# Radiation and hygiene assessment of

# external exposure factors of personnel

# working at experimental facilities in

# the production of mixed nitride

# uranium-plutonium fuel.

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**Abstract**

The purpose of this work was to study and identify the features of the formation of external radiation doses to personnel during the production of a new mixed uranium-plutonium nitride fuel, the production of which is currently being developed in Russia. This paper presents the results of a radiation-hygienic study of the factors of external exposure of personnel at the workplaces of experimental facility carried out in 2018-2019. During the research, dosimetric and spectrometric measurements of operational quantities were carried out at personnel workplaces, as well as the measurement of individual dose equivalents for personnel, including doses to the skin, using individual dosimeters. It has been shown that the neutron component forms about 20 percent of the external dose. The annual equivalent dose of irradiation to the skin of the hands was 190 ± 50 mSv, and with an increase in production volumes, this value will be a factor limiting the operating time. It was also determined that due to the configuration of workplaces, the equivalent dose of radiation to the lower abdomen is about 60% higher than the equivalent dose at the chest level, which is important for individual monitoring of female personnel. The radiation doses to the skin of the face and the lens of the eye turned out to be insignificant due to the sufficient protection provided by the protective glass of the sealed boxes. The results obtained give an idea of the current levels of personnel exposure during the development of new technologies and make it possible to identify the main factors of the harmful effects of external exposure to which the personnel will be exposed during the industrial implementation of the mastered technology.

## INTRODUCTION

At present, the Siberian Chemical Combine JSC is developing a new mixed uranium-plutonium nitride fuel (MNUP fuel). From the point of view of radiation safety, the main difference between MNUP fuel and uranium oxide fuel is the plutonium content in the amount of 10 - 20% [1, 2]. The presence of such an amount of plutonium multiplies the specific activity of the fuel in comparison with uranium fuel, which determines its higher radiotoxicity. The difference from MOX fuel lies in the chemical composition of the fuel matrix, which determines the composition and physicochemical characteristics of radioactive aerosols in the air of industrial premises, as well as the release of ionizing radiation from the fuel due to a number of nuclear reactions on the elements of the fuel matrix.

In 2018-2019 Investigations were carried out at workplaces of MNUP-fuel production from fresh (non-regenerated) raw materials, including studies of photon and neutron radiation fields at workplaces and individual equivalent doses of personnel exposure.

The experience of ensuring the radiation safety of personnel and the public during the development of new technologies, which is accumulated in the process of work on experimental installations, will be further used in the transition to experimental-industrial operation of new modules for fabrication and refurbishment of MNUP fuel, with the subsequent development of production and reaching the design capacity.

In the course of this work, a comparison was also made of the radiation factors of occupational exposure to personnel working at the experimental installation for the production of MNUP-fuel of Siberian Chemical Combine JSC, with the requirements of sanitary standardization documents [3].

## COMPARATIVE ANALYSIS OF IONIZING RADIATION CHARACTERISTICS of MNUP-fuel.

### *Neutron radiation*

Neutron radiation at the fuel production site is formed mainly due to the (α, n) - reaction on light nuclei (O, C, N) and the spontaneous fission reaction. The main contribution to the yield of spontaneous fission neutrons from the fuel is made by the isotopes 238Pu, 240Pu, and 242Pu [4]. The total mass fraction of these isotopes in fuel plutonium varies from one fifth to one third. [5]. Due to this, the yield of spontaneous fission neutrons in the production of fuel containing plutonium is orders of magnitude higher than that of uranium fuel.

Powders of uranium and plutonium oxides are the raw material for the manufacture of MNUP fuel. The (α, n) reaction in them mainly occurs on the isotopes 17O and 18O, which are part of natural oxygen (0.037% and 0.204%, respectively). The threshold for this reaction is about 1 MeV, which is significantly lower than the energy of alpha particles emitted by fuel components.

The energy distribution of the neutron yield of uranium-plutonium oxide, which is the sum of the neutron spectrum formed by the reaction (α, n) on oxygen [6], and the spectrum of spontaneous fission [7], is shown in Fig Fig 1. The average energy is 2.1 MeV, neutrons with energies above 8 MeV are practically absent.

Fig 1. Components of the energy distribution of neutrons emitted by various fuel compositions.

After carbothermal synthesis, the neutron yield of the (α, n) -reaction becomes insignificant, since the threshold of this reaction on isotopes 14N and 15N (the content in the natural mixture is 99.636% and 0.364%, respectively) is already more than 6 and 8 MeV [8], which is higher than the maximum energy of alpha particles for the components of fresh fuel (238Pu - 5.5 MeV).

This reaction can also proceed on the remaining impurities: oxygen and carbon, but their content is insignificant (from 0.01 to 0.15% of the mass.) [9]. Thus, the neutron spectrum of the finished MNUP fuel is characterized by the spectrum of spontaneous fission, and the neutron yield is noticeably lower than that of MOX fuel with an identical fraction and isotopic composition of plutonium.

At workplaces, the energy spectrum of neutrons will be significantly different from the original due to the slowing down, scattering and reflection of neutrons. As a result, the contribution of thermal and intermediate neutrons will increase, depending on the configuration of the workplace.

### *Photon radiation*

The isotopes 238Pu, 239Pu, 240Pu and 242Pu decay with the emission of alpha particles. As a result of the α-decay of plutonium isotopes, with a certain probability (~ 25%), a number of low-energy states of the corresponding uranium isotopes with energies lower than the binding energy of K-electrons are excited (for 238Pu, the level is 43.5 keV, for 239Pu - 51 keV, 240Pu - 45 keV ). The decay of these states occurs through internal conversion of γ-rays with the emission of electrons and subsequently characteristic X-rays with an energy of 13 - 23 keV [10]. The analysis of the nuclide composition showed that more than 90% of the photons formed during the radioactive decay of the MNUP fuel elements have an energy of less than 25 keV.

The effects of self-absorption of radiation and shielding by protective structures will reduce the contribution of low-energy radiation, but in operations with powders, tablets and blocks that require the use of manual labor, high values of the equivalent dose to the skin of the hands can be formed.

In addition, the isotope 241Pu, which is part of energetic plutonium, is a beta emitter; during its decay, 241Am is formed and accumulates over time, which is a soft gamma emitter with a fundamental energy of 59.5 keV.

An insignificant contribution to gamma radiation from fresh fuel will also be made by gamma radiation accompanying spontaneous fission of fuel components, capture gamma radiation (n, γ), bremsstrahlung photon radiation from β-active nuclei, and gamma radiation as a result of the 14N(α,p)17O.

In view of the above, when examining the radiation situation at workplaces, it is necessary to pay close attention to the irradiation of the skin of the hands. The lower limit of the measurement range of the dosimeter of the directional dose equivalent H (0.07) or the individual dose equivalent Hp (0.07), selected to control the irradiation of the skin of the hands, should capture the Lx-radiation of plutonium - 13 - 23 keV. Therefore, in this work, we used the BDKR-01 detection unit of the MKS-AT1117M dosimeter-radiometer, the lower limit of the measured radiation energy range of which is 5 keV, and individual MKD type B hand skin dosimeters, the lower limit of the measured radiation energy range of which is 7 keV.

Since work with MNUP fuel can only be carried out in sealed boxes with an inert (nitrogen) atmosphere, the effect of unshielded radiation of the fuel on the skin of the face and the lens of the eye is excluded. The degree of radiation shielding efficiency in the soft part of the spectrum was estimated by comparing the readings of instruments that make it possible to measure the value of the directional dose equivalent and the ambient dose equivalent in the range of at least 120 keV. As a rule, protective glasses of sealed boxes almost completely absorb low-energy photon radiation.

## RESEARCH METHODS.

When studying the regularities of the formation of the dose of external exposure of personnel, regular monitoring of the dynamics of the ambient dose equivalent rate of photon and neutron radiation at workplaces was carried out. Measurements of the dose rate of neutron radiation were carried out at the level of the worker's chest, and the dose rate of photon radiation at the level of the chest, lower abdomen, feet and head. To measure the ambient dose equivalent rate of photon and neutron radiation, an MKS-AT1117M dosimeter radiometer with a BDKN-03 detection unit was used.

At the workplaces with the maximum values of ambient dose equivalent of neutron radiation, studies of the spectrum of neutron radiation were carried out using a Bonner-sphere spectrometer. The measurements were taken near workplaces where the operations of pressing, crushing and temporary storage of finished products are carried out.

To study the possibility of exceeding the limit of the equivalent dose of irradiation of the hands (500 mSv) while not exceeding the limit of the effective dose (20 mSv), measurements of ambient dose equivalent of gamma radiation were carried out at the chest level and in the glove opening.

As shown in [11, 12], under industrial conditions, one should expect an overestimation of albedo individual dosimeters. To correct the readings of dosimeters in neutron fields with a spectrum different from the spectrum of the calibration facility, correction factors are used, which are applied according to formula (1).

|  |  |
| --- | --- |
|  | (1) |

where E is the estimate of the effective dose of neutron irradiation, M is the indication of the dosimetry monitoring device in the corresponding channel, k is the correction factor for the neutron dose channel.

The values of the correction factor were calculated according to the formula (2) for the purpose of their application for individual dosimetric control at the enterprise.

|  |  |
| --- | --- |
|  | (2) |

where is the measured neutron spectrum, — is the spectrum of the calibration facility with a Pu-Be source, и are the energy dependence of the conversion coefficient of the transition from the neutron flux to the effective dose for the selected geometry and for the anterior-posterior geometry, respectively, и — are the energy dependence of the sensitivity of the individual dosimeter during irradiation in the selected geometry and in the anterior-posterior geometry, respectively.

As part of the research, temporary individual control of equivalent doses of radiation to the skin of the face, skin of the hands and the lens of the eye using individual dosimeters was organized. Estimates of the effective dose of external exposure to personnel were determined by means of individual dosimetric control. All employees of the experimental facility were provided with dosimeters of various types: thermoluminescent gamma-neutron dosimeter DVGN-01, thermoluminescent skin dosimeter MKD type B, electronic direct-reading dosimeter EPD-N2 and electronic cumulative dosimeter of gamma and X-ray radiation DIS-1. The forecast of the values of various effective and equivalent doses in the case of year-round employment of personnel was carried out according to the formula (3).

|  |  |
| --- | --- |
|  | (3) |

where is the duration of the working time during which the worker wore the dosimeter, hours; – is the number of employees participating in the research, – is the value of the individual dose equivalent accumulated by the k-th employee's dosimeter, mSv, is the background dosimeter readings that were exposed at the dosimeter storage site during non-working hours , mSv.

Due to the uneven workload of installations throughout the year, the studies were limited in time, and the results obtained during individual months were applied to the entire calendar year.

## RESULTS.

Measurement of the ambient dose equivalent values of gamma and neutron radiation in the premises of the experimental setup showed that the main sources of penetrating radiation are boxes where tablets are pressed, fuel block and defective tablets are crushed, as well as temporary storage of products.

As can be seen in the distribution histogram of the observed values of the ratio of the ambient dose equivalent rate of neutron radiation to the ambient dose equivalent rate of gamma radiation (Fig. 2) when working with MNUP fuel at an experimental setup, the contribution of neutron radiation to the formation of individual doses is significant and at some workplaces it can exceed the contribution gamma radiation. For comparison, the histogram also shows data obtained at similar workplaces at an enterprise that produces oxide uranium fuel.

‑ experimental stand for the production of MNUP fuel pellets;

– production of uranium oxide fuel pellets (for comparison).

Fig. 2 – Histogram of the distribution of the observed values of the ratio of the ambient dose equivalent rate of neutron radiation to gamma radiation.

Figure Fig. 3 shows a diagram of the scattering of the values of the power of the ambiant equivalent of the dose of gamma radiation in the glove opening and at the level of the personnel's chest. A large number of points are located above the line (),which advises the value of the ratio 25, which corresponds to the simultaneous achievement of the limit of the equivalent exposure dose to the hands (500 mSv) and the limit of the effective dose (20 mSv), controlled by the readings of an individual dosimeter located on the chest. Thus, for these workplaces there is a possibility of exceeding the limit of the equivalent exposure dose to the hands of the hands without exceeding the limit of the effective dose. Two trends stand out from the general set, marked with symbols: ▲ – - caused by contamination of the box during previous operation and ♦ - due to the smaller thickness of the protective walls of the box compared to the rest of the chain.

Fig. 3. ambient dose equivalent rate of gamma radiation in the glove opening and at chest level

For most workplaces, the ambient gamma dose rate at the pelvic level is higher than the ambient dose rate equivalent at the point where the personal dosimeter is located. This is due to the fact that the lower surface of the boxes, where the radioactive material is located, is at the level of the pelvis. This circumstance can lead to unrecorded excess of the main limit of the annual effective dose during individual dosimetry control according to the indications of one dosimeter placed on the chest. When implementing this technological chain in the future, it is recommended to provide for the installation of additional protection against gamma radiation on the front side of the boxes from the bottom of the box to the sight glass. Irradiation of the feet is negligible and does not require control.

The results of reconstruction of the measured spectra of neutron radiation are shown in Table 1. As can be seen from the results, the average energy of the spectrum of the field of neutron radiation formed at workplaces is much less than the average energy of neutrons when irradiated on a verification installation of neutron radiation with a Pu-Be source (3.6 MeV) [13]. The calculated values of the correction factor for the albedo thermoluminescent dosimeter (DVGN-01) used at the enterprise are shown in Table 2.

TABLE 1 - INTEGRAL CHARACTERISTICS OF THE MEASURED NEUTRON RADIATION SPECTRA.

|  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- |
| № | Neutron flux density, 1/(cm2·s) | | | Ambient dose equivalent rate, H\*(10), μSv/h | Specific ambient dose equivalent,  h(10),  pSv/(cm2∙s) | Average energy, MeV |
| less than 100 keV | from 100 keV to 1 MeV | from 1 MeV |
| 1 | 0,22 ± 0,08 | 0,36 ± 0,14 | 0,09 ± 0,11 | 0,58 ± 0,06 | 240 ± 40 | 0,6 ± 0,2 |
| 3 | 1,2 ± 0,3 | 1,8 ± 0,5 | 0,7 ± 0,4 | 3 ± 0,2 | 230 ± 20 | 0,6 ± 0,18 |
| 4 | 0,6 ± 0,15 | 0,5 ± 0,17 | 0,21 ± 0,11 | 0,85 ± 0,07 | 180 ± 20 | 0,6 ± 0,3 |
| 5 | 0,75 ± 0,18 | 1,1 ± 0,4 | 0,4 ± 0,3 | 2 ± 0,14 | 240 ± 20 | 0,6 ± 0,2 |
| Spectrum of a calibration setup with a Pu-Be source | | | | | 406 | 3,6 |

TABLE 2 - RESULTS OF CALCULATION OF THE CORRECTION COEFFICIENT FOR THE INDIVIDUAL DOSIMETER DVGN-01.

|  |  |  |
| --- | --- | --- |
| № | For anterior-posterior geometry,  kAP | For isotropic geometry,  kISO |
| 1 | 0,2 ± 0,04 | 0,29 ± 0,06 |
| 3 | 0,2 ± 0,03 | 0,3 ± 0,05 |
| 4 | 0,17 ± 0,03 | 0,26 ± 0,07 |
| 5 | 0,2 ± 0,03 | 0,3 ± 0,05 |

Based on the results of the analysis and generalization of the obtained data on the regularities of the formation of the fields of photon and neutron radiation at the workplaces of the personnel of the experimental setup, estimates of the external exposure dose were obtained, shown in Table 3.

TABLE 3 - ESTIMATES OF THE EXTERNAL EXPOSURE DOSE TO THE PERSONNEL OF EXPERIMENTAL FACILITIES

|  |  |  |
| --- | --- | --- |
| Irradiated organ | Average value of individual effective/equivalent dose | |
| per working day, μSv/(6 hours) | per year, mSv / year |
| Whole body, gamma radiation | 7,4@ ± 1,5$ | 2,1@ ± 0,4 |
| Whole body, neutron radiation | 1,5@ ± 0,3 | 0,4@ ± 0,1 |
| Lens of the eye | 8,0# ± 1,8 | 2,3# ± 0,5 |
| Skin on the face | 10# ± 2 | 2,7# ± 0,6 |
| Gonads | 12# ± 5 | 3,4# ± 1,3 |
| Hand skin | 680# ± 180 | 190# ± 50 |

@ – is the effective dose; # – equivalent dose; $ – standard deviation estimate.

All the above data and conclusions refer to raw materials that have undergone deep purification from radiogenic impurities. When irradiated nuclear materials are used as a raw material for the manufacture of MNUP-fuel, the levels of gamma-neutron irradiation of personnel are expected to be significantly higher and will differ from the picture presented in the article.

## CONCLUSIONS.

Personnel are exposed to mixed gamma-neutron irradiation with a significant contribution from the low-energy component of photon radiation. Equivalent doses of irradiation of the lens of the eye and the skin of the face are currently far from the established standards and do not require the introduction of instrumental control. It is important to note that the equivalent dose of irradiation of the lower abdomen and skin of the hands, although it is also far from the established standards, but with an increase in the intensity of work and, as a consequence of the intensity of the radiation fields, is able to overcome the established standards much earlier than the effective dose of irradiation determined by the indications chest dosimeter.

The neutron radiation spectra formed in the workplace have a significantly lower average energy than the neutron radiation spectrum of the calibration facility. Therefore, for the albedo thermoluminescent dosimeters used at the enterprise, it is recommended to introduce correction coefficients.

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