

Environmental Water and Urine Sample ^{99g}Tc Analysis Methodology by LSC

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1. Introduction

Technetium-99m (^{99m}Tc) is used in the medical field for diagnosis purposes with a large number of organic carriers. These include ^{99m}Tc -mercaptoactyltriglycine[1], ^{99m}Tc -gluconate[2], ^{99m}Tc -dimercaptosuccinic acid [3,4], ^{99m}Tc -methylene-diphosphonate, ^{99m}Tc -dicarboxypropane-diphosphonate, ^{99m}Tc -ethylenediaminetetramethylene-phosphonate, and ^{99m}Tc -pingyangmycin. ^{99m}Tc is used in 80% of diagnostic nuclear medicine procedures. Almost 30 million examinations are conducted worldwide using this isotope, because ^{99m}Tc has a half-life of 6 hours and decays to ^{99g}Tc . Monitoring and safe disposal of ^{99g}Tc from human urine is very important and concern is increasing every day as global use of ^{99m}Tc has increased by more than 4.5×10^{14} Bq per week and it is increasing continuously. In 2020, demand is expected to increase 32% as compared to current market demand. Patients' excretions are discharged to sewer systems without quantification of ^{99g}Tc due to lack of a rapid procedure. Most of the existing ^{99g}Tc analysis procedures are based on purification of the sample matrix, which requires a trained analytical chemist in a hospital or nearby analytical laboratory. Keeping in view all the practical problems of hospitals and laboratories, a simple and direct analytical method needs to be developed for quantification of ^{99g}Tc in urine as well as municipal waste samples. Previous methods have high minimum detectable activity (MDA) and low figure of merit (FOM).

2. Material and Method

Technetium-99 (which was in chemical form of ammonium pertechnetate, NH_4TcO_4) stock solution of 10 Bq/mL was prepared in DIW from a ^{99g}Tc

ampoule having activity of 185,000 Bq/mL. A working solution of 1 Bq/mL was prepared in DIW. Radioactive solutions of ^{99g}Tc (1 mL) sample having activity 1 Bq/mL were mixed with 15 mL of ULG-LLT cocktail in 20 mL volume glass and polyethylene vials, separately. Blank solutions were also prepared using 1 mL of DIW and 15 mL of ULG-LLT in both glass and polyethylene vials. Using ULG-LLT standard and blank preparation procedures, one ^{99g}Tc standard and one DIW blank solution were prepared with 10 mL of ULG cocktail in glass vials. ULG cocktail was not used at volumes greater than 10 mL because more than 10 mL of ULG produces a high background. Both of these standard solutions were used to optimize the LSC counting methodology for all LSC parameters.

3. Results and Discussion

3.1 Comparison of Cocktails

Among the factors which control the development of the radiometric method, the first thing is to select the right choice of scintillator and its quantity. Careful selection of the cocktail and its volume used influences the radionuclide measurement efficiency. The most important variables governing the superiority of any counting method are B, MDA, ϵ , and FOM. Both B and MDA should be as low as possible, whereas ϵ and FOM should be as high as possible. Ultima Gold was proved to have higher FOM as compare to ULG-LLT in cleaner matrix.

3.2 Environmental Sample Analysis

All samples (Urine, tap water, ground water, sea water, deionized water) were spiked with 1 Bq/mL of $\text{NH}_4^{99g}\text{TcO}_4$, and their activities were found to be

within a very satisfactory range. Error was in the range of ± 0.02 Bq (1.96%) to 0.04 Bq (4%) in five samples for both cocktails. This study shows the validity of the method and quench-curve accuracy for calculating unknown activity of ^{99g}Tc in various samples.

3.3 Quench Variation

The presence of any impurity (chemical or color substances and solution opacity severely affects the yield of light and leads to incorrect quantification of radioactivity. Effects of both inorganic and organic impurities were calibrated for ULG and ULG-LLT. Organic quench has shown more depreciation in efficiency than inorganic matter. Nitromethane was used as quench to calibrate and draw quench curve.

3.4 Inorganic Quench

There are significant changes in sensitivity of the method with artificial organic quench. However, environmental water samples did not show prominent change in sensitivity when these were monitored in the presence of higher TDS. There was no change in tSIE and little change in efficiency with increase in TDS from 1.9 to 11900 ppm. This indicates that inorganic ions present in water samples do not produce appreciable quench. However, organic materials produce higher quench.

3.5 Comparison with Previous Studies

MDA of method reported by Wigley et al. is slightly lower than our method. However our method is better in terms of efficiency, variety of cocktails, much short analysis time, variety of sample matrices, less expensive counting instrument, and emergency analysis. Our method is also better than the method reported by Villar et al.¹⁰ in terms of low MDA, variety of cocktails, no pre-count delay as well as range of sample matrix. Using our method, many samples can be analyzed in a short time period because our method has relatively short sample preparation times. For our method, it is not required to isolate ^{99g}Tc from the sample matrix whereas Villar et al.¹⁰ method needs an automatic isolation assembly which increases the cost of the analysis as well.

4. CONCLUSION

This method emphasizes the selection of cocktail, depending upon the chemistry of matrix and nature of impurities present in various samples. It was revealed that in this study, in order to get better sensitivity and lower MDA, ULG is suitable choice for those matrices which are cleaner with negligible quench species. However for quench containing matrices, ULG-LLT performs better than ULG-LLT. Hence samples like urine are better analyzed with ULG-LLT whereas samples like DIW, Tap water, River, and ocean water are suitable with ULG for ^{99g}Tc analysis

5. REFERENCES

- [1] Itoh, K. *Annal. Nucl. Medi.* **2001**, *15*, 179-190.
- [2] Council of Europe. *European, pharmacopoeia*, 5thed.; Council of Europe, 2004.
- [3] Weyer, K.; Nielsen, R.; Petersen, S. V.; Christensen, E. I.; Rehling, M.; Birn, H. *J. Nucl. Medi.* **2013**, *54*, 159-165.
- [4] Verboven, M.; Ingels, M.; Delree, M.; Piepsz, A. *Pediat. Radiol.* **1990**, *20*, 540-542.
- [5] Wigley, F.; Warwick, P. E.; Croudace, I. W.; Caborn, J.; Sanchez, A. L. *Anal. Chim. Acta* **1999**, *380*, 73-82.
- [6] Villar, M.; Avivar, J.; Ferrer, L.; Galmes, M.; Vega, F.; Cerda, V. *Anal. Chem.* **2013**, *85*, 5491-5498.