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PRESENT STATUS OF RADIOCHEMICAL DOUBLE BETA DECAY STUDY (^{238}U)

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PRESENT STATUS OF A RADIOCHEMICAL DOUBLE ß DECAY STUDY (^{238}U)

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Introduction

Among the candidates for the ßß decay process, ^{238}U and ^{232}Th offer the possibility of a sensitive radiochemical measurement. Because in the $^{238}U \rightarrow {}^{238}Pu$ case, the kinetic energy release (1146 keV) is larger than in the $^{232}Th \rightarrow {}^{232}U$ case (850 keV), the half-life is expected to be shorter and thus more accessible. Several theoretical estimations of the half-life for the ^{238}U ßß(2ν) ^{238}Pu process have yielded the following values : 0.22×10^{20},[1] 1.3×10^{20},[2] and 1.4×10^{20} yr.[3] In 1949, Levine, Ghiorso and Seaborg[4] gave a lower limit of 6×10^{18} yr for the half-life of the ^{238}U ßß decay. Recently, Turkevich et al.[5] gave as a preliminary result a limit of 4.6×10^{19} yr. Taking into account these results and the fact that the calculated ßß(2ν) decay rates may be overestimated,[6] a more sensitive experiment has been designed that will be able to measure an assumed ßß half-life of 1.9×10^{22} yr, a value suggested by Haxton et al.[3] This corresponds to the activity of 27000 ^{238}Pu nuclei formed during a year from 300 kg of ^{238}U, giving 210 alpha decays per year.

Radiochemical separation

The chemical strategy adopted is the following : after initial elimination of ^{238}Pu and ^{239}Pu from a batch of 300 kg of depleted ^{238}U (overall decontamination factor U/Pu = 4×10^6), recovery of ^{238}Pu (and ^{239}Pu) formed during 1 yr of aging. After treatment the uranyl nitrate solution will be stored for one year in a 200 m deep mine to avoid ^{238}Pu production by protons from cosmic ray induced spallation and ^{239}Pu formation by superfluous neutrons.

Because ^{239}Pu formed in the solution by neutron capture in ^{238}U will be used as internal standard, the neutron flux must be exactly known. Neutrons arise essentially from ^{238}U self fission but also from (α,n) reactions.

A volume reduction of $\simeq 2 \times 10^5$ is needed to prepare the ^{238}Pu (^{239}Pu) α-source by electrodeposition or electrospraying. As the determination of ^{238}Pu (and ^{239}Pu) will be done by α spectroscopy, the recovered Pu isotopes must be decontaminated from several α emitters present in natural decay chains. The decontamination factor (Pu/α emitters) needed are fairly high : 3.2×10^{11}(U), 3.4×10^{11} (^{230}Th), 3.6×10^{10} (^{231}Pa).

The chemical system selected to achieve the above mentioned goals is based on extraction chromatography. The columns will be filled with an absorbing phase composed of tri-n-octylammonium nitrate (TOA HNO_3) adsorbed on an inert hydrophobic silica support. This absorbing phase is ideal for the recovery of Pu IV in a highly concentrated U VI solution. To run the process, it will be necessary to adjust the oxidation state of Pu using H_2O_2.

Experimental studies were undertaken to select the best conditions for running the extraction chromatographic cycles. The largest efforts up to now have been related to the initial Pu/U separations (to re-set the ßß decay "counter") and the one to be performed after one year of aging. For these separations the Pu isotopes to be recovered are in the presence of the entire mass of depleted ^{238}U (300 kg).

Figure 1 presents the Pu IV breakthrough curves obtained at 40°C during ^{238}Pu IV/^{238}U VI separation cycles on a 1g TOA HNO_3 silica column. About 60 ml of the initial solution titrating 180 g x l^{-1} of U VI can be decontaminated in Pu. When decreasing the linear velocity of the column feed solution only small improvement of the performance can be obtained as observed in Fig. 1. Thus a linear velocity close to 75 cm x h^{-1} has been selected for Pu IV loading on the column.

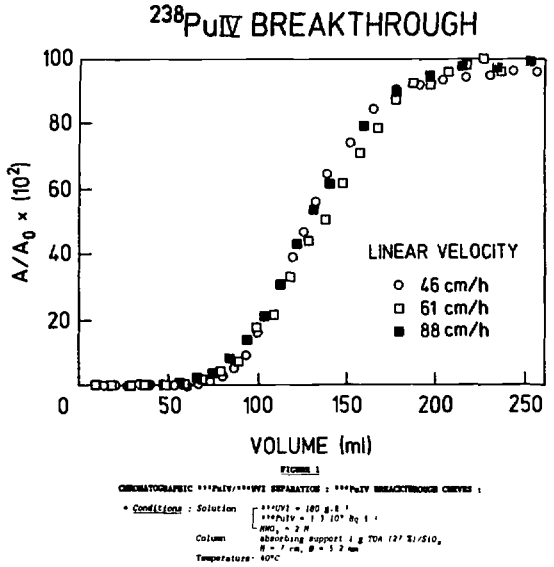

Figure 2 shows the variations of the relative concentrations of U VI, Th IV, Pa V and Pu IV during the washing with a 2M HNO_3 solution of the column loaded with Pu IV. The efficiency (E) varies in the following order $E_{U\ VI} > E_{Th\ IV} > E_{Pa\ V}$. During this step, Pu IV losses are minimal, i.e. < 0.6 % in relative concentration. A volume of washing solution equal to 15 l/kg of support has been selected for the first Pu IV/U VI chromatographic cycle.

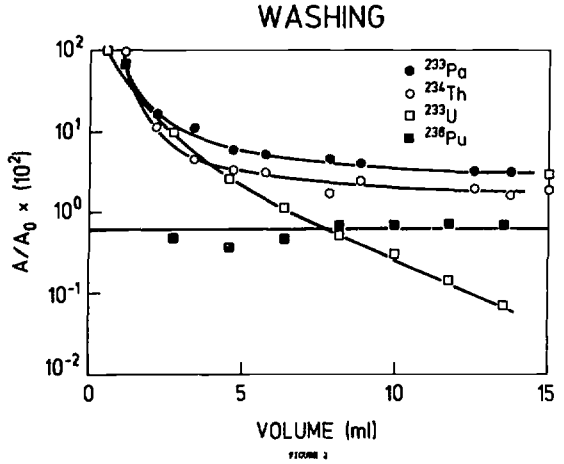

Figure 3 presents the Pu IV elution curve obtained using an H_2SO_4 = 0.5M, HNO_3 = 0.18M eluant. An increase of the temperature reduces the volume of eluant required for a quasi quantitative Pu IV recovery. The conditions adopted for the full scale experiment are : 60°C, eluant 5 l/kg of support, and a linear velocity of the eluant of 75 cm x h^{-1}.

Experiments have yet to be done to define the experimental conditions to be fulfilled for the other chromatographic cycles. The requirements imply running successively five chromatographic cycles on columns with decreasing sizes. For the first full scale cycle, the column will have the following characteristics : H = 210 cm, ø = 21 cm, absorbing support 55 kg.

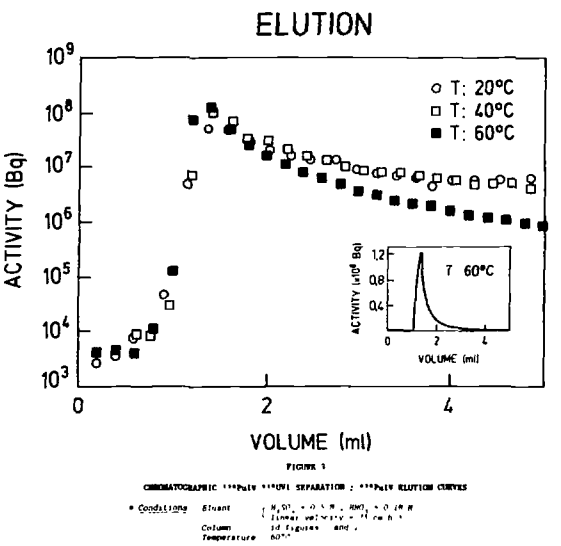

Figure 4 presents a sketch of the separation facility. To reach the performance objectives, two successive full scale chromatographic cycles will be necessary for the initial purification of the 300 kg of ^{238}U, whereas only one full scale cycle will be needed for the recovery of Pu isotopes after the one year of aging. The expected overall chemical yield for the ^{238}Pu is 80% for the five chromatographic cycles (> 95% for each cycle).

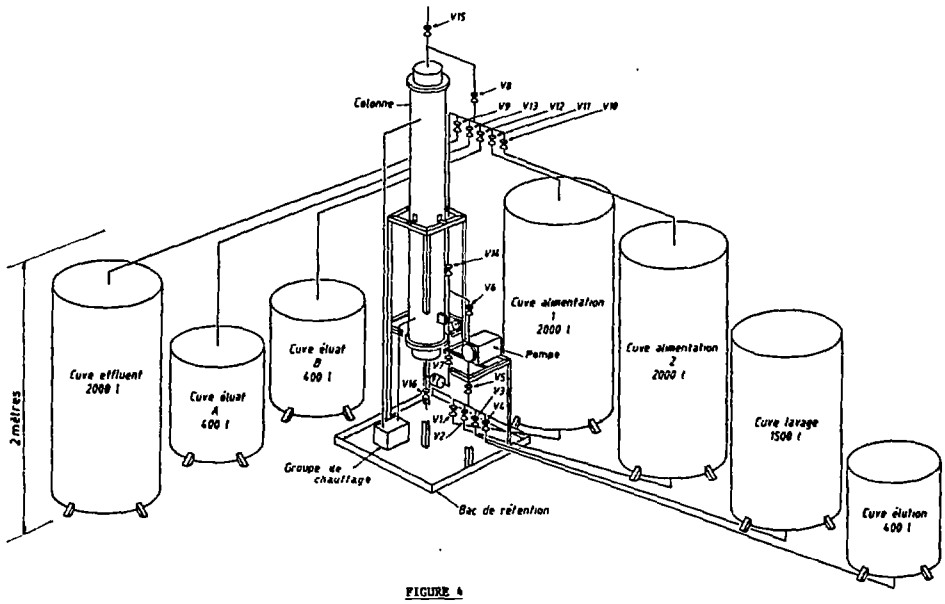

FIGURE 4

ARTIST'S SKETCH OF THE FULL SCALE FIRST CYCLE CHROMATOGRAPHIC FACILITY

Low Background α Spectrometer

In view of the small amount of ^{238}Pu formed by the ββ decay from ^{238}U, the α-particle spectrometer used must have very low background in addition to high efficiency and good resolution. The solution adopted has been to build a $\approx 2\pi$ counter consisting of five implanted Si detectors having a total active area of 15 cm^2. A great deal of effort has been devoted to lowering the background, the study consisting first of all in trying to learn the sources of this background. These can be grouped into the three following categories : radioactive contamination of the materials, atmospheric radon, cosmic rays.

The counter and vacuum chamber have been constructed largely of a Si-Al alloy and of teflon, both selected for their low radioactivity content. The α-particles emitted by ^{220}Rn are always present in the atmosphere, arising from anything containing a trace of Th. A good vacuum (10^{-6} Torr) in the chamber allows to minimize its concentration. It is more difficult to eliminate the ^{210}Po contribution coming from adsorption by surfaces after ^{222}Rn decay. Cleaning with NaOH could eliminate ^{210}Pb, which lies in the decay chain, but this cannot be done on the detector. Another source of

background at sea level comes from cosmic rays. The cosmic neutrons induce nuclear reactions in the spectrometer material (especially Si, Al and B), and produce charged particles which increase the background. In order to essentially eliminate background due to cosmic rays, the α spectroscopic measurements will be performed underground in the Laboratoire Souterrain de Modane.

Under such operating conditions the only contaminations still observed are due to ^{220}Rn and its daughters and to ^{210}Po (Fig. 5).

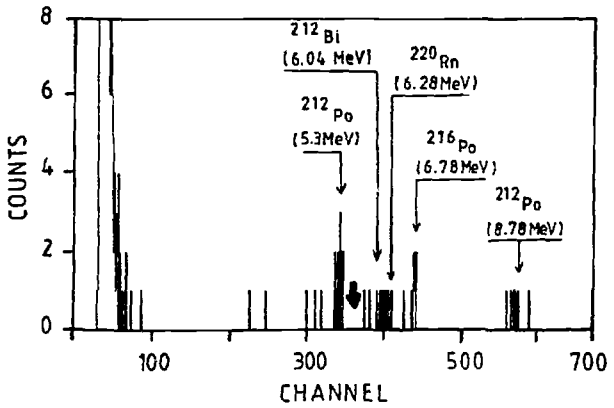

Fig.5.: Background observed during 39 d in the L.S.M. Surface Si detector 4.5 cm^2 - Roughing vacuum.

Considering a value of the ββ half-life $T_{1/2} = 1.9\ 10^{22}$ yr, from the measured background of 60 counts/yr in the region of interest 5.49 ± 15 MeV and assuming an efficiency of 32% (80 chemical, 40% geometrical), the 95% confidence limit of the expected half-life has been estimated as $1.4 \times 10^{22} < T_{1/2} < 2.74 \times 10^{22}$ yr. These limits correspond to a 1 yr formation time and a 1 yr counting period. They have been computed using Bayesian statistics with a uniform a priori distribution for the signal count.

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