FEASIBILITY OF VERY DEEP SELF-DISPOSAL FOR SEALED RADIOACTIVE SOURCES

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ABSTRACT

We analyse the feasibility of self-disposal of spent sealed radioactive sources (SRS) via the "self-burial" technique. We suggest acoustic emission tracking of the self-descending capsules loaded with powerful SRS. Detected signals could also provide information about the deep interior of Earth that is currently unavailable via drilling and other techniques.

INTRODUCTION

Sealed radioactive sources (SRS) have been used extensively in medical, research, industrial and other areas providing indispensable services. For example in the USA each year about 200,000 patients receive radiotherapy most commonly from SRS for treating cancers [1]. The worldwide inventory of SRS is very large. For example, in the European Union about 500,000 SRS have been sold, most of which remain in use or in store [2]. The inventories of SRS in other countries are also very large with no tendency to diminish [3-6]. Many of the sources are characterized by very high levels of radioactivity, e.g., ^{60}Co sources used in sterilisation facilities have activity levels up to 2 PBq during service lives from 3 to 10 years [2]. The decay of radionuclides in SRS diminishes their high levels of radioactivity insufficiently for subsequent safe handling and disposal. Thus most SRS continue to present a high radiological hazard beyond their design lifetime and need to be managed and disposed of safely to ensure the long-term protection of people and the environment. As the radiotoxicity of SRS is very high they require special and expensive procedures for safe and secure storage and most highly active and long-lived SRS remain there pending a suitable disposal option becoming available [7]. One reliable method of ensuring safe and secure long-term storage of SRS is utilisation of metal matrix immobilisation [8]. Immobilisation of SRS in lead and lead based alloys has been used in Russia since 1986 [9] and in Belarus since 2003 [10] while other countries such as Germany, Italy and Portugal are considering or plan to use metals for the encapsulation of sealed sources [2]. However storage can only be considered as an adequate final management option for sources containing short-lived radionuclides that decay to harmless levels in a few decades. After a period of storage, high activity and long-lived SRS will have to be retrieved, transported and disposed of, presumably into deep geological formations, e.g., in deep borehole repositories [7, 11].

Because deep geological repositories are unlikely to become available in the foreseeable future alternative disposal options for SRS are being considered in many countries. The most secure disposal options for highly radiotoxic wastes rely on very deep disposal into the Earth's crust. For example, the concept developed by Gibb [12, 23] examines borehole disposal of the most toxic wastes at depths exceeding 4,000m. Moreover this concept utilises the radiogenic heat of radioactive waste to self-seal the waste into re-melted granite. Another variant of Gibb's very deep disposal concept seeks to use geological pressures to facilitate waste self-sealing by sintering of powder-like disposed materials [13]. Even deeper disposal of radiotoxic waste could be achieved using the radiogenic heat to melt the rock to provide a self-descending mechanism for waste capsules, e.g. the "deep self-burial" concept first proposed by Logan [14, 15]. The purpose of this paper is to analyse the feasibility of self-disposal of SRS via the "deep self-burial" technique and, in addition, suggest acoustic emission tracking of the self-descending capsules.

High Risk Sources

At the end of their operational lifetime powerful SRS containing large amounts of radionuclides retain elevated levels of both radiation and radiotoxicity, thus representing the highest hazards. Table I shows typical data on highest risk SRS giving typical working life times.

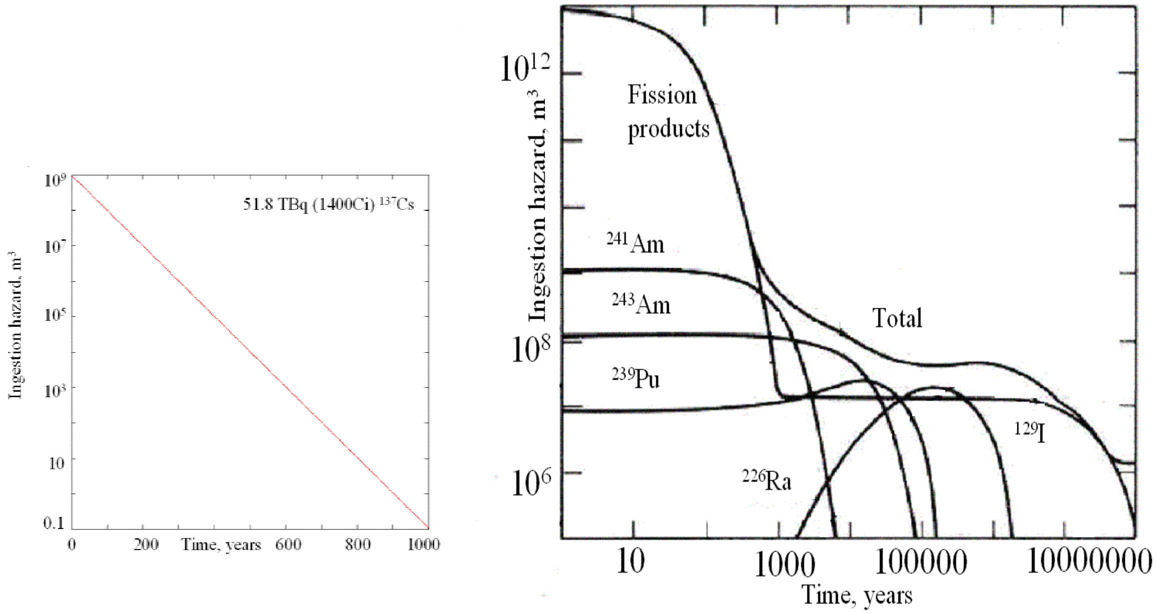
Table I. Typical Parameters of Powerful SRS.

Radionuclide	Uses	Activity	Half-life, y	Working life, y
^{60}Co	High energy radiography	37GBq-8TBq	5.27	5-15
^{60}Co	Gauging. Control of petro-chemical, chemical, coal processes.	370MBq-20GBq	5.27	5-15
^{60}Co	Industrial sterilization.	74TBq-2PBq	5.27	3-10
^{90}Sr	Thermoelectric generators	Up to 30PBq	28.5	
^{137}Cs	Process control in chemical plants	370MBq-100GBq	30.17	10-20
^{137}Cs	Medical	370MBq-8GBq	30.17	5-15
^{238}Pu	Thermoelectric generators	Up to 10TBq	87.74	tens of years

As the working lives do not significantly exceed the half-life (^{60}Co) or are less than the half-life (^{90}Sr, ^{137}Cs, ^{238}Pu), all SRS still contain extremely high amounts of radionuclides at the end of their operational period. The hazard that an SRS poses to the environment, other than from its radiation field, is given numerically by its radiotoxicity index. The radiotoxicity index of nuclear waste, including spent SRS, is the sum of all the toxic constituents of the waste [9]:

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

where $C_i(0)$ is the initial activity (Bq/m^3) for isotope i, IL_i is the intervention level (or maximum permitted activity) (Bq/m^3), λ_i is the decay constant (1/y), t is time (y), Φ_i is the released inventory fraction, which is dimensionless and accounts for the fraction of radionuclides released from the wasteform to the environment. For an aqueous solution $\Phi_i = 1$, whereas durable wasteforms have $\Phi_i \ll 1$. Most spent SRS certainly have values of $\Phi_i < 1$, but some, especially those with active components in the form of readily soluble salts, can have $\Phi_i \to 1$. Consider for example a typical SRS containing ~37GBq (1Ci) of ^{137}Cs in the form of water soluble salts such as ^{137}CsCl or ^{137}CsBr in a volume ~1 cm^3 with a damaged case. For such sources we can assume $\Phi_i \to 1$ in (Eq.1). The intervention level for ^{137}Cs is IL=11 Bq/L [9]. Hence this damaged SRS will have a toxicity index I(0)~3.4 10^{12}, and the potential to contaminate 3.4 million cubic metres of drinking water. Fig.1(a) shows the ingestion toxicity of the ^{137}Cs SRS involved in the 1987 radiological incident in Goiania, Brazil [16]. Fig.1(b) gives for comparison the ingestion radiotoxicity of high-level waste (HLW) produced after reprocessing of spent nuclear fuel from 1 year's operation of a 1GW(e) nuclear power plant (NPP).



(a) (b)

Fig. 1. Ingestion hazard of ^{137}Cs SRS (a) and HLW from 1 GW(e) NPP (b) (adapted from [17]).

Fig. 1 indicates the significant magnitude of hazard that a powerful SRS presents to the environment. It can be seen that one Goiania type SRS has almost the same radiotoxicity as that of all actinides in the HLW produced by a typical NPP after one year. Moreover it takes many hundreds of years for the radiotoxicity of this source to decrease enough for a near-surface disposal. Near-surface facilities are inadequate for the disposal of powerful or long-lived SRS. In contrast, deep geological disposal, such as in very deep boreholes, could provide an adequate isolation of high risk SRS [7].

Assessment of Self-Burial Parameters

Very deep borehole repositories potentially provide the highest degree of safety for highly radiotoxic wastes [11, 12]. Very deep disposal however is not cheap and the deeper the borehole the higher the drilling cost., This may make very deep disposal economically difficult despite the safety achieved except when large numbers of SRS are involved. It should be noted, however, that the relatively small size of SRS would allow use of quite small diameter (and hence less expensive) boreholes and one hole would suffice for very many SRS.

A possible option for access to even deeper layers of the Earth than by boreholes is offered by the "deep self-burial" concept [14, 15, 18-21]. This is based on utilization of the heat generated by the decay of radionuclides but requires sufficiently high concentrations of radionuclides in the waste. Hence self-burial would be most suitable for high heat-generating wastes, such as powerful SRS. (For example, a 2PBq SRS from an industrial sterilization facility provides ~0.8 kW heating power.)

Consider parameters of a self-descending capsule designed to penetrate to very deep in the Earth's crust for self-burial disposal of SRS. The capsule must provide containment of the SRS radionuclides and melt surrounding rocks on its way. It must therefore be constructed from highly–refractory, mechanically strong and radiation durable material(s). It also needs to resist corrosive and erosive destruction in silicate melts at significant, and increasing, lithostatic pressures. Ceramic capsules, although durable, demonstrate significant overheating patterns that limit their heat loading and thus descent velocities [22] and ultimate depths. Metal capsules, on the other hand, would give an almost homogeneous temperature distribution and enable efficient utilization of radiogenic heat.

Self-descent of a spherical body in a melting environment has been considered many times in the contexts of self-burial of radioactive wastes and analyses of the potential nuclear reactor core meltdown (the so-called "China syndrome") [14, 15, 18-22]. Following the results of previous works we examine here the self-burial of a metal capsule of radius R (m) which has an almost homogeneous temperature distribution on its surface.

Let us assume the specific gravity of the loaded capsule, ρ_c, is higher than the specific gravity of the partly melted surrounding rock, i.e., $\rho_c > \rho_m$. If the total power of the heat sources in the capsule = Q (W), then the specific heat power q (W/m^3) =3Q/4πR^3. The capsule partly melts the surrounding rock and descends until its specific heat power is higher than the threshold q_{th} determined by Logan's ratio [14]:

$$q > q_{th} = 3\chi(T_m - T_r)/R^2\,, \qquad \text{(Eq.1)}$$

where χ is the heat conductivity (W/m K), T_m is the melting temperature and T_r is the temperature of the rock far from the capsule (K). It is appreciated that, in reality, rock does not have a single melting temperature but melts over a range between its solidus and liquidus temperatures. However, as a first approximation using a value somewhere between the two is adequate. The heat power generated by decaying radionuclides diminishes with time

$$q(t) = q(0)\exp(-\lambda t)\,, \qquad \text{(Eq.2)}$$

where q(0) is the initial specific heat power (e.g. $q(0)>q_t$), λ is the decay constant = $0.693/T_{1/2}$ and $T_{1/2}$ is the half-life of radionuclides (y). Thus the time of continuous descent, τ (years), can be found by equalizing the actual specific heat power to the threshold $q(\tau)=q_{th}$, which results in the equation:

$$\tau = 1.44T_{1/2}\ln q(0)R^2/3\chi(T_m - T_r)\,. \qquad \text{(Eq.3)}$$

At $t > \tau$ the capsule does not move having achieved its maximum possible depth of penetration $H(\tau)$. $H(\tau)$ (km) can be determined from the equation:

$$H(\tau) \approx 1.9[1 - q_{th} / q(0)]T_{1/2}Rq(0) / \rho_m[L + c_p(T_m - T_r)] \quad \text{(Eq.4)}$$

where ρ_m is the average rock specific gravity, c_p is its heat capacity and L is the heat of fusion of the rock. Thus from initial capsule parameters {R, q(0), $T_{1/2}$} one can determine the depth attained by self-burial.

Self-Burial for Powerful SRS

To carry out self-burial of powerful SRS they could be collected in standard transport containers then re-loaded in the necessary amounts into pre-fabricated spherical capsules made of a refractory metal such as tungsten. Tungsten would be a suitable capsule material due to its high-temperature properties (melting temperature = 3410^{o}C), high specific gravity (19.3 g/cm^3) and (presumed) low corrosion rate in silicate melts. The capsule should have thick enough walls to withstand corrosion damage during sinking: $d > r_{cor}\tau$, where d is the thickness of the walls and r_{cor} is the corrosion rate of the material in partly melted rock. Corrosion of different materials in rock melts under conditions similar to those at very high depths was recently studied in connection with the concept of very deep nuclear waste disposal [23, 24]. Stainless steel for example demonstrated corrosion rates lower than 100μm/year at temperatures around 800°C [24]. The walls of the capsule should also be thick enough to ensure efficient absorption of the radiation emitted by decaying radionuclides and hence an efficient heating of the capsule. Thicknesses in excess of 10 cm are required for this.

Capsules will be heated by the radiogenic heat to high enough temperatures to begin melting the surrounding rock [23] and, when the melt fraction reaches a critical value. The capsule will begin to sink through the partial melt due to its higher specific gravity (Fig.2).

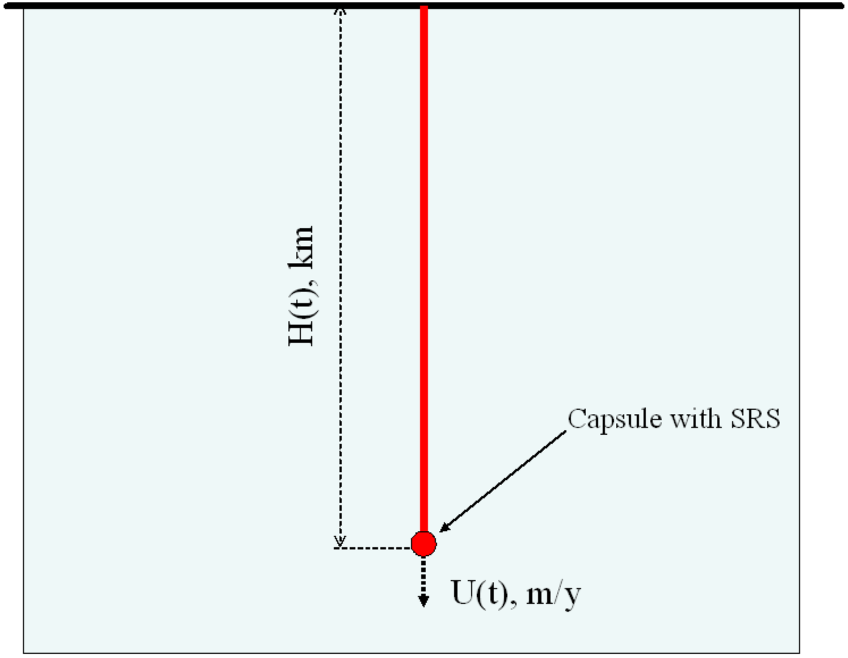


Fig. 2. Schematic of self-burial of capsules containing powerful SRS. U(t) is the velocity of sinking, H(t) is the depth of penetration (burial).

Table II shows estimated self-burial parameters for tungsten capsules filled with SRS containing ^{60}Co radionuclides at two loadings. The following characteristic parameters from works [14, 15, 19-22] were used in our estimations:

- average rock density ρ_m=2.7 g/cm^3,
- heat capacity c_p=1 J/g K,
- heat of fusion L=418 J/g,
- heat conductivity χ=0.01 W/cm K,
- melting temperature T_m =1200°C,
- temperature far from capsule T_r = 20 °C.

The average capsule density was taken ρ_c=12.7 g/cm^3.

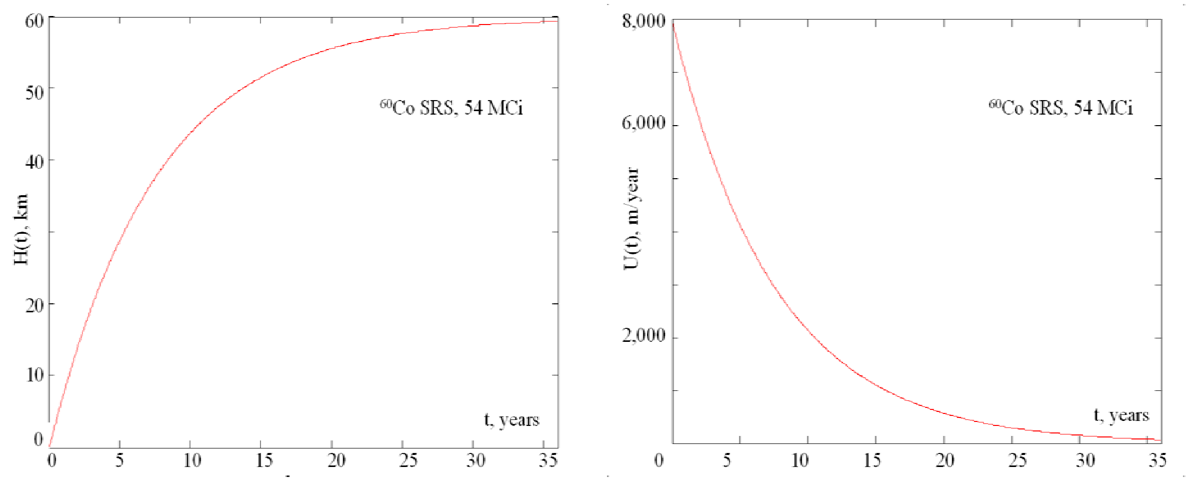
In order to assess the number of SRS needed to achieve the required self-burial parameters we assumed the activity of one SRS is ~2PBq (see Table I). The volume occupied by SRS in the capsule is then calculated from the typical volume of one SRS (about 10cm^3 [25]).

Table II. Self-Burial Parameters for Spent SRS Capsules.

Capsule radius R, m	**Radionuclide**	**Initial activity, PBq (MCi)**	**Number of 2PBq SRS**	**Approximate volume occupied by SRS in capsule, L**	**Initial specific heat power q(0), kW/m^3**	**Total heat power Q, kW**	**Initial descent velocity U(0), m/y**	**Time of continuous descent τ, years**	**Maximum depth of penetration H(τ), km**
0.50	^{60}Co	200 (5.4)	~100	~1-2	162	85	790	18.5	6
0.50	^{60}Co	2000 (54)	~1000	~10-20	1620	850	7900	36	60

Because the heat generating ^{60}Co SRS occupy a very small part of the available space in the capsule (Table II) the remaining volume can be utilized for other SRS containing, for example, long-lived radionuclides such as ^{226}Ra, ^{99}Tc.

Fig. 3 shows the depth reached and sinking velocity as functions of time after start up of self-burial for a 50-cm radius tungsten capsule heated by ^{60}Co SRS of total activity 2,000 PBq (54 MCi), i.e., containing approximately 1000 2PBq SRS.



(a) (b)

Fig. 3. Penetration depth (km) of a self-descending 50-cm radius tungsten capsule heated by ^{60}Co SRS (a) and the rate of self-burial (b).

The final depth of penetration is below the Mohorovicic discontinuity for all but the thickest continental crust. Rocks have never been sampled directly from such depths and knowledge is limited to what can be ascertained by remote geophysical methods such as seismology. Such deep self-burial of spent SRS would have a very small footprint of only a few m^2.

Capsule Tracking

An important issue surrounding the disposal of hazardous waste is its exact location and deep self-burial of SRS is no exception. Exact information would be required on the position of the capsule at any given instant up to the time sinking halts. As it descends the capsule melts the surrounding rock, which then re-crystallizes behind it. Melting and crystallization of materials generates intense acoustic signals (of peak pressure 10^{3}Pa) over a wide spectrum of frequencies due to thermo-mechanical interactions [26]. These are also produced by intense irradiation of materials [27]. It has been argued that a power of about 10 W is sufficient for the detection of acoustic signals from depths hundreds of km below the surface of the Earth [28]. The power of acoustic signals emitted by a descending SRS capsule can be calculated to be ~10$^{2-3}$ W. Inclusion of ^{226}Ra-Be neutron SRS in the capsule could provide additional intense neutron radiation to further strengthen the acoustic signal, which would help continuous tracking of self-burial. Detection of signals from the capsule by a number of coupled detectors would provide permanent information on its motion and position. Moreover these signals would carry information about the layers of the Earth’s crust (and possibly upper mantle) above the capsule that the signals have traveled through and hence on geological structure and, by inference, composition. These might be used, for example, to explore ore mineral resources. As the capsule can be emplaced at a given depth it could provide information about underground motions, which would be particularly useful in seismically active regions.

CONCLUSION

The self-burial concept can be readily adapted to very deep disposal of the most hazardous, powerful, spent SRS. Deep self-burial disposal of SRS involves collection of SRS, their loading into a sealed metal (tungsten) capsule and placing it at the bottom of a shallow borehole. Due to radiogenic heat generation, the capsule will melt surrounding rock and self-descend. Depths of many tens of km are readily achievable for self-burial. These are greater than the levels that have been achieved by deep drilling techniques. The SRS capsules could be tracked via detection of the acoustic signals generated by melting and crystallization of the rocks around and above the capsule. Analysis of the detected signals should also be able to provide information about the deep interior of Earth that is currently inaccessible to direct sampling and augment data from other remote geophysical monitoring techniques.

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