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Technical Note

Spectrometry Analysis of Fumes of Mixed Nuclear Fuel $(U_{0.8}Pu_{0.2})O_2$ Samples Heated up to 2,000°C and Evaluation of Accidental Irradiation of Living Organisms by Plutonium as the Most Radiotoxic Fission Product of Mixed Nuclear Fuel

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ABSTRACT

Purpose: The purpose of this work is to describe the spectrometric analysis of gaseous cloud formation over reactor mixed uranium-and-plutonium (UP) fuel $(U_{0.8}Pu_{0.2})O_2$ samples heated to a temperature > 2,000°C, and thus forecast and evaluate radiation hazards threatening humans who cope with the consequences of any accident at a fission reactor loaded by UP mixed oxide $(U_{0.8}Pu_{0.2})O_2$, such as a mixture of 80% U and 20% Pu in weight.
Materials and methods: The UP nuclear fuel samples were heated up to a temperature of over 2,000°C in a suitable assembly (apparatus) at out-of-pile experiments' implementation, the experimental in-depth study of metabolism of active materials in living organisms by means of artificial irradiation of pigs by plutonium. Spectrometric measurements were carried out on the different exposed organs and tissues of pigs for the further estimation of human internal exposure by nuclear materials released from the core of a fission reactor fueled with UP mixed oxide.
Results: The main results of the research described are the following: (1) following the research on the influence of mixed fuel fission products (radioactive isotopes being formed during reactor operation as a result of nuclear decay of elements included into the fuel composition) on living organisms, the authors determined the quantities of plutonium dioxide (PuO_2) that penetrated into blood and lay in the pulmonary region, liver, skeleton and other tissues; and (2) experiments confirmed that the output speed of plutonium out of the basic precipitation locations is very small. On the strength of the experimental evidence, the authors suggest that the biological output of plutonium can be disregarded in the process of evaluation of the internal irradiation doses.

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Conclusion: The main results of the research are useful for further evaluation of human accidental irradiation in the case of breakdown at fission reactors working on mixed nuclear fuel, and designing medical protection measures for persons irradiated by plutonium isotopes.

Results of worldwide research on the safe use of atomic energy testify that in-pile testing of samples of different nuclear fuels (mixed fuel, fuel with low concentrations of uranium, returned fissile material, etc.) is associated with larger costs and higher hazards in comparison with out-of-pile experiments. To carry out out-of-pile experiments with nuclear fuel samples the authors have elaborated on laser heating assembly allowing the heating of targets up to a temperature of 4,500 degrees Kelvin (K) [1]. The authors' laser heating assembly was mounted at the Satpayev Kazakh National Technical University (Almaty City, Republic of Kazakhstan). The authors also obtained spectrometry results of mixed uranium-and-plutonium (UP) fuel $(U_{0.8}Pu_{0.2})O_2$ heated up to a temperature of over 4,500 K by means of the laser device.

Speculatively, both the human radiation exposure level and the quantitative and qualitative characteristics of the doses, which are absorbed by the reactor personnel and disaster-salvage crew during removal of the consequences of reactor accidents, are determined by the nuclear fuel type in a plant.

In order to determine the anticipated irradiation level of the disaster-salvage crew during the removal of the consequences of reactor accidents, it is primarily necessary to ascertain which fission products are produced during the hybrid fuel reactor operation.

The isotopic composition of emergency emissions from the reactor core with hybrid UP fuel can be forecasted on the basis of the available data on the physical quantities of radioactive materials that belong to the fuel system of one of the most powerful nuclear reactors. The initial quantities of the nuclear fuel components for the 1,000 MW power reactor are presented in Table 1 [2].

In order to artificially create hypothetical emergency conditions for the reactor core destruction, it is necessary to heat a sample of UP oxide up to the fuel temperature (above 2,000°C). UP fuel samples were heated by using a laser-beam evaporation facility, whose functional diagram is depicted in Fig. 1.

The laser beam (1, Fig. 1) is generated using a slightly ionized glow electrical discharge and carbon dioxide (CO_2) heated up to the plasma state. The beam is radiated in the infrared range at a wavelength of 10.5 μm with a power of ~200 MW. To reduce the amplitude of a pulse that is fed to the electrodes of the superficial-discharge plasma injectors, the plasma density and volume, plasma opening switch (POS; 2, Fig. 1) is used. The POS contains a current-pulse source connected to two extended electrodes, which are separated by an insulator and form together with it a vacuum interelectrode gap; a load in the form of a vacuum or plasma diode, which is also connected to the electrodes; and at least one plasma injector arranged on one of the electrodes.

To provide the opportunity of changing the angle of the laser beam focus to a nuclear fuel target sample that depends on the aims and conditions of different experiments, the beam (1, Fig. 1) passes through the shutter (3, Fig. 1), and indirectly enters the vacuum chamber (12, Fig. 1) after experiencing double reflection from the system of mirrors (8 and 11, Fig. 1). The density of the beam of refracted rays that pass through the aperture (9, Fig. 1) into the evacuated chamber (12, Fig. 1) is magnified by means of the collecting lens (10, Fig. 1). Its focal length and lens power can be changed depending on the required heating temperature of the nuclear fuel sample fixed on the turntable (7, Fig. 1). The sample is locally heated to its liquid state through the ballistic collector (6, Fig. 1) to a temperature of over 2,000°C. Owing to the rotation of the movable mirror (11, Fig. 1), the laser focus uniformly moves along a circular path on the sample surface during a laser pulse, thus allowing evaporation of considerable quantities of the liquid target material.

As a result of the evaporation of a liquid nuclear sample in the vacuum chamber (12, Fig. 1), its thermal decomposition and absorption of gases occurs. For gaseous pressure, evaporation rate, and gas permeability measurements during the nuclear fuel evaporation at a temperature of over 2,000°C, the precise vacuum balance (4, Fig. 1), equipped with a quartz spiral with a sensitivity of ~1 mg/mm, is used in the facility (the spiral deformation is registered by cathetometer with an accuracy of up to 0.01 mm). Inasmuch as Vacuum Balance 4 is fed by the current source that is connected in parallel to the

Table 1 – Initial quantities of actinides in the core of the 1,000 MW power reactor operating on the $(U_{0.8}Pu_{0.2})O_2$ hybrid uranium-and-plutonium fuel.

Isotope	Quantity in the core (kg)
^{238}U	42,300
^{237}Np	2
^{238}Pu	0 85
^{239}Pu	7,327
^{240}Pu	2,560
^{241}Pu	0 415
^{242}Pu	0 188
^{241}Am	0 11
^{243}Am	6
^{244}Cm	0.7
^{242}Cm	0.4
Total mass:	52,895

Am, americium; Cm, curium; Np, neptunium; Pu, plutonium; U, uranium.

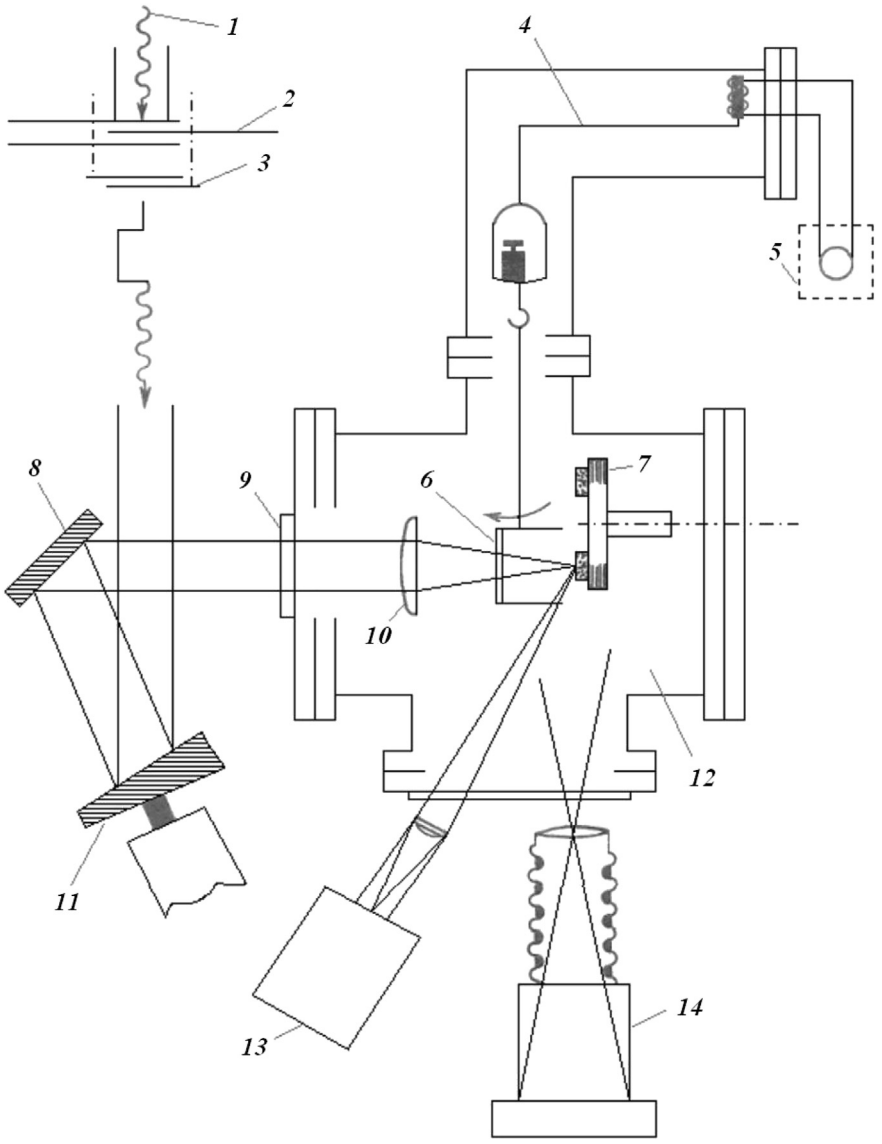

Fig. 1 – Functional diagram of the facility for laser heating of nuclear fuel samples to a temperature >2,000°C. 1, CO_2 gas laser beam; 2, plasma opening switch (POS); 3, shutter; 4, vacuum balance; 5, compensatory circuit; 6, ballistic collector with an aperture for a beam; 7, turn-table for targets; 8, stationary mirror; 9, aperture for a beam; 10, lens; 11, movable mirror; 12, vacuum chamber; 13, fast-acting micropyrometer for measuring the temperature in the focal spot and the evaporation period; 14, gas-jet observation camera.

laser generator, a circulating current arises (a reactive current with oversaturated excitation is fed to one generator, and a capacitive current is fed to the other). In order to reduce the excitation of the generator with the reactive load and increase the excitation of the generator with the capacitive load for the circulating current to become equal to zero, Compensatory Circuit 5 is used.

The purpose of experiments on heating the nuclear fuel samples up to a temperature over 2,000°C is to measure the following characteristics: (1) the evaporation period and area; (2) the mass of the generated gas; (3) the gas jet withdrawal moment, which is measured by the deflection amplitude of a pendulum collector set in Observation Camera 14 (Fig. 1); and (4) spectral incandescence of the evaporation surface, which is measured by fast-acting Micropyrometer 13 in order to identify the fuel-sample evaporation temperature by measuring its spectral emissivity.

If it is necessary to determine not the thermal radiation power but the radionuclide composition of the gas that forms as a result of the evaporation of a nuclear fuel sample, a spectrometer can be used instead of Micropyrometer 13.

This plant differs from its analogs in that before the laser beam enters the evacuated chamber, it is passed through a system of two mirrors, one being movable, and a converging lens. This allows one to heat nuclear fuel samples more uniformly, thus in turn, reducing the error in the data of thermal equilibrium measurements, which are based on the determination of the target-material evaporation rate.

Laser-beam heating to a temperature higher than 2,000°C is the most acceptable method for evaporating samples. It

allows one to obtain data on the thermal equilibrium via determination of the target-material evaporation rate and the vapor pressure.

An electron beam must not be used for heating samples because its interaction with the gaseous cloud above the target influences both the evaporation rate and the reactionary pressure of the gaseous cloud, thus distorting the temperature measurement results. Moreover, the deep penetration of the electron beam will cause indeterminate explosive evaporation.

The sample heating temperature was chosen on the basis of the fact that nuclear-fuel fission products at temperatures below the melting point (~1,800°C) and the irradiated fuel itself at a temperature over 2,000°C [2], play the key role in the gas-pressure increase in irradiated nuclear fuel, which leads to the destruction of fuel slugs and assemblages.

Experiments with the UP oxide samples under close-to-emergency conditions were performed in order to determine the isotopic composition of hybrid-fuel evaporations.

Spectrometry of both the vapors of $(U_{0.8}Pu_{0.2})O_2$ samples and the biological samples has been carried out by means of measuring complex "Progress-2000" (software and equipment itself are manufactured by the Limited Liability Company ≪Scientific and Production Enterprise "Doza"≫, Moscow city, town of Zelenograd, St George's Avenue, Building 6, 124498, Russian Federation).

The complex is a set of measurement (spectrometry or radiometric) tracks, united by a common software, that allows the independent control of all the tracks and process measurements by means of a single PC.

"Progress-2000" provides an automatic account of the density of countable sample matter, monitoring the health of the spectrometer and the stability of metrological characteristics, calculation of uncertainty of measurement results and light emitting diode (LED) stabilization, to guarantee a high temperature stability of the measuring track. The basic measurement error is not more than 30%.

A semiconductor gamma-spectrometer, "Progress-gamma", was used to determine the content of gamma-emitting radionuclides both in biological samples and vapors of $(U_{0.8}Pu_{0.2})O_2$. The scintillation detection units are equipped with a crystal NaI (Tl; Ø63 mm in diameter), integrated power amplification and analog-to-digital converter. Relative energy resolution for the by ^{137}Cs (662 keV) line is not more than 8.5%. Minimum measured activity in the counting sample (by ^{137}Cs) is 3 Becquerel (Bq).

A beta-scintillation spectrometer, "Progress-beta", was used for measuring the specific activities of strontium radionuclides, as well as total beta-activity in biological samples and vapors of $(U_{0.8}Pu_{0.2})O_2$. Measurements of activities of strontium radionuclides in biological samples did not require the special sample preparation due to the use of procedures of accelerated radiochemical concentration. The scintillation unit has a plastic scintillation detector (Ø70 mm in diameter). Minimum activity (by ^{90}Sr) of the counting sample of a mass of 10 g, being measured in a standard cuvette for 1 hour, is 0.1 Bq.

An alpha-spectrometer, "Progress-AR", was used for determining alpha-emitters in biosubstrates after its radiochemical separation and in fumes of samples $(U_{0.8}Pu_{0.2})O_2$, which were obtained by electrolytic deposition (the total alpha-activity deposited on an aerosol filter). The scintillation detector unit is equipped on the basis of ZnS crystal (Ø60 mm in diameter). The measurement range of activities varies between 9×10^{-3} Bq per sample up to 180 Bq/kg.

The results of the spectrometric analysis of $(U_{0.8}Pu_{0.2})O_2$ evaporations at 2,000°C are listed in Table 2.

Table 2 contains the measured volumetric activities of nuclides obtained after processing direct spectrometer readings (measured in Bq per sample). Since the total mass of $(U_{0.8}Pu_{0.2})O_2$ samples was 1 kg, the volumetric activities of radionuclides were derived taking the density of air ($\rho = 1.225$ kg/m^3) into account.

The expected activity A of $(U_{0.8}Pu_{0.2})O_2$ fission products in the 1,000 MW power reactor core with a 50-ton fuel charge can be calculated from the formula:[1]

$$A = 3.1 \times 10^{13} P\, A(i) \times \left(1 - e^{-\lambda(i) T_{eff}}\right). \quad (1)$$

Here P, MW, is the reactor power; $A(i)$, Bq, is the measured volumetric activity of the i-th nuclide; $\lambda(i)$, s^{-1} is the decay constant of the i-th nuclide; 3.1×10^{13} Bq MW is the conversion factor; and T_{eff}, s, is the effective operational time of the reactor, equivalent to the fuel heating period:

$$T_{eff} = B \times M/(1.4 \times 10^{-8}\ W), \quad (2)$$

where B, MW days/t, is the $(U_{0.8}Pu_{0.2})O_2$ hybrid-fuel average burn-up in the core ($B = 50{,}000$ MW days/t for 1,000 MW power reactor); M, t, is the $(U_{0.8}Pu_{0.2})O_2$ mass in the core; and W, MW, is the reactor power.

The anticipated activities of radioactive nuclides in the $(U_{0.8}Pu_{0.2})O_2$ fuel fumes at temperatures > 2,000 °C, which are calculated by the above technique, are presented in Table 3.

Based on the above results, one can speculatively estimate the total activity of emergency emissions from the core of the 1,000 MW power reactor with the $(U_{0.8}Pu_{0.2})O_2$ hybrid UP fuel. It amounts to ~270×10^{15} Bq.

The analysis of the data from Tables 2 and 3 shows that the radioisotope composition of the emergency emissions from the hybrid-fuel reactor core is distinguished by the high content of PuO_2 stable compound and large amounts of long-lived pure plutonium (^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, and ^{242}Pu), which is the most radiotoxic in comparison with all other anticipated emission components.

The obtained results make it possible for a rough forecast to be made of the isotopic composition of accidental emissions from the core of a fission reactor of 1,000 MW power operating on hybrid fuel $(U_{0.8}Pu_{0.2})O_2$.

When calculating radiation doses as a result of leakage of fission products from a mixed fuel reactor core, it is necessary to take into account complex, diverse, and interrelated factors related to plutonium behavior. For calculations, it is necessary to have data or to make appropriate assumptions on exposure conditions, including the composition of radioactive products, particle sizes, concentration of activity, and exposure time.

To assess the degree of plutonium retention in human tissues, organs and their systems, experiments on pigs were

[1] The calculation technique entitled "Estimating the Radiation Consequences of Accidents at Research Reactors" was granted by the Kurchatov Institute of Atomic Energy.

Table 2 – The volumetric activities of radionuclides contained in the $(U_{0.8}Pu_{0.2})O_2$ fuel fumes at 2,000°C.

Nuclide	Half-life (hr)	PVA (Bq/m^3) [3]	Measured volumetric activity of a nuclide (10^{12} Bq/m^3)	Activity of a nuclide, percentage of the total activity (%)
^{133}Xe	1.27×10^2	1.60×10^5	1.6	0.16
^{135}Xe	9.20×10^1	5.18×10^3	1.33	0.14
^{138}Xe	2.33×10^{-1}	7.77×10^3	1.33	0.14
^{85m}Kr	4.50×10^1	2.15×10^3	0.32	0.03
^{87}Kr	1.26×10^1	1.48×10^4	0.53	0.05
^{88}Kr	2.85×10^1	3.00×10^3	0.73	0.07
^{89}Kr	5.00×10^{-2}	1.11×10^3	1.01	0.1
^{238}Pu	7.68×10^5	5.30×10^{-1}	0.002	0.0003
^{239}Pu	2.11×10^8	5.30×10^{-1}	0.024	0.002
^{240}Pu	5.73×10^7	5.30×10^{-1}	0.008	0.0009
^{241}Pu	1.26×10^5	5.00×10^1	0.013	0.001
^{242}Pu	3.29×10^9	5.70×10^{-1}	0.007	0.0007
^{132}Te	7.80×10^1	4.00×10^1	0.001	0.0001
^{131}I	1.93×10^2	7.30	0.003	0.0003
^{132}I	2.30×10^1	1.48×10^2	0.004	0.0004
^{133}I	2.10×10^2	2.04×10^1	0.001	0
^{134}I	8.66×10^{-1}	2.96×10^2	0.001	0
^{135}I	6.60×10^1	6.66×10^1	0.001	0
^{90}Sr	9.50×10^1	3.18×10^2	0.001	0.0001
^{140}Ba	3.12×10^2	2.20×10^1	0.001	0.0001
^{57}Co	6.50×10^3	8.50×10^3	0.009	0.0009
^{65}Zn	5.86×10^3	2.80×10^3	0.004	0.0004
^{147}Nd	2.64×10^2	4.00×10^3	0.004	0.0004
^{147}Pm	2.29×10^4	1.70×10^3	0.003	0.0003
^{145}Sm	2.98×10^6	5.30×10^3	0.007	0.0007
^{235}U	6.16×10^{12}	2.90	0.019	0.002
^{238}U	3.92×10^{13}	2.40	0.019	0.002
^{237}Np	1.87×10^{10}	3.80×10^{-1}	0.001	0.0001
^{242}Cm	1.43×10^6	1.7	0.002	0.0002
Total volumetric activity			6.985×10^{12} Bq	

Ba, barium; Cm, curium; Co, cobalt; I, iodine; Kr, krypton; Nd, neodymium; Np, neptunium; Pm, promethium; Pu, plutonium; PVA, permissible volumetric activity; Sm, samarium; Sr, strontium; Te, tellurium; U, uranium; Xe, xenon; Zn, zinc.

conducted because the human body has more similarities with the pig body than with other land mammals. According to Järvinen and Parviainen [4], the blood, skin, and internal organs of humans and pigs share a number of similar properties, as noted in Table 4.

Thus, the pig is one of the best models in biomedical research for studying human cardiovascular diseases, skin diseases, and diseases of the nervous system and digestive tract. Pigs are used for studying the impact of not only radiation but also alcohol, drugs, and medicines.

In-vivo experiments were conducted with 32 pigs. The authors investigated three general ways of plutonium arrival into a pig's body: through the respiratory tract, through the digestive system, and as a result of precipitation on the carcass surface. To quantify the excretion of plutonium from the living body, 12 experimental animals exposed to plutonium contained in inhaled air, feed, and injected solution, were sacrificed 4 years after the initial experiments were conducted with all 32 pigs.

For inhalation of plutonium by pigs, the experimental animals were placed in an airtight plexiglass chamber (15, Fig. 2), where plutonium was pumped through plenum ventilation (16, Fig. 2). The continuous inhalation period of the plutonium was 2 hours on average, depending on the respiration rate of each experimental animal. The volume of air inhaled by the pigs (with a single breath) was $0.4–0.5 \times 10^{-3}$ m^3, and respiration rate ranged between 15 breaths/min and 20 breaths/min. Therefore, within 2 hours, about 1 m^3 of air containing plutonium penetrated into each pig's body, as required to meet the conditions of spectrometric studies of the samples.

In the course of experiments on laser-induced heating of the samples of mixed fuel $(U_{0.8}Pu_{0.2})O_2$ with a total mass of 1 kg to a temperature above 2,000 °C, it was found that the total volumetric activity of plutonium contained in the resulting evaporations was 54 PBq/m^3 (Table 2). Thus, 54 PBq of plutonium was inhaled by pigs within 2 hours.

The authors experimentally ascertained that the excretion rate of plutonium out of the organism's general precipitation points is very small. Therefore the biological decay could be ignored when the internal irradiation dose was being calculated. Interventions which induce the excretion of plutonium are of medical and therapeutic interest. In order to elaborate a course of therapy for the excretion of radiotoxic plutonium out of an organism, one needs to calculate the radiation exposures of the general points of isotopes' accumulation.

Ingestion of plutonium by a pig body was achieved by feeding experimental animals with grain forage previously placed in Vacuum Chamber 12 over the sample of nuclear fuel $(U_{0.8}Pu_{0.2})O_2$ and irradiated by evaporations containing isotopes of plutonium ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, and ^{242}Pu. The specific activity of ^{239}Pu in irradiated feed was 12 PBq/kg (12×10^{15} Bq/kg); levels of content of other plutonium isotopes

Table 3 – Calculated activities of radioactive nuclides anticipated in the $(U_{0.8}Pu_{0.2})O_2$ fuel fumes at 2,000 °C in the 1,000 MW power reactor core with a 50,000 kg fuel charge.

Nuclide	Half-life (hr)	Calculated activity of nuclide (10^{12} Bq)
^{133}Xe	1.27×10^{2}	6.20×10^{4}
^{135}Xe	9.20×10^{1}	5.15×10^{4}
^{138}Xe	2.33×10^{-1}	5.15×10^{4}
^{85m}Kr	4.50×10^{1}	1.24×10^{4}
^{87}Kr	1.26×10^{1}	2.06×10^{4}
^{88}Kr	2.85×10^{1}	2.83×10^{4}
^{89}Kr	5.00×10^{-2}	3.91×10^{4}
^{238}Pu	7.68×10^{5}	1.07×10^{2}
^{239}Pu	2.11×10^{8}	9.28×10^{2}
^{240}Pu	5.73×10^{7}	3.24×10^{2}
^{241}Pu	1.26×10^{5}	4.95×10^{2}
^{242}Pu	3.29×10^{9}	2.56×10^{2}
^{132}Te	7.80×10^{1}	5.10×10^{1}
^{131}I	1.93×10^{2}	1.13×10^{2}
^{132}I	2.30×10^{1}	1.70×10^{2}
^{133}I	2.10×10^{2}	3.10×10^{1}
^{134}I	8.66×10^{-1}	3.10×10^{1}
^{135}I	6.60×10^{1}	2.60×10^{1}
^{90}Sr	9.50×10^{1}	4.60×10^{1}
^{140}Ba	3.12×10^{2}	5.15×10^{1}
^{57}Co	6.50×10^{3}	3.52×10^{2}
^{65}Zn	5.86×10^{3}	1.39×10^{2}
^{147}Nd	2.64×10^{2}	1.70×10^{2}
^{147}Pm	2.29×10^{4}	1.17×10^{2}
^{145}Sm	2.98×10^{6}	2.80×10^{2}
^{235}U	6.16×10^{12}	7.38×10^{2}
^{238}U	3.92×10^{13}	7.38×10^{2}
^{237}Np	1.87×10^{10}	5.15×10^{1}
^{242}Cm	1.43×10^{6}	7.00×10^{1}
Total activity		270.634×10^{15} Bq

Ba, barium; Cm, curium; Co, cobalt; I, iodine; Kr, krypton; Nd, neodymium; Np, neptunium; Pm, promethium; Pu, plutonium; Sm, samarium; Sr, strontium; Te, tellurium; U, uranium; Xe, xenon; Zn, zinc.

did not exceed the level of spectrometer sensitivity. The feed dosage of each pig equal to 1 kg provided a single ingestion of 12 PBq of ^{239}Pu in the body of each experimental animal.

Because the skin of experimental pigs showed no damage, artificial infliction of which is prohibited by the European Convention for the Protection of Vertebrate Animals used for Experimental and other Scientific Purposes (ETS #123, Strasbourg, March 18, 1986), contact administration of plutonium in pigs was conducted by the intramuscular injection of a physical solution in a volume of 20 mL with ^{239}Pu concentration equal to 1 g/mL.

Results of experiments which had been conducted on samples of mixed nuclear fuel by means of laser-beam heating assembly have shown that ^{239}Pu forms the prevailing contribution into the total activity being measured in evaporations under uranium-and-plutonium dioxide $(U_{0.8}Pu_{0.2})O_2$ [5]. Results of experiments on pigs showed that plutonium generally penetrates an organism through the respiratory tract.

At the present time, exposure doses of internal organs and bones from the particles of any radioactive substance penetrated into the body is calculated based on the assumption that the radiated energy is evenly distributed throughout the organ tissue [6]. The average value of the dose for the whole organ is taken as the resulting value.

Plutonium dioxide emits alpha particles with energy of 5.14 MeV and the free path in the tissue of < 40 microns, the recoil atoms with average energy of ~0.084 MeV and a low-energy X-ray and gamma-ray radiation characterized by the energy of < 0.01 MeV for decay. It is considered that any organ absorbs all alpha particles and recoil atoms, and a portion of X-ray and gamma-radiation [7].

Effective radiation energy ($E_{eff.}$) is calculated for each particular radioisotope in a separate organ. In the case of uniform deposition of radioactive substances in each of several organs, the absorbed dose can be calculated. For convenience of calculations, obtained values of the effective energy are shown in Table 5 [8].

However, radiation loads from the particles of radioactive substance that penetrated into the body should be evaluated based on the equivalent (not absorbed) dose, because, firstly, various types of radiation have different characteristics, and the functional role and sensitivity of each organ varies. Secondly, in most cases radioactive substances are unevenly distributed in the tissue.

For radiation protection, equivalent dose (D_{eqv}, μSv) is defined as the product of the absorbed dose (D_{abs}, μGy), radiation weighting factor (WR) and coefficient of dose distribution (CD) [9]:

$$D_{eqv} = D_{abs} \times WR \times CD \tag{3}$$

Equivalent dose of 1 Bq/h is determined by the following equation [9]:

$$\frac{D_{eqv}}{\text{Bq} \times \text{h}} = 51.2\frac{E_{eff}}{m} \tag{4}$$

where $E_{eff.}$, MeV, is the effective radiation energy; and m, g is the mass of the organ.

The coefficients given in Table 1 were used to determine the effective energies of other components of mixed nuclear fuel. The effective energies for each of the fission products of $(U_{0.8}Pu_{0.2})O_2$ [10] in the lungs, bone tissue, liver, and ovaries are given in Table 6.

In laboratory conditions of exposure, it was found that some fraction of plutonium penetrating with air (f_a) is deposited directly in the lungs, wherein 40% of the initial amount of deposited plutonium disappears with an effective half-life of 1 day (24 hr) and the remaining 60% with the effective half-life of 500 days (12,000 hr). Thus, intake of 1 Bq of plutonium leads to irradiation by dose I [11,12]:

$$I = f_a \times \left(\frac{40\% \times 24h}{0.693} + \frac{60\% \times 12{,}000h}{0.693}\right) = 10{,}404 \times f_a,\ \text{Bq/h}, \tag{5}$$

where f_a is the activity of plutonium that entered into a pig's body with inhaled air, Bq.

The values of internal exposure doses of humans largely depend on the mass of its organs and respiration rate, which in turn, are subject to age-related changes. Radiation doses from plutonium on the internal organs, accumulated 70 years of age depending on the time of ^{239}Pu penetration in the body can be calculated using the equation [11,12]:

Table 4 – Similarities and discrepancies of humans and pigs.

Comparative characteristics	Similarities	Discrepancies
Genetic characteristics	Genetic disorders & dysfunction of proteins, similar diseases: Alzheimer's disease, Parkinson's disease, & obesity.	The proximity of the genomes of humans & pigs is < 90%. Nucleotide texts are different because of substitutions of the Single Nucleotide Polymorphism (SNP), the Copy Number Variation (CNV), & the order of arrangement or orientation of chromosome fragments.
Anatomical characteristics	While in the womb, the embryo of pig has 5-toed arms & a snout similar to a primate muzzle. Apparatus dentition, morphology, & physiology of kidneys, eyes structure, the digestive system, the anatomy & physiology of circulatory system (blood vessels, especially arteries, levels of hemoglobin & proteins, erythrocyte size), the structure of the skin (pigs are the only pets which can sunbathe).	Pigs' hearts weighs 320 g; the weight of human heart is 300 g; lungs mass of a pig is 800 g, human's lungs are 790 g; kidneys' mass: 260 g (pig) & 280 g (human); liver: 1,600 g (pig) & 1,800 g (human).
Physiological characteristics	(1) Insulin for diabetes treatment is extracted from the pancreas of pigs. (2) Pigs liver, kidneys, spleen, & even heart are used for transplantation to human. (3) Sows are used as a surrogate mother for gestation of human embryos.	(1) Porcine differs from human insulin by only 1 amino acid. (2) An obstacle to transplanting pig organs to human was the problem of tissue incompatibility.[a] (3) Pig gestational age is shorter than human; the head of a human baby is larger than a pig's parturient canal; there is a risk of miscarriage.[b]

[a] To solve the problem of transplant rejection, transgenic pigs are incubated. Two human genes are introduced into the germ cells of a pig, and one pig's gene is switched off. The close similarity of human and pig cells allows the growth of tissues of pig's organs that would be suitable for humans.

[b] Healthy female pigs are crossed with male pigs. Then, a week later one pig embryo is removed from uterus under general anesthesia; another pig embryo is left in the uterus. After that, a fertilized human egg is inserted into the uterus of the pig. The piglet, growing along with its human baby brother, is necessary to avoid miscarriage in the early stages of the pig's pregnancy. Pigs are treated by special medication to prevent labor onset, since the gestation period of swine is shorter. At the end of the ninth months delivery occurs by cesarean section: the head of the child is more than pig's birth canal, and the newborn human cannot be born naturally. Mild jaundice is the only trouble that can accompany the birth of human babies. This situation is similar to rhesus conflict between mother and child during normal pregnancy. It is caused by the destruction of pigs' blood cells in the body of the human baby and the start of production of their own red blood cells in the bone marrow.

$$D_{1,2} = K \times \left[\frac{t_2 - t_1}{\ln m_2 - \ln m_1} \times \left(\frac{1}{m_1} - \frac{1}{m_2}\right)\right] \mu Sv/Bq, \quad (6)$$

where $D_{1,2}$ is the dose of multiple human exposures; K is correction factor depending on the organ and tissue: $K = 47.36 \times 10^5$ for lungs [11,12]; $K = 5.30 \times 10^5$ for bone stock [11]; $K = 1.39 \times 10^5$ for liver [11].

Calculation results are given in Table 7.

Besides ^{239}Pu, there are other plutonium isotopes in the mixed fuel of a nuclear reactor with relative amounts depending on the nature of fuel rod use. If we consider the effects of plutonium on humans apart from all other radioisotopes, radiation doses can be calculated on the basis of the data given in Table 8.

If the share of inhaled plutonium (f_a) is not determined, radiation doses from plutonium deposited in the lungs can be determined using the following equation [13]:

$$I = \frac{1}{0.693}\left\{0.4 \times \frac{24 \times T_{1/2}}{T_{1/2} + 24} \times \left[1 - e^{-0.693 \times t}\left(\frac{T_{1/2} + 24}{24 \times T_{1/2}}\right)\right] + 0.6 \times \frac{12,000 \times T_{1/2}}{T_{1/2} + 12,000} \times \left[1 - e^{-0.693 \times t} \times \left(\frac{T_{1/2} + 12,000}{12,000 \times T_{1/2}}\right)\right]\right\} \quad (7)$$

where $t = 50 \times 365 \times 24 = 438,000$ hours, $T_{1/2}$, hours, is half-life.

Thus, plutonium radiation exposure by particles in lungs will amount 10,389.46 Bq/h per Bq of accumulated plutonium:

$$I = 1.443 \times [9.5999 + 7,190.3052] = 10,389.46 \frac{Bq \times h}{Bq_{Pu}} \quad (8)$$

However, only 2.5% of the total activity of plutonium isotopic mixture falls on ^{239}Pu, so the contribution of ^{239}Pu to the total dose will be 259.74 Bq (^{239}Pu)/h per Bq of mix of isotopes accumulated in the lungs [9]:

$$I = 0.025 \times 10,389.46 = 259.74 \frac{Bq_{^{239}Pu} \times h}{Bq_{mix}}. \quad (9)$$

In this case the daily dose from each Bq of ^{239}Pu deposited in the lungs will be 3.82 μSv [9]:

$$\frac{CD}{Bq_{^{239}Pu}} = 51.2 \times \frac{E_{eff}}{m} = 51.2 \times \frac{57}{764} = 3.82 \frac{\mu Sv}{Bq \times h}, \quad (10)$$

and the accumulated dose (AD) due to ^{239}Pu will be almost 1 mSv per Bq of isotopic mixture deposited in the lungs [9]:

$$AD_{^{239}Pu} = 259.74 \times 3.82 = 992.19 \frac{\mu Sv}{Bq} = 0.992 \frac{mSv}{Bq}. \quad (11)$$

Table 9 shows predicted values of radiation doses to the lungs, bone tissue, liver and ovaries of people exposed to radiation at 20 years of age, depending on the activity of isotopic mixture deposited in the lungs, at the end of 50 years.

Data on the total radiation dose to the lungs, skeleton, liver, and ovaries from an isotopic mixture of ^{238}Pu, ^{239}Pu,

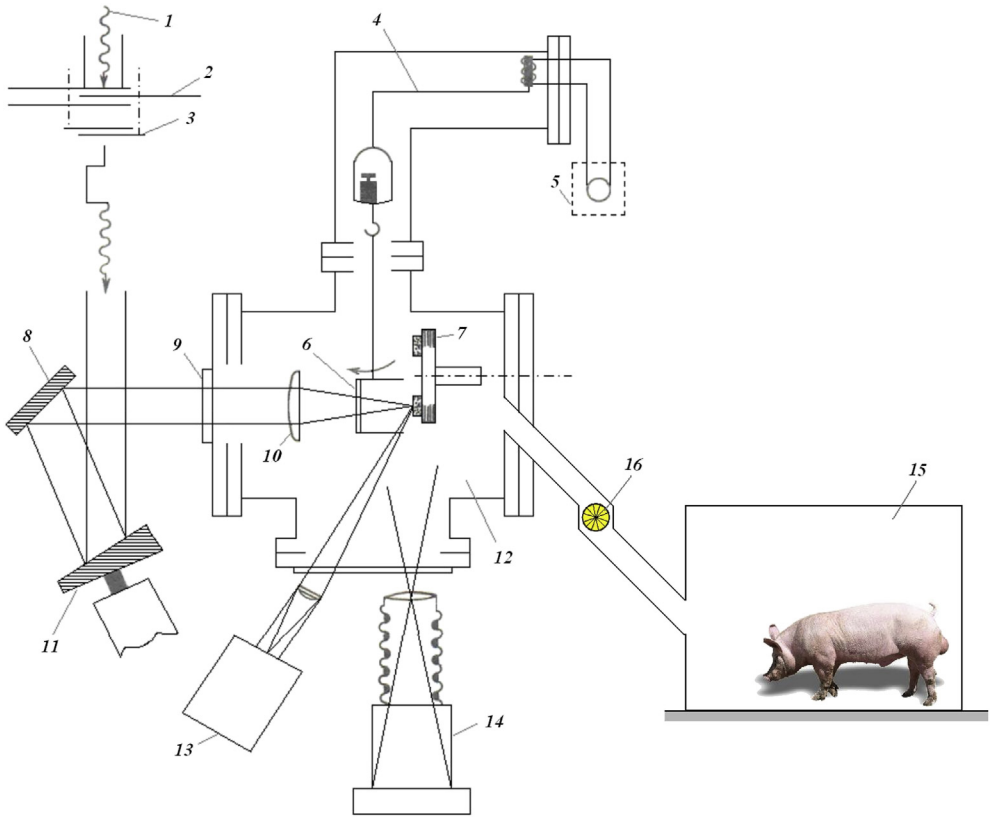

Fig. 2 – Schematic diagram of the installation for laser-induced heating of the samples of nuclear fuel to a temperature >2,000°C. 1, CO_2 gas laser beam; 2, plasma opening switch (POS); 3, shutter; 4, vacuum balance; 5, compensatory circuit; 6, ballistic collector with an aperture for a beam; 7, turn-table for targets; 8, stationary mirror; 9, aperture for a beam; 10, lens; 11, movable mirror; 12, vacuum chamber; 13, fast-acting micropyrometer for measuring the temperature in the focal spot and the evaporation period; 14, gas-jet observation camera; 15, plexiglass chamber for experimental animals; 16, fan providing the influx of evaporations from Chamber 12 into Chamber 15.

^{240}Pu, ^{241}Pu, and ^{242}Pu accumulated in the lungs is given in Table 10.

Taking the reference value of specific activity of plutonium isotopic mixture in biological tissue equal to 17.113 TBq/kg (17.113×10^{12} Bq/kg) [14], and using the formula we can calculate the total radiation dose to lungs, skeleton, liver, and ovaries of a human in relation to the mass, and not to the activity of isotopic mixture accumulated in the lungs. Eq. (12) connects the mass of any radionuclide with its activity value [15]:

$$m = b \times Z \times T_{1/2} \times A \quad (12)$$

where m, g, is the mass of radionuclide; Z is the atomic mass of radionuclide; A, Bq, is the activity of radionuclide; $T_{1/2}$ is the half-life of radionuclide; and b is the factor depending on the measurement unit of half-life: $b = 8.40 \times 10^{-24}$ if $T_{1/2}$ is measured by seconds, $b = 1.44 \times 10^{-22}$ if $T_{1/2}$ is measured by minutes, $b = 8.62 \times 10^{-21}$ if $T_{1/2}$ is measured by hours, $b = 2.07 \times 10^{-19}$ if $T_{1/2}$ is measured by days, $b = 7.56 \times 10^{-17}$ if $T_{1/2}$ is measured by years.

Table 5 – The values of the effective energy of different types of radiation emitted by ^{239}Pu [8].

Radiation type	Energy E (MeV/decay)	Weighting factor (WR)	$E_{eff.}$ (MeV)		
			Bone tissue	Gastroenteric tract	Other organs
Alpha-radiation	5.14	10	257	0.51	51.4
Knock-on atoms	0.084	20	8.4	0.01	1.68
X-ray & gamma radiation	0.0085	1	0.04		
Total			270	0.52	53.0

E_{eff}, effective radiation energy.

Table 6 – The effective energies of various components of mixed fuel $(U_{0.8}Pu_{0.2})O_2$ [10] contained in the lungs, bone tissue, liver, and ovaries.

Radioisotope	Radioisotope activity portion (Bq/Bq of isotope mix)	$E_{eff.}$ (MeV)			
		Lungs	Bone tissue	Liver	Ovaries
^{238}U	1.9×10^{-2}	43	220	43	0.43
^{237}Np	1.36×10^{-4}	49	250	49	0.49
^{238}Pu	2.83×10^{-3}	53	270	53	0.53
^{239}Pu	2.46×10^{-2}	57	284	57	0.57
^{240}Pu	8.58×10^{-3}	53	270	53	0.53
^{241}Pu	1.3×10^{-2}	0.53	14	1.0	0.02
^{242}Pu	6.78×10^{-3}	51	250	51	0.51
^{241}Am	5.89×10^{-4}	57	283	57	0.57
^{243}Am	3.31×10^{-4}	54	272	54	0.54
^{242}Cm	1.84×10^{-3}	64	400	78	0.78
^{244}Cm	5.89×10^{-4}	60	300	60	0.60

Am, americium; Cm, curium; E_{eff}, effective radiation energy; Np, neptunium; Pu, plutonium; U, uranium.

According to the results of calculations conducted by means of the above equation, the activities of plutonium isotopes correspond to the following values of its mass:

1 Bq of $^{238}Pu \equiv 1.58 \times 10^{-12}$ g;
1 Bq of $^{239}Pu \equiv 4.35 \times 10^{-10}$ g;
1 Bq of $^{240}Pu \equiv 1.19 \times 10^{-10}$ g;
1 Bq of $^{241}Pu \equiv 2.62 \times 10^{-13}$ g;
1 Bq of $^{242}Pu \equiv 6.88 \times 10^{-19}$ g;
Total mass $= 5.56 \times 10^{-10}$ g.

The results of calculations of the total radiation dose to lungs, bony skeleton, liver, and ovaries of a human in relation to the mass of plutonium isotopic mixture accumulated in the lungs are given in Table 11.

The data in Tables 9–11 show that plutonium absorption from the lungs causes the deposition of 31% of the amount penetrated into the body in the lungs, 30% in the ovaries, 17% in the liver, 15% in the skeleton, and 7% in other tissues.

A long-term forecast of internal exposure by a mixture of plutonium isotopes is made taking into account a single inhalation of the particles in the body and their subsequent distribution to individual organs. The dose per unit activity of ^{239}Pu (Bq) deposited in the bone tissue, liver and ovaries was 3 mSv. The entire range of plutonium isotopes formed a radiation dose equal to 4.89 mSv/Bq of deposited activity. Predicted radiation doses from a kilogram of plutonium isotopic mixture deposited in the lungs expected at the end of 50 years after accidental exposure of a person should be 15.7 mSv for the lungs; 7.42 mSv for the bone tissue; 8.28 mSv for the liver; and 15.13 mSv for the ovaries.

Thus, human internal plutonium exposure doses contained in evaporations of mixed nuclear fuel $(U_{0.8}Pu_{0.2})O_2$ with a mass of 1 kg, heated to a temperature above 2,000 °C would not exceed permissible limits of radiation safety standards for professional exposure [3]. However, the fuel element column of a nuclear power reactor core can exceed 50 tons; in case of accidental exposure to nuclear fuel fission products of such mass, radiation doses for the human body would increase several thousand times.

Conducted investigations allow one to make the following conclusions:

(1) Internal exposure of plutonium to humans depends on the physical, chemical, and biological properties of the extracted material, local environment, peculiarities of the organism, and the nature of emission.
(2) Since uptake by inhalation, orally, and by contact (as a result of precipitation on the carcass surface) are the general ways of radioactivity penetration into a living organism, the authors investigated the absorption of PuO_2 through the respiratory tract, through the digestive tract, and through deposits on the body's surface.
(3) Only small amounts of plutonium get in the blood by inhalation. Low solubility of plutonium in fluids covering the respiratory tract leads to its deposition and rapid release from nasopharyngeal and tracheobronchial regions. Nevertheless, prolonged retention of the particles in the lung region leads to absorption of approximately 5% of PuO_2 in the blood; about 15% of the deposits in the pulmonary region transferred to pulmonary lymph nodes eventually reach the circulatory system. Thus, approximately 20% of the particles deposited in the pulmonary region after a while will be transferred into other organs with the blood or circulating lymph.
(4) When assessing the internal exposure of pigs after contact with plutonium, the most vulnerable external organs—eyes—were considered. The rate of deposition of plutonium particles on the surface under normal conditions is about 5×10^{-3} m/s, and the area of the pig eye is about 4×10^{-4} m^2. Thus, the total plutonium absorption by the pig eyes was twice integrated exposure

Table 7 – Radiation doses of human lungs, bone tissue and liver expected by the age of 70, depending on the time of plutonium-239 penetration in the body.

Age when activity has penetrated into the body	Proposed mass (g)			Average respiration rate (m^3/hr)	Radiation exposure (μSv/Bq of accumulated activity)		
	Lungs	Bones	Liver		In lungs	In bones	In liver
New-born	66	500	100	0.033	0.03	0.09	0.15
1 yr	170	2,000	250	0.16	0.02	0.09	0.13
10 yr	450	5,900	844	0.62	0.009	0.07	0.09
20 yr (female)	764	6,800	1,450	0.88	0.006	0.06	0.07
20 yr (male)	955	10,000	1,810	0.95	0.005	0.04	0.06

Table 8 – Expected composition of plutonium isotopic mixture in the fuel.

Radioisotopes	Share of activity[a] (Bq / Bq of isotopes mix)	Half-life (hr)	Dose per unit of activity, deposited in the bone tissue, liver, & ovaries (μSv/Bq of accumulated activity)
^{238}Pu	0.0028	7.9×10^5	0.06
^{239}Pu	0.025	2.1×10^8	0.07
^{240}Pu	0.0085	5.8×10^7	0.07
^{241}Pu	0.013	1.1×10^5	0.02
^{242}Pu	0.0068	3.4×10^9	0.07
Total			0.29

Pu, plutonium.
[a] Specific activity is 17.113×10^{12} Bq/kg of plutonium isotopic mixture [2].

Table 9 – Radiation doses to the lungs, bone tissue, liver, and ovaries depending on the activity of isotopic mixture accumulated in the lungs (unit of measurement–mSv/Bq of isotopic mixture deposited in the lungs).

Radioisotope	Radiation dose to the lungs	Radiation dose to the bone tissue	Radiation dose to the liver	Radiation dose to the ovaries
^{238}Pu	0.103	0.059	0.054	0.098
^{239}Pu	0.992	0.555	0.522	0.947
^{240}Pu	0.313	0.018	0.165	0.299
^{241}Pu	0.005	0.014	0.005	0.017
^{242}Pu	0.241	0.132	0.127	0.230
Total	1.65	0.78	0.87	1.59

Pu, plutonium.

Table 10 – Total radiation doses to the lungs, bone tissue, liver, and ovaries depending on the activity of the isotopic mixture accumulated in the lungs (units–mSv/Bq of isotopes mix accumulated in lungs).

	Mass (g)	Deposited dose of plutonium penetrated into the body by inhalation (mSv/Bq of isotopic mixture deposited in the lungs)	Total radiation dose, mSv/Bq of isotopic mixture deposited in the lungs
Lungs	764	–	1.65
Bone tissue	6,800	0.39	0.78
Liver	1,450	0.59	0.87
Ovaries	8	0.00093	1.59

Table 11 – Total radiation doses to the lungs, bone tissue, liver, and ovaries depending on the activity of isotopic mixture accumulated in the lungs (units, mSv / kg of isotopes mix accumulated in the lungs).

	Mass (g)	Total radiation dose (mSv/kg of isotopic mixture deposited in the lungs)
Lungs	764	15.70
Bone tissue	6,800	7.42
Liver	1,450	8.28
Ovaries	8	15.13

(Bq/[s × m^2]). However, it is known that the greatest danger for the living organism is contact introduction of radioactive substances through damaged skin. To simulate the introduction of plutonium through epidermal damage, intramuscular injection of a concentrated solution containing ^{239}Pu was administered to pigs.

(5) Spectrometric analysis of biological samples of pigs slaughtered a day after feeding with grain contaminated with plutonium, showed that absorption of a small amount of PuO_2 from the gastrointestinal tract occurs as a result of partial dissolution. The upper limit of the intestinal absorption of plutonium was 0.007% of the activity entering the pig body orally, and the degree of digestion was not more than 0.003%. Such a low percentage of plutonium absorption can be explained by the fact that the intestinal mucosa is an effective barrier for plutonium absorption during exposure. Plutonium absorbed from the gastrointestinal tract comes into the blood in ionic or diffusing complex form and as a result is deposited in the bones.

(6) Absorbed from the gastrointestinal tract as a result of intramuscular injection and through intact skin, plutonium penetrates into the bloodstream in an ionic or complex diffusing form and concentrates in bones. In human autopsies and some experiments on inhalation of PuO_2 the authors found that plutonium is mainly deposited in the liver. Plutonium can be absorbed from precipitation of PuO_2 in lungs either in the form of Pu^{4+} or microparticles, or in both forms at once.

(7) After making an intramuscular injection of plutonium authors found that about 60% of absorbed plutonium is deposited in bones, about 4% in liver, and 30% is gradually excreted. Such a distribution could be considered as approximately correct when plutonium penetrated a human through wounds or through the gastrointestinal tract.

(8) Absorption of plutonium from lungs leads to the deposition of 31% of plutonium in the lungs, 30% in the ovaries, 17% in the liver, 15% in the skeleton, and 7% in other tissues.

(9) Before the experiments, faster excretion of plutonium was predicted from the liver than from other organs and bone tissue, but the results obtained after the experiments showed a much higher plutonium potential deposition danger in the liver. It turned out that within 4

years only 20% of deposited plutonium was excreted from the liver. The time of plutonium retention in the liver does not differ significantly from time of retention in the bone tissue. Thus, the initial conservative assumption that there is no substantial excretion of plutonium from the liver is apparently true.

(10) Plutonium concentration in the ovaries is approximately 20 times higher than in skeletal muscles, and approximately six times higher than in the heart. This was true for both monomer and polymer plutonium even 4 years after the initial exposure. In fact, when injecting the monomeric form, concentration of plutonium in ovaries is comparable with concentrations in the liver, whereas injection of the polymeric form of plutonium increases its concentration in the liver and exceeds its concentration in the ovaries.

(11) The experiments proved that the rate of plutonium excretion from the main locations of deposits in a living organism is very low, so when assessing internal exposure doses, biological excretion may be neglected; and only therapeutic plutonium excretion acceleration is of practical interest.

Conflicts of interest

The authors report no conflicts of interest. The authors alone are responsible for the content and writing of the paper.

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