EVALUATION OF CHITOSAN-TiO_2/ ZrO_2 MICROSPHERES TOWARDS ITS POTENTIAL APPLICATION IN ^{68}Ge -^{68}Ga GENERATOR SYSTEM

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Efforts were made in the present studies to synthesize the macro-porous chitosan- TiO_2/ ZrO_2 microspheres with the enhanced ability to adsorb the ^{68}Ge. Aiming the separation and purification of ^{68}Ga from its mother ^{68}Ge we further studied the distribution coefficients of ^{68}Ge and ^{68}Ga on the chitosan- TiO_2/ ZrO_2 microspheres in the varying concentration of HCl (0.01 - 1.0 M).The experiments revealed that the uptake of ^{68}Ge/^{68}Ga was strongly dependent on the concentration of HCl and it was possible to separate the considerable amount of ^{68}Ga in both the cases using 0.01 M HCl. The present work can be considered as the initial step towards setting up the ^{68}Ge-^{68}Ga generator system using chitosan- TiO_2/ ZrO_2 composites at the nuclear medicine department.

I. INTRODUCTION

With the capabilities to be produced onsite using radioisotope generator and possessing suitable nuclear as well as radiochemical properties ^{68}Ga radiometal seems to play a pivotal role in near future as a PET isotope.[1] Several generator systems containing various organic and inorganic extractants as the stationary phases for the separation and purification of ultra-high purity ^{68}Ga have been studied in the past.[1-2] However, development of the new and stable material for the isotope production is inevitable processes for the researchers involved. TiO_2 (Ref. 3) and ZrO_2 (Ref. 4) have been used as suitable generator materials and chitosan composites have shown promises towards preconcentration and separation of multitudinous metal ions.[5-6] Considering this common precedence we thought of synthesizing chitosan - TiO_2/ ZrO_2 microspheres for its application in ^{68}Ge-^{68}Ga generator system. A preliminary attempt has been made in the present work to obtain the distribution coefficients of ^{68}Ge and ^{68}Ga on chitosan- TiO_2/ ZrO_2 to check its feasibility as a sustainable generator material.

II. EXPERIMENTAL

II.A. Synthesis of chitosan- TiO_2/ ZrO_2

In the first step, 0.5 g of chitosan was stirred for 24 hours in 50 mL of 5% acetic acid. In second step solution was prepared by addition of 12 mL of ethanol to 0.8 mL of DW and then 2 mL of conc. HCl to which titanium tetraisopropoxide (15 mL) / zirconium phosphate (0.35 g) was added (stirred for 1 hour). Later to this solution 4.75 g of metal precursor P25/ Zr $(HPO_4)_2$ was added and stirred for 15 hours. After this, both the solutions (I step + II step) were mixed for 2 hours. This mixture was then dropped in 28% ammonia solution. The spherical adsorbents were filtered and dried for 6 hours at 80 ^{0}C. Finally, prebuilt materials were sintered for 3 hours at 450 ^{0}C.

II.B. Distribution Coefficients (K_d)

K_d determination was carried out using ^{68}Ge-^{68}Ga with chitosan- TiO_2/ ZrO_2 microspheres. Solutions with desired radioactivity and acidity (0.01- 1 M) of HCl were prepared. 2mL of the solution was agitated in shaking incubator for 1.5 hours with 50 mg of chitosan- TiO_2/ ZrO_2 microspheres. After equilibration, the solution was separated using 0.22 µm PVDF syringe filter and 1 ml of the filtrate was counted for ^{68}Ga activity using gamma counter. Samples were recounted after 48 hours to assay the presence ^{68}Ge if any in the same samples indirectly. K_d was calculated using the formula:

$$K_d = \frac{(C_0 - C)/W}{C/V}\ mL/gm$$

Where C_0 and C are the initial and equilibrium activity of ^{68}Ga, W is the weight of chitosan- TiO_2/ ZrO_2 microspheres and V is the total volume of the acidic phase used during the equilibration experiment.

III. RESULTS AND DISCUSSIONS

As depicted in figure-1 below it is notable that the K_d values of ^{68}Ge as well as ^{68}Ga are strongly dependent on the concentration of HCl and a sharp decrease in K_d value is observed with the increasing acidity range in both the cases. Favorable extraction of the metal ions at lower acidity is a predictable phenomenon with the low H^+ ion concentration and interference. With the separation factor (S.F.) of 15.78 at 0.01 M of HCl, chitosan-TiO_2 appears to be more encouraging as compared to that with chitosan-ZrO_2 (S.F. = 5.4). However, the material under consideration needs to be analyzed under different experimental conditions viz. long term acidic and radiolytic stability, column studies including loading and elution profiles, radionuclidic and radiochemical purity of the obtained ^{68}Ga for radiopharmaceutical applications and life cycle assessment for its further implication as the generator material.

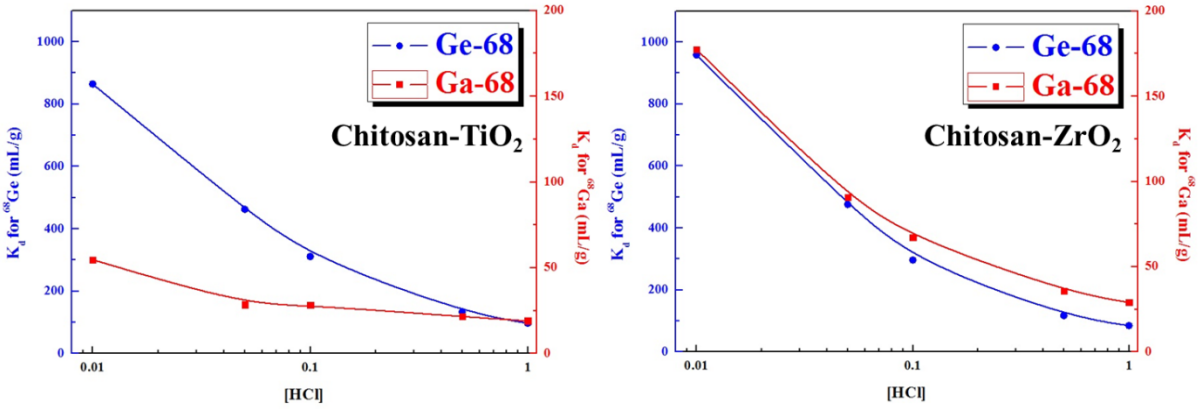

Fig.1. K_d variation of ^{68}Ge/^{68}Ga as function of HCl concentration (C_0 ≈ 300,000 CPM; W: 50 mg; V: 2 mL; T: 298 K).

IV. CONCLUSIONS

A preliminary study with spherical macro-porous chitosan- TiO_2/ ZrO_2 microspheres shows its high retention capabilities towards ^{68}Ge vis-à-vis ^{68}Ga at 0.01 M HCl. With the higher S.F. values, chitosan-TiO_2 appears to be the more promising candidate as a generator material. The further optimization of the process would be carried out to develop efficient generator system for the separation of ^{68}Ga from a secular equilibrium mixture of ^{68}Ge-^{68}Ga using these two materials in HCl medium.

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