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Short Communication

Biosorption of no-carrier-added radionuclides by calcium alginate beads using 'tracer packet' technique

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Abstract

The aim of this study was to determine the adsorption behaviour of various micronutrient elements e.g., ^{61}Cu, ^{62,63}Zn, ^{66,67,68}Ga, ^{66,67,69}Ge, ^{71,72}As present in no-carrier-added state, with calcium alginate (CA) using 'tracer packet' technique. High Ge and Ga and moderate Cu removal were achieved at pH 7 and pH 5, respectively. Results on the studies to recover all the three radionuclides from the calcium alginate beads using desorbing reagents, HCl, thiourea, ammonium oxalate and sodium nitrite showed that 0.1 M HCl and 0.1 M ammonium oxalate removed Cu and Ge moderately. The amount of Ga desorbed by all the washing liquids was almost negligible, except sodium nitrite.
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Keywords: Tracer packet; Micronutrient elements; No-carrier-added; Biopolymer; Calcium alginate beads

1. Introduction

Biodegradable and hydrophilic biopolymers in the form of beads are promising materials competing with the conventional clean up technologies. Alginate is an exo-polymer of natural origin. It is composed of varying composition of β-1,4 linked D-mannurinic and L-guluronic acids. Presence of carboxylic group in the alginate moiety helps to form complexes with cationic metal ions and drawn tremendous attention of scientific community for concentrating metal ions in the alginate beads (Ozdemir et al., 2005; Nestle and Kimmichi, 1996; Min and Hering, 1998). Especially chain segments containing oligopolymeric blocks of guluronic acid exhibit selectivity for metal cations (Nestle and Kimmichi, 1996). Chen and Yiacoumi (1997) studied the sorption of Cu from aqueous solution by calcium alginate. Adsorption property of alginate was also investigated for Cu^{2+}, Co^{2+}, Zn^{2+}, Cd^{2+}, La^{3+} ions by Jang et al. (1991, 1999) and Konishi et al. (1993). However, a limited amount of information is available concerning the uptake behaviors of radioactive isotopes on this polymer gel. The uptake behaviors of ^{241}Am, ^{22}Na, ^{137}Cs, ^{59}Fe, ^{60}Co, ^{85}Sr and ^{152}Eu were studied using alginic acid and alginate polymer gels (Mimura et al., 2001). It was found that calcium alginate exhibited relatively higher uptake rate and distribution coefficient of Am^{3+} than the other metal ions. Quantitative removal of ^{152}Eu was also carried out with alginate bioplymers (Nayak, 2005). Adsorption of no-carrier-added heavy toxic radionuclides e.g., $^{197-200}$Pb, $^{197-200}$Tl and ^{197}Hg with calcium alginate beads was studied by Nayak and Lahiri (2006).

The introduction of "Tracer Packet Technique" by Lahiri and Nayak (2002) gave an impetus in green chemistry research. The "Tracer Packet Technique" has been successfully utilized to explore usefulness of natural reagents, especially in multielemental multidimensional environment, as replacement of established chemicals. This paper reports on the studies of adsorption of radioisotope of micronutrient element e.g., ^{61}Cu, ^{62,63}Zn, ^{66,67,68}Ga,

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^{66,67,69}Ge, ^{71,72}As present together in no-carrier-added state on calcium alginate beads.

2. Experimental

2.1. Preparation of Ca-alginate (CA) beads

Calcium alginate (CA) beads were prepared by drop wise addition of 3% sodium alginate solution into 20% $CaCl_2 \cdot 2H_2O$ solution with constant stirring maintaining a temperature of 4–7 °C. The resultant beads were allowed to stand in the mother liquor for 24 h in a refrigerator and beads were stored after washing with deionized water to remove $CaCl_2$ from the beads surface. $CaCl_2$, $2H_2O$ was obtained from E. Merck, India. Sodium alginate powder was obtained from Sigma Aldrich. All the chemicals were used without further purification.

2.2. Production and separation of no-carrier-added radionuclides

In order to produce 'tracer packet of micronutrient elements', a cobalt foil of thickness 23 mg/cm^2 was irradiated with 96 MeV ^{16}O beam for two hours (average beam current 100 nA), 48 MeV ^{7}Li beam for eight hours (average beam current 110 nA), and 84 MeV ^{12}C beam for two hours (average beam current 50 nA) at BARC-TIFR Pelletron (Lahiri and Nayak, 2002). The following reactions for production of 'tracer packet of micronutrient elements' using ^{16}O, ^{7}Li and ^{12}C beam were considered:

$$^{59}\mathrm{Co}(^{16}\mathrm{O},\mathrm{xn})^{71-73}\mathrm{Br} \xrightarrow{\varepsilon} {}^{71-73}\mathrm{Se} \xrightarrow{\varepsilon} {}^{71-73}\mathrm{As}$$

$$^{59}\mathrm{Co}(^{7}\mathrm{Li},\mathrm{xn})^{61,62}\mathrm{Zn} \xrightarrow{\varepsilon} {}^{61,62}\mathrm{Cu}$$

$$^{59}\mathrm{Co}(^{12}\mathrm{C},\mathrm{xn})^{66-69}\mathrm{As} \xrightarrow{\varepsilon} {}^{66-69}\mathrm{Ge} \xrightarrow{\varepsilon} {}^{66-68}\mathrm{Ga}$$

After irradiation, the cobalt target was removed from the irradiation chamber. The product radionuclides were detected with their corresponding γ-energy peaks. An HPGe detector of 2.13 keV resolution at 1.33 MeV in conjunction with a PC based MCA, PCA2 (OXFORD) was used for γ-spectrometric studies. Batch yield (in kBq) of the no-carrier-added radionuclides after 40 min of end of bombardment for ^{61}Cu, ^{62}Zn, ^{66}Ga, ^{67}Ga, ^{71}As, and ^{72}As were 260, 110, 68, 84, 24 and 42, respectively. Chemical separation of the no-carrier-added radionuclides from the cobalt target was made according to the method described earlier (Lahiri and Nayak, 2002). After the separation of the bulk cobalt, the aqueous phase contained only no-carrier-added radionuclides of ^{61}Cu, ^{62,63}Zn, ^{66,67,68}Ga, ^{66,67,69}Ge, ^{71,72}As, which were further used for adsorption studies onto calcium alginate beads.

2.3. Adsorption study

Adsorption was examined in batch mode. For each batch experiments, 30 CA beads were exposed to a 3 mL of HCl solution containing a measured activity of no-carrier-added radionuclides of Cu, Zn, Ga, Ge and As. The pH of the final solution (i.e., mixture radionuclide + beads) was maintained by dropwise addition of dilute HCl or NH_3. To study the effect of pH on the adsorption of radionuclides in CA beads, different pH levels of 3, 4, 5, and 7 were maintained at the beginning of the experiment. Beads were equilibrated in the respective pH solution for 30 min before exposing to the radionuclide solution. To study the effect of exposure time beads were exposed for 30 min, 1 h, 5 h, 6 h, and 7h to radionuclide solution at room temperature (25 °C).

A 2 mL fraction of the supernatant liquid was removed in all the cases, and assayed for radionuclides by means of gamma spectrometry with a HPGe detector. The activity obtained was compared to the activity of the standard solution, containing the same amount of radionuclide without CA. The radionuclides were monitored by their characteristic photo peaks, e.g., ^{61}Cu(282.95 keV), ^{62}Zn(596.7 keV), ^{66}Ga(1039.35 keV) ^{69}Ge(1106.77 keV) and ^{71}As(174.94 keV). The effect of number of CA beads on the adsorption rate of the radionuclides at pH 5 was investigated in batch experiment with 30, 60 and 90 CA beads.

2.4. Desorption study

Five desorption agents (0.01 M HCl, 0.1 M HCl, 0.1 M ammonium oxalate, 0.1 M sodium nitrite, 0.1 M thiourea at pH 1) were used to study the desorption properties of the ^{61}Cu, ^{62}Zn, ^{66,67}Ga, ^{66,69}Ge and ^{71}As radionuclides from CA beads. After adsorption the supernatant liquid was removed and the beads were washed with deionized water till pH 7. For all desorption experiments, CA beads were shaken with the above reagents for 30 min at room temperature (25 °C). Percentage desorption was calculated as the percentage release of radionuclide initially bound to the beads. Appropriate decay corrections were made in each set of experiments.

3. Results and discussion

3.1. pH variation

Fig. 1 presents the adsorption of Cu, Zn, Ga, Ge and As radionuclides at various pHs after 7 h of exposure time. CA significantly adsorbed ^{61}Cu (65%), ^{66,67}Ga (80%) and ^{66,69}Ge (70%) radionuclides in acidic pH range 3–5. Adsorption of ^{61}Cu and ^{66,67}Ga increased with increasing pH in the acidic pH range. Quantitative adsorption of ^{66,69}Ge was found at pH 7. The pK values of guluronic and mannuronic acids are 3.65 and 3.38, respectively (Chen et al., 1997). These functional groups may be in protonated form for low pH values and in anionic form in high pH values. Thus, a higher adsorption for Cu, Ga was at pH 5 and that of Ge was at pH 7. Ga formed anionic species $Ga(OH)_4^-$ and neutral species $Ga(OH)_3$(aq) at pH 7 (Verweij, 2005), which could not be attached with the COO^-

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