CONSIDERATIONS ON THE ^{90}SR RECOVERY AS PART OF A PROCESS FOR ^{99}MO PRODUCTION FROM LEU FISSION

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Abstract

The possibilities of recovering ^{90}Sr as part of a process for ^{99}Mo production from uranium fission, using low enriched targets, are discussed. The pertinent calculations, concerning the estimated activities and masses of stable isotopes, as well as their evolution with time, were performed. A crown ether, known by the working name: DC – 18 – C – 6, was selected as separative agent. Some experiments are described, in connection with the strontium separation from barium, the influence of uranium on the recovery, and finally, the strontium separation from other fission products present in a real LEU sample. The results show the feasibility of producing ^{90}Sr with good yields and very high degree of purity.

Introduction

The production of ^{99}Mo from ^{235}U fission is routinely carried out in Argentina, at the Ezeiza Atomic Center of Comisión Nacional de Energía Atómica, since more than fifteen years. Although uranium having high degree of enrichment is currently employed for the process, the studies in progress at present foreseen the use of LEU targets as one of the objectives for the near future. Within this frame, the possibility of recovering some radionuclides other than ^{99}Mo, which at this time form part of the wastes, is also explored. This work deals with the investigations oriented to obtain one of them, namely ^{90}Sr, as by-product of the overall process.

In order to evaluate the possibilities of producing ^{90}Sr from uranium fission, the basic requirement is that the method to be proposed should be adapted to the operational conditions accepted for the ^{99}Mo production, i.e. neither the reagents for the chemical attack nor the different stages of the process can be modified. This condition implies severe restrictions to the alternatives for the radiochemical separation. Moreover, the replacement of HEU targets for LEU ones adds an additional difficulty to the separation scheme, since the presence of larger quantities of elemental uranium can affect the chemical purity of the final product, whereas the radionuclides produced by the ^{238}U decay could influence its radionuclidic purity.

The variety of the radionuclides involved in a fission show different nuclear and chemical characteristics; consequently, the isolation of a specific product is usually a cumbersome task. In connection with ^{90}Sr, the most relevant chemical interferences can be originated from both radioactive and stable barium isotopes, due to the similar behavior of the elements. According to the irradiation conditions employed for production of ^{99}Mo, masses of ^{138}Ba as high as 0.19 mg per gram of LEU uranium are formed, together with time-increasing ^{137}Ba minor contents, which are produced by ^{137}Cs decay. If the production of ^{90}Sr is intended after a relatively short cooling time, ^{140}Ba must be added to the list of the potential radioactive impurities.

From the viewpoint of the ^{90}Sr separation as part of an overall process, the insertion of the pertinent steps on line with the ^{99}Mo production, despite its major complexity, seems at first sight more attractive. The limiting factor in this case, rather than the higher global activities and the subsequent need to develop more rigorous chemical procedures, could be the presence of ^{89}Sr in the product. Due to the differences between ^{89}Sr and ^{90}Sr half-lives (respectively 50.5 d and 28.64 y) higher activities of the former can be expected after an irradiation cycle.

The possibilities of the eventual methods for ^{90}Sr separation were primarily assessed as a function of their specificity and the percentages that could be reached in the isotope recovery, as well as their simplicity and applicability as routine procedures. The often expensive and tedious precipitation methods are currently used only for analytical purposes; frequently, the addition of inactive carrier is mandatory, thus limiting the quality of the product. Inorganic absorbers show good selectivity and resistance to high radiation fields, but the elution of the retained species is very complicated, and most of the times almost impossible.

The classical methods based on solvent extraction are normally difficult to implement in cells; however, an interesting possibility is open when the extractant can be supported on an inert

matrix, thus offering the conditions for operational use in chromatographic columns. This is the case of crown ethers, which are recognized as very selective reagents; one of them, known by the working name: DC – 18 – C – 6, appeared as the most suitable for the separation of strontium. Therefore, different sequential experiments were carried out, in order to establish the feasibility of its potential use for the recovery of ^{90}Sr from irradiated LEU targets.

Experimental

The estimated activities of the radionuclides and masses of the stable isotopes involved in LEU uranium fission, as well as their evolution with time, were calculated with the ORIGEN program [1], for conditions close to those corresponding to an actual process of ^{99}Mo production in the RA-3 reactor (thermal flux: 1.1×10^{14} $n.cm^{-2}.s^{-1}$; irradiation time: 120 h).

All the experiments were performed with a commercial reagent from Eichrom Inc. (USA), which consisted in spheres of the inert support that held the crown ether.

A hyperpure Ge detector coupled to multichannel analyzer and a liquid scintillation system were employed, respectively, for measurements of gamma and beta emitters.

Some previous experiments, describing the performance of the columns for the separation of Sr and Ba in nitric concentrated solutions and a study of radiation damage, have been already published [2]. Briefly, the results showed:

- Very good separation between strontium and barium
- Low strontium losses
- High concentration of strontium activity, for elutions with highly diluted HNO_3
- Separation non affected by the involved radiation doses

The influence of uranium on the separation was studied on the basis of the contents present in a mini-plate having 20 % enrichment, whereas the strontium and barium masses corresponded to the estimations for a one-year decay. Two different loading uranium solutions with concentrations higher than 100 g/l were prepared in nitric media similar to those used for the initial experiments, in the presence of strontium and barium carriers as well as ^{85}Sr and ^{133}Ba tracers. The elution medium was diluted HNO_3, in consonance with the results previously found. The tests of retention and elution of strontium were acomplished with standard commercial columns, also supplied by Eichrom, and other columns of larger dimensions, built in the

laboratory. In the course of the first experiments it was observed that the passage of the solution through the columns was extremely slow; thus, a vacuum pump was used for the next ones, so as to reach an adequate elution flow. Fractions corresponding to the loading, washing and eluting solutions were measured by gamma spectrometry.

A pilot experiment of strontium separation from all the other fission products was finally performed. The sample consisted of 20 % enriched metallic uranium, irradiated in conditions similar to those used for ^{99}Mo production in order to test a method of alkaline attack, and the experiment started 17 months after the end of the irradiation. A small volume of the initial solution was adjusted to concentrated nitric medium; 2 ml of this new solution were loaded in a column of the small type, similar to those used in the aforementioned experiment, and the loading waste was collected The column was washed with a solution with similar nitric concentration, the washings being collected in several fractions. Previous experiments demonstrated that barium isotopes, characterized as the limiting interferences, were entirely removed with 20 ml of the washing solution. Nevertheless, and taking into account the complexity of the matrix and the obvious need to eliminate all products other than strontium, the volume was in this case overestimated. The elution was carried out with small aliquots of diluted HNO_3. All the fractions (loading waste, washings and elution) collected in the experiments were measured by gamma spectrometry; those corresponding to the elution were additionally measured by liquid scintillation Cerenkov counting.

Results and Discussion

The estimated activities of ^{90}Sr that can be obtained at the end of the irradiation reach 3700 MBq (100 mCi) per irradiated mini-plate. However, the calculations show that the separation of ^{90}Sr in the course of the ^{99}Mo production is of no practical utility, since the chemical operations turn more complicated and the eventual use of the product is prevented by the presence of ^{89}Sr. The activity of ^{89}Sr will be higher than the ^{90}Sr one even after a one-year decay time, although the ratio of activities will be close to 1. The optimum ^{90}Sr to ^{89}Sr activity ratio depends on the use intended for the former. As illustrative figures, this calculated ratio is 15000, three years after the end of the irradiation; after a period of five years, ^{89}Sr could be considered as completely decayed, in comparison with ^{90}Sr.

The presence of high masses of uranium had been mentioned as an additional complication in the separation scheme. The experiments indicates that both the loading and the washing volumes should augment for the real experimental conditions in order to manage such amounts of the matrix element, thus resulting in a considerable increment in strontium losses. These losses turn intolerable if columns of the small type are used, since only 40 % of the product is recovered after the separation; on the other hand, a yield higher than 85 %, perfectly compatible with the productive requirements, can be obtained with the larger ones.

Some minor activities of ^{235}U and ^{238}U daughters were identified in the separated strontium fractions. One of them, ^{234}Th, is long-lived enough ($t_{½}$ = 24.10 d) to affect the radionuclidic purity of ^{90}Sr. Therefore, it is advisable to perform the radiochemical six months before the delivery of the product, so as to make it possible the decay of this impurity.

Finally, the results of the separation of strontium from the other fission products rendered excellent results. No gamma emitters were found in the eluted fractions, whereas the measurements by scintillation counting show a Cerenkov spectrum attributable to ^{89}Sr; ^{90}Sr in less extent; and its daughter, ^{90}Y.

All these results allow to conclude that ^{90}Sr can be obtained with very good yields and high degree of purity. The experiments performed give a definite ground to support a project for its future routine production.

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