone concentration refers to the similarity of enhancement of both concentrations during the reduction of oxygen gas flow input. The ozone gas which is forced to pass microbubble diffuser should be disturbed by agitation. Eventually, the whole ozone molecule in microbubble form will dissolve into liquid media. Nevertheless, it is too difficult to dissolve all of ozone molecule into liquid media. At this point, microbubble diffuser still produces bubbles which are not interrupted in the liquid media. They move to the surface area of the liquid media and gone afterward to open air. This phenomenon is in accordance with the result shown in Figure 5. The dissolved ozone concentration shows very low concentration at the similar point to ozone gas concentration which shows very high concentration. The concentration of ozone gas and dissolved ozone at the oxygen flow rates of 0.2; 0.4; 0.6; 0.8 L/min are correspondingly 3440 and 0.45 ppm; 1760 and 0.27 ppm; 1173.33 and 0.23 ppm; 900 and 0.11 ppm. Both concentration of ozone gas and dissolved ozone show very distant values. It indicates that not all of ozone molecules produced from the ozone reactor chamber are dissolved in the liquid media.

However, this result reveals similar tendency to the study of dissolved ozone

concentration conducted by Verinda et al., (2022). Their study showed the concentration of dissolved ozone only around 0,05-0,5 mg/L by the similar DDBD machine at 0.3 L/min oxygen gas flow. The single dielectric barrier discharge machine also had quite similar tendency for dissolved ozone concentration, i.e., around 1.8-2.1 mg/L by 10-15 L/min outdoor air flow (Zahar et al., 2019). Additionally, ozone gas concentration analysis run by Nur et al., (2017) similarly showed reduction while its dry air flow rate input in ozone reactor was escalated. Maftuhah et al., (2020) also had similar trend for ozone gas concentration reduction under the increase of oxygen flow rate input. As a note, the ozone gas concentrations ranged from 100-850 mg/L (Maftuhah et al., 2020). All of the findings in the previous research support the correlation between ozone gas and dissolved ozone concentrations in this study. Furthermore, it can be implied from Figure 5 that the increase of oxygen flow rate input will reduce both ozone gas and dissolved ozone concentrations. Figure 5 also showed the positive correlation with R-square value of 0.8934. From Figure 5, it could be informed the correlation between ozone gas concentration and dissolved ozone concentration during ozonation process of coconut water product. By the linear regression in Figure 5, the dissolved ozone concentration in product will be known if the machine is operated on certain ozone gas concentration. By this linear regression, it does not need to analyze the dissolved ozone concentration in product for every ozonation processing. That explanations are valid as long as using a similar machine and conducting by similar ozonation process system.

## Conclusion

Based on the results of this study, it can be concluded that the machine design can perform to produce ozone well. The lower oxygen flow rate brings about higher ozone gas concentration and dissolved ozone concentration in liquid sample. However, the machine has similar capacity for



ozone gas production on all oxygen flow rates, around 42 g ozone/hour. To end with, ozone gas and dissolved ozone concentrations have positive correlation with the R-squared value of 0.8934. The overall results in this study contribute for academics and industry practitioners to characterize the ozon generator machine which will be used for commercial sterility equivalence.

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