RPE | Radiation Physics & Engineering
A Peer-Reviewed Journal
Published by K.N. Toosi University of Technology
http://rpe.kntu.ac.ir
Research Article

Radiation Physics and Engineering 2023; 4(2):45–51

# Calculation of the role of Wigner energy in decontamination of Cs-137 from irradiated Graphite pores

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## HIGHLIGHTS

- The decontamination of Cs-137 from irradiated graphite is investigated using the plasma-sputtering method.
- The results show that the decrease in radiation pollution depends on the release of Wigner energy.
- This study demonstrates the benefits of using noble gases as a plasma feed gas in the plasma decontamination process.
- The results can be used to decontamination of various types of irradiated porous materials used in nuclear systems.
- Researchers of radiation safety, nuclear waste management and nuclear decommissioning will be interested in the results.

## ABSTRACT

During the operation of Graphite -fuel HTGR (High-Temperature Gas-cooled Reactor) nuclear reactors, Graphite used as a neutron moderator, is irradiated and has a variety of contaminants (such as Cs-137, Co-60, and Sr-90) and due to industrial and environmental considerations, decontamination of irradiated Graphite is very important. In this study, the decontamination of Cs-137 trapped in Graphite pores of Graphite -fuel (HTGR) nuclear reactors has been analyzed. The proposed method for decontamination of irradiated Graphite surfaces is the thermal plasma-sputtering method with noble feed gases, which are used to reduce the risk of radioactive Graphite waste and in this regard, a mathematical model was developed to describe the process of decontamination of irradiated Graphite, which is prone to release Wigner energy due to defects and torsion caused by radiation. The results show that the decrease in radiation pollution of irradiated Graphite waste and various parameters of its decontamination process depend on the release of Wigner energy. The results obtained are in good agreement with the other researchers results.

## KEYWORDS

Decontamination
HTGR Reactor
Irradiated Graphite waste
Radionuclides
Wigner Energy
Mathematical model

## HISTORY

Received: 2 May 2022
Revised: 2 August 2022
Accepted: 15 August 2022
Published: Spring 2023

## 1 Introduction

The decommissioning and dismantling of old and worn-out nuclear equipment are as important as the commissioning of nuclear power plants and new nuclear sites (Laraia, 2012; IAEA, 2001). In 2008, the International Atomic Energy Agency published a report in which the high impact of using dry plasma decontamination method in the process of decommissioning of the nuclear equipment is mentioned (IAEA, 2008).

### *1.1 The role of Graphite in the high-temperature gas-cooled reactor (HTGR)*

The HTGR nuclear reactor is a generation IV nuclear reactor that uses Graphite as a neutron moderator, the Uranium fuel cycle, and Helium as coolant. During the operation of HTGR (High Temperature Gas Reactors) nuclear reactor, Graphite is irradiated and after a while, Graphite loses its original properties and needs to be decontaminated or replaced. In addition, after the useful life of the HTGR reactor and during its decommissioning process, in order to comply with radiation and environmental protocols, irradiated Graphite must first be decontaminated and then scrapped (Kuijper et al., 2006; Sato and Yan, 2019). In 2020, Chinese researchers published a report on the decontamination of Graphite surfaces using plasma method in which, factors related to plasma ability to decontaminate radioactive agents from Graphite surfaces were investigated and it acknowledges that the

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https://doi.org/10.22034/rpe.2022.340253.1084
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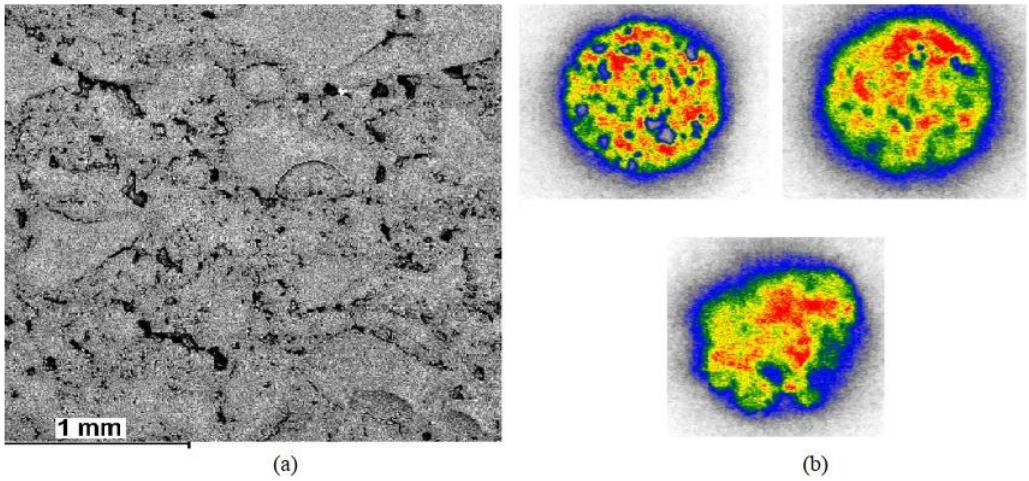



**Figure 1:** a) SEM images of polished irradiated Graphite; b) Autoradiographic images of irradiated Graphite in which, red, blue, and white represent high, medium, and low activity, respectively (Bespala et al., 2019).

plasma method is very effective, inexpensive and promising. Bushuev et al. (Bushuev et al., 2003) presented the results of a study of bushing contamination in Graphite -uranium reactors. These radioactive contaminants are including the radioisotopes of H-3, C-14, Fe-55, Co-60, and etc; and fission fragments such as Ru-106, Sb-125, Ba-133, Cs-137, Eu-154, Eu-155, and the transuranic elements such as Am-241, Am-243, Cm-244, etc. In the meanwhile, the most active radioisotopes are Cs-137, Co-60, Sr-90, Cl-36, and H-3 (Bespala et al., 2018; Pavliuk et al., 2018b). In 2016, the international project of the GRAPA (Irradiated Graphite Processing Approaches) was implemented by the IAEA (International Atomic Energy Agency) with the participation of various countries in which, its content is about the upcoming and new approaches to irradiated Graphite processing (Wickham et al., 2017). Modern knowledge offers a variety of methods for the treatment of Graphite radioactive waste (GRW) and in the meanwhile, the plasma sputtering method using noble feed gas is very effective in decontamination of Cs-137 from irradiated Graphite surfaces (Theodosiou et al., 2018; Vulpius et al., 2013; Fachinger et al., 2008; Dunzik-Gougar and Smith, 2014).

Also it has been shown that Cesium, cobalt, and strontium radionuclides are to be metallic during reactor operation and they have no fixed molecular bond with each other or with Graphite molecules and they are trapped in the Graphite pores and this lack of molecular binding is a good motivation and physical basis for researchers to study the process of decontamination using plasma-sputtering method; Because contamination molecules do not chemically bond with Graphite substrate molecules and thus, we don't need to use plasma-chemical methods for decontamination (Pavliuk et al., 2018a; Theodosiou et al., 2018). Plasma processing is one of the promising methods for decontamination of irradiated Graphite in the nuclear industry and therefore, the simulation and the mathematical study of this process cause that, in addition to detailed analysis of decontamination operations, we minimize the potential effect of the radiation hazards during the actual experimental operation process (Bespala et al., 2016).

### *1.2 Plasma-sputtering method in decontamination of surfaces*

One of the important advantages of plasma in the surface-processing is its major role in minimizing the adverse effects of industrial activities on the environment. Many physical and chemical phenomena are often involved in plasma etching. One of the mechanisms of plasma etching is the etching by sputtering method. In the sputtering method, the positive ions are present throughout the plasma sheath and due to their inherent escape, they accelerate to the boundaries of the plasma and they strike the contamination of substrates with high kinetic energy and some of their energies are transferred to the atoms of the surface contamination, which will lead to the evaporation (or sublimation) of the contamination. This process is different from other mechanisms because its mechanism is mechanical (not chemical), i.e. their escape conditions are provided only by applying force and transferring mass and energy to surface molecules (Auciello et al., 2012). In the present study, the decontamination of Cs-137 from irradiated Graphite is investigated and analyzed using the plasma-sputtering method in which, Thermodynamic processes of heat and mass transfer during the interaction and effect of plasma on the surfaces of irradiated Graphite are analyzed.

## 2 Methods

### *2.1 Selection of reference irradiated Graphite sample*

In reference (Bespala et al., 2019), a Graphite sample (sampling from Graphite stack) of Seversk Uranium-Graphite reactor in the Tomsk region of Russia is experimentally analyzed. This Graphite sample, which has been irradiated in the reactor core for 9 years and was experimentally studied by Russian researchers in 2019, is used

as a reference sample for thermodynamic analysis in this study.

Figure 1-a shows the SEM image of the irradiated Graphite in which, gray, black, and white indicate the irradiated Graphite, pores, and non-carbon solid impurities respectively. The white dots indicate the presence of fission products and neutron activation products at the Graphite surface. However, radioactive contaminants can be located inside the Graphite pores and not appear in the SEM image (Bespala et al., 2019; Bushuev et al., 2015). Radionuclides are impurities that can penetrate into the Graphite substrate and disperse into its pores and we also know that one of the challenges of technology is to remove contaminations from porous materials. The decontamination process requires almost complete removal of contaminants from the Graphite pores and for this purpose, arc discharge plasma and non-thermal atmospheric pressure plasma (NTP) can be used to process irradiated Graphite. In order to reduce the specific activity of irradiated Graphite and also reduce its potential hazards, it is recommended that its heat treatment be performed in an inert gas environment. When a plasma gas stream comes in contact with the Graphite substrate, the Graphite will heat up and when the surface reaches the boiling point of radioactive contaminants, evaporation or sublimation of radionuclides is possible under certain thermodynamic conditions. In most of the experimental findings of other researchers, the results of analysis of the SEM images show that Graphite has an integrated porous structure and most of the non-carbon pollutants are inside its pores.

### 2.2 *Thermodynamic analysis*

In this study, in order to thermodynamic analysis of the Graphite decontamination process, we consider the geometry in such a way that a thin layer of the Cs-137 radionuclides are located in the one of the Graphite pores. We also assume that in during the mathematical modeling process, Cs-137, Co-60, and Sr-90 radiation pollutants dont mix with each other. Thus, in addition to simplifying the form of differential equations, we get closer to the real physical conditions (Bespala et al., 2018, 2019). In references (Bespala et al., 2019; Kane et al., 2013; Barbin et al., 2015; Bespala et al., 2017, 2016; Pavliuk et al., 2018b) efforts for mathematical modeling of thermal processing of the irradiated Graphite are presented and in some of them, the release of Wigner energy is also considered. In our proposed geometry for the analysis in this study, it is assumed that an atmospheric pressure plasma jet with argon as feed gas moves along the Graphite specimen that has a porous surface (Fig. 2).

## 3 Results and discussion

### 3.1 *Thermodynamic analysis*

The plasma decontamination process of irradiated Graphite confirms the effectiveness of the plasma flow on the Graphite surface, which leads to the sublimation of the radioactive contaminants that are trapped in the Graphite pores. When irradiated Graphite is exposed to argon plasma flow, we can get a fairly good estimate of the chemical composition produced (Bespala et al., 2018, 2019). For this purpose, in this study, TERRA software package was used for modeling and thermodynamic analysis of reactions, in which the capabilities of the main version of this software package are mentioned in references (Aref'yev et al., 1974; Young, 1975).

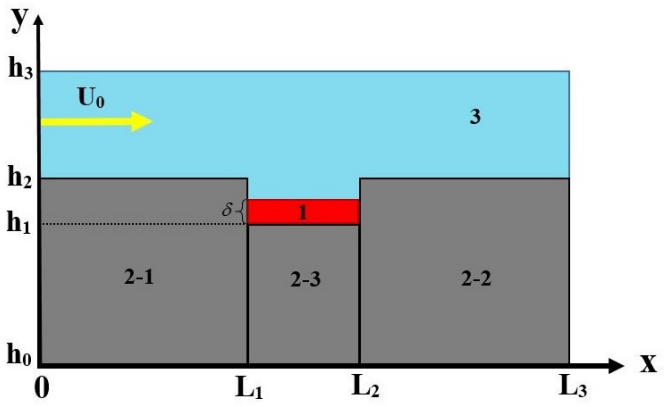



**Figure 2:** Our proposed scheme for analyzing in this study; 1- Layer of Cs-137 (as the radioactive contamination), 2- Graphite (divided into three parts), and 3- Plasma region.

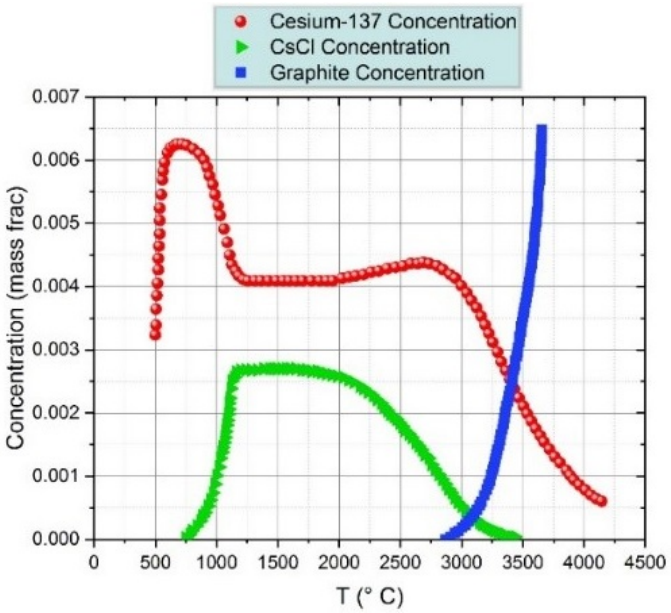



**Figure 3:** Variation of Cs-137, CsCl, and Graphite concentrations during interaction of unit cell with argon atmospheric pressure plasma.

The results show that, in the temperature range of 500 to 2000 °C, the Cs-137 structures are in the form of chloride ($SrCl_2$, SrCl, and CsCl). Figure 3 shows that selective sublimation of radionuclides due to the interaction of plasma flow with irradiated Graphite can be managed by changing the temperature of plasma average mass and this factor reduces GRW activity. The diagrams shown

in Fig. 3 also illustrates that in the plasma decontamination process of irradiated Graphite and over a wide range of temperatures, the Cs-137 radionuclides have high mass concentrations and therefore it is very important and this is another reason that why, of all the radionuclides, we will do the calculation for the Cs-137 radionuclides in this study. Another important conclusion to be drawn from Fig. 3 is the evaporation of irradiated Graphite starts at approximately 3000 °C; Therefore, to avoid damaging the Graphite structure, the temperature of the plasma jet should not be exceeded 3000 °C. In Fig. 3, the mass fraction means the evaporated mass divided by the initial mass (the amount of evaporated material divided by the amount of the initial material).

### 3.2 *Thermodynamic modeling of the plasma decontamination process of the irradiated Graphite*

According to Fig. 2, in the intended geometry for computational analysis, the plasma flow velocity at the input of the desired computational range (has height of $|h_3 - h_2|$ on the Graphite surface) is equal to $u_0$. It is also assumed that the transverse and longitudinal dimensions of entire computational cell are much larger than the thickness of the Cs-137 contamination. The Graphite surface heats up due to contact with the plasma particles and heat transfer. For simulation of the heat and mass transfer processes of Cs-137 in irradiated Graphite pores, their differential heat transfer equations were solved in Cartesian coordinates by Maple 2020 software. The positive direction of the X-axis is assumed along the velocity vector $u_0$ (plasma flow velocity):

$$c_1\rho_1\frac{\partial T_1}{\partial t} = \lambda_1\Big(\frac{\partial^2 T_1}{\partial x^2} + \frac{\partial^2 T_1}{\partial y^2}\Big) \quad (1)$$

in which $h_1 < y < h_1 + \delta$ and $L_1 < x < L_2$.

$$c_2\rho_2\frac{\partial T_2}{\partial t} = \lambda_2\Big(\frac{\partial^2 T_2}{\partial x^2} + \frac{\partial^2 T_2}{\partial y^2}\Big) + Q_{Wig} \quad (2)$$

in which $0 < x < L_1$, $h_0 < y < h_2$; $L_1 < x < L_2$, $h_0 < y < h_1$; $L_2 < x < L_3$, $h_0 < y < h_2$.

$$c_3\rho_3\frac{\partial T_3}{\partial t} = \lambda_3\Big(\frac{\partial^2 T_3}{\partial x^2} + \frac{\partial^2 T_3}{\partial y^2}\Big) - c_3\rho_3 u\Big(\frac{\partial T_3}{\partial x} + \frac{\partial T_3}{\partial y}\Big) \quad (3)$$

in which $L_1 < x < L_2$, $h_1 + \delta < y < h_2$; $0 < x < L_3$, $h_2 < y < h_3$.

$$u = u_0\Big(\frac{T_3}{298}\Big) \quad (4)$$

In these equations, $c_1$, $\rho_1$, and $\lambda_1$ are heat capacity, density, and thermal conductivity of Cs-137 contamination, respectively. $c_2$, $\rho_2$, and $\lambda_2$ are heat capacity, density, and thermal conductivity of irradiated Graphite, respectively. $Q_{Wig}$ is the power density of heat released due to temperature changes of defective graphitic structures (the release of Wigner energy). $c_3$, $\rho_3$, and $\lambda_3$ are heat capacity, density, and thermal conductivity of plasma flow, respectively. $h_0$ is the initial height of inactive Graphite layer (equal to zero), $h_1$ is the height of the Graphite pore from the origin, $h_2$ is the height of the upper surface of the inactive Graphite layer from the origin, $h_3$ is the height of the plasma boundary, $L_1$ and $L_2$ are the longitudinal coordinates of the beginning and end of the Cs-137 contamination layer, and $L_3$ is the length of the Graphite sample. Also, $T_1$, $T_2$, and $T_3$ are the temperatures of Cs-137 contamination, the temperature of irradiated Graphite, and the temperature of plasma particles (at the time $t$), respectively. The right side of Eq. (2) contains $Q_{Wig}$, which represents the release of stored energy (Wigner energy) and during the plasma processing of irradiated Graphite, this parameter affects the thermal balance of the whole system. Additional information on the release of Wigner energy from irradiated Graphite is also given in the reference (Bespala et al., 2018). Calculations show that as a result of the release of Wigner energy, the temperature of irradiated Graphite under the adiabatic heating can be increased up to $\Delta T = 600 - 650$ °C (Bespala et al., 2019).

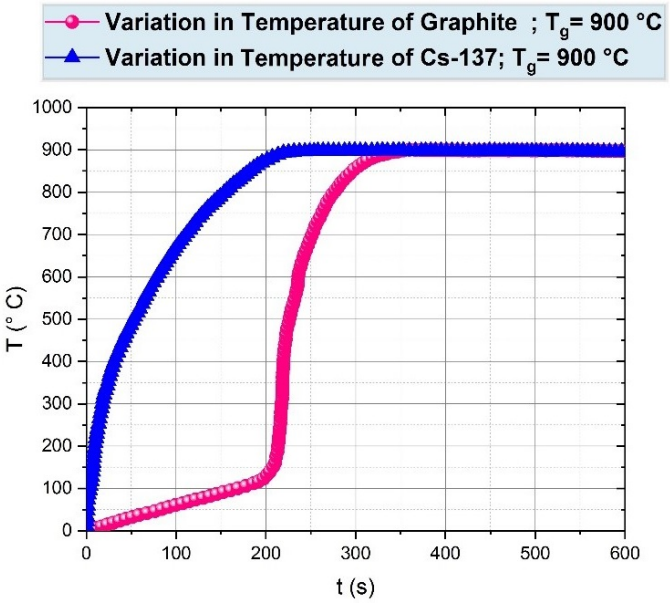


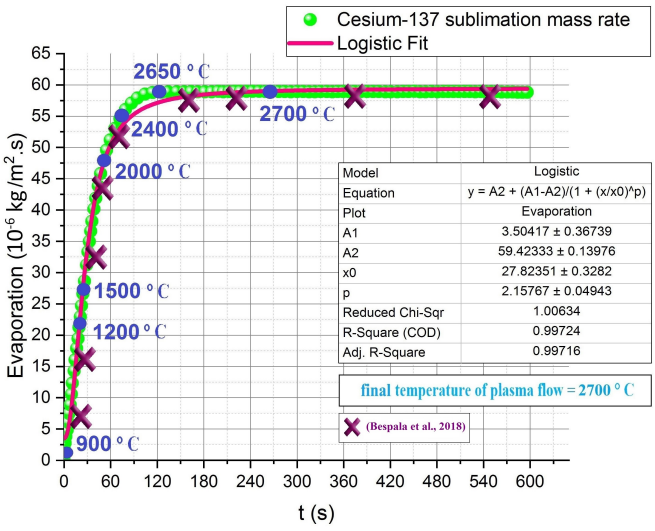

**Figure 4:** Temperature changes over time in Graphite and Cs-137. The sharp variation occurs at $t = 200$ s for Graphite which is due to the release of Wigner energy.

The mass transfer process depends on thermodynamic conditions and diffusion coefficients in which, their values are given in (Bespala et al., 2018, 2019). Also, the thermodynamic parameters of the Cs-137 contamination are mentioned in reference (Bespala et al., 2017). Since the sublimation of Cs-137 leads to its diffusion in the plasma stream, solving the above equations requires the determination of specific parameters of this process and since in this case, it is difficult to accurately calculate the diffusion parameters, the values of the Cs-137 diffusion parameters in Argon gas obtained from the reference (Aref'yev et al., 1974). We assume that at the initial time, for the whole system that it has not yet been irradiated with plasma, the temperature is uniform and equal to $T_0$ ($T_g > T_0$). Equations (1) to (4) were solved using Maple 2020 software under certain boundary conditions. Graphite tem-

perature changes was investigated at $T_g = 900$ °C; The results show that in the first 200 seconds, the graphite temperature changes linearly and in the next 100 seconds, the Graphite temperature changes rapidly and exponentially; The cause of this phenomenon is that in the first 200 seconds, most of the heat given to the Graphite leads to the compensation of lattice defects and as a result of the heated defects, the release of Wigner energy occurs, which is shown as a sharp variation in the Fig. 4. Finally, due to the fact that the heat of the irradiated Graphite is transferred to radioactive contaminants, 300 seconds after starting of the process, the Graphite temperature changes to saturation level. In this case, a sudden rise in temperature has a greater effect on the rate of heating of radioactive contaminants. Among all radioactive contaminants, Cs-137 radionuclide is more affected by the sudden rise in temperature than other contaminants due to its high thermal conductivity (Bespala et al., 2017).

The most meaningful parameter that indicates the efficiency of the irradiated Graphite surface processing is the rate of entry of the mass of radioactive contaminant ($M_i$) into the gas phase (Pavliuk et al., 2018a).

Therefore, after 200 seconds the interaction of argon gas flow with the Cs-137 surface, the Cs-137 temperature will be on the verge of saturation and due to the increase of sublimation capability of radioactive contaminants, plasma processing of irradiated Graphite is suggested at higher temperatures (up to a maximum of 3000 °C). According to Fig. 4, the Wigner energy emission simultaneously affects both the heating of the Graphite and Cs-137, but since the heat capacity of carbon is equal to 8.51 J.mol$^{-1}$.K$^{-1}$ and it is almost four times lower than the heat capacity of Cs-137 (32.210 J.mol$^{-1}$.K$^{-1}$), Carbon is more sensitive to Wigner energy than Cs-137 and thus, its temperature chart has a steeper slope than the temperature chart of Cs-137 and this is an example of the physical interpretation that under the same thermodynamic conditions, the materials with lower heat capacity will have a steeper slope of temperature changes than the materials with higher heat capacity; But in the case of Cs-137, although Wigner heat affects the Cs-137 temperature increase, but due to the high heat capacity of Cs-137, the amount of Wigner heat is not effective enough to show its effects on the Cs-137 temperature diagram and finally, the entire system reaches thermodynamic equilibrium within 300 seconds after the start of the heat treatment operation. Figure 5 shows the $T$ and $M_i$ change for the Cs-137 radionuclide during the temperature range up to 2700 °C. When the plasma flow interacting with surface of the irradiated Graphite pore, the rate of Cs-137 evaporation will soon be changed due to the temperature changes in the surface. Calculations show that the mass transfer rate varies in the range of $(2.5 - 60) \times 10^{-6}$ kg.m$^{-2}$.s$^{-1}$ under the boundary conditions that we have chosen for Graphite processing. As an important result, in the plasma processing of irradiated Graphite surfaces, the radionuclide compounds which have the lowest heat capacity evaporate more quickly, and then the surfaces are clean of them.




ANOVA

| | | DF | Sum of Squares | Mean Square | F Value | Prob>F |
|---|---|---|---|---|---|---|
| Evaporation | Regression | 4 | 266502.59072 | 66625.64768 | 66205.58579 | 0 |
| | Residual | 98 | 98.62179 | 1.00634 | | |
| | Uncorrected Total | 102 | 266601.21251 | | | |
| | Corrected Total | 101 | 35765.15713 | | | |

**Figure 5:** Temperature changes and rate of mass changes of Cs-137 sublimation in noble-gas plasma jet (with Argon as feed gas) over time and in the circumstances that plasma temperature rises from 900 to 2700 °C in a total of 270 s.

In Fig. 5, the vertical axis dimension (rate of mass changes of Cs-137 sublimation) is expressed in $10^{-6}$ g.mm$^{-2}$.s$^{-1}$, which in order to better express the physical concept, is shown in terms of its equivalent, which is $10^{-6}$ kg.m$^{-2}$.s$^{-1}$. In general, we use the sputtering decontamination technique when, from an engineering point of view, the sublimation temperature of the desired radioactive contamination is available. For example, the Co-60 radionuclide has a sublimation temperature of 2870 °C and its sublimation is possible using the plasma technique, but it requires more energy. Also in the case of the possibility of selective decontamination, this is possible for contaminants that have a very long half-life; because it may take a long time to reach the maximum rate of pollutant sublimation. For example, Co-60 radionuclide has a half-life of 5.27 years and can be selectively removed by modifying plasma parameters. It is important to note that in the plasma decontamination technique, the time to reach the highest rate of pollution sublimation should be less than the half-life of the desired pollutant and this issue should be paid special attention (Bespala et al., 2017).

## 4 Conclusions

Plasma processing is one of the promising methods for decontamination of irradiated Graphite in the nuclear industry and therefore, the simulation and the mathematical study of this process cause that, in addition to detailed analysis of decontamination operations, we minimize the potential effect of the radiation hazards during the actual experimental operation process. In this study, the decontamination of Cs-137 from irradiated Graphite

is investigated and analyzed using the plasma-sputtering method in which, Thermodynamic processes of heat and mass transfer during the interaction and effect of plasma on the surfaces of irradiated Graphite are analyzed. In order to reduce the specific activity of irradiated Graphite and also reduce its potential hazards, it is recommended that its heat treatment be performed in an inert gas environment (this action reduces GRW activity). This study demonstrates the benefits of using Argon as a plasma feed gas in the plasma decontamination process.

In the plasma decontamination process of irradiated Graphite and over a wide range of temperatures, the Cs-137 radionuclides have high mass concentrations and therefore it is very important and this is another reason why, of all the radionuclides, we will do the calculation for the Cs-137 radionuclides in this study. Another important conclusion is the evaporation of irradiated Graphite starts at approximately 3000 °C; Therefore, to avoid damaging the Graphite structure, the temperature of the plasma jet should not be exceeded 3000 °C. Results show that selective sublimation of radionuclides due to the interaction of plasma flow with irradiated Graphite can be managed by changing the temperature of plasma average mass and this factor reduces GRW activity.

During the process of decontamination of Cs-137 from irradiated graphite surfaces, using the plasma-sputtering method with noble argon feed gas, most of the heat given to the Graphite leads to the compensation of lattice defects and as a result of the heated defects, the release of Wigner energy occurs. The release of Wigner energy, simultaneously, affects the heating of graphite and Cs-137. Heat released due to temperature changes of defective graphitic structures (the release of Wigner energy) affects the thermal balance of the whole system. But because of the heat capacity of carbon which is about four times lower than that of Cs-137, carbon is more sensitive to Wigner energy and therefore, its temperature behavior will have a steeper slope than Cs-137 temperature. In the case of Cs-137, Wigner's heat has an effect on its temperature rise, but due to the high heat capacity of Cs-137, Wigner heat is not that much which its effects can be seen in the temperature rise diagram of Cs-137 and finally, the entire system reaches to the thermodynamic equilibrium within 300 seconds after the starting at heat treatment operation. the release of Wigner energy was also shown to affect the removal rate and clearance of Cs-137. The results obtained are in good agreement with the other researchers results (Bespala et al., 2018).

The results of this study can be used to decontamination of various types of irradiated porous materials used in other nuclear systems. According to the obtained results on the characteristics of plasma required for decontamination of Cs-137 from irradiated graphite, based on these characteristics, it is possible to design and manufacture useful plasma actuators in this field.

## Conflict of Interest

The authors declare no potential conflict of interest regarding the publication of this work.

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**To cite this article:**

Nikoosefat, M., Bagheri, A., Shakur, H. R., Shahbazi Rad, Z., Javadi, N. (2022). Calculation of the role of Wigner energy in decontamination of Cs-137 from irradiated Graphite pores. *Radiation Physics and Engineering*, 4(2), 45-51.

DOI: 10.22034/rpe.2022.340253.1084

To link to this article: https://doi.org/10.22034/rpe.2022.340253.1084