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# Conditioning and long-term storage of spent radium sources in Turkey

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## Abstract

Conditioning of radium sources is required before long-term interim storage to avoid the release of radioactive material and to limit radiation exposure. In this study, containment of the radium sources was achieved by high integrity encapsulation designed to control the radon emanation problem. The capsules were made of Type 316 austenitic stainless steel with dimensions of 22 mm diameter and 160 mm height. The gas pressures which was caused by encapsulation of different amounts of $^{226}$Ra were determined. The maximum gas pressure found 10 atm for 900 mCi of $^{226}$Ra in one capsule at 20 °C. A lead shielding device was designed to limit radiation exposure. A 200 l drum was used as a conditioned waste package for the radium sources and represents a Type A package under the IAEA transport regulations.

*Keywords:* Radium; Radioactive; Source; Conditioning; Ra-226

## 1. Introduction

The objective of conditioning of $^{226}$Ra sources is to produce a waste package acceptable for handling, transportation and interim storage. The waste package produced in a conditioning process needs to comply with the transport regulations and requirements for long-term storage. Where long-term disposal facilities do not exist, possible future retrieval of the conditioned sources has to be taken in consideration. $^{226}$Ra is in the $^{238}$U decay chain. A radium source always contains $^{226}$Ra and its daughter products. $^{226}$Ra decays by alpha emission to $^{222}$Rn, a noble gas with half-life of 3.6 days. In the decay chain ending with the stable isotope $^{206}$Pb, there are further eight radionuclides of which four are alpha emitters. Thus, each decaying $^{226}$Ra atom gives rise to five alpha particles. In the decay, many low energy gamma photons and beta particles are also emitted has a rather high dose factor [1]. Conditioning of $^{226}$Ra also provides, greater confinement of leaking spent $^{226}$Ra sources and reduces exposure potential. It is required before long-term interim storage to avoid the release of radioactive material and to limit radiation exposure. It is recommended by IAEA [3] that containment is designed to control the radon emanation problem. For this purpose, several techniques (active carbon box, capsules, etc.) have been used in previous applications. In this study, high integrity encapsulation was achieved by special designed capsules. A shielding container was designed to maximize the physical security of radium sources. Some important characteristics of $^{226}$Ra in equilibrium with its decay products are presented in Table 1 [2].

The decay of each atom of $^{226}$Ra yields five helium atoms formed from the alpha particles emitted in the decay chain. These are $^{226}$Ra, $^{222}$Rn, $^{218}$Po, $^{214}$Po and $^{210}$Po. These helium atoms generate overpressure in the sealed radium source (about 0.2 atm/year for 1 g of radium assuming a free volume of 1 cm$^3$) which can result in leakage and spread of contamination. If water of crystallization is present in the source, the alpha particles emitted in the decay chain decompose the water into oxygen and hydrogen, which further increases the overpressure. Leaking radium sources have always been a major radiation protection problem. In the early days of their use, there were a number of explosions of large standard radium sources encapsulated in glass. Explosive ruptures of metal sealed sources have also been reported. This characteristic of $^{226}$Ra is another reason why it is regarded as unsatisfactory from at radiation protection viewpoint [3]. Pressure increase is the major concern for safe storage of sealed $^{226}$Ra sources. Radium sources are used for medical and non-medical applications. Radium sources used for medical purposes have activities in the order of 40–400 MBq (1–10 mg of $^{226}$Ra). Typical uses and activities of radium sources in Turkey are presented in Table 2.

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Table 1
Characteristics of $^{226}$Ra used in sealed radiation sources (IAEA [2])

| Half-life | 1600 years |
|---|---|
| Alpha energies | Up to 7.7 MeV |
| Beta energies | Up to 2.8 MeV |
| Gamma energies | Up to 2.4 MeV |
| Gamma constant | 220 Sv/h at 1 m from a 1 GBq source |
| Half-value layer (HVL) of lead | 14 mm |
| Half-value layer (HVL) of concrete | 70 mm |
| ALI (oral) | $7 \times 10^4$ Bq |
| ALI (inhalation) | $2 \times 10^4$ Bq |

The largest users of $^{226}$Ra sources are hospitals. They are mostly used for brachytherapy and were encapsulated in platinium, platinium–iridium, gold or other alloys. These capsules were mainly divided into two groups needles and tubes depending on their use. Needles were of 1.7 mm diameter and 15–20 mm length. Tubes were 3 mm in diameter and 20–25 mm in length.

## 2. Method of conditioning

In this study, $^{226}$Ra sources were incapsulated in specially designed stainless steel capsules. After this stage, conditioning of $^{226}$Ra sources was achieved by using lead shielding within a prefabricated drum.

### 2.1. Source encapsulation

Capsules were made of Type 316 (AISI 316) stainless steel, which provides superior corrosion resistance when compared to conventional 440 stainless steel. AISI 316 is more resistant to atmospheric and general corrosive conditions than any of the other standard stainless steels. AISI 316 stainless is non magnetic because it contains no iron and has a low carbon content. The basic composition of the metal is a 18–8 chromium–nickel with molybdenum additive. The inner dimensions of each capsule were 22 mm diameter and 160 mm length (Fig. 1).

The inner volume of each capsule was 60.8 cm$^3$. Up to 16.6% of the volume of each capsule was filled with a 11.1 GBq $^{226}$Ra source. Free space of about 50 cm$^3$ (about 83.3% of the volume) remained. The inner pressure was measured in the free space in each capsule. The capsules were sealed using argon welding. Leakage was checked using a bubble test in a vacuum chamber.

Table 2
$^{226}$Ra sources used in Turkey

| Field | Application | Activities (MBq) |
|---|---|---|
| Research and industrial | Moisture detector (Ra–Be) | |
| Industrial | Eliminator for static electricity as lightning rod | 0.01–0.1 |
| Medical | Brachytherapy | 200–300 |
| Instrument dials/watches/clocks | Luminous paint | 0.2–5 |
| Smoke detectors | Silver foils | 0.01–0.1 |

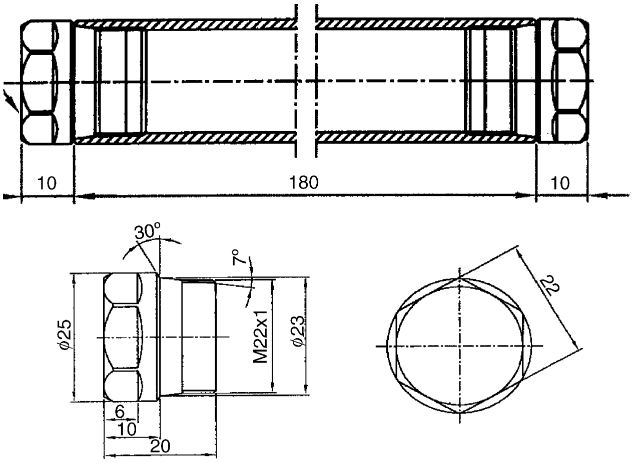

Fig. 1. Stainless steel capsules for $^{226}$Ra sources.

### 2.2. Shielding container

A lead shielding device was prepared for interim storage. Nine holes were constructed for capsules in a lead matrix with one in the centre and others around it (Fig. 2). The container provides shielding and ensures retrievability of capsules.

### 2.3. Waste package

Prefabricated 200 l drums were used for the waste packages. A concrete matrix was prepared to place the lead container in the drum (Fig. 3) and provided extra shielding.

A dose rate of 1.2 mSv/h was measured at the surface of the drum. This was lower than the maximum of 2 mSv/h needed to comply with the transport regulations and long-term storage requirements [4].

## 3. Inner pressure of capsules

Radium is an alkali earth metal. It is very reactive and reacts even with nitrogen. In radioactive sources radium, is therefore, used in the form of salts, which may be bromide, chloride or carbonate. All are soluble in water in amounts which can give

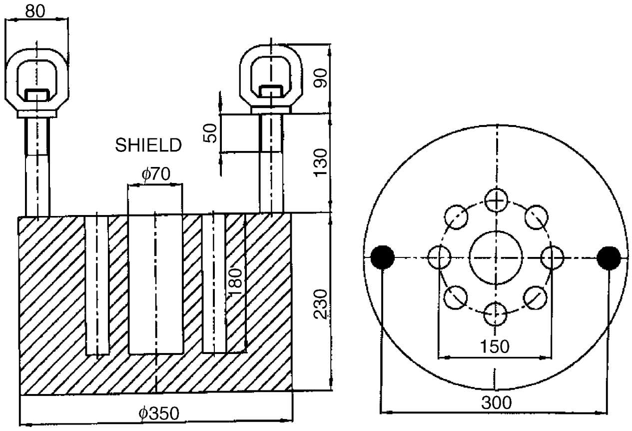

Fig. 2. Lead shielding device.

Fig. 3. Emplacement of capsules in shielding device.

rise to radiological problems. These salts may easily be dispersed as powder if the source encapsulation is damaged. In the body radium behaves like calcium, which means it concentrates in the bone where it has a very long biological half-life [5].

The Ra-226 decay scheme belongs to uranium–actinium series. The most important chains of this scheme leading to gas generation. The decay of one Ra-226 nucleus causes generation of one Rn-222 atom as intermediate product and of five helium atoms ultimately. Hence, the number of helium atoms $N\mathrm{He}$ produced due to the decay of Ra-226 and its daughter products can be written as: $N\mathrm{He} = 5\,N\mathrm{Ra}\,(0)[1 - \exp(-\lambda \mathrm{Ra} t)]$. If the helium atoms are accumulated in a given volume $V$, the gas pressure can be calculated as: $p\mathrm{He} = N\mathrm{He}kT/V$ [6].

$^{226}$Ra atoms initially decay to $^{222}$Rn and ultimately to the stable $^{206}$Pb. The number of $^{226}$Ra atoms in the source decreases according to its half-life (1600 years), but theoretically will not be zero. Therefore, production of helium gas or related pressure increase continues as long as the capsule keeps its integrity. The helium production rate decreases over time with the decreasing number of $^{226}$Ra atoms.

For each 37 GBq of $^{226}$Ra ($\cong 2.7 \times 10^{21}$ atoms), about $1.17 \times 10^{18}$ atoms will have decayed and five times as many (about $5.84 \times 10^{18}$) helium atoms will have been formed at the end of the first year. About $2.92 \times 10^{18}$ helium atoms will be formed after 1600 years. Pressure caused by $5.84 \times 10^{18}$ helium

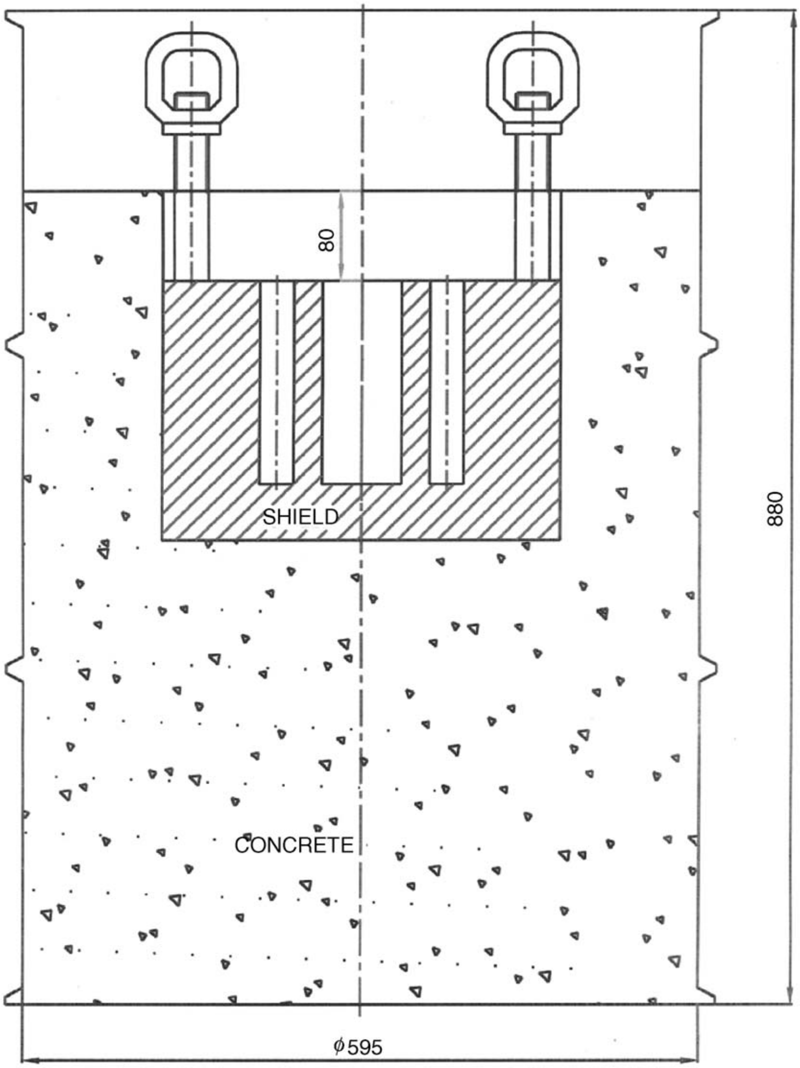

Fig. 4. Retrievable conditioned waste package.

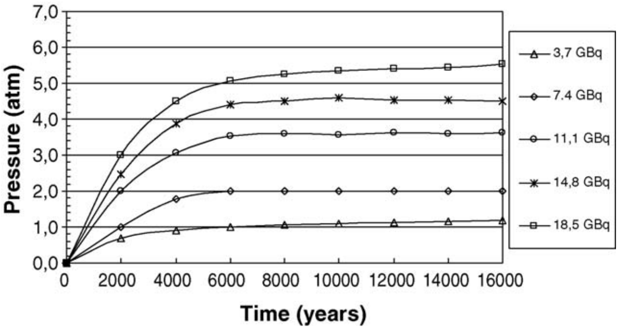

Fig. 5. Inner pressure of capsules.

atoms in 1 $cm^3$ volume is calculated as 0.23 atm at 20 °C. Pressure calculations were made for a period of up to 16,000 years 10 times of the $^{226}$Ra half-life. Projected pressures of helium and radon were calculated for various amounts of encapsulated $^{226}$Ra. Helium pressures in capsule for various amounts of $^{226}$Ra sources are shown in Fig. 4. Retrievable storage is required before the final disposal of $^{226}$Ra sources. A time period of 100 years has been assumed for the interim storage period. For evaluation of the interim storage period, pressure lines were calculated for the first 100 years and are presented in Fig. 5.

## 4. Results and discussions

Capsules are considered as ready for final waste packages without any further handling. For this reason, a time period of 16,000 years was assumed for capsule integrity. Pressure is caused by helium and $^{222}$Rn gas. $^{222}$Rn reaches equilibrium with its parent radionuclide $^{226}$Ra in about 30 days. This is the maximum amount of $^{222}$Rn in the capsule. After this period, $^{222}$Rn amount decreases together with the parent radionuclide. Decline of $^{222}$Rn pressure is not found in the first 10 years (Fig. 4) because of the long half-life of $^{226}$Ra but is found after 1000 years. In the interim storage period, $^{222}$Rn pressure reaches its maximum level at 0.1 year (36.5 days). This is 0.3% of the total pressure of helium and radon. In the 16,000 years evaluation period, $^{222}$Rn pressure decreases to the negligible values (Fig. 6). Helium gas pressure dominates long term. These pressures were calculated at 20 °C. Changing the temperature will result in different pressures (Fig. 7).

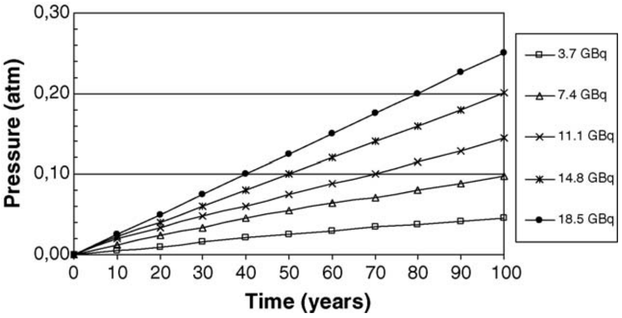

Fig. 6. Pressure changes in the first 100 years period.

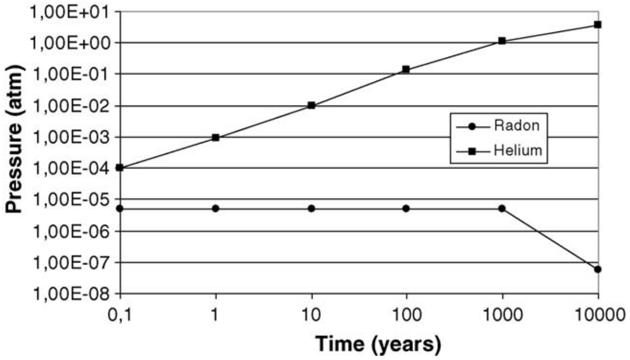

Fig. 7. Helium and Radon pressures for 300 mCi $^{226}$Ra.

## 5. Conclusions

Maximum pressure increases were calculated over a period of 20,000 years. After this time, pressures did not increase. Approximately, 90% of the total pressure rise occurs in the first 5000 years, 10% pressure rise occured in the next 15,000 years. Capsules can be regarded as ready for final packaging without further handling. For his reason, an evaluation period of 16,000 years was considered sufficient to ensure safety of capsules. The results of this study show that internal pressure of capsules can be neglected during this period of time. These results apply under standard conditions (20 °C). In abnormal conditions (fire, volcanic eruptions, etc.), temperatures and pressures will change and the results will not be valid.

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