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LSC methods for analysis of radionuclide impurity of ^{99m}Tc eluate in quality control of ^{99}Tc-^{99m}Tc generators

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The radionuclidic impurities in ^{99m}Tc eluate (sodium pertechnetate injection) used in the medical diagnosis and therapy, especially the long-lived radionuclides is a major concern in the application of this product, because these impurities might interfere the efficiency of diagnosis and therapy, meanwhile the injection of radionuclide impurities will also cause an extra radiation to the patients. Therefore, it is required by the medical authorities that ^{99m}Tc eluate from the ^{99m}Tc-^{99}Mo generator has to contain limited amount of radionuclides other than ^{99m}Tc. According to Ph.Eu Monograph [1] , the major concerning radionuclides in the sodium pertechnetate injection (^{99m}Tc eluate) from a ^{99m}Tc-^{99}Mo generator produced by fission reaction of uranium are ^{99}Mo, ^{131}I, ^{103}Ru, ^{89}Sr, ^{90}Sr, alpha emitting impurities and other gamma (beta) emitting impurities. As a quality control of the ^{99m}Tc-^{99}Mo generator, the ^{99m}Tc eluate from each batch of generator has to be analyzed for the above mentioned radionuclidic impurities. Among all these radionuclides, the gamma emitters such as ^{99}Mo, ^{131}I and ^{103}Ru can be directly measured using gamma spectrometry after 3-4 days cooling of the eluate to remove most of ^{99m}Tc (6.5×10^4 by 4 days through radioactive decay). This work presents a method of chromatographic separation combined with liquid scintillation counting (LSC) measurement for measure beta emitters (^{89}Sr, ^{90}Sr) and total alpha and beta activities.

1. Removal of ^{99m}Tc and ^{99}Mo for determination of total alpha and beta activities

Due to the very high radioactive concentration of ^{99m}Tc in the eluate (1-10 GBq), and might also high ^{99}Mo due to the leakage. An anion exchange chromatographic method was applied to remove ^{99m}Tc and ^{99}Mo in order to measure total alpha and beta activities. The eluate prepared in 0.1M HCl – 0.9%NaCl solution was loaded to a 2 ml anion exchange column (AG1-×4, 50-100 mesh) [2], and the column was washed with 2 ml of 0.1 m HCl solution, the effluent and wash are combined for measurement of total alpha and beta activity and used for further radioactive strontium separation. It was found that the decontamination factor of this procedure to ^{99m}Tc and ^{99}Mo is higher than 10^4, while the most of other possible alpha emitters such as uranium and plutonium isotopes (from the irradiated enriched uranium) are remain in the solution, the recovery of Pu and U are higher than 98%. Meanwhile the most possible beta emitters including ^{89}Sr, ^{90}Sr, ^{134}Cs and ^{137}Cs are not absorbed on the column, it was observed that >98% strontium and caesium remain in the effluent and washes. However, it should be mentioned that ^{131}I, and part of ^{103}Ru, ^{106}Ru as anions are absorbed on the column. Therefore the measured total beta activity in the eluate of 99mTc does not include ^{131}I, ^{99}Mo and ^{103}Ru, which are measured directly by gamma spectrometry.

2. Separation of strontium for determination of ^{89}Sr and ^{90}Sr

Radioactive strontium in the effluent and wash from the anion exchange chromatographic separation was separated using Sr-column (Triskem). Half of the solution (effluent + wash) was taken and prepared in 8.0 HNO_3 by addition of 65% HNO_3, 2 mg Sr and 2 mg Y carrier/hold-back carrier are added. The solution is loaded to a 2 ml Sr column, and the column is washed with 8.0 M HNO_3. Strontium on the column is eluated using 10 ml of 0.05M HNO_3 solution. This eluate is directly used for measurement of ^{89}Sr and ^{90}Sr by LSC. The tracer experiment has shown that the recovery of Sr in this procedure is more than 98%, and the decontamination factor for most of radionuclides including ^{99m}Tc and ^{99}Mo are higher than 10^4. Due to quite quantitative separation of strontium in the two steps above, it is not necessary to measure the chemical yield of strontium.

3. Measurement of total alpha and total beta by LSC using discrimination function

Based on the α/β discrimination function in the LSC instrument, total alpha and beta activities in the separation sample from ^{99m}Tc eluate are measured. To 2 ml separated solution (0.1 M HCl-0.45% NaCl), 10 ml of Ultima Gold scintillation cocktail is added. A satisfied separation of alpha from beta was obtained at SPA of 100. The investigation using ^{90}Sr and ^{242}Pu as representative of alpha and beta radionuclides has shown that the cross-over of alpha to beta window is less than 3% and the beta to alpha window is less than 2%.

4. Measurement of ^{89}Sr and ^{90}Sr using Cherenkov radiation by LSC

The separated radioactive strontium in 0.05M HNO_3 solution was measured by LSC using Cherenkov counting. After Sr column separation, the solution is immediately measured (within 3 hours) to obtain the activity of ^{89}Sr before the ingrowth of ^{90}Y from ^{90}Sr. The solution is then kept for 5-7 days for ingrowth of ^{90}Y, and measured again for ^{90}Y. The Cherenkov counting efficacies for ^{89}Sr and ^{90}Y are measured using standard solution of 89Sr and 90Y to be 42% and 62%, respectively. The ^{90}Sr activity is calculated by the measurement results of two measurements and the ingrowth time from the separation to second measurement. Due to very low counting efficiency of ^{90}Sr by Cherenkov counting, the inference of ^{90}Sr to the measurement of ^{89}Sr is mainly come from the formed ^{90}Y. In the case of radioactive ratio of ^{90}Sr/^{89}Sr lower than 1, this interference is less than 4%. The interference of ^{90}Y to ^{89}Sr measurement is corrected based on the second measurement for ^{90}Y+^{89}Sr.

Summary

With the developed method combined with gamma spectrometry, all required radioactive measurement in the Ph. Eur. Including total alpha, total beta, ^{89}Sr and ^{90}Sr can be completed, the detection limits reached by this method can satisfied with the requirement in the Ph. Eur. 9.0 (6×10^{-7} for ^{89}Sr, 6×10^{-8} for ^{90}Sr and 10^{-9} for alpha emitting radionuclides relative to ^{99m}Tc in the eluate when injection). This method has been successfully used for the quality control of ^{99m}Tc-^{99}Mo generators.

References:

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