R.K İleri Durmuş and S. Akyil Erentürk / Hacettepe J. Biol. & Chem., 2023 51 (1), 51-56

Research Article

Hacettepe Journal of Biology and Chemistry

journal homepage: www.hjbc.hacettepe.edu.tr

# Use of Composite Barrier in Radioactive Waste Repositories

# Radyoaktif Atık Depolarında Kompozit Bariyer Kullanımı

**Rüveyda Kübra İleri Durmuş, Sema Akyil Erentürk***

Department Energy Institute, Istanbul Technical University, İstanbul, Turkey.

## ABSTRACT

In this study, the performance of PAN/Zeolite-based composite barrier material was evaluated for the removal of $^{126}$Sn and $^{79}$Se radionuclides from an aqueous solution. Performance evaluation of the composite barrier material was performed separately for $^{126}$Sn and $^{79}$Se radionuclides using SWOT analysis. In addition, the risks to non-human organisms in case of accidental release of these radionuclides from radioactive waste landfills have been evaluated. These risks were evaluated by calculating the total doses of freshwater and terrestrial biota using the ERICA assessment tool. The exposed total dose range of reference biota as such crustaceans, phytoplankton, and vascular plants in freshwater was decreased to 4.02E6-3.87E9 µG $h^{-1}$ for $^{126}$Sn isotope, and 3.35E07-1.92E12 µG $h^{-1}$ for $^{79}$Se isotope using composite barrier material. In terrestrial reference biota such as annelids, grasses and grasses, lichens and bryophytes, mammals and trees, the exposed total dose range was decreased to 4.51E06-7.41E04 µG $h^{-1}$ for $^{126}$Sn and 2.15E06-5.53E09 µG $h^{-1}$ for $^{79}$Se.

**Key Words**

Radioactive waste repositories, composite barrier, Tin-126, Selenium-79, risk assessment.

## ÖZ

Bu çalışmada, sulu bir çözeltiden $^{126}$Sn ve $^{79}$Se radyonüklidlerin uzaklaştırılması için PAN/Zeolit bazlı kompozit bariyer malzemesinin performansı değerlendirildi. Kompozit bariyer malzemesinin performans değerlendirmesi $^{126}$Sn ve $^{79}$Se radyonüklidleri için SWOT analizi kullanılarak ayrı ayrı yapılmıştır. Ayrıca, radyoaktif atık depolama alanlarından bu radyonüklitlerin yanlışlıkla salınması durumunda, insan dışı canlılara yönelik riskler değerlendirilmiştir. Bu riskler ERICA assessment tool kullarak tatlı su ve karasal biotaların alacağı toplam dozlar hesaplanarak değerlendirilmiştir. Tatlı sudaki kabuklular, fitoplankton ve vasküler bitkiler gibi referans biyotaların maruz kaldığı toplam doz aralığı, kompozit bariyer malzemesi kullanılarak $^{126}$Sn izotopu için 4.02E6-3.87E9 µG $h^{-1}$'e ve $^{79}$Se izotopu için 3.35E07-1.92E12 µG $h^{-1}$'e düşürülmüştür. Annelidler, çimenler, likenler ve briyofitler, memeliler ve ağaçlar gibi karasal referans biyotasında, maruz kalınan toplam doz aralığı $^{126}$Sn için 7.41E04-4.51E06 µG $h^{-1}$ya ve $^{79}$Se için 2.15E06-5.53E09 µG $h^{-1}$'e düşürülmüştür.

**Anahtar Kelimeler**

Radyoaktif atık depoları, kompozit bariyer, Kalay-126, Selenyum-79, risk değerlendirmesi.

**Article History:** Received: Dec 21, 2021; Revised: Jan 18, 2022; Accepted: Jan 28, 2022; Available Online: Oct 5, 2022.

**DOI:** https://doi.org/10.15671/hjbc.1032456

**Correspondence to:** S. Erentürk, Energy Institute, Istanbul Technical University, Istanbul, Turkey.

**E-Mail:** erenturk@itu.edu.tr

## INTRODUCTION

Nuclear power plants seem to key solution for the increasing energy demand by the high capacity factor, preventing climate change; nevertheless, the radioactive wastes problem because of their high radioactivity are waiting for a permanent solution like conducting studies on the storage and disposal of radioactive waste repositories. The main concern of the waste repositories is to attain the water to the waste repository and to migrate the radionuclides in the waste packages to the biosphere. In order to protect the waste repository from the water and prevent the migration of the radionuclides in waste packages, the multi-barrier concepts composed of the engineered barriers and geological formation were developed and has been studied by researchers and authorities since the 1970s [1,2].

The main reason for developing the engineered barrier is the adsorption of the radionuclides having a long half-life and extension of the time that attain the radionuclides to the biosphere; therefore, the concentration of the radionuclides is decreased and they will not jeopardize human and ecosystem health. In order words, Engineered barriers work for the extension of the time to attain the radionuclides to the biosphere and for prevention of the attain the water to the waste repository; therefore, they work bi-directional. In the absence of the engineered barrier, water succeeds to meet the waste repository and radionuclides migrate to the biosphere earlier and more concentration than expected without adsorption [1,2].

In the design phase of the multi-barrier, the design of the engineered barriers is shaped by the geological formation and radioactive waste repositories. However, one of the engineered barriers is generally made of the buffer material where radionuclides contact firstly. The buffer materials should be designed to adsorb more radionuclides having a long half-life. Furthermore, the buffer materials should be designed with high chemical, physical and structural properties such as low permeability, high plasticity, and high cation exchange capacity [3-8].

$^{79}$Se and $^{126}$Sn are remarkable radionuclides in the waste packages because they are long half-live fission products and radiologically harmful in the spent nuclear fuel [9-11]. The fission efficiency for $^{126}$Sn and $^{79}$Se are 0.0236% and 0.04%, respectively [11,12]. The half-life of $^{126}$Sn is $2.35 \times 10^5$ and it is a dangerous beta (max = 252 keV) and gamma (87.6, 86.94 or 64.3 keV) emitter by thermal neutrons although it has a relatively low production of about 0.056% is a fission product. The half-life of $^{79}$Se is 65000 years and it is potentially dangerous for beta radiation (max = 0.056 MeV) [12]. According to the Belgian deep geological repository, $^{79}$Se is seemed to the main contributor to the activity release. Selenium is also redox-sensitive radionuclides that shows oxidation states as -II, 0, IV and VI in aquatic media.

According to the recommendation of the International Commission on Radiological Protection (ICRP) in 1978, while defending the view that “if people are sufficiently protected, other living organisms will also be adequately protected”. In 1991, it was stated that environmental control is also necessary for humans to be adequately protected, that other species should not be at risk, that non-human organisms may occasionally be harmed, but cannot endanger endangered species or create an imbalance between species. This change in the paradigm of the ICRP has accelerated the work carried out within the framework of the protection of non-human species from radiation. Since it will not be possible to evaluate the effects of radiation on all species in the ecosystem, separate assessments are made for reference organisms, exposure routes and terrestrial, freshwater and marine environments. With the ERICA (Environmental Risk from Ionizing Contaminants: Assessment and Management) program, which is a modelling simulation developed for this purpose, it is possible to predict the transfer of environmental or radionuclide pollution on the reference plants or animals, to determine the equivalent dose amount to which the biota will be exposed, and to evaluate the impact [13].

Within the framework of this paradigm, the doses to be exposed to non-human biota as a result of the release of $^{126}$Sn and $^{79}$Se into the environment as a result of an accident that may occur in radioactive waste repository areas were calculated using the ERICA Tool Program. The exposed total doses of selected freshwater and terrestrial reference biota were evaluated separately in the presence and absence of the composite barrier material used in the waste storage area. SWOT analysis was used to show strengths, weaknesses, opportunities, and threats of the composite barrier material.

## MATERIALS and METHODS

ERICA assessment tool is a product developed within the 6th Framework project ERICA and is one of the most inclusive assessments that has been recognized and used by international organizations such as ICRP, IAEA. ERICA Assessment Tool is used for determination of the amount of equivalent dose to which it will be exposed and effect evaluation on reference plant or animal that is determined by IAEA and UNSCEAR by transfer of environmental or radionuclide contamination [14,15]. Ionizing radiation exposure, effects and risks, chemical and environmental risk characterization and environmental management can be performed in order to protect the ecosystem structure and function using this computer-based software [16].

ERICA has 3 tiered approaches and it can be adapted to the specific assessment of using reference organisms. In Tier 1, only the selected elements of a specific area can be examined according to appropriate reference organisms and the risk assessment is made according to result by comparing against Environmental Media Concentration Limits (EMCL). The results are given as Risk Quotients (RQ):

$$RQ = \sum_{1}^{n} \frac{M_n}{EMCL_n}$$

where, RQ= Total Risk quotient, $M_n$= measured or predicted maximal activity concentration for radionuclide “n” in the medium in Bq $L^{-1}$ for water, Bq $kg^{-1}$ (dry weight) for soil or sediment or Bq $m^{-3}$ for air; $EMCL_n$= Environmental Media Concentration Limit for radionuclide “n” (same units as media) [16].

In Tier 2, selected elements in the specific area can be examined according to selected reference organisms and, the risk assessment is made according to the result. Selected reference organisms are representing the range of typical organisms from aquatic, terrestrial and marine environments. The results enable the calculation of doses and identification of the potentially most exposed reference organism by using the concentration value (CR) in Bq $kg^{-1}$ and distribution coefficient ($K_d$) in Bq $kg^{-1}$. Tier 2 is important for determining the uncertainties within the effects analysis part of the assessment [16].

In Tier 3, these results can be evaluated according to probability estimates. This tier enables to enter of the data for the site-specific probability distribution functions and gives the uncertainties in the final dose-rate results. Sensitivity analysis is important for the parameters on the effects on the model output and this helps to quantities the uncertainties especially cost-effective research [16].

In this study, ERICA Assessment Tool is used for the effect of the barrier on the dose exposure in the freshwater biota, i.e., crustacean, phytoplankton, vascular plant, and terrestrial biota i.e., annelid, grasses and herbs, lichen and bryophytes, mammal, shrub, tree. Initial Se and Sn concentration and remaining concentration in the solution after adsorption were converted to activity by using the following formula:

$$A = \frac{ln2}{t_{1/2}} x \frac{mN_A a}{M}$$

where A is activity (Bq), $t_{1/2}$ is half-life of the nuclei (s), M is the molarity of the substance (kg $mol^{-1}$), m is mass of substance (kg), NA is Avogadro’s number ($mol^{-1}$), a is the abundance.

In order to calculate to the subjected dose of the freshwater reference biota in the ERICA Tool Assessment Program, the required distribution coefficient ($K_d$) value was calculated as:

$$K_d = \frac{A_i - A_e}{A_e} x \frac{V}{m}$$

where $A_i$ is the activity concentration of the initial solution (Bq $mL^{-1}$), $A_e$ is the activity concentration of the solution in equilibrium (Bq $mL^{-1}$), V is the volume of the solution (mL), m is the amount of the adsorbent (0.05 g).

## RESULTS and DISCUSSION

In the event of an accident that may occur in radioactive waste storage, the risks of the spread of waste radionuclides to the environment have also been evaluated. The tin and selenium concentrations in the solution before and after the adsorption process were converted into activity units, and the performance of the barrier material to reduce the migration of tin and selenium ions into the ecosystem was investigated.

**Table 1.** Total $^{126}$Sn dose range for freshwater biota.

| Sn | Freshwater | | | | | |
|---|---|---|---|---|---|---|
| | Without barrier | | | With barrier | | |
| | Crustaen | Phytoplankton | Vascular Plant | Crustaen | Phytoplankton | Vascular Plant |
| Max. Dose (µG h$^{-1}$) | 1.24E+10 | 2.41E+08 | 1.08E+10 | 3.87E+09 | 7.49E+07 | 7.49E+07 |
| Min. Dose (µG h$^{-1}$) | 9.44E+08 | 4.01E+07 | 8.32E+08 | 2.37E+08 | 4.02E+06 | 4.02E+06 |

**Table 2.** Total $^{126}$Sn ddose range for terrestrial biota.

| Sn | Terrestrial | | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | Without barrier | | | | | | With barrier | | | | | |
| | Annelid | Grasses & Herbs | Lichen &bryophytes | Mammal | Shrub | Tree | Annelid | Grasses & Herbs | Lichen &bryophytes | Mammal | Shrub | Tree |
| Max. Dose (µG h$^{-1}$) | 1.45E+07 | 5.45E+06 | 6.20E+06 | 1.35E+07 | 5.46E+06 | 4.43E+06 | 4.51E+06 | 1.69E+06 | 1.93E+06 | 4.20E+06 | 1.70E+06 | 1.38E+06 |
| Min. Dose (µG h$^{-1}$) | 9.66E+05 | 3.63E+05 | 4.13E+05 | 9.00E+05 | 3.64E+05 | 2.95E+05 | 2.42E+05 | 9.11E+04 | 1.04E+05 | 2.26E+05 | 9.14E+04 | 7.41E+04 |

**Table 3.** Total $^{79}$Se dose range for freshwater biota.

| Se | Freshwater | | | | | |
|---|---|---|---|---|---|---|
| | Without barrier | | | With barrier | | |
| | Crustaen | Phytoplankton | Vascular Plant | Crustaen | Phytoplankton | Vascular Plant |
| Max. Dose (µG h$^{-1}$) | 7.13E+12 | 1.01E+09 | 1.07E+12 | 1.92E+12 | 2.73E+08 | 2.87E+11 |
| Min. Dose (µG h$^{-1}$) | 4.75E+11 | 6.77E+07 | 7.11E+10 | 2.35E+11 | 3.35E+07 | 3.52E+10 |

**Table 4.** Total $^{79}$Se dose range for terrestrial biota.

| Se | Terrestrial | | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | Without barrier | | | | | | With barrier | | | | | |
| | Annelid | Grasses & Herbs | Lichen &bryophytes | Mammal | Shrub | Tree | Annelid | Grasses & Herbs | Lichen &bryophytes | Mammal | Shrub | Tree |
| Max. Dose (µG h$^{-1}$) | 1.52E+09 | 5.78E+08 | 2.06E+10 | 6.50E+07 | 1.86E+09 | 1.86E+09 | 4.09E+08 | 1.56E+08 | 5.53E+09 | 1.75E+07 | 4.99E+08 | 4.99E+08 |
| Min. Dose (µG h$^{-1}$) | 1.01E+08 | 3.86E+07 | 1.37E+09 | 4.33E+06 | 1.24E+08 | 1.24E+08 | 5.02E+07 | 1.91E+07 | 6.79E+08 | 2.15E+06 | 6.13E+07 | 6.13E+07 |

The different activity ranges between the 1.14E09 and 1.71E10 Bq L$^{-1}$ activity values, the ERICA Tool Assessment program evaluated how much aquatic and terrestrial biota could receive as a result of leakage into the environment without barrier material in the waste area and the presence of barrier material. In the absence of barrier material, crustaceans, phytoplankton, and vascular plants, which are studied as freshwater biota, will be exposed to significant doses. The total dose range to which this biota will be exposed varies between 1.60E7 and 1.24E10 µG h$^{-1}$ from the radioactive isotope of tin, and between 6.77E07 and 7.13E12 µG h$^{-1}$ from the radioactive isotope of selenium (Table 1 and Table 3). In the case of using barrier material, the total dose

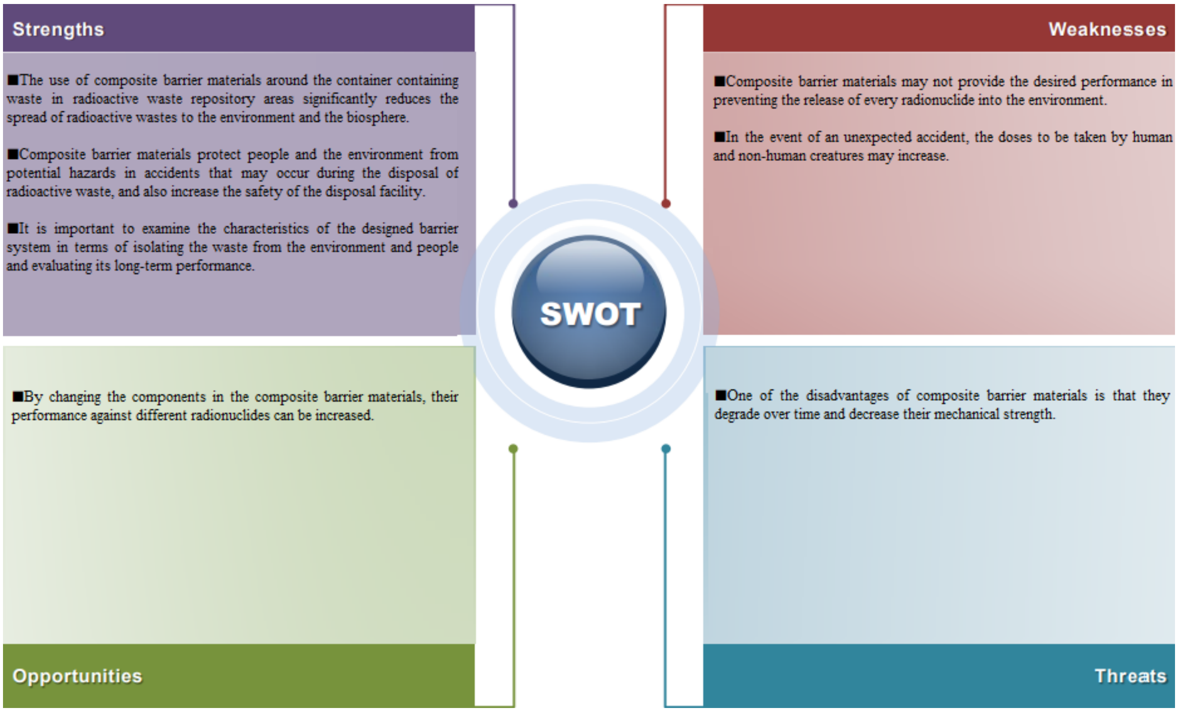


**Figure 1.** SWOT analysis for the composite barrier materials.

range to which this biota will be exposed decreases to 4.02E6 to 3.87E9 µG $h^{-1}$ for radioactive tin isotope, and 3.35E07 to 1.92E12 µG $h^{-1}$ for radioactive selenium isotope. In the case of using a barrier, the external dose is reduced by approximately 70%.

In terrestrial reference biota such as annelids, grasses and grasses, lichens and bryophytes, mammals and trees, the observed total dose range in the case of not using barrier material is between 2.95E05 and 1.45E07 µG $h^{-1}$ for tin, and between 4.33E06 and 2.06E10 µG $h^{-1}$ for selenium (Table 2 and Table 4). In the case of using barrier material, the dose range to which this biota will be exposed has decreased by 70% and it is 4.51E06 to 7.41E04 µG $h^{-1}$ for tin and 2.15E06 to 5.53E09 µG $h^{-1}$ for selenium. Since the exposure to freshwater and terrestrial biota from the total doses originating from radioactive waste repositories has not been examined in another study before with the ERICA program, it was not possible to compare with the literature.

SWOT analysis was used to evaluate the strengths, weaknesses, opportunities, and threats of the use of composite barrier materials in the spread of radionuclides, which are dangerous for living things due to their long half-lives and activities in radioactive waste repository areas. The results of the SWOT analysis are given in Fig. 1. Weaknesses can be improved for different radionuclides by using composite barrier materials of different compositions. In addition, by investigating the interactions of different radionuclides with the barrier materials in more detail, it can be suggested to use different composite barrier materials according to the radionuclide inventory of radioactive wastes.

## CONCLUSION

In this study, the effectiveness of a composite barrier material for $^{126}Sn$ and $^{79}Se$ used in radioactive waste repository areas was evaluated. In accordance with the recommendation of the International Commission on Radiological Protection (ICRP), the ERICA Tool Program was used to assess the total exposures of selected freshwater and terrestrial reference biota. The doses of freshwater and terrestrial biota originating from $^{126}Sn$ and $^{75}Se$ are significantly reduced in the presence of the composite barrier material. Some of the freshwater and terrestrial biota are more exposed to these doses.

In line with the results obtained, the usability of the composite barrier material was evaluated in terms of strengths, weaknesses, opportunities, and threats using SWOT analysis.

**Acknowledgments**

This study is a part of the project supported by the Istanbul Technical University Scientific Research Projects Unit with project number TDK-2018-41684.

## References

1. B. Grambow, Mobile fission and activation products in nuclear waste disposal, J. Contam. Hydrol., 102 (2008) 180–186.
2. C. Bruggeman, A. Maes, J. Vancluysen, P. Vandemussele, Selenite reduction in Boom clay: Effect of $FeS_2$, clay minerals and dissolved organic matter, Environ. Pollut., 137 (2005) 209–221.
3. H.S. Ahn, H.Y. Jo, Influence of exchangeable cations on hydraulic conductivity of compacted bentonite, Appl. Clay Sci., 44 (2009) 144–150.
4. S. Kaufhold, M. Pohlmann-Lortz, R. Dohrmann, R. Nüesch, About the possible upgrade of bentonite with respect to iodide retention capacity, Appl. Clay Sci., 35 (2007) 39–46.
5. B. Riebe, S. Dultz, C. Bunnenberg, Temperature effects on iodine adsorption on organo-clay minerals: I. Influence of pretreatment and adsorption temperature, Appl. Clay Sci., 28 (2005) 9–16.
6. J. Manjanna, T. Kozaki, S. Sato, Fe(III)-montmorillonite: Basic properties and diffusion of tracers relevant to alteration of bentonite in deep geological disposal, Appl. Clay Sci., 43 (2009) 208–217.
7. A. Gosman, I. Hvozdova, J.L.M. Nieto, Measuring The Diffusion and Sorption of the Trace Elements and Radionuclides in Soil, Clay and Bentonite, Czechoslov. J. Phys., 49 (1999) 847-853.
8. C. Hurel, N. Marmier, A. Bourg, F. Fromage, Sorption of Cs and Rb on purified and crude MX-80 bentonite in various electrolytes, J. Radioanal. Nucl. Chem., 279 (2008) 113–119.
9. S.A. Catlow, G.L. Troyer, D.R. Hansen, R.A. Jones, Half-life measurement of $^{126}Sn$ isolated from Hanford nuclear defense waste, J. Radioanal. Nucl. Chem., 263 (2005) 599–603.
10. G. Jörg, R. Bühnemann, S. Hollas, N. Kivel, K. Kossert, S. Van Winckel, C.L. Gostomski, Preparation of radiochemically pure $^{79}Se$ and highly precise determination of its half-life, Appl. Radiat. Isot., 68 (2010) 2339–2351.
11. A.L. Nichols, D.L. Aldama, M. Verpelli, International Atomic Energy Agency INDC International Nuclear Data Committee Handbook of Nuclear Data for Safeguards: Database Extension, INDC (NDS)-0534, Vienna, 2008.
12. V. Silliková, S. Dulanská, Sequential determination of $^{99}Tc$ and $^{126}Sn$ in radioactive concentrate, Acta Chim. Slovaca, 10 (2017) 61–64.
13. ICRP, Publication 29: Radionuclide release into the environment: assessment of doses to man, Pergamon, New York, 1978.
14. J.E. Brown, B. Alfonso, R. Avila, N.A. Beresford, D. Copplestone, G. Prohl, A. Ulanovsky, The ERICA Tool, J. Environ. Radioact., 99 (2008) 1371-1383.
15. N. Beresford, J. Brown, D. Copplestone, J. Garnier-Laplace, B. Howard, C.M. Larsson, D. Oughton, G. Pröhl, I. Zinger, D-ERICA: An integrated approach to the assessment and management of environmental risks from ionising radiation, Description of purpose, methodology and application, EC Contract FI6RCT-2004-508847, European Commission Report, https://wiki.ceh.ac.uk/download/attachments/115017395/D-Erica.pdf, (2004).
16. D.H. Oughton, G. Strømman, B. Salbu, Ecological risk assessment of Central Asian mining sites: application of the ERICA assessment tool, J. Environ. Radioact., 123 (2013) 90-98.