LONG-TERM SAFE STORAGE AND DISPOSAL OF SPENT SEALED RADIOACTIVE SOURCES IN BOREHOLE TYPE REPOSITORIES

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Abstract

Borehole disposal of spent sealed radioactive sources (SRS) has a number of noticeable advantages:

- Effective radiating protection;
- Long-term safety of disposal.
- Operational safety;
- Easy in operation;
- Conceptually simple;
- Simplicity of a design;
- Easy of erection;
- Practicability practically everywhere;
- Low cost;
- Long practice of application;
- Retrievable (the new project);
- Ease in improvement;

Russian Federation has the leading experience in applying borehole storage/disposal method for SRS. A new immobilisation technology for sources being disposed of in underground repositories was mastered by 1986 and since then it is used in the country. This method uses all advantages of borehole type repositories supplementing them with metal encapsulation of sources. Sources being uniformly allocated in the volume of underground vessel are fixed in the metal block hence ensuring long-term safety. The dissipation of radiogenic heat from SRS is considerably improved, radiation fields are reduced, and direct contact of sources to an environment is completely eliminated. The capacity of a typical borehole storage/disposal facility is increased almost 6 times applying metal immobilisation. That has made new technology extremely favourable economically. The metal immobilisation of SRS is considered as an option in Belarus and Ukraine as well as Bulgaria. Immobilisation of sources in metal matrices can be a real solution for retrieval of SRS from inadequate repositories.

Introduction

Safe disposal of SRS is an important issue faced in many countries [1]. Borehole or well type repositories are widely used both for storage and disposal of SRS in many countries of the world: Russian Federation, Ukraine, Belarus, Armenia, Kazakhstan, Uzbekistan, Latvia, Poland, Bulgaria, Hungary, USA, India; at the present the republic of South Africa studies this option. The history of bore hole disposal totals over 40 years of successful operation. Borehole disposal has allowed during many tens of years to store safely SRS, which on their parameters are high-level radioactive waste. Despite of it, borehole storage and disposal of SRS was not especially discussed in publications. Even IAEA, publishing a large number of documents, in any way did not reflect in the publications practice of well storage and borehole disposal of radioactive waste. The storage and disposal of SRS in borehole repositories for the first time was considered in the IAEA by the department of waste technology in 1997, and this year the department of safety of radioactive waste is preparing for publication a TECDOC on the safety of borehole disposal.

Analysis of storage condition of SRS in borehole repositories carried out in 80-s revealed the unsatisfactory degree of safety for free storage of sources in underground vessels. Initially the burial technology assumed simple dump of SRS into underground vessels of disposal/storage facilities. Thereby sources accumulated at the bottom of underground vessel so even at rather low volumetric filling underground capacity the radiation fields in it appeared extremely powerful (units - tens of MR/h). SRS generate radiogenic heat, which is dissipated in an environment, including the way of air convection. There is an ascending stream of warm air from the SRS repository, which can bear radioactive aerosols from the underground vessel. Besides this there are convective counter-currents of air, which are increasing the content of moisture in the repository by condensation of water on the cold walls of the loading channel.

The radiolysis of water occurs in powerful radiation fields of SRS with accumulation of explosive hydrogen (the lower limit of the explosive contents of hydrogen is 4,2 %). The safety of SRS storage is reduced also by accumulation of other radiolysis products.

The State Unitary Enterprise Moscow SIA "Radon" in the beginning of 80-s developed a new SRS disposal technology. In order to ensure long term safety of storage/disposal facilities it was suggested to isolate SRS into an additional matrix material. Taking into account parameters of typical facilities (up to 50 kg - eqv. of radium per one repository) metal matrixes were chosen. The new immobilisation technology for sources being disposed of in underground repositories was mastered by 1986. Ministry of Health of the USSR has certified the new immobilization method to be applied at regional specialized facilities of the "Radon" network. The new method used all available advantages of borehole type repositories supplementing them with excellent long-term safety. Sources thus were uniformly allocated in the volume of underground vessel, being fixed in the metal block inside it. The dissipation of radiogenic heat from SRS is considerably improved, radiation fields are reduced, and direct contact of sources to an environment is completely eliminated. Due to it the capacity of a typical borehole storage/disposal facility is increased almost by 6 times that has made new technology extremely favourable economically [2].

In 1991 the first mobile immobilization plant was mastered. It enabled application of new immobilization method for the entire network of regional facilities of system "Radon". There is no necessity to supply each "Radon" type enterprise by immobilizing complexes since the SRS immobilisation can be done in campaigns. It is more expedient to have some installations in each big region of the country, using it for technological needs as required. To present time the SRS immobilisation was carried out at 5 regional facilities "Radon" in Russia: Sergiev Posad, Volgograd, Nizhni Novgorod, Ekaterinburg, and Ufa. Besides the immobilization of sources was done at Novovoronezh NPP. Total activity of all SRS, which were included in metal matrixes in the Russian Federation, exceeds 1 million Ci. The metal immobilisation of SRS is planned at other regional enterprises in Russia, being considered as an option in Belarus and Ukraine. This method considers as well Bulgaria.

The purpose of this report is to give an overview of currently applied SRS storage/disposal options and to describe the routes of ensuring long-term safety of disposal facilities.

Borehole type repositories

Borehole repositories are of typical design developed by the State Project Institute GSPI at the fall 50-s. The typical bore-hole repository (Fig. 1, left) has an underground stainless steel cylindrical vessel (5) with diameter 200 mm and height 1500 mm which is placed at 4 m depth in a steel-enforced concrete well (3 and 4). The thickness of vessel walls is 5 mm. The stainless steel loading channel (2) of the repository has a form of curved (spiral) tube with the diameter 108x5 mm. At the upper part of repository there is a carbon steel conical socket (1). It provides safe discharging of transport containers. A carbon steel lid closes this socket. The concrete wall of repository is surrounded by a clay-cement (or clay) mixture, which fills the initial construction hole in original soil as a seal material.

Originally typical repositories were designed for the disposal of sources with the total radioactivity corresponding to an equivalent of 50 kg of radium. The maximum dose rate on the surface of repository near the loading channel obeying the design cannot be higher than 0.82 mR/h. The underground reservoir is heated due to radiogenic heat generation by sources. Accordingly with the original design maximum allowable temperature in the reservoir is 230°C.

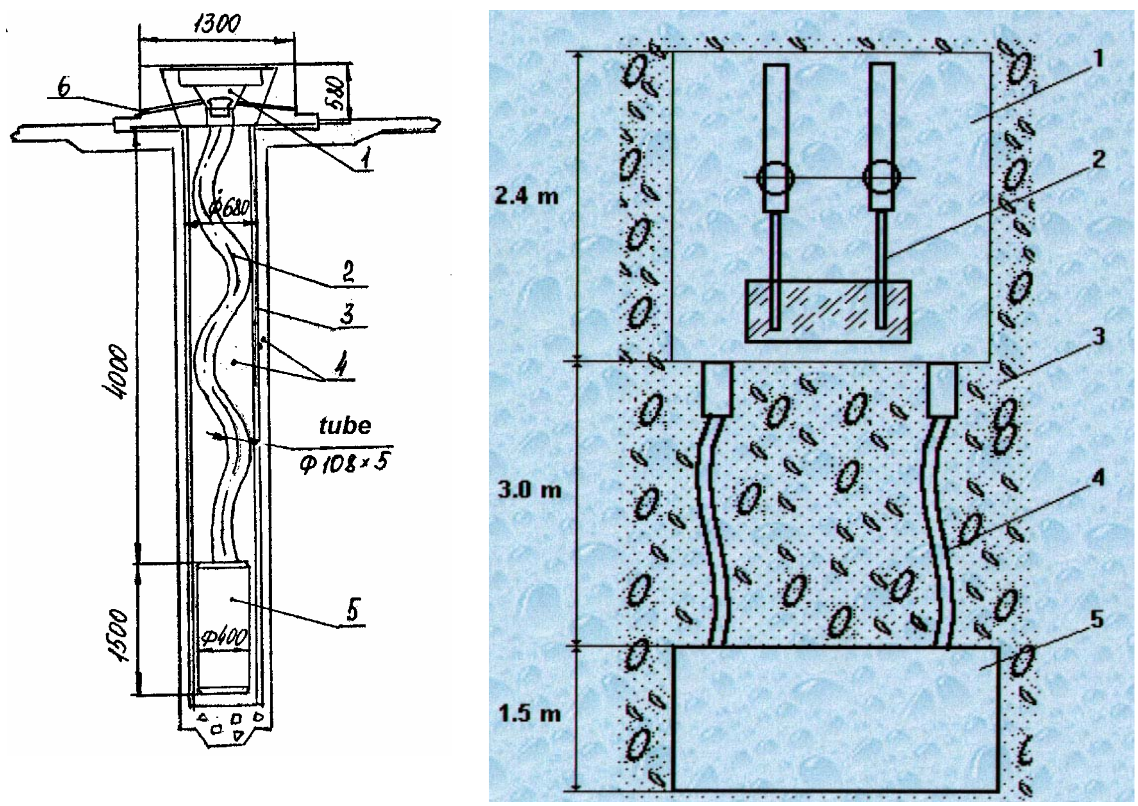

Fig.1. Left: the design of typical bore-hole repository (dimensions in mm).
1 - carbon steel conical socket, 2 - stainless steel loading channel, 3 - steel-enforced concrete well, 4 - concrete, 5 - stainless steel cylindrical vessel, 6 - drainage channel.
Right: the borehole repository at S. Petersburg RSF Radon. 1- hot cell, 2 –manipulator, 3 – concrete, 4 –loading channel, 5 –underground vessel.

As a rule at regional specialised facilities "Radon" there is a few borehole repositories for the disposal of spent sources. In reality there were constructed a number of slightly different borehole repositories with similar design to typical one. Besides some of repositories were constructed with deviation from the typical design (see Fig.1, right).

Special containers (KI-400, KTB-26-12) with upper charging and bottom discharging is provided in order to load bore-hole repositories. Dose rate on the surface of containers shall be not higher than 200 mR/h that corresponds to III transport category accordingly with sanitary rules. For installation of containers at upper part of repository a carbon steel conical socket was designed in order to provide safe discharging of containers. A carbon steel lid closes the socket after the repository loading.

Belarus recently put into operation a new advanced design borehole type repository envisaging the retrieval of sources [3]. Ukraine besides borehole disposal has developed a special container to immobilise SRS using metal matrices [4]. Russia has developed as well a new design for long-lived radioactive sources which enable retrieval of underground container for transportation to final disposal (Fig.2). This repository-container has an underground reservoir for storing sources with the volume 106 dm^3. It is designed to store SRS with total radioactivity 3,7 10^{14} Bq (10 kCi). Supplying these repositories with an appropriate immobilisation technology one can safely store sources for extended period of times.

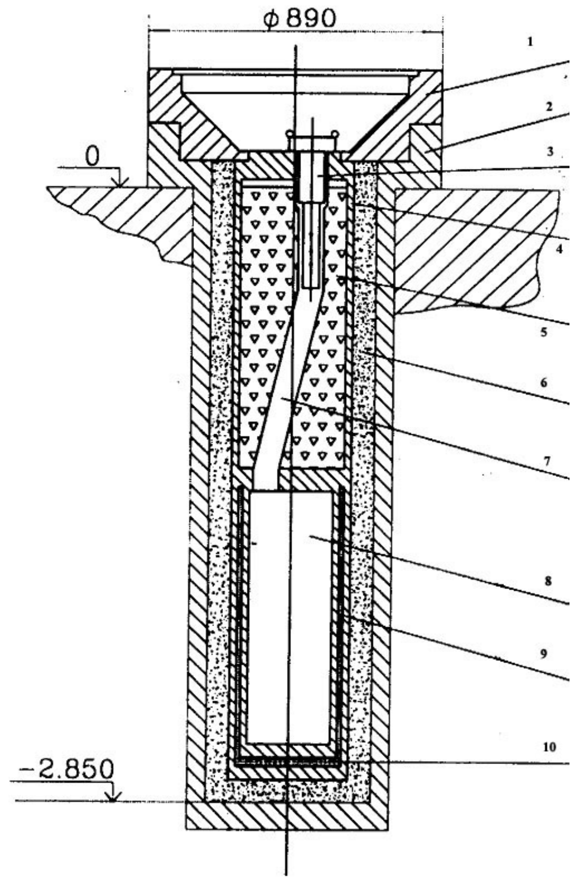

Fig.2. The design of new borehole repository-container for long-lived sources.
1 - conical socket, 2 – steel reinforced concrete tube, 3 – shielding lid, 4 – tube, 5 – shot iron, 6 – filling, 7 - stainless steel loading channel, 8 – vessel, stainless steel walls, 10 – sand.

Status of repositories

Borehole repositories operate since early 60-s. The inspection of repositories using specialised tools revealed extremely high radiation field even the radioactivity of sources was far from the upper limit. (i.e 50 kg-eqv. of radium). Analysis of storage condition of SRS in borehole repositories revealed the unsatisfactory degree of safety for free storage of sources in underground vessels. Initially the burial technology assumed simple dump of SRS into underground vessels of disposal/storage facilities. Thereby sources accumulated at the bottom of underground vessel so even at rather low volumetric filling underground capacity the radiation fields in it appeared extremely powerful (units - tens of MR/h). SRS generate radiogenic heat, which is dissipated in an environment, including the way of air convection. There is an ascending stream of warm air from the SRS repository, which can bear radioactive aerosols from the underground vessel. Besides this there are convective counter-currents of air, which are increasing the content of moisture in the repository by condensation of water on the cold walls of the loading channel. The radiolysis of water occurs in powerful radiation fields of SRS with accumulation of explosive hydrogen (the lower limit of the explosive contents of hydrogen is 4,2 %). The safety of SRS storage is reduced also by accumulation of other radiolysis products.

Small amounts of water were accumulated in repositories due to condensation of water vapours from air on cold walls of loading channel. This is a slow process however during many years of operation in dump conditions when there is a flow of hot air from the bottom part of repository upward and a flow of dump air downward some portion of water is accumulated by condensation in the underground vessel. Large amounts of water in repository indicate on poor operation of drainage system. In any case presence of water in repository and powerful ionising radiation decreases safety of sources disposal.

Radiolysis of air and water occurs under the influence of powerful ionising radiation. Oxides of nitrogen and ozone are the main products of radiolysis. Radiochemical yield of nitrogen oxides is 1.23 molecules/100eV. Concentration of nitrogen oxides in irradiated volume can be determined by formula

$$C=1.6*10^{-4}*D,$$

where D - absorbed dose of radiation in rad. At dose rate about 10^7 rad/h concentration of nitrogen oxides in a closed volume after one day will reach 30 g/l. Ozone is produced with a high radiochemical yield 15 molecules/100 eV. Due to its oxidising properties it contributes to oxidation of nitrogen oxides to NO_2. With water nitrogen peroxide produces nitric acid. This nitric acid during multiple moistening-drying on the surface of sealed source can be concentrated. Formation of nitric acid contributes to acceleration of source case corrosion. Ozonation of water solutions accelerates corrosion also. Besides this mechanical tensions and micro-fissures also contribute to accelerated corrosion of cases. These processes caused a number of accidents with sealed radiation sources during the operation of irradiation installations. Similar conditions are inherent to borehole repositories. In water samples from repositories large amounts of suspensions (up to 34 g/l) were observed which were products of corrosion. Radiometric analysis of water from repositories showed that radionuclides concentration was about 10^2 - 10^3 Bq/l. These concentrations can be caused by radionuclides diffusion through fissures of SRS cases and residual contamination of sources. In one repository concentration of Cs-137 was up to 10^6 Bq/l that can be explained by seal failure. If seal failure occurs borehole repository does not represent a safe barrier and radionuclides release into environment is possible by aerosols during the evaporation of moisture or in the form of hot particles. The sources ought to be immobilised to avoid possible release of radionuclides.

Hydrogen is produced during the radiolysis of water. Radiochemical yield of hydrogen is 0.42 - 0.45 molecules/100 eV. Being accumulated in a closed volume hydrogen can form in admixture with air detonating gas. Inferior limit of explosive danger of hydrogen is 4.2 vol.%. Maximal time of safe accumulation of hydrogen in a closed volume can be determined by formula:

$$t=4.03*10^6*V/(K_\gamma*Q),$$

where V - volume of repository (m^3), K_γ - gamma-constant of radionuclide ($R*cm^2/(h*mCi)$), Q - activity of source (Ci). For activity of Co-60 sources about 10,000 Ci and volume of repository 0.2 m^3 the time of safe accumulation of hydrogen is about 6 h. Maximum concentration of hydrogen about 3.6 vol.% was observed in a closed bore-hole repository. In open repositories the hydrogen concentrations are several times lower.

The status of borehole repositories is shown below in Table 1.

Table I. Status of borehole type repositories at regional specialised facilities (RSF) "Radon".

##	Repository	Operating since	Number of SRS	Total activity, Ci	Filling, %	Dose rate underground, R/h	Dose rate at the entrance, μR/h
Sergiev Posad (Moscow SIA Radon)	A (metal immobilised)	1979	52	18548	35	120000	60
	B (metal immobilised)	1990	992	72996	50	3000000	70
	C (metal immobilised)	1979	405	18649	37	170000	50
	D (metal immobilised)	1985	1845	73312	84	100000	30
Nijny Novgorod	A (metal immobilised)	?	2325	?	17	51	15
Rostov	A	1963	283	2808	43	478	15
	B	1963	18801	2505	100	23	26000
S.Petersburg	A	1963	?	10000	?	?	?
	B	1971	?	73000	3	1380000	60
Samara	A	1985	3283	883	6	58400	11
Saratov	A	1963	2874	6	13	166	16
Ufa	A	1964	?	?	?	?	?
	B (metal immobilised)	1976	?	1400	?	782	48
	C	1964	?	200	100	6	40
Volgograd	A	1990	?	3351	5	36400	5
	B (metal immobilised)	1963	?	1200	53	6	16
Ekaterinburg	A (metal immobilised)	1988	886	837		132	15
	B (metal immobilised)	1992	1972	2555		9000	20

Immobilisation technology

Metals have enough radiation durability to be used as matrix material for the immobilisation of SRS. In order to facilitate the encapsulation process and to minimise the influence of high temperature on radionuclides metals with low melting temperature are used [5]. For damaged sources this minimises the volatilisation of radionuclides in the process of their encapsulation.

Encapsulation involves two steps with melted metal portions varying in volume. In the first stage molten metal of temperature above the melting temperature is poured over the sources in underground repository vessel. Owing to difference in the specific weight, the sources rise to the surface of the melt, and on its crystallization they are fixed on the surface. In the second stage of the technological process a thin layer of melt is poured on the surface of metal block in order to cover some floating sources.

In order to obtain a high-performance metal block the melt quantity is calculated depending on the temperature of the melt, source activity, and repository thermal properties from the following expressions:

$$h_i \cong 8.01*10^{-14} \varepsilon_p A_i / \pi \beta \kappa \Delta T_0,$$

where κ - effective repository heat conductivity, ε_p - average energy of one decay, ΔT_0 - permissible overheating of the repository, A_i - radionuclide activity in specific set of sources, β - in this coefficient the effect of repository geometry is included. For typical underground borehole repositories ΔT_0=503 ^{0}K, κ =(2.8+0.3) W/m*K.

The technological process envisages application of sources immobilisation until the repository is filled up (see Fig. 3).

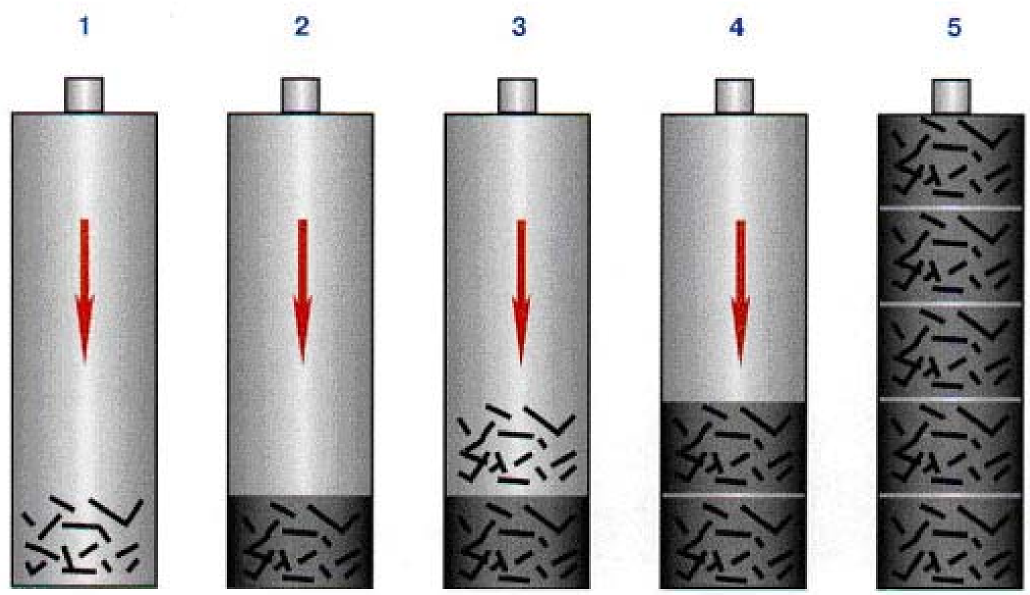

Fig.3. Schematic of SRS immobilisation in borehole repositories.

Structure of the block obtained and coupling between layers were controlled by the use of ultrasonic defectoscopy and testing of the samples for fracture. The measurements showed that only lead and lead based alloys are producing high quality metal blocks without breach of integrity. Some aluminium and tin matrix samples had structural defects in the form of even boundaries between layers. This was caused by rapid oxidation of surface of the metals having the sufficient high reactivity. The lead matrices have a continuous structure with no internal and surface defects.

If SRS contain high volatile radionuclides it is essential to lower melting temperature as much as possible and at the same time to provide consistent cohesion between matrix metal layers. To attain this aim at first sources are poured by lead-based alloy melt (e.g. Bi50.1-Sn14.6-Pb24.9-Cd10.8) having low melting temperature. Upon crystallization of the latter the technological operations above-mentioned are performed. Investigation of the block obtained by ultrasonic defectoscopy has shown no structural defect in the block.

Table 2. Characteristics of immobilising metals and alloys.

Composition, % wt.	Pb	Sn	Pb-32, Sn-68	Bi-50, Sn-22, Pb-28	Bi-50.1, Sn-14.6, Pb-24.5, Cd-10.8
Melting temperature, ^{o}C	327	234	177	100	66

Low-melting alloys with melting temperature below 100 ^{0}C (Table 2) are used for spent sources immobilization buried in the old repositories flooded by water. For this purpose sources are initially poured by melted alloy and if required, water in the repository is heated to temperature higher than alloy melting temperature. Owing to significant difference in the specific weight, water or water-clay suspension is displaced from sources layer by the melt. Upon the pumping out water and drying the repository, metal block melting temperature required is achieved by successive pouring of several melted lead portions.

A series of mobile facilities is available at Moscow SIA "Radon" to immobilise SRS at regional specialised facilities "Radon". One of them is shown in the Fig.4.

Observations of status of repositories after source immobilisation showed absence of hydrogen: its concentration was below registration limits of devices. Besides this both radiation fields and temperatures in repositories decrease considerable. Even in the case of water impact there was no direct contact of water with sources. No contamination of water was observed.

Fig.4. Mobile modular type facility to immobilise SRS in borehole type repositories.

In 1998-1999 the Moscow SIA "Radon", State Scientific Centre VNIINM and Institute of Biophysics of Russian Academy of Science fulfilled a detailed analysis of spent sealed radiation sources safe storage in bore hole repositories. The analysis has showed that reliable radionuclide insulation from environment during the whole storage time of 500 (and up to1000) years is provided through source's immobilisation in a metal matrix material (lead matrix). Even in the hypothetical case of total flooding with simultaneous damaging of all engineering barriers, radionuclides release will cause a summary dose load not exceeding (5.5 - 7.5)x10^{-5} Sv/y [6]. The safety analysis has shown a high degree of safety of SRS borehole disposal in the case when sources are fixed in metal matrices for at least 500 – 1000 years. Even in case of rather improbable events the dose burdens for population will not exceed (5.5 - 7.5) 10^{-5} Sv. As in the case of normal evolution scenarios the burden is nil.

Due to very low corrosion rate of lead and low solubility of lead salts there is practically no impact to environment of lead. This impact was assessed to be very low earlier [7] and confirmed recently by detailed calculations for near surface repositories containing large amounts of lead waste [8]. As for SRS it is deemed that after the decay of radionuclides the metal block will be retrieved and recycled.

Conclusion

Bore hole repositories operate successfully over 40 years. They have allowed during many tens of years to store safely SRS. The conception of disposal of SRS into borehole repositories was developed in former USSR to the end of 50-s - beginning of 60-s. Presently borehole repositories are used at many regional specialised facilities "Radon" in Russian Federation as well as in other countries. The borehole disposal-storage is supplemented by metal encapsulation of sources. Sources are fixed in the metal block hence long-term safety is ensured by high durability of metal matrix immobilising sources. At present time the design of borehole repositories is improved envisaging the possibility to retrieve the metal block with sources.

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