Corona Plasma Multireactors for The Removal of Ammonia in Closed House Livestock

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Abstract: We report the use of cold atmospheric plasma (CAP) generated by a corona reactor to remove ammonia in closed livestock housing. An assembly consisting of a reactor, a power supply, and a fan to circulate air into the reactor is called a cold plasma equipment (CPE). The basic physical parameters of the corona reactor such as I-V characteristics and charge mobility were determined and compared with the results of previous research. Ammonia concentration was measured at positions within a distance of 1 m to 5 m (with 1 m intervals) from the CPE placement. At each position of the CPE the ammonia concentration decreases negatively exponentially as a function of time. The best ammonia gas reduction percentage (91.50%) was obtained in 90 minutes at a distance of 1 m from the CPE. These results prove that CAP is effective in reducing ammonia gas in closed cage livestock. CAP has the potential to be an alternative technology for removing odor-causing gases from gas streams.

Keywords: Ammonia gas; cold atmospheric plasma; cold plasma equipment; corona plasma, negative exponential.

1. Introduction

The livestock business is one of the prospective businesses that can meet human food needs, as they provide both meat and eggs. The fulfilment of these needs is pursued through poultry farming, such as broilers and layers. The chicken farming business increase has grown to a large industrial scale because of the high public interest. The increase in the level of chicken consumption worldwide is very fantastic. In 1960, the total chicken consumption was less than 10 million tons, and it increased to 120 million tons in 2021. Thus, the consumption rate has increased by 12 times (1200%). A study conducted by the Aviagen Broiler Breeding Group, predicted that the consumption level will reach 180 million tons in 2050. The human race will, for the first time, consume far more chicken than any other kind of protein [1]. However, lately, chicken farming is considered one of the causes of environmental pollution. The chicken cage produces a pungent odor produced by chicken manure. Additionally, the poultry industry generates emissions of NH3, N2O, and CH4, and impacts global greenhouse gas emissions, as well as animal and human health. Poultry manure can contain residues of pesticides, microorganisms, pathogens, drugs (antibiotics), hormones, metals, and macronutrients. Further, other pollutants that can cause air, soil, and water contamination, and antimicrobial/multidrug -resistant strains are generated [2]. The odor of unpleasant chicken manure from the chicken farm can disturb the environment. The source of this pollution is the nitrogen, and sulphide contained in the chicken manure, which are produced from the decomposition by microorganisms and form ammonia gas, nitrate and nitrite, and sulphide gas. That gas causes the odor. However, ammonia gas is the most dominant gas that causes odor in chicken cages because of the incomplete digestion process or excessive protein in animal feed. Resultantly, not all proteins are absorbed as amino acids, and the excess is released as ammonia in feces. Thus, the ammonia -gas content in the air around the farm is 40-60 parts per million (ppm), which exceeds the minimum threshold of ammonia in the air by 26 ppm [3]. The intensive farming and livestock sector has attracted attention because the amount of NH_3 emissions from these sectors. is increasing. Furthermore, NH_3 is a precursor to forming particles and a source of acidification and eutrophication in ecosystems [4-6]. Research on the use of aluminosilicate as a natural absorbent of ammonia from poultry manure has been carried out. Ammonia uptake by sodium bentonite and zeolite has been confirmed under ex situ conditions. The average reduction for the entire trial period ranged from 26.41% to 29.04% [7]. Relatively high NH_3 concentrations are associated with adverse health effects, including irritation of the eyes, nose, and skin; headaches; asthma; and other respiratory problems. Further, NH3 is toxic to the brain, perturbing the ability of glial cells to remove potassium. The U.S. Occupational Safety and Health Administration and British Health and Safety Executive have set the limit on NH_3 exposure to 25 (ppm) over an 8 h period and 35 ppm over a 15 -min period [8]. The interaction between plasma and ammonia would cause the ammonia to split into other elements, such as NH_4NO_3. The reduction of NH_3 using non-thermal plasma (NTP) methods has been investigated [9]. Two NTP systems are used in this study, one consisting of a multi- cell plate-to-wire reactor (PTW), and the other consisting of an ozonized chamber and the multi-cell PTW reactor [9]. The plasma decomposition of ammonia is studied as a function of applied voltage/power; residence time, including the length of an inner electrode and flow rate of reactant gases; partial pressure of ammonia; and the amount and metal species of the inner electrode [10]. Another investigation on the plasma application for ammonia decomposition was plasma -driven ammonia decomposition on Fe catalyst [11]. Further, another study that strengthens this research is the characterization of the corona plasma multireactors discharge. Electrical studies on corona discharge have been carried out for a long time, for example by Robinson [12]. Corona discharge has been studied in high -density nitrogen and argon gases [13] and in the air by Sumariyah et.al., [14,15]. The formula related to the current–voltage (I–V) characteristics from Robinson's study has been modified by Nur et.al. [16,18]; thus, the formula also applies to corona discharge with cylindrical wire electrodes and dielectric barrier discharge. The cold atmospheric plasma (CAP) generated by corona discharge has been widely employed, particularly for the environment [18,19,20]. Superheated steam has been decomposed using a cylindrical -type plasma reactor and an atmospheric-pressure plasma [21]. Organic dust poses a health threat to livestock. This organic dust. Contains, in addition to the minerals of soil origin, settled dust particles (including feed particles, litter, excrement, and bits of hair or flaky epidermis). Further, they contain microorganisms (bacteria, fungi, and viruses), harmful gases (NH_3, CO_2, and H_2S)), and chemical particles (e.g., from fertilizers, pesticides, or disinfectants) [18]. The CAP has been employed to control the organic dust containing microorganisms and hazardous chemicals [9,10,18]. Herein, the results of the application of corona plasma multireactors for the production of the CAP to remove ammonia in closed -house livestock. are reported. Further, the electrical studies of macroscopic (I – V characteristics) series and parallel corona discharge reactors are reported.

2. Experimental

2.1. Concept of the electrical corona plasma reactors

Electrical corona plasma reactors have been employed to produce CAPs. Using the I-V characteristic, the average mobility of the plasma species present in the reactor has been determined. This comprehensive study will provide answers to some of the existing knowledge gaps. The knowledge gap that will be covered in this research, includes the Robinson formula, which so far applies only to corona discharge. The modified Robinson formula can determine the average mobility of molecular ions. The most prominent study of corona plasma electricity was carried out by Robinson in 1961.Using a symmetric approximation, Robinson published the formulation of the I-V characteristics [12].

$$I_s = \frac{2\mu\varepsilon_0 V^2}{d} \quad (1)$$

where d is the electrode distance in cm, μ is the value of ion mobility (cm^2/Vs), is the permittivity value (8.85 × 10 F⁄m), and V is the operating voltage (volts). This formula can be used for only point-to-plane electrodes. For corona discharge with cylindrical wire electrodes, the formula must be modified.

$$I = \frac{S\mu\varepsilon_t}{2d^3}(V - V_i)^2 \quad (2)$$

where V_i is the corona threshold voltage (volts); S = 2πρl, S is the surface area of the passive electrode ,in cm^2 ; ρ is the cylinder electrode radius in (cm); l is the length of the electrode (cm), and is the permittivity. In this study, the modified formulation will be comprehensively proven by electrical studying. Further, this formula can be used for dielectric barrier discharge. For a discharge with a dielectric material, known as a dielectric barrier plasma discharge, between the two electrodes, the Robinson, is approximated by equation 2 [16].

2.2. Experimental setup overview

Figures 1 and 2 show the experimental setup of this research. This research. e was performed in a closed -house chicken coop at Diponegoro University. The corona plasma reactor was contracted by an electrode in a stainless steel cylinder with a length of 15 cm, and a diameter of 5 cm as the outer electrode. The inner electrode was a wire with a diameter of 3 mm and a length of 15 cm. This study was conducted in two statges: the first is the characterization of the voltage currents generated at a voltage of 1 – 20 kV with a variation of 1 kV. Using the corona discharge reactors, the (CAP) was produced. The ensemble of the reactor, power supply, and fans to circulate air into the reactor has been called cold plasma equipment (CPE) [18]. The CPE utilized a fan at the top, and this fan could suck air from the room at a rate of 5 m^3/min.

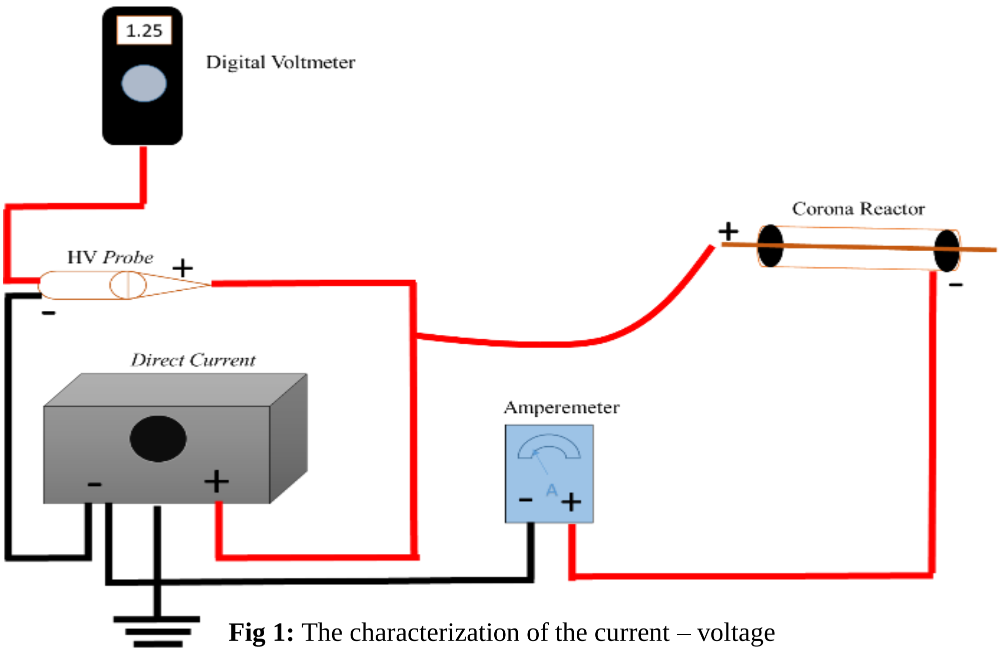


Fig 1: The characterization of the current – voltage

The fan pushed the air that was already contaminated with microorganisms and chemical pollutants into the corona discharge reactor. The cold plasma in the corona discharge converted the dirty air into clean air [20]. Ammonia and other air pollutants that passed through the CAP were cleaned, and the microorganisms died because the polluted air entered the areas containing reactive oxygen species (ROS), reactive nitrogen species (RNS), electrons, ions, and electromagnetic waves [20,23]. After passing through the cold plasma area inside the corona discharge reactor, the air was cleaned and released back into the room through the window at the bottom of the CPE. The concentration of ozone in the room where the CPE was turned on was measured to be 0.04 ppm. This concentration was lower than the standard ozone emission threshold (0.08 ppm) in a contained space with an occupant according to the 2012 US EPA for Health Effects of Ozone in the General Population. The electrical signal from the probe was detected using an Oscilloscope GOS-653, 50 MHz. The electric current that was generated in the reactor was measured using a multimeter (Sunwa TRXn 360) and an ammeter (Kyoritsu, AC/DC Digital Clamp Meter). Direct current (DC) high voltage (DC HV, pulse, up to 20 kV) was used as a power source. A digital voltmeter (Zoyi ZT-Y Avometer Multitester Multimeter HZ) was used for high voltage measurement, after the voltage was scaled down using a voltage divider (HV Probe DC Voltage DC Max 40 kV; 28 kV AC EC code number 1010, En G1010). The second stage is the reduction of ammonia gas, and the best results of characterization will be used in a closed house. The corona reactor, which consists of four reactors, was arranged in the reactor casing; the plasma was ignited for approximately 90 min, and the position of the corona reactor to ammonia ammunition was varied at a distance of 1 – 5 m. The research scheme is as follows.

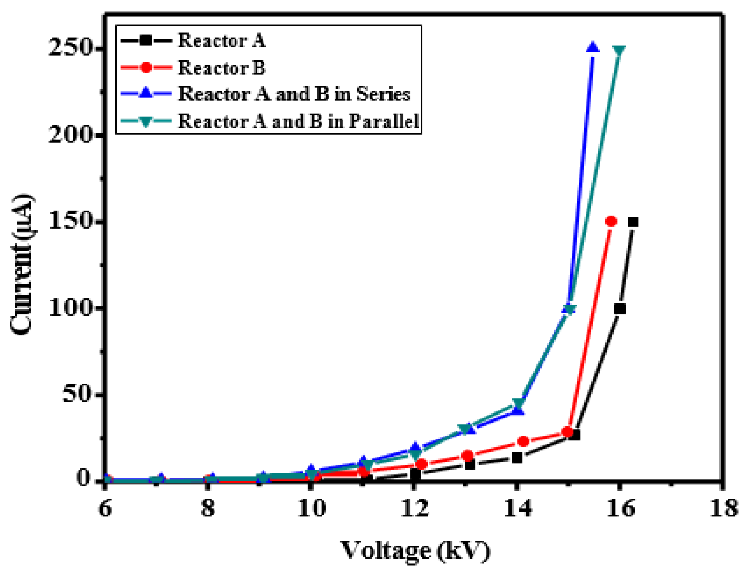
A **B**

Fig 2: The scheme for the reduction of ammonia with 5 measurement positions (A); photo of the close – house livestock coop in Diponegoro University (B)

3. Results and discussion

3.1. The characterization of current–voltage for corona plasma

This study generated corona plasma through a DC high -voltage source at atmospheric pressure. The corona plasma was generated in the space between the cylindrical wire electrodes containing free air, and the analysis of plasma formation was carried out through (I-V) characterization to obtain the optimal plasma generation area, which will be applied to reduce ammonia in a closed house. The characterization will vary the position of the reactor, in series and parallel. The current measurements were carried out in the corona plasma reactor at operating voltages between 6.0 and 16.0 kV. to obtain the I–V characteristics. Figure 3 shows that the characterization of I-–V was carried out in reactors A, reactor B, reactors A and B in series, and reactors A and B in parallel. The graph in Figure 3 shows that the greater the voltage supplied, the greater the current generated. This shows that the DC voltage applied to the corona plasma reactor will increase the amount of electric charge generated during the ionization of the gas molecules, followed by the formation of plasma. The increase in the voltage results in a change in charge because the change in time (capacitive current) produced according to equation ($I = C\frac{dV}{dt}$) will increase. The graph shows that the currents follow a second-order polynomial equation in accordance with the formulation trend (Equation 2) of the modified Robinson [16].



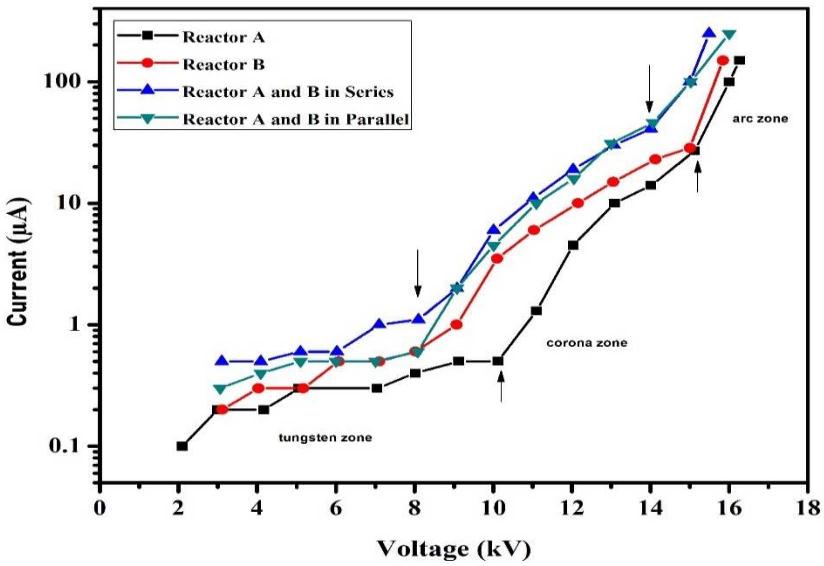


Fig 3: Characterization of the current and voltage in the corona plasma (a) linear scale form (b) semi log form

The emergence of currents in the space between the electrodes was initially due to the movement of charged gas species accelerated to electrodes with different charge properties by non-uniform electric fields; this resulted in collisions between species of gas, which stimulated the process of excitation, de-excitation, ionization, and recombination. The process afforded free electrons that moved toward the electrode, and they were read as currents. This process was referred to as the Townsend discharge. This increase in flow was visually accompanied by a purple glow from the wire electrode, a condition known as the condition of corona incandescent plasma. The increase in current continuously increased the electron and ion densities in the space between electrodes. This was caused by the relatively easy process of excitation, de-excitation, ionization, and recombination. When the electron and ion densities were relatively the same, plasma was formed. The plasma formed was not uniform because it contained electron and ion densities that were centered only on the active electrodes and did not occur in all the spaces between the electrodes [13]. Around the active electrode, the corona discharge has an area where the process of plasma formation occurs. This region is called the active region. The plasma that is formed is (the CAP). When the voltage was increased again above the voltage formed by the discharge of the corona glow, an electrical breakdown occurred. The gas filled the entire space between the electrodes, and it had electrical conductivity properties. This event was followed by a drop in voltage and a drastic increase in the current.

This process was evidenced by a sharp glow emanating from the active electrode. This gas discharge is known as an arc discharge. In reactor A, the arc discharge occurred when the voltage reached 16 kV, and in reactor B, it occurred at 15 kV. When reactors A and B were arranged in series, the arc discharge occurred when the voltage was at 15 kV, and when reactors A and B were arranged parallel, the arc discharge occurred when the voltage reached 15.02 kV. An illustration of a CAP generated by a corona discharge with a wire – cylinder configuration is shown in Figure 4.

The region between the two electrodes outside the active region is known as the remote region. Plasma species are located. in this area. The plasma species are positive ions, negative ions, electrons, photons, radicals, and ion clusters. The chemical pollutants, such as NH_3, will be eliminated. after interaction with these plasma species. Based on the research results of the I – V characteristics, as shown in Figure 3, the (CPE) used for close-house livestock was produced. For the study on the ability of CAP to remove ammonia in livestock, the reactors were arranged in parallel and operated with a DC voltage of 15 kV. The ensemble of the reactor, power supply, and fans to circulate air into the reactor is called the CPE.

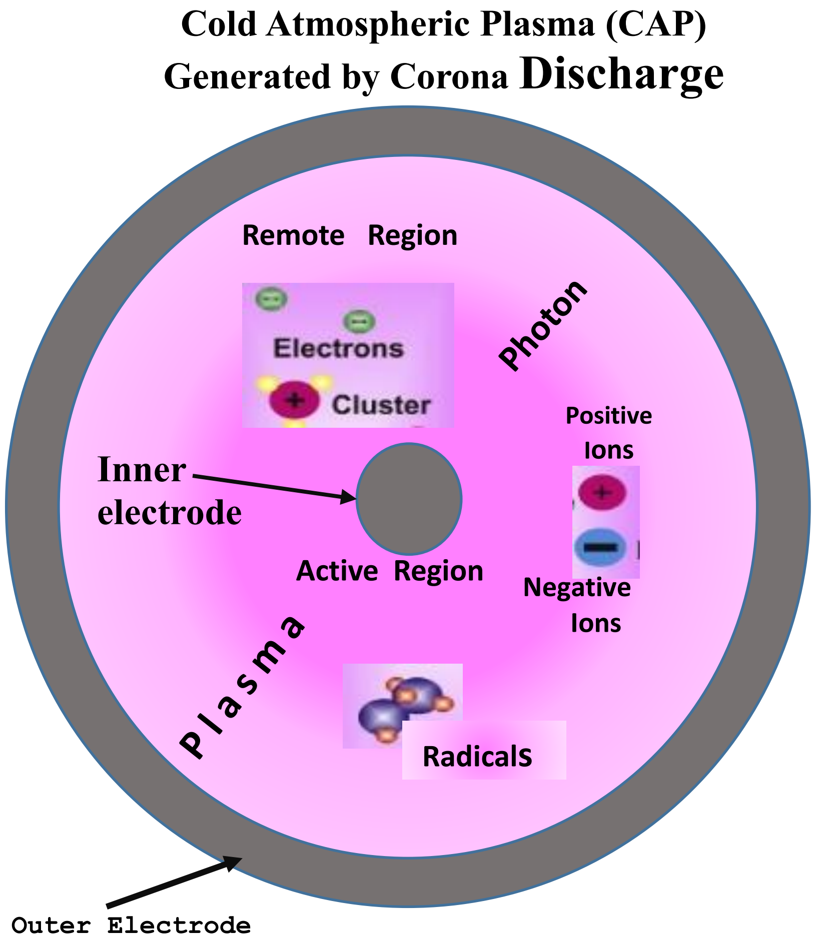


Fig 4: An illustration of a the cold atmospheric plasma generated by a corona discharge with a wire-cylinder configuration

3.2 The Charge Mobility

Based on the characterization of current as a function of reactor voltage A, reactor B; Reactors A and B (in series) and reactors A and B (in parallel) have three plasma discharge regions, namely: Townsend discharge region, corona discharge region, and arc region. In this research, the mobility of charge carriers in the corona discharge region was calculated. This area is the region (I-V) operating cold plasma equipment (CPE) to reduce ammonia emissions in chicken coops. The charge mobility is calculated by determining the gradient value in the current-voltage characterization, obtained by the following linear graph. Equation 2 is then used in calculating the mobility value obtained from the results of characterization of current as a function of voltage via the graph √(I_s) as a function of V (see Fig.5).

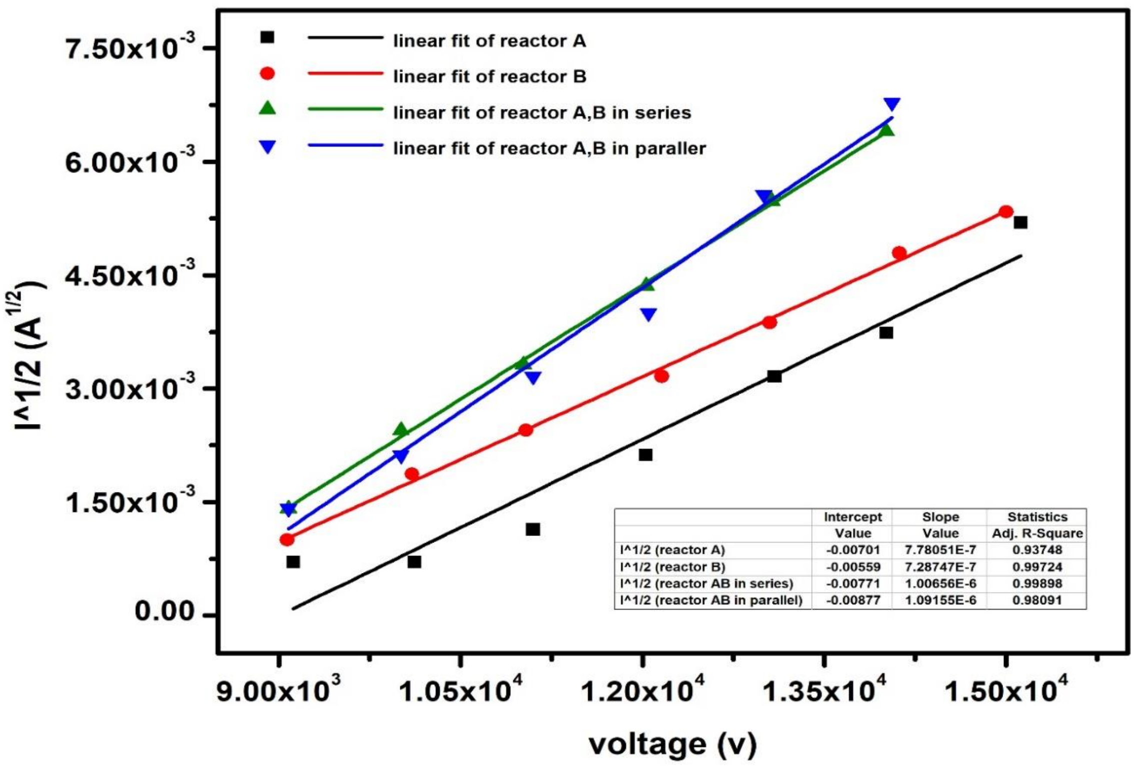


Fig 5: Graph of the square of the electric current as a function of the voltage at the corona reactor

Based on the figure 5, charge mobilities are represented on this figure 6. This figure shows the ion mobility in the corona discharge region for reactor A, reactor B ; reactors A and B (series) also reactors A and (parallel).

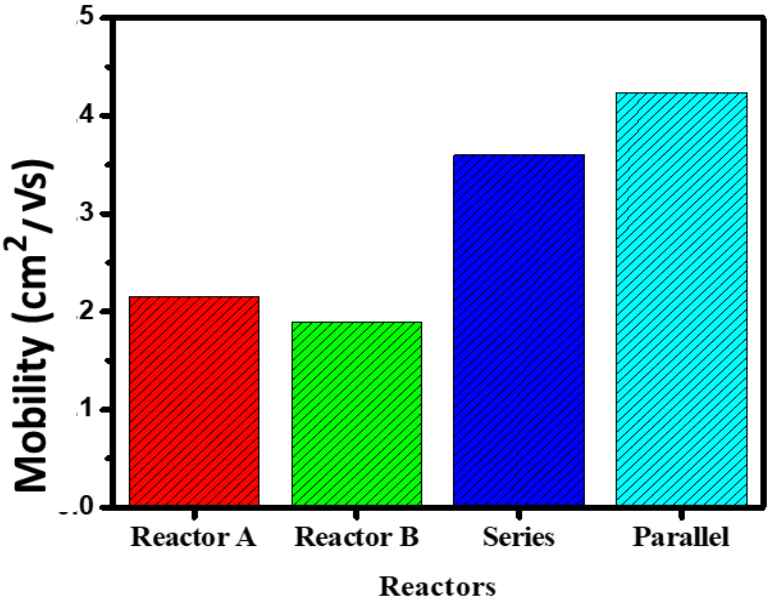


Fig 6: The value of a charge mobility

In this research, air was used as a gas source for cold plasma. The largest content of air is nitrogen, which is more than 78% of the atmospheric volume, oxygen 20% and other gases Ar, He etc. 2%. Due to this, N_2^+ ions will dominate when generating plasma in air, and the ion mobility value obtained from plasma generation in air can be approximated by the mobility value of N_2^+ ions. We found that in reactor A has a charge mobility value of 2.2 cm^2 / Vs. While reactor B has a mobility value of 1.9 cm^2/ Vs. When the two reactors are arranged in series and parallel circuits, parallel reactors A and B have higher charge mobility values than reactors A and B in series which are equal to 4.2 m^2 / Vs, while the reactor in series has a mobility value of 3.6 m^2/ Vs. The results obtained in this study are quite in accordance with the results obtained from the N2+ ion mobility values are in good agreement with measurements of others authors [13,17]. On the other hand if we consider that the charge carriers are ions N_2^+ and its evolved the Langevin model corrected [13], for the effect of charge transfer resonance is applied. The gas source for the formation of cold plasma in a positive corona reactor is air, so ionization occurs around the active electrode with reaction: $e + N_2 \rightarrow N_2 + 2e$. In the plasma, positive nitrogen ions and non-thermal electrons are formed. On the way of electrons to the anode, O_2- ions (oxygen is electronegative) can also occur through electron capture. The mechanism can follow the tree body reaction. : $e + N_2 + O_2 \rightarrow O_2^- + N_2$. Even though O_2– and electrons are formed, the average mobility is contributed dominant by N_2^+[13].

3.2. The impact of distance or reducing ammonia gas

The distance between the (CPE) and ammonia meter varied from 1 to 5 m. The scheme for the reduction of ammonia with 5 measurement positions is shown in Figure 2A. The CPE was turned on for ≥ 90 min. The ammonia concentration when the CPE was turned on at positions 1 to 5, was sequentially 1.53, 1.43, 1.33, 1.26, and 1.23 ppm. The ammonia concentration decreased at a distance of 1-5 m. However, it decreased more at 1 m than at the other positions. as shown in Figure 7. This is because the chain reaction that afforded more gas ammonia molecules was broken down; consequently, the concentration of ammonia gas decreased. The reaction of NH_3 in the reactor is as follows [9].

$$NH_3 + e \rightarrow NH_2 + H + e, \quad (1)$$

$$NH_3 + e \rightarrow NH + H_2 + e, \quad (2)$$

$$NH_3 + O \rightarrow NH_2 + OH, \quad (3)$$

$$NH_3 + OH \rightarrow NH_2 + H2O. \quad (4)$$

In the (CAP) zone where the pollutants from the livestock (close houses) passed, there (RNS) and (ROS). Since the RNS and ROS can generate large numbers of strong oxidizing species (•OH, O3, •NO, •O, NO_2, N_2O, etc.), it has the advantages of a high reaction rate and high efficiency for eliminating pollutants, such as NH_3 [22].

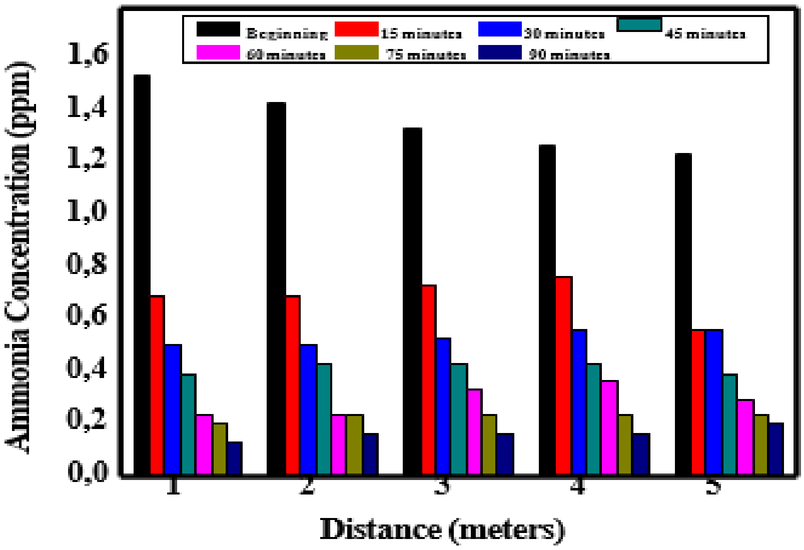


Fig 7: The relationship between the distance and the ammonia concentration after treatment using the CPE for 90 minutes with 15 min interval measurements

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The NH_3 gas molecules entered the reactor, where they first collided with high-energy electrons and active particles (ROS and RNS)), and the N–H bond was broken.

$$2NH_2 + O \rightarrow N_2 + H_2O \quad (5)$$

$$NH_2 + O \rightarrow NO + H_2O \quad (6)$$

$$3NH_2 + 8O \rightarrow 2HNO_3 + 2H_2O \quad (7)$$

$$HNO3 + NH_3 \rightarrow NH_3NO_3 \quad (8)$$

The N–H bond was broken and continued to react with other reactive particles (ROS and RNS) to produce other by-products. The by-products in turn continued to react with strongly oxidizing substances and reactive particles, and the end product was nitrate.

$$NO + O_3 \rightarrow NO_2 + O_2 \quad (9)$$

$$NO + O \rightarrow NO_2 \quad (10)$$

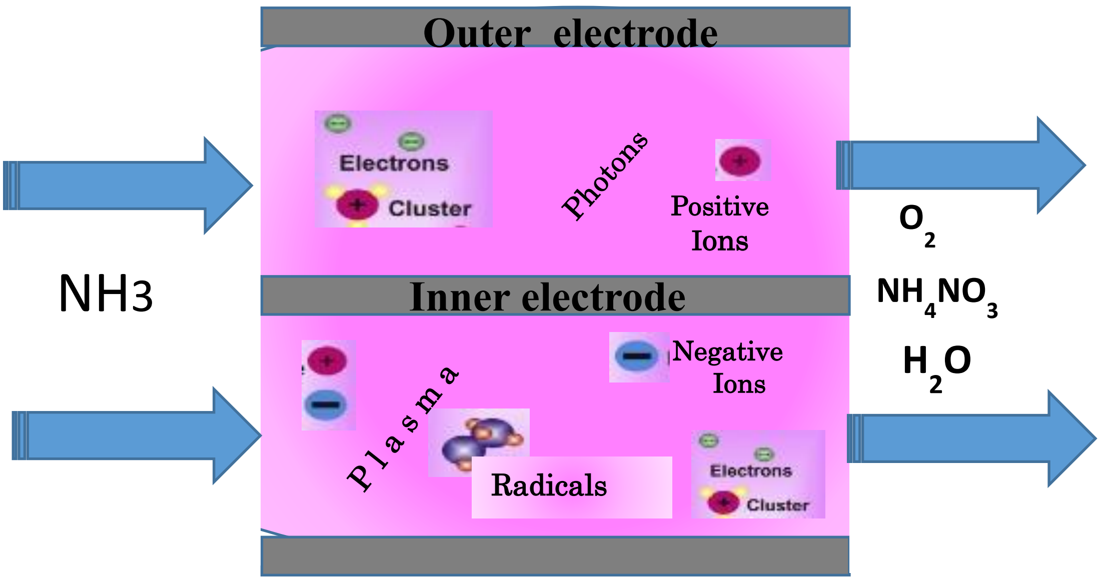
$$NO_2 + OH \rightarrow HNO_3 \quad (11)$$

The corona plasma reactor can produce ROS and RNS; thus, if the ROS reacts with ammonia, it will produce specific molecules as follows.

$$2NH_3 + 4O_3 \rightarrow NH_4NO_2 + H_2O_2 + {}^4O_2 \quad (12)$$

$$NH_4NO_2 + H_2O_2 \rightarrow NH_4NO_3 + H_2O \quad (13)$$

In this case, a more stable compound that does not pollute the environment. can be produced. An overview of (the CAP) with all of its species shows that it can reduce ammonia and produce specific molecules that are more stable and not polluting, as shown in Figure 8.



3.3. The influence of treatment time for the degradation of ammonia gas

In this study, the treatment time for the decrease in ammonia concentration for a distance of 1-5 m was measured. The CAP treatment time was 90 min with an interval of 15 min. This means that every 15 min the amount of ammonia concentration was measured for each distance from the CPE (see Figure 2a). Figure 7 shows the effect of the CPE operating time in a closed house, for the 5 positions where the ammonia meter was placed. The operating time of the CPE significantly affects the removal of ammonia. The longer the CPE operating time, the less NH_3 is retained. Further, the CPE placement distance also affects the ability to remove ammonia. The difference in the removal amount as a function of distance is not very high. At specific treatment times, the value is almost the same. The average of the NH_3 concentrations at the 5 positions of the ammonia meter was obtained. The results of the fitting data are shown in Figure 8.

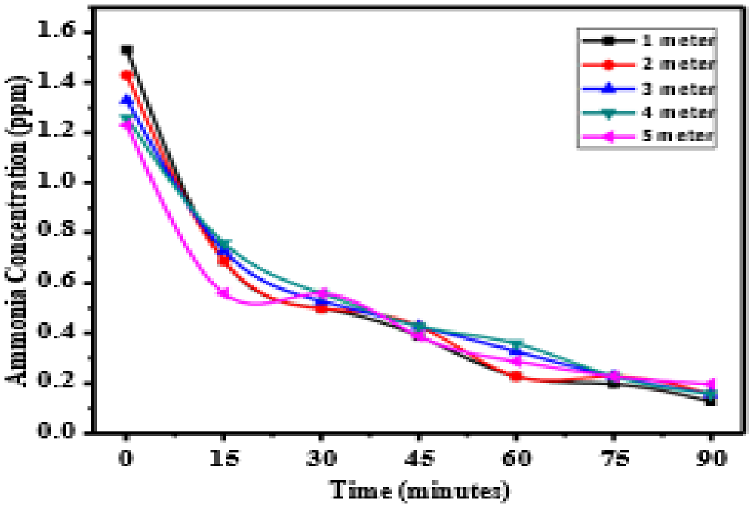


Fig 7: The relationship between ammonia concentration and the operating time of the CPE

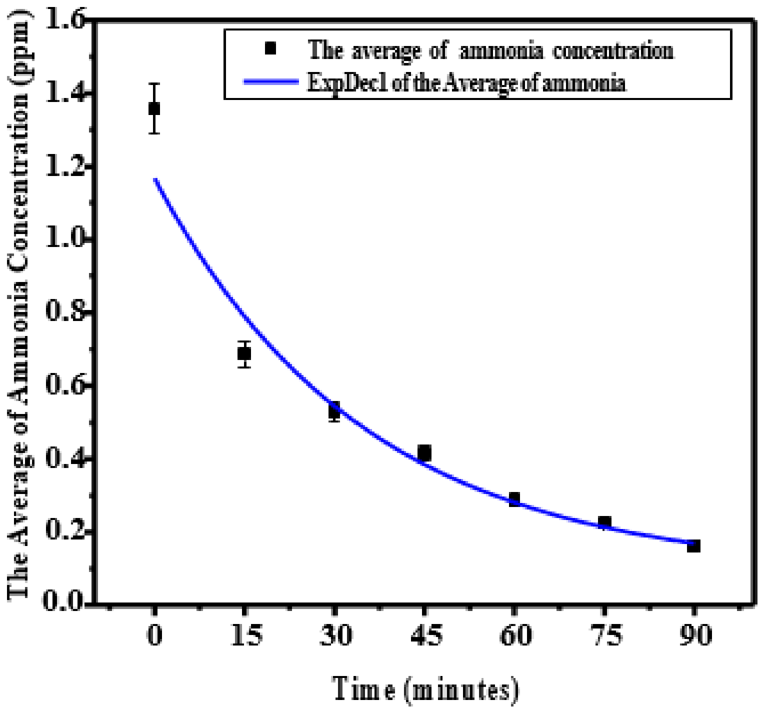


Fig 8: Graph of the average ammonia concentration as a function of the operating time of the CPE

Figure 8 shows the effect of the CPE operating time on the average ammonia concentration. The ammonia concentration decreased with increasing operating time. The concentration of ammonia decreased as a negative exponential function, as shown by the following equation $C = 1.08\, e^{\left(-\frac{t}{35.00}\right)} - 0.09$, where C is the concentration of NH_3, and t is the operating time of the CPE. Based on this equation, we can predict that NH_3 will no longer be found at a specific distance from CPE. All the NH_3 in that distance were removed using the CPE. For a distance of 1 m, NH_3 was completely removed (C = 0) after the CPE was operated for 88.4 min. The graph shown in Figure 9 shows the percentage of ammonia removal for each measurement position as a function of the CPE operating time. The variations in the distance and time significantly influenced the removal of ammonia. The closer the measurement position to the CPE, the more ammonia is removed. Furthermore, the longer the operating time of the CPE, the higher the percentage of ammonia removal. The highest percentage of ammonia concentration reduction occurred at a distance of 1 m with the CPE operating time of 90 min, and it was 91.50 %.

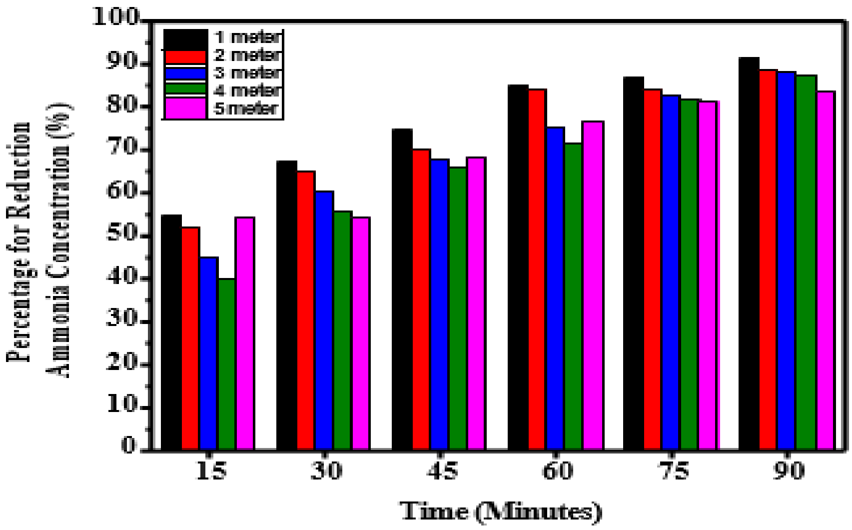


Fig 9: The percentage of the removal of ammonia as a function of the operating time of the CPE

4. Conclusion

In this study, (CAP) has been suggested to remove NH_3 from closed-house livestock. The CAP is produced using a corona discharge. Electrical studies on the corona reactor resulted in a choice of a parallel combination of two reactors with a voltage of 15 kV. It is specifically beneficial for the ensemble of reactors, power supplies, and fans to circulate air into the reactor. This ensemble is called (the CPE), and it is to be installed in a closed-house livestock coop for chickens. Electrical studies show that, the I–V in the corona discharge is directly proportional to the squared value of the voltage (I–V). The removal of ammonia in closed-house livestock directly using CPE was studied. Ammonia concentration measurements were carried out at a distance of 1–5 m (with 1 m intervals) from the CPE placement. The ammonia concentration was decreased following a negative exponential trend as a function of time. The best reduction of ammonia gas (91.50%) was obtained within 90 min at a distance of 1 m from the CPE. These results prove that CAP is effective in reducing ammonia gas in closed – house livestock.

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