

THE MAIN PHYSICAL AND CHEMICAL PROPERTIES OF THE RADIOACTIVE PARTICLES EJECTED INTO THE ATMOSPHERE AT ACCIDENTS

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Abstract: *Physical and chemical properties of radioactive matter which get to the atmosphere at accidents on the examples of accidents at Chernobyl and Fukushima nuclear power plants has been analyzed. The properties determining sedimentation intensity of radioactive matter by an atmospheric precipitation has been analyzed. The chemical composition and superficial properties of the radioactive matter released into the atmosphere has been determined by the accident mode process. The radioactive matter (emissions) has been shown to mainly consist of gaseous inert ^{133}Xe , chemically inert "hot particles" in the form of a $\text{U}_x\text{O}_y + \text{ZrU}_x\text{O}_y$ alloy, microparticles (organic and nonorganic) of dust with vapors of I_2 , TeO_x , CsO_2 radioactive matter condensed on their surface, and microparticles of radioactive graphite.*

Keywords: *Fukushima, Chernobyl, Nuclear plant accident, Radioactive particles, Radioactivity-induced charge, Radioecology, Raindrop, Coagulation, Intensity of excretion.*

INTRODUCTION

Emission in the atmosphere of radioactive matter at large accidents at the objects of nuclear power is possible. Chernobyl Nuclear Power Plant accident (Chernobyl accident) in Ukraine and Fukushima Dai-ichi Nuclear Power Plant accident (Fukushima accident) in Japan are the most large accidents for all history. These accidents have been considered as an example. A large amount gaseous and aerosol radioactive matter falls into the atmosphere as a result of emergency depressurization of a nuclear reactor [1, 2]. Under the influence of convective airflows radioactive matter falls into the top layers of the troposphere and propagation at the long distances. Radius of a zone of radioactive contamination as a result of Chernobyl accident makes about 1500 km [3, 4]. Contamination zone at accident in Fukushima makes 150 km [5]. However the raised background of radiation has been registered at much bigger distance, for example in Norway [2, 6]. The sizes and form of zone contamination have been defined by weather conditions, a district landscape, existence of vegetation, density of buildings and some other factors. Much attention in [7 – 9] has been given modeling of dynamics change of a contamination zone. Atmospheric precipitation over an accident zone significantly reduces the sizes of contamination area and the radiation background. It makes owing to sorption by water drops of radionuclides. Process intensity of radionuclides sorption and purification intensity of the atmosphere from radionuclides depend to aggregate state of radionuclide, its dispersion, and also physical and chemical properties radioactive [10 – 12]. Besides, in articles [13 – 15] it has been determined that radioactive particles can accumulate

charge. Existence at a disperse particle of charge can significantly intensify radionuclide sorption process by rain-drops [10, 16, 17]. Gaseous and radionuclides dispersion influence to process of growth of rain-drops and dynamics of development of a cloud and precipitation intensity. Thus, research of mechanism of influence of precipitation to purification intensity of the atmosphere from radionuclides is an actual task.

PROPAGATION ZONE

A large amount of radioactive gases and solid particles (aerosols) at depressurization of an active zone has been released into the atmosphere. Radioactive matter which has been expelled out rise airflows in the top layers of the troposphere and distribute at the long distances.

The sizes of a contamination zone depend from:

- mode of accident (explosion power, amount of the expelled radioactive matter, accident duration);
- physical and chemical properties of radioactive matter (type of radionuclide, mass, density, water repellent of particles);
- weather conditions (wind velocity and bearing of an apparent wind, humidity, existence of precipitation).

Apparently a large amount of the defining factors has been caused by complex form of a contamination zone. As a result of Chernobyl nuclear accident the zone of radioactive contamination has been made more than 142 000 sq.km (Table 1). Contamination zones only for an isotope ^{137}Cs have been represented on Fig. 1.

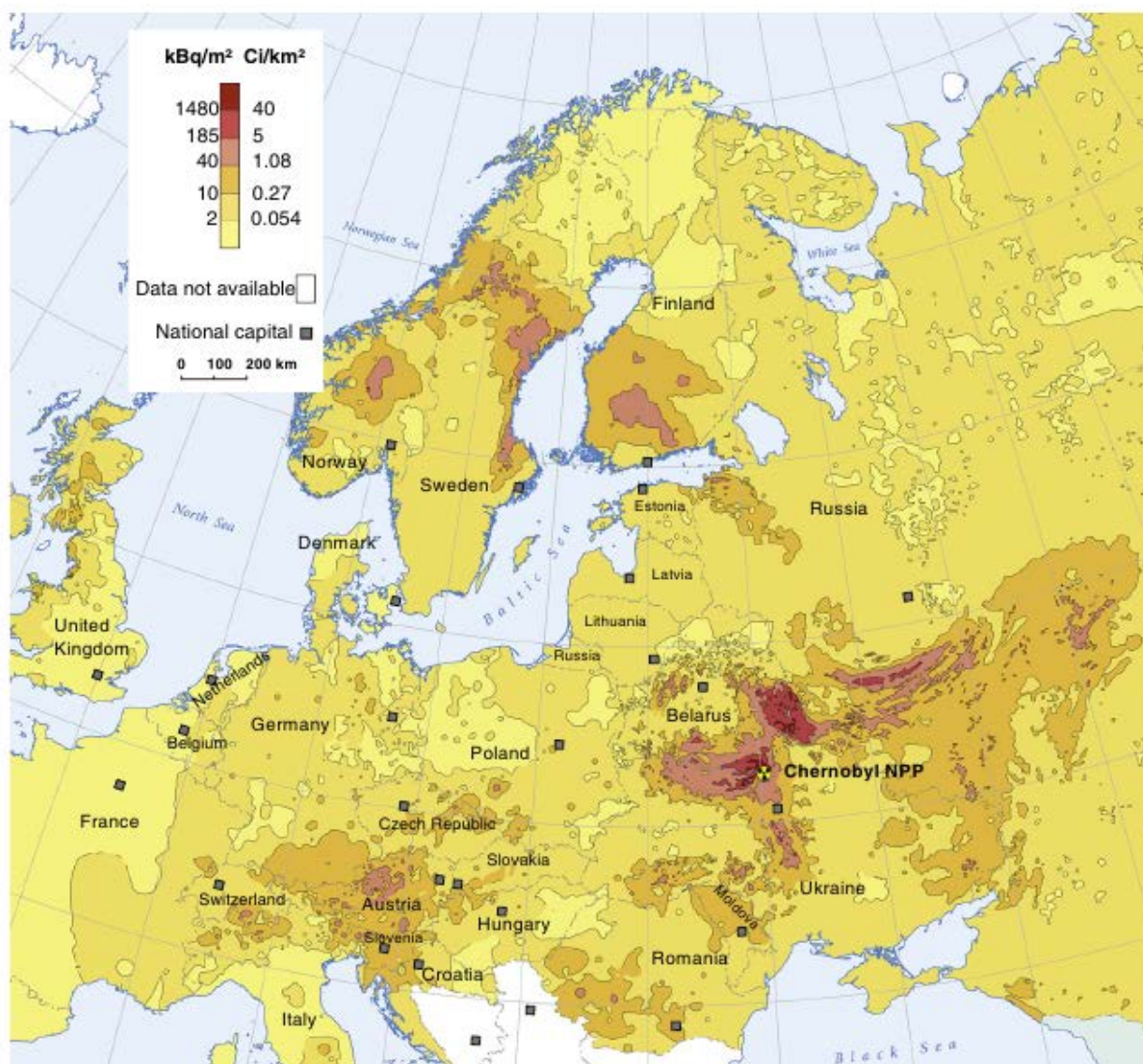


Fig. 1. Surface contamination with ^{137}Cs in Europe after the Chernobyl nuclear accident [18].

Table 1. Contaminated areas in European countries [2, 19].

Country	Areas of the contamination zones (km ²)			
	Zone 4 (37-185 kBq/m ²)	Zone 3 (185-555 kBq/m ²)	Zone 2 (555-1480 kBq/m ²)	Zone 1 (> 1480 kBq/m ²)
Russian Federation	49,800	5700	2100	300
Ukraine	37,200	3200	900	600
Belarus	29,900	10,200	4200	2200
Sweden	12,000			
Finland	11,500			
Austria	8600			
Norway	5200			
Bulgaria	4800			
Switzerland	1300			
Greece	1200			
Slovenia	300			
Italy	300			
Moldova	80			

As a result of Fukushima accident the zone of radioactive contamination has been made much less (~ 7 000 sq.km) (Fig. 2). However, for the small state of Japan ecological aftermaths of accident were catastrophic. However even these data represents the large sizes of a contamination zone and respectively great negative influence to ecosystem of the region and planet in whole. Therefore reduction of the zone's size of contamination matter in close proximity to accident zone is very actual task.

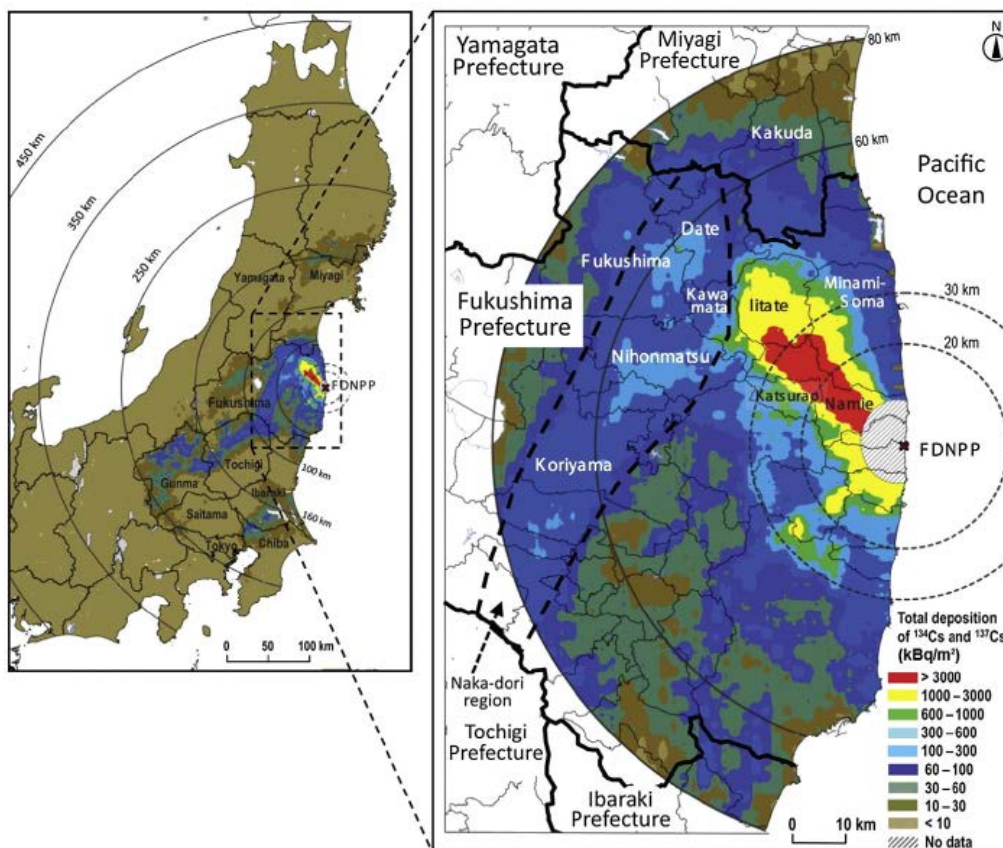


Fig 2. Estimated total deposition of radiocesium after the Fukushima nuclear accident, approximately half of which is ^{137}Cs [20].

MAIN PROPERTIES OF PRODUCTS OF RADIOACTIVE CONTAMINATION

The comparative analysis of the radionuclides which got to environment as a result of Chernobyl accident and Fukushima accident has been carried out in article [21] (Table 2):

Table 2. Main properties of fission products released into the atmosphere as a result Chernobyl and Fukushima accidents.

Radionuclide	$T_{1/2}$	Activity (PBq)			
		Chernobyl accident	Reference	Fukushima accidents	Reference
Noble gases					
^{85}Kr	10.75 y	33	[22]	44	[29]
^{133}Xe	5.25 d	6500	[22]	14,000 15,300	[30] [8]

Volatile elements					
³ H	12.3 y	1.4 (inventory)	[23]		
^{129m} Te	33.6 d	240	[22]	~15	[21]
¹³² Te	3.20 d	~1150 1000	[24] [22]	~180 88	[21] [32]
¹²⁹ I	15.7E6 y	$(4 - 4.8) \times 10^{-5}$ 8.4×10^{-6}	[25, 26] [28]	5.5×10^{-5} 6.6×10^{-6}	[31] [21]
¹³¹ I	8.03 d	~1760 1200–1700	[24] [22]	150 130–160 190–380 65.2 200	[33] [34] [35] [36] [37]
¹³³ I	20.8 h	910 2500	[24] [22]	146	[33]
¹³⁴ Cs	2.07 y	~47	[22, 24]	11.8 18	[21] [34]
¹³⁶ Cs	13.0 d	36	[22]	2.6	[21]
¹³⁷ Cs	30.1 d	85 74–85 98	[24] [22] [27]	12 13 6.1–15	[33, 35] [37] [34]
Elements with intermediate volatility					
⁸⁹ Sr	50.5 d	~115 81	[24] [22]	~0.2	[21]
⁹⁰ Sr	28.9 y	~10 4 8	[24] [26] [22]	~0.02	[21]
¹⁰³ Ru	39.2 d	> 168 170	[24] [22]		
¹⁰⁶ Ru	372 d	> 73 30	[24] [22]		
¹⁴⁰ Ba	12.8 d	240 170	[24] [22]		
Refractory elements					
⁹⁵ Zr	64.0 d	84 87 170	[24] [26] [22]		
⁹⁹ Mo	66.0 h	> 72 210	[24] [22]		
¹²⁵ Sb	2.76 y	0.23	[26]		
¹⁴¹ Ce	32.5 d	84 200	[24] [22]		
¹⁴⁴ Ce	285 d	~50 140	[24] [22]		
¹⁵⁴ Eu	8.60 y	0.13	[26]		
²³⁹ Np	2.36 d	400 1700	[24] [22]		
²³⁸ Pu	87.7 y	0.015 0.03	[24] [22]	$(2-5) \times 10^{-6}$	[21]
²³⁹ Pu	24,100 y	0.013	[24, 26]		

²⁴⁰ Pu	6560 y	0.018	[24, 26]		
²³⁹⁺²⁴⁰ Pu		0.031	[24]	(1.0-2.1)×10 ⁻⁶	[38]
²⁴¹ Pu	14.3y	~2.6	[24]	(1.0-2.1)×10 ⁻⁴	[38]
²⁴² Pu	3.76E5 y	4 × 10 ⁻⁵	[24]		
²⁴¹ Am	433 y	0.0024	[26]		
²⁴² Cm	163 d	~0.4	[24]		
²⁴⁴ Cm	18.1 y	0.0027	[2]		
Total(excludingnoble gases)		~5300	[2]	~520 (340-800)	[21]

Apparently from Table 2 the chemical and quantitative composition of the radionuclides expelled into the atmosphere at Chernobyl and Fukushima accidents has been differed. First of all it has been connected with failure behavior and nuclear fuel composition. So, Reactor of Chernobyl Nuclear Power Plant the of Big Power Channel (RBPC) in which as nuclear fuel the uranium dioxide ²³⁵U low-enriched with UO₂ has been used. As a neutron moderator in RBPC graphite (¹⁴C) has been used. Metal constructional parts in an active zone has been made from zirconium and niobium alloy. Boiling Water Reactor (BWR) has been applied At the Fukushima Dai-ichi Nuclear Power Plant. In such reactors water is a neutron moderator. Therefore at accident there is no large emission of graphite. However a large amount of radioactive water vapor has been expelled into the atmosphere at depressurization of the reactor. A large amount of refractory elements in emissions of Chernobyl accident and much smaller at the Fukushima accident has been connected with that at Chernobyl accident there was a destruction of an active zone of the reactor with emission of radioactive fuel. Besides Chernobyl accident is characterized by a high-temperature stage of accident that results to formation of additional amount of cesium. At the Fukushima accident of emission of parts of radioactive fuel and radioactive graphite didn't occur and the fritting fuel elements accumulated in a trap. The data on the radionuclides expelled into the atmosphere have been represented in the table. These data will be actual only in the first some hours after accident as the most part ¹³³I within the first days breaks up on reaction:



Amount ¹³³Xe in the first days after accident was increased significant as emission ¹³³I in Chernobyl accident was meaning.

Proceeding from it and data of Table 2 emissions of xenon are essential and at the expense of a gaseous state can extend at the long distances. Xenon as inert gas in the lower troposphere is in a free molecular state. Separate molecules don't aggregate in clusters. Therefore the behavior of a molecule ¹³³Xe in the atmosphere as isolated has been considered. Process of radioactive decay is probabilistic therefore the separate molecule of radioactive matter can be considered in two states. The first state – a one-nuclear molecule before radioactive decay that is with standard physical and

chemical properties of xenon. The second – after radioactive decay that is formation of stable cesium (Table 4) with physical and chemical properties of an daughter nuclide.

Table 3. Main nuclear physical properties ^{133}Xe [39]

Nuclide	Half-life	Decay	Daughter nuclide	Deposited energy
^{133}Xe	5.248d	β^- (100%)	^{133}Cs (stable)	425.845 keV

The analysis of kinetics of absorption of radioactive xenon determined that it is possible to use physical and chemical properties of inert xenon without taking of its radioactive decay. However the most part of radionuclides is in a solid state in the form of aerosol microparticles. Therefore, researches of atmosphere purification dynamics of radioactive matter it is expedient to consider properties of the solid microparticles of an aerosol containing radionuclides.

In a zone of radioactive contamination from the Chernobyl accident there is a large number of fine particles of the fritted radioactive fuel. These particles have been created as a result of the first high-temperature explosion and the subsequent stage of reactor's elements burning. These particles have been denominated "hot particles". Research of structure of fuel particles has been described in [40 – 50]. Hot particles represent impregnations of the particles fuel element UO_2 in a cover of the uranium's higher oxides and zirconium – U_xO_y , ZrU_xO_y (Fig. 3, 4). Existence of a highly stable cover of ZrU_xO_y leads to increase in chemical persistence of "hot particles" in the atmosphere, the soil and basins. Therefore "hot particles" will be in a stationary condition from the moment of their formation after nearly 30 years after accident.

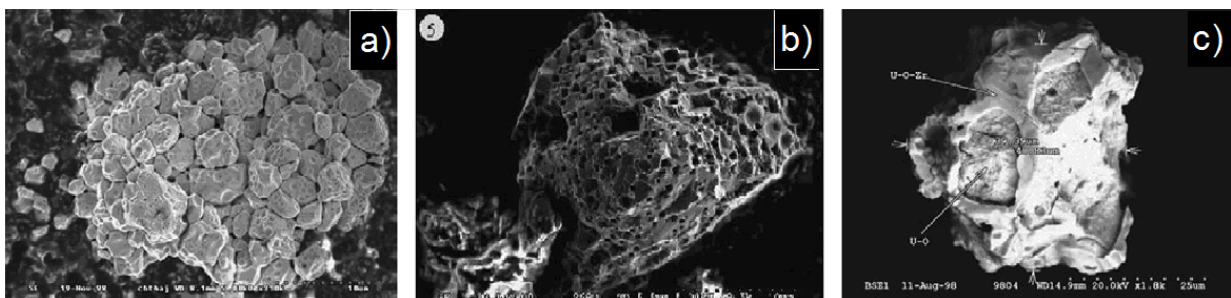


Fig. 3. SEM images of fuel particles: (a) UO_2 fuel particles; (b) UO_{2+x} fuel particles; (c) ZrU_yO_x fuel particles [40]

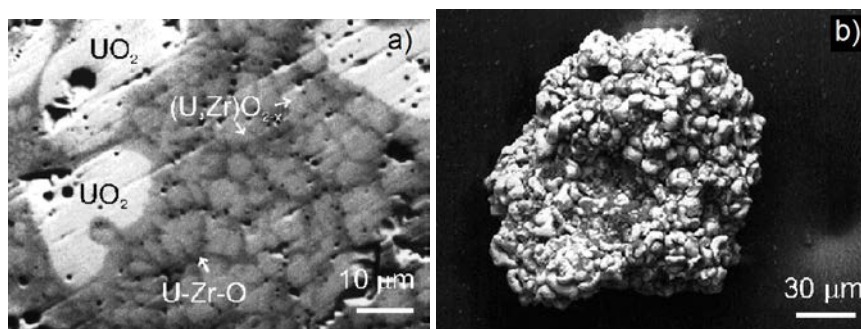
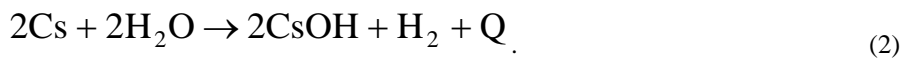


Fig. 4. SEM backscattered images of Zr-U-containing hot particles and inclusions in Chernobyl fuel particles (a) razrez toplyvnoy particles ; (b) appearance of a fuel particle [50]

Such radionuclides as ^{95}Zr , ^{95}Nb , ^{99}Mo , $^{141,144}\text{Ce}$, $^{154,155}\text{Eu}$, $^{137,139}\text{Np}$, $^{238-242}\text{Pu}$, $^{241,243}\text{Am}$, $^{242,244}\text{Cm}$ have been expelled from the emergency block only consisting of fuel particles. Also more than 90% of leakage of activity $^{89,90}\text{Sr}$ and $^{103,106}\text{Ru}$ are the share of fuel particles [41, 42]. The disperse composition of fuel particles in initial radioactive precipitation at a distance of 2 – 60 km from the CNPP has been described by the logarithmically normal law of distribution with a median radius of fuel particles about 3 microns [44]. On the basis of multitudinous results of researches of properties of "hot particles" the "Hot particles" the database [51] has been created. Besides as a result of the fire at the Chernobyl accident and high temperature at the Fukushima accident vapors of fission products (radioisotopes of iodine, tellurium, cesium and in much smaller extent of strontium and ruthenium) which in the atmosphere settle subsequently on various surfaces (water microparticles, organic have been expelled into the atmosphere and inorganic raise dust, a smoke) [26, 52]. Iodine evaporates in the form of the molecules I_2 and weakly reacts with atmosphere oxygen. Such metals as tellurium and caesium are oxidized more actively and are expelled into the atmosphere in the form of oxides – TeO_x , CsO_2 . Also at contact with vapors of atmospheric water C_5OH on reaction can be formed:



Emission of a large amount of radioactive graphite was characteristic of the Chernobyl accident. At operation of a nuclear reactor as a result of neutrons reactions with impurity of isotope ^{13}C which is present at graphite in number of 1,1% in the last radionuclide ^{14}C accumulates:



This radionuclide belongs to low-radio toxic and its admissible concentration in air makes $5 \text{ Bq}\cdot\text{m}^{-3}$. Basic nuclear properties fyzicheskye ^{14}C in Table 4:

Table 4. Main nuclear physical properties ^{14}C [39]

Nuclide	Half-life	Decay	Daughter nuclide	Specific activity
^{14}C	5730y	β^- (100%)	^{14}N (stable)	$1.65 \cdot 10^{14} \text{ Bq}\cdot\text{kg}^{-1}$

Extent of accumulation ^{14}C in graphite depends on time of operation of the reactor. For graphite of the 4th power unit Chernobyl Nuclear Power Plant specific activity of graphite on radionuclide ^{14}C was equal to 10^7 - $10^8 \text{ Bq}\cdot\text{kg}^{-1}$. At the Chernobyl accident in environment more than 800 tons of radioactive graphite has been expelled. It is big threat of ecological safety of the region. At the same time the part of graphite in the form of a fine aerosol has got to the atmosphere and has been extended by airflows to the large territory. Graphite particle as high-porous substance in the

structure may contain both microparticles of radioactive fuel and practically whole range of radionuclides which has been represented in table 2. Therefore radioactive graphite can be presented as a graphite carcass with impregnations of radioactive materials of their oxides and oxidic complexes. Thus, when modeling process of sedimentation of radioactive matter as object of sedimentation by an atmospheric precipitation we will consider:

- 1) gaseous inert gas xenon;
- 2) chemically inert "hot particles";
- 3) microparticles of organic and inorganic dust with vapors of radioactive matter condensed on a surface;
- 4) the liquid microdrops and liquid clusters containing the condensed radioactive matter;
- 5) microparticles of radioactive graphite.

The analysis of solubility and hydrophobic properties of radioactive particles has allowed prognosticating behavior of a particle after trapping by a rain-drop (Fig. 5).

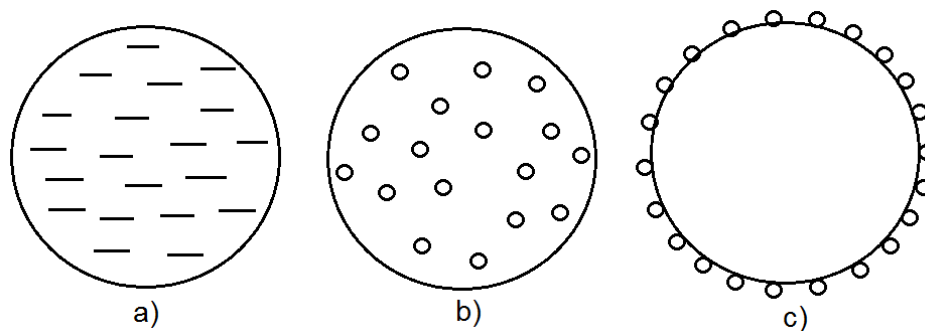


Fig. 5. Absorption rain-drop radioactive particles with various superficial properties: a) the dissolved radioactive matter; b) hydrophilic insoluble radioactive matter; c) hydrophobic insoluble radioactive matter.

Soluble particles are dissolved evenly on all volume of a drop changing at the same time its physical and chemical properties (**a**). Process of absorption of soluble particles is much more intensive, than process of absorption insoluble other conditions being equal. The dissociation of crystals of salts occurs rather quickly and removal of soluble substance from a drop surface in its volume has been defined by volume diffusion of the hydrated ions. It is the result of the small sizes the diffuse of particles that there is much higher than the velocity of diffusion of large insoluble radioactive particles. As it was noted earlier the liquid micro-drops and liquid clusters containing the condensed radioactive matter belong to soluble radioactive particles. Water is present at the atmosphere as a liquid drop carrier. Among solid soluble radioactive particles the main part was made by salts NaCl, MgCl₂, CaSO₄, Na₂SO₄, K₂SO₄ and others which have got to the atmosphere at evaporation of sea water and a soil erosion. An insoluble hydrophilic particle penetrates into drop volume with a velocity corresponding to their degree of hydrophilily (**b**).

Volume diffusion of such particles has been determined by coefficient of a superficial tension on border water / an insoluble radioactive particle. Owing to lower speed of penetration of the radioactive particles adsorbed on a surface of a drop trapping intensity of such particles will be less, than soluble particles. The main part of the solid radioactive particles extending to long distances is made by SiO₂ microparticles with vapors of radioactive matter condensed on a surface (¹³⁷Cs). Organic dust, pollen and microparticles of vegetation have been carried to insoluble hydrophilic particles. Ashes particles which contain burning products at the fires possess hydrophilic properties. Particles of ashes have been offered as complexes from oxides of metals with spherical porous structure. [53, 54]. The dominating components of ashes complexes are CaO, SiO₂, Fe₂O₃, Al₂O₃. The main volume of radioactive particles in a near zone to epicenter of emission has been made of insoluble hydrophobic particles. Graphite and chemically inert "hot particles" have been carried to these particles. Insoluble hydrophobic particles have been accumulated up for drop surfaces (*c*). At the expense of a high superficial tension they don't penetrate into drops. In a limit occurrence when all surface of a drop becomes covered by solid particles that such drops on superficial properties can be considered as solid radioactive particles. Accordingly over time the free area of a rain-drop will decrease it reducing trapping intensity of radioactive particles. Thus, the greatest trapping intensity a drop of radioactive particles will be at soluble particles in water under other identical conditions. Insoluble hydrophobic particles will have the smallest trapping intensity.

CONCLUSIONS

The chemical composition and physical and chemical properties of radioactive particles which have been expulsed at accident depends on nature of course of accident (temperature, pressure in a destruction zone of the reactor). The radioactive matter expulsing to the atmosphere at emergence of nuclear plant accidents on nature of interaction with rain-drops have been divided into 5 classes: inert gas xenon; solid aerosol fuel particles; graphite particles; particles of organic and inorganic dust and liquid aerosols. On their surface flying radioactive matter I₂, TeO_x, CsO₂ I₂, TeO_x, CsO₂ are condensed. Most quickly at precipitation soluble radioactive particles will be precipitated. For insoluble radioactive particles rain scavenging velocity precipitation falls with growth of their hydrophobic behavior.

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