METAL MATRIX IMMOBILISATION OF SEALED RADIOACTIVE SOURCES FOR SAFE STORAGE, TRANSPORTATION AND DISPOSAL

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ABSTRACT

Metal matrix immobilisation of spent sealed radioactive sources (SRS) is described aiming to ensure safe and secure conditions of storage and to enable consequent retrieval, transportation and disposal of sources into suitable a disposal facility. Metal matrix immobilisation of highly radioactive SRS was developed in the mid 1980s to ensure safe conditions of borehole-type repositories in the former USSR. The immobilisation can be carried out *in situ* utilising the shielding properties of borehole repositories and enhancing operational safety. Lead and lead alloys have been identified as the most suitable materials for spent SRS immobilisation. Immobilisation ensures safe conditions of long-term storage and considerably enhances the level of physical protection of SRS.

INTRODUCTION

Sealed radioactive sources (SRS) have been used extensively in medical, research, industrial and other areas providing indispensable services. Many SRS continue to present a high radiological hazard beyond their design lifetime and need to be safely managed and disposed of to ensure long-term protection of people and the environment [1]. Levels of radiation fields and concentrations of radionuclides from SRS may far exceed high-level radioactive waste from spent nuclear fuel reprocessing so their radiotoxicity may be immense. Safe and secure storage and disposal of SRS currently is a priority in many countries and has focused the attention of the IAEA.

Borehole storage and disposal is one of the most efficient options for spent and disused SRS. More that 40 years of successful experience of borehole storage of high activity SRS in many countries convincingly demonstrates its viability including operational safety and efficiency [2]. Currently, borehole disposal is considered one of the most promising options for SRS including high activity and long-lived sources [3]. Most highly active and long-lived SRS remain in storage pending a suitable disposal option becoming available. Storage can only be considered as an adequate final management option for sources containing short-lived radionuclides, which decay to harmless levels in a few years. High activity and long-lived SRS will after a period of storage be retrieved, transported and disposed of, presumably into deep geological formations, e.g. deep borehole repositories.

However, deep geological repositories are unlikely to become available in the foreseeable future in many countries. Hence the storage times for SRS may be extended and storage facilities ought to provide adequate safety levels over tens of years. Moreover, the security of storage facilities becomes an important issue for SRS, being an attractive target for terrorist attack or as suitable materials for dirty bombs. Thus additional barriers enhancing both safety and security of SRS are of primary importance.

This paper describes the metal matrix immobilisation of SRS aiming to ensure safe and secure conditions of SRS storage and enable subsequent retrieval and re-disposal of SRS into a suitable disposal facility.

CLASSIFICATION OF SRS

The classification of radioactive wastes in the Russian Federation is based on the nature and concentration of radionuclides [4]. There are 3 categories of solid and liquid radioactive wastes as specified in Table I:

Table I Classification of radioactive waste in Russian Federation

Category of Waste	Specific radioactivity, kBq/kg		
	beta	alpha	transuranic
Low Level	$< 10^3$	$< 10^2$	10
Intermediate Level	$10^3 <...< 10^7$	$10^2 <...< 10^6$	$10 <...< 10^5$
High Level	$> 10^7$	$> 10^6$	$> 10^5$

Typical SRS having 10g weight of active part is categorised as high level waste (HLW) if its total activity is higher than 100 MBq for beta, >10 Mbq for alpha and >1 MBq for transuranic radionuclides. Hence many spent SRS, accordingly to the Russian regulatory document, belong to the category of HLW, despite not having arisen from nuclear fuel reprocessing.

SRS have always treated separately from other radioactive wastes due to the enhanced hazard that they represent in many aspects similar to HLW. However, in contrast to HLW, a certain number of SRS have in the past been disposed of in shallow ground repositories. Consequently the status of some of these inadequate disposal facilities was changed to a long-term storage, so at some point in the future these SRS will need to be retrieved for disposal into a suitable geological formation.

The current waste management scheme (Fig. 1) also provides separate processing of spent SRS from other radioactive wastes. This scheme comprises metal matrix immobilisation of SRS followed by long term storage.
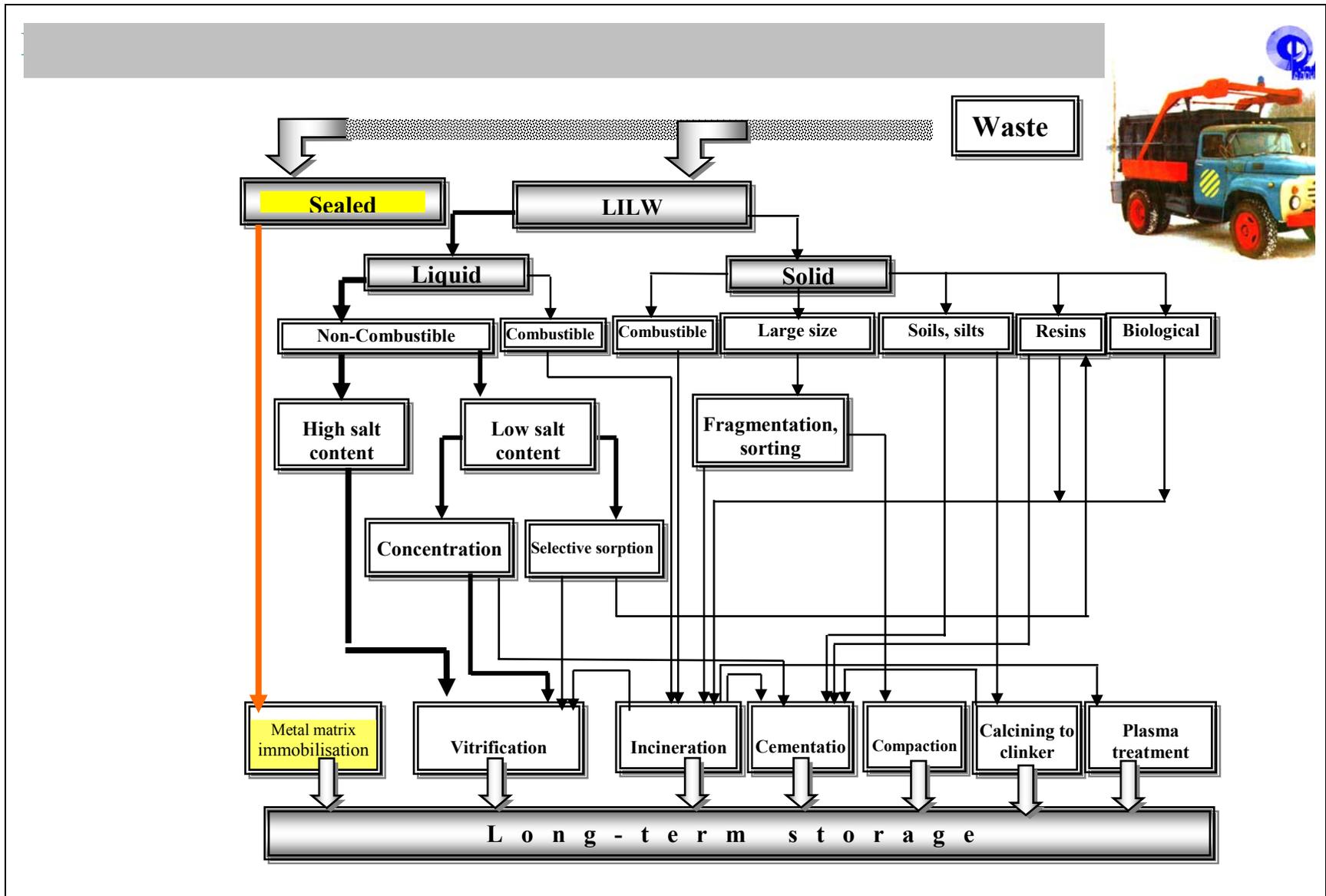
Fig. 1 Schematic of radioactive waste processing

POTENTIAL HAZARD OF SRS

The index of radiotoxicity (or hazard) of nuclear waste is the sum over all toxic constituents of waste [5-8]:

$$I(t) = \sum_i \frac{C_i(0)\exp(-\lambda_i t)}{IL_i} \Phi_i \qquad\qquad \text{(Eq. 1)}$$

where $C_i(0)$ is the initial concentration of the *i-th* radionuclide (Bq/m^3), IL_i is the intervention level (or maximum permitted concentration), that can be taken from national regulatory documents (Bq/m^3), λ_i is the decay constant (1/s), Φ_i is the released inventory fraction, which is dimensionless and accounts for the fraction of radionuclides released from the waste form to the environment. The inverse of the released inventory fraction Φ_i^{-1} is the retention coefficient or retardation factor [7, 8]. For an aqueous solution (liquid waste) obviously $\Phi_i = 1$, whereas durable waste forms hold $\Phi_i \ll 1$. Most spent SRS certainly have low values of $\Phi_i < 1$, but some, especially those with active parts in the form of ready soluble salts, can have $\Phi_i \to 1$. A typical SRS containing ~1 Ci of ^{137}Cs in a volume ~1 cm^3 has the initial concentration $C_i(0)$ ~10^3 Ci/l, whereas typical HLW has lesser typical concentrations ~1-10 Ci/l. Hence the potential hazard of SRS can be similar or even higher that of HLW.

Consider a SRS containing the active part in form of water soluble salts as ^{137}CsCl or ^{137}CsBr with a damaged case. For such sources we can assume $\Phi_i \to 1$ in (1). The intervention level for ^{137}Cs is IL=11 Bq/dm^3 according to [9]. Hence this SRS supposing a damaged case will have the index of toxicity $I(0)$~3.4 10^{12}, meaning the potential of contaminating 3.4 million cubic metre of drinking water.

The index of toxicity (1) diminishes with time due to natural decay of radionuclides, however for extended periods of time $I(t) \gg 1$, which shows the potential hazard of contamination. Times T_i needed to isolate an SRS from the environment can be assessed assuming its radioactivity $A_0 \exp(-\lambda_i t)$ becomes below the exemption activities EA_i (Bq), where A_0 is the initial SRS radioactivity (Bq). This gives the required retention (storage or disposal) time T_i (years) in terms of half life of radionuclides $T_{1/2,i}$ (years):

$$T_i = 1.44\, T_{1/2,i}\, \ln\left(\frac{A_0}{EA_i}\right) \qquad\qquad \text{(Eq. 2)}$$

If $t > T_i$ the SRS is exempt from regulatory control and is not considered as a radioactive waste. Values of EA_i can be taken from national regulatory documents. Table II shows for radionuclides most frequently used in SRS numeric values of $A_{exe,\,i}$ and intervention levels IL_i according to [9].

Table II   Exemption activities and intervention levels of some radionuclides used in SRS.

Radionuclide	Half life $T_{1/2}$, years	Exemption activities EA_i, Bq	Intervention levels IL_i, Bq/kg
^{3}H	12.3	10^9	7.7 10^3
^{60}Co	5.27	10^5	41
^{90}Sr	28.6	10^4	5
^{137}Cs	30.2	10^4	11
^{226}Ra	1600	10^4	0.5

Hence a typical SRS with ~1 Ci ^{137}Cs will become exempt in Russia after a period of time exceeding ~653 years. Figure 2 shows the retention times for SRS containing short-lived ^{3}H or ^{60}Co, and long-lived

[226]Ra radionuclides depending on the initial level of radioactivity. Required periods of isolation are of the order of several tens- hundreds of years for typical short-lived SRS. Much longer times (proportional to half life) are required for SRS containing long-lived radionuclides.
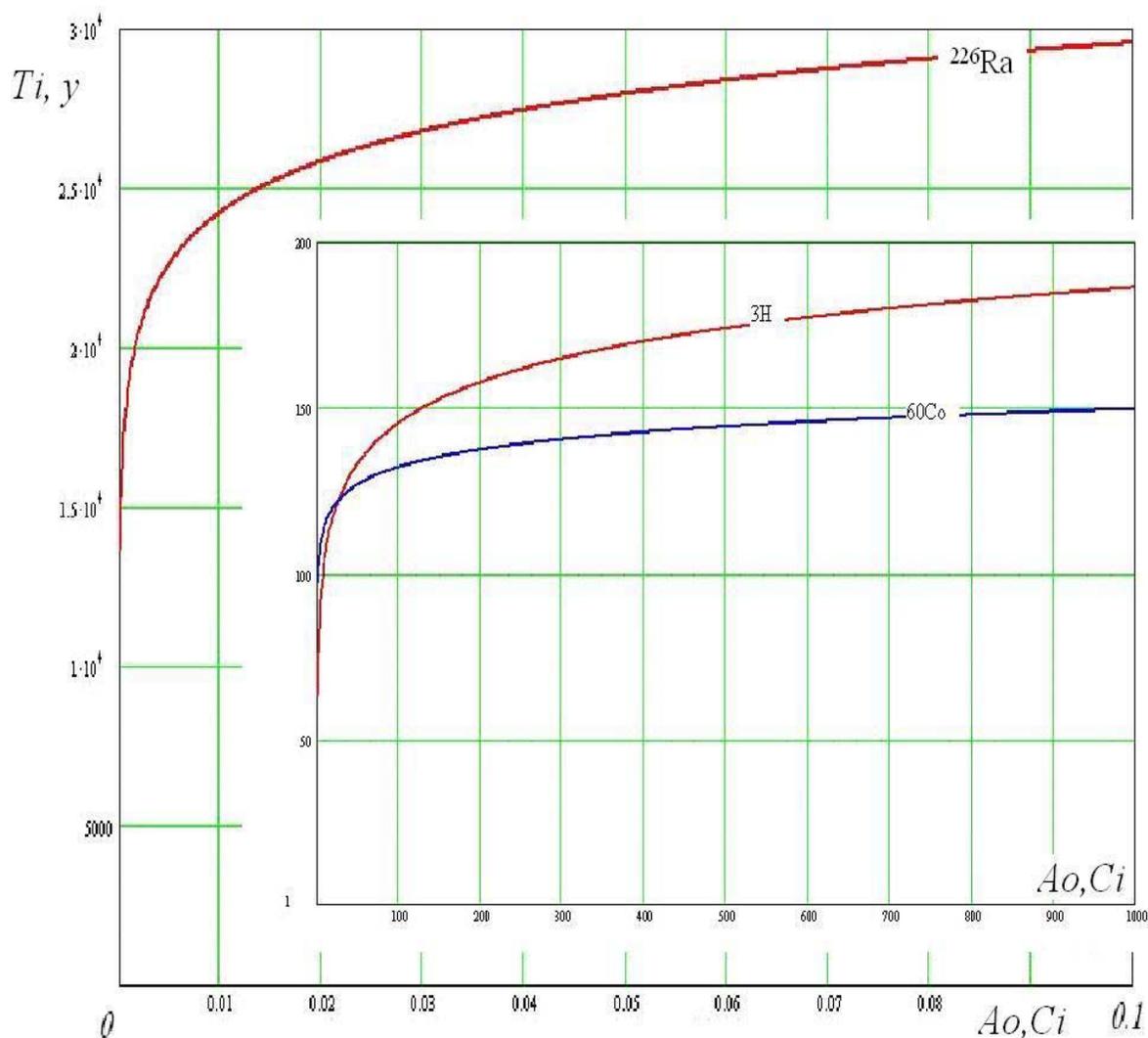

Fig. 2　Necessary retention Times for SRS to achieve exemption Activities [9] as a function of initial radioactivity.

Safe storage or disposal for such extended periods of time requires durable immobilising materials. Spent SRS often have damaged or weakened cases. Even SRS with undamaged cases are not able to survive in a corroding medium for hundreds of years. SRS will sooner or later leak resulting in $\Phi_i \rightarrow 1$ and high potential for contamination of the environment. A sensible option is to place the SRS in a durable matrix able to resist the environment for the necessary storage or disposal time.

Immobilisation of SRS in a matrix material not only reduces the environmental contamination hazard but also diminishes the dose rates of ionising radiation. The reduction of contamination hazard is accounted for in (1) by the coefficient Φ_i.

Consider a SRS which consists of an active part enclosed in a protective case of thickness d and suppose the source is placed in an immobilising matrix with wall thickness L. There are a number of barriers preventing the release of radionuclides from the source: (1) the form of the active part, which can be a soluble or insoluble salt, enamel, ceramic, glass or metal, (2) the case, which usually is stainless steel, (3) the matrix, which is lead or a lead based alloy. All of these barriers play their role. The active part ensures retention of radionuclides until case failure. The case ensures complete retention of radionuclides during the operational life-time of SRS. The matrix ensures retention of radionuclides during extended periods of storage, transportation and potential disposal.

The index of toxicity (or hazard) of immobilised radioactive wastes including SRS can be calculated from (1) accounting for the correct values of released inventory fraction from the immobilising barriers. To calculate the value of Φ_i it is necessary to know the values of released inventory fractions for every barrier. A comprehensive method of calculating release of radionuclides using computer-based codes from immobilised radioactive wastes was developed by Sullivan *et.al.* [10, 11]. We use herein a similar estimation method to assess the value of summary released inventory fraction from a SRS.

The resulting released inventory fraction is:
$$\Phi_i = \Phi_{ai}\, \Phi_{cs}\, \Phi_m \tag{Eq. 3}$$

where Φ_{ai}, Φ_{cs} and Φ_m are released inventory fractions of the form of the active part, case and immobilising matrix respectively.

The active material may be a water soluble salt, in which case $\Phi_{ai} = 1$. If the active part is a metal, Φ_{ai} can be estimated based on the corrosion behaviour. For a steel we have:
$$\Phi_{ai} = (S/V)D_o t^a, \tag{Eq. 4}$$

where D_o is a coefficient (m/y^a), (S/V) is the active part surface to volume ratio (1/m), $a=0.61\pm0.03$ [12].

If the active part is a glass the released inventory fraction can be assessed from [13]:

$$\Phi_{ai} \approx \frac{S}{V}\left[\left(\frac{D_i}{\lambda_i}\right)^{1/2} + \frac{r_c}{\lambda_i} + \frac{n_i k_i}{k_i + \lambda_i}\right], \tag{Eq. 5}$$

where (S/V) is the active part surface to volume ratio (1/m), D_i is the diffusion coefficient (m^2/s), r_c is the glass dissolution rate (m/s), k_i is the rate of instantaneous dissolution from the surface (1/s), n_i is the initial relative surface concentration (m): $n_i = n_{si}/C_i(0)$, and n_{si} is the initial surface concentration of radionuclides (Bq/m^2).

The released inventory fraction of the metallic case and immobilising matrix are given by formulae:

$$\Phi_{cs} = \Theta(t-\tau_{cs}), \quad \Phi_m = \Theta(t-\tau_m), \tag{Eq. 6}$$

where $\Theta(t)$ is the Heavyside function ($\Theta(t)=1$ if $t>0$ and is 0 otherwise), τ_c (y) and τ_m (y) are the retention times for the casing material and immobilising matrix respectively. These can be found from the dependences of corrosion depth with time: $D=D_o t^a$ [12]:

$$\tau_{cs} = \left(\frac{d}{D_{ocs}}\right)^{\frac{1}{a_{cs}}} \qquad \tau_{cm} = \left(\frac{d}{D_{om}}\right)^{\frac{1}{a_m}} \qquad\qquad\qquad\qquad\qquad \text{(Eq. 7)}$$

where D_{ocs} ($m/y^{a_{cs}}$), D_{om} (m/y^{a_m}), a_{cs} and a_m are the coefficients of case and immobilising metallic materials respectively.

Substituting (3) in (1) we can see that the contamination hazard is nil until the most durable immobilising barrier is entirely corroded. The retention time for the casing material τ_c cannot be longer than several tens of years. By choosing a low corrosion rate immobilising material one can ensure long retention times τ_m for the matrix material and hence practically zero hazard of contamination from an immobilised SRS. For example, a lead immobilising matrix which has extremely high corrosion resistance ensures a minimum 500 - 1000 years of complete isolation in a near surface repository [14].

METAL MATRIX IMMOBILISATION

Metal matrix immobilisation technology was developed in the mid 1980s to ensure safe conditions of borehole-type repositories in the former USSR. These have been constructed at 35 regional centralised facilities for radioactive waste management using the "Radon" system which in many aspects is similar to the system of compacts in the USA. At this time borehole repositories were considered as facilities for the disposal of short-lived SRS however after 1990 practically all borehole repositories both in Russia and other independent states changed the status of disposal to storage. The new borehole-type repositories currently deployed provide the possibility of retrieving the SRS. Hence many of the highly active SRS, which are currently being stored, will eventually be retrieved, transported and disposed of in dedicated geological repositories providing the necessary level of safety.

The Russian Federation uses borehole type repositories for SRS at 11 Regional Specialised Facilities "Radon" since 1963. SRS are stored underground in stainless steel cylindrical vessels located in steel-reinforced concrete wells which are the main parts of a typical borehole repository. Special containers are used to transport SRS to borehole repositories. These provide for upper source loading and bottom unloading from the container. The SRS loading channel into the repository has the form of a spiral tube (Fig. 3) . A conical socket provides safe loading of SRS into repositories from the transport containers. The capacity of typical borehole repositories corresponds to an equivalent of 50 kg of ^{226}Ra. The maximum dose rate in the socket near the SRS loading channel does not exceed 8.2 µGy/h.

Metal matrix immobilisation is a well-known method of nuclear waste immobilisation used for many years, e.g. metallic containers, capsules, *vitromet* at PAMELLA HLW vitrification plant. This method has also been recommended for SRS immobilisation [15]. Other materials including glasses are not able to withstand the enhanced radiation doses and heat generation from highly radioactive sources. Investigations of the possibility of using lead as a matrix for immobilisation of spent fuel and high-level radioactive waste shows that lead holds much promise for radioactive waste disposal [16-18]. The lead is suitable as a SRS immobilising matrix due to its: high radiation stability, ability to withstand deformation without loss of integrity, high corrosion resistance, high heat conductivity, effective filling of free space between sources, compatibility with other construction materials. Lead also does not contaminate groundwater due to its high passivity and low corrosion rate. A number of investigations assessed the impact of lead from the disposed wastes and demonstrated no impact of the lead onto environment [18-20]. This is an expected result that results from the high corrosion resistance of lead in groundwater.

SRS immobilisation involves two steps. In the first stage a large volume of molten metal is poured over the sources in an underground repository vessel. Owing to their lower density the sources rise to the

surface of the melt, and it solidifies they are fixed in the metal. In the second stage of the technological process a thin layer of melt is poured on the surface of the metal block. The structure of the resulting block and coupling between the layers are controlled by the use of non-destructive testing (ultrasonic defectoscopy) and mechanical testing of the samples for fracture. These revealed that some aluminium and tin matrix samples had significant defects at the boundaries between layers. This was caused by rapid oxidation the reactive metal surface. However lead and lead-based alloys showed no such effects.

If spent SRS contain highly volatile radionuclides it is essential to keep the metals melting temperature as low as possible but at the same time to provide reliable cohesion between matrix metal layers. To achieve this, the SRS are enveloped in a low-melting lead-based alloy. Upon crystallization of the melt the above technological operations above-mentioned are performed. Ultrasound analysis of the resulting block shows no structural defects in the block.

Table III shows SRS used immobilising metals including alloys with melting temperatures below 100 ^{0}C. These can be used for spent SRS immobilisation buried in repositories flooded by water. For this purpose the sources are initially surrounded by low-melting alloy and if required, water in the repository can be heated to a temperature above the alloys melting temperature. Owing to significant density differences, the water or water-clay suspension is displaced from sources by the melt. Upon pumping out the water and drying the repository, final sealing is achieved by successive pouring of several layers of molten lead.

Table III   SRS immobilising metals and alloys.

Composition, % wt.	Al	Pb	Sn	32Pb68Sn	50Bi22Sn28Pb	50.1Bi14.6Sn 24.5Pb10.8Cd
Melting temperature, ^{o}C	660	327	234	177	100	66

A mobile facility has been developed for SRS immobilisation using metal matrices [21]. This facility allows traditional SRS disposal technology to be used and SRS immobilisation directly in an underground vessel in a repository (Fig. 3). The facility contains a joining unit and a gas purification system, which insulates the inside of the repository from the atmosphere. In addition, maintaining reduced pressure eliminates the possibility of radionuclide release into the environment during immobilisation. Matrix metal melts are prepared outside of the repository in a special technological unit. So the duration of thermal impact on spent SRS is minimised. The prepared melt is fed into the repository by a flexible heat-resistant hose lowered through the SRS loading channel of the borehole. The existing biological protection of the repository functions as an ionizing radiation shield, ensuring an enhanced degree of operational safety.
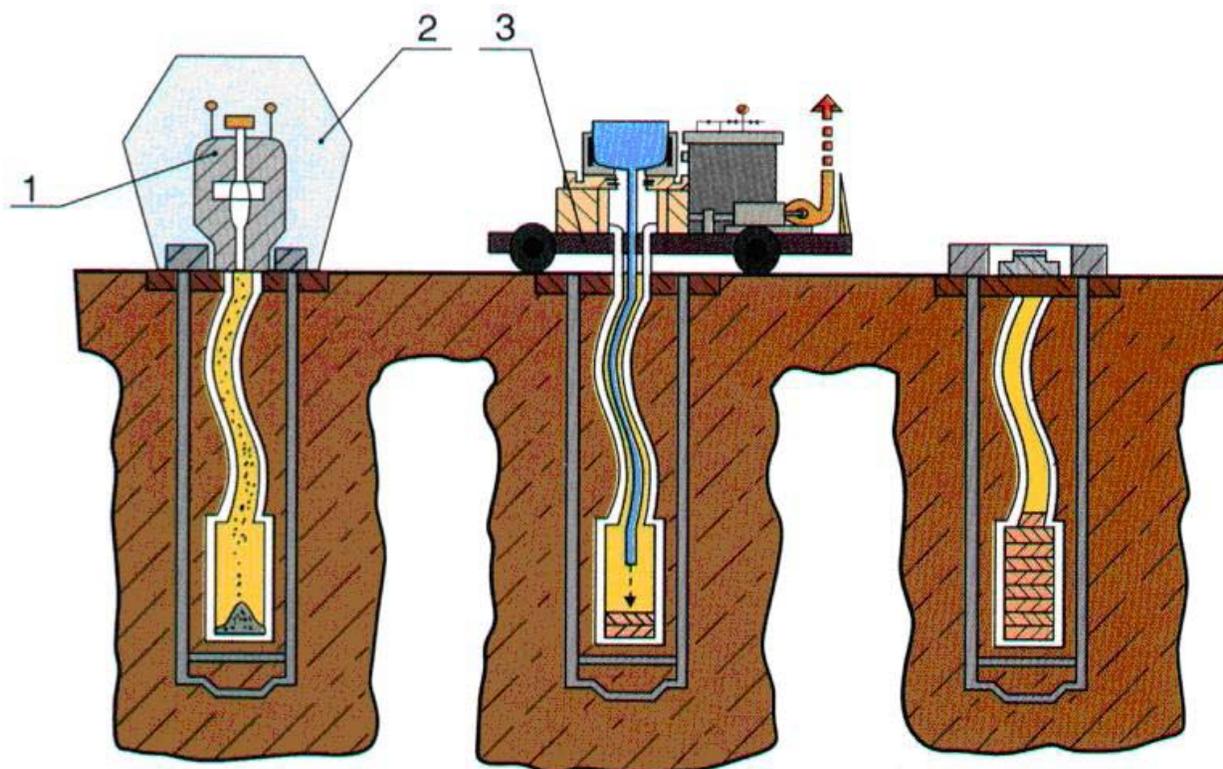
Fig. 3   Schematic of *in situ* metal matrix immobilisation of SRS. 1 – transport container, 2 – gas purification unit, 3 – technological unit.

Metal matrix immobilisation technology has been used by Moscow SIA RADON to immobilise SRS since 1986. From 1990 a new technique has been used at the regional centres of radioactive waste processing such as Volgograd, Nizhny Novgorod, Ufa and Ecaterinburg regional repositories and at Novovoronezh NPP. To date spent sealed sources with total radioactivity over 1 million Ci have been encapsulated into metal matrices [2].

Recently metal matrix immobilisation has been used in Belarus to prepare SRS for retrieval and disposal. Other countries also regard metal matrix immobilisation as a potential option [22, 23]. In addition metal matrix immobilisation is an excellent method to prepare long-lived SRS for safe storage and disposal [24, 25].

POTENTIAL RETRIEVAL OF SRS

Highly active SRS, which require long periods of isolation (equation (1)) exceeding reasonable periods of institutional control, will eventually be retrieved, transported and disposed of in dedicated geological repositories providing the necessary level of safety. The retrieval of SRS requires its preliminary immobilisation to ensure safe conditions for technological operations and transportation. Metal matrix immobilisation also provides safe conditions for transportation and considerably enhances the level of physical protection of SRS.

Immobilisation of SRS enables safe retrieval after reasonably short-term storage periods which are significantly less than the decay time necessary to achieve exemption levels. Assessment of this time has been carried out assuming dose rates less than 2mSv/h on the surface of immobilising metal blocks. Figure 4 shows the required delay time to achieve either exemption or safe radiation dose rate levels on the surface of metallic blocks (right) as a function of activity of ^{60}Co SRS. The delay time to achieve safe

handling conditions, e.g. 2mSv/h, is an order of magnitude less than the time required for free release of SRS. Hence the retrieval in contrast to exemption can be carried out within institutional control time.
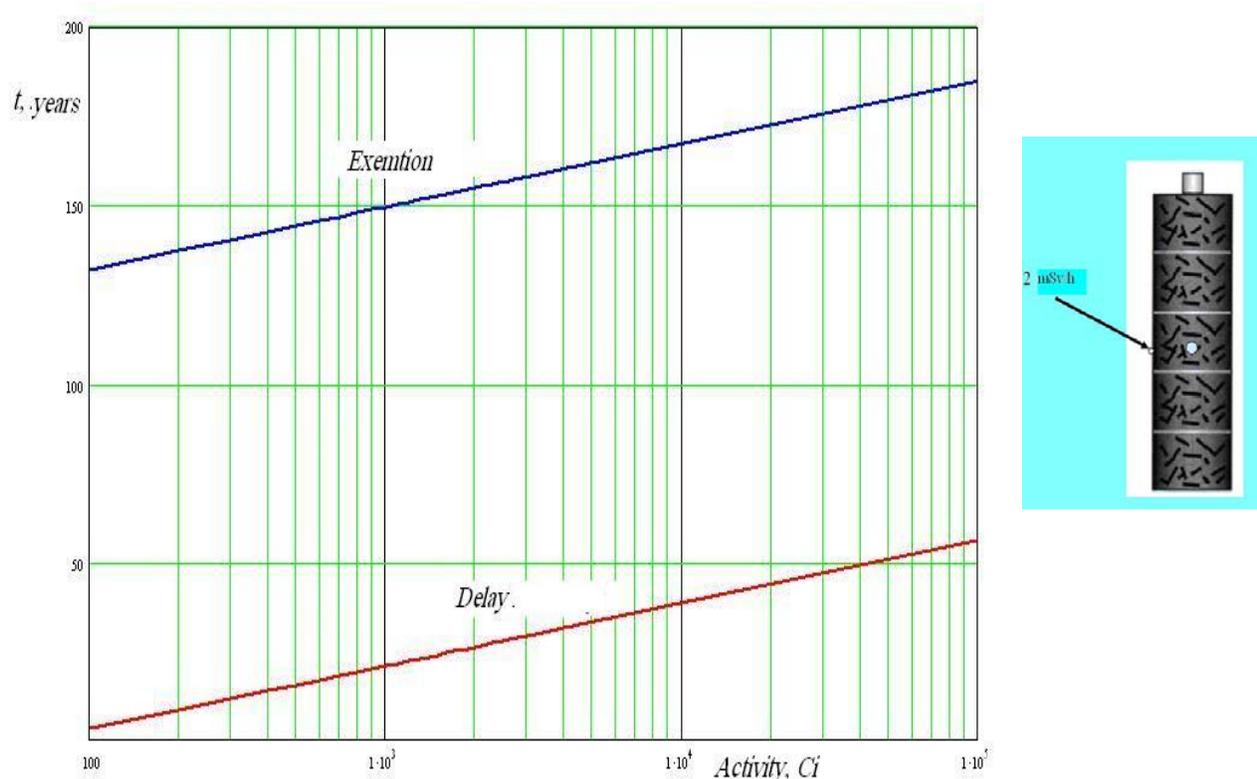

Fig. 4   Necessary Delay Time to Achieve Safe Dose Rates at the Surface of SRS Immobilising Metal Blocks as a Function Of SRS Activity.

CONCLUSIONS

Metal matrix immobilisation enhances both the safety and security of spent SRS and facilitates consequent retrieval and final disposal. The immobilisation can be carried out *in situ* utilising the shielding properties of borehole repositories and enhancing operational safety. Lead and lead alloys have been identified as the most suitable materials for SRS immobilisation. Metal matrix immobilisation has been licensed and used in the Russian Federation since 1986. Metal matrix immobilisation has been recently used in Belarus to prepare SRS for retrieval and disposal. Other countries also use or consider metal matrix immobilisation as a potential option. Experience of metal matrix immobilisation demonstrates its high efficiency at providing safe conditions for long-term storage, retrieval, transportation and re-disposal of SRS.

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