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Increasing Peroxide Number of Sumbawa Oil Through Ozonation Process Using Double Dielectric Barrier Discharge (DDBD) Plasma Reactor

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Abstract: Oil is one of the most popular media for the ozonation process compared to other media such as water or saline methods or direct exposure to blood. Ozonated oil has an advantage over other ozone therapy methods because ozonated oil has an antiseptic activity that is several hundred times higher than ozonated saline. Sumbawa oil is one of the local wisdom products of the Sumbawa tribal community, which is generally used as medicine or massage therapy to heal injuries such as broken bones, tendonitis, bruises, back pain and treatment of various skin wounds. An ozonation process can be carried out to increase the ability of Sumbawa oil to regenerate skin and repair tissue. Ozonation uses a Double Dielectric Barrier Discharge (DDBD) plasma reactor. The DDBD Plasma Reactor is a reactor that can produce ozone gas with very high purity. Pure ozone gas is very well applied in the medical world. This research aims to increase the peroxide levels in Sumbawa oil by ozonation using a Double Dielectric Barrier Discharge (DDBD) Plasma reactor. The flow rates used were 0.1 L/minute, 0.4 L/minute and 0.7 L/minute for 1 hour, 3 hours and 5 hours. The results showed that the peroxide value increased with increasing oxygen flow rate and ozonation time. The highest peroxide value was obtained at a flow rate of 0.7 L/minute for 5 hours, namely 332.03 mg Eq/kg Sumbawa oil. Increased by 282.71 mg Eq/kg; the peroxide figure for Sumbawa oil without ozonation was 49.32 mg Eq/kg.

Keywords: DDBD Plasma Reactor; Ozonation; Ozone Gas; Sumbawa Oil; Peroxide Number.

Introduction

Ozone has many benefits in various fields. In the medical field, ozone can be used for cancer, diabetes, and heart and liver therapy, inhibiting aging and healing chronic wounds such as ischemic and diabetic wounds, which require a long healing time. This is because ozone functions as an inactivating agent for bacteria, viruses, fungi, and several types of protozoa, which can be used as clinical therapeutic agents [1], [2]. Ozone compounds for wound therapy can be carried out in several methods, such as ozone being exposed directly to the skin [3], Extracorporeal blood oxygenation and ozonation (EBOO) [4] and ozonated water [5]. Excessive methods of using ozone directly in gas form can harm human health. Therefore, the alternative use of water, saline and ozonated oil is preferred.

Ozonated oil has antiseptic activity several hundred higher than ozone saline. Ozonated oil also lasts longer, up to 2 years, than ozonated water, which quickly decomposes into oxygen. Ozonated oil can function as an effective disinfectant because it contains unsaturated fats. For the health sector, ozonated oil can act as an antibacterial [6], [7], [8]. The reaction of ozone with oil or ozonation of oil containing unsaturated fatty acids leads to the formation of oxygenated compounds such as hydroperoxides, ozonides, and aldehydes, which reduce bacterial infections in wounds. The ozonation process is widely applied to various types of oil, both for medical, energy and other purposes, such as kapuk oil [9], rice bran oil [10], sunflower oil [11-13], olive oil [14-20], vegetable oil [21-22], palm seed oil [23], waste cooking oil [24] and bulk frying oil [25].

Sumbawa Oil (SO) is one of the local wisdom products of the Sumbawa people in West Nusa Tenggara (NTB) Province. Sumbawa oil is used as medicine or massage therapy to heal injuries such as broken bones, sprains, whiplashes, bruises, back pain, skin diseases and wounds. Sumbawa oil, which is made from various types of Sumbawa mountain plants, contains lauric acid and other fatty acids such as capric acid, palmitic acid, linoleic acid, myristic acid, linolenic acid, which can inhibit the growth of bacteria and fungi in wounds [26]. Sumbawa oil contains polyunsaturated fatty acids (PUFAs). When it reacts with ozone compounds, it will form ozonide compounds, which can stimulate tissue regeneration and repair, as evidenced by decreased iodine levels and increased peroxide levels. Peroxide species are the most important product in the oil ozonation process as Reactive Oxygen Species (ROS) compounds for antibacterial action in wound healing.

Ozone used for medical applications such as wound healing must be produced from pure oxygen through silent electrical discharge/DBD. The ozone generator and delivery system as an oxygen source must have a medical level of purity to avoid nitrogen and other impurities because nitrogen can produce NO, which is toxic to tissues [27]. The Double Dielectric Barrier Discharge (DDBD) Plasma Reactor qualifies as a medical reactor because there is a space between the two barriers for the flow of pure oxygen

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that is not mixed with nitrogen so that when it reacts with an electric field, high-purity ozone is produced.

This research aims to increase the peroxide number in Sumbawa oil through ozonation using a DDBD Plasma reactor. The results of this research can provide additional references for future research related to the ozonation of Sumbawa oil for wound healing.

Research Methods

Materials

The tools and materials used in this research are DDBD plasma reactor, oxygen gas, sodium thiosulfate solution, potassium iodide solution, sodium hydroxide solution and Sumbawa oil. The Sumbawa oil used is Sumbawa oil, which is made traditionally.

Determination of Oxygen Flowrate to Produce Ozone Dose

Flowrate This oxygen is carried out at a flow rate of (0.1-0.8) L/minute with a scale increase of 0.1 L/minute. The ozone concentration, capacity, and dose produced for each oxygen flowrate supplied are calculated. Calculation of ozone concentration using the equation:

$$C_{O_3} = \frac{m_r.V.N}{v.e.t}$$

Where CO_3 is the concentration of ozone gas (ppm), Mr is the relative molecular mass of ozone gas (gram/mol), V is the volume of $Na_2S_2O_3$ solution (mL), N is the concentration of $Na_2S_2O_3$ solution (mol/L), v is the oxygen flowrate (m/s), e is a constant with values two and t is the dissolution time of ozone into the KI solution. Meanwhile, the ozone capacity calculation uses the equation:

$$Cap = C_{O_3}.v$$

Where Cap is the ozone capacity (mg/minute), C_{O_3} is the ozone concentration (ppm), and v is the oxygen flow rate (L/minute). Then, measure the ozone dose using the equation:

$$D = Cap.t$$

Where D is the ozone dose (mg), Cap is the ozone capacity (mg/minute), and t is the time (minutes).

Preparation of Ozonated Sumbawa Oil

Ozonated Sumbawa Oil (OSO) is made by flowing ozone produced from the DDBD plasma reactor with an oxygen flow rate variation of 0.1, 0.4, and 0.7 L/minute and a time variation of 1, 3, and 7 hours. The series of tools for making ozonated Sumbawa oil can be seen in Figure 1 and Figure 2.

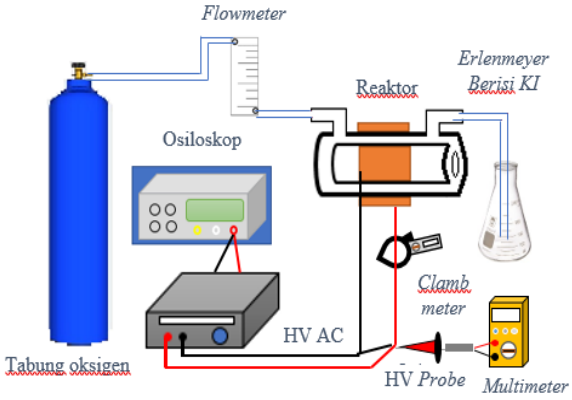

Figure 1. Sumbawa oil treatment scheme

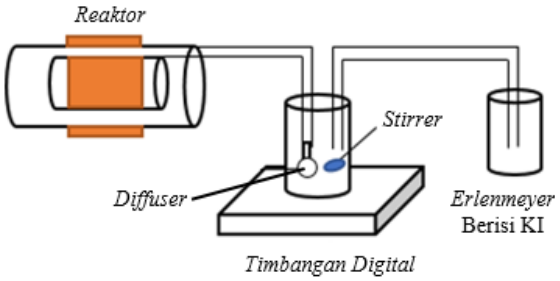

Figure 2. Set up of experimental equipment [31]

Characterization of the physicochemical properties of Ozonated Sumbawa Oil

Peroxide Number

The peroxide number (PV) indicates the total peroxide compound formed. Peroxide value was measured using the iodometric titration method. Determination of peroxide number using the equation:

$$PV = \frac{1000.V.c}{m}$$

Where PV is the peroxide number, V is the volume in mL of $Na_2S_2O_3$ solution used for titration, c is the concentration of $Na_2S_2O_3$, and m is the amount of sample (grams).

Acid Number

The acid number shows the base mass needed to neutralize the free fatty acids in 1 gram of oil. The acid number was measured using a titration method using a NaOH solution in Sumbawa oil added with hot, neutral alcohol and phenolphthalein (PP) indikator.

Viscosity

Viscosity measurement indicates peroxide produced during the ozonation process of Sumbawa oil. Viscosity measurement using a viscometer.

Results and Discussion

Effect of flowrate on ozone concentration

The ozone formation process uses a DBBD plasma reactor through oxygen ionization, recombination, dissociation and association. The ozone produced by the DBD reactor has high purity. The ozone gas produced

between the two electrodes is not contaminated with toxic-free air such as nitrogen oxide (NO). The main gas source is oxygen gas, which is supplied to the DDBD plasma reactor.

The following reaction equation can explain the ozone formation process:

$$e + O_2 \rightarrow O + O$$
$$O + O_2 + M \rightarrow O_3 + M$$

The ozone concentration produced by a DDBD plasma reactor depends on the reactor's voltage, dielectric material, pressure, and system configuration,s introduced into the reactor [28].

Table 1. Oxygen flowrate to produce ozone dose

Flowrate (L/min)	Concentration (ppm)	Capacity (mg/min)	Dosage (mg/ml)
0.1	432.00	43.20	86.40
0.2	288.00	57.60	115.20
0.3	224.00	67.20	134.40
0.4	180.00	72.00	144.00
0.5	124.80	62.40	124.80
0.6	96.00	57.60	115.20
0.7	89.14	62.40	124.80
0.8	78.00	62.40	124.80

In this research, oxygen flowrate optimization was carried out to determine the concentration, capacity and dose of ozone produced, starting from 0.1 L/min to 0.8 L/min with a flowrate increase of 0.1 L/min. The concentration, capacity and dose of ozone produced can be seen in Table 1.

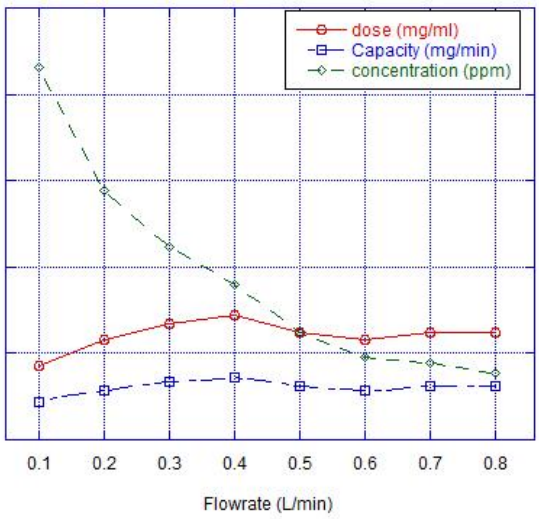

Figure 3. Ozone Concentration, Capacity and Dosage on Variations in Oxygen Flowrate

In Figure 3. It can be seen that the concentration of ozone formed becomes smaller with increasing flow rate. This is because the oxygen residence time in the reactor is getting shorter, so the amount of oxygen ionized, recombined and dissociated is smaller [28], [29], [30], [31]. Meanwhile, the ozone dose obtained increased from a flow rate of 0.1 L/minute to 0.7 L/minute. However, the increase is not linear or exponential. As shown in Table 1, the dose at a flow rate of 0.4 L/min is the largest, namely 144 mg/ml, greater than at a flow rate of 0.7 L/min, namely 124.80 mg/ml.

Based on the optimization results, three flowrates were selected to be applied to the Sumbawa oil ozonation process, namely 0.1 L/min, 0.4 L/min and 0.7 L/min.

Physicochemical properties of ozonated Sumbawa oil

Ozonated oil is formed by bubbles of ozone compounds (O_3) in oil with a portion of oleic acid or unsaturated fatty acids of 65-85%. The ozone reaction with oil often occurs with the double-bonded carbons in unsaturated fatty acids [19]. This interaction forms many oxygenated compounds, such as hydroperoxides, ozonides, aldehydes, peroxides, and poly peroxides, as seen in Figure 4.

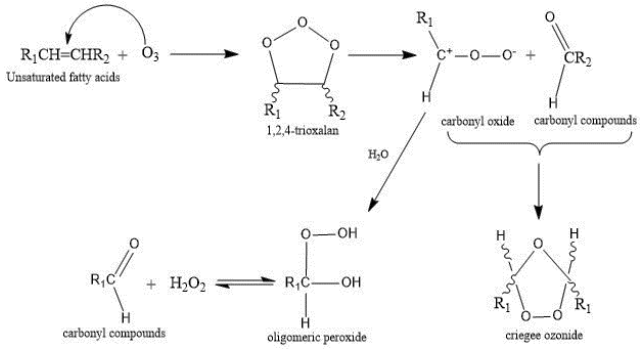

Figure 4. Reaction of unsaturated fatty acids with ozone

Sumbawa oil is coconut oil that is added with various spices. It is made traditionally and is used as medicine, such as massage oil and wound treatment. Sumbawa oil has antibacterial and cytotoxic properties [26,32].

Table 2. Physicochemical properties of ozonated Sumbawa oil

Flowrate (L/Min)	Time (h)	Peroxide Number (mgEq/kg)	Acid Number (mg NaOH/g SO)	Viscosity (cP)
0	0	49.32	0.65	29.6
0.1	5	187.32	1.95	29.23
0.1	3	183.05	1	31.53
0.1	1	164.57	1	33.17
0.4	5	304.12	2.1	31.53
0.4	3	293.30	1.6	29.03
0.4	1	251.92	1	30.67
0.7	5	332.03	3.45	31.23
0.7	3	325.16	1.2	29.47
0.7	1	316.73	1.75	30.83

After being supplied with ozone, Sumbawa oil will experience changes in its physicochemical properties, such as peroxide number, acid number and viscosity. Changes in the values of peroxide number, acid number and viscosity of Sumbawa oil before and after ozonation can be seen in Table 2.

Peroxide Number

The peroxide number indicates the amount of saturated fatty acids that undergo oxidation [12]. The results of measuring the peroxide number of ozonated Sumbawa oil can be seen in Table 2. In Figure 5, it can be seen that the peroxide level of Sumbawa oil before and after ozonation has increased. This increase in peroxide levels indicates that the ozonation process was successful.

The oxygen flow rate and the length of ozonation time influence the increase in peroxide levels in ozonated Sumbawa oil. The greater the oxygen flow rate, the greater the peroxide number and the longer the ozonation process, the greater the peroxide number. The highest peroxide number was obtained at a flow rate of 0.7 L/min for 5 hours, 332.03 mg Eq/kg. Then, a flow rate of 0.4 L/min for 5 hours was followed, and a flow rate of 0.1 L/min for 5 hours, respectively, was 3014.12 mg Eq/kg and 187.32 mg Eq/kg.

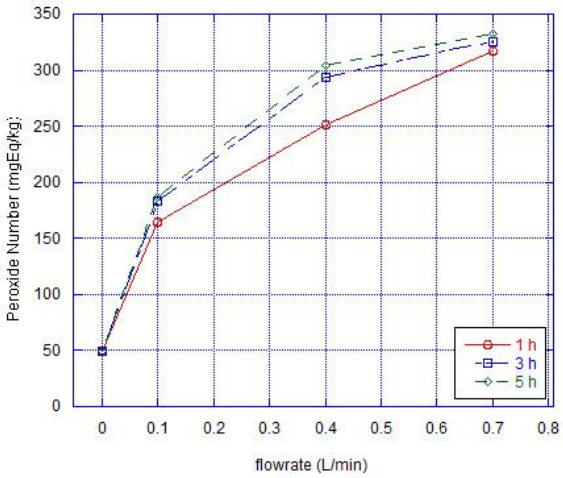

Figure 5. Effect of flow rate on the peroxide number of ozonated Sumbawa oil

From Figure 5, the change in peroxide number from a flow rate of 0.4 L/min to 0.7 L/min is insignificant compared to a flow rate of 0.1 L/min to 0.4 L/min. Even though the peroxide number at the flowrate of 0.7 L/min is the highest, the flowrate of 0.4 L/min has the potential to be applied for the Sumbawa oil ozonation process, because the ozone dose produced is the highest, namely 144 mg/ml, as shown in Table 1. To get maximum results, it is necessary to conduct studies to obtain optimum conditions at a flow rate of 0.4 L/min, such as current strength, voltage and ozonation time.

Acid Number

The acid number is a parameter that shows that the ozonation process was successful. The increase in peroxide number is generally directly proportional to the increase in acid number. The formation of new carbonyl compounds such as aldehydes and trioxalan causes the increase in acid number. Ozonides and aldehydes obtained from the hydrolysis reaction of the product with oxygen increase the medium's acidity [19].

In Figure 6, we can see that there is an increase in the amount of acid in ozoneated Sumbawa oil. Oxygen flow rate and ozonation time also affect the acid number. The highest acid number was obtained at an oxygen flow rate of 0.7 L/min for 5 hours, namely 3.45 mg NaOH/g SO.

Based on the experimental results, the acid number increased with increasing time. The acid number rises during the ozonation process, and this can occur because, in the ozonation process, the decomposition of hydroperoxides forms carboxylic acids as a result of secondary oxidation [15]. Measuring the acid number can assess whether the tested oil is fresh, spoiled, and stored properly. The increase in the acid number indicates that the ozonation process was successful [8].

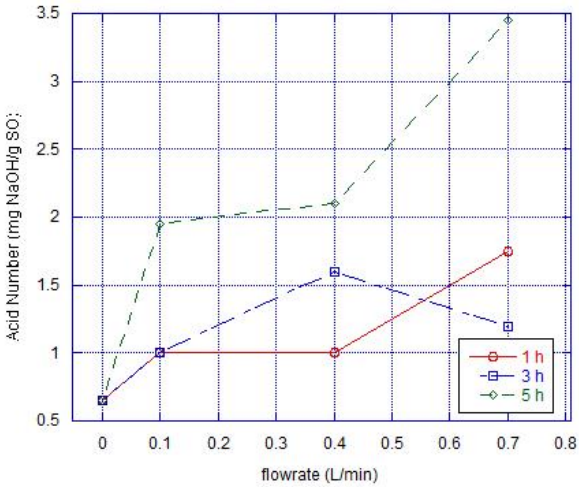

Figure 6. Effect of flowrate on the acid number of ozonated Sumbawa oil

Viscosity

Viscosity measurements of ozonated Sumbawa oil confirm the formation of peroxide compounds. In the ozonation process, ozone will interact with the double carbon bonds and increase the van der Waals bonds, increasing viscosity. The increase in viscosity due to polymerization during the ozonation process produces polymer peroxide [19].

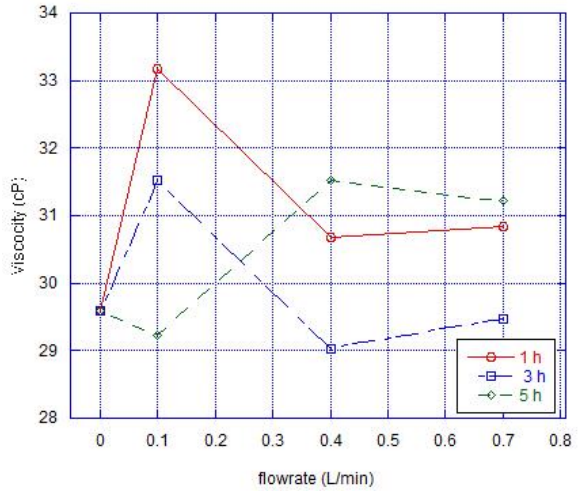

Figure 7. Effect of flow rate on the viscosity of ozonated Sumbawa oil

From Figure 7. we can see an increase in the viscosity of ozonated Sumbawa oil. This corresponds to an increase in the peroxide number.

Conclusion

Based on the research results, it can be concluded that the ozonation process in Sumbawa oil can increase the peroxide number significantly. The increase in peroxide number is greater with increasing oxygen flow rate and ozonation time. The highest peroxide number was obtained at a flow rate of 0.7 L/min for 5 hours, 332.03 mg Eq/kg. The acid number also increased in ozonated Sumbawa oil, namely 3.45 mg NaOH/g SO at a flow rate of 0.7 L/min for 5 hours. The viscosity of ozonated Sumbawa oil also increased compared to before ozonation. This indicates that the ozonation process was successful.

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