

Application of Polymers for the Emergency Damage Control and Remediation at Nuclear and Chemical Industry Sites – 15259

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ABSTRACT

From the standpoint of possible accidents and the following complex emergency damage control the nuclear and chemical industry sites are of the primary concern. Among the possible causes of accidents and emergency situations are technological and waste solution spills. Polymers can be used for this problem resolution and the spilled solutions could be effectively isolated. The apparent advantage of polymer materials is the possibility of solidifying solutions with rather high acidity and high salt bearing.

To date the situations with the RW storage tank decommissioning in Russia and in the United States have much in common: the lack of the waste management strategy developed at the initial stage of production, the end of the storage tank lifetime. Development of the *in-situ* solidification technique for the mobile (liquid) components of heterogeneous waste will provide a good option to contain the radio nuclides within the storage site boundaries.

The presented work was aimed at the search for the simple engineering decisions that would provide the waste immobilization in case of the accidental liquid spills. The application of polymers for the purpose indicated would allow in case of emergency to reduce the radioactive aerosol concentration in the air and prevent the distribution of aerosols on the premises and in the environment. Another promising area of the polymer application could be the technical challenges associated with the decommissioning of NPPs and other objects and facilities including the large volume LRW storage tanks.

INTRODUCTION

Industrialization of the modern society and the ever increasing sophistication of the production technology inevitably lead to the negative developments that occur in the emergencies and under extraordinary conditions. And from the standpoint of possible accidents and the following complex emergency damage control the nuclear and chemical industry sites are of the primary concern.

Among the possible causes of accidents and emergency situations are technological and waste solution spills. The resulting radioactive contamination area causes the apparent possibility of the biologically hazardous component contact with the environment.

If an option to collect the spilled solution into waste tanks is not available, the primary task is to contain the solution followed by the conversion thereof into a stable form thus reducing the potential risks of the spread radioactive contamination.

Materials for the localization of spilled solutions should be chosen with regard to the composition and specific activity of radionuclides, their chemical speciation and physical

properties. Those materials should be relatively easy in the subsequent processing, and the application thereof should not lead to a significant increase in the final waste product volume as compared to the volume of original waste. When choosing the material, its commercial availability and cost should be taken into account, too.

Among the other requirements one may indicate the resistance to the environmental impact (chemical, biological factors) as well as the thermal and radiation stability, the storage quality. The cementation is the most widely spread technique for radioactive waste conditioning [1, 2]. This method is economically efficient for immobilizing various waste types into the safe solid waste form suitable for the long-term storage.

Shortcoming of cementation consists in material incompatibility with solutions of acids. It is worth to mention that, in the case of spills of organic liquids using cement impossible so Portland cement alone is not effective for solidification of any organic liquid wastes [3]. Polymers can be used for this problem resolution [4] and we believe the spilled solutions could be effectively isolated and the obtained solid products successfully contained with the aid of the polymer materials manufactured in industrial scale by the NOCHAR Company (U.S.) [5-16]. The principal advantages of liquid wastes processing through their immobilization within a polymer matrix are as follows:

1. First, the apparent advantage of polymer materials is the possibility of solidifying solutions with rather high acidity and high salt bearing. It is an option for localization of waste of practically any composition including organic liquids and mixtures of aqueous and organic solutions.

2. Possibilities to perform the process of waste immobilization directly in containers, resulting products remaining in containers for further storage.

It is worth noting that the application of polymers provides a possibility to reduce the solidified waste volume coming to the repository. The waste volume reduction factor achieved could be as high as 5 thru 10 subject to the LRW salt bearing and activity [14, 17]. (In paper [18] radiation and thermal stability of polymers have been presented.)

On the other hand, we didn't meet any example of polymer use in case of emergency at nuclear sites or at any chemical industry objects.

The presented work was aimed at the search for the simple engineering decisions that would provide the waste immobilization in case of the accidental liquid spills.

The application of polymers for the purpose indicated would allow in case of emergency to reduce the radioactive aerosol concentration in the air and prevent the distribution of aerosols on the premises and in the environment.

For the first time the possibility of the polymer application as a remedy in the emergency situations resulted from spills of the radioactive solutions is justified and confirmed experimentally.

RESULTS OF EXPERIMENTS AND DISCUSSION

As we noted beforehand the possible causes of accidents and emergency situations are technological and waste solution spills. The resulting radioactive contamination area causes the apparent possibility of the biologically hazardous component contact with the environment. From the standpoint of possible emergencies and incidents the most serious challenge is associated with the liquid radioactive waste handling, the volume and total activity of it greatly exceeding those of the solid radioactive waste.

The process of radioactive waste isolation is aimed at the conversion of waste into the stable final waste forms to reduce potential hazards associated with the temporary waste storage, transportation, and final disposal. The spill response process tentatively consists of three phases: the first – the spill localization, the second – the collection and removal of the product from the contaminated surface, and the third – the transportation of the collected product to the processing or disposal site. If an option to collect the spilled solution into waste tanks is not available, the primary task is to contain the solution followed by the conversion thereof into a stable form thus reducing the potential risks of the spread radioactive contamination.

We believe the spilled solutions could be effectively isolated and the obtained solid products successfully contained with the aid of the third generation of the high-tech polymers manufactured in industrial scale by the NOCHAR Company in the U.S.

Solidified products in the form of solid waste can be easily assembled using simple devices or mechanisms and be shipped to storage. (If necessary, the obtained solid waste may be subjected to further conditioned, e.g., by cementation.)

It should be noted the following performance criterions were selected for LRW immobilization methods:

- The materials, used in operation, should ensure strong fixation in a polymer matrix of all radionuclides, occurring in the solutions in the case of aqueous solutions, organic solutions and mixed (aqueous – organic) solutions solidification as well;
- The composition and the amount of reactants to be used should not hamper further waste processing and increase significantly their volume in comparison with the volume of liquid spills;
- Products solidified have sufficiently high resistance to radiation and are not flammable.

That's why the choice has been made in favor of using the third generation of the high-tech polymers offered by the 'Nochar' company. Results of experiments on solidifying solutions of the different composition are given in Table 1 [14- 16].

The experimental results obtained it has been shown that Nochar's polymers have a versatile affect and are capable to solidify aqueous solutions of various acidities and specific activities; organic liquids (solvents and extractants); and suspensions and sludges of different compositions. If apart from organic liquids the liquid waste contains an aqueous phase, a combination of various polymer materials is used to solidify the whole mass of the liquid waste, without additional stages associated with the mix separation and the liquid phase removal.

The solidification of liquid waste for the subsequent conditioning and transportation to the long-term storage sites the mass of polymer to the mass of the liquid phase ratio (S/L) should be maintained within the range of 1/1 – 1/3. If polymers are used to prevent the radiochemical or the other toxic waste spills the S/L ratio could be much less within the range of 1/5 – 1/10 and lower. That amount is sufficient to make the gel, which unlike the liquid is non-fluid and blocks further spread of either radioactive or hazardous chemical pollution.

In case of an accidental spill of radioactive solutions, beside the hazards associated with the spread of the contamination on the premises, there is always the hazard of the radioactive aerosol entrainment with the air flow, which makes the additional threat for the site service personnel.

As long as in the process of solidifying aqueous solutions the first stage is the formation of a colloidal system followed by the formation of a gel one may assume that the evaporation of

liquids and the release of radioactive aerosols into the gaseous phase is going to be very different as compared to the true solutions behavior.

TABLE I. The description of some real waste been used in experiments.

The LRW composition	Solidification conditions	
	Specific activity of solution	The mass of the polymers to the mass of the liquid ratio, S/L, and polymers composition
Acidic drains. (HNO ₃ concentration more than 3 mole/L)	Total β-activity 0.4·10 ⁶ Bq/L	1:4 Polymer No960 – 90% Polymer No910 – 10%
The aqueous solution (the evaporator cube residue). pH = 12.7. COD* = 1900 mgO/L	Total α-activity 2.5·10 ⁶ Bq/kg, the total β-activity 7.3·10 ⁷ Bq/kg.	1:5 Polymer No960 – 100%
The aqueous solution (the evaporator cube residue). pH = 12.7. COD = 1900 mgO/L	Total α-activity 2.5·10 ⁶ Bq/kg, the total β-activity 7.3·10 ⁷ Bq/kg.	1:5 Polymer No960 – 95% Polymer No910 – 5%
The aqueous solution from the evaporator	Total α-activity 5·10 ⁴ Bq/kg, the total β-activity was 1.1·10 ⁷ Bq/kg.	1:3 Polymer No960 – 80% Polymer No910 – 20%
Low-level decontamination washing solutions (about 5% by volume of organic admixtures in the sample)		1:4 Polymer No960 – 80% Polymer No910 – 20%
Perlite slurry	Total β-activity 1.1·10 ⁴ Bq/kg. Activity of Am-241 was 3.1·10 ⁴ Bq/kg	1:3 Polymer No960 – 80% Polymer No910 – 20%
An organic precipitate from the LRW collector tank	Total α-activity 6.6·10 ⁶ Bq/kg, the total β-activity 1.1·10 ⁷ Bq/kg.	1:3 Polymer No960 – 80% Polymer No910 – 20%
Drains of organic liquids (water phase is presented, according manual observation it's something 3 -10%)	Total β-activity 3.6·10 ⁶ Bq/L.	1:3 Polymer No960 – 20% Polymer No910 – 80%
Spent extractant (17% TBP in a hydrocarbon solvent).	Total β-activity 1.4·10 ⁶ Bq/kg	1:10 Polymer No910 – 100%
The scintillator liquid.	Total β-activity 5.9·10 ⁶ Bq/L (including Cs-137 - 2.1·10 ⁶ Bq/L, Eu-154, 2.6·10 ⁶ Bq/L)	1:2 Polymer No960 – 4% Polymer No910 – 96%

* Chemical Oxygen Demand

The experimental results obtained in our attempt to determine the aerosol entrainment during the solidification of the solution with the polymer and during the evaporation of liquid without it are given below.

In run 1 the plastic beakers containing the solution with the polymer additive and without it were covered by paper filters; after the experiment was accomplished, the alpha- and beta- activity of the filters was measured.

In the process of the run the loss of the solidified sample mass and the mass loss of the solution without the polymer additive were measured. The experimental conditions and the aerosol entrainment data are given in Table 2.

TABLE II. Experimental conditions and the data on the aerosol entrainment in the process of drying samples in the air atmosphere.

Experimental conditions		Alpha-activity of the filter, relative units	β -activity of the filter, relative units
The composition of solution	The mass of the polymers to the mass of the liquid ratio, S/L		
A sample of the aqueous solution, α-activity was $7.5 \cdot 10^4$ Bq/kg; the total β-activity was $1.1 \cdot 10^7$ Bq/kg.	1:3	$3.0 \cdot 10^{-5}$	$2.0 \cdot 10^{-5}$
A sample of the aqueous solution, α-activity was $7.5 \cdot 10^4$ Bq/kg; the total β-activity was $1.1 \cdot 10^7$ Bq/kg.	-	$1.2 \cdot 10^{-4}$	$6.1 \cdot 10^{-6}$
An organic precipitate from the LRW collector tank, α-activity was $6.6 \cdot 10^6$ Bq/kg; the total β-activity was $1.1 \cdot 10^8$ Bq/kg.	1:3	$2.5 \cdot 10^{-7}$	$7.1 \cdot 10^{-7}$

From the data given in Table 2 one may suppose that the aerosol entrainment factor is lower for the evaporation of water after the addition of the polymer in comparison with water evaporation from the surface of liquid spills (waste).

However the experiments we performed should have been considered as the preliminary ones and the data obtained needed thorough checking. It was necessary to perform additional experiments to find out the effect of a whole number of factors, namely, the solution composition, S/L ratio, drying temperature, etc.

In the experiments we used the actual liquid radioactive waste arising at the Radium Institute Applied Radiochemistry Department building. Radioactivity of the solution was as follows:

$A_{sp. \beta} = 1.5 \cdot 10^8$ Bq/L; $A_{sp. \alpha} = 1.3 \cdot 10^6$ Bq/L; $A_{sp. Cs-137} = 6.3 \cdot 10^7$ Bq/L; $A_{sp. Am-241} = 7.8 \cdot 10^5$ Bq/L; dry residue = 19.1 g/L; pH = 12.6; COD (Chemical Oxygen Demand) = 160 mgO/L.

An aliquot weight of polymer No.960 and an aliquot of LRW were put into a 150 sm^3 polyethylene beaker. The analytical filter was put onto the beaker and secured by the prepared lid. The beaker was weighted and the holding period started.

Repeated weighing of the beaker was done in 1, 3, 7, 15 and 30 days; the filter was replaced by a fresh one. The removed filter was assayed with the aid of the radiometric facility and a gamma-

spectrometer. The gross β -activity, gross α -activity, the radioactivity of ^{137}Cs and ^{241}Am radionuclides, and the waste entrainment rate were determined.

All in all, the entrainment of radionuclides and the total beta- and alpha-activity is negligible that is associated to some extent with the low specific activity of feed solutions. A lot of the readings were within the measurement inaccuracy range.

On account of the low radionuclide entrainment value in the next run we had to use the same analytical filter during the entire sample holding period to obtain more or less reliable results.

The performed work resulted in evaluation of the water and radionuclide entrainment rate from the compounds containing polymer No.960 and the actual LRW as a function of the polymer -to-LRW ratio and the holding time duration at the temperature of 20°C and 80°C.

The experimental conditions and the aerosol entrainment data are given in Table 3 and 4.

TABLE III. The radionuclide entrainment as a function of the holding time at the $t=22\pm 2^\circ\text{C}$.

The composition of sample, S/L	The entrainment of total β -activity, % of the initial activity in the sample			The entrainment of total α -activity, % of the initial activity in the sample		
	Holding time, days					
	1	3	35	1	3	35
1:1	2.0×10^{-4}	3.1×10^{-4}	5.3×10^{-4}	$\leq 9.6 \times 10^{-4}$	$\leq 9.6 \times 10^{-4}$	$\leq 9.6 \times 10^{-4}$
1:6	1.1×10^{-4}	1.6×10^{-4}	2.0×10^{-4}	$\leq 3.2 \times 10^{-4}$	$\leq 3.2 \times 10^{-4}$	1.4×10^{-4}
1:1+2g FS-2	1.4×10^{-4}	1.4×10^{-4}	8.5×10^{-5}	$\leq 9.6 \times 10^{-4}$	$\leq 9.6 \times 10^{-4}$	1.5×10^{-4}
1:6+6g FS-2	$\leq 1.4 \times 10^{-5}$	$\leq 1.4 \times 10^{-5}$	$\leq 1.45 \times 10^{-5}$	$\leq 3.2 \times 10^{-4}$	$\leq 3.2 \times 10^{-4}$	$\leq 3.2 \times 10^{-4}$

TABLE IV. The radionuclide entrainment as a function of the holding time at the $t=80\pm 2^\circ\text{C}$.

The composition of sample, S/L	The entrainment of total β -activity, % of the initial activity in the sample			The entrainment of total α -activity, % of the initial activity in the sample		
	Holding time, hours					
	7	31	77	7	31	77
1:1	1.0×10^{-3}	3.5×10^{-4}	$\leq 5.9 \times 10^{-5}$	1.5×10^{-3}	1.3×10^{-4}	$\leq 3.2 \times 10^{-4}$
1:6	6.9×10^{-5}	1.7×10^{-4}	8.3×10^{-5}	4.2×10^{-5}	2.9×10^{-5}	3.4×10^{-5}
1:1+2g FS-2	2.7×10^{-5}	4.3×10^{-5}	$\leq 5.8 \times 10^{-5}$	$\leq 3.1 \times 10^{-4}$	$\leq 3.1 \times 10^{-4}$	$\leq 3.1 \times 10^{-4}$
1:6+6g FS-2	1.5×10^{-4}	1.9×10^{-4}	3.7×10^{-4}	1.6×10^{-4}	1.4×10^{-4}	2.6×10^{-4}

During the experiments it has been shown that from the aerosol entrainment reduction standpoint the good results could be obtained if the FS-2 sorbent is added to the solution.

At the final stage of work the radioactive aerosol entrainment was determined by the direct measurement of the activity of the condensed vapor phase yielded from the solidified samples in the process of drying rather than by the analysis of filters. To accomplish that, the experiment on freezing water vapors with the reservoir trap cooled by the liquid nitrogen has been performed.

A liquid nitrogen cooled trap was used in the experiment for freezing the water vapors.

Drying of the solidified sample was performed at the temperature of 95°C. The activity of the actual solution used to run the experiment was as follows: the total α -activity of $5.7 \cdot 10^6$ Bq/L and

the total β -activity of $4.5 \cdot 10^6$ Bq/L, Cs-137 - $4,2 \cdot 10^6$ Bq/L, Am-241 - $5,5 \cdot 10^6$ Bq/L, the salt bearing of 100.0 g/L, pH = 9.9.

The water vapors were distilled off daily for 7 hours until the solidified sample was completely dried.

By the end of every working day the facility was disassembled and the volume of the obtained condensate was measured. Every portion of the condensate and the bubbling flask water were analyzed daily by means of a gamma-spectrometer.

During the entire time span of the run the solidified sample remained incoherent in its upper part, and in the bottom it was a viscous monolith.

The experiment was run for 9 days, and in all the samples taken the activity was equal or less than the detection limit of 0.5 Bq/L for ^{137}Cs and 2.8 Bq/L for ^{241}Am , respectively.

In the case of the accidental LRW spills the techniques proposed for calculating the radionuclide release rate into the air and the radionuclide concentration in the air of working rooms may appear to be useful. Yet the experiments performed should be considered as the preliminary ones. The data obtained need thorough checking by the additional experiments with the other waste compositions with different specific activity.

However if the planned experiments appear to be successful, the possibility will appear to propose the application of polymers during the up rise and remediation of emergency situations when the radioactive aerosol content in the air must be reduced and the distribution thereof in the working areas and the environment must be prevented.

The results obtained allow a conclusion about the high efficiency of the third generation high-tech Nochar polymers application for the liquid radioactive waste spill response in the emergency situations at the atomic and chemical industry sites.

The sequence of operations when using polymers for the emergency damage control and remediation at nuclear sites is shown in Fig. 1.

In the meantime, the selection of conditions for the hazardous liquid spill response should be made in every particular case with regard to the chemical composition and specific activity of waste and in view of the technology intended for the following collection and removal of the solidified products to the repository.

That's exactly why additional experiments should be performed with the other waste compositions including the solidification of heterogeneous waste (sludge, slurry) of the different specific activity in the presence of toxic substances – the chemical industry waste. If the proposed experiments are successful, the real possibility will arise of using polymers for the prevention and remediation of emergencies at many sites of atomic and chemical industry.

In the conclusion of this section one more issue associated with the safe maintenance and decommissioning of large liquid radioactive waste storage tanks should be addressed. In our opinion the use of polymers could be the most effective with these very objects.

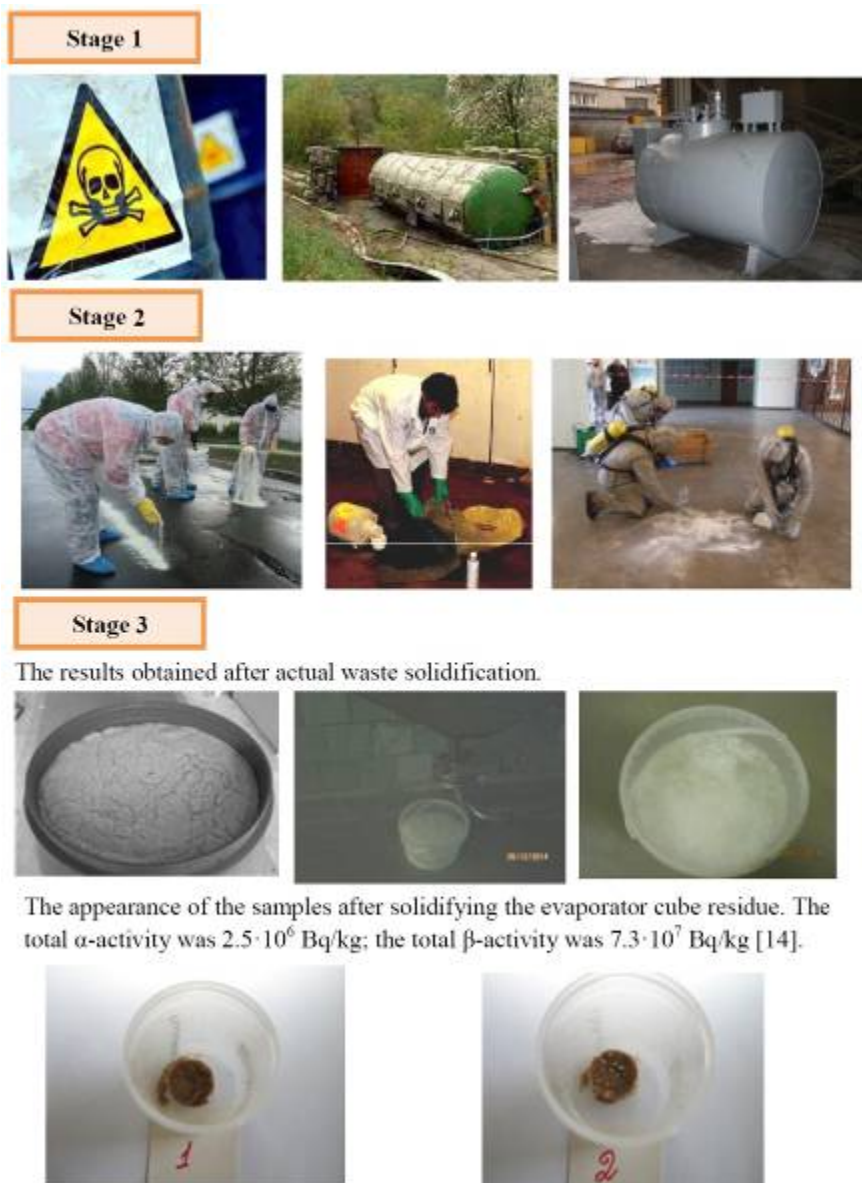


Fig. 1. How we can use polymers for the emergency damage control and remediation at nuclear sites.

To date the situations with the RW storage tank decommissioning in Russia and in the United States have much in common: the lack of the waste management strategy developed at the initial stage of production, the end of the storage tank lifetime, and the associated environmental issues. At the initial development and assimilation stage of the radiochemical defense plutonium production technology the primary type of waste was suspensions, i.e. the suspended matter containing liquids. That waste was the source of the heterogeneous radioactive waste, namely, sludge. The challenging issues associated with handling radioactive waste accumulated in the course of the defense nuclear programs are important not only for Russia and the U.S., but for other countries possessing the fissile material production technologies, too.

Since 1948 about 100 tanks of 300 thru 1300 m³ volume have been built in the former Soviet Union at PA “Mayak”.

The problems associated with the heterogeneous waste in Russia have much in common with those of the Hanford Tank Farm, Hanford, USA. To date they have in operation the waste tanks of 210 thru 3800 m³ volume erected in the period of 1943-1964.

Hanford’s waste tanks contain complex mixtures that fit into 89 separate chemical profiles. Chemical concentrations in each of the tanks widely vary by as much as 100percent [19-22]. Although radioactive materials make up about one percent to of Hanford’s waste volume, they are enough to make the wastes highly dangerous, with expose relieves inside the tanks as high has 10,000 rad per hour [19].

There are several forms and layers of wastes, which are “heterogeneous in all phases, both within a given tank and among different tanks [23].

Any leak of this waste results in the radioactive contamination of tens thru hundreds meter cubic of soil. The leak is rather difficult to fix, especially in an underground tank, and the most acceptable way of the emergency response is to retrieve the waste, i.e. to pump the waste over from the old leaky tank to another one.

The authors do not believe the application of polymers is likely to trigger a revision of the waste handling technology used to date for processing the overall volume of waste at similar facilities either in Russia or in the United States and other countries. On the other hand, the application of polymers may be effective enough to prevent the possible leaks and the following migration of radio nuclides into the environment.

CONCLUSIONS

The actual situation very often differs greatly from the picture established in the public conscience. And indeed the results obtained allow a conclusion about the high efficiency of the third generation high-tech Nochar polymers application for the liquid radioactive waste spill response in the emergency situations at the atomic and chemical industry sites.

First time substantiated and experimentally demonstrated the possibility of using polymer materials as a means of liquidation of emergency situations caused by leaks of radioactive solutions. On the whole number of reasons the economic assessment in this focus area is very hard to accomplish, but it requires very thorough and weighted approach anyway.

Another promising area of the polymer application could be the technical challenges associated with the decommissioning of NPPs and other radiation hazardous objects and facilities including the large volume LRW storage tanks.

Development of the *in-situ* solidification technique for the mobile (liquid) components of heterogeneous waste will provide a good option to contain the radio nuclides within the storage site boundaries.

Another field of their application could be the utilization as components of engineering barriers for improving the safety at the most vulnerable areas of waste storage and processing.

In conclusion it is worth to mention also that besides the atomic industry applications the polymers could find their use at various enterprises for localizing toxic waste, namely, heavy-metal containing sludge of the sewage treatment plants, galvanic sludge, etc., severely hazardous when accumulated in the environment.

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