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NUCLEAR ENVIRONMENT RECONSTRUCTION ON THE TERRITORY OF CHERNOBYL APP ACCORDING TO GAMMA-SPECTRAL ANALYSIS OF SOIL SAMPLES AND LITERARY DATA

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ABSTRACT

Radiative conditions reconstruction at the place of radioactive accident according to research results of sample radionuclide composition was done by example of the Chernobil atomic power-station failure happened in April 1986. The Chernobil atomic power-station failure took place 20 years ago. A lot is comprehended and summed up. But the failure and its consequences are still an actual theme for studies.

Radiation exposure estimation on human, radiation protection, consequence forecast, somehow, are connected with radioactive features changes and dose calculation using physical and biophysical models. Here we face with common problem - impossibility to estimate reliability of results acquired without alternative way to check selected models. Radiochemical and spectrometric method for radionuclide determination in samples, taken at the places of radioactive accident, conducted some years ago after that, give an opportunity to calculate independently radiation fields and compare it with data, received during the accident. Such work is made at radiochemistry department chemistry faculty of MSU named after M.V. Lomonosov.

19 years later the accident its possible to determine gamma-emitting radionuclide composition in sample of pulverescent sandstone taken in July 1986 on wayside of Pripyat –the Chernobil AS road 1,5 km from 4-th power unit. It's interesting to determine relatively long-lived radionuclides such as Eu-154, Eu-155, Co-60, Am-241, which became possible, because majority of shot-lived radionuclides had decayed. Using reference and literary data radionuclide sample reconstruction was done on first and ten day after the accident. Possible variations of gamma radiation dose were determined for thin film model. Based on received data variations of absorbed gamma radiation dose were estimated, which is necessary for urgent decision making in the beginning of radiation accident. Received results are coordinated with published dose value data and validate a decision about people evacuation from the city Pripyat.

Key words: radiative conditions, the Chernobil atomic power-station failure, gamma radiation, dose, radiochemical and spectrometric measurements.

ACCIDENT ON THE CHERNOBYL APP

In 1986 on the Chernobyl APP 4 CPCT (high-power channel-type) reactors 1000 MW power each functioned. The reactor has worked for 865 calendar days (715 effective days), during this time 1500 MCu fission and activation products were accumulated.

The accident happened in 01 hour 23 min 40 sec of Moscow time.[7,1] Cover of the reactor 2000 t weight was moved away by two successive heat explosion, active zone was thrown out to the reactor hall, red-hot concrete and graphite pieces were throw away through a hole in the cover, as a consequence 30 places of fire occurred around a neighbor reactor and turbine blocks. At the same time there was a radionuclide release from an active zone of reactor to the environment during explosions in the form of a cloud that spread at a height of some kilometers. At later stages of the accident it was in the form of jet. At initial stage of the accident the jet was 1200 m height (30m to north-west), next days the jet height did not exceed 200-400 m. Sharp release stopping occurred on May 6 due to full melting of the covering materials that «vitrificated» damaged fuel. [10]. First hours after the accident the jet at a height of 700- 1500 m spread mainly in north-west direction, than the direction changed to north, since 15.00 April 27 it changed on north-west, south-east. In surface air (up to 200 m) on April 26-27 radioactive mass transfer was in the beginning in the west and north-west direction, than on April 27-29 discharge propagation was mainly in the north-east direction, since April 29 it was in south and later in east direction.[1]

According to specified estimations [1] the value of fuel release outside 4^{th} block reactor is (3,5±0,5)% i.e. 52,5 MCu or $2*10^{18}$Bq of radioactive substance. From them ≤0,3% on industrial area, ≤1,5% – on adjacent 80 km territory (near zone), ≤ 1,5%- on the ot territory, ≤1%- beyond the bounds of the USSR territory.

According to literary data (6) initial stage of relative radioactive products release (since 26.04 until 30.04) is well approximated by a dependence:
$Q(t) = 0{,}25\exp(-0{,}28 \cdot t)$, where t is a number of days, passed after the accident, i.e. $\lambda_{эфф} = 0{,}28$ (1/day) , but it has « shortage » on 6 MCu for the first day.

For the period from 1.05 till 5.05 relative day release is approximated by dependence:
$Q(t) = 0{,}09 \exp 0{,}35(t-5)$, where t is a number of days after the accident = 5, 6…9

According to experimental data of work [2] average radioactive products specter energy on near track in the period of 5-120days after the accident changed in the limits 0,9 – 0,6 MeV/ decay.

CRITERION OF RADIOACTIVE INFLUENCE UPON HUMAN.

According to RF Law «About social protection of citizens exposed to radioactive influence due to Chernobyl APP accident» [9], and also to Norms of radiation safety, (NRS-99) [8], additional (upon the level of natural radiation background for present area) exposure to radiation of citizens due to Chernobyl APP accident is accessible and allowed that formed

in 1991 year and during next years average annual effective dose that does not exceed 1 mSv.

To prevent exposure of population on polluted due to the accident territories NRS-99 the criterions for urgent decision making, where prompt interference is necessary, are introduced into practice. Interference criterions are based on the value of radiation absorbed dose for first 2 days, 10 days, and year. These condition highs requirements for determination of radiation dose correction.

DOSE POWER CALCULATION DURING USAGE OF DIFFERENT MODELS

Estimation of radioactive influence on human, radiation protection, forecast of consequences and so on, in any way, we have deal with radioactive characteristics measuring and calculation using physical and biophysical models. Here we encounter common problem - impossibility to estimate the truth of received results with no alternative way to check selected models.

Recommended models to calculate dose power in the air by fission product density on polluted territory [1, 2, 3, 4, 9] meant, as usual, for real distribution of specific radionuclids in soil on line. All models are based on some theoretical conclusions, experimental data and postulated regulations. In works under review contribution to dose power, created separately by each radionuclide that was taken into account during estimation and forecast of radioactive pollution characteristics of the area, is examined. Difference between dose coefficients used in various estimation models, for instance, for ^{137}Cs that is one of the most dangerous fission product that has a simple decay scheme and well studied, reaches more than three [3]. Depending on real radionuclide distribution model along soil profile, difference between dose coefficients of transition from area pollution density to dose power could achieve an order, to a lesser degree, the difference is determined by soil moisture, density and composition [3, 11].

In calculations, as usual, exponential law of artificial radionuclide distribution in depth of virgin soils is used. Absorbed dose power in the air (mkP/year) could be calculated by a formula:

$$P = 18{,}6F\sum_i E_{0i} n_i \mu_{en,m,i}^{air} fi$$

Where;

F - radionuclide surface activity (мCu/km^2), E_{0i} – i-th energy of initial radionuclide γ-quantum (MeV), n_i – photons with energy for one decay, $\mu_{en,m,i}^{возд}$ - mass coefficient of energy absorption in the air (cm^2/g), f_i - dimensionless function, depended from kind of radionuclide distribution in soil and calculation model parameters.

Function f (h,k,E_0) in the limits of 20% error is constant in the energy range from 0,1 to 3,0 MeV, so it's average meaning could be accepted at energy E_0=0,5 MeV. Within the limits of error ±5% f (h,k,E_0), calculated for ^{137}Cs could be used to estimate MED from any other radionuclide. So dose power could be calculated by formula:

$$P = 0{,}096 K_\gamma f F \quad [(\text{mkGy/year})/(\text{mCu/km}^2)],$$

where K_γ - full gamma - radionuclide constant ($R \cdot sm^2/h \cdot mCu$).

For flat infinite source calculation formula for MED for i radionuclide with pollution density F_i (Cu/km^2) is: $P_i=\varepsilon_i F_i$, where ε_i – conversion factor [(mkR/h)/(Cu/km^2)]. In works [3,9] the values of conversion factor are given for possible fission and activation products that pollute environment. So for ^{137}Cs this value is 10,7 [(mkR/h)/(Cu/km^2)] or 0,56 [(mkGy/year)/(mCu/km^2)].

Besides vertical radionuclide distribution in soil, microrelief, snow cover presence and thickness, buildings shielding are also important factors that influence on absorbed dose weakening. In a work [3] the following selected model factors estimation is offered. Microrelief region influence is in the range k1=0,82-1,01 (let's take it = 0,9). Due to snow cover annual dose decreases on 23-33 %, k2=0,75. During transition from absorbed to effective dose it is offered to take k3=0,6. Buildings shielding coefficient for city is k4=0,25, for village it is k4=0,7. So for city η_r will be: 0,10. For village η_c will be 0,28. Therefore, for averaged dose estimations η_r=0,15 and η_c=0,3 could be taken.

Radiochemical and spectrometric radionuclide determinations in samples taken at the places of radiation accident, that were made some years later after the accident, allow to conduct independent radiation field calculation and compare it with radiation characteristics received during the accident. Besides, presented below model could be used for operative estimation of radiation dose on population, as a part of information, according to which radiation safety measures are taken.

Sample Gamma- spectrometric measures

A sample of dusty sand material was taken from the Pripet – Chernobyl APP wayside in 1,5 km from the 4th block in July 1986. The square of sample splitting is approximately 100 - 150 cm^2 like the size of adult palm. The tests of the sample was done in CGSEN in Moscow by the detector from super-pure germanium (spectrometer «DSPEC jr» with specter processing program «GAMMA-VISION-32» by firm «ORTEC») in punctual geometry.

Test results of radionuclide specific activity from sample №1 of dusty sand material is represented in Table 1.

Table 1.: Test results of radionuclide specific activity of sample №1-05 of dusty sand material.

Nuclide	T, year	A, Bq/kg 31.05.2005	A, Bq/kg 26.04.1986	A_i / A_{cs-137} 26.04.1986
^{125}Sb	2,77	$(3,1 \pm 1,8) \cdot 10^{+4}$	$(3,6 \pm 1,8) \cdot 10^{+6}$	0,06
^{137}Cs	30,17	$(3,9 \pm 0,27) \cdot 10^{+7}$	$(6,1 \pm 0,42) \cdot 10^{+7}$	1
^{134}Cs	2,07	$(4,2 \pm 0,36) \cdot 10^{+4}$	$(2,6 \pm 0,22) \cdot 10^{+7}$	0,44

^{241}Am	433	$(9,0 \pm 0,74) \cdot 10^{+5}$	-	
^{241}Pu	14,7	-	$(2,2 \pm 0,2) \cdot 10^{+6}$	0,04
^{154}Eu	8,5	$(1,6 \pm 0,18) \cdot 10^{+5}$	$(7,4 \pm 3,4) \cdot 10^{+5}$	0,01
^{155}Eu	4,96	$(1,3 \pm 0,28) \cdot 10^{+5}$	$(1,9 \pm 0,4) \cdot 10^{+6}$	0,03
^{60}Co	5,27	$(2,1 \pm 0,19) \cdot 10^{+4}$	$(2,6 \pm 0,23) \cdot 10^{+5}$	0,004

*Activity ^{241}Pu is calculated through the activity of ^{241}Am.

Determination of europium-154 and 155 that were never cited in tables of Chernobyl radionuclide, and also cobalt-60 is very interesting. Europium -155 was determined by lines 86,5 keV with 30 % outlet and 105,31 keV with 21,8 % outlet, europium -154 was determined by lines 123,10 keV, 1274,80 keV, 723,30 keV with 40,46 %; 35,5 %; 19,7 % outlet; correspondingly. Now refractory radionuclide caesium-144 (T=284,3 days) and zirconium-95 (T=64,4 days) decayed, so we normalized the determinate radionuclide activity on caesium -137, as radionuclide, presented in all tables and that could be well determined even at the present time. It is important that radionuclide-caesium ratio in sample corresponds to ratio in fuel composition (Table 2.) of the reactor 4th block on the moment of release.

Table 2.: Radionuclide and Cs-137 ratio in active reactor zone at the moment of the accident in coarse hot parts, fine-dyspersated parts (data of ИОЯФ РНЦ КИ , 1992[4]) and in tested sample №1-05 of soil-dust material.

Nuclid	T, year	$A_i/A_{cs\text{-}137\ on}$ 26.04.86			
		In active zone of reactor	Big hot particle	Little particle	Sample №1-05
^{125}Sb	2,77	0,07	0,07	7,4	0,06
^{134}Cs	2,07	0,58	2,5	0,2	0,44
^{137}Cs	30,17	1	1	1	1
^{241}Am	433,0	$0,5 \cdot 10^{-3}$			
Eu-154	8,5		0,04	0,06	0,01
Eu-155	4,96				0,03
Co-60	5,27			0,08	0,004

Specific activity Cs-137 in tested sample on 26.04.1986 equals $(6,1 \pm 0,42) \cdot 10^4$ Bq/g, corresponds to average density of surface pollution 610 Bq/sm^2, on conditions that soil density is 1 g/cm^3, that corresponds $6,1 \cdot 10^6$ Bq /m^2 or 6100 kBq/m^2. (Global background by cesium -137 is determined as 2,0 kBq /m^2 , [1]).

According to reference data [5, table 4.1.1.] relative content of caesium-137 in inviolate fuel composition of the 4th reactor block is 0,43%, i.e.

$$\frac{A_{Cs-137}}{\sum Ai} = 4,3*10^{-3} ,$$

So, using surface pollution density data by caesium-137, we estimate total nuclear fall-out density by the following formula:

$\Delta A_i = \frac{6{,}1 \cdot 10^6}{0{,}0043} = 1{,}4 \cdot 10^9$ Bq/m^2.

Dose power calculation

To calculate dose power we estimate decay speed constant and gamma-constant for radionuclide mixture.

Table 3.: According to table 4.4.1.[5] Integral radioactive characteristics per 1 ton of irradiated fuel (actinium series + fission products) of fuel assembly concentration 2 % of reactor CPCT -1000 (A irradiation conditions).

t, days	Activity, Bq/g	M, g-eqv Ra/t	E, MeV
0,25	$1{,}21 \cdot 10^{18}$	$7{,}63 \cdot 10^6$	409
0,5	$1{,}08 \cdot 10^{18}$	$6{,}54 \cdot 10^6$	388
1,0	$9{,}35 \cdot 10^{17}$	$5{,}54 \cdot 10^6$	379
5,0	$5{,}16 \cdot 10^{17}$	$3{,}29 \cdot 10^6$	451
10,0	$3{,}67 \cdot 10^{17}$	$2{,}42 \cdot 10^6$	528

Proceeding from this data we receive effective decay constant:

$\ln \frac{A}{A_0} = -\lambda_e t$, отсюда $\lambda_e = 0{,}11$ (1/d).

We calculate γ-constant, using the sane reference data [5] Total actinium series and fission products activity per 1 day (see table 3):
$A = 9{,}35 \cdot 10^{17}$ Bq/t,

gamma – equivalent $M = 5{,}54 \cdot 10^6$ g-eq Ra, so we have:

$$\Gamma_x = \frac{M}{A} \cdot 8{,}4 \rightarrow 1{,}84 \text{ (R} \cdot \text{sm}^2\text{)/(h} \cdot \text{mKi)} \approx 12{,}4 \text{ (aGr} \cdot \text{m}^2\text{)/(s} \cdot \text{Bq)}.$$

γ- radiation power dose calculation on first day after the accident

For local release in the initial period of the accident (until the first rain) dose power at a height h could be calculated on the model of radiant thin disc with radius R [6]:

$P_\gamma = \pi \cdot \Gamma_{СИ} \cdot A_s \cdot \ln\left(\frac{h^2 + R^2}{h^2}\right)$,

$\Gamma_{СИ}$ - γ - constant, (Gy· m^2)/(sec· Bq); A_s - pollution density, Bq /m^2;
h - detector height above the disc centre, m; R – radiant disc radius, m.

For instance, with h = 1 m; R = 50 m; $A_s = 1{,}4 \cdot 10^9$ Bq /m^2 ; $\Gamma_{СИ} = 12{,}4$ aGy· м^2·/(sec· Bq),

$P_\gamma = 3,14 \cdot 12,4 \cdot 10^{-18} \cdot 1,4 \cdot 10^{9} \cdot 7,82 = 4,3 \cdot 10^{-7}$ Gy/s ≈ 1,58 mGy/h→0,17 R/h.

In a work [4] it is offered to calculate absorbed dose estimation under uniformly polluted infinite surface of the earth at a height of one meter in the energy range 0,5-3,0 MeV by the following formula:
$P=1,1 \cdot 10^{-11} S \cdot E$ (Gy/s),

Where;
E- emitted gamma- quantum (MeV), in our case 0,4 MeV [5].
S- surface pollution (Bq /cm^2), in our case – $1,4 \cdot 10^{5}$ Bq /sm^2.
So $P = 6,2 \cdot 10^{-7}$Gy/sec, that coincides with result of our calculations $P= 4,4 \cdot 10^{-7}$ Gy/sec in the limits of 30%.

Meeting the population irradiation limitation requirements in conditions of an accident

According to the NRS-99 (Table 6. «Predictable exposure levels, when urgent interference is necessary») for the whole body absorbed dose for two days should not exceed 1Gy. When these levels are exceeded, clinical determinate effects are possible. Let's compare this criterion with estimate data.

According to materials about accident consequences liquidation [7] the γ- radiation dose power level in city Pripyat during first day of the accident varied:
Pγ(t=1) =180 ÷ 500 mR/h→ 1,6 ÷ 4,4 mGy/h

On the second day of the accident:
Pγ(t=2) = 400 ÷ 1500 mR/h →3,5 ÷ 13,2 mGy/h

According to our calculations dose power on the first day was ≈1,6 mGy/h. Let's estimate prevented dose for first two days using calculated value *λэфф=0,11 (1/day*) and literary data about dose power.

For the first day:

$$D1 = \frac{P_\gamma}{\lambda} \cdot (1 - e^{-\lambda t}) = \frac{(1,6 \div 4,4) \cdot 24}{0,11} \cdot (1 - e^{-0,11 \cdot 1}) = 230 \div 634 \text{ mGy}$$

For the second day prevented dose was:
D2 = 504 ÷ 1900 mGy

So, in accord to [8, table 6.1.] with any dose power variations population evacuation must be obligatory!

27.04.86 in 14.00 o'clock the evacuation of Pripyat citizens began and in 19.00 o'clock it was over.

2.05.86 Government commission made a decision to evacuate citizens of 30km zone. In all 116000 persons from 188 settlements were evacuated.

Application Conditions of the Present Model

1. It is considered that tested sample is representative sample – it means that it has radionuclide composition and activity typical for the local area, because it is obvious that sample composition and activity depend on time and place of sampling.

2. To apply the present model it is necessary that isotope ratios in sample correspond to release isotope ratio, and release isotope ratio should correspond to isotope ratio in reactor active zone at the moment of release, because for calculations we use reference data.

3. To estimate release density it is necessary to know the square of sampling. Sampling square in this work has the size of adult palm, i.e. it is near 100 -150 cm^2. The calculation is made for the 100 cm^2 square.

4. Chernobyl fall-out is characterized by «mezo» and «micro heterogeneity» - during sampling with «envelope» on the square 100 m^2 the results of sample analysis differ several times [1,2], this gives the main error (mistake) during dose power determination on the sampling territory and it cover all other errors. Let's remind that dose power variations in city Pripyat were 1,6–4,4 mGy/h.

3. Moscow;
   According to work [12] average specific activity value of ^{137}Cs in Moscow soils (1-15 cm) on 2005 year on experimental data is 6 Bq/kg. So the specific activity of cesium in 1986 year was 11 Bq/kg. Absorbed dose power for 2-π geometry (semi-infinite radiant space):
   $P_\gamma = 2\pi * A * Гси / \mu^*_e$

   Where;
   absorption coefficient for air μ^*_e=0,0028 m^2/kg
   gamma-constant=21,33*10^{-18}(Gy*m^2)/(sec*Bq)
   P_γ=2 π*11*21,33*10^{-18}/0,0028 =5,26*10^{-13} Gy/sec=1,03*10^{-9} Gy/h=8,8 mkGy/year.

   According to [3] we apply coefficient 0,15 (mkSv/ mkGy) for city, and receive average effective dose - 1,3 mkSv/year.

   Let's compare it with literary data. According to [13] pollution surface density in Moscow by ^{137}Cs was A_s=630 Bq/m^2 or 17 mCu/km^2, conversion factor [3] - 0,565 (mkGy/year)/(mCu/km^2)–then P_γ=0,565*17=9,6 mkGy/year, if we multiply this value on coefficient for – 0,15 (mkSv/ mkGy) we will have dose equals 1,44 mkSv/year, that is well coordinated with received data.

Compare it with dose that is formed by natural radionuclides (natural background) in USSR in 1980-1981 years – effective equivalent dose – 1 mSv/year[6]. Effective dose, formed by Chernobyl cesium is 0,13 % of background irradiation.

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