MODELING OF SPONTANEOUS PLUTONIUM PARTICLES SPUTTERING FROM PLUTONIUM DIOXIDE MATERIAL

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ABSTRACT

Theoretical study of spontaneous emission (sputtering) of radionuclide from plutonium dioxide material due to radioactive decay is performed by means of Monte-Carlo calculation. The resulting sputtering yield is 0.98 plutonium particles and 3.23 oxygen particles per decay of the source material. For a typical plutonium dioxide sample (density — 11.5 g/cm^3, activity — 2 GBq/g) these values relates to $3.40 \cdot 10^5$ and $1.11 \cdot 10^6$ cm$^{-2} \cdot$s^{-1} for plutonium and oxygen particles, respectively. The mean energy of sputtered particles is found to be 370 eV for plutonium and 120 eV for oxygen.

KEYWORDS

Plutonium, Sputtering, Monte-Carlo, Radioactive Aerosol

1. INTRODUCTION

The effect of spontaneous sputtering of radionuclide from substance is of considerable concern for correct radiation dose assessment for nuclear plant workers. Due to this effect any radioactive material may become a permanent source of new atmospheric radioactive aerosols. Moreover, inhaled and deposited activity in human body may also produce bound-free radioactive particles that migrate independently towards different tissues and organs.

Spontaneous displacement of some radionuclides was already observed in the earliest studies of radioactivity. However, these processes are still poorly investigated. There are only few publications on the subject of plutonium sputtering, although this effect may create a considerable radiation hazard for professional radiation workers and public population, as well.

The main objective of this work is numerical modeling of generation, interaction and transport of recoil cascades in plutonium-containing materials and theoretical calculation of trajectories, energy and angular distributions of particles, emitted from the outer surface of the material.

The results of this research in the form of simulation model and numerical theoretical estimates are used for further industrial radioactive aerosol studies and improvement of internal dosimetry methods for professional workers. These results can also be useful for the sputtering analysis of other hazardous radioactive elements of different origin.

2. METHODS

Modeling methodology used in this work is based on a mathematical calculation of nuclei trajectories in matter by Monte-Carlo simulation, based on the modern models and conceptions of the processes of nuclear and electronic stopping of ions in matter. Computer simulation is widely used for study of ion implantation, radiation damage, sputtering, reflection and transmission of ions in matter. The Monte-Carlo method has several distinct advantages over analytical formulations based on transport theory. It allows solving more rigorously problems of elastic scattering, to explicitly take into account surfaces and interfaces, to determine more easily energy and angular distributions. The main limitation of this method is that it is basically a time-consuming calculation.

Monte-Carlo method implies a large number of individual particle “histories” in the target material. Here, each history begins with radioactive decay of a plutonium nucleus that starts a chain of events in the original material leading sequentially to plutonium nuclei sputtering from the material surface. Initial position of the plutonium nucleus is assigned randomly (uniform distribution of activity within the object) with random initial direction of emitted decay products (isotropy). Further processes are associated with the movement of decay products within the object. It is assumed that particles change their direction due to binary nuclear collisions and move along straight free path between collisions. The energy decreases as a result of nuclear and electronic energy losses, and each history ends either when the energy drops below a predetermined value or when the particle’s position appears to be out of the target. The target is considered as an amorphous object with atoms at random locations, i.e. directional properties of crystal lattice are ignored. This method is applicable to a wide range of energies of incident particles from about 0.1 keV/u up to several MeV/u, depending on the masses involved. The lower limit is due to the consideration of binary collisions only, and the upper due to the exception of nuclear reactions and relativistic effects. The processes of nuclear and electronic stopping are considered independently.

In this work the transport of ions in matter was calculated using SRIM-2011 software [1]. This software package is well-known and acknowledged by scientific community as a tool for theoretical calculations of the stopping and range of energetic ions in matter. The package SRIM-2011 embodies the latest theoretical approaches for the structure of matter and processes of interaction of particles that are empirically linked to the data of numerous experimental studies. Detailed description of scientific background of this software package can be found in [2]. Additional software modules for random assignment of initial positions and angles of particles of interest, input files generation for SRIM-2011 and output files analysis were developed.

The total number of simulation sets carried out in this work amounts to 120 with 10 000 independent histories of events in each set (1.2 million individual histories altogether).

3. RESULTS

The decay of plutonium nuclei is followed by formation of alpha particles and uranium recoil nuclei with mean energies of 5.15 MeV and 87.7 keV, respectively. These particles transfer their energy to the plutonium dioxide material differently. The energy transfer of alpha particles is insignificant along the entire particle path except for the terminal part. As for uranium nuclei, the energy transfer has a maximum at the beginning of the path and slowly decreases to its end. Due to the energy transfer, recoil nuclei in the original material receive enough energy for its displacement within the material and formation of displacement cascades. The ranges of displaced nuclei are small and, in general, recoil cascades are located in the close proximity of the primary particles trajectories.

The emission of the near-surface nuclei from the matter takes place when recoil cascade transfers the energy high enough to overcome the surface binding energy. The sputtering yield is defined as the average nuclei number sputtered from the matter per one primary particle (an alpha particle or uranium recoil nucleus), that is per one decay act of plutonium. Detailed description of sputtering mechanics can be found, for example, in [3, 4].

The probability of particle sputtering is higher if the location of decaying plutonium nucleus is closer to the outer surface of the substance. Dependence of the sputtering yield on the depth of decaying atoms is shown in Fig. 1. One can see that sputtering occurs mainly due to decay of atoms locating within a distance of not more than 20-30 nm from the surface, and the dominant contribution is formed by atoms decaying within the depth of about 2 nm.

In order to simulate a real plutonium dioxide sample, initial locations of plutonium decaying nuclei were assigned uniformly along its volume. Figure 2(a) shows that the sputtering yield decreases with the extension of the sample thickness that corresponds to distribution of plutonium nuclei over a larger volume. To obtain comparable values of sputtering yield for different sample thicknesses, all the results were normalized to an appropriate value of the sample specific activity. All the values were subsequently evaluated for PuO_2 with density of 11.5 g/cm^3 and corresponding specific activity of $2 \cdot 10^{12}$ Bq/kg. The resulting dependence of the sputtering yield on the thickness of the simulated sample is shown in Fig. 2(b).

As can be seen, for sample thickness of about 20-30 nm, which corresponds to the mean free path of uranium nucleus in the material, sputtering yield saturates and slightly changes up to thickness of about 150 nm. From 150 nm and more the sputtering yield becomes extremely small, and the statistical accuracy is greatly reduced, so that in Fig. 2(b) there are some scatters around this area. In general, we found that in the wide range of sample thicknesses (from 30 to 150 nm), the simulation results remain representative and all final results were obtained by averaging over this range.

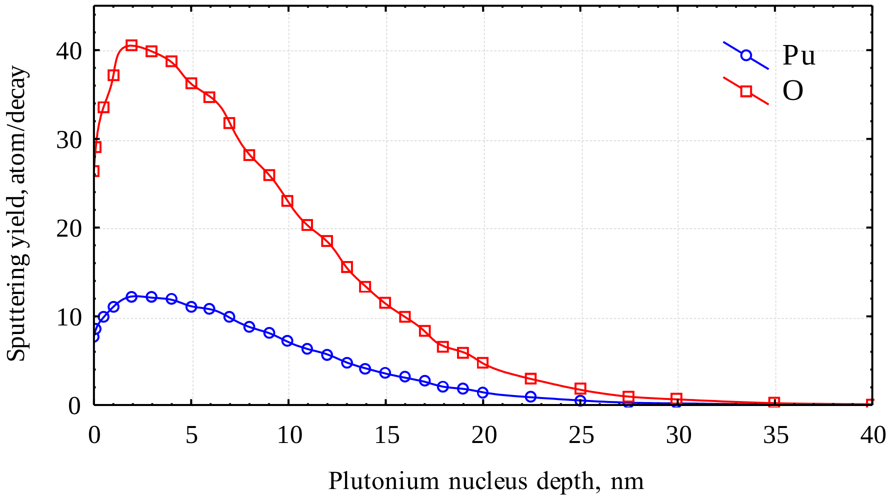

Fig. 1. Variation of sputtering yield vs. nucleus depth

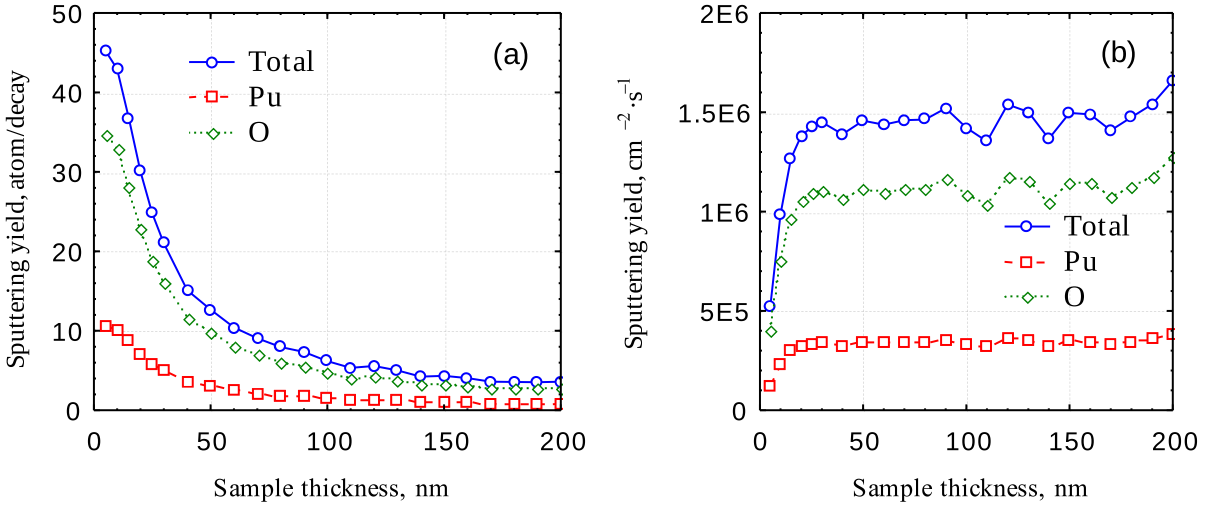

Fig. 2. Variation of sputtering yield vs. sample thickness (a — relative values, b — normalized values)

The resulting energy of sputtered particles was distributed in a wide range. Fig. 3 shows this distribution for sputtered plutonium and oxygen nuclei. There are some particles in the spectra with sufficiently large energy (60 keV for plutonium nuclei and 20 keV for oxygen nuclei), but nonetheless the yield of the distribution is completely determined by particles of much lower energies (hundreds of eV).

The final values of the sputtering yield and the average energy of sputtered particles are presented in Table 1. The data in the 2nd and 5th columns relates to the modeling for a sample with thickness of 150 nm on the base of 10 000 histories. Converted values for the typical plutonium dioxide material are presented in the 3rd and 6th columns. It can be seen, in particular, that alpha particles contribution is insignificant in comparison with uranium recoil nuclei and the mean energy of sputtered particles is also significantly lower.

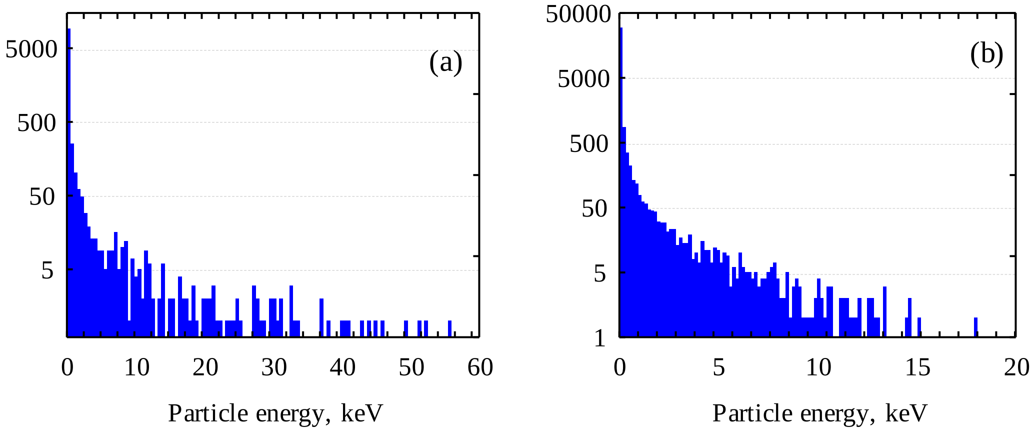

Fig. 3. Energy distribution of sputtered uranium (a) and oxygen (b) nuclei

Table 1

The main results of simulation

Sputtered particle	**Alpha particles contribution**			**Uranium recoil nuclei contribution**		
	Sputtering yield		Mean energy, eV	Sputtering yield		Mean energy, eV
	particle/decay	particle/cm^2·s		particle/decay	particle/cm^2·s	
Pu	0.0006	$2.07 \cdot 10^2$	140	0.98	$3.40 \cdot 10^5$	370
O	0.0033	$1.14 \cdot 10^3$	30	3.23	$1.11 \cdot 10^6$	120
Total	0.0039	$1.35 \cdot 10^3$		4.21	$1.45 \cdot 10^6$	

4. CONCLUSION

In this paper, by of Monte-Carlo simulation a study of the processes of spontaneous emission (sputtering) of particles from plutonium dioxide material was performed. The modeling of processes of particle transport, energy transfer and recoil cascades formation was fulfilled. It was shown that alpha particles do not contribute to the sputtering, and it is entirely determined by the influence of heavy recoil nuclei of uranium.

The final obtained values of the sputtering yield for PuO_2 material are as follows:

- $3.40 \cdot 10^5$ particles/cm^2·s for plutonium recoil nuclei,
- $1.11 \cdot 10^6$ particles/cm^2·s for oxygen recoil nuclei.

The average energy of sputtered particles:

- 370 eV for plutonium recoil nuclei,
- 120 eV for oxygen recoil nuclei.

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