

Feasibility Study on the Radionuclide Generator Based Radiotracer for the Industrial Process Diagnosis Technology

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1. Introduction

Radioactive tracers have been widely used throughout industry for almost 70 years to solve problems, improve productivity, save energy and reduce pollution [1]. However, in spite of their manifest benefits, radiotracer techniques continue to be underutilized, not only by developing countries but also by more industrialized nations. There are a number of factors that restrict the usage of radioisotope techniques, but chief among them is the timely availability of the tracers themselves. Firstly, it is hard to deliver the short-lived radioisotopes to industry field. Secondly, there is no possibility of offering industry a quick response to problems of an urgent nature.

Making use of tracers from radionuclide generators can alleviate the difficulties associated with radioisotope supply. Medical radionuclide generators such as ⁹⁹Mo/^{99m}Tc and ¹¹³Sn/^{113m}In provide a partial solution to the problem of industrial radiotracer supply, but unfortunately the radioisotopes they produce have limited applications in industrial and environmental radiotracer studies because of their relatively short shelf lives and/or relatively low gamma energies.

In this paper, the feasibility studies were performed with prototype of ⁶⁸Ge/⁶⁸Ga and ¹³⁷Cs/^{137m}Ba generators which produced in China (Atom Hightech Co.).

2. Methods and Results

2.1 Performance test of the generators

The generators were eluted by a peristaltic pump for elution efficiency test. The elution efficiency is defined as ratio between the activity from designed specification and activity of the daughter nuclide by elution. In order to measure activity of the eluted daughter nuclide, dose calibrator (Capintec, INC., CRC-25R) was used. The breakthrough is defined as ratio between activity of daughter nuclide and mother nuclide in an eluted nuclide. For measuring the breakthrough of the mother nuclide, the activity of the mother nuclide in the eluted sample was measured by a calibrated HPGe detector (Canberra, GC2019) after long time passed from elution not to be interfered by daughter's presence. In addition to elution efficiency test and breakthrough, elution profiles were measured. The concentration of daughter nuclide varied in its eluted point during the elution. The concentration of daughter nuclide according to the amount of eluent was

measured as the elution profile. The characteristics of the generators are shown in table 1.

Table 1. The characteristics of the generators

Generator	Activity	Elution efficiency	Breakthrough h	Main energy
Ge/Ga	20mCi	52~67%	$< 10^{-6}$	511keV
Cs/Ba	50mCi	17~25%	$< 10^{-5}$	662keV

2.2 Preparation and test of Ga-DOTA

In order to stabilize the Ga³⁺ ion in water phase, the macrocyclic chelate DOTA (1,4,7,10-tetraazacyclododecane-1,4,7,10-tetraacetic acid) was used. After synthesis the thin layer chromatography (TLC) was performed on 0.8×7.5 cm TLC plates (Kieselgel 60 F-254, Merck) with 75% methanol. The radiation depending on the position was measured by TLC-radioisotope scanner (TRACERMASTER20, Berhold). Fig. 1 shows TLC chromatograms of Ga ion and Ga-DOTA complex. In case of Ga ion, Ga ion remained the spotting point of plate (R_f=0). On the other hand, there is no significant peak at origin with DOTA. The Ga moved to center of plate (R_f=0.45). It means that Ga ion become Ga-DOTA complex.

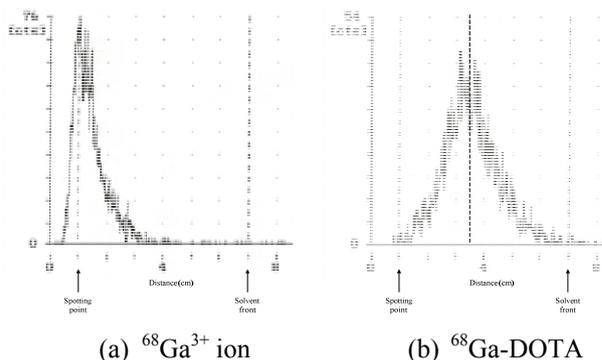


Fig. 1 TLC chromatograms of ⁶⁸Ga ion and ⁶⁸Ga-DOTA

2.3 Industrial emission tomography experiments with IRG tracers

In order to examine the spatial distribution of tracer, we used the radionuclides from the generators (Cs/Ba, Ge/Ga) for the emission tomographic tests with the In-SPECT (industrial single photon emission computed tomography) system [2]. The In-SPECT system consists

of 30 detectors, 30 single channel analyzers and the DAQ system (Fig. 2). The detectors are 2×2 inch NaI(Tl) scintillator coupled with PM-tube and shielded with Pb collimator to minimize scattered radiation detection. The measured counts in each detector were processed with the expectation maximization (EM) algorithm [3] that reconstructs the cross-sectional image of the distribution of radiotracer in the vessel. The experiments were carried out for the static phantom and dynamic flow. A cylindrical vessel was filled with water ($\Phi = 60$ cm) containing two cylindrical sources. These cylindrical sources were placed with 17 cm interval. Table 2 shows RI concentration of static phantoms and counting mode.



Fig. 2 Experimental apparatus of Industrial Single Photon Emission Tomography

Table 2. The experiment conditions for the In-SPECT tests with the radiotracer

No.	Nuclide (energy, keV)	Counting mode	RI specific activity(mCi/L)	
			Pipe 1	Pipe 2
1	^{68}Ga (511)	Gross	2.2	-
2	^{68}Ga (511)	Gross	1.1	3.0
3	$^{137\text{m}}\text{Ba}$ (662)	Gross	6.2	10.4
4	$^{137\text{m}}\text{Ba}$ (662)	Photopeak	4.0	11.0

Fig. 3 shows the reconstructed images by the In-SPECT for four different cases: (a) only the cylinder 1 is filled with ^{68}Ga , (b) ~ (c) both cylinders are filled with ^{68}Ga and $^{137\text{m}}\text{Ba}$, respectively and (d) both cylinders are filled with $^{137\text{m}}\text{Ba}$ and counting mode is only photopeak collecting. In case of (a), the image quality is marginally acceptable; however, in cases of (b) and (c), the image quality is hardly acceptable and it is almost impossible to figure the distribution of the radioactive tracer from the given image. In case of (d), two cylinders are distinguished and concentration difference of tracer is shown also. The performance of In-SPECT system was significantly deteriorated by the scattered photons.

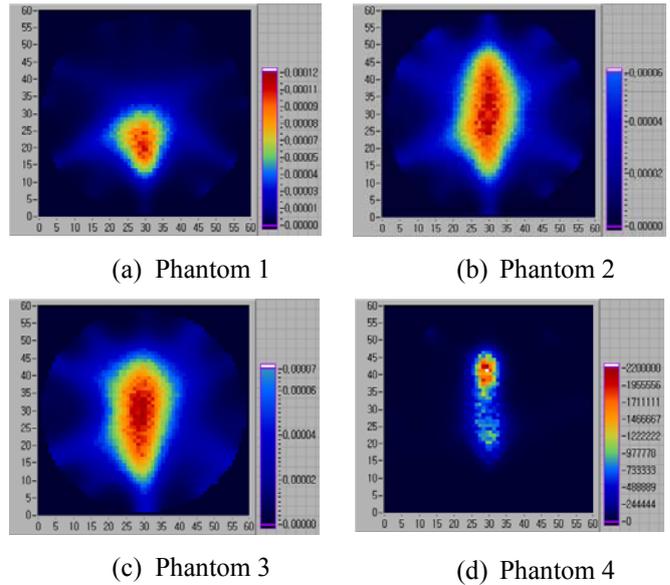


Fig. 10 The spatial distribution of radiotracer

3. Conclusions

Two different kinds of radioisotope generators supplied by the IAEA, $^{68}\text{Ge}/^{68}\text{Ga}$ and $^{137}\text{Cs}/^{137\text{m}}\text{Ba}$, were tested in terms of the elution efficiency, breakthrough and the potential for the industrial applications. The generators have potential possibilities as industrial tracers and were used as tracers in the industrial emission tomography experiments. The reconstructed images of tracer distribution were successfully obtained by In-SPECT system.

Acknowledgement

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REFERENCES

- [1] G. Axelsson, B. J. Barry, P. Berne, T. Bjørnstad, R. Cameron, S. Charlton, G. E. Maggio, Z. Pang, J. Thereska, X. Vitart, Radiotracer Applications in Industry – A Guidebook, IAEA Technical Report Series No. 423, IAEA, Vienna, Sept. 2004.
- [2] S. Legoupil, G. Pascal, D. Chambellan, D. Bloyet, Determination of the Detection Process in an Experimental Tomograph for Industrial Flow Visualization Using Radioactive Tracers, IEEE Transactions on Nuclear Science, Vol. 43(2) p. 751-760, 1996.
- [3] K. Lange, R. Carson, EM reconstruction algorithms for emission and transmission tomography, J. Comput. Assist. Tomogr. 8(2) p. 306-316, 1984.