PHYSICOCHEMICAL PROCESSES OCCURING UNDER ACTION OF IONIZING RADIATION IN SARCOPHAGUS

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The result of analysis of environment ionization process inside Sarcophagus owing to alpha-, beta- and gamma-radiation processes with forming of ions. It is shown that as a result of ionization and physicochemical transformations gaseous mixtures, which are dangerous for personnel's health and can influence upon general technical safety of Sarcophagus, can release into atmosphere.

The significant part of inspected premises of Chernobyl Sarcophagus is the container of water and damp (about 3000 m^3), spent nuclear fuel (approximately 56 tons), Fuel Containing Masses (FCM) (more 800 m^3), fine-dispersed fuel dust - 10 tons, graphite blocks and graphite dust, volume 700m^3. In given premises the dose rate of radiation makes up from 10 up to 3000 R/hour. At the interaction of the ionizing radiation of complex structure i.e. alpha-, beta- and gamma-radiation with environment the ionization process occurs.

The run of the energetic electron in air causes, basically, to breaking of two-atomic molecules on the positively charged ion of nitrogen (or oxygen) and neutral atom of nitrogen (or oxygen).

Knoched - on electron with the energy up to several keV runs the significant distance ionizing the other molecules with generation of free electrons, energy of which is sufficient for appreciable expulsion from its maternal atom.

Whereat, except positive charged atom and neutral atoms will product. As a result of ionization the ions O^-, H^+, N^- occur. The effect of the recombination of positive ion with electron is not significant, as far as electron has time to escape on appreciable distance.

The aqueous vapor as a result of ionization includes the ions: H^+, OH^-, O^-. While the electron adherence results to forming of H_2O. The random encounter of ions and single atoms leads on first stages to the appearance of the molecules: NO, NO_2, H_2O and O_3.

In media where β-radiation acts process of condensations of molecules NO, NO_2, H_2 and O_3 is significantly intensified.

The influence of α-radiation with strong ionic density in the layer of graphite several microns (10 - 20 micrometers) thick consists that as a result of elastic collisions of α- particle with carbon atoms (graphite) at the surface layer of air the ions of C^+ and atoms of C, as well as hydrocarbon molecules, CO, CO_2, CH_2, C_2H_2, H_2 etc. occurs.

Hereinafter the molecules: NO, NO_2, CO_2 interacting with damp, through series of chemical reactions result to formation of acids, which interacting with metal-constructions bring to their corrosion. In addition, the collateral effects of such reactions - is release of hydrogen.

Thus, the yielding of combustible gases (CO, CH_4, C_2H_2, H_2 etc.) from environment can lead to the formation of explosive mixtures. On this reason it is necessary to estimate the rate of recombination and adhesion in dependence on concentration of ions and neutral molecules. Let us note some features of these processes.

1. The recombination coefficient of free electrons and positive ions appears for majority of gases identical one.

$$A_{(e^- + z^+)} = \mathbf{2 \cdot 10^{-10}}\ \text{cm}^3\ \text{sec}^{-1}.$$

2. The recombination coefficient of positive and negative ions are almost uniform for all gases

$$A_{(z^- + z^+)} = \mathbf{1.5 \cdot 10^{-6}}\ \text{cm}^3\ \text{sec}^{-1}.$$

3. The "sticking" probability of electrons to molecules is depended on electrons velocity and strongly depended on the nature of gases: for inert gases, H_2, N_2, CO_2 the sticking probability is equal to zero.

For oxygen $\quad A_{(e^- + O_2)} = \mathbf{(1\text{–}2) \cdot 10^{-4}}\ \text{cm}^3\ \text{sec}^{-1}$.

For the vapors of water $\quad A_{(e^- + H_2O)} = \mathbf{0.5 \cdot 10^{-5}}\ \text{cm}^3\ \text{sec}^{-1}$.

Given work makes the approach of valuation, as a first approximation, of yielding of ions at interaction with the ionizing radiation of environment, internally of premises of Sarcophagus.

Let us make the following assumptions:

1. Average energy of γ-radiation - 600 keV, β-radiation - 400 keV, α-radiation - 5 meV
2. Gamma-radiation induces ionization uniform on all the extent of air in the premises, β-radiation 0.5 meter, α-radiation - 2 cm.
3. Ionization process makes a sensible contribution into destruction of molecules of nitrogen, oxygen - the main components of air and water vapor.

Assume that in some initial point in time the concentration of two types of ions and molecules N_1^0 and N_2^0. To be clear, let us assume that $N_1^0 \le N_2^0$. The quantity of recombined molecules $dN_p(t)$ in the point in time t is equal to:

$$\mathbf{dN_p(t) = AN_1(t)\ N_2(t)\ dt}, \qquad (1)$$

where **A** - is sticking probability $(\text{cm}^3\ \text{sec})^{-1}$;

In the proportion N_1 and N_2 it may be assumed two types of the dependence of quantity of recombined molecules in time:

1. Rates of decrease for N_1 and N_2 are equal and constant;
2. Concentration of N_1 and N_2 are changed in time exponentially;

a) Examine the first case, $\mathbf{N_1^0 \le N_2^0}$:

$$\mathbf{N_1(t) = N_1^0 - Bt}$$
$$\mathbf{N_2(t) = N_2^0 - Bt,)} \qquad (2)$$

where **B** - coefficient of stick $[\ 1/(\text{cm}^3\ \text{sec})]$;

$$\mathbf{N_P(\tau) = N_1^0 N_2^0 A\tau - (N_1^0 - N_2^0)\ (AB/2)\ \tau^2 + (AB^2/2)\ \tau^3}. \quad (3)$$

Relation between **A** and **B**:

$$B = [N_1^0(3N_1^0 - N_2^0)] \cdot A/6 \tag{4}$$

Full time of recombination of τ_0 :

$$\tau_0 = [1/(3N_1^0 - N_2^0)] \cdot 6/A \tag{5}$$

b) Examine the second case, $N_1^0 \leq N_2^0$:

$$N_1(t) = N_1^0 e^{-\mu t};$$

$$N_2(t) = N_2^0 - N_1^0(1 - e^{-\mu t}) = (N_1^0 - N_2^0) + N_1^0 e^{-\mu t}; \tag{6}$$

$$N_p(\tau) = \eta/\mu\, [N_1^0(N_2^0 - N_1^0)(1 - e^{-\mu t})] + \eta/2\mu\, [(N_1^0)^2(1 - e^{-2\mu t})], \tag{7}$$

where μ, η - coefficient of dimension (sec^{-1}).

Relation between μ and η:

$$\mu = \eta/2\, (2N_2^0 - N_1^0); \tag{8}$$

As concentrations are changed in time exponentially, so let us define the time τ_0, for which:

$$N_p(\tau_0) = N_1^0/2; \tag{9}$$

from the equation:

$$e^{-\mu\tau_0} = \{[(N_2^0 - N_1^0)/2]^2 + S\}^{-S} - (N_2^0 - N_1^0)/2\, N_1^0 \tag{10}$$

When small τ_0 both distributions result to the same expressions:

$$N_p(\tau_0) = N_1^0\, N_2^0 At \tag{11}$$

Let us calculate the process of ionization of the environment when α, β, and γ - radiation and possible (α, β) - reactions.

Let us assume that FCM are covered by the layer of fine - dispersed graphite dust with the size of grains about 1-3 mm. Alpha-particles escaping out from the surface of the FCM stratum with the unit activity of trans-uranium products per 1 gram of uranium of $4{,}37 \cdot 10^6$ Bq/cm^3 FCM pierce several grains of graphite. At that due to Coulomb scattering on the border of grains some individual atoms of carbon can be knocked out. After a small time these atoms come to the air of the premises. The main process defining the magnitude of mentioned molecules is Coulomb scattering of nucleus scattering of the atoms of helium on carbon. As in course of movement the α-particle losses its energy, such a nucleus scattering can happen by any energy between Ea - the energy of α-particles when decay, and E_{min} - the energy sufficient for knocking out the atom of carbon from the crystal lattice.

Differential cross-section for the System of Center of Mass (SCM) can be presented as follows:

$$d\Phi(\theta) = 0.8139 \cdot 10^{-26}\, L^2\, \sin\theta d\theta/\sin^4\theta/2, \tag{12}$$

where $L^2 = z^2 Q_z^2/E^2$,

z - charge of α-particle;

Q_z - charge of target nucleus;

θ - α-particles scattering angle.

Integral cross-section of scattering of α-particles in angles superior to the θ is equal to:

$$\int_0^{\pi} d\Phi(\theta) = \mathbf{1.6278} \cdot 10^{-26}\ \mathbf{L^2\ ctg^2\theta/2}. \qquad (13)$$

For α-particles of big mass it is necessary to generate a multiplier $(1+m/M^2)$. Relation between the angle θ - in FCM and φ - angle of a recoil nucleus in Laboratory System of Masses (LCM):

$$\cos\varphi = \sin\theta/2 \qquad (14)$$

For ψ - angle scattering angle in LCM for colliding α-particle the following relation exists:

$$\cos\psi = (m + M\cos\theta)\ /\ [\ m^2 + M^2 + 2m\,M\cos\theta]^{-1/2}, \qquad (15)$$

where:

m - mass of particle;

M - mass of target nucleus.

For **m**=4, **M**=12, θ=20°, φ=80°, quantity of ψ equal 15°.

So when angles $\theta<20^0$ give the angles φ of scattering of recoil nucleus more then 80^0. In our case such processes are not very important as the nucleis of carbon can come to the air only when they are scattered forward with angles φ<80°. Energy of recoil nucleus can be defined from the following relation:

$$E_{rec} = E_0\ [2mM/(m + M)^2](1 - \cos\theta) \qquad (16)$$

For $\theta=20^0$, $E_{rec}=0{,}02E_0$, it means that for Ea = 1 keV atom of carbon will receive the energy E ≈ 20 eV - enough for its escape out from a crystal lattice. Integral scattering cross-section α-particles by angles $\theta>20^0$ for Ea = 10 keV is 10^{-19} cm^2, for Ea = 1 keV it will be - 10^{-17} cm^2.

Experimental data on recombination features point out that adherence of electrons happened only to molecules of oxygen and water. Parameters of ionization effect due to γ-radiation in the humid air are presented in the table 1:

Table 1

Gas	*Density, mg/cm³*	*Number of molecules, 10^{19} cm^{-3}*	*Relative percentage*	*Number of pairs of ions in 1 cm³ by dose (R/hour)*	*Ion, atom, molecula*
N_2	0.836	1.80	0.735	$0.426\cdot10^6$	e^-, N, $^-N_2$
O_2	0.225	0.48	0.196	$0.113\cdot10^6$	e^-, O^-, O_2
H_2O	0.051	0.17	0.069	$0.04\cdot10^6$	e^-, OH^-, H^+, H_2

In table 2 the estimation of time τo (the half decreating) of concentration of electrons calculated on the basis of data from the Table 1 and formula (5) at the different exposure dose rates is given.

For the course of (α, p)-reaction it is necessary that the energy of α-particles exceeded Coulomb barrier. Low energy of α-particles points out that this light nu-