PREPARATION OF A GEL OF ZIRCONIUM MOLYBDATE FOR USE IN THE GENERATORS OF ^{99}Mo-^{99m}Tc PREPARED WITH ^{99}Mo PRODUCED BY THE ^{98}Mo(n, γ)^{99}Mo REACTION

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IPEN develops a project concerning the preparation of a gel of Zirconium Molybdate for use in the generators of ^{99}Mo-^{99m}Tc. ^{99m}Tc is the most used radioisotope in Nuclear Medicine diagnosis procedures and nowadays the generators are being prepared with imported ^{99}Mo, produced by ^{235}U fission. The production of ^{99}Mo by the ^{98}Mo(n,γ)^{99}Mo reaction is now possible because of the power upgrade of IPEN's IEA-R1 reactor, from 2 to 5 MW. This work describes the preparation method of Zirconium Molybdate gel that will be used in the ^{99}Mo-^{99m}Tc generators. The gel is prepared by the chemical reaction between Mo, in MoO_3 form, and Zr, in $ZrOCl_2.8H_2O$ form. After the reaction, the gel is filtered, dried and cracked with saline solution. The product is then loaded into glass columns for use as ^{99m}Tc generator. The results showed the good quality of the gel prepared at laboratory level and of the generators evaluated.

INTRODUCTION

^{99m}Tc is the most used radioisotope for diagnosis exams performed in Nuclear Medicine worldwide due to its favorable physical decay properties and its distribution in the generator form of ^{99}Mo-^{99m}Tc. ^{99}Mo can be produced by two different reactions that take place at a nuclear reactor: the fission of Uranium and the activation of Molybdenum ($^{98}Mo(n,\gamma)^{99}Mo$)[1-3]. The first method produces ^{99}Mo with higher specific activity, what is preferable for assembling chromatographic generators, but on the other hand requires a very sophisticated chemical separation and radioactive waste management. The (n,γ) reaction is a cleaner method, but produces ^{99}Mo with a low specific activity. The generator technology developed to solve this disadvantage is the preparation of a gel containing Mo and also with ion exchange properties. The gel of zirconium molybdate (MoZr) fulfills these characteristics.

Nowadays IPEN imports ^{99}Mo produced by the fission of uranium and prepares the generators with in house technology, and in parallel has been developing the nationalization of the ^{99}Mo production. The opted route was the (n,γ) reaction on natural Mo at the IEA-R1 reactor that just had a power upgrade from 2 to 5 MW.

This work describes the several parameters involved in the production of ^{99}Mo and the preparation of the gel of MoZr and posterior assembling and testing of the generators of ^{99}Mo-^{99m}Tc.

EXPERIMENTAL

Preparation of MoZr Gel

The preparation of the gel of MoZr followed at principle the work developed at China[3], and it was initially developed with cold MoO_3, then with tracer amounts of ^{99}Mo and finally with irradiated MoO_3.

Mo reacts with Zr, in the form of $ZrOCl_2.8H_2O$ in suitable conditions of concentration, pH, temperature and stirring. The gel is further filtered, dry and finally cracked with saline solution. This product can be loaded onto glass columns either dry or wet.

Two techniques have been studied for the gel preparation: 1) irradiation of MoO_3 and further gel preparation and 2) cold preparation of gel and further irradiation.

The parameters of the reaction analysed were: speed of filtration, speed of drying, uniformity of cracking, and mass and radioactivity balances.

Irradiations

All irradiations were performed at the IEA-R1 reactor at IPEN-CNEN/SP, with power varying from 2 to 5 MW. All the targets were irradiated inside aluminium targets either directly or inside a quartz tube. A Beryllium irradiator was specially fabricated for high power irradiations of MoO_3 targets.

Preparation of the generators of ^{99}Mo-^{99m}Tc

The generators of ^{99}Mo-^{99m}Tc were prepared using glass columns similar to the ones used in the routine production of fission ^{99}Mo generators at IPEN-CNEN/SP. First 1.0 g of Al_2O_3 were loaded onto the column (to retain Mo) and then the necessary mass of dry or wet MoZr gel. After the growing of ^{99m}Tc, the elution was performed with saline solution and the eluate collected onto evacuated flasks.

Gel Characterization

<u>Structure identification</u>: X-Ray difraction with alpha CuK radiation

<u>Particle size determination</u>: Sieve distribution apparatus, weighing the sieves before and after the distribution

<u>Determination of Mo, Zr and impurities</u>: Neutron activation analysis and UV-visible spectrometry.

<u>Elution profile of ^{99m}Tc</u>: Activity determination using a dose calibrator Capintec

<u>Quality control of ^{99m}Tc</u>: Radiochemical, chemical and radionuclidic purity and pH determinations

RESULTS AND DISCUSSION

Gel Preparation

Up to now, the studies showed that the gel of MoZr can be produced in a reliable and reproducible way, with fast speeds of filtration and drying and uniformity of cracking. The mass balance was in average - 2% and the radioactivity -5%.

The results also showed that the best way of preparing the MoZr gel was the irradiation of MoO_3 and further preparation of the gel.

Irradiations

The results of the high power irradiation of MoO_3 showed that a specific activity of 1 Ci ^{99}Mo / g Mo can be achieved. The irradiation in quartz reduced the ^{99}Mo activity in up to 20%.

Characterization of the gel

X-ray difraction studies showed that the MoZr has an amorphous form, as expected.

The particle size distribution for MoZr gel can be seen in figure 1. This distribution is appropriate to give a good performance of the gel type generators.

The neutron activation analysis of the gel showed one impurity, Hf, with a level of 0.62%. This impurity comes along with the Zr salt that is used in the preparation. The amount of Mo and Zr in the gel are, in average, 26.16% and 26.95%, respectively. These values confirm the proposed chemical form of the gel, i.e., $MoZrNa_2O_6$.

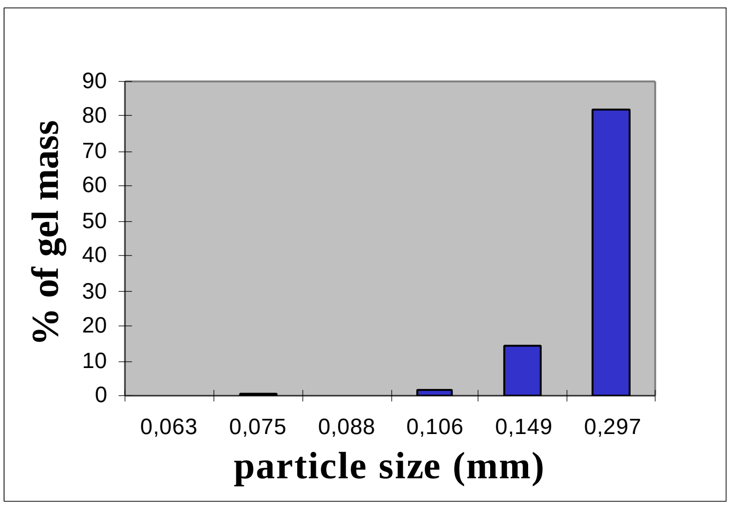

Figure 1. Particle Size Distribution of MoZr Gel

A typical elution curve of a gel type generator is shown in figure 2. It can be seen that more than 80% of ^{99m}Tc is eluted in the first 6 mL, reaching more than 96% in 12 mL. These numbers are the same for a dry or wet generator. If the total activity of ^{99}Mo present in the generator is taken into account, the elution yield is higher than 75% for the dry generator and higher than 85% for the wet generator. These results are always better than the ones achieved in China.

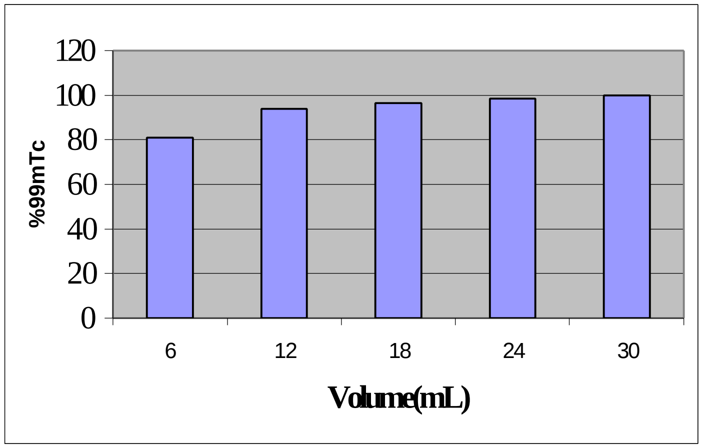

Figure 2. Elution curve of gel type generator

The ^{99m}Tc eluted from the gel generators was approved by all the quality control tests. The radiochemical purity was higher than 99.4% (TcO_4^-), the radionuclidic impurity of ^{99}Mo was lower than 10^{-4}%, the level of Mo and Zr was lower than the permissible, and the pH was between 5 and 6.

CONCLUSIONS

A method for the production of ^{99}Mo and the preparation of the MoZr gel was successfully developed at IPEN-CNEN/SP at laboratory level. The method is efficient and reliable, and the gel prepared has the adequate technical characteristics. The performance of the ^{99}Mo-^{99m}Tc generators prepared with the gel confirmed the good quality of the product, that is similar to the generators prepared with fission ^{99}Mo. The results so far show that is possible to prepare generators with activity up to 1 Ci that represents 50% of the present market.

As a final test for its quality, kits of MDF and coloidal S were labelled with ^{99m}Tc eluted from the gel type generator. The labelling yield was identical to the ones achieved with the fission generator, i.e., more than 98% for MDP and more than 95% for coloidal S.

The next steps in the project are: 1) improve the specific activity of ^{99}Mo, using different geometry of irradiation and testing metallic Mo as target; 2) design, construction and assembling of the equipments for the routine production and 3) improve the conditions of reaction in order to produce the gel with higher content of Mo.

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