

SUMMARY

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The thesis entitled "Production of Some High Specific Activity Radioisotopes" comprises three chapters; introduction, experimental, and results and discussion.

Chapter 1, introduction, includes brief accounts on nuclear fission reactions, uranium targets, chemistry of uranium and fission products, chemical separation concepts, review of some fission-products separation procedures, and radiochemical processing plants.

Chapter 2, which includes the experimental work, is divided into three main parts: (1) preparation and irradiation of the uranium targets, (2) chemical processing, including (i) digestion of the irradiated uranium targets, (ii) separation and recovery of radioiodine, (iii) separation and recovery of radoruthenium, (iv) batch separation of Sr, Ba, La, and Ce radionuclides, (v) batch separation of Zr, Nb, Te, and Np radionuclides, (vi) separation and recovery of radiocesium, and (vii) recovery of radiomolybdenum, and (3) quality control investigations of the recovered radioisotopes, including the separation yield, recovery yield, radionuclidic purity, radiochemical purity, and/or pH-value. It includes also a list of chemicals and a description of the equipments and apparatuses used in the present work.

Chapter 3 presents results and discussion. When uranium targets (uranium trioxide and uranyl nitrate hexahydrate) were irradiated in the 22 MW-water-cooled ETRR-2 Research Reactor (Egypt) for 4 h at a thermal neutron flux of 1×10^{14} n/cm².s, cooled for one week, and analyzed by γ -ray spectroscopy, the following radionuclides were appeared: ⁹⁵Zr ($T_{1/2} = 64.02$ d), ⁹⁷Zr ($T_{1/2} = 16.9$ h), ^{97m}Nb ($T_{1/2} = 58.1$ s), ⁹⁷Nb ($T_{1/2} = 1.23$ h), ⁹⁹Mo ($T_{1/2} = 2.75$ d), ^{99m}Tc ($T_{1/2} = 6.01$ h), ¹⁰³Ru ($T_{1/2} = 39.27$ d), ¹³¹I ($T_{1/2} = 8.04$ d), ¹³²I ($T_{1/2} = 2.28$ h), ¹³²Te ($T_{1/2} = 3.26$ d),

^{137}Cs ($T_{1/2} = 30.17$ y), $^{137\text{m}}\text{Ba}$ ($T_{1/2} = 2.55$ min), ^{140}Ba ($T_{1/2} = 12.75$ d), ^{140}La ($T_{1/2} = 1.68$ d), ^{141}Ce ($T_{1/2} = 32.5$ d), ^{238}U ($T_{1/2} = 4.47 \times 10^9$ y), ^{235}U ($T_{1/2} = 7.04 \times 10^8$ y), and ^{239}Np ($T_{1/2} = 2.36$ d). Many other radionuclides did not appear in the gamma spectrum of the irradiated uranium target because of one or more of the following reasons: (i) low fission-yield, (ii) low-energy or low-abundance gamma rays, (iii) interferences between γ -photopeaks, and (iv) absence of gamma-ray emission. Some of these radionuclides were successfully appeared in gamma spectra with the successive separation processes.

The irradiated targets were digested in 10 ml of 2 M NaOH solution for ~12 h. At end of the digestion process, the radionuclides of $^{137}\text{Cs}/^{137\text{m}}\text{Ba}$ and $^{99}\text{Mo}/^{99\text{m}}\text{Tc}$ were completely included in the supernatant. The uranium bulk (^{238}U and ^{235}U) was included in the undissolved residue. The other radionuclides distributed themselves between the supernatant and the residue with different ratios.

To separate and recover ^{131}I , the supernatant obtained from the digestion process was acidified by addition of H_2SO_4 , along with H_2O_2 . The acidified fission-products solution (15 ml containing 20 % H_2SO_4 and 0.5 ml of 30 % H_2O_2) was boiled for 3.5 h. The volatilized iodine was recovered in two alkali receivers (each containing 0.1 M NaOH-0.01 % $\text{Na}_2\text{S}_2\text{O}_3$ solution), after passage by two successive acid filters (each containing 3 M H_2SO_4). Separation and recovery yields of ^{131}I received in the alkali solution were found to be > 99.99 % and 73.6 %, respectively, with radionuclidic purity of 98.28 % (1.72 % of the total γ -radioactivity of the radioiodine product was due to ^{132}I). Radiochemical purity of the radioiodine product (as iodide) was determined by the ascending method of paper chromatography (using Whatman No. 1 paper) and TLC (with silica gel) chromatography. Radiochemical purity was found to be 99.81 % with Whatman No. 1 paper and 99.76 % with TLC. pH-value of the radioiodine

product solution was found to be 12.8. Calculated radioactivities of the recovered ^{131}I were found to be < 898 , < 2694 , and < 4490 μCi , while the calculated specific radioactivities were found to be < 45 , < 135 , and < 225 $\mu\text{Ci/ml}$ with weights of 0.02, 0.06, and 0.1 g of the irradiated UO_3 targets, respectively.

To separate and recover ^{103}Ru , the fission-products solution obtained after separation of ^{131}I was treated with excess of H_2SO_4 , along with KMnO_4 . The fission-products solution (15 ml containing 40 % H_2SO_4 and 0.01g KMnO_4) was then boiled for 40 min. The volatilized ruthenium was recovered in an alkali receiver (containing 0.1 M NaOH). Separation and recovery yields of ^{103}Ru was found to be > 99.99 % and 65.03 %, respectively. Radionuclidic purity of the recovered ^{103}Ru was found to be 91.78 % (4.81 % and 3.41 % of γ -radioactivity of the recovered radoruthenium were due to ^{106}Rh and ^{132}I , respectively). pH-value of the recovered radoruthenium solution was found to be 12. Calculated radioactivities of the recovered ^{103}Ru were found to be 293, 880, and 1466 μCi , while the calculated specific radioactivities were found to be 16, 49, and 81 $\mu\text{Ci/ml}$ with weights of 0.02, 0.06, and 0.1 g of the irradiated UO_3 targets, respectively.

After separation of ^{103}Ru , two successive batch separation processes were carried out to discard two groups of elements. In the first process, a solution containing 20 mg of BaCl_2 was added to the fission-products solution and, then, the formed precipitate was separated after ~ 18 h. The isotopes of ^{140}Ba and ^{140}La disappeared completely from the filtrate and detected in the formed precipitate. The precipitate also contained $^{90\text{m}}\text{Y}$, ^{141}Ce , ^{143}Ce , ^{144}Ce , and traces of ^{132}I and $^{137\text{m}}\text{Ba}$. In the second batch separation process, a solution containing 20 mg of FeCl_3 was added to the fission-products solution. Then, pH-value of the solution was risen to ~ 7 . The formed precipitate was separated after about one hour. The isotopes of

^{95}Zr , ^{97}Zr , ^{95}Nb , $^{97\text{m}}\text{Nb}$, ^{97}Nb , ^{105}Rh , ^{132}Te , ^{132}I , and ^{239}Np disappeared completely from the fission-products solution and detected in the formed precipitate.

To separate and recover ^{137}Cs , the fission-products solution obtained from the second batch separation process was treated with a solution of 0.01 M (30 mg) sodium ferrocyanide. After ~ 15 min, a solution of 0.02 M (26 mg) nickel chloride was added. The solution was left again for ~15 min before raising its pH-value to 10. After about one hour the formed precipitate was separated and dissolved in dilute H_2SO_4 . ^{134}Cs and ^{137}Cs disappeared completely from the filtrate and detected in the formed precipitate. The precipitate also contained ^{136}Cs . Separation and recovery yields of ^{137}Cs were found to be > 99.99 % and 98.3 %, respectively. Radionuclidic purity of the recovered ^{137}Cs was found to be 84.5 % (15.46 % and 0.04 % of the total γ -radioactivity of the recovered radiocesium were due to ^{134}Cs and ^{136}Cs , respectively). Calculated radioactivities of the recovered ^{137}Cs were found to be 4, 11, and 19 μCi , while the calculated specific radioactivities were found to be 1, 2, and 4 $\mu\text{Ci/ml}$ with weights of 0.02, 0.06, and 0.1 g of the irradiated UO_3 targets, respectively.

After separation of radiocesium, only ^{99}Mo radioactivity was remained in the fission-products solution in equilibrium with its $^{99\text{m}}\text{Tc}$ daughter. Finally and after carrying out all of the aforementioned separation processes, recovery yield of ^{99}Mo was found to be ~ 70 %. Radionuclidic purity of the recovered ^{99}Mo was found to be > 99.9 %, while pH-value of its solution was found to be 10. Calculated radioactivities of the recovered ^{99}Mo were found to be 1368, 4104, and 6841 μCi , while the calculated specific radioactivities were found to be 137, 410, and 684 $\mu\text{Ci/ml}$ with weights of 0.02, 0.06, and 0.1 g of the irradiated UO_3 targets, respectively.