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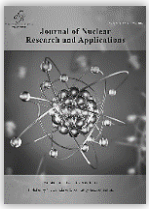

Simulation Investigation of Fluorine Cold Plasma Interaction with Cobalt Oxide as an Etchant for Decontamination Metal Waste

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A B S T R A C T

Fluorine cold plasmas are recognized as effective dry processing techniques for removing surface contaminants with high bond energy. Among the radionuclides, ^{58}Co and ^{60}Co pose significant challenges. The removal of Co isotopes from contaminated metal surfaces is a key focus in developing metal surface cleaning processes. This study aimed to investigate the plasma reaction resulting from the discharge of a dielectric barrier in fluorine gas. This plasma was interacting with a Cobalt oxide film on the surface of stainless steel 304 through experimental observation and simulation study. It was observed that the Cobalt oxide transformed into a powdered form after exposure to plasma irradiation, enabling easy separation from the base metal surface. The efficiency of clearance from the metal surface depends on the generation of fluorine (F) radicals within the plasma. To identify the crucial parameters influencing plasma formation and optimization, the plasma created using CF4 etching gas was simulated using Comsol Multiphysics software. The simulation study reports there are optimal values for voltage, frequency, and the distance between the two electrodes, which enhance cleaning speed. In addition, increasing the temperature will speed up the etching process.

Keywords: Fluorine Cold Plasma, Metal decontamination, Plasma etching, Cobalt oxide, Comsol Multiphysics Software.

1. Introductions

Plasma etching has been suggested as an alternative option for solid nuclear waste removal since the early nineties [1-16,18,22,24-26,28]. Various research groups have reported on its effectiveness, including Martz and colleagues in 1990 [2], who removed Pu using low-pressureradio frequency glow discharge. They found that plasma worked at least 200

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times faster than conventional chemical processes. However, their measurements may not have been precise due to a lack of precision instruments. The data presented in [3] by Tatenuma et al. suggests that chemically reactive plasma treatment is a viable gas-phase decontamination method. This method can be performed using non-toxic gases, ensuring safety and mild conditions such as reduced pressure, shorter treatment durations, and ambient temperature. Other researchers, including Kim et al. [4], have also utilized CF_4/O_2 plasma and demonstrated its ability to convert UO_2 to UF_6 in a two-step process, which facilitates working with radioactive materials. Following their work, Veilleux et al. achieved successful etching of UO2 in 1999 using radio frequency glow discharge plasma, but with the gas switched from CF4 to NF3. They reported their findings [6].

In 2000, Windarto et al. conducted a study that demonstrated, for the first time, the effectiveness of a microwave atmospheric pressure plasma jet in cleaning radioactive materials. They utilized a CF_4-O_2-based microwave discharge to decontaminate radioactive CoO_2 from a stainless steel surface at 1.5 kW [7].

Among other related research, in 2000, researchers from the Hicks group [9] demonstrated that CF_4/O_2 plasma at atmospheric pressure and a frequency of 13.56 MHz, effectively etched Ta instead of Pu. Kim et al. conducted a fluorination reaction of UO_2 in $CF_4/O_2/N_2$ RF plasma under a total gas pressure of 0.3 Torr [10]. They also investigated the decontamination rate of metallic Cobalt surfaces through surface etching with a CF_4-O_2 mixed gas plasma. Their findings revealed that by applying a negative 300 DC bias voltage to the substrate, the reaction could occur even at 290°C, with a rate reaching 0.43 μm/min at 380°C [11].

In another study, Yong-Hwan Kim et al. developed an atmospheric pressure plasma torch source for decontaminating Cobalt-contained oxide layers on metal surfaces [12]. They utilized helium- and argon-baseddischarge with O_2 and CF_4 as reactive gases. The researchers noted that achieving stable plasmas with such reactive gases at atmospheric pressures was challenging, especially without sufficient helium gases in most cases. Furthermore, they observed that the introduction of CF_4 gases to an Argon-air-basedplasma torch, which generates an arc plasma with relatively high gas temperatures, resulted in significant instability. In fact, ^{58}Co and ^{60}Co are the most troublesome radioactive nuclides to be decontaminated in many cases, including spent Inconel steam generator tubes as shown in Fig. 1 [12]. Building upon this research, Yang et al. in 2004 utilized a similar plasma composition (CF_4/O_2/He) to discharge UO_2 from a stainless-steel surface. Their findings [13] led them to conclude that atmospheric pressure plasma holds promise as a technology for transuranic waste decontamination.

Suzuki and his colleagues have also compared different methods of removing radioactive contamination with plasma. They evaluated atmospheric pressure methods as suitable methods for cleaning wide surfaces such as inside the reactor vessel. They formed a DBD with an applied voltage of 6.5 kV and a frequency of 3 kHz with a power of 7W at a pressure of 1 atm. They also used a gas composition of 70% He, 24% CF_4, and 6% O_2 to remove Cobalt oxide pollution [14].

In 2009, Xie et al. conducted experiments using a microwave atmospheric pressure plasma jet at 1000 W. Their findings, published in [19], indicated that an optimal flow of O_2 is necessary to enhance the etching efficiency of the device. The aforementioned studies suggest that a microwave-based atmospheric pressure plasma jet has the potential to serve as an alternative method for radioactive decontamination. This is due to its high chemical conversion rate, cost-effectiveness,

ease of operation, compact size, and low operating cost. In 2018, Karbalai Akbar designed a dielectric discharge barrier plasma generator in Iran to eliminate uranium contamination from surfaces [23]. In 2019, Jaleh Semler and colleagues conducted a study on the decontamination of large surfaces following a radiation accident. Among the methods examined, the plasma method was highly effective, albeit complex [13].

NF_3 gas is an efficient etchant gas in plasma processing, particularly for Cobalt and its oxides, due to its higher fluorine dissociation rates compared to CF4 and SF_6 [18,17]. In a recent study [25], researchers examined the plasma etching reaction of Cobalt oxide film grown on Inconel base metals using NF_3 gas to demonstrate the high decontamination rate of steam generator tubes contaminated with radioactive Co isotopes. They conducted surface analysis to understand the surface reaction mechanism with three experimental parameters: temperature, power, and negative bias. The results showed that the contamination was successfully removed, and this method could be applied in maintaining aging nuclear power plants [25]. In their latest work published in 2022, they studied the NF_3 plasma etching reaction with Cobalt oxide films grown on inorganic compounds using 150 W of RF power. The study showed that plasma surface decontamination is highly effective in removing contaminated nuclides such as Cobalt attached to aggregate in concrete generated during the decommissioning of nuclear power plants [31]. Despite the successes, however literature survey showed that high enough plasma etching rates have not been achieved yet to practically decontaminate Cobalt [12,17-18] and Cobalt oxides [11].

Cobalt removal from metal surfaces can be achieved through chemical reactions occurring in the plasma. This results in the production of volatile compounds or powders that can be easily separated. Fluorine plasma facilitates the formation of detachable powders like CoF_2 and CoF_3 through fluorination. As can be seen from the reviewed articles, the commonly used gases for cold plasmas include fluorine, such as C4F8, CF4, and SF6 gases. These gases are used in plasma generation through electrical discharge. The resulting plasma finds applications in material etching in microelectronics technology and surface property modification [28, 29]. However, halogenated metal-binding compounds possess high boiling points, leading to low volatility. This low volatility is undesirable for the etching process in microelectronics technology [22]. Despite the importance of volatile and reactive products in semiconductor etching, this research focuses on the production of nonvolatile radioactive products. From a treatment view, nonvolatile radioactive waste is better as it can be more easily contained and stored without the risk of releasing harmful gases into the environment. Volatile radioactive waste, on the other hand, requires more specialized treatment processes to prevent hazardous substances release [8]. In this study, among various plasma generators, DBD (dielectric barrier discharge) is selected due to its ease of installation in steam generator tubes and the ability to operate. While the microwave process exhibits fast etching speed, it consumes a lot of energy. On the other hand, the dielectric barrier process requires minimal power and can be considered a preferred choice. Moreover, atmospheric pressure method is selected because it is appropriate for cleaning large surfaces, such as inside reactor vessels, since it is easy to use.

To facilitate the production of fluorine (F) radicals, CF4 gas was selected as the etching gas based on previous reports highlighting its abundant generation in plasma and its effectiveness in the current cleaning process [4,7,10]. Subsequently, a DBD with CF4 etching gas was simulated using Comsol software. This was done to determine optimal parameters in terms of dimensions, dielectric thickness,

source power, and frequency. In addition to simulations, an experimental setup was implemented using available equipment and gas in the laboratory to observe Cobalt removal.

This research indicatesthat plasma surface decontamination is a highly effective and efficient method for removing contaminated nuclides, including Cobalt, from metal surfaces during the decommissioning process of nuclear power plants.

2. Material and methods

2.1 Experimental set up

A Cobalt oxide film was created on stainless steel 304 by starting with the cutting of square pieces from a stainless steel 304 plate. These pieces had dimensions of 3 x 3 cm and 0.5 mm thickness. The samples were then polished using sandpaper with three grain sizes: 320, 600, and 1200 microns. To obtain a mirror-like surface, the samples were cleaned with a 50:50 solution in an ultrasonic cleaner. Finally, the samples were left to air-dry. After drying, a solution of Cobalt nitrate hexahydrate ($Co(NO3)2{\cdot}6H2O$) with a concentration of 300 mg/ml was applied onto the surface of each sample (Fig.1.a). 300 microliters of the solution were poured onto each sample (Fig.1.b). The samples were then placed in an electric oven and baked at a temperature of 500 °C for 2 hours. This process allowed the Cobalt oxide films to form and grow on the surfaces of the stainless-steel base metal (Fig.1.c) [32].

In this study, a plasma etching device is designed and implemented. The experiment setup and the schematic diagram of the device can be seen in Fig.1.d and Fig.1.e respectively. The supply of 300 SCCM of reaction gas to the reaction chamber, with a capacity of 3.4 liters, was received through the MFC (Mass Flow Controller) unit. An electrical discharge was created by employing a 13-liter capsule containing 95% helium gas and 5% fluorine gas. The power required for the discharge between parallel electrodes was provided by a 300W neon transformer. Its output was regulated by an autotransformer. To achieve optimal performance, the power supply output voltage was set at 4.5 kV, with a frequency of 25 kHz. The distance between the dielectric and the sample was maintained at 0.5 cm. The estimation of the reactor's active power can be obtained from the area of the Lissajous figure formed by the V-Q traces, and it is approximately equal to 20 W [26]. The similar characteristics of the applied power supply are discussed in more detail [34].

To ensure safety and prevent fluorine gas activity, the gas capsule was placed in an open area while maintaining a closed reaction path. This arrangement prevented fluorine leakage into the laboratory environment. To evacuate the gas, direct release to the environment was avoided. Instead, the gas was first introduced into a NaOH solution. The fluorine gas combined with the solution to form HF acid, eliminating its release as a dangerous gas into the environment. Despite these precautions, all procedures were performed wearing chemical masks.

2.2 Simulation of designed apparatus

In this study, simulations were conducted on a cold plasma reactor utilizing fluorine gas. Plasma is generated through electrical discharge in CF4 gas. The production of fluorine (F) radicals in the plasma is the primary factor influencing the rate at which radioactive metal is cleared from the contaminated metal surface. The objective was to design a plasma DBD reactor that maximizes reaction rate. The impact of key parameters in the simulation was examined due to the highly sensitive and dangerous nature of working with fluorine gas. This investigation aimed to reduce costs but also to minimize the duration of exposure to fluorine gas. The simulation of the designed system was carried out using COMSOL 5.6 software. For this purpose, a dielectric barrier

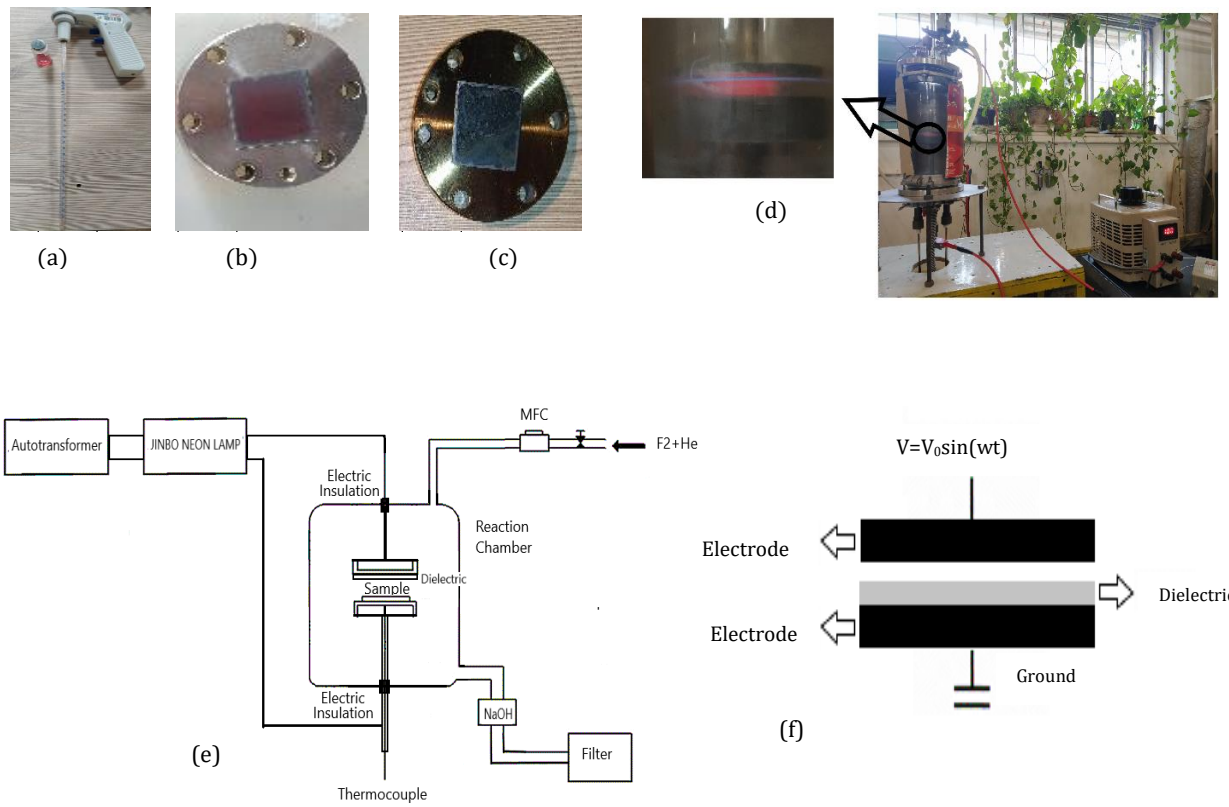

Fig. 1. (a) Image of electric pipette and Cobalt nitrate hexahydrate solution. (b) Image of 300 µl of Cobalt nitrate hexahydrate solution on each sample before being placed in the furnace. (c) Samples after leaving the furnace and Cobalt oxide formation. (d) experiment set up. (e) Schematic of the experiment. (f) DBD simulated geometry.

discharge plasma generator with a circular geometry was simulated, featuring a 10 cm diameter and CF_4 gas (From the cross-section import, Table 1-3 reactions are added to COMSOL). Quartz (SiO_2) was selected as the dielectric material, possessing a relative electrical conductivity of 3.8. It was placed on one of the applied electrodes. The secondary electron coefficient remained constant for both electrodes at 0.0007. The simulation geometry can be observed in Fig.1. f. Triangular grid has also been used to create a balance between the duration of the program and proper review.

For several different geometries, the effect of dielectric thickness, frequency and electrode distance from each other was investigated. The amount of fluorine ion moles was calculated as the main cleaning factor near the ground electrode where the sample is to be placed.

Table 1. Electron reactions used in simulations [30].

Reaction	Formula	Type	∇E(ev)
R1	$e + CF_4 \Longrightarrow CF_3(-) + F$	Attachment	0
R2	$e + CF_4 \Longrightarrow F(-) + CF_3$	Attachment	0
R3	$e + CF_4 \Longrightarrow CF_4^-$	Attachment	0
R4	$e + CF_4 \Longrightarrow e + CF_4^*$	Excitation	0.05
R5	$e + CF_4 \Longrightarrow e + CF_4^*$	Excitation	0.112
R6	$e + CF_4 \Longrightarrow e + CF_4^*$	Excitation	8
R7	$e + CF_4 \Longrightarrow e + CF_4^*$	Excitation	12.5
R8	$e + CF_4 \Longrightarrow e + e + CF_4^+$	Ionization	16.25
R9	$e + F \Longrightarrow e + F$	Elastic	m/M=0.0000288751
R10	$e + F \Longrightarrow e + F$	Excitation	12.818
R11	$e + F \Longrightarrow e + e + F^+$	Ionization	17.687

Table 2. Chemical reactions used in the simulation [19].

Reaction	Formula	Rate ($m^3.s^{-1}$ OR $m^6.s^{-1}\ mol$)
R1	$CF_4 + e \Longrightarrow CF_3 + F + e$	$2 \times 10^{-15}\exp(-13/\mathrm{Te})$
R2	$CF_4 + e \Longrightarrow CF_2 + 2F + e$	$5 \times 10^{-15}\exp(-13/\mathrm{Te})$
R3	$CF_4 + e \Longrightarrow CF + 3F + e$	$4.8 \times 10^{-17}\exp(-20/\mathrm{Te})$
R4	$CF_3 + e \Longrightarrow CF_3 + F + e$	3.3×10^{-16}
R5	$CF_2 + e \Longrightarrow CF + F + e$	3.3×10^{-16}
R6	$CF_4 + e \Longrightarrow CF_3^+ + F + 2e$	$7 \times 10^{-14}\exp(-17.4/\mathrm{Te})$
R7	$CF_4 + e \Longrightarrow CF_2^+ + 2F + 2e$	$7 \times 10^{-15}\exp(-24.7/\mathrm{Te})$
R8	$CF_4 + e \Longrightarrow CF^+ + 3F + 2e$	$9 \times 10^{-15}\exp(-30/\mathrm{Te})$
R9	$CF_3 + e \Longrightarrow CF_3^+ + 2e$	$8 \times 10^{-15}\exp(-12.2/\mathrm{Te})$
R10	$CF_2 + e \Longrightarrow CF_2^+ + 2e$	$2.5 \times 10^{-14}\exp(-12.2/\mathrm{Te})$
R11	$CF + E \Longrightarrow CF^+ + 2e$	$2.5 \times 10^{-14}\exp(-15.3/\mathrm{Te})$
R12	$CF_4 + e \Longrightarrow F^- + CF_3$	$4.6 \times 10^{-15}\mathrm{Te}^{-1.5}\exp(-7/\mathrm{Te})$
R13	$CF_3^+ + e \Longrightarrow CF_3$	4×10^{-14}
R15	$CF^+ + F^- \Longrightarrow CF + F$	4×10^{-13}
R16	$CF_2^+ + F^- \Longrightarrow CF_2 + F$	4×10^{-13}
R17	$CF_3^+ + F^- \Longrightarrow CF_3 + F$	4×10^{-13}
R18	$CF_3^+ + F^- \Longrightarrow CF_4$	5×10^{-14}
R19	$CF_3 + F^- \Longrightarrow CF_4 + e$	5×10^{-16}

Table 3. Surface reactions in the simulated design [33].

Reaction	Formula	Sticking coefficient
R1	$CF_4^+ \Longrightarrow CF_4$	1
R2	$CF_4S \Longrightarrow CF_4$	1
R3	$CF_3^- \Longrightarrow CF_3$	1
R4	$CF_4^- \Longrightarrow CF_4$	1
R5	$F^- \Longrightarrow F$	1
R6	$F \Longrightarrow F$	1
R7	$CF_3 \Longrightarrow CF_3$	1

3. Result and discussion

Experiment: It was anticipated that plasma irradiation would cause the Cobalt oxide layer to be pulverized and detached from the surface of the sample. This is precisely what occurred and can be observed with the naked eye. Optical microscope images of the sample surface, before Cobalt is oxidized on the sample surface, after Cobalt oxidation on the sample surface, and after plasma irradiation are shown in Fig.2. The resulting powder can be effortlessly gathered from the surface using a basic vacuum surface sweeper. EDX analysis has been done on the surface of a sample before and after the test. Our results conclude that Co Atomic% reduce from 22.21% to %1.16.

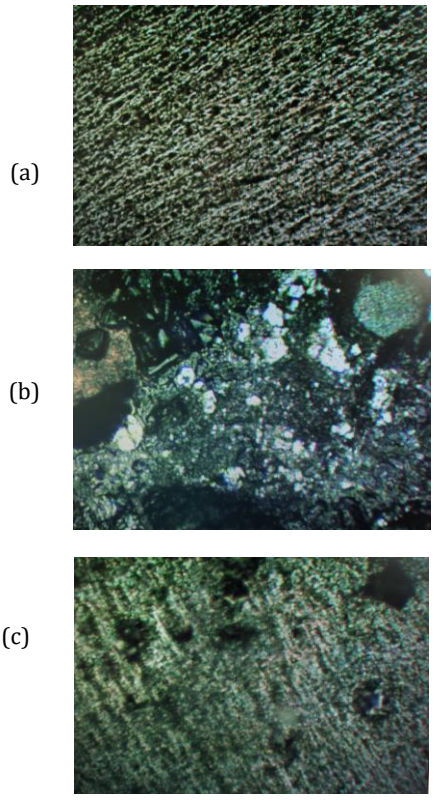

Fig. 2. Optical microscope images of the sample surface. (a) Before Cobalt is oxidized on the sample surface. (b) After Cobalt oxidation on the sample surface. (c) After plasma irradiation with DBD plasma resulting from a mixture of fluorine and helium gas with an input gas flow of 300 SCCM, at a voltage of 4.5 kV, a frequency of 25 kHz and 5 min duration.

Simulation: For the simulation of the designed apparatus to create an electric field between the electrodes, it is necessary to connect them to a voltage source. Increasing the voltage results in a higher electric field, which significantly affects the discharge process. To investigate the voltage effect, the temperature was fixed at 600 Kelvin, the frequency at 60 kHz, the distance between the electrode and dielectric at 2 cm, and the dielectric thickness at 1 mm. The voltage was then varied within the range of 400-1000 V. The results are depicted in Fig. 3.a. According to the diagram, the quantity of fluorine ion moles showed a certain degree of increase as the voltage increased to 700 V. This can be attributed to the enhanced ionization resulting from a stronger electric field, leading to the production of a higher number of fluorine ions. Similarly, the intensified electric field, resulting from the increased voltage, raised the temperature of electrons and facilitated the recombination of electrons and ions. However, when the voltage exceeded a certain threshold (700 V), the atom required electron loss from its second and third layers to further enhance ionization. Consequently, a stronger electric field was necessary. As the voltage was not sufficiently high, the recombination rate increased, causing a decrease in fluorine ions quantity [6]. The applied voltage in this experiment follows a sinusoidal waveform and can have various frequencies. The purpose is to examine how frequency affects fluorine ion moles. Additionally, several other parameters were considered. These include a temperature of 600 degrees Kelvin, a voltage of 600 volts, a 2 cm distance between the electrode and the dielectric, and a dielectric thickness of 1 mm. Next, the impact of frequency was studied within the range of 30-80 kHz. The findings of the simulation are displayed in Figure 3.b. The results indicate that frequency plays a significant role alongside voltage. Specifically, increasing the frequency leads to a higher rate of voltage direction changes between the electrodes. Consequently, fewer electrons reach the positive electrode. This causes a greater number of electrons to become trapped in the plasma, leading to an increased electron density within the plasma. The ionization of fluorine atoms is consequently enhanced. However, once the frequency surpasses 70 kHz, electrons no longer have the opportunity to move. Consequently, the current decreases significantly, resulting in a sharp decline in ionization.

When the gap contains a dielectric material, plasma formation results in the low mobility of electric charges on the dielectric surface. Consequently, these charges accumulate on the dielectric surface, generating a field that opposes the main field and effectively reducing its strength. The strength of this opposing field increases with the dielectric thickness. To study the influence of dielectric thickness within a specific range of 0.5-10 mm, a simulation was conducted at a temperature of 600 degrees Kelvin. This simulation was conducted with a voltage of 600 volts, an electrode-to-dielectric gap of 2 cm, and a frequency of 60 kHz. The findings are depicted in Fig. 3.c. The results revealed that as the thickness decreases, the number of fluorine ion moles increases. This relationship can be observed through the dielectric loss coefficient. However, it should be noted that reducing the thickness compromises the material's strength and may result in dielectric breakdown.

Additionally, another simulation was conducted at a temperature of 600 degrees Kelvin. This simulation had a voltage of 600 volts, a dielectric thickness of 1 mm, and a frequency of 60 kHz. This time, the investigation focused on the gap space between the electrode and the dielectric, ranging from 0.5 to 4 cm. Fig. 4.d. illustrates the results. The findings demonstrate that there exists an optimal distance between the electrodes, which leads to increased cleaning speed. Increasing this distance beyond the optimal value diminishes

the field, potentially leading to plasma shutdown. Conversely, decreasing the distance reduces the volume of available gas, resulting in a decrease in the number of fluorine ion moles.

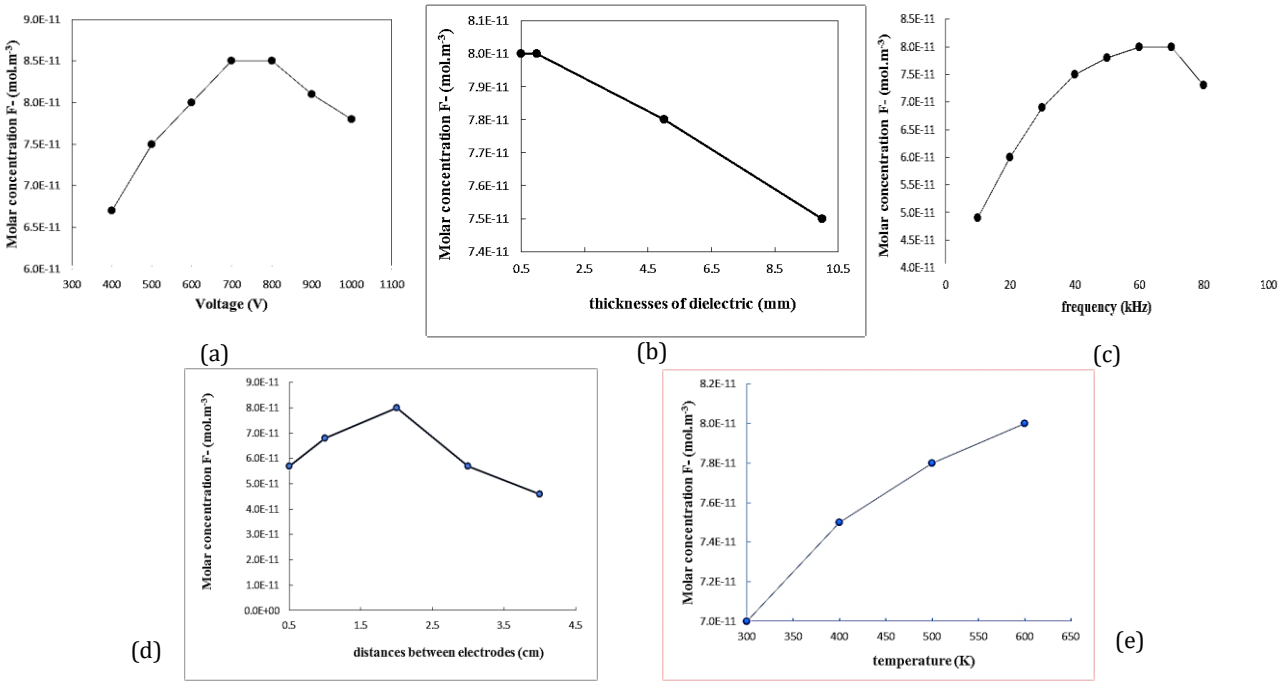

Fig. 3. (a) Diagram of the amount of moles of fluorine ion in terms of voltage. (b) Diagram of the amount of moles of fluorine ion in relation to frequency. (c) Diagram of the amount of moles of fluorine ion according to dielectric thickness. (d) Diagram of the amount of moles of fluorine ion according to the distance between two electrodes. (e) The diagram of the amount of moles of fluorine ion relating to temperature.

At a voltage of 600 volts, a dielectric thickness of 1 mm, a distance between two electrodes of 2 cm, and a frequency of 60 kHz, the temperature in the range of 300-600 degrees Kelvin has been investigated. The results are shown in Fig. 4.e.

As anticipated, it was observed that an increase in temperature led to a corresponding increase in the quantity of fluorine ion moles. This can be attributed to the rise in electron energy within the plasma as the temperature increases. Essentially, elevating the electron temperature promotes higher collision rates and consequently enhances plasma ionization [25, 27]. To verify the accuracy of the simulator code, we simulated the test set-up using reference voltage and frequency characteristics described in ref [14], along with the temperatures presented in figure 3 of that reference. Our simulation showed a similar behavior of the change of fluorination rate as observed in the experimental results presented in figure 3 (ref [14]), where the fluorination rate increases with increasing temperature. Fig.4 illustrates the simulated result.

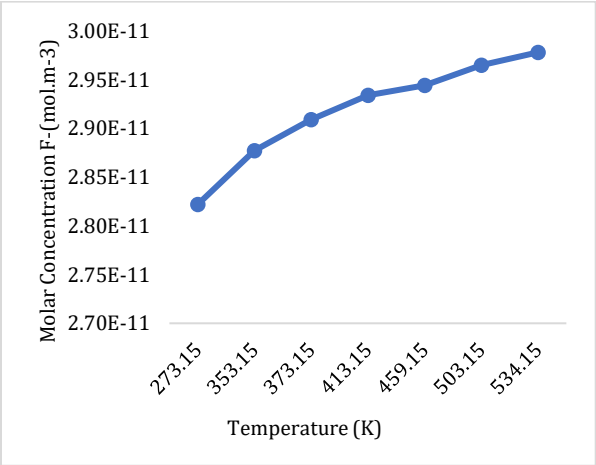

Fig. 4. result of moles of fluorine ion according to temperature simulation for DBD by characters indicated in ref [14] (6.5 kV and 3kHz).

4. Conclusions

The use of reactive plasma gas for cleaning or etching metal surfaces is an emerging dry processing technique aimed at effectively removing surface contaminants with strong bonding, particularly for cleaning used parts and equipment [3, 4, 7, 10, 11]. This method minimizes secondary waste generation while achieving similar levels of efficiency as wet cleaning techniques [11].

The final goal of this research was to design a suitable plasma apparatus for removing cobalt pollution from wide metal surfaces. For this reason, by studying various references and to reduce the cost, the DBD structure has been considered at atmospheric pressure. Then, in order to conduct an experimental feasibility study, an initial layout has been implemented. Samples of cobalt oxide were prepared and exposed to plasma to observe oxide removal from the surface. A mixture of 95% helium and 5% fluorine was used. The experimental results demonstrated that the Cobalt oxide transformed into a powder upon exposure to plasma irradiation. This made it easily detachable from the metal surface. The optimal conditions for generating plasma through the dielectric barrier discharge (DBD) were atmospheric pressure, a voltage of 4.5 kV, and a frequency of 25 kHz.

Using the Comsol simulator software, the optimal conditions and effective parameters were identified assuming the use of suitable gas (CF4). Based on the simulation results there are optimal values for voltage, frequency, and the distance between the two electrodes, which enhance the cleaning speed.

Furthermore, it was indicated by the obtained results that an increase in temperature can moderately enhance the number of moles of fluorine ions near the dielectric surface. This leads to a higher decontamination rate.

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