Experimental geoecology

Galstyan A.V., Gevorkyan R.G., Sarkisyan S.E. Sulfuric gases neutralization of non-ferrous metallurgy by natural zeolites of Armenia

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Abstract. Neutralization of sulfuric gases of non-ferrous metallurgy is a difficult task due to the large volume, high temperature, low concentration of sulfur dioxide in the flue gas emissions into the atmosphere. The utilization of sulfur dioxide should be organized with the use of solid adsorbents, one of which is a natural zeolite. According to its adsorption properties Armenian zeolites are an indispensable raw material for extraction of flue gas desulfurization in non-ferrous metallurgy.

Key words: zeolite, adsorbent, metallurgy, sulfuric gases, temperature, utilization.

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In Armenia, non-ferrous metals occupy an important place in the industrial production and have a huge tendency of growth (compared to 1990 increased from 8 to 31% [2nd National Messages, 2010]). At the same time, this industry is one of the major pollutants of air basin by sulfur dioxide. The neutralization of sulfur dioxide in non-ferrous metallurgy is a difficult task, due to the large volume, high temperature and low concentration of sulfur dioxide in the flue gas emissions into the atmosphere. The utilization of sulphurous anhydride in the latest would solve several problems at once: to protect the environment and help to avoid irretrievable loss of valuable raw materials, that can be used in the future for the production of sulfuric acid and other useful sulfur compounds.

Currently, there are two directions in the utilization of sulfur dioxide: wet process related to SO_2 absorption by various solutions and processes of dry or adsorption purification, based on availability of cheap sorbents that have significant SO_2 [Anurov S.A., Torocheshnikov N.S., Smola V.I., 1999] activity.

The utilization of sulphurous anhydride should be organized with the use of natural sorbents, one of which are zeolites. At present there are more than 40 types of zeolite minerals, of which only six are of industrial importance (clinoptilolite, mordenite, erionite, chabazite, ferberit, philipsite), and among these, the most significant role is of clinoptilolite [Chelishchev N.F., Berenshtejn B.G., Volodin V.F., 1987]. Zeolites are water aluminosilicates, with infinite openwork frame structure, formed by the junction of a common vertices of SiO_4 and AlO_4 tetrahedra, where are interconnected cavities occupied by large ions and water molecules. The term "molecular sieve" is applied, as dehydrated zeolite crystals, due to the system of channels and cavities penetrating crystals, have well developed internal surface, accessible to the adsorbed molecules [Alietti A., 1972].

In Armenia there are known major deposits of zeolite rocks: Noyemberian, Alaverdi, Shirak, Ijevan, Tavush region, Vayk, Kapan. Of these, the most studied is Noyemberyan's, containing its composition 65–80% of clinoptilolite.

The adsorption properties of natural zeolites by SO_2 are caused by: clinoptilolite content in the rock, the silicium oxide, the temperature of adsorption and the type of zeolite modification. At the same time the adsorption capacity for SO_2 is directly dependent on the clinoptilolite content in the breed [Tsitsishvili G.V., Andronikashvili T.G., KirovG.N., Filizova L.D., 1985]

Table 1. Adsorption capacity for SO_2 due to the clinoptilolite content

Deposite	Clinoptilolite Content in the Breed, %	Adsorption Capacity for SO_2, g/100g
Sokirnitsina	75	6,3
Noyemberyan	65	4.1
Akhalcikh	50	2.6
Ay-Dag	75	6.2
Hot Spring	80	6.0
Beretian	40	2.4

The very important factor to arrange SO_2 recovery is in the formation of kremen content as lawflint zeolites are characterized by a high adsorption capacity and low acid resistance, while the High-silicons – have a low adsorbability to SO_2 and high acid resistance.

Thus lawflint zeolits can be used for the extraction of the SO_2 from pre-dehydrated systems, high-silikons – for catching SO_2 from wet environments, since the action to them of acids even slightly improves their absorption capacity [Tamboli J.K., Sand L.B., 1970]. Acid stability is a very important factor in the organization of SO_2 recycling, as in the regeneration process, due to high acidity the SO_2 zeolitic structures collapse and, therefore, can not be used for multicycle processes.

Table 2. Comparative chemical composition of major oxides (SiO_2, Al_2O_3) containing clinoptilolite tuff from various fields in the world

Oxide %	Noyemberyan field, Armenia					"Hektor" California, USA	Hokaido, Japan	"Dzegvi", Georgia	"Aydag", Azerbayjan
	"New", hole № l8	"Nor Kogxb"	"Central"	"South" hole №7	"East West"				
SiO_2	68.87	68.75	68.23	69.31	66.80	66.82	65.27	61.30	65.90
Al_2O_3	12.20	11.92	12.48	12.09	10.58	12.24	14.84	13.00	12.90
SiO_2/Al_2O_3	9.6	9.8	9.3	9.8	10.7	9.3	7.5	8.0	8.7

Table 2 shows the content of the basic oxides (SiO_2, Al_2O_3) in klinoptilolic tuffs of different regions, from which it follows that the Armenian zeolites are the richest with silicon and this is why they are high resistance of acids.

The sorption of SO_2 on clinoptilolite at low SO_2 concentrations and with temperature up to 200^0C remains high. This allows you to recommend the clinoptilolite as an efficient absorber of SO_2 at low concentrations of SO_2 and of high temperatures, which is important for the production [Liang P.M., 1989]. The adsorption capacity for SO_2 largely depends on the ion exchange shape of the modified zeolite, thus the exchange of Na^+ to Ca^{2+} and Mg^{2+} ions in the zeolite structure significantly increases the adsorption capacity. Obviously in exchange of ions two sodium cations are replaced by a cation of calcium, and the input window expands, and this facilitates the penetration of SO_2 molecules in the cavity of the zeolite structure [Barrer R.M., 1956]

Widespread use of zeolites in the industrial scale is due to the possibility of complete and SO_2 desorption at 350–400^0C. Desorption of SO_2 and appropriate regeneration of the "spent" zeolite is one of the important steps that basically defined the technical and economic value of the adsorption process.

In order to obtain by desorption concentrated gas various variants of zeolites' regeneration were investigated:

1) the use of hot air as heat carrier and stripping gas leads to the dilution of desorbed gas;

2) uniformly intense heating of the sorbent layer (5, 10, 20 deg/min) with simultaneous stripping, SO_2 evolvment forms gas with a maximum concentration of about 24%;

3) the desorption by gas, circulating in a special closed circuit with 300–35^0C allowes to extract SO_2 with the concentration of about 30%;

4) purging of the spent layer by the superheated steam (220–260^0C) the SO_2 may be allocated in a concentrated form after the condensation:

Three last ways of the regeneration of natural zeolites can allow to utililize SO_2 by next using it in the sulfuric acid production, or in the production of liquid sulfur dioxide [Tsitsishvili G.V., Andronikashvili T.G., Kirov G.N., Filizova L.D., 1985].

When using zeolites in many cycles its activity decreases only for the first 5–10 cycles and only for 10–12%, but in subsequent cycles this capability remains unchanged. The main conditions for the use of zeolites for the neutralization of non-ferrous metallurgy's waste gases are:

- the ability to light thermo-chemical modification of the zeolite, which is very important for the selective adsorption;

- the adsorptive properties of zeolites depend little on up to 200^0C flue gas temperature and therefore a wide temperature range without changing the adsorption capacity applies;

Some properties of Armenian zeolites better promote the adsorption of SO_2:

- high content of clinoptilolite in raw (65–80%), due to which the adsorption of SO_2 is in a direct linear relationship;

- high content of silicium oxide in the mineral, on which the use of zeolite multicycle is directly dependent on;

- zeolite rocks of Noemberian deposits of Armenia is readily available in the extraction since come to the surface, and the prospective reserves are of 400–500 million tons.

Therefore, the Armenian zeolites are an indispensable raw material for the organization of recycling waste sulfuric gases of ferrous metallurgy.

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Gevorkyan R.G., Harutyunyan R.S., Sargsyan H.O., Sargsyan O.H., Badalyan G.G., Gevorkyan M.R. Extraction of radionuclides and heavy metals from industrial water using natural and modified zeolites of Armenia

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Abstract. In Armenia are known many wide-spread deposits of zeolite type rocks of volcanoes-sedimentary origin. The main zeolitomaker and bentonitomaker minerals are clinoptilolite (heulandite), mordenite, stilbite, phillipsite, chabazite and bentonite-montmorrilonite. Adsorption properties of zeolites are studied but the highest selectivity for the radionuclides and heavy metals showed the clinoptilolite.

Key words: zeolite, radionuclide, metals, clinoptilolite, sorbent.

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Many wide-spread deposits of zeolite type rocks (also referred to aszeolites) of volcanoes-sedimentary origin (type of Noyemberyan zeolite) are known in following regions of Armenia: Tavush (Noyemberyan–Nor Kokhb, Paravaqar, Idjevan–Kuybishev, Dilidjan), Ashotsk (Sarchapet), Shirak (Keti, Kaps, Krashen, Krasar and e.t.), Lori (Alaverdi), Kotayk (Garni–Azat), Vayq (Martiros),

Syuniq (Kapan–Shikakhokh) [Petrosov I.Kh., Djrbashyan R.T., Mnatsakanyan A.M., 1999].

The main zeolitomaker and bentonitomaker minerals of these groups are clinoptilolite (heulandite), mordenite, stilbite, phillipsite, chabazite and bentonite-montmorrilonite.

Cation-Exchanging Properties. The adsorption of cations on natural clinoptililite and the ion-exchanging selectivity of clinoptilolite to Cs^{137}, Cs^{134}, Sr^{90}, Co^{60}, Mn^{54} and other radioactive isotopes in model solutions are studied. The properties of none treatment, thermal treatment and electron radiated samples are studied. The adsorption properties of samples of clinoptilolite which are subjected to ion-exchanging by different cations of metals and treatment with acids, radiated with the electron beam and γ-radiation are reported.

Receiving the monocationic forms of zeolites is of interest for fundamental and applied investigations. For this purpose, the initial clinoptilolite was subjected for long time and multiple ion-exchanging (8–15 times) in concentric solutions of chlorides of inputting cations at 80–90^{0}C during 110–120 hours. In dependence on type of cation the rate of ion-exchanging is changed; the rate of inputting of "guess" cation is changed and the constancy of balance of exchange of initial cation by "guess" (contre-cation) is changed. From the above, the problem of formation of maximum monocationic forms of clinoptilolite from Noyemberyan deposit and investigation of ion-exchanging properties of these samples is solved.

The highest rate of adsorption has the samples that are subjected to chemical treatment with NaOCl. But Ba^+ form of clinoptilolite shows the lowest cation-exchanging capacity (CEC) and slow saturation as father as cations of Ba^+ is formed more strong bonds with carcass of structure and difficultly are subjected to changing. The samples, the treatment of which by acids has low values of CEC due to dealumination of clinoptilolite and destruction of carcass of structure which reduce the adsorption properties [Gevorkyan R.G., Sargsyan H.H., Karamyan G.G., Keheyan Y.M., Yeritsyan H.N., Hovhannesyan A.S., Sahakyan A.A., 2002].

The Extraction of Radinuclides. The methods of deactivation of radioactive wastes and wastewaters of atomic industry and nuclear power engineering were successfully used in 1986–1992 in the period of accident at Chernobyl nuclear power by staff of GEOCH AS USSR, where, particullary, zeolites from Noyemberyan deposits of Armenia were also used. Development of nuclear power engineering highlights the searching of effective and cheap ways for purification of radioactive wastewaters and waste disposal. It becomes particularly actual in Armenia after the restart of nuclear power station. In the present the effective technology of application of thermo-chemically and radiation-modified samples of clinoptilolite of Armenia is developed by the staff of YSU as ion-exchanging sorbent for treatment of Cs^{137}, Cs^{134}, Sr^{90}, Co^{60}, Mn^{54}-containing radioactive solutions. The results of experiments allow us to offer zeolites for further application for the purpose of disposal of liquid radioactive wastes.

Based on experimental data it is developed experimentally-pilot technique, from 4 columns, for purification of liquid radioactive wastes on Armenian atomic station. Using big series of modified and granulated zeolites, the investigations of the process of purification of liquid radioactive wastes on territory of Armenian nuclear power plant are carried out.

After some cycles of purification decreasing of radioactivity of Cs^{137} and Cs^{134} isotopes up to 2500 times compared with initial is pointed. It makes possible to exclude liquid radioactive wastes, decrease the volume of solid radioactive wastes (up to 25 times) and decrease ecological risks. Besides this, for the first time the microwave effect is used on the ion-exchange process and the positive interesting results obtained [Gevorkyan R.G., Sargsyan H.H., Karamyan G.G., Keheyan Y.M., Yeritsyan H.N., Hovhannesyan A.S., Sahakyan A.A., 2002; Gevorkyan R.G., Mazmanyan L.G., Sargsyan A.H., 2007; Harutyunyan R.S., Gevorkyan R.G., Badalyan G.G., Sargsyan A.H., Akhalbedashvili L.G., 2012].

The ion-exchanging properties of natural zeolites are also used as ion-exchangers for protection of environment.

Clinoptilolite and its variety-heulandite are zeolites which have low force of field and for them the activity of cations is located by the following: $Rb^+ > NH_4^+ > K^+ > Na^+ > Li^+ > H^+$ и $Ba_2^{+2} > Ca^{2+} > Mg^{2+}$. The experiments show that $Pb^{2+} \approx Ba^{2+} >> Cu^{2+}$, Zn^{2+}, Cd^{2+}.

By using Na-clinoptilolite as the etalon the series of activity is obtained:

$NH_4^+ > Pb^{2+} > Na^+ > Cd^{2+} > Cu^{2+} \cong Zn^{2+}$, $Pb^{2+} > NH_4^+ > Cu^{2+} \cong Cd^{2+} > Zn^{2+} \cong Co^{2+} > N_1^{2+} > Hg^{2+}$.

The ecological danger of anthropogenic geochemical anomalies and barriers is determined primarily by the fact, that they are supplier of great quantity of tocsin gases and metals which are involved in the distribution of the entire ecosystem; atmosphere–soil–plant–human. Particularly, the technogenic anomalies of heavy metals are described many times for ore districts of Alaverdy and Zangezur of Armenia. The special investigation about monitoring of technogenic geochemical anomalies can be successfully used for solving series of geoecological problems [Gevorkyan R.G., Mazmanyan L.G., Sargsyan A.H., 2007].

The Extraction of Heavy Metals From Technical Waters. There are many data in literature about wide application of zeolites for extraction of heavy metals from industrial waters. Particularly, the powder of phillipsite-chabazite composition is used for extraction of Pb^{2+} and the zeolites-phillipsite, shabasite, clinoptilolite, mordenite are tested for extraction of Cr^{3+}. The Na-chabazite is used for extraction of Cd.

The acidic mine waters were subjected to purification with the following composition: 2<pH<3. Metals: Al, Ca, Cd, Co , Cu, Fe, K, Mg, Mn, Na, Ni, Pb, Zn.

The long time contact is needed when passing through the bilayer zeolite construction for purification. The ability of clinoptilolite tuffs to extract cations of metals which usually are present in acidic drainage waters is high enough [Gevorkyan R.G., Mazmanyan L.G., Sargsyan A.H., 2007].

In general, comparison of the adsorption properties of clinoptilolite, mordenite, chabazite and activated carbon showed that clinoptilolite exhibits the highest selectivity for the radionuclides and heavy metals.

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Maghidov S.Kh. Economic use of underground waters in RF and exhaustion elastic potential of the Earth's bowels

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Abstract. Based on the analysis of underground water extraction in the Russian Federation problems are discussed related to their protection and management. An examination of the dynamics of water use and compare them with changing the share of flowing wells in the oil and gas industry (OGI) allow to obtain some information about the adverse changes in the Earth's bowels, which is necessary to produce science-based predictions about the geohydrosystem transformation under various scenarios. Concern is expressed about the current state of water infrastructure, particularly aspects such as the depletion of groundwater resources and their pollution, as well as the reduction of the elastic energy of bowels. In this connection, there is the need for extensive study of the problems and the adoption at the federal level urgent measures to drastically improve the situation in this sphere.

Key words: *undergr*ound water, drinking water supply, compressibility potential *of bowels, the proportion of* flowing wells, *oil and gas industry,* geo-ecological catastrophe.

Citation: Maghidov S.Kh. (2013). Economic use of underground waters in RF and exhaustion elastic potential of the Earth's bowels

Underground freshwater is the most valuable of natural resources and require careful treatment. It is the main reserve for drinking water supply of population in the future. This is connected with the fact that considerable part of surface watercourses is much polluted and doesn't meet sanitary and hygienic demands. In the arid regions underground water (UW) UW provides most of the population with fresh water. The share of UW in drinking water supply of Daghestan exceeded 70% in some years. By the end of 80s the portion of UW in drinking water supply of the Soviet Union towns was 65-70% according to the famous Soviet hydrologist V.M.Goldberg [*Golgberg, 1987*]. In 2011 needs in drinking water more than 60% of urban population including settlements was met by underground sources, about 20% of them have mixed sources of water supply. Rural population had the share of UW for potable water supply of 80% that time. Moreover, the tendency of growth of this index is shown in many regions, despite of more intense pollution of underground hydrosphere due to intensive engineering [*Golgberg, 1987; Plotnikov, 1983*].

There are two ways of UW extraction: production of UW and water drainage. In 2009 these parameters were accordingly 22,9 and 4,6 mln. m^3 a day.

In the Russian Federation 3 types of water supply are allocated conditionally: drinking (potable) water supply (DWS), industrial and technical water supply (IWS) and agricultural water supply (AWS), containing irrigation of grounds and flooding of pastures. The portions of UW used for different needs are shown in the Fig. 1.

Analysis of information for the last 20 - 30 years allows identifying unfavorable tendencies in the production and use of UW. Dynamics of these parameters is shown in Fig. 2 and 3.

Despite of growing requirements in freshwater and increasing in underground water resources, the Fig.2 shows that the extraction and use of them have been reducing, and the rate of decline of the needs ahead of the production decline. The dynamics of UW extraction for population water consumption and withdrawal of liquid in oil and gas sphere (oil + associated water) are given in the Fig.3. The Fig.3 shows that the curve of UW extraction for considered period demonstrates steady descent at the time when extracting the liquid in oil and gas industry (OGI) has been increasing. Besides, the information from the Fig.3 testifies that the scales of extraction of UW used by population are higher than volumes of extraction of liquid by oil and gas industry. That's why the influence of water management activity on underground hydrosphere, including compressibility potential (CP) of bowels, may be considerable.

The development of oil and gas deposits leads to depletion of CP of the Earth' bowels, and there is a number of publications about it [Magidov, 2011; Magidov and Musaev, 2011]. If to consider the proportion of flowing wells (FW) exploited by oil and gas industry as parameter illustrating exhaustion of bowels and follow 50-years dynamics of this index in such a large region as the Russian Federation, it turns out that there were considerable changes for this comparatively short period of time. This tendency is represented graphically in the Fig.4. The progressing depletion of oil and gas deposits results in transformation of hydrogeological systems, that is expressed in decreasing of elastic energy of hydrostatic head systems in the zones of oil and gas extraction; this, in turn, leads to reducing of the FW share and respective increasing the amount of oilfield wells exploited by pumps and other means. The similar tendency is observed in the analysis of gas extraction dynamics.

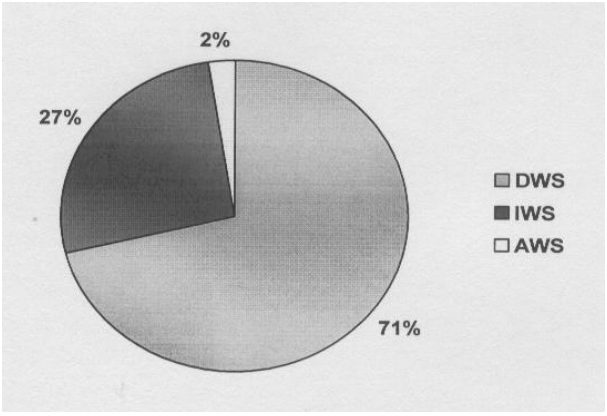

Fig. 1. Use of underground water in the Russian Federation in 2009.

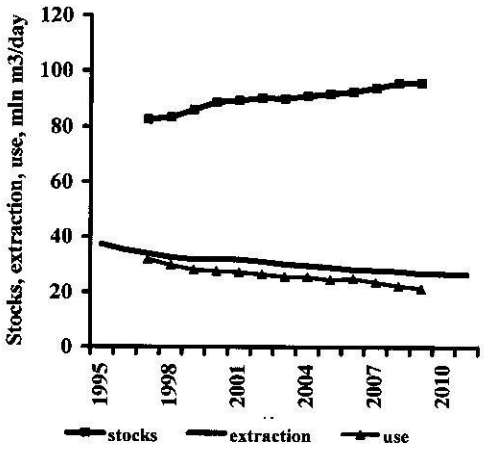

Fig.2. Resources, production and use of underground water in Russia, mln m^3/day.

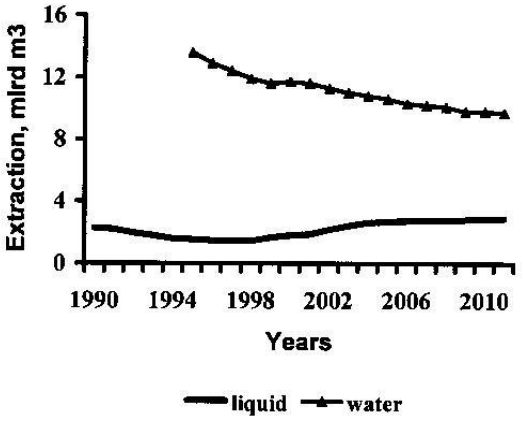

Fig.3. Production of underground water in the Russian Federation and withdrawal of liquid in oil and gas branch.

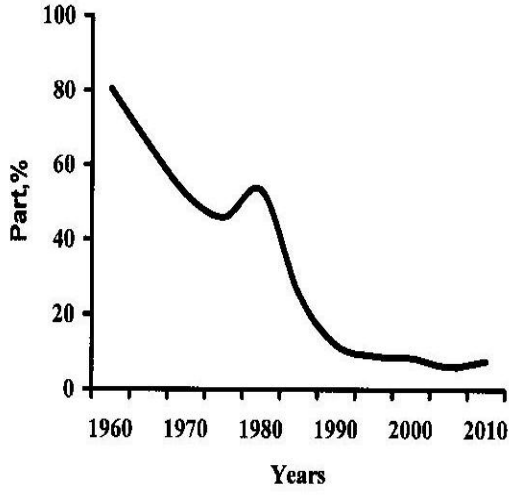

Fig.4. The proportion of flowing wells in the Russian Federation.

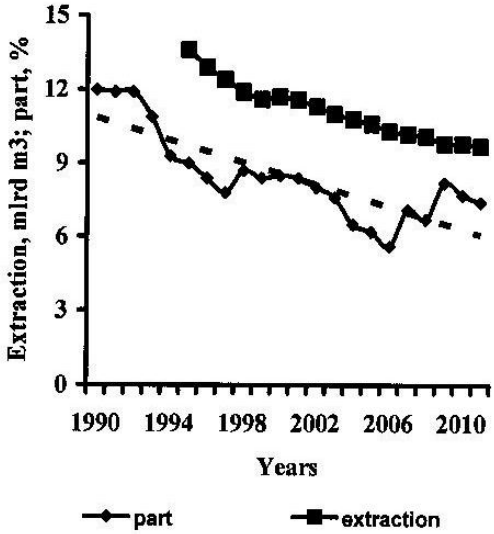

Fig.5. The dynamics of the share of flowing wells in the oil and gas branch and production of underground water in the Russian Federation.

Significant drop in the share of UW demonstrated in the Fig.4 shows major changes taking place in geohydrosystem. It should be considered that there are not some local changes in a small area, but affecting the areas of greater than 10% of the Earth's surface. And when one considers the fact that there are similar changes in the subsurface hydrosphere of other regions, including such large ones as the United States, one probably can already speak about global changes of the geological environment. Apparently, these changes will only grow with time, because the further increasing in demand for underground fresh water, which is better than the surface water, is expected in the future. We examined the effect of oil and gas production to reduce CP of bowels in the previous works. Obviously, the production of UW will also cause a change in temperature and pressure conditions in the bowels, so there must be some correlation between the dynamics of UW extraction and changing of parameters characterizing the geophysical fields in the geohydrosystem. The Fig.5 tells about it.

If to compare the curves of UW production and change in the proportion of FW in the oil and gas industry, their correspondence is evident, especially if to follow the tendency. Trend in the Fig. 5 is almost parallel to the curve of UW extraction. It may reflect the fact that reduction of stratal pressure, which is an indicator of change in the share of FW, makes extraction of UW difficult. On the other hand, perhaps, the withdrawal of fluids reached such proportions that even drop in UW production for potable purposes can't compensate the further adverse changes in the subsurface hydrosphere. Moreover, that although potable UW extraction is decreasing, but the total withdrawal for OGI is growing, as it is evident from the data of the Fig.3.

It is also necessary to take into account the increasing extraction of natural gas. Thus, the total volume of the fluid extracted can increase that explains the continuing decline in the share of the FW (trend). You also need to take into account man-made emergency defluidization of subsurface connected with material wear of old wells equipment (in operation, conserved and liquidated).

In this context, the challenge is to evaluate the contribution that water management in changing of compressibility potential of underground geohydrosphere. From the Fig. 3. it follows that the effect of water management on CP of subsurface can be stronger than the

impact of the oil and gas industry. However, here, it is necessary to take into account the depth of wells. If the majority of potable groundwater is extracted from the zone of intensive water exchange, the damage to the underground hydrosphere may be not so significant. The situation worsens and the tendency to increase with time, the share of UW for potable drinking consumption extracted from the zone of slow and not intensive water exchange. Considering that the rate of replenishment of stocks in these two areas can be different by two or three mathematical order, it becomes clear all danger of such development of events. In this case, there is not only the probability of pre-exhaustion of groundwater used for drinking, but can be reduced significantly CP of bowels, which can result in geo-environmental disasters, both regional and global scale. Calculations carried out for the last five years (2006-2010) indicate that now the proportion of groundwater extraction in Russia amounted to 27.3% of the operational reserves (average).

Therefore, to obtain reliable forecasts and the adoption of effective measures that prevent the development of adverse events the science-based parameters in hydrogeology must be firmly established. After all, there are differences even on such an important indicator as the rate of water movement in the zones of intensive water exchange. Different groups of influential scientists of Russia give the data, which can vary by orders of magnitude even in the handbooks. So, according to the reference book [Zorkin, 1989], in the zone of free (intensive) water exchange movement of water occurs at a rate of tens of centimeters to meters per year. In another handbook [Chernikov, 1988] the rate of water movement in the zone of intense water exchange are defined within meters to hundreds of meters per year. There are also other publications, in which the maximum rate in this region is an order of magnitude higher. After all on this benchmark index depends the other parameter - the duration of the water exchange. According to the reference data for the zone of active water exchange the duration of water exchange is hundreds of years, and for hindered water exchange zone – tens and hundreds of thousands of years [Chernikov, 1988]. This means that the underground water, particularly in the regions of difficult and rather hindered water exchange we can very conditionally consider as renewable resources. From this it follows indisputably that it is necessary to take drastic measures for the protection and sustainable use of groundwater.

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Martynov K.V., Konstantinova L.I., Proshin I.M., Zakharova E.V. Solutions composition effect on plutonium leaching from the aluminophosphate glass

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Abstract. ^{238}Pu leaching from model aluminophosphate glass was studied at contact with different leachant: "bentonite" and "kaolinite" water in a static mode under normal conditions. The expected indicators of ^{238}Pu leaching in "bentonite" water calculated on experimental data were 10^6 times higher than the same indicators in "kaolinite" water. Thus, bentonite use for creation of engineering protective barriers of storages constitutes potential danger to HLW disposal in the form of aluminophosphate glass. At the same time, kaolinite is quite acceptable inert material in this regard.

Key words: aluminophosphate glass, leaching, plutonium, bentonite, kaolinite.

Citation: Martynov K.V., L.I. Konstantinova, I.M. Proshin, E.V. Zakharova (2013). Solutions composition effect on plutonium leaching from the aluminophosphate glass.

Primary leaching tests for stability of matrix materials can be carried out in the distilled water as leachant. The following stage is the leaching in the natural or model ground water corresponding on composition to the water, circulating in the rocks containing HLW storage. The modern concept of solidified HLW disposal in geological formations provides a creation of the multibarrier protection consisting from natural (the massif of rocks) and engineering barriers. Most important of the last ones is clay filling, providing filtration (due to low water penetration) and migratory (due to high sorption ability in relation to radionuclides) protection. As a material for clay filling can be used different in a mineral and chemical composition of clay: bentonite and kaolin. Bentonite clay consists mainly of montmorillonite $(Na,Ca,K)_{0.3}(Mg,Al,Fe)_2[Si_4O_{10}](OH)_2 \cdot nH_2O$, which artificially transfer to a sodium form for swelling capacity strengthening at hydration and increases in sorption capacity. The main mineral of kaolin clay is kaolinite: $Al_2Si_2O_5(OH)_4$ with impurity of Fe, Mg, Na, K, and Ca. In an emergency ground waters as a result of their contact with a material of clay barrier will get specific composition which can determine the rate of radionuclides leaching from the solidified HLW. Thus, for understanding of real leaching rates it is necessary to study interaction in the system: water - rocks - a clay barrier - HLW matrix material.

Table 1. Composition of glass batch for preparation of model glass

Component	Wt.%	Mol.%	Reagent
P_2O_5	53.9	39.4	$NaPO_3$
Na_2O	23.5	39.4	
Al_2O_3	14.5	14.7	$Al(OH)_3$
Fe_2O_3	1.4	0.9	$Fe(NO_3)_3 \cdot 9H_2O$
Cr_2O_3	0.2	0.1	$Cr(NO_3)_3 \cdot 9H_2O$
NiO	1.1	1.6	$Ni(NO_3)_2 \cdot 6H_2O$
Cs_2O	0.5	0.2	$CsNO_3$
CaO	1.0	1.9	$Ca(NO_3)_2 \cdot 4H_2O$
SrO	0.5	0.5	$Sr(NO_3)_2$
La_2O_3	1.5	0.50	$La(NO_3)_3 \cdot 6H_2O$
Nd_2O_3	1.5	0.4	$Nd(NO_3)_3 \cdot 6H_2O$
SO_3	0.4	0.5	Na_2SO_4

Table 2. Cationic composition and pH of the model ground water and leachants, mg/l

Solution	Na	Mg	Al	Si	S	K	Ca	Fe	Sr	pH
GW	73	8.5	0	0.38	12	0	11	0	0.007	7.21
KW	73	5.4	0	2.4	14	1.2	20	0	0.091	7.8
BW	220	1.2	0.18	5.2	21	2.2	1.9	0.14	0.025	9.72

The main industrial technology of liquid HLW solidification in our country is their processing in aluminophosphate glass in a ceramic melting furnace at production association Mayak. The chemical and radiochemical compositions of processed waste and the main components of glass are given in [Polyakov, 1994]. According to these compositions the glass batch (tab. 1) was prepared. The reagents were carefully mixed, crushed and calcinated at temperature 120-400°C in some stages. After that glass batch was placed in corundum (Al_2O_3) crucible. ^{238}Pu (IV) nitrate solution having activity of $1 \cdot 10^4$ Bq/g of glass batch was added in it in quantity sufficient for uniform treatment of glass batch, and then the added solution was evaporated. ^{238}Pu doped glass batch was melted in the closed crucible at 1000°C during 4 hours. The melt was spilled in the graphite molds heated to 400°C, and it was annealed at this temperature within 1 more hour. Translucent dark green tablets with a smooth surface having density of 2.0 g/cm^3 were received as a result.

Model ground water (GW) was prepared by dissolution of $CaCl_2$, $MgSO_4$ and $NaHCO_3$ reagents in distilled water considering data on a chemical composition of the natural ground waters which have been selected at drilling of wells in the rocky massif intended for placement of underground HLW storage. "Bentonite" (BW) and "kaolinite" (KW) waters were prepared at contact within a day model ground water with the clays (1 liter of water on 20 g of clay) at periodic stirring. Then suspensions were centrifuged (8 thousand turns per minute, 45 minutes), a liquid phases were decanted and filtered via two-layer filter "The white ribbon". The received solutions had no opalescence. Chemical compositions of prepared water solutions were defined by inductively coupled plasma mass spectrometry (Perkin Elmer Elan-6100) and inductively coupled plasma emission spectrometry (Perkin-Elmer Optima-4300 DV) methods, the results are presented in tab. 2.

The leaching tests were carried out on the state standard specification (GOST) R52126-2003 procedure at 25°C in a drying box with temperature control in 50 ml volume polypropylene hermetically being closed test tubes. The ratio of water solutions volume to geometrical surface area of glass tablets made 5:1-7:1 ml/cm^2. ^{238}Pu activities in leachates were determined by liquid scintillation spectrometry method on Perkin Elmer Liquid Scintillation Analyzer Tri-Carb 3180 TR/SL.

In accordance with GOST R52126-2003 as characteristic parameter of material leaching resistance the radionuclide leaching rate normalized to the content of radionuclide in solidified waste form is used:

$$R^i_n = a^i_n / (A^i_0 \cdot S \cdot t_n), \text{ g/(cm}^2 \cdot \text{days)}, \quad (1)$$

where a^i_n – activity (Bq) or weight (g) of separate radionuclide i leached for a certain time interval, A^i_0 - specific activity (Bq/g) or mass concentration (g/g) of a radionuclide in solid waste form, S – an area of open geometrical surface of a sample (cm^2), t_n - duration of n leaching interval (days). However the leaching rate in an increment form as it is written down in expression (1) is not the most convenient parameter for processing of leaching tests results because, firstly, it is continuously changing parameter, but test samples are selected discretely. Therefore when studying dependence of $R^i_n = f(\tau)$, where $\tau = \Sigma_1^n t_n$, there is a systematic error which is especially great at small values of τ. Secondly, this function isn't integrated on time from zero as it has uncertainty at $\tau = 0$. As a result it is impossible correctly to calculate amount of leaching radionuclide for this period. To eliminate these problems and to have more opportunities for interpretation of leaching tests results, it was offered [Kotelnikov, 2012] and it was proved in more detail [Martynov, 2012] to use cumulative parameter – leaching depth as characteristic one:

$$L^i_\tau = \Sigma_1^n (R^i_n \cdot t_n / \rho), \text{ cm}, \quad (2)$$

where ρ – matrix density (g/cm^3). For approximation of dependence of a matrix leaching depth from the time the quasidiffusive model well proved:

$$L^i_\tau = \omega^i \cdot \tau^{k^i}, \quad (3)$$

where ω^i and k^i – model parameters. To calculate of values of incremental leaching rate according to (1) expression (3) is easily differentiated:

$$R^i_n = \rho \cdot \partial L^i_\tau / \partial \tau = \rho \cdot k^i \cdot \omega^i \cdot \tau^{(k^i-1)} \quad (4)$$

However it is more convenient in our opinion to use a cumulative leaching rate:

$$R^i_\tau = \rho \cdot L^i_\tau / \tau = \rho \cdot \omega^i \cdot \tau^{(k^i-1)}. \quad (5)$$

The quantity (weight, activity) of the component leached from a matrix is as

$$a^i_\tau = A^i_0 \cdot \rho \cdot \omega^i \cdot \tau^{k^i}, \text{ g/cm}^2. \quad (6)$$

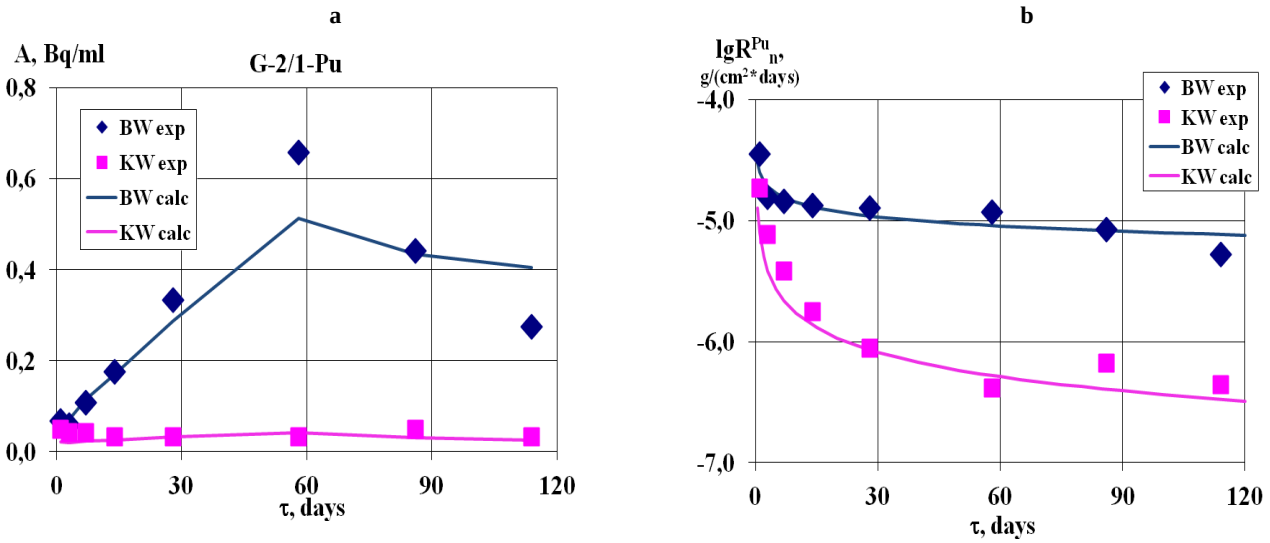

Fig. 1. The activity in leachates (**a**) and incremental leaching rate of ^{238}Pu from aluminophosphate glass **(b)** in "bentonite" (BW) and "kaolinite" (KW) waters on experimental data (exp) and calculation for quasidiffusive model (calc)

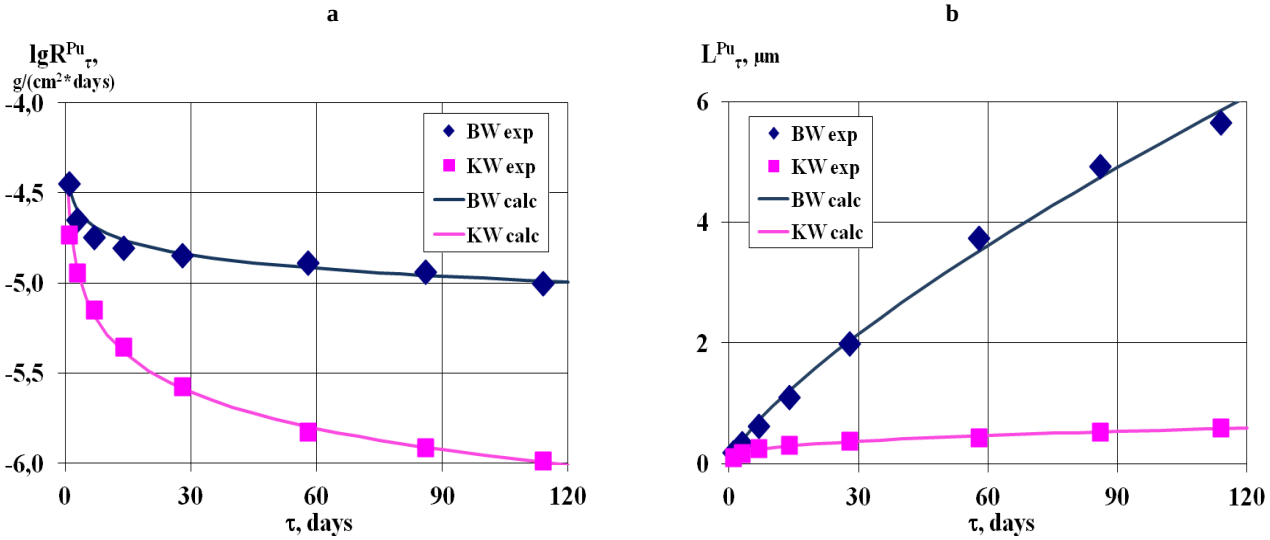

Fig. 2. The cumulative rates (**a**) and depths (**b**) of ^{238}Pu leaching from aluminophosphate glass. Designations see in fig. 1

Table 3. The quasidiffusive model parameters and indicators of ^{238}Pu leaching from aluminophosphate glass in "bentonite" and "kaolinite" waters at the time of achievement of incremental leaching rate $R^{Pu}_n = 1 \cdot 10^{-7}$ g/(cm^2·days) admissible in accordance with GOST R50926-96

$L^{Pu}_\tau = \omega^{Pu} \cdot \tau^{k^{Pu}}$	ω^{Pu}, μm	k^{Pu}	R^2	$R^{Pu}_n = 1 \cdot 10^{-7}$, g/(cm^2·days)		
				τ, days	L^{Pu}_τ, μm	a^{Pu}_τ, Bq/cm^2
BW	0.17(3)	0.75(4)	0.995	$4.34 \cdot 10^9$	$2.887 \cdot 10^6$	$5.78 \cdot 10^6$
KW	0.12(1)	0.33(2)	0.982	690	1.05	2.1

Basic experimental data are presented in fig. 1a. Changes of the leachants were carried out in 1, 3, 7, 14, 28, 58, 86 and 114 days from the beginning of experiments. Durations of the leaching intervals made $t_{1 \ldots n}$ = 1, 2, 4, 7, 14, 30, 28 and 28 days. ^{238}Pu activity in leachates grew with increase in t_n and began to decrease at consecutive exposures of equal duration. It is especially brightly expressed for "bentonite" water in which activity leached ^{238}Pu exceeded this indicator for "kaolinite" water much. Thus, any signs of saturation of solutions at

leaching experiments we didn't observe that is a basic condition of carrying out the tests.

The incremental leaching rates of ^{238}Pu from aluminophosphate glass in accordance with GOST R52126-2003 calculated according to expression (1) are shown in fig. 1b. In both leachants they decrease over time but in "kaolinite" water the rate decreases quicker and to the end of experiments the difference makes more than 10 times. The same effect is more visually expressed for the cumulative leaching rates calculated by the left part of expression (5) and shown in fig. 2a. It occurs owing to that integrated function is more smooth than differential one and the first of them experiences less fluctuations connected with an errors of experimental data. For calculation of the cumulative leaching rates previously according to expression (2) the cumulative depths of leaching were calculated (fig. 2b). It is obvious that the leaching depth of aluminophosphate glass in "bentonite" water increases over time quicker, than it in "kaolinite" water, and during experiment the difference reaches one order.

In comparison with "kaolinite" water, "bentonite" water possesses bigger aggression first of all because of the increased alkalinity. The difference of pH of the prepared leachants makes two units (tab. 2). It remains fairly not only for the modified bentonite processed by caustic sodium solution, but also for the natural clay containing impurity up to 2 wt.% of sodium carbonate.

Experimental data on the leaching depths (fig. 2b) were a basis for calculation of quasidiffusive model parameters (3) by means of the regression analysis. The model parameters and coefficients of determination R^2 are presented in tab. 3. Curves of leaching parameters calculated on expressions (3)-(6) are shown in the figures. Apparently on figures also it is confirmed by R^2 values close to unit, settlement curves well describe experimental data. It allows to hope that their extrapolation on time also will be sufficiently correct.

In accordance with GOST R50926-96 the value of admissible leaching rate of Pu makes $1 \cdot 10^{-7}$ g/(cm^2·days). The forecast on quasidiffusive model shows that this value of leaching rate for aluminophosphate glass in "bentonite" water will be reached only through 11.9 million years. During this time the cumulative leaching depth of aluminophosphate glass will make 2.89 m, and a quantity of Pu taken out from the matrix – $5.78 \cdot 10^6$ Bq/cm^2 at specific activity in the solid equal $1 \cdot 10^4$ Bq/g. For "kaolinite" water these indicators make respectively 690 days, 1.05 microns and 2.1 Bq/cm^2 that is six orders less.

Thus, bentonite use for creation of engineering protective barriers of storages constitutes potential danger to HLW disposal in the form of aluminophosphate glass from possible essential acceleration of processes of radionuclides leaching at contact of the matrix with water solutions in an emergency. At the same time, kaolinite is quite acceptable inert material in this regard.

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Rudakov V.P. Time-space structure of the emanatsion fields in the radioecological monitoring problem

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Abstract. Long-term experience of the geodeformation processes monitoring in the geodynamically (seismically) active and platform regions of the former USSR with use of the emanation method has allowed reveal some time-space regularities of the emanation field's formation, which are offered to be used at realization of radioecological researches. These regularities, displaying the fluid-transportation dynamics under influence of the geodeformation processes, predetermine determinacy of the emanation field's formation in each concrete region that allows to define places of the radioecological measurements realization a priory.

Key words: emanation monitoring, geodeformation processes, mosaic of emanation fields, radioecology.

Citation: . Rudakov, V. P. (2013). Time-space structure of the emanatsion fields in the radioecological monitoring problem.

Introduction. At the beginning of 90th years of the last century in Russia under influence of " radonic boom", initiated in the western countries, the State program "Radon", implicated realization of researches on studying influence of the radioactive emanations on the formation of a negative background of an inhabitancy of the person was created. However because of the limited financing this program has not received wide development, and then and in general was closed. Nevertheless, interest to a problem has not died away, to what the publications connected to attempt of the account of time factors of receipt of an emanation from rocks to the near surface layer of atmosphere [Klimshin; Mareniy; Zaitsev, Rogalis, Kuzmich, 2008]. Thus it is marked, that for practical purposes, connected to questions of an estimation of dynamics and scales of radon receipt in building constructions, long-term monitoring practically it is not real. This circumstance has induced the author, basing on experience more than 30-years researches of geodynamic processes with use emanation (radonic) measurements [Rudakov, 2009] to specify those regularities which predetermine of the emanation fields behavior, and which are expedient for using at realization of radioecological researches, essentially having limited volumes of spent measurements.

Time-space mosaic of emanation fields and its radioecological consequences. The main feature of the emanation fields is, that in the geodynamically active (seismoactive) regions and in the conditions of platforms the variation of radon fields are hierarchically structured both in time and in space, submitting to a wave nature of processes inducing them. Second and not less important circumstance it - that variations of radon are subjected to a modulating influence both internal (geodynamic) and external (meteorological) factors, so hierarchically structured. But, perhaps, the most important is, that in

zones of dynamic influence of tectonic faults the seasonal (year) course of subsoil radon variations is caused not by thermal diffusion as usually is considered, but buy the geodeformation wave migrating on the part of equator to poles of the Planet. This wave operates the appropriate processes of the fluid-transportation in the sedimentary cover of the earth's crust and can reach the levels, capable to provoke various geodynamic events [Рудаков, 2009] which periodicity in a seasonal cycle precisely follows the moments of passage in each concrete territory of region (latitude on which that territory is) of the geodeformation wave's extremes.

In figure 1 is shown the graphic dependence of time arrival on concrete latitude of extreme levels of the seasonal geodeformation wave, received according to the subsoil radon monitoring on various grounds of territory of the former USSR. This dependence is of interest to the radioecologists as a priori enables to establish in each concrete territory the periods of the maximal receipt of emanations to the near surface layer of the atmosphere. So, for example, if for Caucasus it is the winter months, at latitude of Moscow it is July - August, i.e. summer. The given diagram allows determine the periods of year when receipt of radon from rocks to the near surface layer of the atmosphere is maximal for any territory of Northern hemisphere and, being guided by them to carry out practical measurements of the emanation streams.

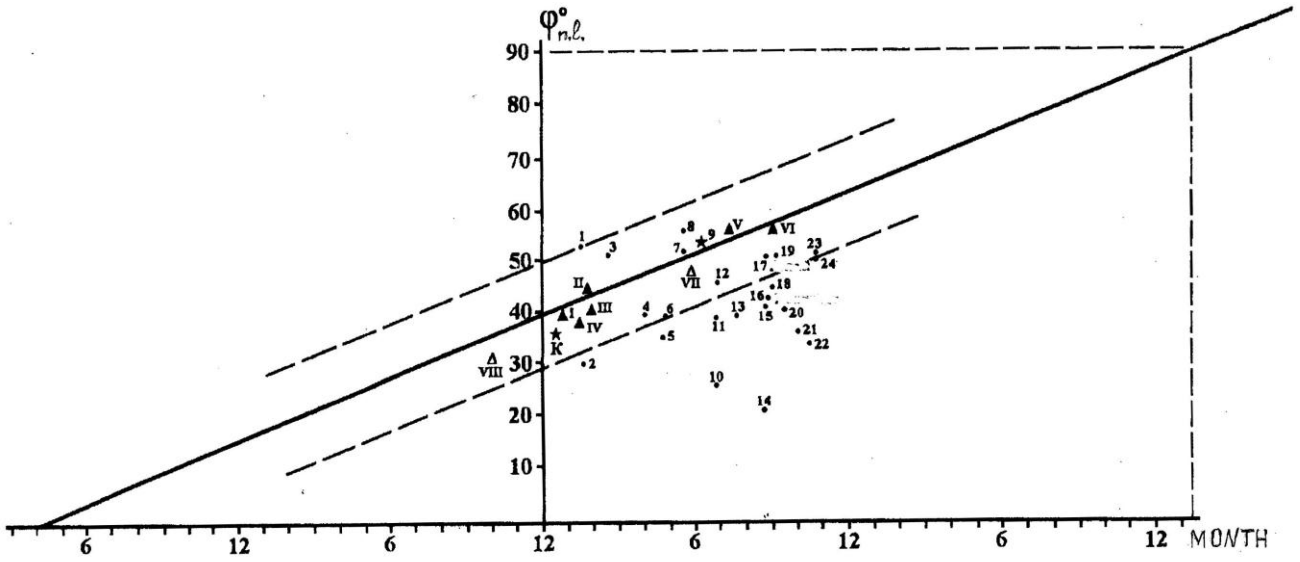

Fig.1. The diagram of time dependence of the seasonal geodeformation wave extremes passage from the latitude of point of supervision.

However, if these periods steadily keep the position in an annual cycle peak parameters of them vary, submitting to modulating influence of processes of higher hierarchical levels, such as processes 4, 6, 8 years periodicity etc. Thus the amplitude of the exhalation emanations variations in an annual cycle in the same region can vary more than on 30 % [Rudakov, 2009].

From the periodic processes shown inside a year cycle, from the point of view of radioecological estimations the variations connected to tidal influence of the Moon at the moment of approach of a full moon and a new moon, and also intraday tidal variations with the periods of 24 and 12 hours are of interest. And those and others in an obvious kind show themselves in zones of dynamic influence of tectonic faults with small on capacity a zone of aeration, i.e. in zones of faults strongly irrigated. Such faults are usually traced by channels of the rivers, streams, ravine net etc. Also such territories more often are allocated by a network under construction of habitation as are zones of biological comfort.

On fig.2 are shown examples of the subsoil radon variations in the faulting zones, preliminary revealed by the emanation survey, in time of preparation and realization of various geodynamic events, including, in one of a similar zones in territory of Moscow from which it is visible how grate the scope of the variations connected, in particular, with tidal lunar-solar deformations of an earth's crust. These zones, being strongly irrigated, are channels of intensive gas exchange in the rocks of a sedimentary cover. In consequence of the aggression rocks destructing processes in them is much higher, than in the not broken blocks of the rocks that promotes formation of the linearly extended zones of carst-suffosion processes development determining, in turn, the appropriate configuration of territories of ecological risk. The high level of intensity of gas exchange in the rocks of a sedimentary cover loosened by the geodeformation processes defines the appropriate intensity of carrying out of the radioactive emanations (radon and toron), and also electrically charged products of their disintegration, assimilated by the ascending streams of the subsoil fluids. Thereof in the certain periods of year the radio-activity of atmospheric air above zones of the geodynamically weakened lineaments the geological environment usually essentially (sometimes on orders) exceeds maximum permissible levels of concentration of emanations which in Russia for inhabited objects make 100 Bk/m^3, i.e. 100 disintegrations per one second in cubic meter of air. The given circumstance speaks that in the not broken rocks the emanations, transferred to the surface by means of diffusion, practically completely breaks up, not having reached the ground, and in conditions of the geodynamically active zone the emanations are transferred compulsorily to the account pnevmo-diffusion and

convective streams of the subsoil fluids. And both and in other conditions even typical sandy-argillaceous accumulations (depending on their mineral structure and open porosity) on depth more than one meter each cubic meter contains in those from 2 up to 10 thousand breaking up atoms of radon and approximately as much toron. So in conditions of the geodynamically active zone in the near surface layer of an atmosphere are transported significant volumes of the emanations repeatedly raising a radio-activity of the air. Thus it is abnormal a high level of a radio-activity in such places because of extreme big nuclear weight and concerning the big half-life period of emanations it can be kept during long (about one month) time intervals.

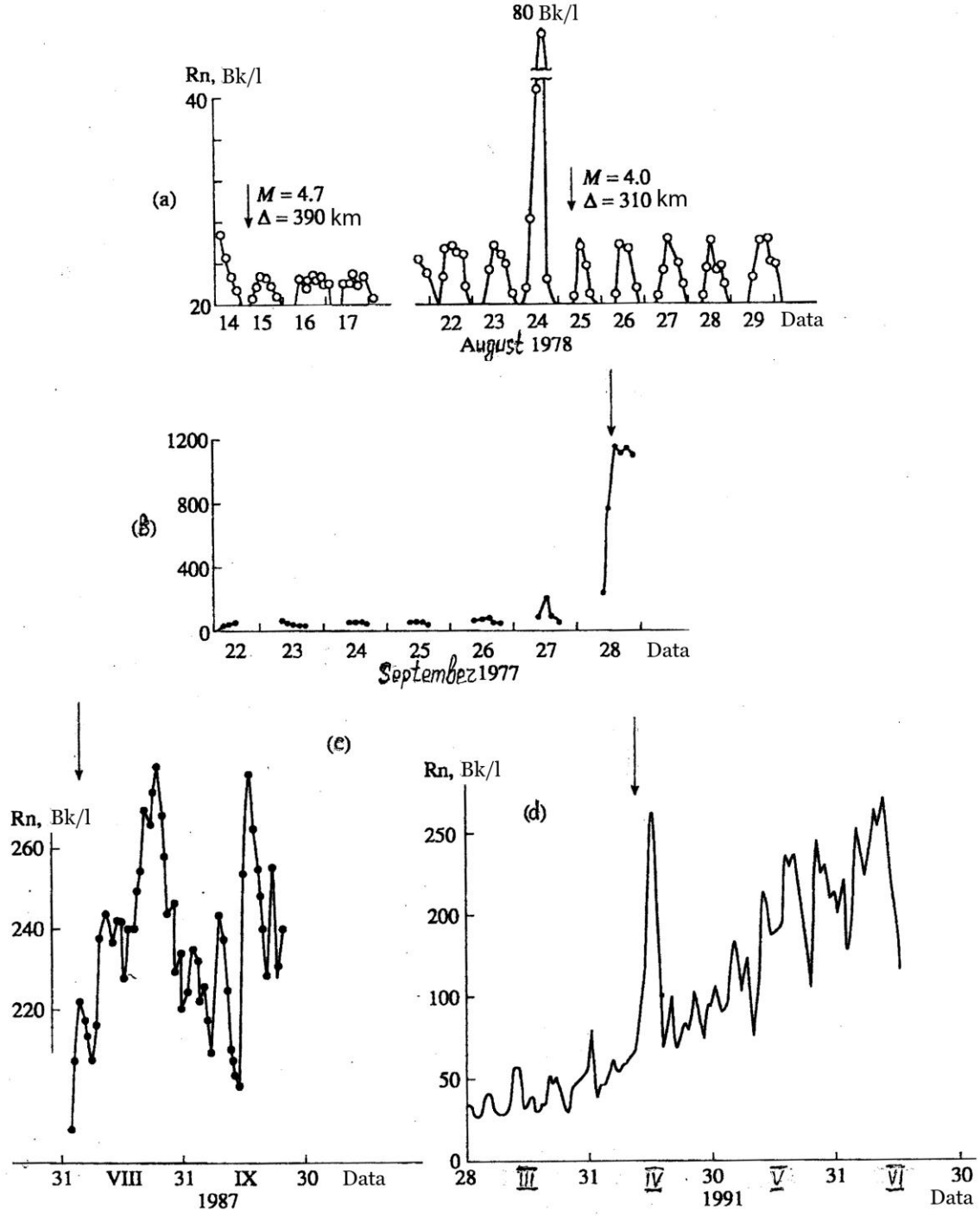

Fig.2. Examples of short-term abnormal effects in the subsoil radon field from various geodynamic events:

a) On the Paratunka (Kamchatka) deposit of the termomineral waters before the remote earthquakes;
b) on the landslide slope in Dilizan (Armenia) before and at the moment of activization of a landslip; в) at the foot of a volcano Klyuchevskaya Sopka before and at the moment effusive-explosion phases of eruption; г) in a zone of the carst-suffosion processes development in territory of Moscow after powerful seismoprospecting explosion in the Ryazan area.

In conditions of mega polices effects of compulsory carrying out of emanations to the surface amplify by the "pump" effects of underground, especially in places of crossing of its lines with zones of geodynamic activity. Moreover, results of complex monitoring have shown [Rudakov,Tsiplakov, 2011], that the level of the radon

exhalation is in direct dependence on intensity of seismic noise and fields of the magnetotelluric current, producing by the various enterprises of city, and, first of all, by the underground.

The named factors are one of the seen reasons of the destructive processes development processes accompanying with infringement of stability of the geological environment in zones of geodynamic activity, and, attracting for themselves destruction of technical constructions (underground communications, road coverings, the bases of buildings etc.). These processes, from our point of view, are also a source of the various geopathogenic displays determining a level of the social - medical parameters degradation and biological comfort close to the geodynamically active territories of the city [Melnikov, Rudnik a.e., 1994; Rudakov, 2000].

Structural features of geoenvironment in the emanation fields mosaic. Using of the emanation survey at the structural – geodaynamic mapping of various elements of the geological environment has allowed to establish, as it is marked above, that emanation fields are hierarchically structured even within the limits of insignificant territories, submitting to the features of the tectonic structures dynamic, in a zone of which dynamic influence there is a concrete area [Rudakov, 2009]. I.e. it was established, that as a result of the wave geodeformation processes influence in tectonic faults of the crystal base, in the thickness of the sedimentary accumulations overlying them are formed the fluid-transmition channels which, in turn, determine the density's heterogeneity mosaic, apparently, petrophysically homogeneous sediments. And these features are precisely enough displayed in results of the emanation surveys. And from the point of view of radioecological estimations interest is represented with territories of the output on a surface of channels increased fluid-transparency through which more intensive carrying out of emanations in the near surface layer of the atmosphere is carried out. These places in results of the emanation surveys are allocated more than triple (concerning background variations) with increase of concentration of radon at searching profiles that testifies to their unequivocal connection with channels of the increased fluid-transparency of the sediments.

Conclusion. Proceeding from long-term experience of the emanation method use at the decision of the seismo-prognostic, explorative and other problems, some of which are shown above, with reference to the decision of problems radioecological it is necessary to mean the following.

1) At realization of the radonometric control on the areas of construction of inhabited objects it is necessary to carry out emanation surveys for definition of the fluid-transparency mosaic of sediments. 2) In the revealed zones of increased fluid-transparency it is expedient to carry out the estimation of dynamics of receipt of the radioactive emanations to the near surface layer of the atmosphere by means of continuous and passive (without compulsory choose of samples) measurements. 3) At performance of radioecological estimations in each concrete region it is necessary to be guided by character of development of the seasonal geodeformation processes determining dynamics and a level of the exhalating emanations.

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Rudakov V.P., Tsyplakov V.V. Barometric effects in variations of subsoil radon and of the rock's sample containing radium

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Abstract. By the studying of the variations influence of the atmospheric pressure on the rocks emanation abilities change were the comparative measurements of the subsoil radon variations in situ and variations of radiation of the rock's sample containing radium in laboratory carried out. Measurements were accompanied by measurements of variations of atmospheric pressure. Results of the analysis have allowed to reveal some common regularities of the variations of radon both in situ and in the radioactive sample, connected with dynamics of disintegration of the emanation products. Besides components of variations of the radon, connected with variations of atmospheric pressure, including, caused by influence of lunar-solar tides were revealed. And, tidal components are revealed and in the spectrum of the subsoil radon variations radon and in the variations spectrum of the radioactive sample radiation. Moreover, effects which for their explanation need realization of more refined researches are revealed.

Key words: rocks emanation, baric variations, fluid-transportation, seismic noise, subsoil radon.

Citation: Rudakov, V. P., V.V. Tsyplakov (2013). Barometric effects in variations of subsoil radon and of the rock's sample containing radium

Introduction. By the problem of research of influence of various factors on the rocks and minerals emanation began to be engaged from the very beginning of radiometry development when there was a necessity of the estimation of scales of the radioactive ores prospecting deposits [Shashkin, Prutkina. 1979]. In a more later time this problem was rised, when it was required to explain abnormal behavior of radon in underground waters in connection with catastrophic earthquakes and volcanic

eruptions [Gratsinsky, Gorbushina,Timinsky, 1967]. Also it is necessary to note, that till now in this problem there is a lot of open questions which demand the decision, is especial when these questions concern radioecological estimations [Klimshin, 2008;Marenny, 2008]. Research of the geodynamic processes, carried out by us last years with use of the emanation method [Rudakov,Tsyplakov, 2011;Rudakov, Tsyplakov, 2012], also suggest about necessity of the more deep analysis of those factors which influence on the emanation dynamics and transportation of emanations to « a day time surface ». Therefore the present work is the next attempt to continue studying of these factors with use of results of long-term monitoring.

Technology of experiment and results of researches. Measurement of subsoil radon variations is carried out by us in the territory of EPY RAS already more than 10 years, however at qualitatively new level these measurements became possible due to use of the independent portable registrar the "E-Clerk", allowing to carry out every minute interrogation of the detecting devices, long-term storage of the information and subsequent its copying to a personal computer. Simultaneously with record of the subsoil radon variations the measurements of variations of gamma-radiation of a sample of ore from the Transbaikalian uranium deposit placed in a hermetically sealed flask to limit influence of atmospheric pressure began to be carried out. Besides with the help of the electronic converter constructed on the basis of a barograph the measurements of variations of atmospheric pressure began to be carried out. On fig. 1 fragments of record of the named parameters by duration two weeks (from 19.02 to 04.03.2013г.) are given, which were analyzed with use of the spectral-correlation analysis. On fig. 2 results of calculation in a 9-percentage window of function of sliding correlation of the named time series are given, and in table 1 their spectral characteristics are given.

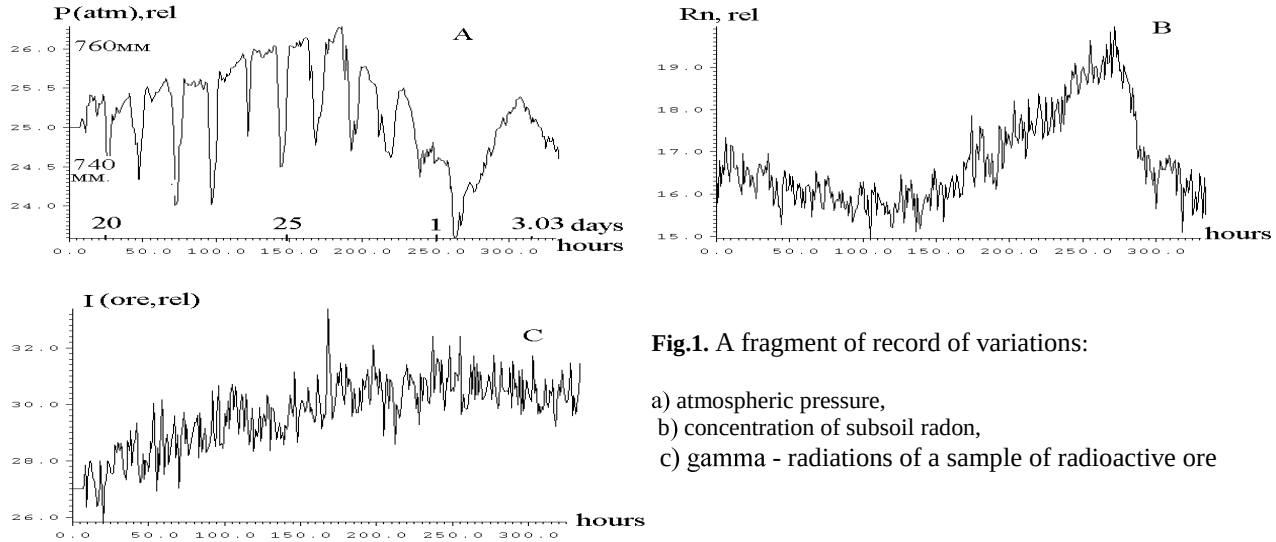

Fig.1. A fragment of record of variations:

a) atmospheric pressure,
b) concentration of subsoil radon,
c) gamma - radiations of a sample of radioactive ore

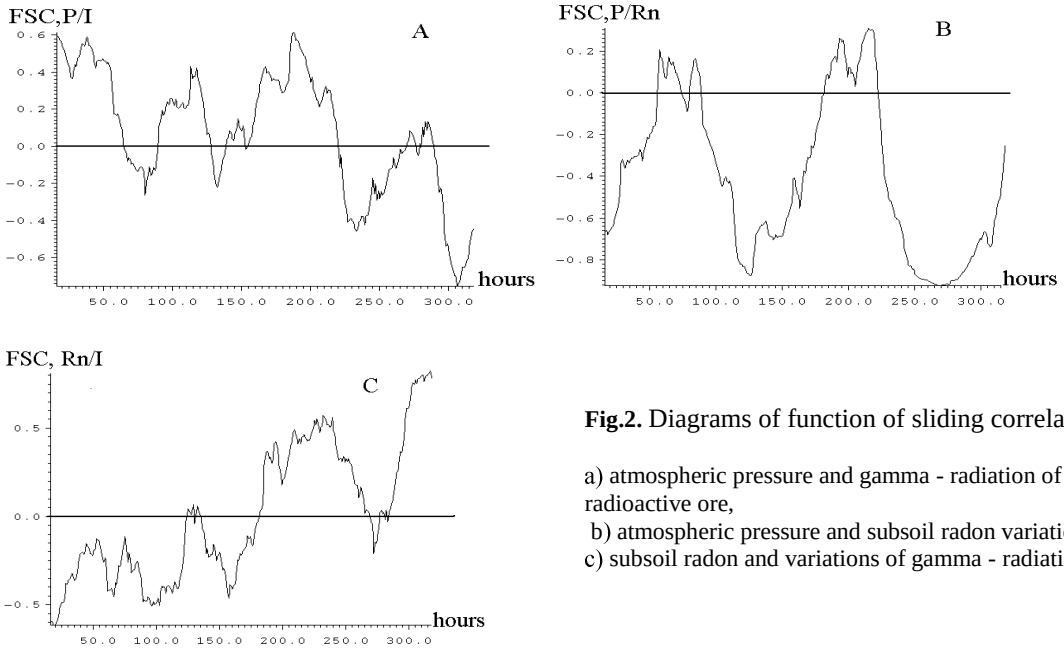

Fig.2. Diagrams of function of sliding correlation of time series:

a) atmospheric pressure and gamma - radiation of a sample of radioactive ore,
b) atmospheric pressure and subsoil radon variations,
c) subsoil radon and variations of gamma - radiation of ore.

The attention pays, first of all, return correlation dependence of subsoil radon variations from variations of atmospheric pressure which in an obvious kind was not reflected in radiation of a sample of the radioactive rock. In too time from results of sliding correlation it is visible, that such connection exists and has the expressed variable character. And, if correlation between the time series of pressure and radiation of a sample changes a sign in 2 days between time series of pressure and subsoil radon it occurs in 3.5 days, i.e. in the first case the cycle is 4 days, and in the second - 7 days, that corresponds to a quarter of a lunar cycle of change of phases. Meanwhile, as follows from table 1, components of variations of tidal genesis with the periods in the field of a daily and semidiurnal range are allocated in variations and atmospheric pressure and subsoil radon and gamma-ray's of the radioactive sample, sealed in a metal flask of the thermos. And, if the given circumstance is easily explained for variations of the subsoil radon which measurement is assumed with a technique of a choice of the detector sites installation [Rudakov, 2009] occurrence of the tidal components in variations of the radioactive sample radiation in laboratory conditions causes, at least, misunderstanding. Therefore in the analysis of results of experiment were involved also records of seismic noise of city by the seismometer, established in a window aperture of laboratory. However, and in variations of seismic noise also were allocated making the daily and semidiurnal range, testifying about their possible tidal genesis.

Table1. The most significant (P > 0.95) components of spectra of variations of atmospheric pressure, subsoil radon and radium-containing ore, and also spectra of functions of the sliding correlation**.**

Spectra name	**F1 hour**	**F2**	**F3**	**F4**	**F5**	**F6**
Atm. pressure	59.7	23.4		11.95	7.9	4.7
Radon	59.7	23.4			7.3	3.95
Ore		23.4	17.3	10.8	7.2	5.0;4.0
Pressure/radon	128	25.6	13.8	12.2; 9.0	7.2	
Pressure/ore	170.6	24.4		12.2	7.4	
	85.5					

As to the character of dependence of the subsoil radon variations from the variations of atmospheric pressure, in that area where it (character) is negative, the interrelation speaks influence on process of carry of an emanation of the subsoil moisture reducing with increase of atmospheric pressure receipt of fluids to the near surface layer of the atmosphere. When this interrelation has positive character, i.e. the increase of atmospheric pressure causes increase of the subsoil radon concentration to explain this phenomenon it is possible only from a position of a hydraulic drive of front of pressure in an installation site of the detector as it was earlier marked in conditions of Kamchatka [Rudakov, 2009]. Besides these effects are formed by variations of seismic noise in relation to which the subsoil radon variations have also sign-variable character [Rudakov, Tsyplakov, 2012].

Conclusion. It agrees to the data of experiment the variations of the subsoil radon and of the gamma - radiations of the radioactive ore's sample are in strong dependence on variations of atmospheric pressure. And this dependence reaching periodically 90 %-s' levels, has the sign-variable character changing a sign in 2 and 3,5 days. A nature of the established connections yet it is not possible to explain proceeding from the existing representations about the processes of the emanation and transportation of the radioactive gases in the natural environments.

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