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Monte Carlo simulations of radioactive waste encapsulated by bisphenol-A polycarbonate and effect of bismuth-III oxide filler material

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ABSTRACT

Radioactive waste generated from the nuclear industry and non-power applications should carefully be treated, conditioned and disposed according to the regulations set by the competent authority(ies). Bisphenol-a polycarbonate (BPA-PC), a very widely used polymer, might be considered as a potential candidate material for low level radioactive waste encapsulation. In this work, the dose rate distribution in the radioactive waste drum (containing radioactive waste and the BPA-PC polymer matrix) was determined using Monte Carlo simulations. Moreover, the change of mechanical properties of BPA-PC was estimated and their variation within the waste drum was determined for the periods of 15, 30 and 300 years after disposal to the final disposal site. The change of the dose rate within the waste drum with different contents of bismuth-III oxide were also simulated. It was concluded that addition of bismuth-III oxide filler decreases the dose delivered to the polymeric matrix due to photoelectric effect.

1. Introduction

Radioactive waste is inevitably generated from the nuclear industry and nuclear applications; the generated radioactive waste should be managed with a safe and secure manner to minimize its current and future potential risks on both human beings and the biotic/abiotic environment. Although, many more classification schemes have been developed (IAEA, 2009), the radioactive wastes that are generated specifically from nuclear industry is mainly classified as; (a) low, (b) intermediate and (c) high-level waste regarding to the activity levels of the wastes. Different physical and chemical methods (such as evaporation, precipitation, ion exchange, adsorption on clay surface) are employed to concentrate wastes for further treatment or conditioning (Valdovinos et al., 2014). Waste immobilization methods that include embedding of radioactive waste into cement is the main practice used in the radioactive waste management. In addition, bitumen is also used for the immobilization purpose. Although, the frequency of use was not as much as that of cement or bitumen, polymers were also used as the immobilization matrix (Day et al., 1985; Kalb et al., 1996; Debré et al., 1997; Cota et al., 2009). Cementation could, however, be ineffective for different cases (Cummins, 2011) such as (a) leaching of resins during the initial period of cementation process resulting with presence of mobile radionuclides (b) interference of mobile radionuclides during the curing of cementation, for these cases use of polymeric materials could be an alternative solution to the conventional cementation process. Although, the solution of the problem with the use of polymeric encapsulant is generally more expensive compared to that of using conventional cementation process, the safety perspective might require the selection of expensive and convenient alternative. Polymers with a considerable degree of radiation stability have been used as encapsulants for radioactive waste management (IAEA, 1988; Debré, O et al., 1997). On the other hand, polymeric materials have also been considered for use as transport containers for radioactive waste (Bonin et al., 2004). The employment of polymeric materials in radioactive waste management as encapsulant or as radioactive waste transport container have been considered by many studies (Baluch et al., 1977; Nicaise et al., 1986a, 1986b; Debré´ et al., 1997; Damian et al., 2001; Sakr et al., 2003; Özdemir and Usanmaz, 2007; Özdemir and Usanmaz, 2008; Özdemir, 2008; Özdemir and Usanmaz, 2009a, Özdemir and Usanmaz, 2009b, Özdemir and Usanmaz, 2009c; Cummins, 2011; Hacıoğlu et al., 2013; Hacıoğlu et al., 2016). It should be clarified that the polymeric encapsulant could mainly be considered for the encapsulation of low and intermediate level radioactive wastes. The polymeric materials, for the time being, have limitations for the high level waste management mainly due to radiation and thermal stability considerations.

Treatment of radioactive waste includes following steps of; (1) embedding waste into the container pre-filled with PC, and (2) container fulfilment with polycarbonate. Physical disintegration of embedding matrix could produce preferential pathways for radionuclide migration.

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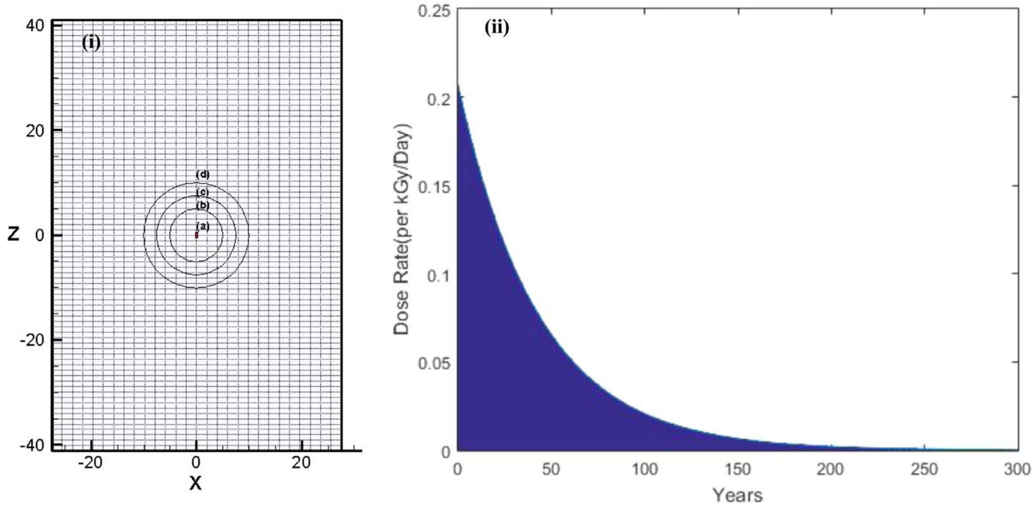

Fig. 1. (i) Cylindrical (a) and spherical (b, c, d) waste geometries (ii) Change of initial dose rate (kGy/day) with the time period of 300 years.

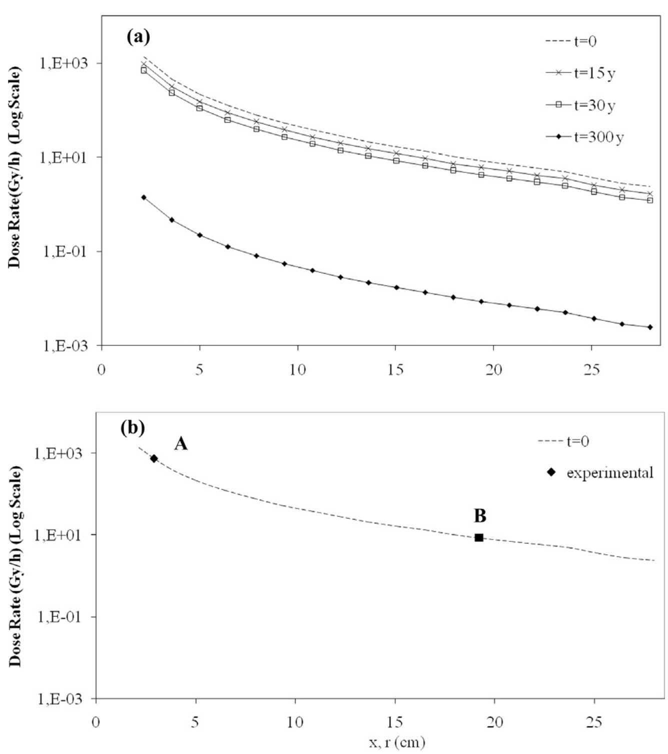

Fig. 2. (a) Change of dose rate (Gy/h) with the distance from the center in the waste drum embedded with BPA-PC (b) semi logaritmic plot of initial dose rate.

Final repositories for the disposal of radioactive waste should be composed of multi-barrier system to isolate the radioactive waste from the biosphere for the safety purposes both for the present and future generations. As it is well known, the multi-barrier concept typically comprises a natural geological formation and engineered barrier systems. The engineered systems might consist of many different components, such as waste canisters, backfill material, seals and plugs (NEA, 2003). The main function of the engineered barrier system is to prevent and/or trap the possible release of radioactive nuclides from the waste forms to the repository's host rock for several hundred years after repository backfill and closure (NEA, 2003). Consequently, radioactive waste that is encapsulated in a polymer matrix could possibly be a part of the engineered barrier system and could help delay of radionuclide migration for a considerable time or better prevent the release of radioactive nuclides into the local environment of the repository.

Polycarbonates are polyester type polymeric materials and they are the second largest used engineering thermoplastics in the World (Ebewele, 2000). In addition, bisphenol-a polycarbonate has a high toughness, a good optical transparency, and inherited flame resistance. Polycarbonates are used for variety of applications while they are utilized extensively in medical, automotive, electrical, electronic, and technical applications (Mark and Kroschwitz, 2004). Compact discs (CD/DVD) made of bisphenol-a polycarbonate were the popular data storage media for about two decades and million tones of polycarbonate was used for the manufacture of the those discs. Currently, many of the used discs became waste since no further use is foreseen for them. Proper way of utilization of these waste products could be a potential environmentally friendly way that could employ the reuse of polycarbonate recovered from waste discs for the purpose of radioactive waste embedding.

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 (Anon, 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) and poly(bisphenol a-*co*-epichlorohydrin) (Özdemir and Usanmaz, 2009b), PMMA (Özdemir and Usanmaz, 2009c) and EPDM (Özdemir, 2014). 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, gamma radiation stability of the BPA-PC was also studied by our research group (Hacıoğlu et al., 2016) and it was concluded that BPA-PC could resist to the gamma radiation dose of 3280 kGy. This dose was achieved with a Co-60 gamma irradiation source and irradiations were carried out with a dose rate of 737 Gy/h. It should be noted that the the type of radiation, dose rate and the period of irradiation are important factors for determining the resistible radiation dose.

Additives are used to change the properties of polymers or other materials to obtain desired properties. Additives could functions as a radiation protection agent in the polymeric matrix; this could be achieved by means of either (a) internal protection, or (b) 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 limit the irradiation dose to the polymer matrix. There is a good correlation between the photoelectric absorption and the atomic