# Investigation of Fluorine Cold Plasma Interaction with Cobalt Oxide as an etchant for decontamination metal waste

Askari M.[1], Darestani Farahani N. Correspondent[2*], Bakhshzad Mahmoudi M.[2]

[1] *Radiation Application Department, Shahid Beheshti University, P.O. Box: 1983963113, Tehran, Iran*

[2] *Plasma and Nuclear Fusion Research School, Nuclear Science and Technology Research Institute, B. O. Box: 14399-51113, Tehran, Iran*



## Abstract

Fluorine cold plasmas are being 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, and 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 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 depended on the generation of fluorine (F) radicals within the plasma. To identify the crucial parameters influencing the 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 the cleaning speed. Also increasing the temperature will speed etching rate.

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

## INTRODUCTION

Plasma etching has been suggested as an alternative option for the removal of solid nuclear waste 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 pressure radio frequency glow discharge. They found that plasma worked at least 200 times faster than conventional chemical processes, although 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. in 1999 achieved successful etching of $UO_2$ using radio frequency glow discharge plasma, but with the gas switched from $CF_4$ to $NF_3$. 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 researches, 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-based discharge and Argon-based discharge 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 based plasma 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 and evaluated atmospheric pressure methods as suitable method for cleaning wide surfaces such as inside the reactor vessel. He from 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. A gas composition of 70% He, 24% CF4 and 6% O2 has been used 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 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 identified as highly effective, albeit complex [13].

$NF_3$ gas has been found to be 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].

The removal of Cobalt from metal surfaces can be achieved through chemical reactions occurring in the plasma, resulting 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 it can be seen from reviewed articles, the commonly used gases for cold plasmas include fluorine, are such as C4F8, CF4 and SF6 gases. These gases are utilized in the generation of plasma through electrical discharge. The resulting plasma finds applications in material etching within 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 the release of hazardous substances [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 utilize. While the microwave process exhibits fast etching speed, it consumes a significant amount of energy. On the other hand, the dielectric barrier process requires minimal

power and can be considered a preferred choice. Also atmospheric pressure method is selected because it is suitable for cleaning large surfaces like inside reactor vessels.

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 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.

The findings of this research indicate that 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.

## MATERIAL AND METHODES

### a. 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 a thickness of 0.5 mm. The samples were then polished using sandpaper with three different 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 a duration of 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 set up 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, and 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 active power of the reactor can be obtained from the area of the Lissajous figure formed by the V-Q traces, and it is approximately equal to 20W [26]. The similar characteristics of the applied power supply are discussed in more detail in reference [ 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 any 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, where 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 while wearing chemical masks.

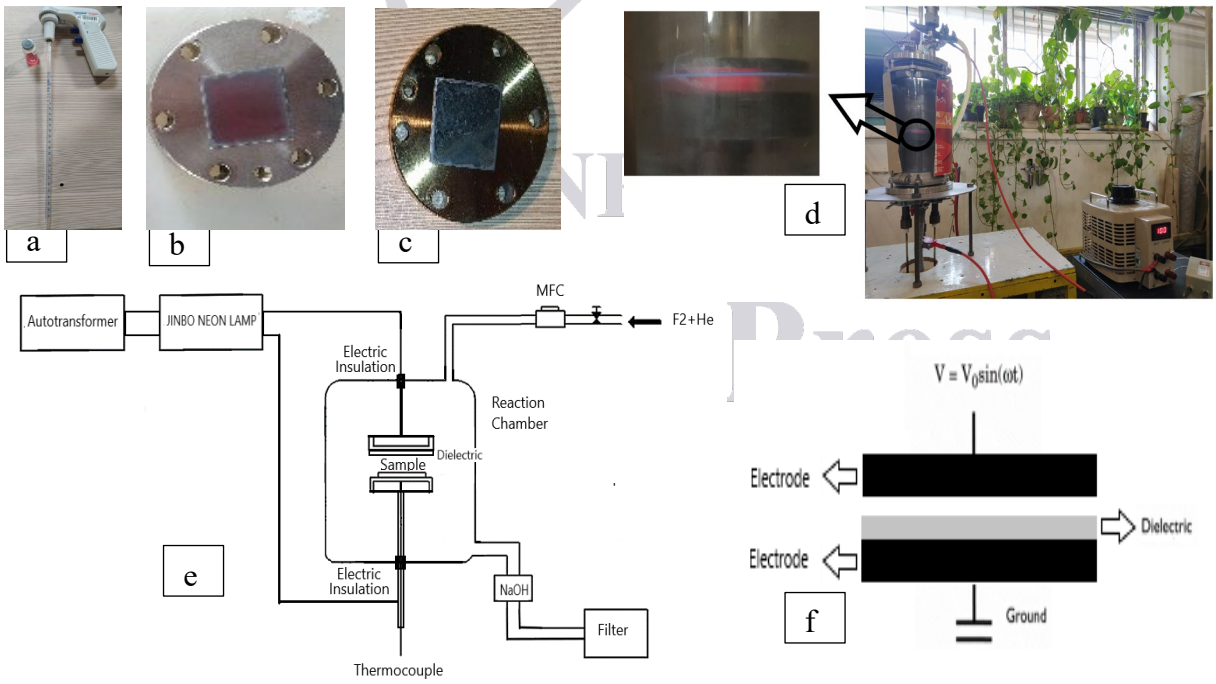


**Fig 1.** a) Image of electric pipette and Cobalt nitrate hexahydrate solution. b) Image of 300 µl of Cobalt nitrate hexahydrate solution on each sample before being placed in the furnace. c) Samples after leaving the furnace and Cobalt oxide formation. d) experiment set up. e) Schematic of the experiment.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

### b. Simulation of designed apparatuse

In this study, simulations were conducted on a cold plasma reactor utilizing fluorine gas. The plasma is generated through an 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 the 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. The aim of this investigation was not only 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 discharge plasma generator with a circular geometry was simulated, featuring a 10 cm diameter and $CF_4$ gas (From the cross section import, the reactions of Table 1-3 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 a value of 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 and 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/Te) |
| R2 | $CF_4 + e \Longrightarrow CF_2 + 2F + e$ | $5 \times 10^{-15}$exp(−13/Te) |
| R3 | $CF_4 + e \Longrightarrow CF + 3F + e$ | $4.8 \times 10^{-17}$exp(−20/Te) |
| R4 | $CF_3 + e \Longrightarrow CF_3 + F + e$ | $3.3 \times 10^{-16}$ |
| R5 | $CF_2 + e \Longrightarrow CF + F + e$ | $3.3 \times 10^{-16}$ |
| R6 | $CF_4 + e \Longrightarrow CF_3^+ + F + 2e$ | $7 \times 10^{-14}$exp(−17.4/Te) |
| R7 | $CF_4 + e \Longrightarrow CF_2^+ + 2F + 2e$ | $7 \times 10^{-15}$ exp(−24.7/Te) |
| R8 | $CF_4 + e \Longrightarrow CF^+ + 3F + 2e$ | $9 \times 10^{-15}$exp(−30/Te) |
| R9 | $CF_3 + e \Longrightarrow CF_3^+ + 2e$ | $8 \times 10^{-15}$exp(−12.2/Te) |
| R10 | $CF_2 + e \Longrightarrow CF_2^+ + 2e$ | $2.5 \times 10^{-14}$ exp(−12.2/Te) |
| R11 | $CF + E \Longrightarrow CF^+ + 2e$ | $2.5 \times 10^{-14}$exp(−15.3/Te) |
| R12 | $CF_4 + e \Longrightarrow F^- + CF_3$ | $4.6 \times 10^{-15}$Te$^{-1.5}$exp(−7/Te) |
| R13 | $CF_3^+ + e \Longrightarrow CF_3$ | $4 \times 10^{-14}$ |
| R15 | $CF^+ + F^- \Longrightarrow CF + F$ | $4 \times 10^{-13}$ |
| R16 | $CF_2^+ + F^- \Longrightarrow CF_2 + F$ | $4 \times 10^{-13}$ |
| R17 | $CF_3^+ + F^- \Longrightarrow CF_3 + F$ | $4 \times 10^{-13}$ |
| R18 | $CF_3^+ + F^- \Longrightarrow CF_4$ | $5 \times 10^{-14}$ |
| R19 | $CF_3 + F^- \Longrightarrow CF_4 + e$ | $5 \times 10^{-16}$ |



**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 |

RESULTS 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 is 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. it is concluded from results that Co Atomic% reduce from 22.21% to %1.16.

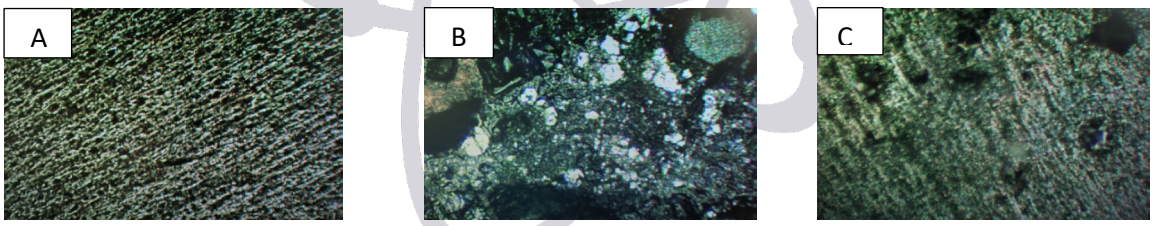


**Fig. 2.** Optical microscope images of the sample surface. A: Before Cobalt is oxidized on the sample surface. B: After Cobalt oxidation on the sample surface. C: After plasma irradiation with DBD plasma resulting from a mixture of fluorine and helium gas with an input gas flow of 300 SCCM, at a voltage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

**Simulation:** For the simulation of the designed apparatuse, 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 obtained from the simulation 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 much stronger electric field was necessary. As the voltage was not sufficiently high, the recombination rate increased, causing a decrease in the quantity of fluorine ions [6].

The applied voltage in this experiment follows a sinusoidal waveform and can have various frequencies. The purpose is to examine how frequency affects the number of fluorine ion moles. Additionally, several other parameters were taken into account. 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, and 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 are able to 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. As a result, the ionization of fluorine atoms is 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, the formation of plasma results in a 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 reduces its strength. The strength of this opposing field increases with the thickness of the dielectric. 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, 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 moles of fluorine ion increases. This relationship can be observed through the dielectric loss coefficient. However, it should be noted that reducing the thickness compromises the strength of the material and may result in dielectric breakdown.

Additionally, another simulation was conducted at a temperature of 600 degrees Kelvin, with 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. The results are illustrated in Fig. 4.d. The findings demonstrate that there exists an optimal distance between the electrodes, which leads to an 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.

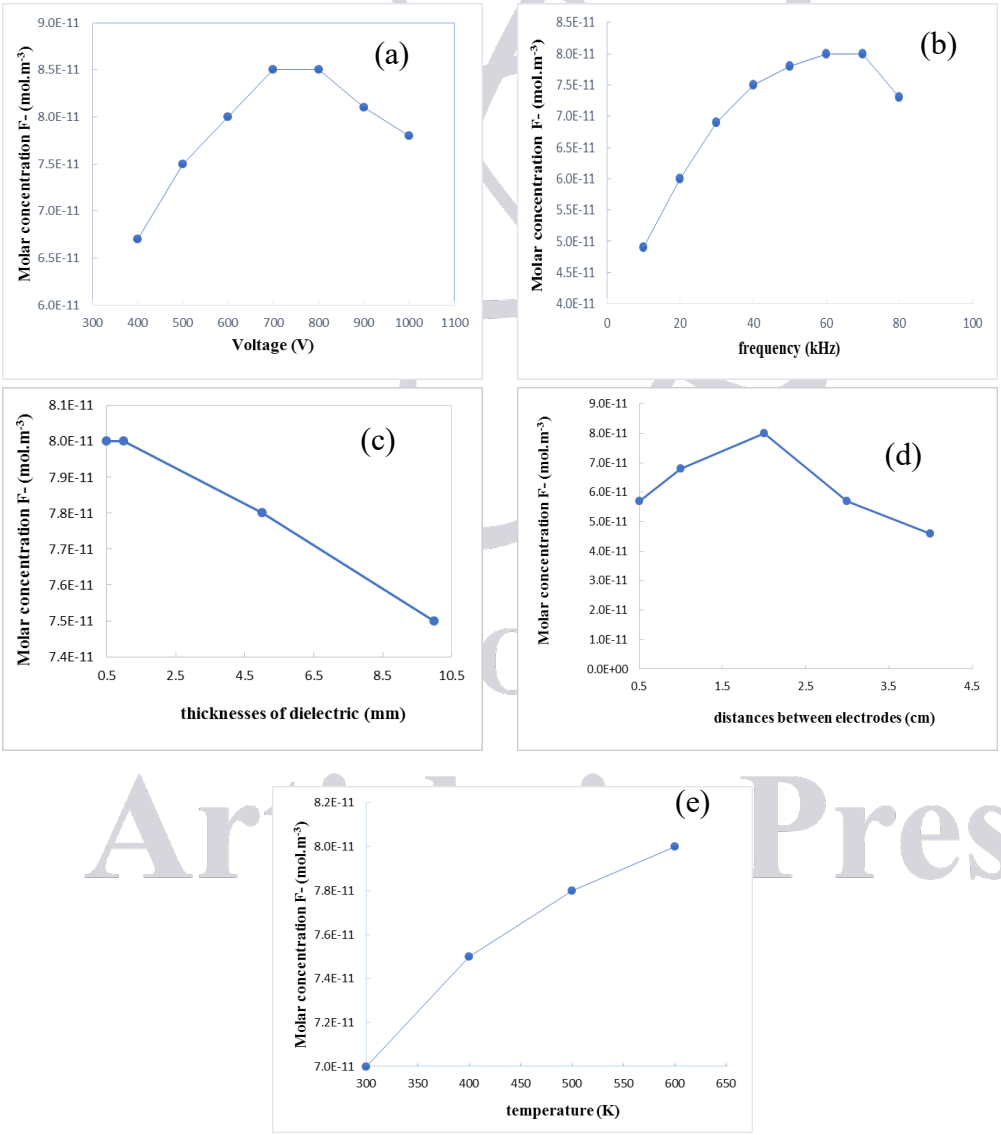


**Fig. 3. a)** Diagram of the amount of moles of fluorine ion in terms of voltage. b) Diagram of the amount of moles of fluorine ion according to frequency. c) Diagram of the amount of moles of fluorine ion according to dielectric thickness. d) Diagram of the amount of moles of fluorine

ion according to the distance between two electrodes. e) The diagram of the amount of moles of fluorine ion according 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, and 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 temperature of the electrons 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. The simulated result is illustrated in Fig.4.

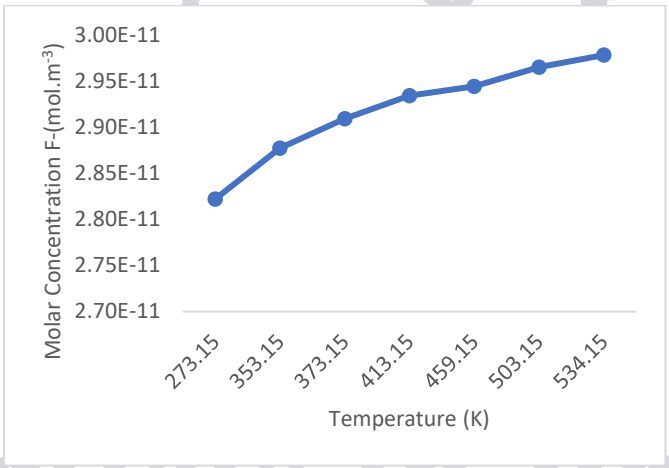




**Fig.4.** result of moles of fluorine ion according to temperature simulation for DBD by characters indicated in ref [14] (6.5 kV and 3kHz).

## 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 has the advantage of minimizing 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 the removal of oxide 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, making it easily detachable from the metal surface. The optimal conditions for generating the plasma through the dielectric barrier discharge (DBD) were determined to be 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 ion near the dielectric surface, leading to a higher decontamination rate



ACKNOWLEDGMENTS



The work presented in this study was conducted within the Plasma Processing Laboratory of the Plasma and Nuclear Fusion Research School. The authors express their gratitude to Dr. Amir Charkhi from the Nuclear Science and Technology Research Institute in Iran for supplying the F2-He mixture and offering valuable guidance regarding the handling of F2 gas.

REFERENCES

1. Martz JC, Hess DW, Haschke JM, Ward JW, Flamm BF. Demonstration of plutonium etching in a CF4/O2 RF glow discharge. Journal of nuclear materials. 1991;182:277-80.

2. Schutze A, Jeong JY, Babayan SE, Jaeyoung P, Selwyn GS, Hicks RF. The atmospheric-pressure plasma jet: a review and comparison to other plasma sources. IEEE Transactions on Plasma Science. 1998;26(6):1685-94.

3. Tatenuma K, Hishinuma Y, Tomatsuri S, Ohashi K, Usui Y. Newly Developed Decontamination Technology Based on Gaseous Reactions Converting to Carbonyl and Fluoric Compounds. Nuclear Technology. 1998;124(2):147-64.

4. Kim YS, Min JY, Bae KK, Yang MS. Uranium dioxide reaction in CF4/O2 RF plasma. Journal of nuclear materials. 1999;270(1-2):253-8.

5. Tu VJ, Jeong JY, A S, Babayan SE, G D, Selwyn GS, Hicks RF. Tantalum etching with a nonthermal atmospheric-pressure plasma. Journal of Vacuum Science & Technology A: Vacuum, Surfaces, and Films. 2000;18(6):2799-805.

6. Veilleux JM, El-Genk MS, Chamberlin EP, Munson C, FitzPatrick J. Etching of UO2 in NF3 RF plasma glow discharge. Journal of Nuclear Materials. 2000;277(2-3):315-24.

7. Windarto HF, Matsumoto T, Akatsuka H, Suzuki M. Decontamination process using CF4-O2 microwave discharge plasma at atmospheric pressure. J Nucl Sci Technol. 2000;37(9):787-92.

8. AGENCY IAE. Management of Radioactive Waste from the Mining and Milling of Ores. Vienna: IAEA; 2002.

9. Hicks RF, Herrmann HW. Atmospheric-Pressure Plasma Cleaning of Contaminated Surfaces. United States; 2003. Contract No.: DOE/ER--45857.

10. Kim YS, Jeon SH, Jung CH. Fluorination reaction of uranium dioxide in CF4/O2/N2 r.f. plasma. Annals of Nuclear Energy. 2003;30(11):1199-209.

11. Kim Y-H, Choi Y-H, Kim J-H, Park J, Ju W-T, Paek K-H, Hwang YS. Decontamination of radioactive metal surface by atmospheric pressure ejected plasma source. Surf coat technol. 2003;171(1-3):317-20.

12. Kim Y-s, Seo Y-d, Koo M. Decontamination of Metal Surface by Reactive Cold Plasma: Removal of Cobalt. J Nucl Sci Technol. 2004;41(11):1100-5.

13. Yang X, Moravej M, Babayan SE, Nowling GR, Hicks RF. Etching of uranium oxide with a non-thermal, atmospheric pressure plasma. Journal of nuclear materials. 2004;324(2-3):134-9.

14. Suzuki M, Kadowaki M, Windarto FH, Mori S. Comparison of Different Types of Plasma in Radioactive Surface Decontamination Process. Materials Science Forum. 2005;502:321-6.

15. Alim F, Richard A, Adam J, Laurel OC, Charlotte L, Corrie W. Guide for the Selection of Chemical, Biological, Radiological, and Nuclear Decontamination Equipment for First Responders: DHS Guide 103-06; 2007.

16. AGENCY IAE. Managing Low Radioactivity Material from the Decommissioning of Nuclear Facilities. Vienna: IAEA; 2008.

17. Jeon SH, Kim YS. A Study on Plasma Etching Reaction of Cobalt for Metallic Surface Decontamination. Journal of the Korean Radioactive Waste Society. 2008;6(1):17-23.

18. Jeon SH, Kim YS, Jung CH. Cold Plasma Processing and Plasma Chemistry of Metallic Cobalt Surface. Plasma Chemistry and Plasma Processing. 2008;28(5):617-28.

19. Fukumoto H, Fujikake I, Takao Y, Eriguchi K, Ono K. Plasma chemical behaviour of reactants and reaction products during inductively coupled CF4 plasma etching of SiO2. Plasma Sources Science and Technology. 2009;18(4):045027.

20. Xie HD, Sun B, Zhu XM, Liu YJ. Influence of O2 on the CF4 Decomposition by Atmospheric Microwave Plasma. International Journal of Plasma Environmental Science and Technology. 2009;03(01):39-42.

21. Veilleux JM, Kim Y. Can plasma decontamination etching of uranium and plutonium be extended to spent nuclear fuel processing? 52nd Annual Meeting, Institute of Nuclear Materials Management; 2011-06-03; United States: N. p.: USDOE; 2011.

22. CHEN K-C. Generalized Approach for Selecting Plasma Chemistries in Metal Etch. CALIFORNIA: UCLA; 2016.

23. Karbalai Akbar M. design of a Dielectric Discharge Barrier (DBD) Plasma Generator to Remove Uranium Contamination from Surfaces: Payame Noor University; 2018.

24. Semmler J, Kuang W, Volchek K, Toor A, Snaglewski A, Khan Z, Azmi P. Large area decontamination after a radiological incident. Journal of Environmental Radioactivity. 2019;199-200:66-74.

25. Lee J, Kim K, Kim Y-S. A Study on the NF3 Plasma Etching Reaction with Cobalt Oxide Grown on Inconel Base Metal Surface. Plasma Chemistry and Plasma Processing. 2019;39(4):1145-59.

26. Taran A, Lozina A, Garkusha I, Taran V, Krasnyj V, Gnidenko Y, et al. Evaluation of active power in dielectric barrier discharge with pulse power supply. Journal of Instrumentation. 2021;16(12):T12019.

27. Lee J-Y, Kim K-M, Ko M-S, Kim Y-S. A study of the NF3 plasma etching reaction with Cobalt oxide films grown on an inorganic compounds. Nuclear Engineering and Technology. 2022;54(12):4449-59.

28. Kim SJ, Jeong JW, Park SY, Chung CW. High-density plasma etching of Cobalt thin films using C2H5OH/O2/Ar gas mixture. Materials Science and Engineering: B. 2023;293:116494.

29. Kim Y, Chae S, Ha H, Lee H, Lee S, Chae H. Thermal atomic layer etching of Cobalt using plasma chlorination and chelation with hexafluoroacetylacetone. Applied Surface Science. 2023;619:156751.

30. LXCAT, electron scattering database, University of Toulouse, France. www.lxcat.net. Accessed 1 Nov 2017

31. Lee JY, Kim KM, Ko MS, Kim YS. A study of the NF3 plasma etching reaction with Cobalt oxide films grown on an inorganic compounds. Nuclear Engineering and Technology. 2022 Dec 1;54(12):4449-59.

32. Askari M, Farahani ND, Davani FA, Mahmoodi MB, Alam ZR. Experimental Investigation of the Decontamination of metal Cobalt from Steel 304 Surface by the Dielectric Barrier Discharge Plasma 9th Iranian Plasma Engineering and Plasma Physics Conference; Gonbad Kavous University: ISC; 2022.

33. Askari M, Farahani ND, Davani FA. Modeling and simulation of dielectric barrier discharge with CF4 etching gas by Comsol Multiphysics. ICNST22; NSTRI, Tehran2022.

34. Sohbatzadeh F, Mirzanejhad S, Shokri H, Nikpour M. Inactivation of Aspergillus flavus spores in a sealed package by cold plasma streamers. Journal of Theoretical and Applied Physics. 2016 Jun;10:99-106.