RADIOLOGICAL ASPECTS OF THE USE OF FAST REACTORS IN A CLOSED NUCLEAR FUEL CYCLE UNDER THE "PRORYV" PROJECT

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Abstract. The development of nuclear power engineering with closing of a fuel cycle and the use of fast reactors must ensure a higher level of ecological safety of the population and the environment. The highest ecological effect is achieved by recycling of spent fuel and isolation of long-lived radionuclides (^{90}Sr, ^{137}Cs and ^{99}Tc) and transmutation of 99% of americium.

In normal operation of CNFC facilities exposure doses to the population are formed via different critical pathways: for a reactor plant – due to inhalation intake of ^{3}H, for fabrication and refabrication module – due to inhalation of Pu aerosols, for SNF recycling module – due to external radiation from the soil and ingestion in case of surface contamination of plants.

The use of nitride fuel generates large amounts of ^{14}C (270 g per 1 ton fuel). Insolubilizing most of ^{14}C ensures compliance with the project standards for the population exposure to the gas phase of release.

The development of nuclear energy with a closed fuel cycle using fast reactors should provide a higher level of population and environment ecological safety, that it is he basic principles of the PRORYV Project - a natural safety in the reactor operation and radiological equivalence for the final nuclear waste disposal. Taking into account the ability of radionuclides to migrate from radwaste underground location to biotic layers of the earth surface under certain conditions, the principle of radiation equivalence became known as the radiation-migration equivalence. Consider the all pathways of human exposure, according to the discharge of radionuclides into the top layer of fertile soil, this concept is called radioecological equivalence. Simultaneously, this concept is applicable not only to humans but also to natural organisms as bodies, subjected to greater radiation exposure then humans in consequence of environmental contamination under certain radioecological situations.

Features of technical solutions for pilot demonstration energy complex creation at the site of the Siberian Chemical Combine to determine the specificity of the environmental aspects of population and environment radiation safety. Thus, the use of lead as coolant in the BREST-OD-300 reactor allows you to avoid accidental releases of radioactive substances into the environment, leading to the population evacuation.

According to the requirements of inherent safety of nuclear facilities (without serious accidents) the first place in the ranking of radioecological challenges is taken with the problem of isolating radioactive waste. The basic requirements for their disposal is based on the principles of preadoptive and radiation equivalence and determined by the environmental safety need in the long term development of nuclear power engineering through the final disposal of long-lived high-level waste in deep geological formations [1 – 4]. This concept can be realized only with a closed nuclear fuel cycle using fast reactors, as a part of the long-lived radionuclides can be utilized in the transmutation process.

Realization of radioecological equivalence concept allows us to minimize the waste to be acceptable level for the society and, therefore, it works to implement one of the basic principles of radiation safety ALARA (As Low As Reasonably Achievable) principle.

The first insight into the radiological significance of different radionuclides by effects on living organisms gives the value of their activity. In the long term horizon, the greatest

danger in radioactive waste after reprocessing of spent fuel is represented by long-lived fission products and minor actinides. In Fig. 1 it is shown the main contribution of long-lived fission products activity, includes ^{90}Sr, ^{137}Cs, and ^{151}Sm for 500 years.

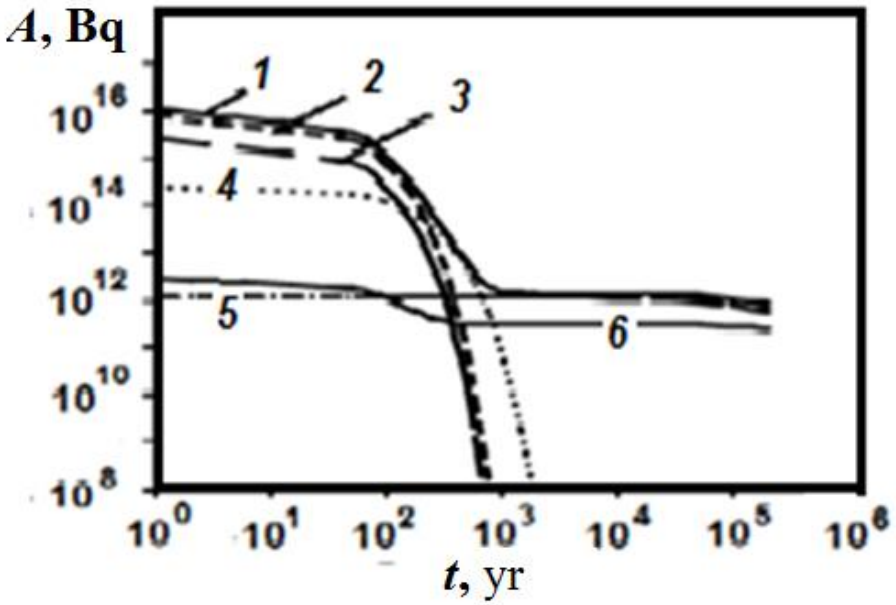

Fig. 1. Variation in activity of all long-lived fission products (1), ^{137}Cs (2), ^{90}Sr (3), ^{151}Sm (4), ^{99}Tc (5), other long-lived (6) in the spent fuel of the BREST-OD-300 reactor for 1 ton heavy at. after the launch of the campaign in 1500 days

The fission products in contrast to activity of some transuranic radionuclides may increase in time due to the accumulation of actinides decay in the other chain, for example, activity of ^{239}Pu and ^{240}Pu increases in time almost 10 times (Fig. 2). Fig. 2 also shows the total activity is driven more by Am in the period 100 – 10,000 years.

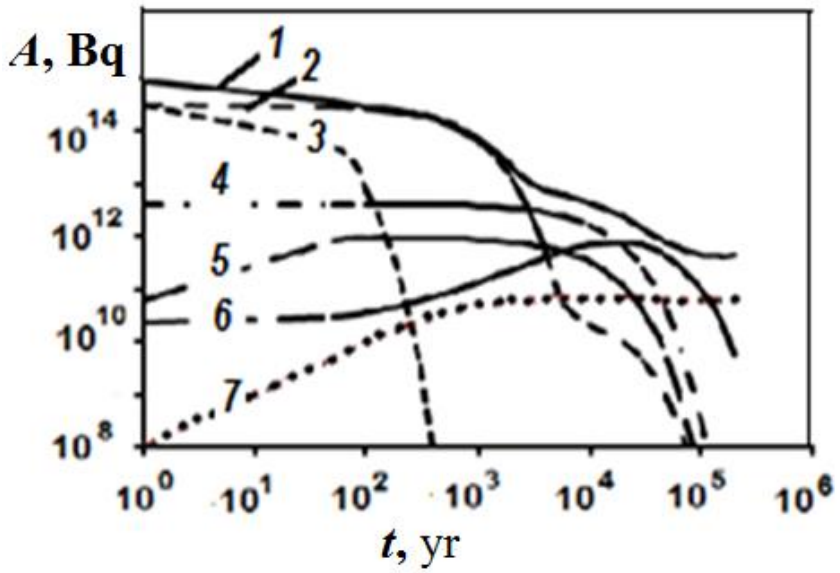

Fig. 2. Variation in the activity of minor actinides and their decay products (1), ^{241}Am (2), ^{244}Cm (3), ^{243}Am (4), ^{240}Pu (5), ^{239}Pu (6), ^{233}Pa (7) in the spent fuel of the BREST-OD-300 reactor for 1 ton of heavy at.

The importance of radionuclides contribution to the dose characteristics of spent fuel is different to their activity. Thus, the contribution of ^{151}Sm in the fission products activity is significant, while the dose is small (Fig. 3).

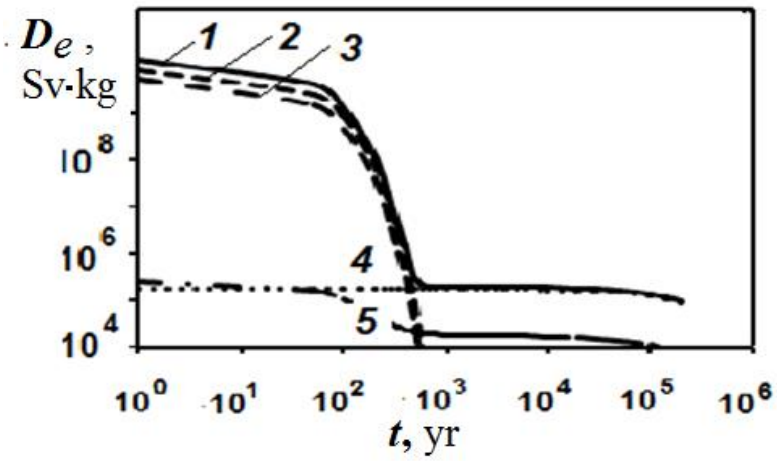

Fig. 3. Dose equivalent of all long-lived fission products (1), ^{137}Cs (2), ^{90}Sr (3), ^{99}Tc (4), other long-lived fission products (5) in the spent fuel of the BREST-OD-300 reactor, normalized to 1 ton of heavy at.

The greatest contribution after decay of ^{90}Sr and ^{137}Cs, to the dose makes ^{99}Tc by an order of magnitude larger in comparison with other radionuclides. Consequently, it is possible to determine which radionuclides need to be recovered from the spent fuel, recycled in a transmutation process and transformed into the insoluble form by special techniques, forward to long-controlled storage for ensure the requirement of radiation equivalence to the disposal of long-lived high level waste.

As the characteristics of the radionuclides potential biological hazards at the disposal or uranium ore was "dose equivalent", we defined it as a product of activity by the total dose conversion coefficient of external and internal human exposure when soil is contaminated . Dose equivalent migration included the amendment as a ratio of the radionuclides distribution coefficients and U in the system water – rock.

The analysis shown that according to the dose equivalent estimation both by taking into account and not the migration factor the same radionuclides still stay as the most dangerous, and biohazard of ^{99}Tc remains at the same level as far as the migration coefficient assumed equal to uranium (Fig. 4).

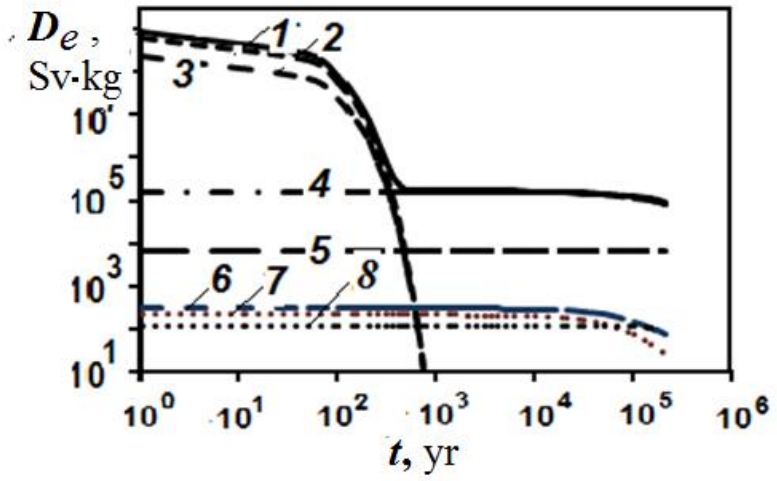

Fig. 4. Dose migration the equivalent of all long-lived fission products (1), ^{137}Cs (2), ^{90}Sr (3), ^{99}Tc (4), ^{129}I (5), ^{126}Sn (6), ^{79}Se (7), ^{135}Cs (8) in the spent fuel of the BREST-OD-300 reactor, normalized to 1 ton of heavy at.

The similar calculations of dose equivalent for minor actinides and decay products are shown in Fig. 5, 6. Comparison of the data shows that in contrast to the fission products migration factor significantly reduces the potential biological risk of minor actinides.

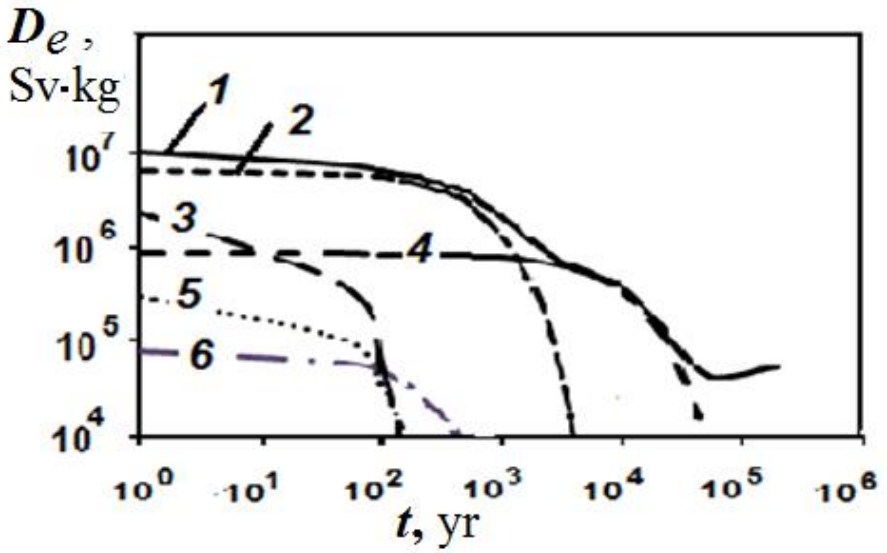

Fig. 5. Dose equivalent of minor actinides (1), ^{241}Am (2), ^{244}Cm (3), ^{243}Am (4), ^{243}Cm (5), ^{242}Am (6) in the spent fuel of the BREST-OD-300 reactor, normalized to 1 ton of heavy at.

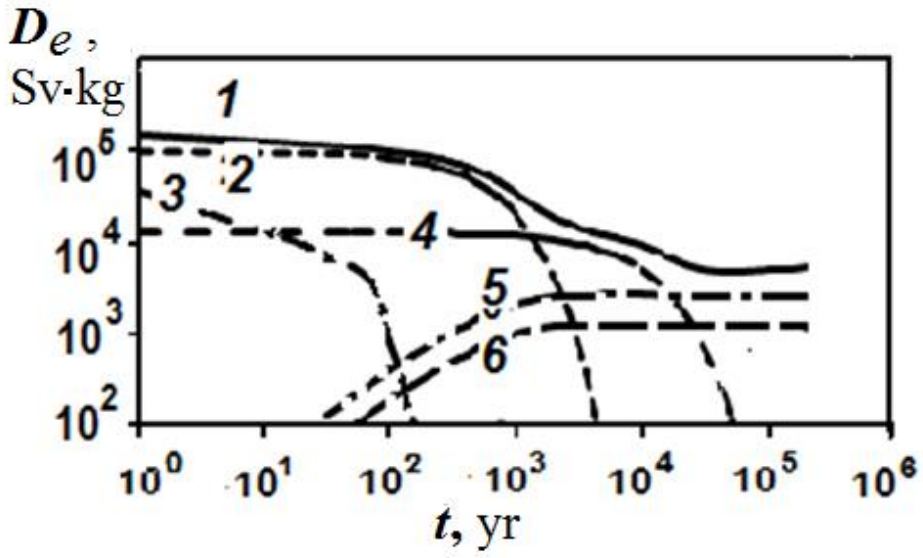

Fig. 6. Migration of the dose equivalent of minor actinides mixture (1), ^{241}Am (2), ^{244}Cm (3), ^{243}Am (4), ^{233}Pa (5), ^{237}Np (6) in the spent fuel of the BREST-OD-300 reactor, normalized to 1 ton of heavy at.

Dose-equivalents are separated by chains of the parent excluding actinides decay less regard to the migration factor, and they are shown in Fig. 7, 8. It can be observed that the burning of the Am nuclides – the most preferred option to reduce the potential biological hazard of radioactive waste.

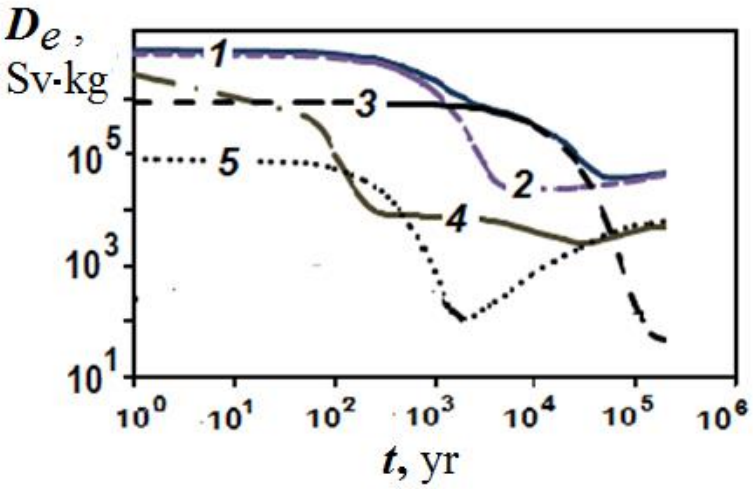

Fig. 7. Contribution to dose equivalent for all isotopes of americium (1), ^{241}Am (2), ^{243}Am (3), all isotopes of curium (4), ^{242m}Am (5) in the spent fuel of the BREST-OD-300 reactor, normalized to 1 ton of heavy at.

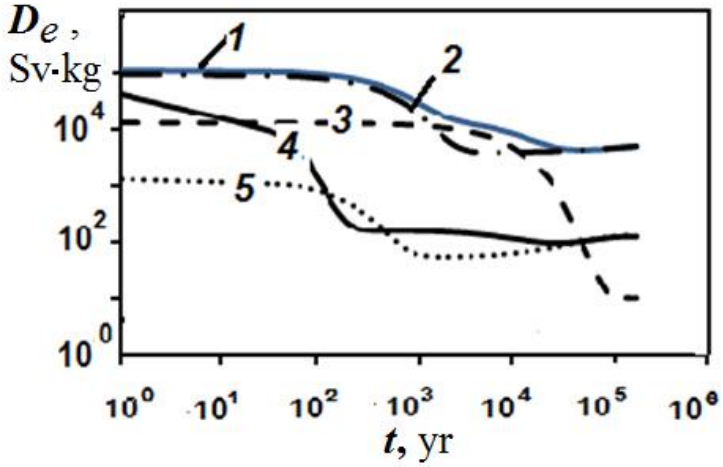

Fig. 8. Contribution of all the isotopes of americium (1), ^{241}Am (2), ^{243}Am (3), all isotopes of curium (4), ^{242m}Am (5) to dose migration equivalent in the spent fuel of the reactor BREST-OD-300, normalized to 1 ton of heavy at.

Another one of the radioecological aspects on the Pilot Demonstration Energy Complex (PDEC) associated with the use of mixed nitride uranium-plutonium fuel in the BREST–OD–300 reactor. Its usage leads to the formation of ^{14}C by the reaction ^{14}N(n,p)^{14}C. The total number of accumulated ^{14}C per 1 ton of fuel is 274 grams, the equivalent of 20 tonnes will be made of 1.37 kg per year, what is comparable to the annual flow in the Earth's atmosphere ^{14}C due to its formation from cosmic radiation – 8.2 kg [9]. Environmental hazard of this radionuclide occurs in the case of its discharge in the gas phase, mostly in the form of dioxide. With the constant release of ^{14}C as CO_2 is the predominant way of human exposure is oral. At photosynthesis CO_2 comes into the plant and further into plant and animal products.

Calculations show that the equilibrium concentration of ^{14}C in the atmospheric air, leading to the formation of the annual dose of 1 mSv, is 3.2 Bq/m^3, thus the annual emission, leading to the formation of doses of 10 μSv/year, at the border of sanitary protection zone is equal to 2.4 g. Conservatism of the calculations can be greatly reduced by taking into account the characteristics of the territory adjacent to the fuel re-processing module , for example, features of local food products production and consumption.

Reviewing the Siberian chemical plant the nearest agricultural lands are located approximately 8 km away from PDEC. In this case it is allowable to reduce the conservatism of the calculations by taking into account the scattering of the gas jet up to the border of agricultural lands. It's allowed by regulations. So, it is stated [10] that in general, the dose via food chains is not determined by a person's location in the territory, but spatial distribution of crop areas and other agricultural lands, and population exposure dose for all pathways, including food chains, is determined by the place of his residence in the territory.

The evaluation showed that the ratio of the average value of a function of scattering gas jet calculated in accordance with IAEA recommendations [11] at a distance of 8 – 10 km, to a maximum at a distance of 1 km from the stack is 0,043, on the border of 8 km to a maximum is 0,053. Thus, at large distances a distribution over the area has little effect on the amendment to the calculation of the permitted discharge. Revised permissible emission of ^{14}C will be equal to 2.4/0,043 = 55 g. The precision of the Pasqill method by which the calculation was made is estimated at ± 50% of the average [12]. Thus, adjusted for the error of the annual emission of 22.5 g of ^{14}C will not lead to exceeding the annual dose of 10 μSv for the population.

The main technical solutions to exclude the impact of ^{14}C on the population and the environment should be aimed at preventing its release at the gas phase during the reprocessing of spent fuel above the permissible limits and as whell as the transfer of the solid phase in the form of fullerene soot in an insoluble state at the burial.

Due to the different composition of the radionuclides in the discharges of fuel fabrication module, reactor facility and fuel re-processing module the structure of the radiation doses of the population during normal operation of the above-mentioned units will be different. Thus, for a reactor facility the main contribution to the population exposure dose is formed due to inhalation of ^{3}H, for the fuel fabrication/re-fabrication module due to inhalation of aerosols of Pu, for fuel re-processing module due to external radiation from soil and ingestion of receipts with the surface pollution of plants.

The contribution of individual radionuclides to the total dose of irradiation is presented in the Fig. 9. The contribution to dose of ^{14}C was taken into account.

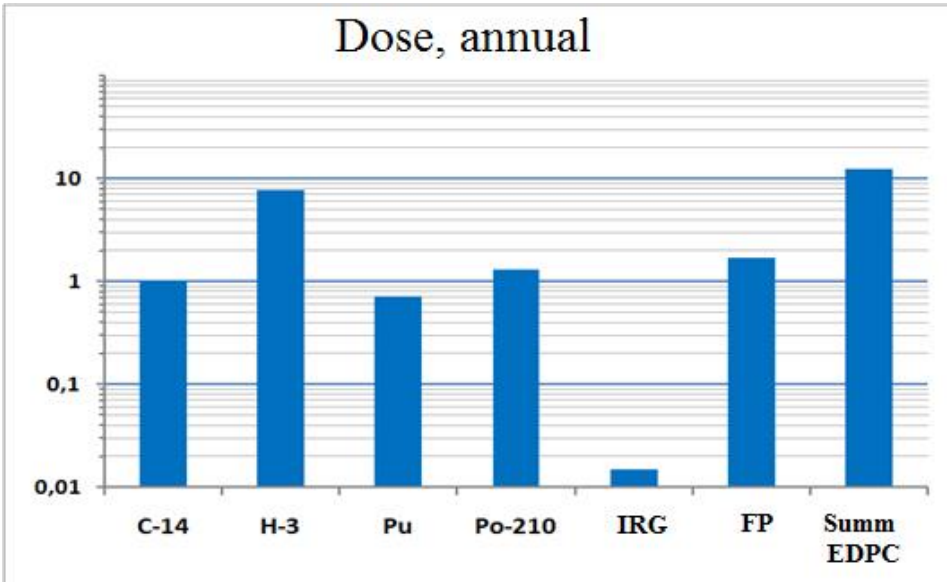

Fig. 9 Contribution of components of the radionuclide to the total dose of population exposure in the zone of maximum surface concentration of atmospheric discharges of PDEC

Thus, the greatest ecological effect during normal operation of a closed fuel cycle is achieved by spent fuel reprocessing and selection of long-lived radionuclides for transmutation. The extraction of 99% of the americium from spent nuclear fuel for transmutation with the additional extraction of ^{90}Sr, ^{137}Cs and ^{99}Tc for the controlled storage or the usage achieves compliance with a principle of radioecological equivalence for potential hazards of external and internal exposure of the population and biota.

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