MANAGEMENT OF SPENT RADIATION SOURCES AT REGIONAL FACILITIES "RADON" IN RUSSIAN FEDERATION

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ABSTRACT

The system of management of spent sealed radiation sources in Russian Federation is based on centralised approach. This system comprises 16 regional facilities of the system "Radon" which provide safe management of spent sealed radiation sources. The scientific and methodical guidance on activities of regional facilities "Radon" carries out the Scientific and Industrial Association "Radon", Moscow. At regional facilities "Radon" spent sealed radiation sources are disposed of in bore hole type repositories. They are also stored in containers being placed into repositories for solid radiation waste. Spent sealed radiation sources which contain long lived radionuclides are stored until the decision on their final disposal into a deep geological formation. At five regional "Radon" facilities (Zagorsk branch of Moscow SIA "Radon", Volgograd, Ekaterinburg, Nizhny Novgorod, and Ufa) the sources were immobilised into Metallization matrix materials (lead or lead based alloys). More than 1 million Ci of spent sealed radiation sources are stored at present time in conditioned form in bore hole repositories. The analysis of the work on spent sealed radiation sources management shows that the operating system is able to solve basic problems, providing safe conditions for the environment. The paper aims to give an overview of current practice in this area.

INRODUCTION

The system of management of radioactive waste including spent sealed radiation sources in Russian Federation is based on centralised approach. This system was established in the beginning of 60-s on the regional basis and comprises a network of 16 regional specialised facilities "Radon", which are located near large industrial centres. The regional specialised facilities "Radon" provide safe management of spent sealed radiation sources.

About 80% of radioactive waste from scientific and industrial centres in the central region of Russian Federation manage the Scientific and Industrial Association "Radon". It serves almost 2000 enterprises, which use radionuclides in their activities. The main part of radioactive waste here is collected from Moscow and Moscow Region (where mainly the waste is collected from Moscow). The volume of radioactive waste to be treated at SIA "Radon" is up to 6000 m^3 per year. Some 18% of radioactive waste are collected in the north-west region of Russian Federation by St. Petersburg regional facility "Radon". It has also experimental plant for treatment and conditioning of waste. Total volume of radioactive waste to be treated at the Leningrad regional facility "Radon" is 3000 m^3 per year. Radioactive waste from scientific and industrial centres on the remaining territory of Russian Federation is collected by other facilities of the system

"Radon". The total inventory of radioactive waste at “Radon” type enterprises is estimated to be $2.0*10^5$ m^3 with the total radioactivity $7.77*10^{16}$ Bq [1].

The basic technical characteristics of RSF "Radon" and their servicing zones (regions) are given in the Table I. Radioactive waste at all RSF “Radon is disposed of in conditioned form. At some RSF “Radon” additional treatment of waste is carried out (see Table I). .

Table I. Regional Specialised Facilities "Radon" in Russian Federation.

	RSF "Radon"	Max. Capacity, m^3/year	Treatment method	Operation region
1	Moscow SIA "Radon"	6000	Incineration Compaction Bituminization Cementation Purification Metallization.	Moscow, *Regions:* Moscow, Briansk, Vladimir, Tver, Kaluga, Ryazan, Smolensk, Tula, Yaroslav, Kostroma
2	St. Petersburg	3000	Evaporation Incineration Bituminization	S.Peterburg, *Regions:* Vologda, Leningrad, Novgorod, Pscov, Kaliningrad, *Republic* Karelia
3	Kazan	20	Cementation	*Republics:* Maryi El, Chuvashia, Tatarstan, Udmurtia
4	Volgograd	10	Cementation* Purification* Metallization.*	*Regions:* Astrahan, Volgograd, *Republic* Kalmykia
5	Nijnyi Novgorod	25	Purification* Metallization.*	*Republics:* Komi, Mordva *Regions:* Ivanov, Kostroma, Kirov, Nijnyi Novgorod
6	Groznyi	30	no	*Republics:* Dagestan, Kabardino-Balkaria, North Osetia, Chechnya
7	Irkutsk	10	no	*Republics:* Buryatia, Tuva, Saha *Regions:* Irkutsk, Chita
8	Samara	10	no	*Regions:* Samara, Ulianov, Orenburg
9	Murmansk	up to 50	no	*Regions:* Arhangel, Murmansk
10	Novosibirsk	up to 40	Cementation*	*Lands:* Altai, Krasnoyarsk *Regions:* Kemerovo, Omsk, Tomsk, Novosibirsk
11	Rostov	up to 20	no	*Lands:* Krasnodar, Stavropol *Region* Rostov
12	Saratov	up to 50	no	*Regions:* Orlov, Belgorod, Voronej, Kursk, Lipetsk, Tambov, Penza, Saratov
13	Ekaterinburg	up to 150	Cementation Metallization*	*Regions:* Perm, Tumen, Ekaterinburg
14	Ufa	up to 10	Metallization*	*Republics:* Udmurtia, Bashkortostan
15	Cheliabinsk	up to 50	no	*Regions:* Kurgan, Orenburg, Cheliabinsk
16	Habarovsk	up to 60	no	*Lands:* Primorie, Habarovsk *Regions:* Amur, Kamchatka

*These works were provided by mobile units of Moscow SIA "Radon".

Recently the Moscow SIA "Radon" examined activities at regional facilities "Radon" accordingly with the program of State Supervision Authority of Russia (GAN). It was revealed that the main type of radiation waste at these facilities comprises spent sealed radiation sources. This is the case both on summarised quantity of accumulated radioactivity (more than 99%) and on general quantity of the containers with waste (more than 80%). Cs-137 (40%), Co-60 (25%), Sr-90 (22%), Ir-192 (8%) and Tm-170 (4%) represent the radionuclide composition of the spent sealed radiation sources. Other radionuclides in spent sealed radiation sources comprise not more than 1%.

Thus from the point of view of radiation safety the conditions of storage of spent sealed sources are of primary importance.

SEALED RADIATION SOURCES MANAGEMENT SYSTEM

The scheme of the management of spent sealed radiation sources in Russian Federation is given in the Fig.1. Radiation-chemical plants produce sealed radiation sources. The main dealer of sealed radiation sources in Russia is the firm ISOTOP. Currently the company NPP DOZA is also a large source delivering company. The ISOTOP company supplies sealed radiation sources to users. Those are scientific and research institutes, clinics, production plants, etc.

After applications or in the case of end of operation time the radiation sources are considered as spent. Transportation of spent sealed radiation sources is carried out in transportation protective containers. Spent sources are loaded in container by a special company SMNU (Special Installation-Arranging Board of Ministry of Atomic Energy) the radiation control being provided during all operations. Users, which have necessary equipment, can carry out loading of spent sources in transportation containers themselves. All further operations with spent sources are within the sphere of activity of regional specialised facilities "Radon".

Special vehicles are used for the transportation of containers with spent sealed radiation sources. Routes of spent sources transportation from users to regional specialised facilities "Radon" are co-ordinated by Departments of State Auto Police. Patrol cars accompany special vehicles that transport containers with spent sealed sources along the route. At regional specialised facilities "Radon" spent sealed radiation sources with short lived radionuclides are disposed of into shallow ground repositories. Long lived radionuclides are stored in shielded containers until the decision on their final disposal into deep geological formation.

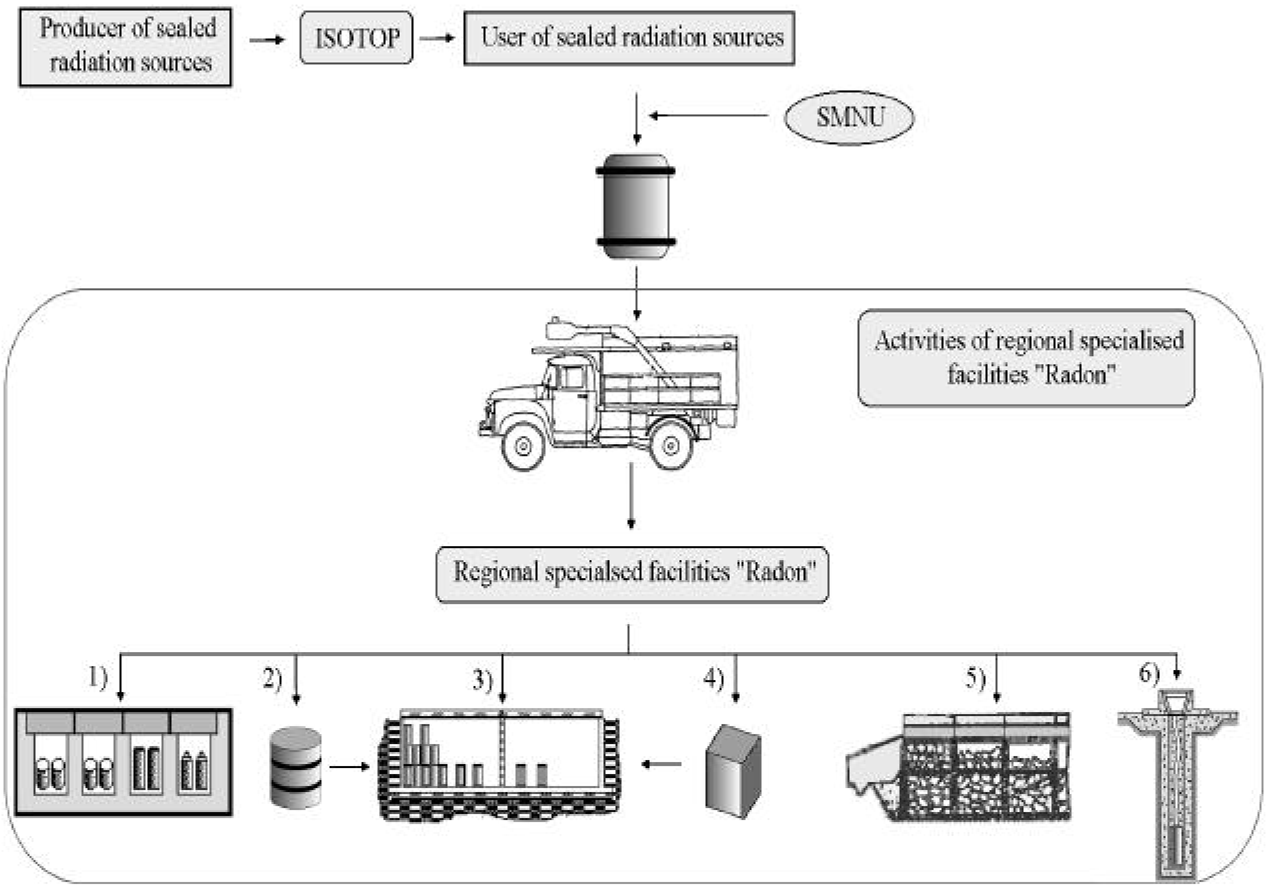

Fig. 1. Schematic of radiation sources management in Russian Federation
1 - interim storage of spent radium-226 sources, 2 - immobilisation of spent sources by concrete in casks, 3 - interim storage of spent sources in containers, casks or capsules, 4 - immobilisation of spent sources by metal in capsules, 5 - immobilisation of spent sources by concrete in shallow-ground solid radioactive waste repositories, 6 - disposal of spent sources in shallow-ground bore-hole repositories

As one can see from Fig.1 the following options are possible for spent sealed sources at regional specialised facilities “Radon”:

- interim storage in containers, casks or capsules,
- interim storage of spent radium sources,
- immobilisation by concrete in casks,
- immobilisation by metal in capsules,
- immobilisation by concrete in shallow-ground repositories for solid radioactive waste,
- disposal of in shallow-ground bore-hole repositories with or without additional conditioning.

Different regional specialised facilities “Radon” applies different methods of spent sources handling. The radiation control is always provided during transportation and handling of spent sources.

TRANSPORTATION

Protective containers are used fort the transportation and storage of sealed beta and gamma-radiation sources. Construction material for these containers is steel, protective material is lead. Sealed radiation sources are loaded into containers by top and unloaded out either by top or bottom. During the transportation the containers additionally are packed up in wooden or metal packages. Wooden or plastic containers are used for the transportation and storage of sealed alpha-radiation sources. During the transportation these containers additionally are packed up in wooden boxes. Neutron sources are transported in steel containers with the paraffin protective material. During the transportation these containers additionally are packed up in wooden or metal packages. Maximal radioactivity loading for containers is 22000 Ci of Co-60 for gamma-radiation source containers and 10^7 n/s for neutron sources containers. Special vehicles are used for the transportation of containers with spent sealed radiation sources to regional specialised facilities "Radon". Vehicles OT-20 provided by stainless steel containers with the volume 3 m^3 are used for transportation of sealed sources.

INTERIM STORAGE

The interim storage of spent sealed radiation sources as a rule is carried out in repositories for solid radioactive waste. The typical repository is an underground reservoir with rectangular cross-section. The reservoir consists of a few boxes. The sizes of these boxes are 6x6x3 m. The cover of reservoir is in the form of concrete slabs with the thickness35 cm. Walls and bottom are monolithic concrete. Thickness of the walls is 25 cm, thickness of bottom is 15 cm. The walls have bitumen water-resist protection. The bottom is situated on reinforced concrete bed impregnated with bitumen. Thickness of the bed is 10 cm. Repository of interim storage of radium sources operates only at the Moscow SIA "Radon".

DISPOSAL

Reinforced concrete units construct walls, bottom and cover of the repository. Thickness of the walls is 60 cm, thickness of the cover is 35 cm. Walls and bottom have water-resist protection, which consists of cement and bitumen layers.

Bore hole type repositories were designed for the disposal of spent sealed radiation sources that are not serviceable for further usage. The design of repository was calculated on the basis of maximum allowable temperature, which is obtained as a result of heat generation by sources. The temperature in the underground steel vessel of the repository is limited by 230°C. The radioactivity loading limit per one repository is 50,000 g-equivalent of radium. The soil near the repository (with a thickness of a few tens cm) is clay or cement-clay mixture, which fill the initial construction bore-hole in the original soil as seal material. The repository was designed in the form of cylindrical vessel with diameter 400 mm and height 1500 mm made of stainless steel. From the top lid of vessel there is a stainless steel tube-loading channel with diameter 108x5 mm curved in the form of a spiral. The maximum dose rate on the surface of repository near the tube, taking into account absorption of radiation in the loading channel and four reflections of radiation at elbows of tube, is 0.82 mR/h.

A special container with upper charging and bottom discharging is provided in order to load borehole 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 SP-349-60. 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.

As a rule at regional specialised facilities "Radon" there are a few typical borehole repositories for the disposal of spent sealed sources (Table II).

Table II. Bore hole repositories at the Regional Specialised Facilities "Radon" in Russian Federation.

Regional Specialised Facilities "Radon"	Number of repositories
Ekaterinburg	2
Habarovsk	2
Irkutsk	1
Moscow	18
Nijnyi Novgorod	1
Novosibirsk	2
Rostov	2
S.Petersburg	3
Samara	2
Saratov	3
Ufa	2
Volgograd	2

In the case when bore hole repositories are not provided the disposal of spent sources is carried out directly in containers into the repository of solid radioactive waste. In case, when it is impossible to unload transportation containers with spent sources, they are deposited directly in containers in shallow-ground repositories for solid radioactive wastes jointly with other solid radioactive wastes and immobilised by concrete. Otherwise spent sealed sources are stored in containers until their disposal.

IMMOBILISATION

In order to ensure long term safety of both stored and disposed sealed sources an additional immobilisation of sources is required. An appropriate matrix material for this aim shall be chosen in dependence of radionuclide used and initial radioactivity. The most suitable matrix for all types of sealed sources is the metal matrix. For example, a lead matrix has a very high corrosive stability: in water lead has corrosion rate less than 5 μ/year. For safe sources immobilisation into an easy melted metal matrix a technology of encapsulation was used at Moscow Scientific and Industrial Association "Radon" since 1986. The spent sources encapsulation into the metal matrix is carried out directly in the underground vessel of repository. This procedure allows following traditional technology scheme of spent sources disposal as well as using the radiation repository-self biological shielding.

Encapsulation technology comprises basically 3 steps:

- pumping out of water
- repository drying
- immobilisation of spent sources

After this the repository can be used again with following fixation of sources into the metal matrix (stage D in the Fig.2). Immobilisation is repeated for every repository until it is filled completely by sources (stage F in the Fig.2).

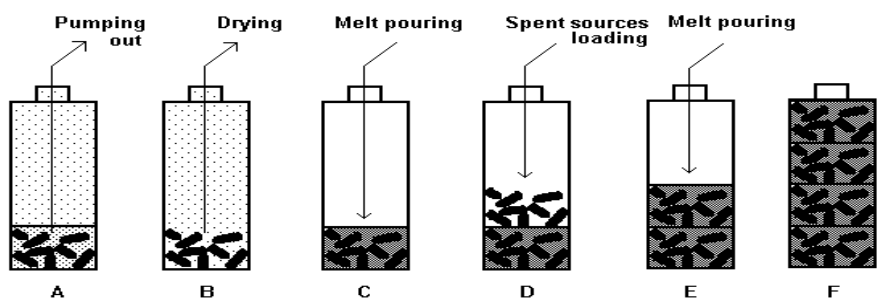

Fig. 2. Immobilisation stages

Researches revealed high safety assets of proposed technology such as radiolysis gases absence, considerable radiation and temperature fields decreasing in the repository and hence, increasing of repository capacity up to 5-6 times.

The necessity to carry out analogous activities at other regional specialised facilities “Radon” caused to develop a universal mobile unit. It allows both to encapsulate spent sources into metal matrix and to perform a range of preparations. Besides, the number and time history of spent sources receipt showed that the use of stationary facility for the purpose is not expedient at every regional disposal site. Following this an industrial mobile unit was designed for immobilising into metal matrices. The unit allows serving disposal sites of one or several regions. The unit consists of several separate technological modules fixed on a transportable truck.

State Supervision Authority and Ministry of Health licensed the immobilisation method and technical documentation for the unit. Such a unit was used for spent sources immobilisation into the metal matrix at Moscow SIA "Radon" since 1986. Besides, since 1991 the unit has been used also at some regional specialised facilities “Radon” (Volgograd, Ekaterinburg, Nizhny Novgorod, and Ufa)

STATUS OF REPOSITORIES

Status of bore hole repositories at Moscow SIA "Radon" was investigated in the beginning of 80-s. Bore hole repositories operate at Moscow SIA "Radon" about 30 years. Some of repositories contain powerful sources. Dose rates up to 20 MR/h were detected in the underground vessels of borehole repositories. Contaminated water, which was accumulated due to condensation on cold walls of loading channel, was observed in repositories. Radionuclides concentration was about $8*10^3$ Bq/l. In some repositories direct inflow of water was observed also. Accumulation of radiolysis hydrogen was observed with concentration up to 3.5 vol.% explosive harmful concentration being 4.2 vol.%.

Investigations of status of borehole repositories at other regional specialised facilities "Radon" in Russian Federation were begun at the beginning of 90-s. A special program was developed by the State Supervision Authority (GAN) in order to clarify the real situation with sources at regional specialised facilities “Radon”.

The main results of examination of repositories status are summarised in the Table III.

Table III. Status of bore hole repositories at the Regional Specialised Facilities "Radon".

	Repository	Beginning of disposal	Number of Sources	Radioactivity, Ci	Filled volume, %	Dose rate in the repository, R/h	Dose rate loading socket, μR/h
Moscow	A (metal matrix)	1979	52	18548	35	120000	60
	B (metal matrix)	1990	992	72996	50	3000000	70
	C (metal matrix)	1979	405	18649	37	170000	50
	D (metal matrix)	1985	1845	73312	84	100000	30
Nijny Novgorod	A (metal matrix)		2325		17	51	15
Rostov	A	1963	283	2808	43	478	15
	B	1963	18801	2505	100	23	26000
St. Petersbourg	A	1963	undefined	10000	undefined	undefined	undefined
	B	1971	undefined	73000	3	1380000	60
Samara	A	1985	3283	883	6	58400	11
Saratov	A	1963	2874	6	13	166	16
Ufa	A	1964	undefined	undefined	undefined	undefined	undefined
	B (metal matrix)	1976	undefined	1400	undefined	782	48
	C	1964	undefined	200	100	6	40
Volgograd	A	1990	undefined	3351	5	36400	5
	B (metal matrix)	1963	undefined	1200	53	6	16
Ekaterinburg	A (metal matrix)	1988	886	837		132	15
	B (metal matrix)	1992	1972	2555		9000	20

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

$$C=1.6*10^{-4}*D, \qquad \text{(Eq. 1)}$$

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 microfissures of 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.

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:

$$t=4.03*10^6*V/(K_g*Q), \qquad \text{(Eq. 2)}$$

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.

In order to ensure safe condition of disposal and long term storage it was proposed to fix sources into a metal matrix. 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. Therefore no contamination of water was observed.

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 the Metallization 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 [3].

CONCLUSION

The conception of disposal of spent sealed radiation sources into bore hole repositories was developed in former USSR to the end of 50-s - beginning of 60-s. Currently bore hole repositories are used at many regional specialised facilities “Radon” in Russian Federation as well as in New Independent States. Investigations of repositories showed insufficient high level of safety for long term storage of powerful sealed radiation sources in bore hole repositories. A technological scheme was proposed in order to correct deficiencies of initial conception with open storage of sources in underground vessels of bore hole repositories. The technology provides fixation (immobilisation) of sources into a metal matrix after their free loading into the bore hole repository. This scheme is also applied for sources, which are stored before their final disposal (long lived radionuclides) in order to ensure safe conditions of long term storage. Now this technological scheme is applied at many regional specialised facilities “Radon” in Russian Federation providing safe conditions for operation of bore hole repositories and long term storage of sources.

The analysis of the work on spent sealed radiation sources management shows that the operating system is able to solve basic problems, providing safe conditions for the environment

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