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Monte Carlo simulations of radioactive waste embedded into EPDM and effect of lead filler

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HIGHLIGHTS

- EPDM candidate polymeric material to be used in the radioactive waste management.
- Activity of the radioactive waste to be embedded into the EPDM matrix was simulated.
- Effect of lead filler to the polymer matrix was determined.
- Simulations shown that lead filler decrease dose delivered and surface dose rate.

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ABSTRACT

Radioactive waste is generated from the nuclear industry and should be processed and disposed of according to the regulations set by the appropriate regulatory authority. Ethylene propylene diene monomer (EPDM) is a widely used polymer and might be considered as a potential candidate radioactive waste encapsulation material. In this study, the dose rate distribution in the radioactive waste drum (containing radioactive waste and the polymer matrix) was determined using Monte Carlo simulations. The change in the dose rate within the waste drum with different amounts of lead filler was also simulated. It was seen that lead filler would decrease the dose delivered to the polymer by means of energy dissipation. Moreover, the change of mechanical properties of EPDM was estimated and their variation within the waste drum was determined for the duration of 15, 30 and 300 years after embedding.

1. Introduction

Radioactive waste is inevitably generated from the nuclear industry and nuclear applications; the generated radioactive waste should be managed in a safe and secure manner to minimize its current and future potential risks on people and the environment. The wastes that are generated by the nuclear industry are generally classified into levels: low, intermediate and high. Different physical and chemical methods (such as evaporation, precipitation, ion exchange, adsorption on clay surface) are used to concentrate wastes for further treatment or conditioning. Waste immobilization methods that include embedding of radioactive waste into cement, polymer (Day et al., 1985) or bitumen are the practices used in the radioactive waste management applications. Cementation could be ineffective, such as for the cases of (1) wastes containing metallic parts due to corrosion of metallic parts, and (2) waste containing ion exchange resins due to the release of mobile radionuclides during the curing of cementation process, for such cases use of polymeric materials could be an alternative to the cementation process. Polymers with a considerable degree of radiation stability have been used as encapsulants for radioactive waste management (IAEA, 1988; Debré, O et al., 1997; Özdemir, 2008). Polymers have also been considered for use as transport containers for radioactive waste (Bonin et al., 2004). The use of polymeric materials in radioactive waste management as encapsulant or for use in radioactive waste transport has been considered by many authors (Sakr et al., 2003; Damian et al., 2001; Nicaise et al., 1986a,b; Baluch et al., 1977; Debre´ et al., 1997; Özdemir and Usanmaz, 2007, 2008, 2009a, 2009b, 2009c; Özdemir, 2008; Hacıoğlu et al., 2013).

Repositories for the final disposal of radioactive waste include a multi-barrier system to isolate the waste from the biosphere. The multi-barrier concept typically comprises a natural geological formation and an engineered barrier system. The engineered system might consist of many different components, such as the waste form itself, waste canisters, backfill, seals and plugs (NEA, 2003). The main function of the engineered barrier system is to prevent and/or delay the possible release of radionuclides from the

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waste forms to the repository host rock for several hundred years after repository backfill and closure (NEA, 2003). Consequently, radioactive waste that is encapsulated in a polymer matrix is part of the engineered barrier system and should delay or preferentially prevent the release of radionuclides into the local environment.

MCNP is a general-purpose Monte Carlo N-Particle input that could be used for neutron, photon, electron or coupled neutron/photon/electron transport simulations (Briesmeister, 1997). Monte Carlo simulations for wastes encapsulated in polymer matrices have been studied, the materials that have been studied with Monte Carlo studies include: polycarbonate urethane (PCU), poly (bisphenol a-co-epichlorohydrin) (Özdemir and Usanmaz, 2009b), and polymethylmethacrylate, (PMMA) (Özdemir and Usanmaz, 2009c). In those studies, the activity of wastes that could be embedded into a drum and the dose rate distribution in the drum arising from the wastes were simulated. On the other hand, radiation stability of the EPDM was also studied (Özdemir, 2008; Hacıoğlu et al., 2013) and it was concluded that EPDM could resist to the gamma radiation dose of 2156 kGy. In the previous study (Hacıoğlu et al., 2013), the compounding of the EPDM was containing ethylene, propylene, diene source, vulcanizing agent (2,5-di-(t-butylperoxy)-2,5-dimethylhexane (DMBPHa) or di-(tert-butylperoxyisopropyl)-benzene (BBPIB)), coagent, zinc oxide, plasticizer, carbon black. High dose rate irradiations carried out in that study with an initial dose rate of 993 Gy/h and low dose rate irradiations had been carried out with an initial dose rate of 63.16 Gy/h. The highest dose that the samples had been irradiated was 2156 kGy.

It is well known that additives are used to change the properties of polymers or other materials to obtain desired properties. Additives can also act as radiation protection agent in the polymeric matrix; this could be achieved by means of either internal protection or external protection (Ivanov, 1992). In this regard, additives to the polymer matrix such as metal filler in the forms of either micro, nano or flake form may decrease the dose absorbed by the polymer. There is a good correlation between the photoelectric absorption and the atomic number that makes lead an appropriate material for radiation shielding applications (Cember and Johnson, 2009). Lead additives are often used in EPDM compounding for wire and cable applications (Kang, D.H., 2006). It is well known that, shielding of electromagnetic radiation is achieved with use of high atomic number materials with high density (El-Hosiny and El-Faramawy, 2000).

In this study, based on previous experimental result of 2156 kGy (Hacıoğlu et al., 2013) of total allowable exposure dose of EPDM, the maximum allowable waste activity than could be embedded into a waste drum was determined. The dose rate distribution within the waste drum (containing waste and the polymer matrix) was also simulated via Monte Carlo simulation. The change in the mechanical properties of the EPDM was determined and their variation within the waste drum was obtained for the cases of 15, 30 and 300 years after embedding waste in drum. In addition, the effect of the lead filler to the polymer matrix was simulated and the corresponding dose rates were calculated for the cases of different lead filler compositions.

2. Materials and methods

For this work program, radioactive waste structures with (1) spherical and (2) cylindrical geometries embedded into polymer matrices were studied. For the cylindrical geometry, radioactive waste was taken as a fixed size/geometry Cs-137 source with the radius of 0.5 cm and height of 0.5 cm and located at the center of the waste drum. On the other hand, for spherical geometry, radioactive waste was taken also as Cs-137 with the radius values of 5, 10, 15 and 20 cm. The radius of the waste drum was taken as 28.5 cm with a height of 83 cm. Monte Carlo simulations were carried out to determine the maximum allowable waste activity that could be embedded into the waste drum without completely degrading the structural integrity of the package after a 300 year control period. As the radiation stability of the EPDM matrix had been determined as 2156 kGy, a 20 cm radius was selected as the boundary for the dose due to waste. This meant that the total absorbed dose at a distance of 20 cm from the center of drum would be equal to the maximum allowable dose, 2156 kGy, for the polymeric material after 300 years exposure. By setting a 20 cm distance from the center, an annulus of 8.5 cm thickness of polymeric material (annular region) would be the geometry that would provide a barrier to prevent the release of the radionuclides to the environment.

Using the Monte Carlo method, the maximum allowable dose rate was then calculated. The dose rate distribution for the waste drum was then determined for the EPDM matrix at the initial time of embedding this activity in the waste. Dose rate distributions along the drum radius for different exposure periods of 15, 30 and 300 years after initial encapsulation were also determined. In addition, a feature called mesh tally was used in order to obtain a contour plot of the dose rate distribution.

The total dose to the immobilization matrix during the active institutional control period was calculated via Eq. (1) (Özdemir and Usanmaz, 2007), where TD is total dose (kGy), DR_o is initial dose rate (kGy/year), t is period (year) and k is decay constant. The active institutional control period is defined as the period during which the low and intermediate level short lived radioactive waste category disposal facilities should be properly controlled and regulated (Debré et al., 1997) by the facility operator and the regulatory authority, respectively.

$$TD = \int_0^T DR_o \times e^{-kt}\, dt \tag{1}$$

As described earlier, changes in the mechanical properties of the EPDM as a function of radiation dose up to 2156 kGy when irradiated at a dose rate of 993 Gy/hr were obtained in previous work (Hacıoğlu et al., 2013). Consequentially, knowledge of the change in properties of the EPDM with radiation dose was available. The initial dose rate distribution within the waste drum containing the polymeric matrix was found as a function of radial distance from the Monte Carlo method. The total dose as a function of radial distance was calculated from Eq. (1) for the time periods of 15, 30 and 300 years. As a result, the changes in mechanical properties as a function of radial distance for the time periods of 15, 30 and 300 years were obtained. In other words, mechanical properties were found as a function of total dose from the experimental results, total dose within the drum was found as a function of radial distance from simulations, these two results were combined to obtain the change of mechanical properties as a function of radial distance. Moreover, simulations were carried out to find the dose rate distribution within the waste drum by means of the addition of lead filler with amounts of 1, 5 and 10% by weight.

3. Results and discussions

The maximum allowable activity of the Cs-137 source with cylindrical geometry that could be embedded into EPDM filled waste drum was found to be 6.05 TBq. The radiation dose rate associated with this source is the maximum rate that can deliver a total dose of 2156 kGy at 20 cm from the center of the drum after 300 years. 2156 kGy is the total allowable dose for the EPDM estimated from the previous study. Fig. 1(a) shows the estimated reduction in dose rate distributions from such a source in a drum after time periods of zero