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# Removal of radioactive contaminants by polymeric microspheres

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**ABSTRACT**

Radionuclide removal from radioactive liquid waste by adsorption on polymeric microspheres is the latest application of polymers in waste management. Polymeric microspheres have significant immobilization capacity for ionic substances. A laboratory study was carried out by using poly(*N*-isopropylacrylamide) for encapsulation of radionuclide in the liquid radioactive waste. There are numbers of advantages to use an encapsulation technology in radioactive waste management. Results show that polymerization step of radionuclide increases integrity of solidified waste form. Test results showed that adding the appropriate polymer into the liquid waste at an appropriate pH and temperature level, radionuclide was encapsulated into polymer. This technology may provide barriers between hazardous radioactive ions and the environment. By this method, solidification techniques became easier and safer in nuclear waste management. By using polymer microspheres as dust form, contamination risks were decreased in the nuclear industry and radioactive waste operations.

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**KEYWORDS**
Cs-137; liquid waste; polymer; radioactive; removal

## Introduction

Radionuclide contamination of waste water is a significant issue for the environment because of their potential health effects. Various treatment technologies exist for removing radionuclide from the waste water. Some of these technologies are ion-exchange, reverse osmosis, filtration and chemical precipitation. Most of these treatment technologies require an investment for a plant. In addition, after these processes another type of waste was generated. By these methods, volume reduction and concentration were achieved. In this study, basic properties of polymeric hydrogels were used for immobilization of radionuclide in waste water. Polymer microspheres have basic parameters which affect their overall performance. The basic parameters are the size and distribution of particles, chemical structure and reaction conditions. By adding into aqueous medium, ions in the liquid will be encapsulated by surrounding polymer microspheres. Adsorption and flocculation of polymers and polymer mixtures were studied theoretically [1–3] and practically [4–8] by numerous researchers. Specific polymers were prepared by using gamma rays, and their radioactive intake capacities were investigated. Poly(acrylic acid–maleic acid) $SiO_2/Al_2O_3$ was prepared as a novel composite material for the removal of cesium from acidic solutions.[9] Microcrystalline Naphthalene was studied for extraction of cobalt, strontium and cesium from liquid radioactive waste, and adsorption capacity of Cs was found as 16 mg/g.[10] Depending on the poly(itaconic acid) graft chains poly (*N*-isopropyl acrylamide) and its copolymers were studied in the development of temperature-modulated drug delivery systems.[11,12] Sorption properties were investigated by using resorcinol-formaldehyde polymeric resin.[13,14] Researchers started to use polymers in radioactive waste management and decontamination of radioactive materials.[15–17] Crown ethers impregnated polymeric resin was used for treatment of low-level liquid radioactive waste.[18] Kinetic and thermodynamic studies for cesium removal from low-level liquid radioactive waste using impregnated polymeric material were carried out. According to studies, polymers can be used as an alternative sorbent material for the treatment of liquid waste.[19] Polymer-assisted ultrafiltration was investigated for the removal of ions from water.[20] Effects of polymers were determined on the phase separation of aqueous solutions.[21] In case of addition of polymer in an aqueous solution, it swells and becomes an elastic gel depending on the aqueous radioactive waste. These properties of polymers are the motivation of this study and utilization of polymers in solidification process of the waste management is the novel of this study. In this study, microspheres were prepared by the inverse suspension polymerization technique. Encapsulation is a process to entrap radioactive nuclides within a carrier polymer and it is a useful tool to transfer hazardous nuclides to solidification process.

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One of the most important reasons for encapsulation of radioactive nuclide is to provide improved stability in the final waste form. By this way, evaporation is decreased and contamination of radioactive materials is prevented.

## Materials and methods

### *Preparation of microspheres*

Microspheres which were prepared previously by the inverse suspension polymerization technique were used in this study.[22] The diameter of the microspheres ranged from 71 to 500 µm, as shown in Figure 1. The particles were sieved and separated into particle distribution range groups. Each group was dried at 40°C in a vacuum oven for 24 hours.

Physical properties of this polymer were determined and mean values are presented in Table 1.

### *Solidification and polymerization process*

Polymeric material was taken and mixed with pure Portland cement. Homogenous mixture was prepared and dried at 40°C in vacuum oven for 24 hours. This procedure was applied to all size ranges of microspheres. This adsorbent can selectively adsorb radionuclide from radioactive liquid waste through cementation process. The cementation process is used in management of liquid radioactive waste for long-term stability. In this experiment, real liquid radioactive waste was used as 10 kBq/L specific activity of Cs-137 at the waste treatment facility. The radionuclide capacity was found on this adsorbent as 50 kBq/g at cesium-137 concentration of 10 kBq/L. Two sample groups were performed for tests. For the first-group samples, distilled water was used for solidification of polymer/cement mixture. Tests were applied to this group without using any radioactive liquid. SEM image of polymer/cement mixture solidification of distilled water is shown in Figure 2.

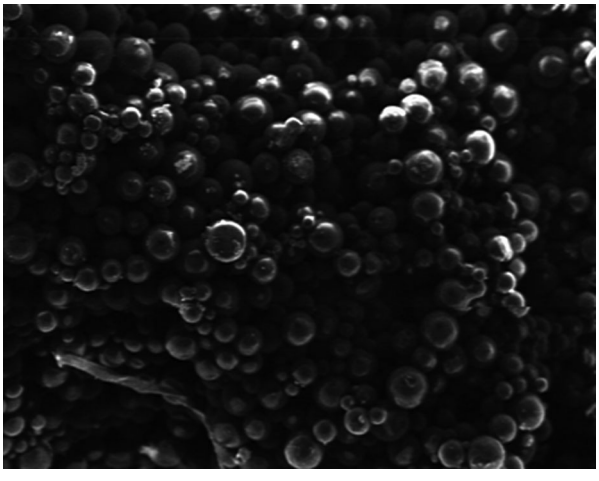

**Figure 1.** Microspheres.

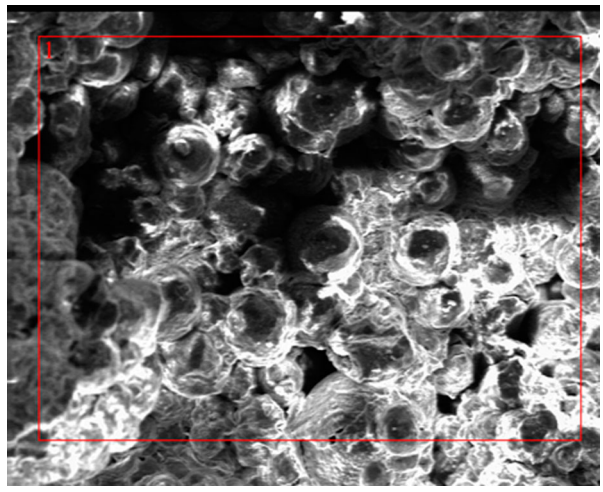

**Figure 2.** SEM image of polymer/cement mixture.

**Table 1.** Properties of Poly(*N*-isopropylacrylamide) microspheres.

| Physical form | Microspheres |
|---|---|
| Diameter of microspheres | 71–500 µm |
| Total exchange capacity | 0.60 meq/ml |
| Radionuclide capacity | 50 kBq/g for Cs-137 (10 kBq/L radioactivity) |
| Density | 1.20 g/cc |
| Water retention capacity | 70% |

### *Solidification process*

By solidification process, voids and surfaces of microspheres covered by cement. For the second-group samples of polymer/cement mixtures, Cs-137 radionuclide solution with 10 kBq/L specific activity was used for solidification (Figure 3).

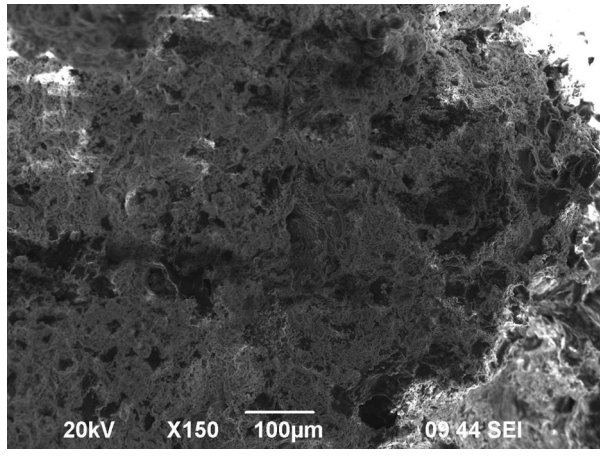


**Figure 3.** SEM image of polymer/cement/radionuclide mixture.

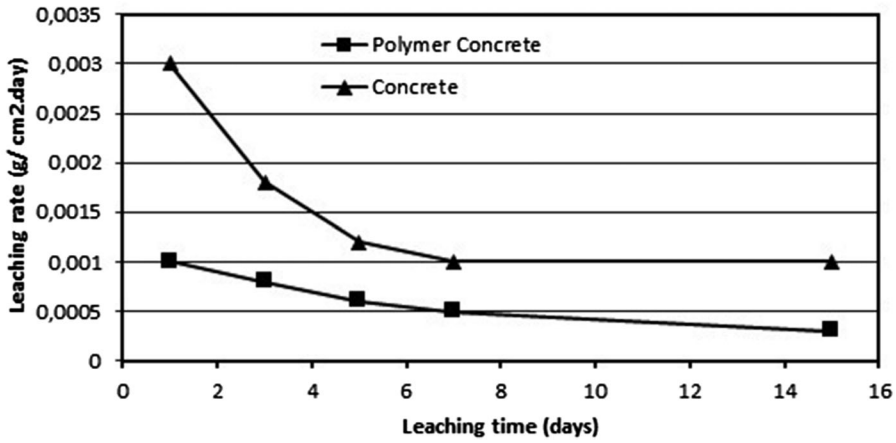


**Figure 4.** Leaching rate of solidified samples.

## Results

Leaching tests were applied to both groups of samples. Leaching test results at different pH levels show that maximum leaching rates were achieved at pH 5.5. Leaching media was arranged at pH 5.5. Cylindrical samples were immersed into the water and leaching amounts during the 15 days were determined. Leaching rates were measured in g/cm$^2$ day. In addition, specific activity of radionuclide was monitored by gamma spectrometry analysis. The gamma ray spectrometry system consisted of a HPGe coaxial GCD-80-210 detector with %80 efficiency coupled with a multispectral hybrid converter (MCA), high voltage (5000 V) with negative polarity. LSRM SpectraLineGP software was used to record the intensity of the incident and transmitted gamma rays. The resolution of the system was 1.9 keV at 1332.5 keV gamma peaks from a Co-60-point source kept at a distance of 10 cm in front of the detector face. The whole system was housed in a lead shield with varied thickness. The shield also contained 1.5-mm-thick copper and 1.0-mm-thick aluminum to suppress annihilation photons and X-rays. Liquid passed from the solidified samples was collected and measured during the test time. The leaching behavior of solidified waste containing different amounts of radionuclides were expressed in g/cm$^2$ day according to IAEA test protocol [23] and ISO 6961-1982. Changing of leaching rate, according to the time is shown in Figure 4. In the first five days, leaching amounts were significantly decreased to 0.001 g/cm$^2$ day. After 10 days, leaching rate was slightly decreased to 0.0005 g/cm$^2$ day for polymer/concrete waste form. Results show that solidified waste form without polymer has a low resistance to leaching rate than polymer-added solidified waste form. NX-type cylindrical samples were prepared for uniaxial compressive strength tests. Effect of polymer addition to the strength of solidified waste form was determined.

Results show that polymer has a positive effect on leaching stability when compared to regular cemented radioactive waste form. Compressive strength values of

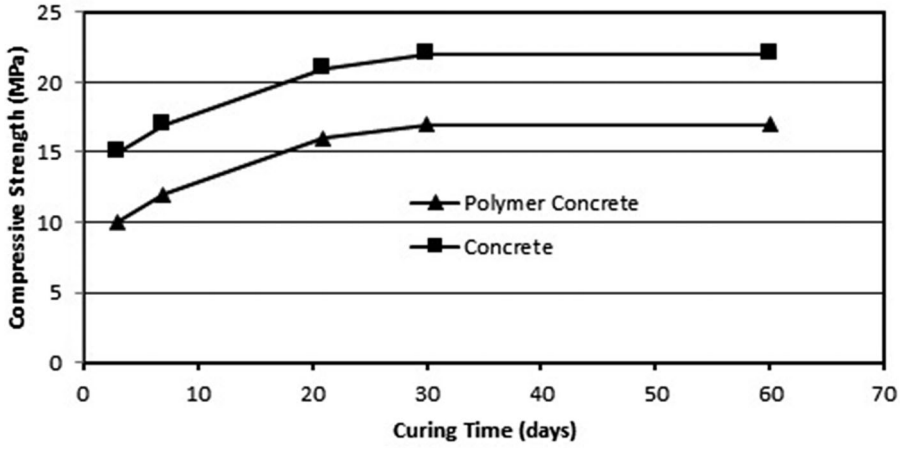


**Figure 5.** Compressive strength of solidified samples.

the concrete without polymer were measured and they were higher than those of polymer-added samples. The difference of these strength values was presented in Figure 5.

## Conclusion

Previous studies show that polymers can be used as an alternative sorbent material for the treatment of radioactive liquid waste.[9–20] Basic advantage of polymerization is minimization of operational contamination risks during waste management. Especially, polymers in microsphere scale can be used for decontamination operations in the nuclear industry and radioactive waste management. In these studies, these properties were used in solidification process of waste management as a novel study and two solidified radioactive waste forms were evaluated. Leaching rates and compressive strength values of the concrete without polymer were measured and compared. First form includes microspheres and second waste form was without polymer. Polymer-added solidified waste form has a significant advantage on leaching resistance. But decreasing of strength is disadvantage for the long-term integrity of polymer-added waste forms. Eventually high strength with low leach rate waste form production could be obtained by the optimal amount of polymer. Implementation of this method makes solidification technique easier and safer by using polymer microspheres in nuclear waste management. The previous studies in literature have shown that polymers have sufficient adsorption capacity of radionuclide, and this study shows that adsorption mechanism of polymers has similar effects on cementation process. The prospective work in future should investigate adsorption properties of other matrix materials in solidification process.

## Disclosure statement

No potential conflict of interest was reported by the author.

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