UDC [622.831.325:532.528].003.1 DOI: https://doi.org/10.15407/geotm2025.175.014

CONTAINERIZATION OF RADIATION-HAZARDOUS WASTE FOR PROTECTION AGAINST GAMMA RADIATION: POSSIBILITIES ASSESSMENT AND RESEARCH PROSPECTS

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Abstract. The article provides a comprehensive analysis of modern approaches to the storage of low-level radioactive waste using protective containers that ensure reliability, durability and environmental safety. The basic principles of containerization are considered, in particular the concept of multi-barrier protection, according to which the isolation of radioactive materials should be guaranteed by a combination of natural and man-made barriers, including the physical and chemical form of waste, a hermetic casing and protective coatings. The modern classification of materials used for the manufacture of containers, their advantages, disadvantages and factors limiting the efficiency of operation under long-term storage conditions are analyzed.

A new approach to the creation of structural materials based on compositions with the introduction of rare-earth elements is proposed, which allows significantly improving their physical, mechanical and radiation-protective properties. It is shown that the use of ultrafine particles of a metal filler in the matrix structure provides effective attenuation of gamma radiation due to a combination of the processes of multiple reflection, scattering and absorption of quanta in the metal, as well as ohmic losses caused by the excited electromagnetic field. The dependence of the lead equivalent on the thickness of the sample with different contents of rare-earth components was experimentally confirmed, which indicates an increase in the effective thickness of the protective layer without an increase in the mass of the product.

Based on the results obtained, a new design of a container for the storage and transportation of radiation-hazardous waste was developed, which is characterized by the unification of constituent elements, the use of a material with increased protective properties, low cost and high manufacturability. The proposed container is environmentally friendly, has an increased service life, and after the end of its service life it can be recycled or reused as a secondary raw material for the production of new composite materials.

Keywords: container, radiation-hazardous waste, storage, gamma radiation, rare-earth elements.

1. Introduction

Recently, many countries around the world have seen an increase in the share of nuclear generation in the energy supply structure. It currently provides approximately 17% of total needs. At the same time, the issue of radiation-hazardous waste management generated during the operation of nuclear power plants is becoming particularly relevant. For example, in Ukraine an average of 0.15 m³ to 0.35 m³ of liquid and 0.1 m³ to 0.3 m³ of solid radioactive waste is generated per megawatt of electricity every year [1,2].

The problem of radiation-hazardous waste management is complex, multifaceted and requires an integrated approach. In Ukraine, at the present stage, there is a complication of the radiation and ecological situation, which is due to the accumulation of significant volumes of radioactive waste generated as a result of the activities of nuclear fuel cycle enterprises, the defense industry, as well as the use of radioactive substances in medicine, science, agriculture and other areas. Reliable processing, safe storage and effective isolation of such materials, taking into account the specifics of radionuclides, their physicochemical and biological properties, are of crucial importance for environmental protection [3].

In international practice, radiation-hazardous waste is disposed with taking into account its radioactivity. In particular, low- and medium-level waste with short-lived

Received: 04.02.2025 Accepted: 15.12.2025 Available online: 19.12.2025

radionuclides (half-life up to 30 years) is usually isolated in special containers placed in near-surface repositories. On the other hand, high-level waste with long-lived radionuclides (half-life of more than 30 years) is subject to long-term storage in deep geological formations. Containerization is a key element in ensuring radiation safety at all stages of waste management – from initial collection, diagnostics, transportation, sorting, storage and processing to final disposal in an acceptable state.

One of the promising directions for solving this problem is the development of highly efficient waste isolation systems using modern materials characterized by increased radiation-protective properties. At the same time, it is necessary to form a conceptual basis for the containerization of radiation-hazardous waste at the phenomenological level, which will make possible to substantiate and implement effective technological strategies for countering the risks associated with their long-term storage.

2. Methods

Analysis of scientific sources indicates the presence of several most appropriate directions for improving technologies for long-term and safe storage of radiation-hazardous waste, which are based on the concept of using a system of natural and artificial barriers. Natural barriers include geological formations within which the repository is located, which ensure the restriction of radionuclide migration by reducing their mobility in potential transfer routes to the biosphere. Artificial barriers include the conversion of radiation-hazardous waste into stable physicochemical forms with low leaching and dispersion ability, the use of containers, as well as the introduction of additional engineering protective structures as part of the repository, including special geochemical barriers. This approach contributes to increasing the level of environmental and radiation safety during the storage of waste with a long period of danger.

Analysis of the regulatory documents governing hazardous waste management indicates the presence of contradictory provisions and the need of their further improvement [4]. In particular, the current classification of radiation-hazardous waste by activity level does not contain clear instructions on the required periods of its isolation. Most of such waste is currently stored in temporary storages. One of the main reasons for the accumulation of significant amounts of radioactive materials in storages is the inefficiency of the current system for their management. Today, the approach is practiced, according to which all waste is subject to storage for 30–50 years with the possibility of extending the life of the storages. Such a strategy does not provide a final solution to the problem of safe isolation of radiation-hazardous waste, and requires significant financial funds and resources for maintaining the infrastructure without a clearly defined perspective for the waste liquidation. With this approach, making a final decision on the further status of this waste is actually transferred to future generations [5].

Today, a number of technologies for immobilization of radiation-hazardous waste have been developed, and various approaches to their safe storage are also actively investigated. The main criteria for choosing appropriate solutions are minimizing financial costs for the implementation of measures for radiation-hazardous waste management and reducing the volume of secondary radioactive materials. The method of containerization of radioactive waste meets these requirements most fully.

In this regard, the purpose of this study is to summarize modern experience in container storage of radioactive materials, to analyze scientific approaches to creating composite materials with increased radiation-protective properties, and to improve the design of containers that would combine a high level of radiation protection, low cost, environmental safety, and the possibility of their further disposal.

3. Theoretical part

Containers play a key role in improving the safety of hazardous waste at all stages of their handling: from collecting untreated waste to its disposal in a suitable way [6]. The use of containerization has a number of significant advantages [6], including:

- increasing safety during collection, transportation, long-term storage of waste;
- the possibility of control during temporary storage and, if necessary, elimination of emergency situations;
- possibility of further processing or re-burial of radiation-hazardous waste;
- simplification and unification of engineering and technical decisions;
- use of typical hoisting machines and mechanisms;

Additionally, the protective container has a restricted service time that takes into account the half-life of radioactive isotopes contained in the radioactive waste intended for storage. In this case, the container provides protection against ionizing radiation, so that the power of the equivalent dose from any outer surface of the container at a distance of 1 m does not exceed 0.5 mSv per year. Moreover, the container provides reliable isolation of the radiation-hazardous waste contained therein from the environment, and withstands the action of a column of water at a height of at least 3 m with the probable flood without leakage [7].

Depending on the frame material, there are metal, reinforced concrete, steel concrete and steel-reinforced concrete containers (Fig. 1).

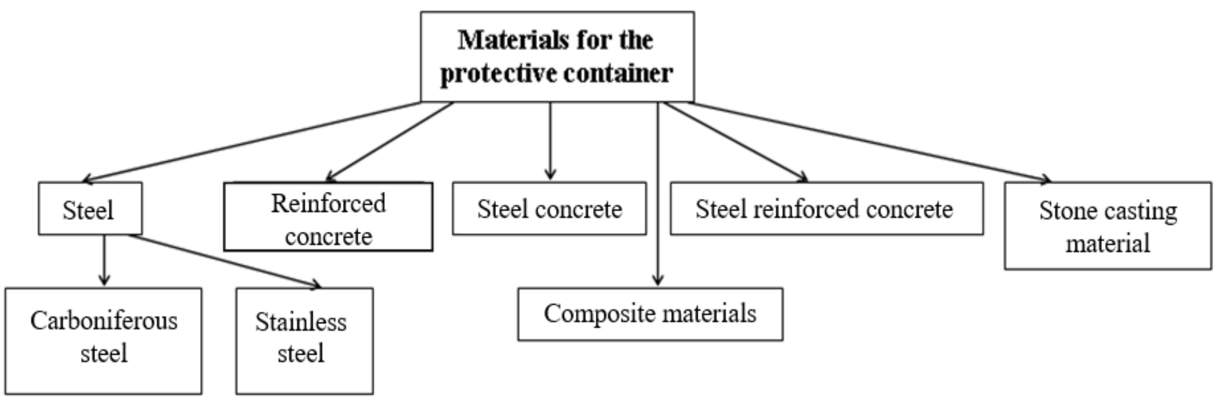


Figure 1 – Modern classification of materials for the protective containers manufacturing in order to keep radiation-hazardous waste

Given the variety of container designs used for storage and disposal of radiation-hazardous waste, the choice of the optimal option requires taking into account not only the physical-mechanical and protective properties of the materials, but also technological and operational factors. Reinforced concrete, steel and stone casting containers have their own unique advantages and limitations, which can significantly affect the safety, durability and cost of implementing projects in the field of waste management. To systematize these characteristics, a comparative analysis of the main types of containers was carried out, the results of which are given in Table 1 [7-9].

Table 1 – Comparative analysis of data on reinforced concrete, steel and stone casting containers for radiation-hazardous waste storage

Container type	Advantages	Disadvantages
Reinforced concrete	- high protective properties against ionizing radiation; - resistance to external factors (temperature, humidity); - relatively low manufacturing cost; - long service life (up to 300 years).	- high labor intensity of production and the need for formwork; - insufficient crack resistance; - low resistance to mechanical damage; - in prefabricated structures - the problem of sealing joints, which require a large number of embedded parts.
Steel	- high strength and manufacturing accuracy; - possibility of mass production and reduction of dimensions with the same strength; - ease installation and dismantling.	- possible loss of local stability of thin walls under concentrated loads; - low fire resistance; - susceptibility to corrosion; - higher cost compared to reinforced concrete.
Stone casting	- high density and homogeneity of the material; - resistance to chemical corrosion and atmospheric influences; - high crack resistance; - good compatibility with other materials.	- large mass of structures; - complexity of casting technology, need for specialized equipment; - relatively high cost of production; - limited experience of application in the field of storage of radiation-hazardous waste.

A comparative analysis of the containers for the storage and disposal of radiation-hazardous waste shows that each type has a specific set of operational characteristics, determined by the physical, mechanical and technological properties of the materials. Reinforced concrete containers remain the most common solution due to the combination of high radiation-protective properties, durability and relatively low cost. At the same time, their use is limited by the complexity of manufacturing and insufficient crack resistance, which can affect tightness during long-term storage. Steel containers demonstrate advantages in manufacturing accuracy and compactness of structures, but their effectiveness is reduced by high cost, susceptibility to corrosion and low fire resistance. Stone-cast containers are a promising option due to their high density, resistance to chemical corrosion and the possibility of creating frameless forms, but they require additional technological development, in particular, reducing mass and optimizing casting processes. Thus, the choice of the optimal type of container should be based on a balance between the requirements of radiation protection, operational reliability, and economic feasibility, taking into account the conditions of a specific facility for storing or disposing of radiation-hazardous waste.

The conceptual principles of radiation-hazardous waste containerization are based on the principle of multi-barrier protection, according to which isolation should be provided by a system of engineering and natural barriers, including considering the physical and chemical form of waste, and a corrosion-resistant container. Therefore, the development of radiation-protective materials for creating such a container for the purpose of immobilizing radionuclides should be aimed at creating matrices that must meet the following basic requirements [9]:

- bind and retain as many radionuclides and their fission products as possible (waste minimization), as well as other waste components, for a long (according to geological scales) time;
- be resistant to the processes of physical and chemical interaction under conditions of long-term storage and disposal;
- have a complex of physical and mechanical properties to ensure transportation, long-term storage and disposal.

Along with these requirements, one of the requirements for the method of obtaining matrices is its efficiency. To do this, it is necessary to determine the optimal parameters for matrix synthesis that will ensure their production with the specified properties at the lowest energy consumption.

4. Results and discussion

In order to determine the key parameters of containers to be developed there were conducted some investigations related to creation of a gypsum-based composite material with enhanced radiation-protective characteristics. To do this, particles of a polydisperse mixture of rare earth elements were introduced into the gypsum matrix, followed by their fixation by forming strong phase contacts.

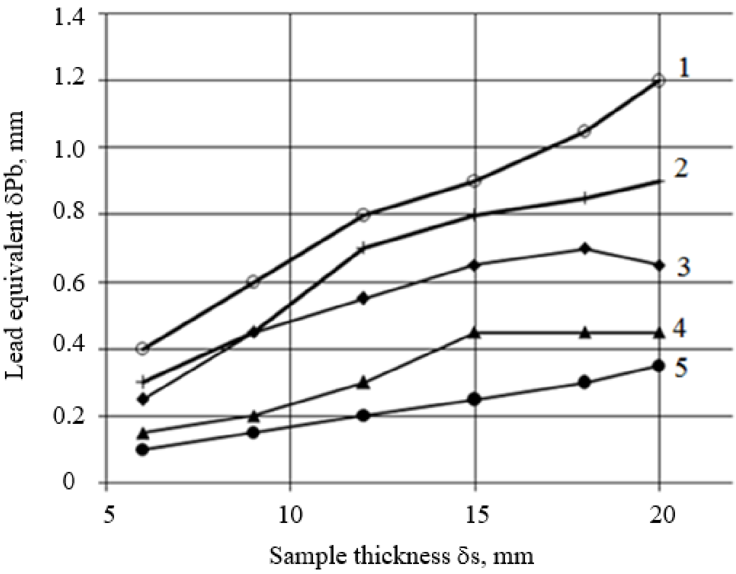
The obtained results confirmed that the development of highly effective radiation-protective materials is possible on the basis of polydisperse systems, which include ultradisperse particles (UDP) with a size of less than 100 nm. The specific properties of such systems are due to the special nature of atomic and electronic states. With the optimal selection of the matrix material and the filler converted into an ultradisperse state, some conditions are created for the transfer of electrons from the UDP to the matrix material, which causes the redistribution of charges and the formation of local electric fields of high intensity. As a result of such interaction, the particles of the polydisperse mixture are able to self-organize into energetically coherent ensembles that effectively deflect ionizing radiation. This leads to an increase in their path length in the material, which is equivalent to an increase in the effective thickness of the protective layer.

As one of the components of the UDP, metals can be used, in particular tungsten, cerium, barium or zinc, as well as intermetallic compounds, oxides, carbides, nitrides, borides, hydrides. The optimal mass ratio of the matrix and the polydisperse mixture, provided that they are thoroughly mixed, provides physicochemical activation, which is accompanied by the avalanche-like formation of a system of energetically consistent ensembles. Their characteristics can be changed by adjusting the degree of dispersion, segregation processes and spatial distribution of particles in the matrix.

To achieve maximum radiation protection efficiency of composite materials based on polydisperse powders with ultrafine fraction, it is necessary to control not only the granulometric composition of the filler, but also the formation of its optimal structure. This makes possible to prevent the occurrence of inhomogeneities caused by coagulation and aggregation of particles, which, otherwise, leads to a decrease in the protective properties of the material [10].

The research methodology was of a computational-experimental nature and was based on a comparative analysis of the radiation-protective characteristics of the composite material and its individual components [8]. The experimentally obtained values of the protective parameters of samples with a given volumetric content of the filler were compared with the calculated values determined on the basis of the characteristics of the material components, selected in proportions corresponding to the composition of the experimental samples.

Figure 3 shows the dependences that reflect the change in lead equivalent on the thickness of the sample for different percentages of rare-earth elements.



1– 39 %; 2 – 31 %; 3 – 26 %; 4 – 20 %; 5 – 9 %

Figure 3 – Dependence of the change in lead equivalent (δPb) on the sample thickness (δs) at different percentages of rare earth elements

Analysis of the obtained dependences shows that the value of the lead equivalent increases with increasing sample thickness, which is natural for radiation-protective materials. At the same time, a significant influence of the volume fraction of rare-earth elements is observed: an increase in their content in the composition of the polydisperse material leads to an increase in the lead equivalent at a fixed sample thickness. The most pronounced effect of enhancing the protective properties is manifested in samples with the maximum percentage of rare-earth impurities, which confirms the feasibility of their use as effective matrix fillers.

The effects revealed during experimental studies are primarily due to different mechanisms of interaction of gamma quanta with the protective material, which manifest themselves depending on the radiation energy and the content of finely dispersed metal filler. The qualitative features of these processes differ from each other, while their quantitative characteristic is the effective atomic number of the polycomponent material.

In addition, the energy losses of gamma radiation are explained by its quantum-dualistic nature. If we consider radiation as a stream of quanta, then the absorption of energy in metal-saturated gypsum occurs due to multiple reflections, scattering and absorption, which actually lengthens the trajectory of their penetration. In the wave interpretation, the decrease in the radiation intensity in the conductive matrix is due to the excitation of the opposing electromagnetic field and the occurrence of ohmic losses.

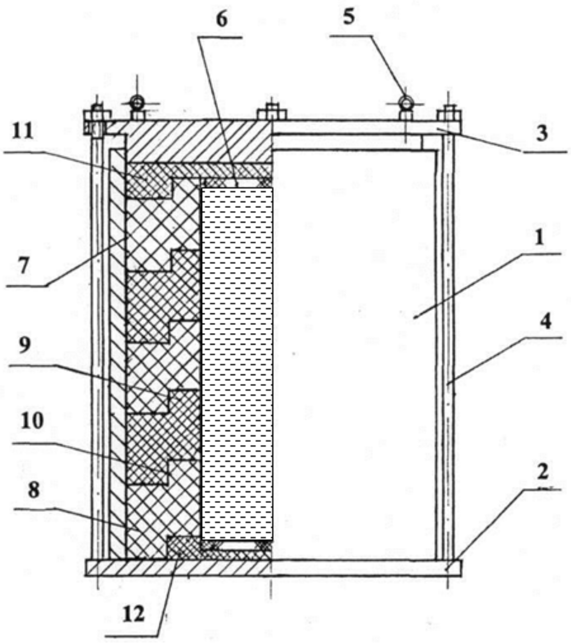
The conducted studies confirmed the increase in the effective cross section of the interaction of gamma radiation with the composite radiation-protective material due to an increase in the concentration of ultrafine filler particles, in particular rare earth elements.

According to the analysis of the possibilities of containerization of radiation-hazardous waste, it was found that such containers must necessarily have a number of properties - resistance to chemical agents, durability, tightness and mechanical strength, which depend on the complexity of the design, manufacturing technology and cost of the materials used, as well as the ability to recycle. This became a prerequisite for the development of an improved container design for long-term storage and transportation of radiation-hazardous waste [11].

The advantage of the proposed container is the unification of its components with the use of a new composite material containing organosilicon compounds with a rare-earth filler. This material is characterized by increased radiation-protective properties, differs from known analogues by its lower cost, high operational reliability and environmental safety, and also provides the possibility of further disposal.

Fig. 4 shows a cross-section of the container along its axis.

As one of the variants of the example of using the model, a container in the form of a cylindrical metal container body 1 with a rigid lower support bottom 2 and an top cover 3, fastened together by tie bolts 4, is proposed. Eye bolts 5 are placed on the cover 3 for transporting the container. Inside the container body 1, a container 6 with radiation-hazardous waste is placed, which is separated from the wall of the container body 1 by a replaceable unified radiation-protective shell 7, which is made in the form of assembly sleeves 8 by the pressing method from a material containing organosilicon compounds with a rare-earth filler, which makes it possible to significantly reduce the thickness of the shell, ensuring its high radiation-protective properties, which leads to a reduction in the weight of the structure itself and its cost. In addition, this makes it possible to make the shell 7 with different radiation-protective properties, which contributes to its unification during storage and transportation of radiation-hazardous waste. In this case, the end surface of each sleeve 8 is provided on one side with a protrusion 9, and on the other with an internal groove 10, for interaction with the protrusion of the adjacent sleeve, creating a single design of the radiation-protective shell 7, which allows it to be unified during storage and transportation of radiation-hazardous waste and ensures the versatility of the container design.



1 – container body, 2 – support bottom, 3 – top cover, 4 – tie bolts, 5 – eye bolts,
6 – container with radiation-hazardous waste, 7 – radiation-protective shell, 8 – assembly sleeves,
9 – sleeve protrusion, 10 – internal groove in the sleeve, 11, 12 – rubber seals

Figure 4 – Scheme of a protective container for storing radiation-hazardous waste

For hermetic fastening of the container body 1, radiation-protective rubber seals 11 and 12 are used, which are installed under the top cover 3 and on the support bottom 2, respectively, which makes it possible to obtain a hermetic structure of the container and contributes to increasing the reliability of its design as a whole.

Before closing the cover 3, the tightness of its installation and conformity on all surfaces should be controlled. The claimed design allows preventing the occurrence of any gap.

After the end of the storage period of radiation-hazardous waste, the container 6 can be recycled or used as a secondary filler thanks to the use of a composite material based on chemically stable polyester resin.

Thus, the use of the proposed container simplifies the design due to unified individual components with the use of an innovative material with increased radiation-protective properties. This contributes to increasing the reliability, environmental safety and durability of the container, and also creates opportunities for its environmentally acceptable recycling after the end of its service life.

5. Conclusions

1. It was established that the majority of radiation-hazardous waste is located in temporary storage facilities. The reason for the accumulation of large volumes of radiation-hazardous waste in storage facilities is an inefficient approach to their management. It is currently accepted that all generated waste should be stored for 30-50 years with the possibility of extending the storage period. This approach does not ultimately lead to a safe problem solution and requires significant costs for the operation of storage facilities without a clear prospect of the waste liquidation. The final solution of managing the radiation-hazardous waste accumulated in this way is entrusted to future generations.

2. Containers play a key role in improving the safety of the radiation-hazardous waste management system, namely, from the collection of untreated waste to its burial in a conditioned form. A comparative analysis of the containers designs for the storage and burial of radiation-hazardous waste shows that each type has a specific set of operational characteristics, determined by the physical, mechanical and technological properties of the materials.

3. The obtained results confirmed that the use of polydisperse systems with ultradisperse particles less than 100 nm allows to form an energy-coordinated ensembles capable of effectively deflecting ionizing radiation, which ensures an increase in the radiation-protective properties of the material due to an equivalent increase in the thickness of the protective layer. The absorption of gamma radiation energy in metal-saturated gypsum is explained both by multiple reflections, scattering and absorption of quanta by the metal (in the pseudoparticle model), and by the excited electromagnetic field of counteraction due to ohmic losses (in the wave model).

4. The obtained results show that the content of rare-earth elements significantly affects the radiation-protective characteristics of the material, which is confirmed by the change in the lead equivalent depending on the thickness of the sample. Thus, the analysis of the obtained dependencies show that the value of the lead equivalent increases with increasing sample thickness, which is natural for radiation-protective materials. At the same time, a significant effect of the volume fraction of rare-earth elements is observed: an increase in their content in the composition of the composite material leads to an increase in the lead equivalent at a fixed sample thickness. The most pronounced effect of enhancing protective properties is manifested in samples with the maximum percentage of rare-earth impurities, which confirms the feasibility of their use as effective matrix fillers.

5. An improved container design for long-term storage and transportation of radiation-hazardous waste was developed, which is distinguished by the unification of its individual components using a new material containing organosilicon compounds with rare-earth filler and has increased radiation-protective properties, low cost, high reliability and environmental friendliness with the possibility of its further disposal.

Conflict of interest

Authors state no conflict of interest.

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КОНТЕЙНЕРИЗАЦІЯ РАДІАЦІЙНО-НЕБЕЗПЕЧНИХ ВІДХОДІВ ДЛЯ ЗАХИСТУ ВІД ГАММА-ВИПРОМІНЮВАННЯ: ОЦІНКА МОЖЛИВОСТЕЙ ТА ПЕРСПЕКТИВИ ДОСЛІДЖЕНЬ

Клюєв Е., Возіянов В., Сапунова І., Мірзаєва Е.

Анотація. У статті проведено комплексний аналіз сучасних підходів до зберігання радіаційно небезпечних відходів низької активності із застосуванням спеціалізованих контейнерів, що забезпечують надійність, довговічність і екологічну безпечність процесу. Розглянуто основні принципи контейнеризації, зокрема концепцію мульти-бар'єрного захисту, згідно з якою ізоляція радіоактивних матеріалів має гарантуватися поєднанням природних і техногенних бар'єрів, включно з фізико-хімічною формою відходів, герметичним корпусом і захисними покриттями. Проаналізовано сучасну класифікацію матеріалів, що використовуються для виготовлення контейнерів, їх переваги, недоліки та чинники, які обмежують ефективність експлуатації в умовах тривалого зберігання.

Запропоновано новий підхід до створення конструкційних матеріалів на основі композицій із введенням рідкісноземельних елементів, що дозволяє суттєво підвищити їхні фізико-механічні та радіаційно-захисні властивості. Показано, що використання ультрадисперсних частинок металевого наповнювача в структурі гіпсової матриці забезпечує ефективне ослаблення гамма-випромінювання завдяки поєднанню процесів багаторазового відбивання, розсіювання та поглинання квантів у металі, а також через омічні втрати, викликані збудженим електромагнітним полем. Експериментально підтверджено залежність свинцевого еквівалента від товщини зразка при різному вмісті рідкісноземельних компонентів, що свідчить про збільшення ефективної товщини захисного шару без зростання маси виробу.

На основі отриманих результатів розроблено нову конструкцію контейнера для зберігання та транспортування радіаційно небезпечних відходів, яка характеризується уніфікацією складових елементів, використанням матеріалу з підвищеними захисними властивостями, низькою собівартістю та високою технологічністю. Запропонований контейнер є екологічно безпечним, має збільшений термін експлуатації, а після завершення строку використання може бути утилізований або повторно застосований як вторинна сировина для виготовлення нових композиційних матеріалів.

Ключові слова: контейнер, радіаційно небезпечні відходи, зберігання, гамма-випромінювання, рідкісноземельні елементи.