

IN VITRO SOLUBILITY OF CHERNOBYL NUCLEAR FUEL AEROSOL WITH RESPECT TO COLLECTIVE BEHAVIOR OF ITS RADIONUCLIDES

Vladimir A. Kutkov,¹ and Olga I. Komaritskaya²

¹Russian Research Center "Kurchatov Institute", Moscow, Russia,

²State Scientific Center "Institute of Biophysics", Moscow, Russia

INTRODUCTION

The Chernobyl accident was the first one, when big groups of peoples were affected by aerosol containing poorly investigated chemical speciation of fission products – matrix-bound radionuclides. Due to uranium-oxide matrix of the spent nuclear fuel, the behavior of the nuclear fuel particle (NFP) radionuclides both in the environment and in the body acquires a collective character. Bound by the matrix, the radionuclides do not escape from the fuel particles until the matrix is chemically destroyed. If any nuclear fuel radionuclide tracer was found in the sample from environment or from human barrier organ e.g., lungs or skin, all another NFP radionuclides have to be expected to present in the sample too. Their normalized activities have to be closed to those from Kutkov et al. (1) having regard to the radioactive transformation.

Cuddihy et al. (2) give the results of a study on the radionuclide leaching rate from the particles found in late May 1986 on a wooden container which was about 80 km from Chernobyl at the time of the accident. Approximately 90% of activity were slow to leave the particles, with a leaching half-time of about 160 days and leaching constant of about $0.004 d^{-1}$. This was characteristic of all the fuel particle radionuclides including ^{131}I , ^{134}Cs and ^{137}Cs commonly have the relatively soluble chemical compounds. Such an effect of *U* substrate on leaching of radionuclides has been reported earlier by Dua et al. (3) and from our viewpoint it is the result of binding the fission products by the matrix of uranium oxides.

MATERIAL AND METHOD

The strength of a radionuclide-matrix bond in the nuclear fuel particles was studied using dialysis of an aerosol sample in Ringer's solution. Since all commonly occurring *Cs* compounds are soluble, 100% of the ^{137}Cs activity could be expected to pass into solution in a short time. However, Chernobyl on-site samples collected in June 1986 contained about 40% of aerosol *Cs* in a relatively insoluble form, probably, as part of fuel particles, the remainder being soluble (4). In 1990-1991, the "Shelter" (special entombment of damaged Chernobyl reactor) samples showed that at least 30% of *Cs* were still in a nontransportable form, as part of nuclear fuel particles and did not pass in 2 days into aqueous phase through a membrane with a pore diameter of about $0.5 \mu m$.

A special investigation of longtime dialysis kinetics for "Shelter" samples was carried out in 1992. The test procedure resembled the one described by Eidson and Griffith (5). Smears from vertical surfaces with an area of up to $100 cm^2$ were collected on ash-free aerosol filters in premises of the "Shelter". All the samples spread to contain nuclear fuel

radionuclide tracers (^{144}Ce , $^{154,155}\text{Eu}$). The activities of gamma-emitting fission products normalized to that of ^{144}Ce in the smears showed no statistically significant difference from those in the representative NFP at the time of sample measurement.

Disks of 35 mm in diameter made out of filter sections chosen for the heaviest surface contamination were fixed between two membrane filters MFA-MA-12 (Russia) with a pore diameter of about $0.5\ \mu\text{m}$, fixed between closely assembled Teflon rings and placed for dialysis into thermostatic vessels containing 150 ml of Ringer's solution at room temperature.

The leaching of radionuclides from samples into aqueous phase (dialyzate) was investigated on 2, 7, 21, 35, 49, 63, 77, 92, 106 by 122 days after onset of dialysis. Every time assembly extracted from a vessel, it was washed up by 10 ml of distilled water and examined by γ -spectrometry without disassembling. Every spent portion of dialyzate was acidulated by 5 ml concentrated nitric acid and examined with γ -spectrometry in parallel with sample examined. Then the total α -activity in dialyzate was determined. After that procedure the contaminated solution was changed for fresh one and dialysis continued.

Till the ending of experiment the assemblies were disassembled, and the filters with membranes were used for determination of total α -activity, remaining in the sample.

RESULTS

The data obtained on the levels of ^{137}Cs and total alpha emitters were used to determine a constant for the sample-solution transfer of the radionuclides. The transportability of over 70% of ^{137}Cs was found to be weak in the sample, with a leaching constant of $0.002 \pm 0.001\ d^{-1}$. The remaining Cs fraction was 20 times as transportable. For the total of alpha emitters, the dialysis constant was $0.005 \pm 0.002\ d^{-1}$. The difference between the leaching constants for ^{137}Cs and total alpha emitters is not statistically significant ($p > 0.05$). This confirms a supposition that these radionuclides have similar chemical speciation, being matrix bound. Using the summarized data on total alpha and Cs leaching, the constant for radionuclide leaching from nuclear fuel particles is estimated at $0.003 \pm 0.002\ d^{-1}$ and closed to $0.004\ d^{-1}$ from Cuddihy et al. (2).

CONCLUSIONS

It is evident that an *in vitro* experiment cannot simulate the processes of interaction between aerosol particles and intrapulmonary medium; the findings, however, evidence in favor of a hypothesis that there is an equally strong bond between the matrix of the nuclear fuel and fission products of different nature. The results are in good agreement with earlier data on the binding of fission products by U matrix from Eidson and Griffith (5) and Dua et al. (3). This led us to develop a special dosimetry model to describe the behavior of the nuclear fuel particle radionuclides in the human respiratory system (6).

It stands to reason that the NFPs are the crystals produced by fragmentation of fuel pellets. Their lattice forms a fuel matrix and strongly holds fission products and transuranium radionuclides. By analogy with uranium oxides assigned by ICRP to inhalation class Y by their transportability in the respiratory system (7), all radionuclides entering the respiratory system as a part of fuel particles should be assigned to the same inhalation class.

On penetrating the body, the matrix-bound radionuclides of the nuclear fuel particles exhibit collective behavior but in the respiratory system, the gastrointestinal tract and skin, until the bonds between them are broken due to chemical destruction of the nuclear fuel matrix that takes place in these barrier organs. Chemical destruction of aerosol particles in these organs is a fundamental process resulting in transfer of radionuclides to body fluids where they become independent.

Therefore, a model for the behavior of matrix-bound radionuclides in the body can be proposed as follows.

1. Due to the matrix, the behavior of fuel particle radionuclides in the body acquires a collective character. Matrix-bound radionuclides penetrate to "body fluids" only after a chemical destruction of the fuel matrix.
2. The matrix-bound radionuclides acquire biokinetic properties of uranium oxides, normally unusual for them, so that they should be assigned to nontransportable compounds of inhalation class Y. When fuel particles enter the gastrointestinal tract, the fractional absorption of matrix-bound radionuclides does not exceed that of uranium oxides.
3. Distribution and retention of radionuclides, penetrating into "body fluids" after NFP destruction has individual peculiarities and is determined by biokinetic properties of the related elements only.

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