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STUDY OF DIFFERENT ADSORBENT MATERIALS FOR THE PREPARATION OF GENERATOR SYSTEMS OF ^{99}Mo - ^{99m}Tc AND ^{188}W-^{188}Re

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ABSTRACT

Amongst some currently radioisotopes, ^{99m}Tc and ^{188}Re present adequate decay properties for use in Nuclear Medicine. In the current times the most diagnosis examinations are performed with ^{99m}Tc and ^{188}Re is one radioisotope of potential use in therapy techniques. The objective of this work consists of determining the capacity of some adsorbent materials for retention of molibdenum and tungsten, aiming the optimization of generator systems of ^{99}Mo-^{99m}Tc and ^{188}W-^{188}Re with suitable characteristics for application in Nuclear Medicine. Known amounts, in mass, of molybdenum and tungsten were percolated through different chromatographic columns containing different commercial adsorbent materials such as: PZC (poly-zirconium compound), acid alumina and calcinated alumina used in the routine preparation of ^{99}Mo-^{99m}Tc generators at IPEN. The tungsten (^{188}W), as well the PZC used in this project, supplied by Russia and Japan, respectively, through the International Atomic Energy Agency (IAEA) and used without any previous preparation. Also trace amounts of ^{99}Mo and ^{188}W were added to the initial solutions and the generators were assembled. The ^{99}Mo-^{99m}Tc generators were then eluted with known volumes of 0.9% NaCl solution every 24 hours whereas the ^{188}W-^{188}Re generators were eluted every 48 hours. The eluted samples were analyzed by Gamma Spectroscopy and later submitted to quality control evaluation. The results showed that the PZC presents superior retention capacity for Mo of 97.50mgMo/gPZC, higher than in acid and calcinated alumina, however the elution efficiency at lower pHs is not so high. With regards to the experiments carried out with ^{188}W and alumina, it was verified that the elution efficiency of ^{188}Re was not reproducible and the retention capacity of W was 90.23mgW/gAl_2O_3 at pH 7.

1. INTRODUCTION

The nuclear medicine can be defined as a clinical application of radioactive materials, to diagnosis or to treat distinct pathologies, produced in its majority of artificial form. Amongst the some currently existing radioisotopes, ^{99m}Tc and ^{188}Re presents properties respectively sufficiently specific physicist-chemistries that characterize them for use in Nuclear Medicine in the areas of diagnosis and therapy. Beyond this, these radiopharmaceuticals, can be distributed to the medical classroom under the form of generator systems of ^{99}Mo -^{99m}Tc e ^{188}W-^{188}Re, guaranteeing autonomy and praticity in its use.

^{99m}Tc essentially is used in nuclear medicine under the form of sodium pertechnetate ($NaTcO_4$) in scintilographic studies and ^{188}Re is being using in therapy techniques.

The traditional chromatographic generator system that it uses ^{99}Mo from the nuclear reaction ^{235}U (n, f)^{99}Mo, adsorbed in aluminum oxide, constitutes the main currently employed method for attainment of ^{99m}Tc. This technique involves the elution of pertechnetate with saline solution [1].

However, recent studies point a new trend of substitution of alumina as material adsorber for a inorganic material known as composed of poly-zirconium compound (PZC) [2].

Essentially a chromatographic generator system of ^{188}W-^{188}Re presents similar internal configuration as the generator of ^{99}Mo -^{99m}Tc, being composed basically of a glass column, in which the radionuclide father is adsorbed in an inert material (such as alumina) at the same time that decays to the element son [1-3].

The research proposal involved the evaluation of the capacity of molybdenum and tungstenium retention in presence of different adsorbent materials, as well as the preparation of generator systems of ^{99}Mo -^{99m}Tc and ^{188}W-^{188}Re, the analysis of elution profile and the quality control of the eluted samples containing ^{99m}Tc and ^{188}Re.

2. EXPERIMENTAL PROCEDURE

2.1. Generators Systems of ^{99}Mo-^{99m}Tc and ^{188}W-^{188}Re

The tungsten (^{188}W), as well as the PZC used in this work, came from Russia and Japan, respectively and were yielded by the International Atomic Energy Agency (IAEA) and used in the experimental assays without previous preparation.

Solutions of molibdate and tungstate were prepared by adding known amounts, in mass, of molybdenum (Mo) and tungsten (W) non-radioactive, being these in turn later percolated throughout different devices of retention.

Tracers of ^{99}Mo and ^{188}W were also added to the these solutions initially prepared, before the percolation process, and the loaded solutions were analyzed by Gamma spectroscopy , using the hiperpure germanium detector (HPGe) of Canberra (Mod. 747) or by means of a dosecalibrador from Capintec (Mod. CRC 15R) depending on the nature of the radionuclide studied.

The loaded solutions used in the percolation process of alumina or PZC, were prepared with values of pHs between 1 and 7. The elution processes of ^{99m}Tc was performed using a solution of sodium chloride (0.9%) as eluent and carried out four times consecutively

always with an interval of time of approximately 24 hours between each elution, being this the necessary time for the maximum growth of ^{99m}Tc, whereas the elution of ^{188}Re was carried out in intervals of time of 48 hours.
The eluted samples of both generator systems containing ^{99m}Tc or ^{188}Re were submitted to quality control tests aiming the determination of the radionuclidic, radiochemical and chemical purities of the eluates, as follows:

2.2. Quality Control

2.2.1. Radionuclidic Purity

The eluted samples from the generator systems of ^{99}Mo -^{99m}Tc and ^{188}W-^{188}Re were submitted to radionuclidic control tests, in order to identify the presence, as well as the amount of ^{99}Mo and ^{188}W existing in the solutions.
The eluted samples containing traces of ^{99}Mo were first analyzed into a closed special lead shield as known as canister in order to detect the ^{99}Mo energy emitted by them. Following, the same samples were once again analyzed using the same system, however the device were kept opened aiming at quantify the ^{99m}Tc presence.
Due to the close energies of ^{188}W and ^{188}Re radionuclides, the ^{188}W eluted samples were analyzed in a different way, by Gamma Spectroscopy using the hiperpure germanium detector (HPGe) to detect the presence of the radionuclide father and also to quantify the son.

2.2.2. Radiochemical Purity

The radiochemical control had objectified to identify the radiochemical impurities such as TcO_2 (technetium in the colloidal form) and ReO_2 (rhenium in the colloidal form), using the technique of paper chromatography with Whatmann 3 MM paper and sodium chloride solution as solvent.

2.2.3. Chemical Purity

The possible existing chemical contaminants in the eluted samples are molybdenum (Mo) and tungsten (W), as well as aluminum (Al), proceeding from the different adsorbent materials used in the preparation of the generator systems of ^{99}Mo -^{99m}Tc and ^{188}W-^{188}Re. The amount of these elements was determined by the analytical technique of optic emission with plasma connected inductively spectroscopy (ICP-OES), using the equipment from Varian (Vista-MPX-CCD Simultneous), which is based on the emission of electromagnetic radiation of the visible and ultraviolet regions of the electromagnetic spectra for excited neutral atoms or ions.

3. RESULTS

3.1. Retention Capacity of ^{99}Mo

3.1.1. Chromatographic Columns of Alumina and PZC

The accomplishment of this experimental assay involved the study of the capacity of retention of ^{99}Mo for alumina in the calcinated and acid forms as well as for PZC.
Figures 1 and 2 show the capacity of retention of Mo by alumina and PZC in different experimental conditions.

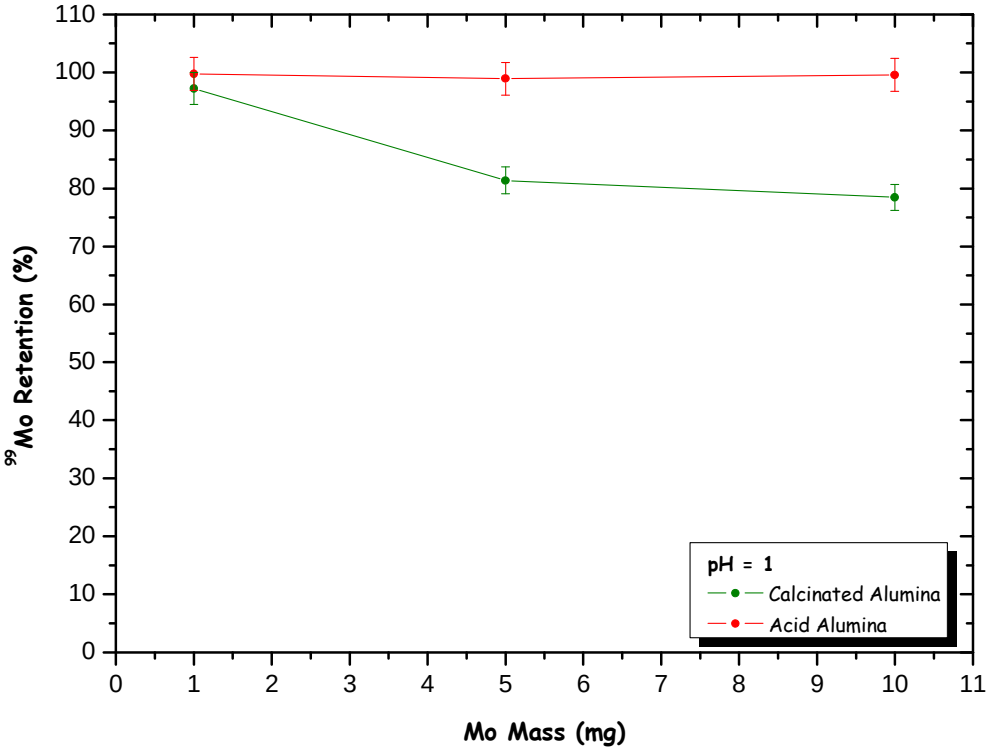

Figure 1- Curves of retention of Mo versus molybdenum mass (non-radioactive) by alumina columns at pH 1.

Acid alumina has higher capacity for Mo than calcinated alumina, as well as PZC as the highest capacity among all adsorbers employed.

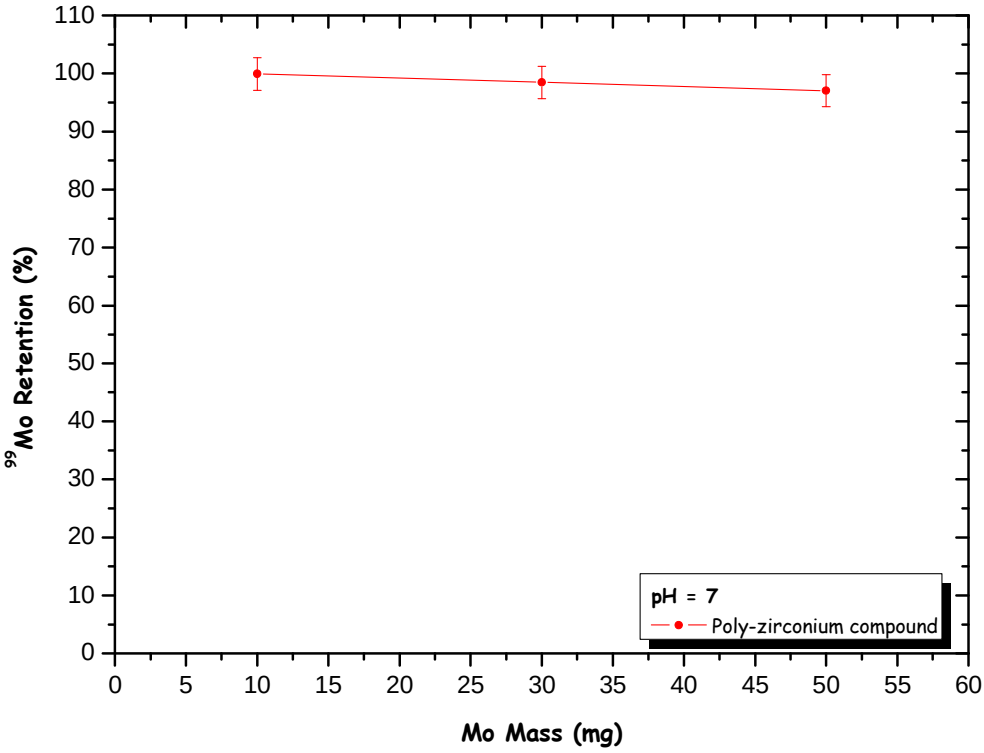

Figure 2- Curve of retention of Mo versus molybdenum mass (non-radioactive) by PZC columns at pH 7.

The capacity of Mo by PZC varied at between 96 ± 1mgMo/gPZC and 99 ± 2mgMo/gPZC, as shown in Figure 2, demonstrating that PZC constitutes a very efficient adsorbent material for retention of ^{99}Mo at higher pH values.

3.2. Retention Capacity of ^{188}W

The experimental assay involving the study of the capacity of retention of ^{188}W for alumina in the calcinated and acid forms was carried out in neutral solutions (pH 7), as shown in Figure 3.

According to Figure 3, it is observed that the capacity of retention of ^{188}W for calcinated and acid alumina is significantly lower than the capacity of retention observed for ^{99}Mo using the same adsorbent materials. It also can be seen that the capacity of retention for calcinated alumina was between 50 and 60% for all the concentrations of ^{188}W solutions percolated and the efficiency of retention for acid alumina was higher than 60%.

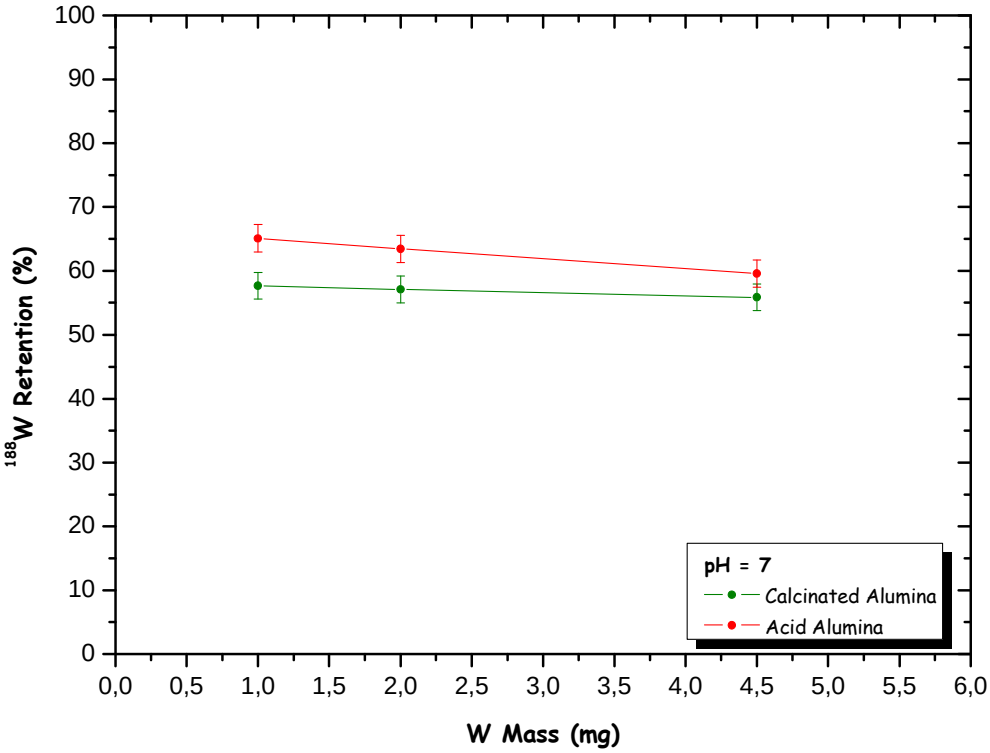

Figure 3- Curves of retention of ^{188}W versus tungsten mass (non-radioactive) by calcinated and acid alumina columns at pH 7.

3.3. Elution Profile of ^{99m}Tc and ^{188}Re

The elution profiles of ^{99m}Tc and ^{188}Re did not suffer any kind of alteration by the addition of carrier material to the loaded solutions independently of the adsorbent material used.

3.4. Quality Control

The quality control tests in the eluted samples of ^{99m}Tc and ^{188}Re showed that they did not have significant contamination with molybdenum (Mo) and technetium in the colloidal form (TcO_2), tungsten (W) and the colloidal chemical species of rhenium (ReO_2) and aluminium in all samples.

4. CONCLUSIONS

In general, the elution efficiency of ^{99m}Tc remained next to 100% for the generators systems prepared with calcinated and acid alumina.

Acid alumina presented a higher capacity of retention of ^{99}Mo than calcinated alumina, and it shows to be an interesting option to substitute the calcinated alumina used in the routine production of IPEN.
Despite the capacity of the PZC being lower than the capacity reported in other works [4-5], it is superior to the other materials.
Concerning the retention of ^{188}W it was observed again that acid alumina presented a better performance than calcinated alumina for all the analyzed situations. With regard to the tungsten samples, it was verified that the capacity was lower than for Mo.

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