Risk Assessment for Underground Disposal of Radioactive Wastes in Clay Strata

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Abstract. Radioactive wastes (RAW) storage in clay strata followed by long-term radiation effect on clays results in a noticeable decrease of shear strength and deformation property, with intensification of microbiota activity being a negative factor. Clay properties change provokes the disturbance of hydrodynamic condition and enhances a contamination hazard of water-bearing horizons. Fundamental hydrogeological and geoenvironmental problems require implementation of special engineering treatments for protection of water-bearing horizon used for water supply.

Introduction

Accumulation of considerable amount of radioactive wastes (RAW) as a result of various manufactures activity in the Leningrad region, should stimulate the solution of the problem of its safe storage and / or processing in the short time. RAW have been buried in surface storages of the specialized industrial complex "Radon" and in burials of the Leningrad atomic power plant (LAPP) for a long period of time. At present due to absence of RAW storage the problems of disarrangement of nuclear pipelines which service garantees have been expired at LAPP and nuclear fuel removal can't be cardinally solved.

Some experts in nuclear physics recommend underground disposal of RAW of the Leningrad region to be embedded within Vendian Upper Cotlin clays and Lower Cambrian blue clays , while the blue clays are preferred ones due to its higher plasticity, content of clay particles as well as to its surface occurrence over the pre-glintal territory, where the wastes are supposed to be buried.

As the thickness of blue clays does not exceed 130 m in the region observed, RAW must be buried in special underground workings.

General features of Lower Cambrian blue clays.

In the design of RAW underground disposal in clay massif is based on the assumption that such deposits are fine-porous non-cracking medium with the low filtration capacity, low diffusive permeability and high sorption capacity. Besides, development engineers declare that clays are resistant to the action of radioactive irradiation, high temperatures; at the same time they are inert to corrosive mediums.

The studies have shown that Lower Cambrian blue clay strata should be considered as a unitized medium with a deep zone structure. Each zone is characterized by the intensity of different genesis of fissuring: tectonic, glacio-tectonic, litho-genetic, weathering, reconsolidation and others. Tectonic and litho-genetic fissuring form an unitized structure of blue clay massif: a bloc size increases regularly from the top downward. The depth of a reconsolidation zone usually does not exceed 25 m and is characterized by higher values of moisture content and lower values of density. Each zone distinguished according to its depth has a definite variability of parameters of physical and mechanical properties depending on fissuring intensity and a degree of clay reconsolidation.

Radiation stability of rocks and soils.

At present there are few researches done concerning the influence of radioactive irradiation on rocks and soils, including clay deposits and rock-forming minerals. Radiation stability of minerals usually reduces through quartz to alumosilicates: quartz – microcline – kaolin – hydromica – montmorillonite. According to the statements of the theory of radiation damages in a solid body, crystalline structure of minerals is changed owing to interaction among radiating particles and mineral constituent elements, which are disturbed from equilibrium condition at the points of the lattice and may cause the displacement of other elements. Such processes lead to crystalline structure disordering, vacancies making and forming defects on the micro-level, finally all these phenomena cause the change of mineral properties owing to their amorphisation. Special investigations have shown that amorphisation of quartz after being irradiated is accompanied by an increase in its volume to more than 17%. First of all, the irradiation influences on structural bonds existing between minerals and its aggregates. For example, granite samples are desintegrated to pieces, the size of grains being 1-5 mm at 2.8×10^{20} neutron/cm^2 irradiation.

The exposure of different types of concrete to radioactive irradiation results in the decrease of its strength from 20 to 90% depending on the conditions of irradiation, its action duration, cement composition and filling materials.

It is necessary to pay a special attention to radioactive stability of clay minerals, which interlayer bonds usually results from unlike charged surfaces, or cations which are between the same charged layers. Similar bonds are easily broken down when exposed to radioactive irradiation simultaneously with the structural bonds existing between mineral particles. Change in valent bond vibrations of Si - O and Al - O inside a layer up to its fracture is observed on long-term radioactive exposure. Besides there is dehydratation of clay minerals with consecutive isolation of crystallization water, and structural water in the form of OH^- – groups from the crystal lattice. The removal of crystallization water has an impact on chemical, physico-chemical and physical and mechanical properties of clays, and the removal of structural water leads to a complete fracture of clay mineral.

Being part of clay minerals of Lower Cambrian blue clays are represented by high-alkaline minerals, interlayer links destruction will take place at low enough radiation doses. The main structural defects in clay minerals are developed on radiation exposure, which is 10^5-10^6 Grey. Thus the process of destruction of silica-alumina nucleus of clay minerals is becoming more active in a particular sequence of cation outlet of crystal lattice: $Fe^{3+} > Ca^{2+} > Mg^{2+} > Na^+ > K^+ > Si^{4+}$. Except for the destruction of a solid phase of clays, there is radiolysis of pore water, which results in the formation of free radicals and the molecular components causing the change of acid-alkaline and oxidation-reduction conditions in a clay strata. In blue clays organics and sulphides may be oxidized actively while forming the process of radiolysis of pore water at absorbed doses up to 10^2-10^3 Grey. Besides, a rise in temperature of a soil massif with radioactive decay generally causes activation of the process of water radiolysis. For example, when the chain process of radiation oxidation of substances is in progress, a rise in temperature from 20 up to 80 °C results in approximately a twice rate increase. Complex action of radiation and temperature factors also promotes a deeper degradation of clay minerals.

Transformation of blue clays under the influence of radioactive irradiation.

Studies of Lower Cambrian clays, which have been exposed to radioactive irradiation for 50 years at the bottom of one of RAW storages, have shown considerable transformations of these clays. It is necessary to note that low-active wastes (LAW) with the α – radiation level that did not exceed 10^5 Bk/kg and β – radiation level, less than 10^6 Bk were disposed in the considered storages. Under the action of long-term irradiation the amount of clay minerals, which are identified by means of X-defraction analysis has reduced; in d < 0,002 mm fraction composition only illite and a muscovite were found, while unirradiated clays contained chlorite, hydromuscovite, hydrobiotite, glauconite, less often montmorillonite. Amorphisation of clay minerals is confirmed by the results of analysis for adsorption capacity of blue clays. In blue clays the value of adsorption capacity is 12 mg-eqv/100 gr of soil, in irradiated ones the value of this index has practically double increased - 20-22 mg-eqv/100 gr of soil. Amorphisation of clay minerals has also led to a rise of hydrophylity of blue clays and it has invoked the increase of plasticity index and capacity of these soils to swelling. The value of free swelling of irradiated blue clays has considerably increased in comparison with unirradiated soils. Besides the long-term radiation has essentially lowered soils. Besides the long-term radiation has essentially lowered parameters, such as shear strength and deformation capacity of blue clays (Table 1).

Table 1. Blue clays parameters modification on exposure to radioactive influence.

Region of research	Depth, m	Moisture content	Plasticity index	Value of swelling, %	Shear strength parameters		The module of deformation, E, MPa
					C, MPa	φ0	
Storage of wastes (Sillamyaye)	5.0-0.0	0.19-0.23	0.31-0.41	14.0-20.0	0.019-0.075	0	2.9-7.8
Narva region	5.0-0.0	0.16-0.28	0.14-0.22	4.5-6.0	0.10-0.14	3-4	10.0-28.0
Nikolskoye town, the Leningrad region	3.0-8.0	0.20-0.23	0.19-0.27	5.0-8.0	0.075-0.117	3-8	19.0-24.0

As shear strength and deformation capacity determines the stability of rocks and soils in underground workings, it is necessary to take into consideration a decrease of such parameters on exposure to radiation while designing both underground storages of the radioactive wastes storages in blue clays and underground atomic power stations. Even visual evaluation of Lower Cambrian blue clays that suffered the long-term radioactive irradiation, reveals an intensive degree of their desintegration and fissuring.

Intensification of microbiological activity under the influence of low-active wastes.

While arranging and using LAW storages special attention should be paid to a possible change in micribiological activity in enclosing soil massif. From the point of view of survival rate of microorganisms on exposure to radioactive irradiation, first of all, it is necessary to distinguish the microorganisms in an active state. The former which are continuously capable of elliminating radioactive damages and slumbering ones, in which there is no metabolism and reproduction. Radiation intensity is more important for «active» microorganisms than the rate of the processes of damages recovery. The integral dose, obtained in “a rest” state, is much more important for “slumbering” microorganisms.

It should be noted that only recendly researchers have started to investigate the problem of microbiological activities at RAW burials, whereas a few studies paid attention to the role of microorganisms at deep RAW disposal. As a rule the field of microbiological activity studies should include: a) survival of microbes in irridiation conditions; b) possibility of formation for products of their metabolism and finally biochemical gas; c) role of corrosion of container's walls, caused by microbes; d) microbiological degradation of waste products and materials.

The microbiological degradation of waste products draws a special concern at burial of low-active wastes as they frequently contain a significant amount of organic materials, which are subjected to biochemical degradation. It is necessary to note that according to the data of some specialists the microbiological activity in the zones of burial of low-active wastes exists everywhere and has a significant impact on the geoenvironment. The studies have been conducted in Great Britain in the zone of the underground burial of low-active wastes for 42 years, have shown that microbiological disintegration and corrosion of materials have caused a considerable deterioration of the natural ecological situation. The general trend of the processes was bound up with the formation of reduction conditions and methane generation. The latter could lead to its self-ignition and was especially dangerous.

In our case the first stage of study of the microbiological activity at the bottom of the tailing dump of the low-active wastes storage Sillamyaye was held to estimate bacterial mass at the blue clays bottom section. The value of bacterial mass was estimated by Bredford's biochemical method, which allows to obtain the content of microbial protein in blue clays (Table 2).

Table 2. Microbial protein (MP) content in blue clays

Depth from the roof of blue clays	1,8-3,4	3,5-11,0	12,0-19,0
MP, microgr/gr	204-465* / 284/9	150-365 / 234/10	104-275 / 172/10

* In numerator: minimum - maximum values of an index; in denominator: average value and number of analyses.

Precise regularity of MP values decreasing with the depth (Table. 2) may be explained by the existence of the surface radiation source. As a rule MP does not exceed 10-30 microgr/gr for the considered clays outside irradiation zones and other technogenic effects, where the depth influences insignificantly.

The presence of microbiota, the cells of which are sorbed on dispersed particles, operates as a negative factor, promote additional strength loss, development of plastic properties and generating biocorrosive attack to building and defensive constructions. All these factors should be taken into consideration while designing RAW storages and atomic power stations.

Hazard of water-bearing horison contamination.

It should be noted that Lower Cambrian blue clays serve as the upper confining layer occuring at the top of the Lomonosov water-bearing horizon, which is used for water supply in the western parts of the Leningrad Region (the towns of Ivangorod, Kinguiseppe, Slantsy) and in Estonia (Narva).

At present it is planned to bury RAW of the “Radon” plant and Leningrad atomic plant within the Koporye area, which is 10 km from Finland Bay, where the Lomonosov water-bearing horizon is discharged. Therefore, its contamination has a regional significance resulting not only in contamination of water-bearing horison, used for water suply, but also in the waters of Finland Bay. The considered area of burial is located within the limits of two transcontinental lineaments covering, one of which – Lapland- Nilsk with kinematics of shift and contraction has the meridional direction, and the second one - a northwest direction of strike (extent), - Narva-Amudarya is characterized by the existence of gapping deformations. The territory is crossed by a system of regional and transregional fractures of sublatitude submeridional and northwest directions. At present in the clays of the sedimentary blanket of Pre-quarternary deposits the intensification of fractures takes place when while the intensity of fissuring of water-bearing and waterproof soils increases accordingly.

Desintegration increase of waterproof blue clays strata on exposure to radiation and temperature causes the growth of clay massif permeability, where RAW are to be buried. Besides, it should be noted that the active use of the Lomonosov water-bearing horizon for the purpose of water supply will predetermine the development of a depression, which will cover the Koporye area. Deterioration of waterproof properties of blue clays, the change of the hydro-dynamic situation caused by intensive use of the Lomonosov water-bearing horizon will result in the flowing from upper underground waters. Descending water flow passing through the zone influence of radioactive wastes storage will be enriched by radionuclides and transfer them to the water-bearing horizon used.

Such serious hydrogeological and geotechnical problems require the development of special engineering means of water-bearing horison protection with regard to long-lasting negative changes in waterproof clay strata.