MODELING OF SPONTANEOUS PLUTONIUM PARTICLES
SPUTTERING FROM PLUTONIUM DIOXIDE MATERIAL

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INTRODUCTION

Theoretical research of spontaneous emission (spattering) of radionuclide from plutonium dioxide material due to radioactive decay is performed by means of Monte-Carlo calculation. The effect under study is of considerable concern for correct radiation dose assessment for nuclear plant workers, since any radioactive material may become a permanent source of new atmospheric radioactive aerosols. Inhaled and deposited activity in human body may also produce bound-free radioactive particles that migrate independently towards different tissues and organs. The values of the sputtering yield for plutonium and oxygen particles and their energy distributions are obtained for typical plutonium dioxide samples.

METHODS

Modeling methodology based on mathematical calculation of nuclei trajectories in matter by Monte-Carlo simulation was used. This approach implies a large number of individual particle “histories” in the target material. Here, each history begins with radioactive decay of a plutonium nucleus and generation of an energetic alpha particle and uranium recoil nucleus. Decay products transfer its energy to nuclei of the matter and produce recoil cascades. When a near-surface nucleus receives the energy, that is high enough to overcome the surface binding energy, sputtering takes place. The sputtering mechanics description can be found in [1, 2]. Transport of particles in matter was calculated using SRIM-2011 software [3] — a well-known and acknowledged tool for theoretical calculations of stopping and range of energetic ions in matter. Detailed description of scientific background of this package can be found in [4]. 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).

RESULTS

Dependence of sputtering yield and decaying nucleus distance from the surface is shown in Fig. 1. Sputtering occurs mainly due to decay of atoms locating within a distance of not more than 20-30 nm from the surface, dominant contribution is formed by atoms decaying within the depth of about 2 nm.

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

Fig. 3 shows resulting energy distributions for sputtered particles. Some particles have sufficiently large energies (60 keV for plutonium nuclei and 20 keV for oxygen nuclei), though the yield is completely determined by particles of much lower energies (hundreds of eV).

Resulting values for sputtering yield and average energy of sputtered particles are presented in Table 1. The data in the 2nd and 5th columns relates to modeling for a sample with thickness of 150 nm on the base of 10000 histories. Converted values for a 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.

Table 1. The main results of simulation

<table>
<thead>
<tr>
<th>Sputtered particle</th>
<th>Alpha particles contribution</th>
<th>Uranium recoil nuclei contribution</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Sputtering yield (particle/decay, particle/cm²·s)</td>
<td>Mean energy, eV</td>
</tr>
<tr>
<td>Pu</td>
<td>0.0006, 2.07·10²</td>
<td>140</td>
</tr>
<tr>
<td>O</td>
<td>0.0033, 1.14·10³</td>
<td>30</td>
</tr>
<tr>
<td>Total</td>
<td>0.0039, 1.35·10³</td>
<td>4.21</td>
</tr>
</tbody>
</table>

Mean energy of (a) and oxygen (b) nuclei.

REFERENCES