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Performance of $^{113}Sn/^{113m}In$ Generator Prototype based on Zirconium Oxide for Radiotracer Applications in Industry

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Abstract: *This paper describes the results of the research of $^{113}Sn/^{113m}$ radioisotope generator at the Centre for Applied Nuclear Science and Technology - National Nuclear Energy Agency with the aim of making radioisotope generator for radiotracer applications in industry. This research discussed the need of short half-life radiotracer for several material phases such as oil, water, gas and solid. Based on desired physical properties of half life, radiation type and energy, ^{113m}In radioisotope was selected. Thus, the performance of $^{113}Sn/^{113m}$In generator prototype based on zirconium oxide was determined for this purpose. The final product specification in the form of $^{113m}InCl_3$ is clear solution, pH 2 with obtained yield of 95 %, radionuclide purity of 95 % and radiochemical purity of 95 %.*

INTRODUCTION

Radiotracer is a nuclear engineering method using radioisotopes that are excellent for measuring flow and tracking leaks in industrial facilities. Among the advantages of radiotracer techniques are high accuracy, low detection limits, measurement without disturbing the processing system. Until now, the use of tracing methods using radioisotopes with a short half-life is still relatively few. The problem is that they cannot be used in locations which are far from nuclear facilities, because their short half-life limits the time for transport the tracer to measurement site. Therefore, the radiotracer availability with required radioactivity at measurements/investigations site is one of the most important challenge for the utilization of tracer based on short-lived radioisotope (1,2).

The existence of a radioisotope generator is an effective strategy in solving practical problems to facilitate the measurement in locations that far from nuclear facilities. The right selection of radioisotope generator will avoid problems associated with radiotracer, such as radioactive waste and radiation exposure which can be minimized or even eliminated.

Radioisotope Generator

A radioisotope generator is a chemical / physical / mechanical functioning device that is based on the parent-daughter radioisotope relationship. In radioisotope relationships the parent produces daughter radioisotopes by means of radioactive decay. In a radioisotope generator, the core properties of the parent radioisotope must have a longer half-life than that of the daughter radioisotope. This requires that the daughter radioisotopes will grow until equilibrium is reached between the two radioisotopes.

The relationship of parent-daughter radioisotopes with a much longer half-life of the parent than that of the daughter is called secular equilibrium, or the half-life of the parent being equal to or slightly longer than that of the daughter is called transient equilibrium. The most useful system in radioisotope generators for applications in industry away from nuclear facilities is the secular equilibrium (2). Due to the nature of the radioactive decay between parent and daughter there will be many different chemical elements and therefore can be chemically, physically or mechanically separated. The generator system of most parent radioisotopes is attached to a column based on a matrix of arsorbenic material, and the daughter radioisotopes are eluted with a liquid flowing through the column leaving the parent radioisotope. The isolated daughter radioisotope

can then be used as a direct tracer or chemically bonded in molecular form.

Selection of Radioisotope Generator.

Many of the physical properties of a radioisotope generator are governed by the half-lives of the parent and daughter radioisotopes. If the half-life of the parent is longer than that of the daughter, the daughter radioisotope can be repeatedly eluted because the parent radioisotope will produce daughter radioisotopes continuously. Beside parent radioisotope's half-life, generator lifespan is also regulated by the possibility of radiolysis and other chemical processes that can reduce the milking efficiency and/ or induce the escape of the parent radioisotope, thereby negatively affecting the safety of generator operation. The operating period of radioisotope generators for many practical systems is limited to 2-3 times of half-lives of the parent radioisotope.

The half-life of the daughter radioisotope regulates how often the generator can be eluted and how long the daughter radioisotope will be available as tracer. The shorter half-life means the radioisotope will decay more rapidly and this limits tracer use, but it also has advantages associated with radioactive waste.

Indium-113m radioisotope, ^{113m}In.

The ^{113}Sn - ^{113m}In generator is commercially available for industrial use with an operating time of about 0.5 - 1 year, that is, based on the parent radioisotope half-life (^{113}Sn) of 115 days. The parent radioisotope (^{113}Sn) decays to produce a daughter radioisotope (^{113m}In) which has a half-life of 1.7 hours and emits a gamma radiation energy (g) of 392 keV from the internal transition from the ^{113m}In (isomeric state) to the ^{113g}In non-radioactive (ground state). The radioactive decay kinetics and equilibrium times for parent radioisotopes that have a much longer half-life than daughter radioisotopes are called secular equilibrium (fig.1) (2,3).

Figure (Fig.1.a) shows that the radioactivity of the parent remains almost constant, while the radioactivity of the daughter grows until equilibrium with the radioactivity of the parent. ^{113m}In daughter radioisotope every 14 hours will grow about 80% of the total radioactivity of the generator (parent radioisotope), and after more than 20 hours, ^{113m}In radioactivity will be run out. The Figure (Fig.1.b) shows the maximum equilibrium time to produce a daughter radioisotope ^{113m}In every 18 hours (4,5).

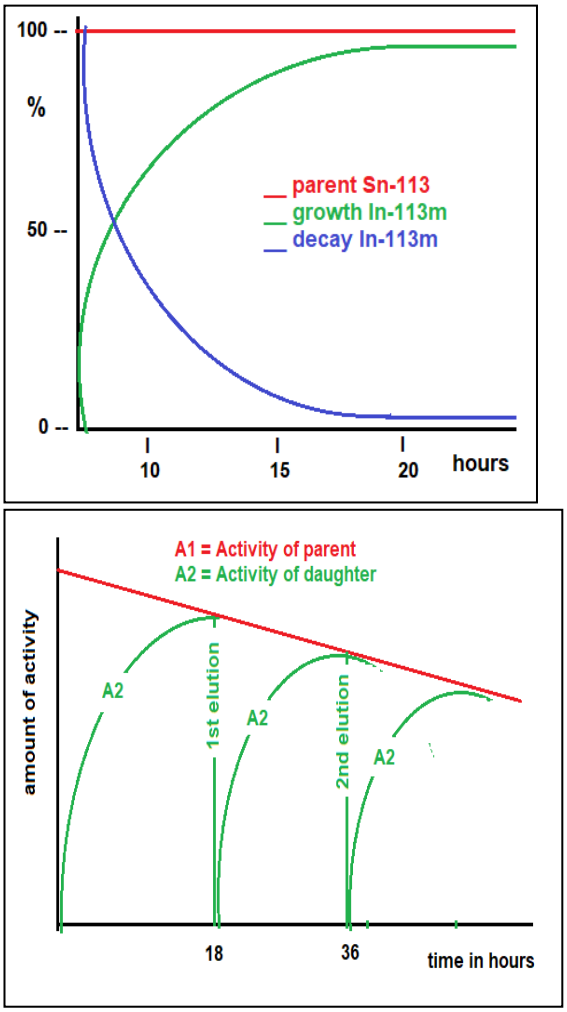

Figure 1. Growing and decay of the eluted ^{113m}In in the generator ^{113}Sn – ^{113m}In: (Top). secular equilibrium, (Bottom). maximum equilibrium time.

EXPERIMENTAL SECTION

Materials

The materials used in this study are tin (Sn) metal from E. Merck, hydrochloric acid, zirconium oxide (100-200 mesh) (6), hydrogen peroxide (30% H_2O_2), distilled water, ammonium molybdate (1%), phosphate buffer (Na_2HPO_4), universal pH indicator paper, dry methanol, ITLC chromatography paper, aluminum inner / outer capsule.

Instrumentation.

Meanwhile, the equipment used in this study are mettler toledo AL 204 analytical balance, Nouva II heater (Thermolyne), ORTEC model 402 A single channel (SCA) counter, multi channel γ spectrometer counter (MCA) with

HPGe detector Canberra DSA-1000, Heraeus T5050 oven, TRIGA 2000 nuclear reactor BATAN Bandung, hot cell, box glove..

The $^{113}Sn/^{113m}In$ prototype design based on zirconium oxide

Zirconium dioxide (ZrO_2) with a size of 100-200 mesh (± 1.5 grams) was inserted into the glass column (Figure 2). Then the column and its contents were conditioned with HCl 0.05 N then the stock solution ($^{113}Sn/\ ^{113m}In$) was passed through the column. The column was eluted with 10 mL HCl 0.05 N. The eluate was stored 1 mL each in a 10 mL vial (n fraction = 10). The separated eluate was stored as a $^{113m}InCl_3$ solution.

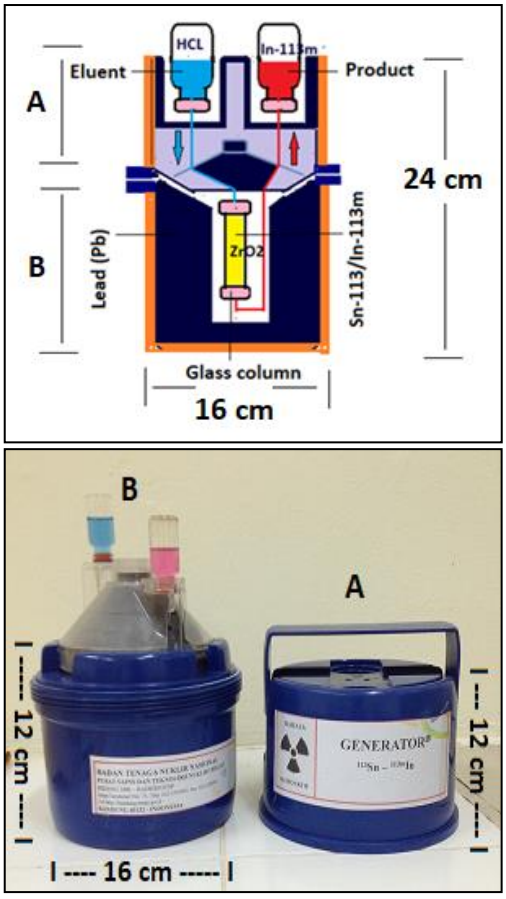

Figure 2. (Top). Design of ZrO_2 column, shielding lead, (Bottom). Prototype of $^{113}Sn/^{113m}In$ generator system.

^{113m}In radioisotope characterization and elution profile

Determination of radionuclide purity using a spectrometer-γ HPGe-MCA. 10 µL of prepared radionuclide $^{113m}InCl_3$ in 1mL vial was counted for 300 seconds. γ-ray energy of $^{113m}InCl_3$ at peak energy of 393 keV. Determination of radiochemical purity using paper chromatography method with the ITLC I as stationary phase and dry methanol as mobile phase. Determination of radiochemical purity was carried out by dripping a $^{113m}InCl_3$ solution at a distance of 2 cm at the bottom of the chromatography paper, then the paper was inserted into the chamber which was saturated by the mobile phase. Elution was carried out until the mobile phase migration distance reached 16 cm. The chromatogram was dried, cut every 1 cm and counted with γ-sepctrometer-SCA for 60 seconds. The value of Rf was determined by comparing the migration distance traveled by the $^{113m}InCl_3$ to the migration distance of the mobile phase.

RESULTS AND DISCUSSION

The elution profile of the ^{113}Sn - ^{113m}In generator uses ZrO_2 matrix column

The separation results are shown as ^{113m}In elution profile in Figure 3.

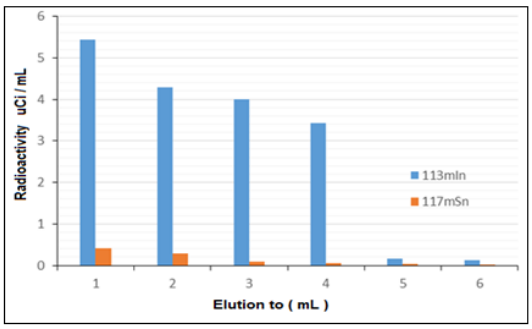

Figure 3. The elution profile of the ^{113m}In generator system $^{113}Sn/^{113m}In$

Figure 3 shows the ^{113m}In radioisotope elution which has a high radioactivity in the 1st fraction (elution), while the 2nd, 3rd, 4th fraction and so on show a decrease in radioactivity. The result of the calculation is that the percent yield for separation of ^{113m}In is 95%. Figure 3 showed the presence of other radionuclides than ^{113m}In which formed, were ^{117m}Sn radionuclides which derived from the core reaction of their isotopes by neutron at irradiation process. ^{117m}Sn came from the decay of irradiated result ^{116}Sn (natural isotopic abundance of 14 %). ^{116}Sn isotopes were contained in metal Sn target (7).

^{113m}In radioisotope characterization

Radionuclide purity of ^{113m}In

The purity of radionuclides in the $^{113m}InCl_3$ was tested using the MCA-HPGe method

with the gamma ray spectrum results as shown in Figure 4:

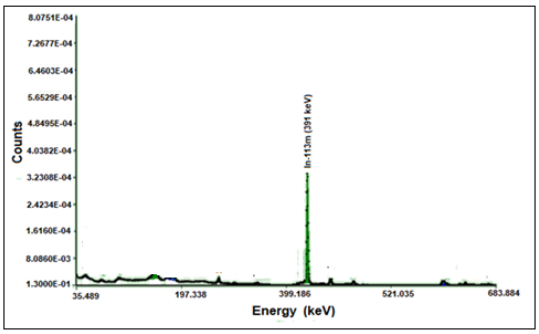

Figure 4. Gamma ray spectrum for radioisotope $^{113m}InCl_3$

Figure 4 shows the characteristic gamma ray spectrum of $^{113m}InCl_3$ at a peak energy of 393 keV. This is consistent with the explanation that the radioisotope ^{113m}In has a half-life of 1.6 hours on a gamma emition (393,675 keV) (8). The results of the analysis and calculations from the experiment showed that the radionuclide purity (KRN) was above 95%.

Radiochemical purity of ^{113m}In

Using the ITLC stationary paper chromatography method with dry methanol as the mobile phase. ^{113m}In radioisotope chromatogram as shown in Figure 5:

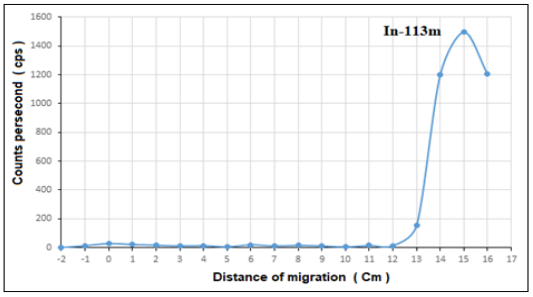

Figure 5. ^{113m}In chromatogram using ITLC and dry methanol

Figure 5 shows the radioisotope ^{113m}In migrated to reach Rf values of 0.7-0.8 (migration distance of 13-15 cm, using the ITLC stationary phase and dry methanol mobile phase) and the radiochemical purity was above 95%.

CONCLUSION

$^{113}Sn/^{113m}In$ radioisotope generators for industrial needs is based on this research results provide commercial performance. ^{113}Sn - ^{113m}In generator, meet the requirements regarding the final product specification obtained ^{113m}In radioisotope in chemical form $^{113m}InCl_3$, clear solution, pH at 2, separation yield 95%, radionuclide purity of 95% and radiochemical purity of 95%.

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