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Analysis of physical processes that may cause the fragility and destruction of the lava-like fuel containing materials (FCM) of 4-th unit of Chernobyl Nuclear Plant (ChNP) has been carried out. The following processes were considered: influence of electric fields arising in medium with incorporated radionuclides, the role of the defect creation under irradiation of incorporated nuclides, residual mechanical strains caused by their cooling from the melting temperature in the time of the accident to the ambient temperature at present. It is shown that mechanical strains of such kind might be one of the causes of degradation and destruction of FCM. Different structure modifications of radiolysis of hydrogen sources are discussed. It is shown, that the β -irradiators ^{137}Cs and ^{90}Sr bring the cardinal contribution in generating of hydrogen. Depending on spent nuclear material dispersity, concentration of soluble forms of radionuclides, moisture saturation degree of FCM and water quality the account of radiolysis hydrogen yield rate is presented. The estimations of the safe content of hydrogen in the Sarcophagus premises air is estimated depending on leaktightness and the gas exchange rate.

Keywords: Sarcophagus, physic-chemical processes, radiolysis of hydrogen.

Fuel containing masses in the premises of the 4-th unit of the Chernobyl Nuclear Plant (ChNP) are the unique object, that is the mixture of the concrete, nuclear fuel and metal constructions in the fields of intense radiation. Study of the fuel containing materials (FCM) is important for determination of the nuclear and radiation safety of the object, reconstruction of the accident scenario, forecast of the FCM future behavior, understanding of the properties of this new object. A strong degradation of FCM with time is observed.

The importance of understanding of this process cannot be underestimated because it brings about the formation of the radioactive dust dangerous for the human beings and the environment. So in this paper special attention is paid to the determination of the physical mechanisms leading to the degradation of the FCM, namely, electric fields, defect formation and mechanical strains emerging due to the gradual cooling of the masses. Possible mechanisms connected with the water penetration into FCM are not considered here.

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 study of fuel-containing masses interaction with water in premises of the 4-th unit of ChNP with forming of radiolytic hydrogen is the important task for determination of the general safety in Sarcophagus. If the investigation yielding rate of hydrogen in dependence on nuclear spent fuel dispersity,

concentration of soluble forms of radionuclides, ionizing radiation dose rates, moisture saturation of FCM and quality of water a significant number of works is devoted [1—4], the data on the estimation of the safe maintenance of radiolytic hydrogen in every premises of Sarcophagus in dependence of genneticity degree and interchange of air degree, possibility of the appearance of the explosive and fire — dangerous situation are practically absent.

In the given work the numerical modeling of different variants of generation sources of radiolytic hydrogen at the action of radiation is carried out, and just as the qualitative estimation yielding rates of hydrogen at the interaction of water with FCM.

The characteristic of FCM and dose rates in the sarcophagus premises. Since the external protective containment of the Sarcophagus reactor 4 at the ChNP is not hermetically, it is impossible to exclude the penetration of water into puddles. The upper estimation of average annual penetration of water and moisture from atmosphere by rain and snow into the Sarcophagus premises, being assessed as long ago as to the beginning of the work upon hermetization of the roof is such:

- about 3000 m³ was received on the surface of the former reactor hall;
- 1800 m³ — on the surface of deaerating stand.

Penetrating into Sarcophagus water on its path from upper points into bottom ones gradually ruins the constructions, washes away FCM, carries away with itself the particles of nuclear fuel and soluble radionuclides. Gross activity of water on all γ -irradiating radionuclides is in the range from micro curie to two tens of millicurie per meter. The base component of activity is ^{137}Cs .

After the accident the rise of ground water have occurred, foundation-basement part have been incompletely or totally filled up by water, probably included ground one.

In unit "D" (turbine hall and deaerator) below zero point there are approximately 2500 m³ of water with activity from $2.7 \cdot 10^{-7}$ to $1.9 \cdot 10^{-6}$ Ci/l on ¹³⁷Cs. The water in turbine hall in its content approximates to industrial one, main contamination is oil and mast oils on it surface. There are about 500 m³ of water, draining down there from the reactor hall (unit "B"), in the compartment 001/3 of unit "VSRO".

In Table 1 the distribution of FCM, fuel assemblies and other sources of ionizing radiation in the premises of Sarcophagus are offered.

In the compartments at bottom points of unit "B" Up to 200 m³ of water accumulate when penetrating into them of precipitations and also of industrial water from system of Fuel dust depression, disposed in the central hall (see Fig. 1). Water accumulations in units "B" and "VSRO" compartments classify with medium-active radioactive wastes from $2.2 \cdot 10^{-7}$ to $2.9 \cdot 10^{-2}$ Ci/l. In this water, containing besides ¹³⁷Cs and ⁹⁰Sr, alpha-active and fission uranium ²³⁵U, ²³⁸U and plutonium products ²³⁹Pu, ²⁴⁰Pu, since every year content of the latter increases [5].

Various modifications, of structure of hydrogen sources are considered. The main radionuclides benign the puddles of Sarcophagus α , β - and γ -irradiators are taken into consideration. It is shown that β -irradiators ⁹⁰Sr and ¹³⁷Cs make the cardinal contribution into hydrogen generation.

Rated values of hydrogen yield rate depending on spent nuclear fuel and fuel containing masses dispersity, soluble forms of radionuclides concentrations, ionizing radiation dose rate, moisture-saturation of FCM and quality of water are shown. The estimation of safe content of radiolytic hydrogen in Sarcophagus air assumption of hermeticity degree and interchange of air degree.

The average values of chemical characteristics of water: solid residue 0.2 mg/cm³; pH — (7.3—9.6); hardness — (0.8—2.5) mg-ekv/kg; ox disability — (5—10) mg (O₂)/kg.

Table 1. The distribution of FCM, fuel assemblies and other sources of ionizing radiation in the Sarcophagus premises [5, 6]

The name of premises (premise, mark)	Form of FCM, fuel assemblies and its condition	Estimation of fuel quantity in FCM, fuel assemblies (on uranium), tons	Volume content of UO ₂ , %	Maximum exposure, R/hr
Central Hall 914/2 (35.50)	Fragments of reactors core	60—70	<14	~3500
Under premises of unit 4 (in shame «E») (35.50—40.00)		10—36	<14	~3000
South cooling pond	About 140 of fuel assemblies with spent fuel	~20	—	~3200
Under reactor premises 305/2, 307/2, reactor vault, shame «OR» (9.00—11.00)	Lavous FCM, fragments reactor core, dispersed fuel	~25	<9.0	1200—1300
Steam distributing corridor 210/5, 210/6, 210/7 (6.00)	Lavous FCM, dispersed fuel	25±11	2.4	~1200
Pressure suppression pool, 2 nd floor 012/13, 012/14, 012/15 (2.20—3.00)	Lavous FCM	8±3	1.1	~2000
Pressure suppression pool, 1 st floor 012/5, 012/6, 012/7 (0.00)	Lavous FCM	1.5±0.7	3.4	1200—1300
Premises of reactor unit "elephant foot" 217/2, 304/3, 303/3, 301/6 (6.00—9.00)	Lavous FCM	11±5	<8	~3000
Premises of reactor unit (35.00—68.00)	Fuel dust, "hot" fuel particles	~10	6—14	400—1200

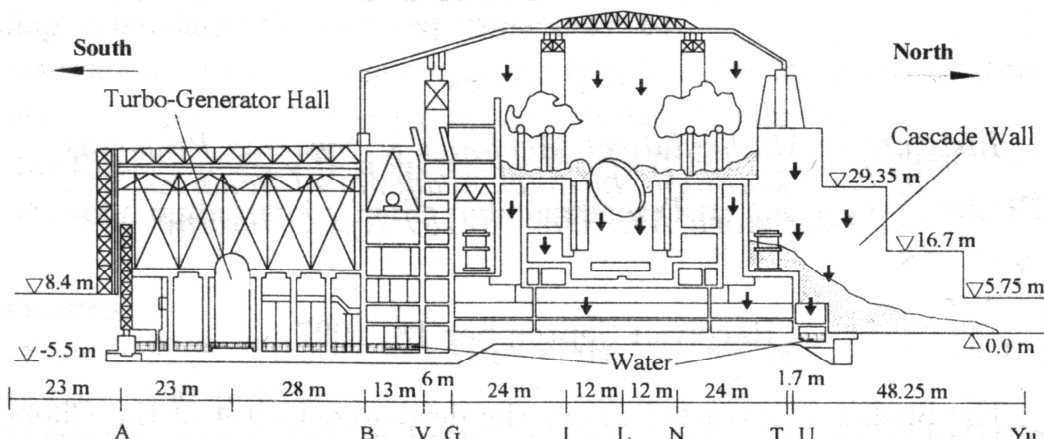


Fig. 1. Schematic representation of the Sarcophagus. Water pathways and locations within the premises (North-south cross — section along Axis 47)

Table 2. Specific α -, β - and γ -activities of FCM, fuel assemblies and other sources of radiation (in 25 years after accident)

Nuclide	Emission	Activity, Bq/g	Disintegration constant, sec ⁻¹	Averaged energy of disintegration, MeV	Specific doze rate, MeV/(cm ³ ·sec)
¹³⁷ Cs	γ	0.41—10	$7.3 \cdot 10^{-10}$	0.565	$0.53 \cdot 10^8$
⁹⁰ Sr	β	$8.3 \cdot 10^6$	$7.82 \cdot 10^{-10}$	0.196	$0.38 \cdot 10^8$
¹³⁷ Cs	β	$4.1 \cdot 10^6$	$7.28 \cdot 10^{-10}$	0.180	$0.17 \cdot 10^8$
²³⁸ Pu	α	$5.36 \cdot 10^6$	$2.5 \cdot 10^{-10}$	5.49	$6.73 \cdot 10^6$
²³⁹ Pu	α	$4.54 \cdot 10^6$	$9.0 \cdot 10^{-10}$	5.15	$5.35 \cdot 10^6$
²⁴⁰ Pu	α	$6.32 \cdot 10^6$	$3.3 \cdot 10^{-10}$	5.15	$7.4 \cdot 10^6$
²⁴¹ Am	α	$1.22 \cdot 10^6$	$5.1 \cdot 10^{-10}$	5.50	$1.5 \cdot 10^7$

Isotope specific activity makes [6]: ¹³⁷Cs — (3—8)·10⁸ Bq/l; ⁹⁰Sr — (4—9)·10⁶ Bq/l; ^{239,240}Pu — 850 Bq/l; Uranium content — (6—9)·10 μg/l.

In Table 2 specific α -, β - and γ -activities of FCM, fuel assemblies and other sources of radioactive ionizing (in 25 years after the accident) are offered.

Radiolysis of water in puddles under the action of ionizing radiation may be accompanied by molecular hydrogen formation, which will be educed and accumulated in stagnating zones of Sarcophagus. The probability of such process depends upon dispersity of spent nuclear fuel and FCM, being in various structural and composite forms, moisture-saturation degree of fuel-containing structures, radiation dose rate, quality characteristic of radiolysis water, value of radiation chemical hydrogen yield (G_{H_2}). For transuranic elements ^{238, 239, 240, 241}Pu, for α -particles with averaged energy of radiators $E_0=0.021$ MeV — $G_{H_2}=1.7$ mol/100 eV; for β -particles of ⁹⁰Sr and ¹³⁷Cs with $E_0=1$ MeV — $G_{H_2}=0.1$ mol/100 eV; for γ -radiation of ¹³⁷Cs with $E_0=0.66$ MeV — $G_{H_2}=0.3$ mol/100 eV.

Some mechanism of FCM to destruction. The following processes are considered: influence of electric fields arising in medium with incorporated radionuclides; the role of the defect creation by irradiation of incorporated nuclides; residual mechanical strains caused by their cooling from the melting temperature in the time of the accident to the ambient temperature at present.

1. Electric fields in FCM

One of possible mechanism of reduction of the mechanical durability of FCM is occurrence of significant electrical fields as a sequence of the accumulation of charges generated by ionizing particles [7] and others causes.

A general model of FCM is a SiO₂ matrix with impurities, structural defects, inclusions of various nature, radionuclides, nuclear fuel [8].

Microstructure of FCM is very complex, physical parameters of such highly inhomogeneous structure

are unknown. Therefore, only estimates are possible and at present stage of research it is expediently to use simple models with the minimum number of parameters.

The following processes have been considered:

1. Accumulation of charge in “hot” particles. For “hot” particles with the radius R , the activity A , atomic number z , the conductivity a and the density ρ one can obtain for electric field at the particle matrix boundary:

$$E = \frac{A \cdot z \cdot a \cdot \rho \cdot R}{3\beta} \quad (1)$$

The estimates for the parameter values $\rho=10$ g·cm⁻³; $\beta=l/10^{12}$ ohm·cm, $R=10$ μm, $A=10^8$ sec·g gives $E=0.053$ V/cm.

2. Typical processes for inhomogeneous and disordered solids: fields connected to inclusions of other phase and regions of radiation damages, electric fields, caused by a contact-potential difference. Typical values of electric fields in inhomogeneous semiconductors are up to 10² V/cm. So electric fields are small and cannot significantly influence the destruction of FCM.

2. Radiation induced defect formation doe to the incorporated radionuclides

Effect of the radionuclides incorporated in the FCM on the atom displacements is calculated. Main contribution into the displacement comes from the α -particles. The displacement rate is calculated for the Si and O atoms in the vicinity to the UO₂ inclusion. The estimates are fulfilled for the parameters: activity $A=2.7 \cdot 10^7$ sec·g, inclusion density $\rho=10$ g·cm⁻³, particles energy $a=5$ MeV, the threshold displacement energy will 16.5 eV [9].

The calculations result in displacement rate $5 \cdot 10^{-12}$ dpa/sec. This value of the displacement rate during the entire time of the irradiation after the accident may significantly change electro-physical, optical and other properties of dielectrics but it is not sufficient for the significant effect on the mechanical properties, i. E. To explain the observed degradation of the FCM.

3. Residual strains in FCM emerging at cooling

Let us estimate another of the possible mechanisms for the cracking of the FCM containing masses. It is connected to the residual strains that emerge in the lava due to the cooling. Important thing here is that lava consists of great variety of inclusion.

Let us consider an inclusion in the FCM that was formed in the melted lava at high temperature T_0 . It is natural to assume that during formation of an inclusion at high temperature mechanical strains are absent. Due to the different thermal expansion coefficient of the FCM matrix and material of the inclusion mechanical strains appear leading to the microcracks and finally,

to the destruction of the FCM. To do estimates we consider a spherical inclusion in an isotropic medium. The equations of mechanical equilibrium are solved waffle boundary conditions of continuity of the displacement field vector and the pressure at the boundary. The material is let to expand (contract) free at the infinity.

Solving the appropriate equilibrium equations and satisfying the boundary conditions one obtains mechanical strains outside the inclusion with radius R at foe point:

$$\delta = -\frac{8}{3}\mu_g \cdot (T - T_0) \cdot \frac{R^3}{r^3} \cdot K_n \cdot \frac{a_n - a_g}{3K_n + 8\mu/3}, \quad (2)$$

where T — the recent ambient temperature; r — the radius dislocation; μ and K — the elastic modules of materials a is the thermal expansion coefficient; indexes g and n stand for materials of the matrix and the inclusion, respectively.

One can clearly see from the obtained formula that mechanical strains are proportional to the difference of the temperatures and difference in the expansion coefficients of the inclusion and the material of the matrix. It is interesting to note that the strains at the boundary do not depend on the radius of the inclusion.

Let us perform some estimates that will show magnitudes of the appearing strains. It is widely known that FCM were formed from the lava at temperature of order of 1500 °C. At present the temperature of the FCM is about 50 °C. For the estimates we will take silicon dioxide (which is the main constituent part of the FCM) as the matrix material. The thermal expansion coefficient varies in the broad range depending on the glass type (from $6 \cdot 10^{-7} \text{ K}^{-1}$ to $6 \cdot 10^{-6} \text{ K}^{-1}$).

And for the fuel UO_2 inclusion it is according to different authors from $9 \cdot 10^{-6} \text{ K}^{-1}$ to $13 \cdot 10^{-6} \text{ K}^{-1}$. So according to formula (2) the emerging strains can reach up to $10^{-2} \mu\text{g}$. This value by its magnitude is of order of the glass fragility threshold.

The calculation of radiolysis water. Power density of sources of α - and β -radiation was calculated by formula [1, 10]:

$$Q_m = g_m \cdot E_0 \cdot X_{\text{UO}_2} \cdot P_{\text{UO}_2}, \quad (3)$$

where g_m — m -isotope specific activity; E_0 — average disintegration energy; X_{UO_2} — volume content of nuclear fuel in puddles; P_{UO_2} — nuclear fuel density.

The rate of emitting hydrogen was determined by formula [11]:

$$W_{\text{H}_2}^m = \frac{d(\text{H}_2^m)}{dt} = \frac{G_{\text{H}_2}^m \cdot Q_m}{100N_a}, \quad (4)$$

where N_a — Avogadro number.

Potential explosive-danger from radiolytic hydrogen depends on hydrogen yield and Interchange of air rates ratio in the Sarcophagus premises as a result of natural convection, for calculation of the outlay of air for blowing off of hydrogen was used the formula:

$$L_m = k \cdot W_{\text{H}_2}^m, \quad (5)$$

where k — coefficient equal to 250.

Interchange of air degree was determined by formula [10]:

$$N = \frac{L_m}{V}, \quad (6)$$

where V — the volume of examined premises.

An attempt of estimation as in the first approximation of the rate of hydrogen yielding as a result of water radiolysis in the Sarcophagus environment under the following conditions is made in this paper: fuel dust is under a layer of water; a layer of water is covered by spent nuclear field cluster; water mass ratio of which is given is in pore space of FCM.

The quantity of fine-grained aerosol particles (fuel dust) in the reactor pit and under reactor premises (rooms 504/2 and 305/2), south and north premises of former central hall and others is 10 tons [5].

The first type of fuel dust is the fragments of Irradiated nuclear fuel, consisting of UO_2 . The initial microstructure of fuel tablets is distinctly traced in particles measuring 80—700 kVt. Dimensions of separate mycroblocs are varied from 2 till 7 kVt with density (7—9) g/cm^3 and relative hardness 490—510 kg/cm^2 .

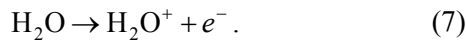
The second type — the particles of mixed composition, in which the main matrix component are Uranium oxides — (U_4O_9 and UO_2) and zirconium, the ratio of which is changed in wide range. The largest dimension of dispersed irradiated fuel particles is (1.0—1.2) μg .

The structure of “hot” particles is different-cellular, cavity-porous etc. in the calculations the layer of dust 10^{-3} m thick was studied, which is covered by water layer 10^{-2} m thick and surface area 1 m^2 . Let us suppose, that α -irradiation is utterly absorbed inside fuel particles, and release energy is spent on heating of water. Half energy of β -radiation is absorbed inside fuel particles, the second half by the layer of water.

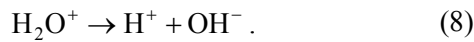
We neglect self absorbing of γ -radiation inside fuel particles.

Nuclear fuel, total quantity of which is estimated at 56 tons consists of fragments of active zone of the reactor spent fuel pools, fuel channel assemblies, fuel tablets, with initial proton content: ^{238}U — 98 %, ^{235}U — 2 %. Uranium dioxide, high uranium oxides and uranium hydro carbonates form chemical composition of fuel. Let us suppose, that nuclear fuel layer $5 \cdot 10^{-2} \text{ m}$ thick and 120 kg/m^3 density is overflowed

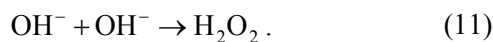
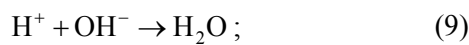
with water layer 10^{-2} m thick and surface area 1 m^2 . In the process of radiolysis of water hydrogen, hydrogen peroxide and oxygen will be generated at final stage [10]:



The ions H_2O^+ can be in different electron states ones of which are stable and others break down. Concentration of oscillation energy on certain bonds may results in ion decomposition:



Free radicals H^+ and OH^- doesn't satirically separated, therefore they can with the great probability recombine with forming of molecular products [11]:



Radioactive-chemical yields and water radiolysis products lifetime are offered in the Table 3.

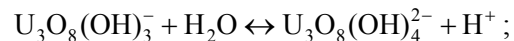
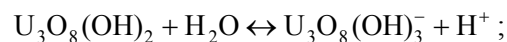
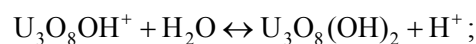
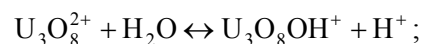
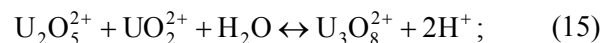
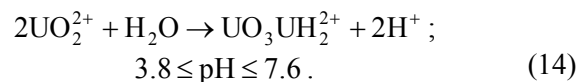
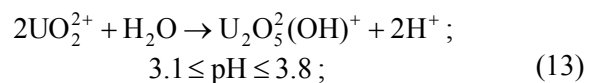
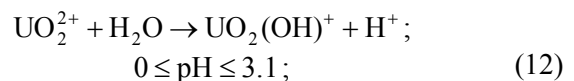
It is obvious, hydrated electronrons and OH^- radicals form in deficiency per every 100 eV of absorbed radiant energy at neutral pH value.

The specific feature of aqueous solutions, containing Tran Uranium Elements (TUE) — ^{235}U , ^{238}U , ^{239}Pu , ^{240}Pu , ^{241}Am — is generation of different forms of hydrated ions. Hydrolysis of TUE with forming of hydrogen ions may occur according following reactions [12].

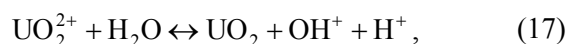
The experimental investigations of the object's aqueous solutions of radioactive elements, with the average concentration of uranium 10 mg/1 and more, show that their pHs are usually in the intervals of (6.5—7) and (9—10). The most interesting are solutions in those places of interaction of the nuclear magma with water, where we observe a substantial yield of uranium in water and a high level of the α -, β - and γ -radiation of water. High values of pH are pointing to a certain shift of equilibrium (due to chemical reactions) between groups OH^- and H^+ , and the fact that ions of hydrogen, which are formed in the process of radiolysis, hydratation and other characteristic reactions, quickly react transforming into molecular

hydrogen and molecules in water. Besides, atomic hydrogen can be formed in reduction reactions. This value of pH corresponds to the basic solutions where the concentration of OH^- groups is dominant and there is a deficit of hydrogen ions H^+ . From this point of view it is necessary to analyze chemical reactions with UO_2^{2+} , PuO_2^{2+} , AmO_2^{2+} in the system “nuclear magma-basic solution” taking into account radiolysis and alkalization processes.

In aqueous solutions U, Pu and its isotopes, strontium Sr, cesium Cs and other radioactive elements produce different forms of hydrated ions, molecules, double and mixed complexes, mono- and polynuclear products hydrolysis and colloid particles. Let us analyze a set of reactions where uranium is involved. The approximated schematic set of reactions can be seen as follows. In aqueous solutions uranium has +3, +4, +5, +6 states of oxidation. The stability of valence states of uranium in a solution is characterized by the sequence $\text{U}(6) \geq \text{U}(4) \geq \text{U}(3) \geq \text{U}(5)$. The stabilization of the large positive charge of uranium $\text{U}(6)$ takes place because in aqueous solutions urinal ion UO_2^{2+} is produced. The hydrolysis of urinal ions is governed by the reactions [1]:



The hydrolysis of uranium UO_2^{2+} can be explained in terms of the formation of monomer $\text{UO}_2(\text{OH})^+$ by the reaction:



and dimer UO_2 , UO_2^{2+} by the reaction:

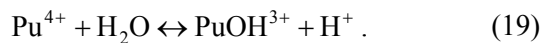


Table 3. Radioactive chemical yield and lifetime of primary products of water radiolysis

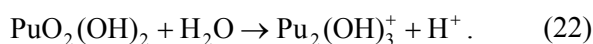
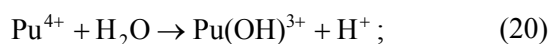
Products of water radiolysis	Value G_{H_2} , molecules/100 eV	Lifetime τ , sec
e^-	2.6	10^{-15}
H^+	0.6	10^{-13}
OH^-	2.6	10^{-11}
H_2O_2	0.75	$8.6 \cdot 10^5$
H_2	0.45	10^2

A lot other possible reaction channels can fit this scheme, thus constituting a complicated reaction network.

Plutonium in aqueous solutions has five degrees of oxidation: +3, +5, +6, +7. Pu^{4+} has the most interesting properties. It is in the form of hydrated ions $\text{Pu}(\text{OH})_{6-8}^{4+}$ in polymer-acidic solutions. Also for some concentrations of Pu, when pH of the solutions increases, the hydrolyses of Pu^{4+} ions leads to the formation of mononuclear hydroxocomplexes $\text{Pu}(\text{OH})^{3+}$, $\text{Pu}(\text{OH})_2^{2+}$, which further can exhibit polymeric and colloid properties. The hydrolysis of Pu(4) takes place at the concentration of H^+ ions less than 0,3 M [1]:



Here we have to distinguish between two types of reaction products: monomer hydrated ions of $\text{Pu}(\text{OH})_n^{(4-n)+}$ type and products of polymerization, which are formed at the same values of acidity. A slow decrease of acidity in the region of H^+ hydrolysis below 0,3 pH helps the formation of polymers. The dissolving of acidic solutions with water causes an instantaneous local decrease of acidity to the values favorable for polymerization. The formation of polymers is also accelerated when moderately acidic solutions of plutonium are heated [1, 10]. The process of polymerization is accompanied by polarization of a water molecule by two plutonium ions. Six-valiant plutonium, producing plutonic PuO_2^{2+} with oxygen, actively hydrolyses in water according to the reactions [1]:



At some particular values of pH this leads to the formation of colloidal complexes and polymer structures.

It was supported in the calculations, that the conditions of absorption of radiation energy of a flat extended source are realized.

Total absorption of radiation energy for α - and β -irradiators takes place (thickness of the water layer is larger than α - and β -particles. For γ -radiation absorption depends on thickness of the water layer. It was supported, that dose, rate of γ -irradiators was about $5 \cdot 10^{13}$ eV/(g·sec).

As a result of high-temperature processes during accident melting of zirconium casing of fuel channel assembly, interaction of zirconium melt with uranium dioxide with forming of uranium-zirconium oxygen phase take place. This melt formed FCM 800 m³ in number during the interaction with construction materials (serpentines, concrete, sand) and air. It is

possible to choose conditionally the most typical modifications of FCM with mass ratio of the uranium: brown ceramics 9—10 %; black ceramics 4—8 %; slageous and pumise-stoneous <1 %.

Tree processes, leading to destruction of monolithic FCM are revealed: destruction of superficial part and forming fuel containing particles on it; eliminating radioactive hydrogen during interaction with water; cracking of matrix because of inner stress.

The most real of all processes leading to brittleness is the mechanism related to getting cold of FCM and impregnating of them with water and moisture.

The calculation of hydrogen generation rate was carried out for water and moisture being in cracks of FCM, quantity of which is one ton. It was supposed that FCM contains such oxides in percents by mass: UO_2 — 10 %, CaO — 10 %, K_2O — 2 %, Fe_2O_3 — 8 %, ZrO_2 — 4 %, SiO_2 — 60 %.

Water with ox disability 2 mg/l — 10 % contains admixtures of organic compounds — oil and surface active substances — 1 %. Let us suppose that absorption of radiation by water depends on nature of radiation: α -irradiators are regarded as surface irradiators and total absorption of energy occurs in the layer of water and FCM, equal α -particle range.

Let us consider, that absorbed by water energy which is in proportion to electronic (or mass) ratio of water in FCM, is conditioned by influence only of β - and γ -radiation. The results of calculation of hydrogen generation rate from puddles in the Sarcophagus under given initial data are shown in the Table 4.

Table 4. Calculation of H_2 generation rate from puddles in the Sarcophagus under given initial data

	Specific activity, Bq/kg			W_{H_2} , m ³ /hr
	g_α	g_β	g_γ	
Fuel dust	$3.65 \cdot 10^3$	$4.28 \cdot 10^3$	$0.36 \cdot 10^6$	$1.24 \cdot 10^{-4}$
Spent nuclear fuel	$6.32 \cdot 10^3$	$8.31 \cdot 10^3$	$0.42 \cdot 10^6$	$5.53 \cdot 10^{-4}$
Fuel-containing masses	$5.22 \cdot 10^2$	$7.93 \cdot 10^2$	$0.21 \cdot 10^5$	$0.98 \cdot 10^{-3}$

The determination of the fire-explosive dangerous situation. Fire/explosive ones are events, realization of which results in forming of an explosive mixture (burnable mixture) and the appearance of a source (a cause) inciting an explosion (striking).

A probability of the appearance of the explosion in Sarcophagus during a year is calculated by the formula [13—15]:

$$Q(Sr) = 1 - \prod_{i=1}^h [1 - Q_i(HF)] , \quad (23)$$

where $Q(Sr)$ — a probability of the appearance of the explosion in Sarcophagus during a year; $Q_i(HF)$ — a probability of the appearance of an expulsion in a i -th

compartment of Sarcophagus during a year; h — quantity of the Sarcophagus compartments; HF — factor of fire/explosive danger.

We rely to Explosive/Fire Hazards factor $HF(E/F)$ an indication on an array of indications whose change in time may cause explosive/fire or ignition if not to suppress tm dynamics of the event [13]. Explosive/fire category is determined of specifications and depends upon standards of protected object.

Thus $HF(E/F)$ identify possible explosive/fire causes (combustible media, oxidizer, ignition source, energy transfer mechanisms) and the character of consequences.

As an information toil term $HF(E/F)$ include two parts — “a priori” (characterizing possible explosive/fire hazards level and consequences). “A posteriori” part in $HF(E/F)$ is derived by prognostic simulation or real explosive/fire damage estimation. Typically the premises of Sarcophagus differ in structure of $HF(E/F)$ from other objects in “a priori” (specific technologic midis, nuclear fuel, FCM, sources of radiolytic hydrogen, radioactive elements etc.) therefore in “a posteriori” (ecology, social and material damage from explosive/fire, in Sarcophagus is much higher than in other nuclear objects). Presently there exist various methods for classification and analysis of $HF(E/F)$ indication.

The inner events introduced are[14]: technical or technological out coming e. g., in premises Sarcophagus processes (radiolysis, radiation heating of structure materials, Viegner’s effect chain reaction in a FCM etc.); In — plant management events (personnel feather; inadequate regulations and guidelines).

External events may cause explosive/fire are kept with natural phenomena such as earth quake water spout storm, flood lightning tornado etc. and events of technological origin — aircraft accidents, transport accidents gas clouds of chemical origin, shock wave impact etc. We also refer here nuclear substance explosive/fire geared with sabotage and terrorism. Ignition source indication are divided into nuclear (radiation heating, heat transfer, convection etc.), thermal (self ignition, heated surface), mechanical (friction, strike, wear), electric (lightning strikes; electrostatic charges, spark discharges, short circuits etc.), physicochemical (vapor of water, reaction, explosive mixture origin, radiolysis of water etc.).

Combustible items media exhibit the following substance status: gas items (oxygen, hydrogen explosion mixture, carbon etc.), FCM and nuclear fuel (hard from, structural material, graphite, cable insulation etc.), liquid items (transformer oil, lubricant oil, acids, alkalis etc.).

Factor “a posteriori” indication in Sarcophagus fall into four categories: factors impacting nuclear safety, factors impacting nuclear safety indirectly, factors impacting radiation safety. The first and second categories

should be listed in guideline regulations accounting individually each object.

Though preserving nuclear safety explosive/fire may cause radiation safety failure therefore assuming radionuclides release and encompassing of urgent measures performance (repair, technological, deactivation, rescue etc.) in radiation dangerous conditions.

Let us make some assumption for eyetic compartment of Sarcophagus [15]: K_0 — concentration of hydrogen, which can form an explosive mixture (detonating mixture); N_0 — quantity of explosion causes; M_0 — quantity of explosion sources; L_0 — quantity of energy transfer mechanisms by the explosion. Then

$$Q(HF) = 1 - \prod_{K=1}^{K_0} \prod_{N=1}^{N_0} \prod_{M=1}^{M_0} \times \prod_{L=1}^{L_0} [1 - Q(HF_K) \cdot Q(HF_N) \cdot Q(HF_M) \cdot Q(HF_L)], \quad (24)$$

where $Q(HF_K)$ — a probability of forming of a detonating mixture; $Q(HF_N)$ — a probability of the appearance of a cause of N -type explosion; $Q(HF_M)$ — a probability of the appearance of a cause of M -type explosion; $Q(HF_L)$ — a probability of transfer of the mechanical energy of L -type explosion; K_0, N_0, M_0 and L_0 are the maximum magnitudes of either events.

Assuming that every $Q(HF) < 0.1$, i. e. at a sufficient protection with K, M, N, L of a compartment (at a small probability of display of either event in a given compartment) it is possible to assume that

$$Q(HF) = \sum_{K=1, N=1, M=1, L=1}^{K_0, N_0, M_0, L_0} Q(HF_K) \cdot Q(HF_N) \cdot Q(HF_M) \cdot Q(HF_L). \quad (25)$$

Let us imply that concert of branch is a product under the $\Sigma(\dots)$ — signee for concrete K, N, M, L .

Let us examine more minutely the concrete magnitudes of separate probabilities:

$$Q(HF_K) = Q(B_K) \cdot Q(E_K) \cdot Q(O_K), \quad (26)$$

where $Q(B_K)$ — a probability of the appearance of k -th quantity of hydrogen for forming of a detonating mixture; $Q(E_K)$ — a probability of forming in k -th detonating mixture of stored energy; $Q(O_K)$ — a probability of the appearance of an oxidizer for k -th detonating mixture, $Q(O_K) = 1$.

$$Q(B_K) = Q^{cond} \cdot \sum_{p=1}^3 Q(A_p), \quad (27)$$

where Q^{cond} — a conditional probability of the appearance of any N -cause (internal, external), which is able

to provoke the explosion of k -th detonating mixture; $Q(A_p)$ — a probability of the permanent presence or the appearance of k -th detonating mixture.

$$Q(E_k) = \int_{W_{\min}}^{\infty} f(W) dW, \quad (28)$$

where W — a stored energy of the detonating mixture; $f(W)$ — density of distribution of detonating mixture stored energy.

The conditions inside containment prior to hydrogen combustion may include elevated temperature, elevated pressure, and the presence of steam. The flammability limits widen with increasing temperature. For example, at 100 °C the lower limit for downward propagation is approximately 8.8 % (see Fig. 2).

Values for hydrogen flammability in air saturated with water vapor at room temperature and pressure are given in Table 6. These limits may vary slightly during accident conditions. There may be scale effects due to the large size of reactor containments as well as variations in flammability due to the ignition source strength.

The triangular diagram of Shapiro and Moffette indicates regions of flammability of hydrogen:air:steam mixtures [14]. It has been widely reproduced and appears as Fig. 3.

Ignition of dry hydrogen: air mixtures, particularly when the mixtures are well within the flammability limits, can occur with a very small input of energy. Common sources of ignition are sparks from electrical equipment and from the discharge of small static electric charges. The minimum energy required from a spark for ignition of a quiescent hydrogen:air mixture is of the order of 10^{-2} (a very weak spark). The ignition energy required as a function of hydrogen concentration is shown in Fig. 4.

For a flammable mixture, the required ignition energy increases as the hydrogen concentration approaches the flammability limits. The addition of a diluent, such as steam, will increase the required ignition energy substantially. As mentioned previously, high energy ignition sources can cause mixtures outside the flammability limits to burn for some distance.

The presence of moist air in the Sarcophagus premises simultaneously raises the threshold magnitude of hydrogen concentration and the magnitude of necessary initiating energy.

The next initial data were used in the calculations.

It should be noted that, talking about “explosion”, it should be accentuated the difference between two phenomena of fast combustion — deflagration and detonation — conditions of occurrence and consequences of which differ one from another.

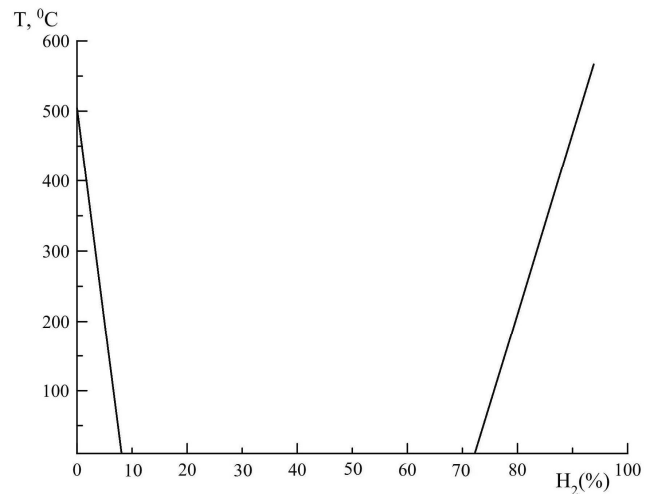


Fig. 2. Effect of initial temperature on downward propagating flammability limit in hydrogen

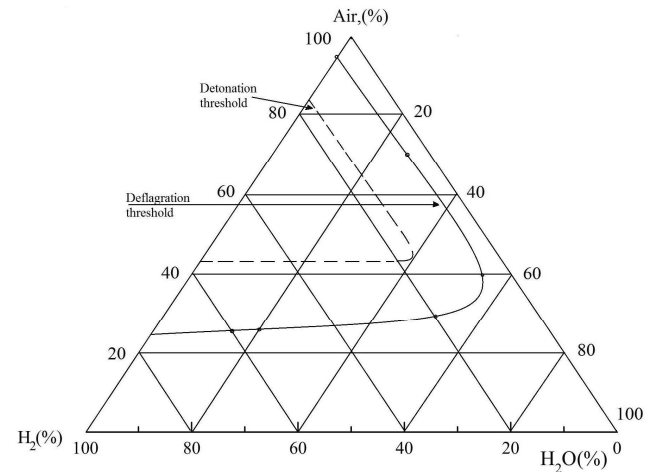


Fig. 3. Flammability and detonation limits of hydrogen, air, moisture

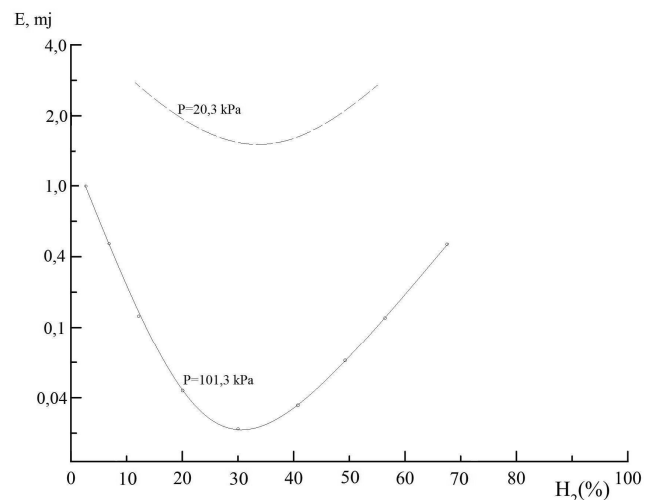


Fig. 4. Spark ignition energies for dry hydrogen air mixtures

The deflagration or fast burning down is the instantaneous combustion without forming of blast and consequently without explosion by which the flame front spreads in a gaseous mixture at a speed of several metres per second as a result of gas heating owing to heat-conductivity and diffusion of free radicals in a region of unhurt gas.

The deflagration can occur at comparatively low hydrogen content in dry air. Shapiro diagram shows the magnitude about 4 %. Necessary initiating energy is very small and makes the magnitude not exceeding one mill joule. In the absence of moisture the hot point about 500 °C can provoke self-ignition of hydrogen.

The detonation is combustion, taking place at a thin layers boundary, spreading with a constant supersonic speed along an unhurt gaseous mixture.

The detonation is a power blast, beyond the front of which the sharp chemical reaction of substance decomposition, attending by heat and gaseous products release, takes place.

By expansion of compressed detonation products explosion occurs.

Much higher concentrations of hydrogen are necessary for occurrence of detonation than for deflagration, in Shapiro diagram the explosive region corresponds to the presence in dry air 18—55 % of hydrogen (Fig. 3).

The experiments show that by very large volumes the explosive threshold is lowered.

Necessary initiating explosion energy makes up 5 kg at hydrogen concentration 30 % and reaches 250 kg at 20 % of H₂ concentration. The presence of moisture in premises raises the threshold magnitude of hydrogen concentration and initiating energy.

The upper limit of inflammability of hydrogen in a mixture with air: bottom 4.10 % upper — 74.0 % ignition temperature in air (530—590) °C, and combustion rate — 30 000 kg/(m²·sec).

The risk is a probabilistic measure, punitively expressing the effect of either human activity or natural phenomenon. At risk estimation the probability of negative effect with regard for probability of the realization of this effect. The risk is also the damage probability which is usually determined as a product of the probability of a certain process of given energetic class (in our case — the probability of the appearance of the an explosion (a fire) in Sarcophagus during a year $Q(Sr)$) and an expected damage [13]:

$$R(Sr) = Q(Sr) \cdot Y, \quad (29)$$

where Y — a prognosticated damage from an explosion (fire).

The risk level, the source of which is the artificial inhabitation environment, is in the limits $3 \cdot 10^{-6}$ — 10^{-3} , and level $5 \cdot 10^{-4}$ is the socially acceptable professional risk during prolonged action of unhealthy factors.

A situation, influencing on nuclear, radioactive and general technical object safety can appear during explosion (fire) in Sarcophagus.

Nuclear accident may be associated with FCM and nuclear fuel damage with following personnel overexposure violation of chain nuclear fission reaction control, critical mass formation, heat removal from FCM violation etc.

The real danger of displacement of fuel-containing masses and neutron-absorbing materials with following realization of self-sustaining chain reaction exists in the case of an explosion/a fire. For fresh fuel assemblies the critical mass is 315 kg, and for utilized low burn-up assemblies and with enrichment 2 % the critical mass is 200 kg, when penetrating of water into homogenous mix of FCM the critical mass is ~600 kg of uranium. The action of high temperatures at a fire can result in falling the main constructions of Sarcophagus, the strength of supports of which was not determined and permanently declines under the action of radiation and environment.

The danger of magmatic differentiation and forming of a phase essentially enriched by ²³⁹Pu (in reactor core before the accident it was produced more than 450 kg) may result in critical mass dozens kilograms.

Radioactive accident may result in release of radioactive products and ionizing radiation over stipulated object operation borders in quantities, exceeding the limits of its safe operation. At exposure dose at the industrial site of ChNP border making up 50 mSv/year which corresponds to dose limit of total external and internal exposure of personnel during a calendar year the general risk of the potential death will be expressed by magnitude $3 \cdot 10^{-7}$.

This magnitude is calculated in assumption that radioactive effect continues over the whole life. Let us section on three types explosions (fires) in Sarcophagus, resulting in breaking up of object's construction and release of radioactive substances into external environment in dependence of character and scales of radioactive consequences.

The first type — “local” explosion, when release of radioactivity occurs over stipulated borders of the object. Incidentally the quantity of released radioactive substances will exceed fixed values, but zone of external environment contamination is not out of the bounds of the industrial site of ChNP. The probability of the occurrence of an explosion (fire) in Sarcophagus at 1-st category doesn't exceed $R(Sr) \leq 10^{-3} \text{ year}^{-1}$.

The second type — “domestic” explosion (fire) is the incident when the release of radioactivity is possible over the industrial site bounds, but inside of controlled access area. The radioactive consequences are limited by controlled access area, where personnel exposure is possible in doses, exceeding allowed ones. The possibility of the occurrence of the 2-nd type accident is in the limits $10^{-5} \leq R(Sr) < 10^{-3} \text{ year}^{-1}$.

The third type is a large explosion (fire) when the radioactive contamination area goes out controlled access area limits, but is localized in the nearest limits at the level of total exposure, of population during a year more than 0.03 Sv. The probability of the occurrence of the 3-rd type accident is in the limits $10^{-7} \leq R(Sr) < 10^{-5} \text{ year}^{-1}$.

Radiation risk in the main is connected with the probable occurrence of potential dangerous initial events and development of their consequences. The quantitative evaluation of the radiation risk probability is determined by the risk function of individual sources of dangers, expressed as absolute mathematical anticipations of the radionuclide release into environment (Table 5), moreover the probability of an initial event was calculated by formulae (3—8, 10—15).

Table 5. The probabilities and the risk functions of individual sources of dangers, expressed as absolute mathematical anticipations of the radionuclide release

Initial event	Probability, year ⁻¹	Risk function, Ci/year
A fire inside Sarcophagus premises	$10^{-5}—10^{-3}$	$4 \cdot 10^{-4}$
An explosion inside Sarcophagus premises	$10^{-6}—10^{-4}$	$3 \cdot 10^{-5}$
Coming down of constructions by a fire	$10^{-6}—10^{-5}$	$6 \cdot 10^{-4}$
Coming down of constructions by an explosion	10^{-7}	$0.2 \cdot 10^{-5}$

Table 6. Danger ranking of some Sarcophagus premises

Height mark, m	Premises number	Danger factor			Danger category		
		A	B	C	I	II	III
-0.65—2.20	012/6			+			+
	012/7			+			+
	012/8		+	+		+	
	012/9			+			+
2.20—6.00	012/13			+			+
	012/14		+	+		+	
	012/15		+	+		+	
	012/16			+			+
6.00—8.00	210/5		+	+		+	
	210/6	+	+	+	+		
	210/7	+	+	+	+		
6.00—9.00	217/2		+	+		+	
6.00—9.00	303/3		+	+		+	
8.00—11.00	305/2	+	+	+	+		
9.20—12.50	304/3	+	+	+	+		
14.00—32.00	504/2	+	+	+	+		
18.50—35.50	504/3		+	+		+	
18.50—35.50	504/4	+	+	+			
35.50—62.00	914/2	+	+	+	+		

A — the influence of a fire/an explosion on the nuclear safety;

B — the influence of a fire/an explosion upon radiation safety;

C — the influence of a fire/an explosion upon generally technical safety.

Danger ranking of some Sarcophagus premises obtained by calculation, is adduced in the Table 6.

The main types of the danger in Sarcophagus are nuclear, radioactive and general technical ones. The radioactive danger is the threat of the exceeding determined occupational doze limits as a result of release of radionuclide's into environment. The potential radioactive danger is determined by total activity about 20 MCi containing in Sarcophagus premises.

The analysis of various components of danger factor in the total risk shows that the greatest danger is the coming down of constructions by a fire, and the main mechanism of the realization of negative consequence is forming of radioactive combustion products and following release of them over Sarcophagus boundary.

By its nature particles of soot and ashes in a smoke can be conditionally separated two fractions: rather big "fuel" particles with an average median diameter 30 μm and density 10.8 g/cm^3 ; fine-dispersed particles and aerosols with average median diameter 5 μm and density 6.2 g/cm^3 .

Conclusions

Sarcophagus can be regarded as a system, containing sources of the nuclear and radiation danger and having one protective barrier — external entombment (covering), having no internal fire-prevention obstacles, fire gaps and fire-blocking barriers. Radioactive water is in insulated premises where external genetic walls in direct contact with FCM and highly active dust are absent. The continuous process of radiolysis of water with yielding of radiolysis hydrogen and forming explosive detonating mix occurs.

Therefore of all the considered physical processes the most significant for the degradation of FCM is the emergence of the mechanical strain that appear at the FCM cooling from the high post-accident temperatures to the present ones due to the difference of the thermal expansion coefficients of the FCM components.

Preliminary calculative estimations on every premises of Sarcophagus, in which fuel dust, nuclear fuel or FCM are found, showed according formulae (21—24), that there is no potential explosive danger, e. i. interchange of air degree is more than 0.01. For securing of explosive safety in Sarcophagus premises, volume of which is 100 m^3 , containing 10 tons FCM, when hydrogen concentration is below 0.04 % (by volume) interchange degree of volume of gas phase only 10^{-3} hr^{-1} is enough. The probability of the critical state occurrence during an explosion/a fire in Sarcophagus because of radiation-chemical destruction of water, as a result of which the occurrence of radiolysis hydrogen can happen, can't be evaluated as a negligible magnitude.

The probability of the occurrence of such accidents in Sarcophagus is evaluated as average magnitude

10^{-4} year⁻¹ at the risk function. It results from the data of the Table 6 that the following premises are the most potentially dangerous from the stand-point of an explosion/a fire: the central hall, 505/4, 504/2, 305/2, 304/3, 210/6, 210/7 and in the given premises the regular control of hydrogen concentration should be carried out. It is also necessary to adjust the periodical control of hydrogen concentration in the rooms 012/8, 012/14, 012/15, 210/5, 217/2, 303/3 and 505/3. Thus, hydrogen forming rate as a result of radiolysis of water

under the action of radionuclide radiation will not exceed permissible values.

It should be ranked to measures on protection from explosive detonating mixture forming: prompt reveal of hydrogen forming seats and qualitative evaluation of explosive concentrations; gas-isolating and forced ventilation of bottom parts of premises situated in explosive zones; draining of radiolysis hydrogen on the path of its migration by special wells.

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