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Modeling and simulation of dielectric barrier discharge with CF$_4$ etching gas by Comsol Multiphysics

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Abstract

In nuclear waste management problems, removing radioactive contaminants from various components before the final waste disposal is difficult. Recently, plasma cleaning or etching techniques have been considered to remove contaminated used nuclear instruments and equipment. Molar value measurement of Fluorine ions in plasma formed with CF$_4$ etching gas and optimizing parameters experimentally is not economical, so simulations will be useful and cost-effective. In this study, the influence of distance between electrodes, temperature and dielectric thickness on fluorine ion molar changes in CF$_4$ plasma was investigated. Also, when the arc length increases, there is an optimum value for fluorine ion amount. This is the optimal value at a voltage of ~600 V with a dielectric thickness of 1(mm) and a temperature of 600 (° K) and Arc length equal 2 (cm).

Keywords: ***DBD, Radioactive Decontamination, CF$_4$ Etchant, Comsol ultiphysics***

Introduction

Many major instruments and equipment, especially in the primary circuit of nuclear power plant, are gradually contaminated by the adsorption of radioactive isotopes under continuous energy utilization. If these surface contaminants can be selectively removed, the radioactive substrate metal can be converted to non-radioactive or low-level radioactive material, which leads to huge reduction in the amount of radioactive waste. Also, significant economic benefits achieved if the substrate is recycled [1]. Secondary waste production is one of the disadvantages of cleaning processes techniques, such as wet chemical process, mechanical machining, and high-reliability gas phase cleaning processes [10-11].

Dry processing technique selectively selects surface contaminants, then converts them to volatile compounds through the catalytic surface, and finally removes them from the surface. Since the early 1990s, various studies have been reported the new method for nuclear applications using various plasma torches [2-9]. In the present study, clearance rate from the metal surface depends on the production of fluorine (F) radicals in the plasma, and the goal of this study is to design a plasma DBD with maximum reaction rate. CF$_4$ gas was selected as the etching gas due to the others report about fluorine (F) radicals (the target product in this study) that can be abundantly produced in the plasma and in the current cleanup process.

Simulation

In present research, using COMSOL 5.6 software, a dielectric barrier discharge plasma generator with circular geometry with a diameter of 10 cm with CF$_4$ gas have been simulated (reactions used in Tables 1-3). The chosen dielectric is Quartz (SiO2) with a relative electrical conductivity of 3.8 and it is placed on one of the applied electrodes. The distance between the two electrodes is examined in several different geometries. The secondary electron coefficient for both electrodes is a constant value 0.0007, and the voltage amplitude is equal to 600 V and frequency equal 60kHz. The simulation geometry can be seen in Figure 1.

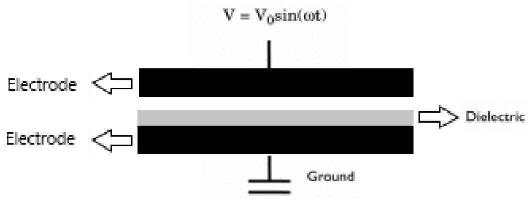

Figure 1. DBD simulated geometry

The reactions used in this simulation are described in the following tables 1-3:

Table 1. The reactions of electron impact [12]

Reaction	**Formula**	**Type**	**∇E(ev)**
R1	E+CF4 =>CF3(-)+F	Attachment	**0**
R2	E+CF4 =>F(-)+CF3	Attachment	**0**
R3	E+CF4 => $CF4^{-}$	Attachment	**0**
R4	E+CF4=>E+CF4	Excitation	**0.05**
R5	E+CF4=>E+CF4	Excitation	**0.112**
R6	E+CF4=>E+$CF4^{*}$	Excitation	**8**
R7	E+CF4=>E+$CF4^{*}$	Excitation	**12.5**
R8	E+CF4=>E+E+$CF4^{*}$	Ionization	**16.25**
R9	E+F=>E+F	Elastic	**m/M=0.0000288751**
R10	E+F=>E+F	Excitation	**12.818**
R11	E+F=>E+E+F^{+}	Ionization	**17.687**

Table 2 .Two and three body reactions between the atoms and molecules, the unit of the two and three body rates are m^3/s and m^6/s, respectively[13]

Reaction	Formula	Rate($\mathbf{m^3}$/s OR $\mathbf{m^6}$/s mol)
R1	CF4+E=>CF3+F+E	$2\times\mathbf{10^{-15}}\exp(-13/Te)$
R2	CF4+E=>CF2+2F+E	$5\times\mathbf{10^{-15}}\exp(-13/Te)$
R3	CF4+E=>CF+3F+E	$4.8\times\mathbf{10^{-17}}\exp(-20/Te)$
R4	CF3+E=>CF3+F+E	$3.3\times\mathbf{10^{-16}}$
R5	CF2+E=>CF+F+E	$3.3\times\mathbf{10^{-16}}$
R6	CF4+E=>CF3++F+2E	$7\times\mathbf{10^{-14}}\exp(-17.4/Te)$
R7	CF4+E=>CF2++2F+2E	$7\times\mathbf{10^{-15}}\exp(-24.7/Te)$
R8	CF4+E=>CF++3F+2E	$9\times\mathbf{10^{-15}}\exp(-30/Te)$
R9	CF3+E=>CF3++2E	$8\times\mathbf{10^{-15}}\exp(-12.2/Te)$
R10	CF2+E=>CF2++2E	$2.5\times\mathbf{10^{-14}}\exp(-12.2/Te)$
R11	CF+E=>CF++2E	$2.5\times\mathbf{10^{-14}}\exp(-15.3/Te)$
R12	CF4+E=>F-+CF3	$4.6\times\mathbf{10^{-15}}\boldsymbol{Te}^{\mathbf{-1.5}}\exp(-7/Te)$
R13	CF3++E=>CF3	$4\times\mathbf{10^{-14}}$
R15	CF++F-=>CF+F	$4\times\mathbf{10^{-13}}$
R16	CF2++F-=>CF2+F	$4\times\mathbf{10^{-13}}$
R17	CF3++F-=>CF3+F	$4\times\mathbf{10^{-13}}$
R18	CF3++F-=>CF4	$5\times\mathbf{10^{-14}}$
R19	CF3+F-=>CF4+E	$5\times\mathbf{10^{-16}}$

Table 3 .Surface reactions

Reaction	Formula	Sticking coefficient
R1	CF4+=>CF4	1
R2	CF4S=>CF4	1
R3	CF3-=>CF3	1
R4	CF4-=>CF4	1
R5	F-=>F	1
R6	F=>F	1
R7	CF3=>CF3	1

Results and discussion

Figure 2, 3, and 4 show the effect of temperature, dielectric thickness and gap length on the molar value of fluorine ions, respectively.

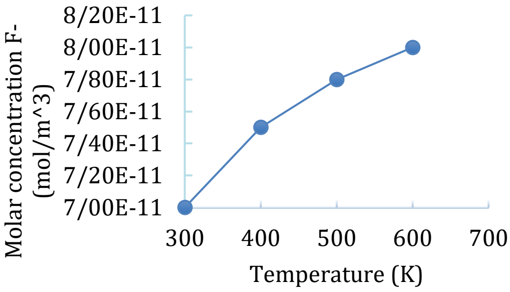

Figure 2. The effect of electrode temperature on the molar value of fluorine ions in dielectric=1mm and arc length=2 cm.

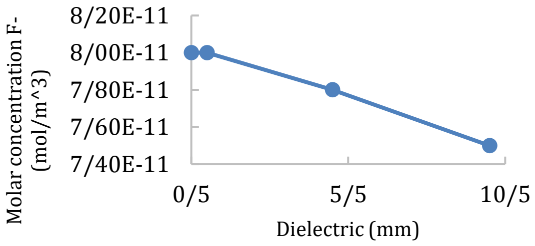

Figure 3 . The effect of dielectric thickness on the molar value of fluorine ions in temperatur=600° K and arc length=2 cm.

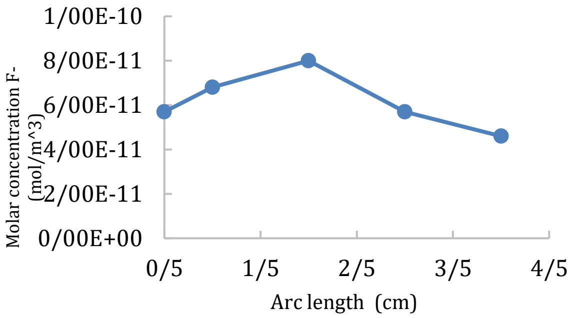

Figure 4 . The effect of gap length between DBD electrodes on the molar value of fluorine ions in in temperatur=600° K and dielectric=1mm.

As you can see, the effect of the arc length (length of the produced plasma) is greater for a voltage of ~-600 V at an atmospheric pressure and the temperature of 600 ° K. So, there is an optimal value for our research.

Conclusion

We simulated several different geometries to examine the number of fluorine ions mol, which is an important factor in cleaning decontaminations. Our results showed that the distance between the electrodes has an optimal value, which increases the cleaning speed. Increasing this distance leads to plasma extinction and decreasing it will reduce the molar amount of fluorine ion. We also found that where the arc length has the lowest amount or the plasma is uniform (without arc), the amount of fluoride ions in the plasma volume has not uniform distribution. The results showed increased temperature could increase the amount of fluorine ions mol near the dielectric surface to some extent and the rate of decontamination would be higher. In Figure 3, the effect of dielectric thickness on the amount of fluorine ions was investigated and it was shown when the thickness decreased, the amount of fluorine ions mole increases, which can be found in the dielectric loss coefficient. Dielectric thickness decreases and the amount of fluorine ions increases, but it should be noted that low thickness reduces its strength and causes dielectric breakdown.

References

[1] OECD/NEA, Nuclear Decommissioning: Recycling and Reuse of Scrap Metals, A Report by a Task Group of the Co-operative Programme on the Decommissioning, OECD, (1996).

[2] INEL, Idaho National Engineering Laboratory Decontamination and Decommissioning Technology Logic Diagram,Vol. 3, EGG-WTD-11104, (1994).

[3] Y. S. Kim, S. H. Jeon, C. H. Jung, ‘‘Fluorination reaction of uranium dioxide in CF4/O2/N2 r.f. plasma,’’ Ann. Nucl. Eng., 30, 1199 (2003).

[4] Y. H. Kim, Y. H. Choi, J. H. Kim, J. K. Park, W. T. Ju, K. H. Paek, Y. S. Hwang, ‘‘Decontamination of radioactive metal surface by atmospheric pressure ejected plasma source,’’ Surf. Coat. Technol., 171, 317 (2003).

[5] H. F. Windarto, T. Matsumoto, H. Akatsuka, M. Suzuki, ‘‘Decontamination process using CF4–O2 microwave discharge plasma at atmospheric pressure,’’ J. Nucl. Sci. Technol., 37, 787 (2000).

[6] Y. S. Kim, J. Y. Min, K. K. Bae, M. S. Yang, ‘‘Uranium dioxide reaction in CF4/O2 rf plasma,’’ J. Nucl. Mater., 270,253 (1999).

[7] K. Tatenuma, Y. Hishinuma, S. Tomatsuri, ‘‘Newly developed decontamination technology based on gaseous reactions converting to carbonyl and fluolic compounds,’’ Nucl. Technol., 124, 147 (1998).

[8] E. B. Munday, D. W. Simmons, Oak Ridge K-25 Site, K/TCD- 1048, (1993).
[9] J. G. Malm, P. G. Eller, L. B. Asprey, ‘‘Low temperature synthesis of plutonium hexa-fluoride using di-oxygen di-fluoride,’’ J. Am. Chem. Soc., 106, 2726 (1984).
[10] U. Chun, K. Choi, K. Yang, J. Park, M. Song, Waste Manage.18 (1998)183.
[11] J. Veilleux, M. El-Genk, E. Chamberlin, C. Munson, J. Ritz-Patrick, J. Nucl. Mat. 277 (2000) 315.
[12] LXCAT, electron scattering database, University of Toulouse, France. www.lxcat.net. Accessed 1 Nov 2017
[13] Hiroshi Fukumoto, Isao Fujikake1, Yoshinori Takao, Koji Eriguchi and Kouichi OnoPlasma chemical behaviour of reactants and reaction products during inductively coupled CF4 plasma etching of SiO2(2009)