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ON COMPREHENSIVE APPROACH TO USE CLAY MATERIALS AS NUCLEAR FACILITIES SAFETY BARRIERS

Linge I. I., Ivanov A. Y., Kazakov K. S.

Nuclear Safety Institute of RAS, Moscow, Russia

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The article discusses topical issues and problems associated with the use of clay materials as safety barriers in the construction and operation of nuclear facilities. Comprehensive measures are proposed to increase the efficiency of clay material use in addressing long- term nuclear and radiation safety issues.

Keywords: *clay, bentonite, clay materials, barrier materials, radioactive waste (RW) management, radiation safety.*

Introduction

Useful properties of clay-based materials applied to ensure the safety of radioactive waste (RW) management were discussed in [1]. Unique properties of clays and clay-based materials have been known since ancient times resulting in their wide application in different industries. The global volume of clay production is huge and is even considered as a marker demonstrating the national economic growth in different countries. However, in nuclear industry particular features of clay-based materials associated with raw material characteristics and their processing methods have been consistently ignored as such, thus, often resulting in the underuse of the high clay potential.

This paper presents a summary on the state of art in using clay and clay-based materials in construction of new nuclear facilities, as well as during the reconstruction of already existing ones. It also focuses on the lines of actions developed to increase the efficiency and to expand their use.

Effects associated with the use simplified technological approaches in the design solutions suggesting the use of clays have fully manifested themselves at so called first generation nuclear facilities. For example, waterproof clay screens were provided for in the designs of RW storage pools at the Siberian Chemical Combine, constructed in early 1960's. Further operation has revealed that these screens were not able to fulfill their function resulting in the spread of radioactive waste components into aquifers. To mitigate these consequences a set of measures was implemented enabling to arrange some additional impervious screens. Thus, pool B-25 [2] was fitted with 320-meter U-shaped purpose-designed barrier requiring 220 special injection wells to be drilled and to inject over a thousand cubic meters of a gel-forming solution. The results obtained were considered as a success: hydraulic conductivity of the aquifer in the central and middle parts of the barrier associated with the predominant contaminant flow was reduced by 10,000 times [3]. These costly measures, as well as soil contamination near the pool could have been avoided, if the safety barriers were initially constructed using higher-quality barrier materials with proper quality control arrangements in place.

The second, yet more notable case in point is associated with the post-accident clean-up at the Chernobyl NPP [4]. In the shortest possible amount of time, a clay "curtain" type barrier was built to a depth of 30 meters along the outer perimeter of the ChNPP industrial site to prevent the discharge of contaminated groundwater from the industrial site and adjacent territories to the Pripyat River. However due to the following reasons, in terms of attaining adequate radiation safety level these activities have proved to be extremely ineffective:

1) Expected ground water inflow being relatively clean was far not the most important source of the Pripyat River contamination;
2) Construction of the "curtain wall" barrier resulted in the flooding of the ChNPP industrial site, as well as of the adjacent territories imposing risks for the NPP's reactors constructed below the ground level.

To mitigate these consequences a high-capacity dewatering system had to be installed to pump out already highly contaminated ground water (due to the flooding of the contaminated lands and additional contamination of the ground water due to dewatering wells operation) and to discharge it into ChNPP storage pool channel.

At present time, clays and clay-based materials are widely used at nuclear facilities and enterprises, also enabling to address some unique challenges associated with attaining the environmentally safe level at some RW sites. However, usually no comprehensive in-depth consideration is given to the specific properties of the clays applied.

We believe that significant improvement in this respect can be attained by introducing the following elements at an industry-wide level:

- Comprehensive and regularly updated knowledge base on characteristics, properties and application of clays in the considered areas with due account of the particular activities being implemented;
- Incentives should be specified in the provisions of federal rules and norms and other regulations provided that clays and clay-based materials are used in keeping with relevant instructions and recommendations;
- Updating the entire legal framework on engineered activities (in particular, development of design solutions) by introducing relevant data on clays;
- Certain arrangements on introducing the best practices and quality control over the developed designs and implemented activities.

A brief overview of these components is presented below considering two contexts — current situation and what needs to be done.

Comprehensive source of knowledge on the use of clays in atomic energy and competence center

There's a huge amount of scientific papers and reference guides on clays constantly growing in their number. It should be noted that according to statistics: over the past 20 years, more than 50 monographs on the use of clays and clay-based materials was published in Russia. Almost 40 % of these were dealing with the medical applications, 25 % — with pottery and other crafts, slightly more than 10 % — with the use of clays as a raw material for construction, less than 10 % — the use of clays in agriculture and veterinary. Whereas, there were only 4 monographs on the topic of the sorption properties of clays (not associated with nuclear industry). Clay application in nuclear power field is virtually absent in this list. The only exception is the overview of clays as a medium for RW disposal featuring some monographs [5, 6], thematic collection [7] and a number of overview papers. According to the electronic data base of scientific papers, over 10,000 articles on clay-based materials and their applications (covering all fields of their application) have been published in the last 15 years with only some 200 papers associated with the nuclear field.

In a number of cases, active competence centers can be identified, for example: the Republic of Tatarstan can be considered as such when it comes to the use of clays as sorbents in veterinary medicine; experts from Belgorod possess extensive know-how on the use of clays as sorbents for water purification purposes. Based on the literature sources review, one can state that no integrational analytical center on the use of clays at nuclear sites is available. All publications on this topic, except [1], are confined exclusively to specific facilities and sites.

For instance, as part of the FTP NRS, large scale efforts were implemented also suggesting that the issues associated with the use of clay-based materials were addressed for particular sites and facilities allowing for more in-depth scientific study. The latter produced a tangible effect in terms of ensuring the long-term safety. The unique nature of the activities performed and their self-sufficiency can be considered as a particular feature of the activities performed. In all the case studies provided (see Figure 1), the amount and the depth of research demonstrating the feasibility of particular clay-based materials and relevant methods of their processing and installation was defined and limited solely by particular features of the facility and the competencies of the design development and/or operating organization. Obviously, there are also broader prospects for the use of clays similarly to other industries.

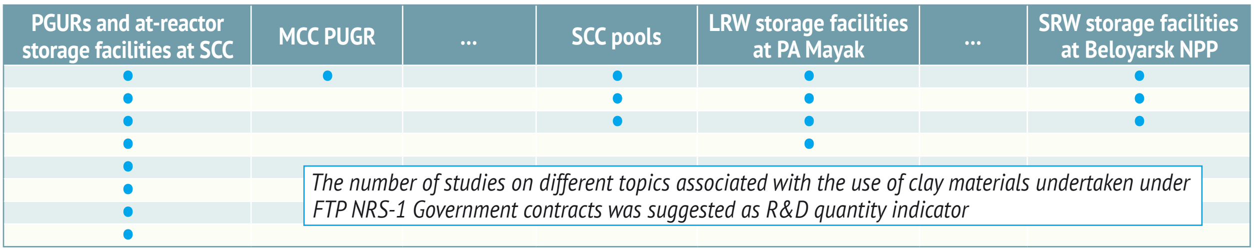

Figure 1. Number of R&Ds substantiating the use of clay-based materials at different facilities implemented under FTP NRS

For instance, a technology exists suggesting the use of bentonite as a structure material for oil and petroleum product storage facilities and tanks at petrol stations providing that an impervious screen (IS) is installed under the soil layer. In some European countries, geosynthetic clay-based materials (certificate number 148-CRP0413/Z) are used to cover the slopes and cuvettes at motorways. It is probably advisable to implement this technology at motorways where intensive LRW and radioactive substance shipment operations potentially resulting in accidents are performed. In case of spill, clay components of the above-mentioned materials will significantly mitigate the effects of such an emergency facilitating the elimination of its consequences.

Moreover, considering the existing international experience, the use of complex combined screens is believed to increase IS performance. Such combined screens involve bentomates, clay retainers and geomembranes in various combinations depending on in situ conditions [8].

To increase the efficiency of efforts addressing nuclear legacy challenges, as well as of those associated with construction of new nuclear facilities, it's considered important to analyze and summarize the experience, as well as to provide the integration and availability of knowledge in this area. It should be also noted that application of new materials is not limited only to construction of RW storage and disposal facilities for radioactive waste, including the operations on achieving long-term safe configuration of facilities holding special (non-retrievable) RW (in Russian literature sources the term referred to as "conservation") and nuclear decommissioning. No less relevant may be the use of modern clay-based materials at other nuclear fuel cycle facilities, including operating and planned NPPs, uranium mining enterprises and other nuclear fuel cycle facilities.

Let us dwell on two more examples. Extensive subsoil monitoring efforts are performed at nuclear sites [9]: these are performed both in relevant regions and directly within their territories. Such monitoring revealed that the actual state of affairs in terms of radioactive contamination is far from the ideal one. At the same time, presence or installation of compact sorbing screens at structures potentially causing the contaminated water discharge is not provided for. Spillages or spills of radioactive materials during their management, including their transportation is yet another case in point. This case requires prompt and competent measures to be taken enabling to retrieve the contaminated soils, thus, requiring the development (or availability at best) of an appropriate action plan. The use of bentomates may be seen as an effective solution providing that the radioactive substance, discharged into the external environment without any permit, is contained, protected from water inflow and is well absorbed. The only point required is a comparatively small stock of special bentomate materials available at relevant facilities (at transportation stations) or on the vehicles themselves.

Despite the abovementioned examples demonstrating the effectiveness of clay material application at particular facilities, the overall situation cannot be characterized as an optimal one due to the lack of a unified information resource and absence of tools enabling to share the best practices on the use of clay-based materials as safety barriers in nuclear and other industrial areas as well.

Summarizing the needs for changes set forth in this area, the following priorities should be noted:

- preparation and publication of a comprehensive monograph — a reference book on the subject presented in a modern information format;
- availability of an expert group composed of a few specialists who would monitor the situation on the use of clay materials. This group could be a prototype of an industry-wide competence center.

Regulatory incentives on clay use

This area suggests that regulatory provisions should contain references stating the possibility and necessity of using clays and clay-based materials as effective natural insulation materials. This point is considered below briefly with no account for the hierarchy and not limited to the topic of radioactive waste management. Such overview provides an answer to the question on the availability of and opportunities for introducing these incentives.

Some regulations involve recommendations and/or permitting statements. For example, construction of engineered barriers associated with RW management operations suggests the use of clays ensuring the maximum ability to hold and absorb radionuclides. On the other hand, the use of clays is considered, first of all, as an insulating material, i. e. used in hydraulic structures. A particular case study has showed that these clays should include those from montmorillonite group of minerals, or bentonite clays. However, regulatory provisions include no specific statements on the allowed types or origin of the applied clays and clay-based materials. The range of characteristics and parameters reflecting the changing properties of clays and clay-based materials is literally ignored. It should be noted that even the density of clays varies by several times — from 1.3 to 3.45 g/cm^3. As it comes to the characteristics describing the valuable properties of the material, such as low water permeability, plasticity, swelling capacity, high absorption capacity, resistance to climatic factors these could vary by thousands and tens of thousands of times. No direct reference is provided to the documents reflecting the abovementioned properties, nor the practical guidelines and documents describing the variability of clay-based material properties upon their interaction with isolated substances are available. Montmorillonite content is seen as the main indicator of the bentonite clay quality. Technical specifications on material supply and design documentation developed for the organizations of the State Corporation Rosatom usually either contain no requirements on the content of montmorillonite, or indicate the requirements set forth in the foundry industry standard GOST 28177-98 Bentonite Molding Clays. General Technical Specifications (not less than 30 %).

However, studies of clay-based material focused on their application for radioactive waste disposal purposes conducted all over the world unanimously rely on the montmorillonite content of at least 70 % [10]. Summary of the requirements on clay-based materials applied during design development and implementation of relevant practical efforts is presented in Table 1.

The materials of interest are those presented under the general name of “insulating” [NP-055-04] or “low-permeable” materials, as, for example, provisions of [RB-078-12] indicate that: “materials characterized with low permeability such as clay, loam, construction and artificial materials with a low diffusion coefficient may be used as such”. The larger part of recommendations is reduced to the possibility of using clays as a material enhancing the insulating (filtration and sorption) properties of natural barriers, without taking into account their specific characteristics.

The list of relevant terms is extremely limited and virtually undefined. It involves compacted and (or) pug clay, bentonite-cement, pellets, powders, well-absorbing backfill with different particle sizes, etc. However, neither the clay composition nor their types are not specified which may be required for their practical application. Moreover, regulatory provisions provide no clear straightforward definitions of such terms as “compacted” and (or) “pug” clay, bentonite, nor the references to relevant provisions indicating their quality characteristics.

Quantitative characteristics of the permitted or recommended materials are presented in an extremely simplified form. A case in point, provisions on the establishment of landfills for toxic waste disposal in permeable soils. The document states that a pug clay screen with a hydraulic conductivity of 10^{-9}—10^{-9} cm/s along the bottom and 0.5–0.8 m thick layer slopes should be provided for [9]. The following questions can arise: whether the clay used for this purpose (commonly available in the region) meets the requirements, what kind of reactions occur between clay and toxic solutions to which extent the isolation safety is provided, what are most feasible solutions enabling the construction of such screens, as well as other questions that commonly arise in design development or contractor organizations involved in such projects.

Pure clays are used for rather simple, but large-scale engineering structures — tailing dumps of mining industries, artificial reservoirs, ash and slag dumps of thermal power plants and other hydraulic structures.

Many documents contain no provisions on the opportunities for using clay-based materials, although their application, taking into account specific features of the facilities developed seems to be quite feasible. Moreover, most of the documents are based on a simplified principle of “reasonable sufficiency” with no opportunities for introducing excessive/additional elements, including the construction of engineered barriers. We believe that in the design development of nuclear enterprises and facilities [11], including relevant safety arrangements based on the joint use of geological and engineering barriers, it’s quite reasonable to emphasize the importance of these barriers and clay-based material properties with due account of the specific features of such enterprises. References to relevant regulatory documents, including guidelines and recommendations on the comprehensive use of clay as a structure component material for of these barriers should be also provided.

Table 1. Practical application of the requirements on clay materials

Indicator/conditions	Unit	Function/application							
		Backfilling of inter-container space in deep RW disposal facility (RW DDF)			Clay screen in deep RW disposal facility (RW DDF)		Barrier construction		LRW cementation
Material, purpose		Mud powder in Novouralsk RWDDF	Bentonite molding clays, P1T2 in Ozersk RW DDF	Mud powder in Seversk RWDDF	Ozersk RW DDF	Seversk RWDDF	Bentonite clay PBMG	Mud powder for maintenance activities at KhTO-2	Bentonite clay of SK mark. Cementation
Author of the requirement		NO RAO operating organization	Design development organization – Ural branch of JSC FCNIVT SRPA "Eleron" – UPII VNIPIET	Design development organization JSC CPTI	Design development organization – Ural branch of JSC FCNIVT SRPA "Eleron" – UPII VNIPIET	Design development organization JSC CPTI	FSUE Radon		JSC Concern Rosenergoatom, Novovoronezh NPP
Regulations on the material			GOST 28177-98 Bentonite Molding Clays. General Technical Specifications »				TU 39-0147001-105-93 or TU 2164-005-04002160-2007		GOST 7032-75 Bentonite clay for fine and building ceramics»
Technical specifications for the material									
Mass fraction of montmorillonite	%	20–30	No less than 30		No less than 30				
Mass fraction of kaolinite	%	20–40							
Total content of clay materials	%		No less than 75		No less than 50				
Concentration of exchange cations	mg-eq/100 g dry clay	No less than 25	No less than 30						
Bentonite number (swelling capacity)	ml								No less than 75
Adsorption rate	mg/g								No less than 150
Mass fraction of carbonates in terms of $CaCO_3$,%, not more than	%		No more than 10						
Mass fraction of sulfide sulfur	%		No more than 0.3						
Mass fraction of iron in terms of Fe_2O_3,%	%		No more than 12.0						
Total iron and titanium oxides content (Fe_2O_3-fTiO_2)	%								No more than 2.25
Sulfuric Anhydride (SO_3) Content	%								No more than 0.75

Continue of Table 1

Indicator/conditions		Unit	Function/application							
			Backfilling of inter-container space in deep RW disposal facility (RW DDF)			Clay screen in deep RW disposal facility (RW DDF)		Barrier construction		LRW cementation
Colloidity		%	No less than 10	No less than 10						
Water absorption		pt.	No less than 1.5	No less than 1.5						
Compressing strength		Pa (kgf/cm^2)		No less than $8.826 \cdot 10^4$ (0.9)						
Tensile strength in the zone of moisture condensation		Pa (kgf/cm^2)		No less than $0.275 \cdot 10^4$ (0.028)						
Thermal resistance		pt		No less than 0.3						
Average flexural strength (binding power)		kg/cm^2								No less than 20
Oversize product	0.1 mm	%	No more than 30		Mud powder					
	0.16 mm			No more than 10						
	0.4 mm			No more than 3						
	0.5 mm		No more than 20							
	1.0 mm		No more than 15							
	5.0 mm		No more than 3							
Undersize	0.1 mm	%	No less than 35							
Content of pieces by size	50 mm	%								No more than 10
	300 mm									No more than 10
Sand content		%					No more than 10–15		No more than 5	
Hydraulic conductivity		m/day	No less than 10^{-5}			No more than 10^{-5}	No more than 10^{-5}			
Index of plasticity						No less than 20	No less than 27			
Moisture content		%	3.0–5.0	6.0–10.0				No more than 13	No more than 10	No more than 20
The output of the clay solution with a viscosity of 20 MPa		m^3/t						No less than 8	No less than 8	
Suspension density		g/cm^3						1.079–1.053	1.079–1.032	
Filtration rate		cm^3						No more than 15	No more than 15	
Bulk density		t/m^3	0.9–1.2						0.9–1.0	
Treating agent								Na_2CO_3		

Thus, the regulatory framework contains rather simplified and conservative approach to the recommendations on materials, which, in fact, should be recognized as ineffective. Current recommendations assume the reproduction of this approach and the use of the same materials that were used decades ago. At the same time, it's worth emphasizing that release of radioactive substances was identified at all facilities built 50 or more years ago. Sub-soil monitoring, development of corrective measures and relevant engineering solutions, including comprehensive R&Ds and other similar actions are required. It should be noted that many of the cases discussed refer exclusively to the effects associated with the possibility of detecting low, ultra-low or background concentrations of radioactive substances. But regardless of this, there is an obvious need and possibility for providing more specific references to more up-to-date materials, including the clay-based ones. The first reasonable step seems to be the expansion of clay use applications by introducing relevant amendments to the regulatory framework with due account of the specific characteristics of clays, their properties when interacting with isolated substances and barrier construction materials, design purpose of facilities, goals and objectives.

Documentary support for design activities

The use of clay-based materials in relevant design solutions is considered to be impossible without a large amount of reference materials presented in the form of new and updated construction norms and rules, GOSTs, various kinds of pricing and estimated data. Recent experience along with already mentioned purpose-developed reference guides will enable to shape the initial form of such documentation base. Introduction of such regulatory incentives will promote its continuous expansion and upgrading. The documentation base should involve data and knowledge bases presented in an up-to-date format ensuring the efficiency of its application.

Stimulation of best practices and special quality control (conformity assessment)

Availability of various classification guides on clay-based materials developed with due account of nuclear industry needs will enhance relevant conformity assessment potential accumulated by the industry. In this case, development of specific requirements to particular operations can be based on a list of materials with known properties and characteristics, rather than referring to raw materials for drilling fluids (PBMG bentonite clay, TU 39-0147001-105-93), molding clay for the foundry industry (GOST 28177-98), clay for building ceramics (GOST 7032-75) and grounded refractory clay used in construction of insulating screens and backfilling of void spaces in RW storages as it is done today in the same way as it was done some thirty years ago.

Special and expanded requirements in place will enable to fine-tune the conformity assessment and quality control system. Ideally, as manufacturing of well-known modern clay-based materials is a rather complicated technological process and proper use of these materials requires particular know-how and certain skills, demonstration centers should be established.

Conclusion

The proposed set of measures suggests a large amount of scientific, regulatory and managerial efforts stretched in time, and, thus, with no immediate result achieved to be implemented. However, in some cases, it allows to avoid mistakes that could've resulted in some negative effects in some 5—10 years, including significant radiation and economic detriment. Moreover, the expected effect will clearly reveal itself in terms of ensuring the long-term safety and its justification, also covering RW management issues. It should be noted that clay is considered as a promising bedrock option in the construction of geological disposal facilities for RW [12]. Based on the study of the terms of references on the development of DDF RW, one can conclude that the decisions regarding the selection of particular materials are going to be quite shallow with no in-depth analysis to be applied.

The use of special clay materials in nuclear industry presents a quite promising, but so far poorly studied area with scientific and practical efforts aimed at development of new purpose-designed clay-based materials, such as: cement-clay materials, zeolite-containing clays, treatment technologies for clays and their blends, manufacturing mixtures composed of clay-based materials with specified properties and characteristics, manufacturing compacted pellets, briquettes and other compacted bentonite products.

References

1. Krupskaya V. V., Biryukov D. V., Belousov P. E., Lekhov V. A., Romanchuk A. Yu., Kalmykov S. N. The use of natural clay materials to increase the nuclear and radiation safety level of nuclear legacy facilities. *Radioactive Waste,* 2018, no. 2 (3), pp. 30—43. (In Russian).

2. *Problemy yadernogo naslediya i puti ih resheniya.* Ed. by A. M. Agapova, L. A. Bol'shova, E. V. Evstratova, N. P. Laverova, I. I. Linge. Moscow, "Ehnergopromanalitika" Publ., 2012. Vol. 1. 356 p.

3. Zaharova E. V., Kozyrev A. S., Zubkov A. A., Aver'yanov B. Yu. Sozdanie vneshnih bar'erov bezopasnosti kak sposob predotvrashcheniya migracii radionuklidov iz hranilishch RAO. *Nuclear and Radiation Safety of Russia,* Moscow, Ehnergopromanalitika Publ., 2012, issue 13, pp. 133—139.

4. Aleksahin R. M., Buldakov L. A., Gubanov V. A. et al. *Krupnye radiacionnye avarii: posledstviya i zashchitnye mery.* Ed. by L. A. Il'in and V. A. Gubanov. Moscow, IzdAT Publ., 2001. 752 p.

5. Savonenkov V. G., Anderson E. B., Shabalev S. I. *Gliny kak geologicheskaya sreda dlya izolyacii radioaktivnyh othodov.* SPb: Info Ol, 2012. 215 p.

6. Rumynin V. G., Nikulenkov A. M. Tendencies in the change in physical and mechanical properties of the vendian clay formation (North Western edge of the Russian platform). *Journal of Mining Institute,* 2012, vol. 197, pp. 191—196. (In Russian).

7. *Rossijskaya konferenciya "Fundamental'nye aspekty bezopasnogo zahoroneniya RAO v geologicheskih formaciyah"*: Moscow, October 15—16 2013. Moscow, "Granica" Publ., 2013. 158 p. ISBN 978-5-94691-585-4.

8. Cebakovskaya N. S., Utkin S. S., Ivanov A. Yu., Saharov V. K., Polunin K. E. *The best foreign practice of decommissioning nuclear installations and rehabilitation of contaminated areas*: Vol. 1. Under the general editorship of I. I. Linge and A. A. Abramov. Moscow, IBRAE RAN Publ., 2017. 336 p. (in Russian). ISBN 978-5-9907220-6-4.

9. SNiP 2.01.28-85. *Poligony po obezvrezhivaniyu i zahoroneniyu toksichnyh promyshlennyh othodov. Osnovnye polozheniya po proektirovaniyu.* Moscow, CITP Gosstroya SSSR, 1985. 16 p.

10. *Design, production and initial state of the buffer.* SKB 2010. Svensk Kärnbränslehantering AB. Technical Report, TR-10-15. 2010. 89 p. Available at : http://www.skb.com/publication/2151517/TR-10-15.pdf (20.09.2017).

11. SanPiN 2.6.1.07-03. *Gigienicheskie trebovaniya k proektirovaniyu predpriyatij i ustanovok atomnoj promyshlennosti.*

12. Trudy Radievogo instituta im. V. G. Hlopina, v. XI, 2006. — 130 p.

Information about the authors

Linge Igor Innokentevich, Doctor of Technical Sciences, Deputy Director, The Nuclear Safety Institute of the Russian Academy of Sciences (52, Bolshaya Tulskaya St., Moscow, 115191, Russia), e-mail: linge@ibrae.ac.ru.

Ivanov Artem Yurievich, Head of Department for Information Support of Nuclear and Radiation Safety Programs, The Nuclear Safety Institute of the Russian Academy of Sciences (52, Bolshaya Tulskaya Str., Moscow, Russia, 115191, Russia), e-mail: aivanov@ibrae.ac.ru.

Kazakov Konstantin Sergeevich, Deputy chief of NRS programs development and analytical support Laboratory, The Nuclear Safety Institute of the Russian Academy of Sciences (52, Bolshaya Tulskaya Str., Moscow, Russia, 115191, Russia), e-mail: kks@ibrae.ac.ru.

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