Experiments for separation and purification of Mo-99 from uranium solution with fission products as tracers

Gabriela Elena ANDRONE, Ana MATEI
Romanian Authority for Nuclear Activity- Institute of Nuclear Research
România

ABSTRACT

Technetium – 99 m (**$^{99}Tc^{m}$,$T_{1/2}$ = 6 hours**), one of the most utilised radioisotopes in nuclear medicine, is generated through the beta desintegration of **^{99}Mo ($T_{1/2}$ = 66 hours) and which will desintegrate through isomer transition to ^{99}Tc ($T_{1/2} = 2 \cdot 10^5$ years) through the emission of a gamma radiation with the energy of** 0,140 Mev. The work presents the phases of the process of Mo separation and purification at a tracer level. The tests performed in the laboratory have established the optimum conditions for the separation and purifcation of Molybdenum. To establish the separation and purification parameters,a a synthetic solution which contains the elements which result following the irradiation of a low enriched Uranium foil weighing 10 g (~20 % U^{235}). To mark this solution, approx. 13 mg of UO_2 10% U^{235} was irradiated for 2000 s , at a flow of approx. $7x10^{12}n/cm^2s$. This quantitiy of UO_2 will be aded to the above-mentioned solution after dissolution. The method for separating the Molibdenm from irradiated Uranium solution is one of selective precipitation of Mo with α-benzoin-oxyme (α-BO). To purify the Molybdenum solution, two purification columns will be utilised. Their role is to absorb the impurities remained in the mass of the precipitate. They and the Molybdenum have passed into the solution simultaneously, allowing the Molybdenum to pass. These columns are : the column with active charcoal(AC)+ active charcoal covered with silver (AgAC); active charcoal column(AC)+active charcoal covered with silver(AgAC)+hydrated zirconium oxide(HZO). Although all the phases of the process are performed with high yields, the final yields of recovery of Mo from U solutions are higher than 80%.

INTRODUCTION

Technetium-99 (^{99}Tc) is mainly an artificially produced radioactive metal. ^{99}Tc also occurs naturally in very small amounts in the earth's crust. ^{99}Tc was first obtained from molybdenum but is also produced as a nuclear reactor fission product of Uranium and Plutonium. All isotopes of Technetium are radioactive, and the most commonly available forms are ^{99}Tc and $^{99}Tc^{m}$. Technetium-99 is an excellent superconductor at very low temperatures. In addition, ^{99}Tc has anti-corrosive properties. Five parts of Technetium per million will protect carbon steels from corrosion at room temperature. $^{99}Tc^{m}$ is used in medical therapy in brain, bone, liver, spleen, kidney, and thyroid scanning and for blood flow studies. $^{99}Tc^{m}$ is the radioisotope most widely used as a tracer for medical diagnosis.

EXPERIMENTS FOR ESTABLISHING THE PARAMETERS OF PURIFICATION AND SEPARATION OF THE MOLYBDENUM AT A TRACER LEV*EL.*

To establish the Mo separation parameters, 6 rings of Uranium dioxide (11,3767 g de UO_2 ≈ 10 g U), which will have a mass that is equivalent to the processing of 10 g of LEU Uranium foil. The volume HNO_3 that is necessary for the dissolution is 40 ml. A synthetic solution is prepared(Table 1). 1 ml of this solution corresponds approximately to the composition of a

solution resulted following the dissolution of a 10 g LEU Uraium foil(~ 20% U^{235}), that is irradiated fot 7 days in the TRIGA reactor at a flow of ~$7x10^{12}n/cm^2s$.

Table 1. The compozition of the synthetical solution . Solution volume 100 ml.

Element	U	Mo	Sr	Zr	Te	Cs	Ba	La	Ce	I
The concentration of the element M	0.42	1,04E-3	5,82E-5	6,53E-5	2,35E-5	6,77E-5	5,82E-5	5,03E-5	6,06E-5	5,50E-4

To mark this solution , approx. 13 mg de UO_2 10% U^{235} was irradiated for 2000 s, at a flow of de ~$7x10^{12}n/cm^2s$. This quantity of UO_2 will be added to the above-mentioned solution after dissolution. The activity of the initial solution is presented in table 2, and its spectre in fig. 1.

Table 2. The activity of the initial solution.

Isotope	Zr-95	Nb-95	Mo-99	Te-132	I-131	I-132	Ba-140	La-140
μCi	3.2	6.6E-18	69.08	119.78	36.71	68.94	52.62	43.22

I. The separation of the Molybdenum from Uranium solutions

The method of separation of Mo from irradiated U solutions that is utilised in this experiments is the selective precipitation with α-benzoynoxyme (α-BO). For the quantitative separation of Mo, the following reagents have been added for the adjusting of the solution resulted after the dissolution of the target : 4 g I carrier (4 ml solution 10 mgI/ml); 0.51 g HNO_3 (1,35 ml HNO_3 6N); 0,01 g $AgNO_3$ (1 ml solution 0,01g $AgNO_3$/ml); g HCl (2,27 ml HCl conc.); 1 g Mo carrier (1 ml solution 10 mg Mo/ml); 3 g solid Na_2SO_3..

25 ml of α-benzoinoxyme 2% have been added to NaOH 0.4 N, slowly near the walls, for the precipitation of the Mo. The solution was left intact for 30 minutes, so that the precipitate can mature. After the filtration, the precipitate was washed, to clean the U traces and the fission products remained in its mass. The precipitate of Mo with α-benzoinoxyme was dissolved in an alkalyne solution of NaOH 0.4 N and 1% H_2O_2 with which Mo forms a strong $H_2MoO_2(H_2O_2)_2$ compound .

The results of the experiments are illustrated in table 3 and in figures 2-4. After the analysis performed on the filtrate and on the reunited washing solutions, it has been found that the Mo loss is of approx. 2%. For the dissolution of the Mo precipitate with α-BO, the Mo recovery yield is of 89%.

II. The purification of the Molybdenum solution

To purify the Mo solution, two purification columns whose were used. Their role is to absorb the impurities stuck in the Mo precipitate mass . They and the Mo have passed simultaneously into the solution. The columns have been prepared like this: a layer of glass wool, respectively a 1:1:1 mix of active charcoal(AC)+ active charcoal covered with silver(AgAC) for the first column, respectively a 1:1:1 mix of active charcoal(AC) +active charcoal covered with silver(AgAC)+ hydrated zirconium oxide(HZO) for the second column. The columns were conditioned through washing with 50 ml NaOH 0.2 M followed by washing with 50 ml demineralised water . The Mo separated solution was passed on the first column with a speed of 3 ml/min. For the total recovery of the Mo, the column will be washed with 10 ml of NaOH 0.2 N. If it is necessary, a pump for the total recovery of the solution can be used. 40 ml of yellow-pale Natrium Molybdate solution in NaOH 0.25 M will result

Before passing onto the second column, a new reprecipitation of the iodine was made, by adding iodine carriers4 ml of NaI 1mg/ml and 0.5 ml of $AgNO_3$ 10 % in HNO_3 0,1 M. After 5 minutes, the solution is passed onto the second purification column at the speed of 3ml/min. For the complete recovery of the Mo, the hydrated zirconium oxide column will be washed with 10 ml of NaOH 0.2 N. If it is necessary,a pump can be used for the complete recovery of the solution. 55 ml of yellow-pale Natrium molybdate in NaOH 0.25 M will result. The results of the purification experiment are illustrated in table 3 and figure 5.

The recovery yield of the Mo is 92.95% for the *AC+AgAC column and 98.56% for the second column, with AC+AgAC+HO, and throughout all the purification process the yield is 91.59%. The Mo loss is ~8.4 %and can be explained by the fact that not all the efluent from the columns has not drained from the columns. If a pump had been used, this effect would not have occured.*

Tabelul 3. Separarea şi purificarea Mo-lui din soluţie de uraniu iradiat la nivel de trasor

No.	Isotope	Initial solution Λ [µCi]	Filtrate Λ [µCi]	Washing Λ [µCi]	Precipitate dissolution Λ [µCi]	The purification of the Mo solution	
						Purification on AC/AgAC Λ [µCi]	Purification AC/AgAC/ HZO Λ [µCi]
1	Zr-95	3.2	1.00E-03	0.21	0.00	0.0	0.0
2	Nb-95	6.659E-18	3.22E-11	1.23E-08	3.79E-07	4.56E-10	8.43E-12
3	Mo-99	69.08	1.18	-	62.26	8.05	7.53
4	Tc-99	-	3.001E-11	4.751E-12	5.97E-09	3.16E-09	5.74E-09
5	Te-132	119.78	19.59	0.67	1.97	1.87E-02	1.05E-02
6	I-131	36.71	10.261	0.48	2.41	0.00	0.00
7	I-132	68.94	15.25	1.01	8.62	0.165	2.95E-02
8	Ba-140	52.62	4.98	0.38	0.00	0.00	0.00
9	La-140	43.22	13.70	0.349	0.00	0.00	0.00

CONCLUSION

After the analysis performed on the solution at the Mo precipitate dissolution with α-BO, the recovery yield of the Mo is 89%. This can be explained by the fact that, in the moment of the dissolution of the precipitate in 1 N NaOH +1% H_2O_2 , a black precipitate appears – silver telluride which contains the Mo. Similarly, the fact that only 16.35 % of the Te-132 activity is found in the filtrate.

The analysis of the after-purification solutions show that the yield of Mo recovery is 92.95% for the AC+AgAC column and 98.56% for the second column with AC+AgAC+HOZ, and throughout all the process, the yield is 91.59%.

Although all the phases of the process are performed with high yields, the final yield of the recovery of Mo from U solutions are higher than 80%.

REFERENCE

1. D. Wu, S. Landsberger “ Processing of LEU targets for ^{99}Mo production-testing and modificatione of the CINTICHEM process”, International Meeting on Reduced Enrichment for Research and Test Reactors, 18 -21 septembrie, 1995
2. IAEA - TECDOC -1065 ‘”Production technologies for molibdenum -99 and technetium -99m” , februarie, 1999.

3. G.F. Vandergrift, Yoshikazu Koma, H. Cols,C. Conner, S. Aase, M Peter, D. Walker, R.A. Leonard, J.L. Snelgrove “Progress in chemical processing LEU - targets for ^{99}Mo production”, International Meeting on Reduced Enrichment for Research and Test Reactors, 5-10 octombrie , 1997
4. C. Conner, M. W. Liberatore, J. Sedlet, D.E. Walker, A. Mutalib si G. F. Vandergrift, “ Progress in developing processes for converting ^{99}Mo production from higt- to low - enriched uranium “ 1998
5. Deenis W. Wester, " Production and applications of molybdenum -99" Piteşti, 13-15 septembrie.2004.
6. G.F. Vandergrift, C. Conner, J. Sedlet, D.G.Wygmans," Progress in chemical processing of LEU targets for ^{99}Mo production, 1997.
7. H. Arino and P.M. Madigan " Production of Hight-Purity Molybdenum Using Silver -Coated Carbon as Asorbent" U.S. Patent 3,745119
8. S. El-Bayoumy and M. El Kolaly, " Some Radiochimical Studies on the Adsorption Behavior of Molybdenum-99 on Silver -Coated Carbon Granules and Activitated Carbon ", J. Radioanal. Chem. 60, 7 _1982)

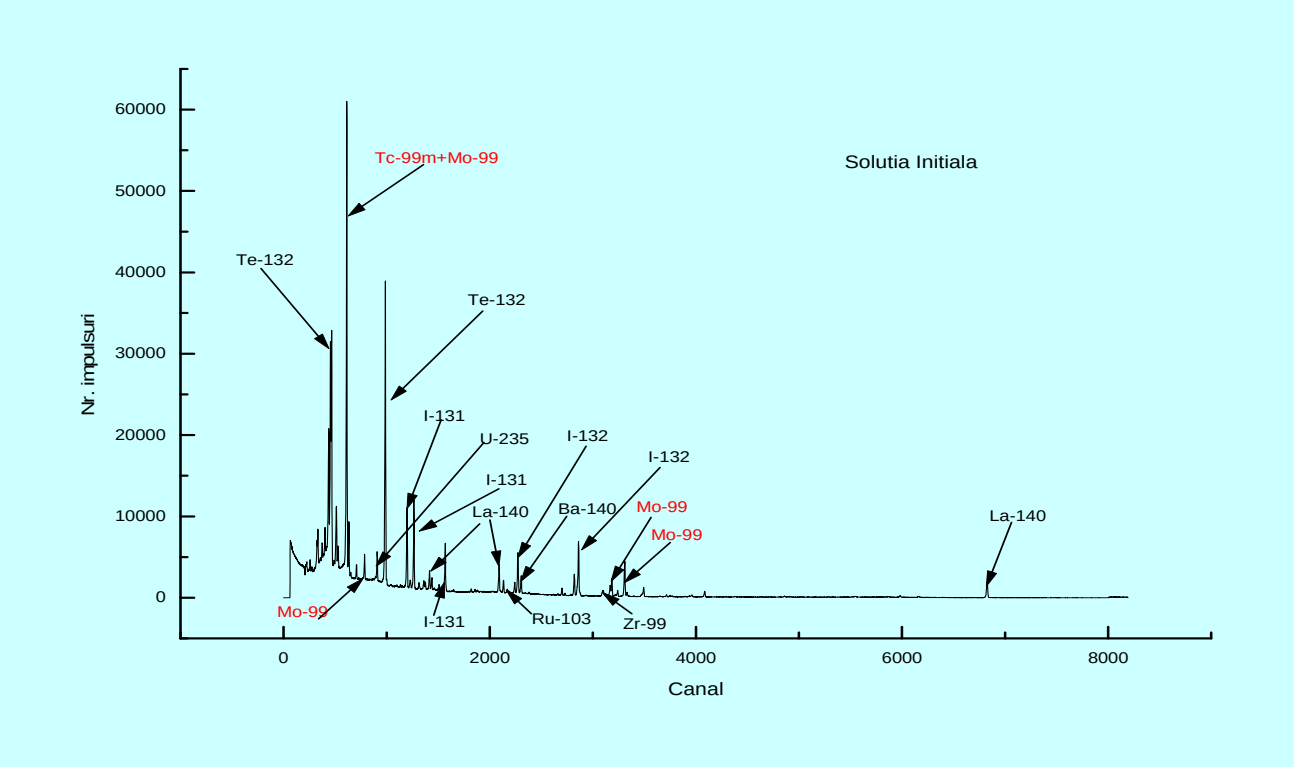

Figure 1 The spectre of the initial solution

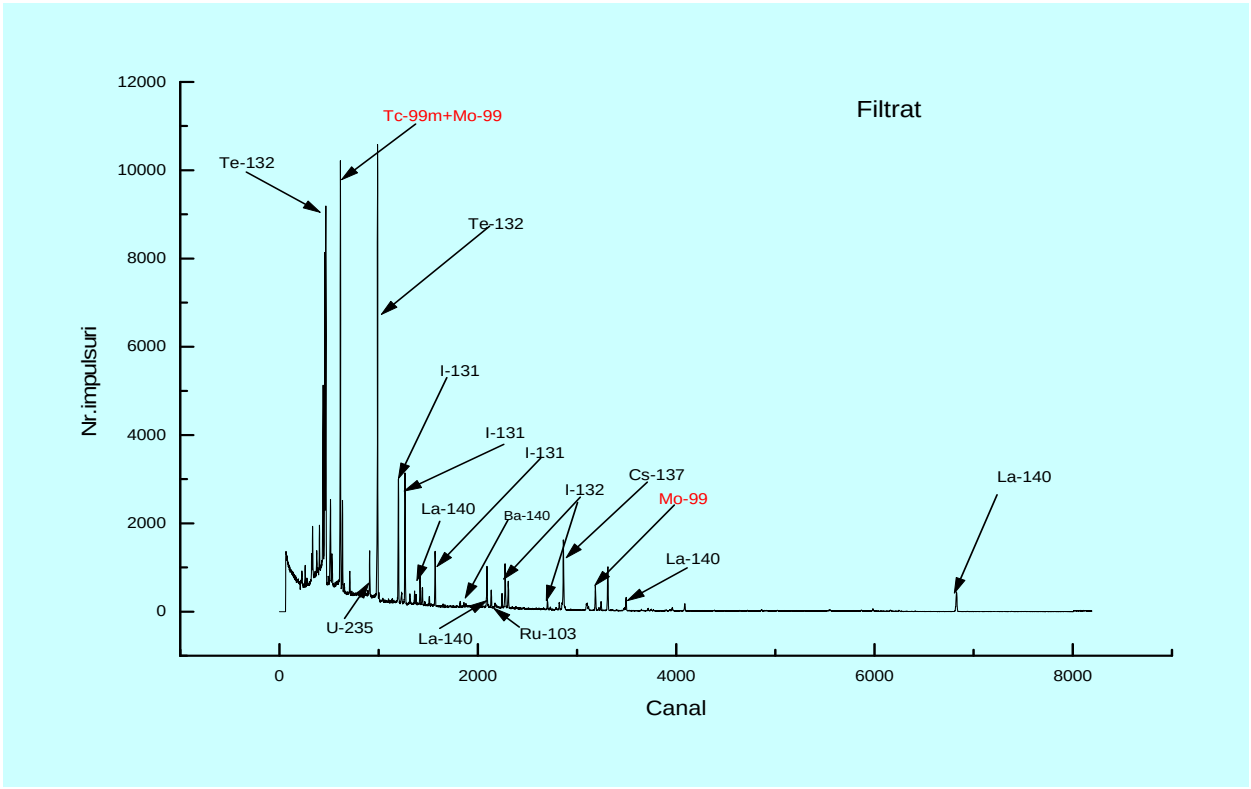

Figure 2. The spectre of the filtrated solution

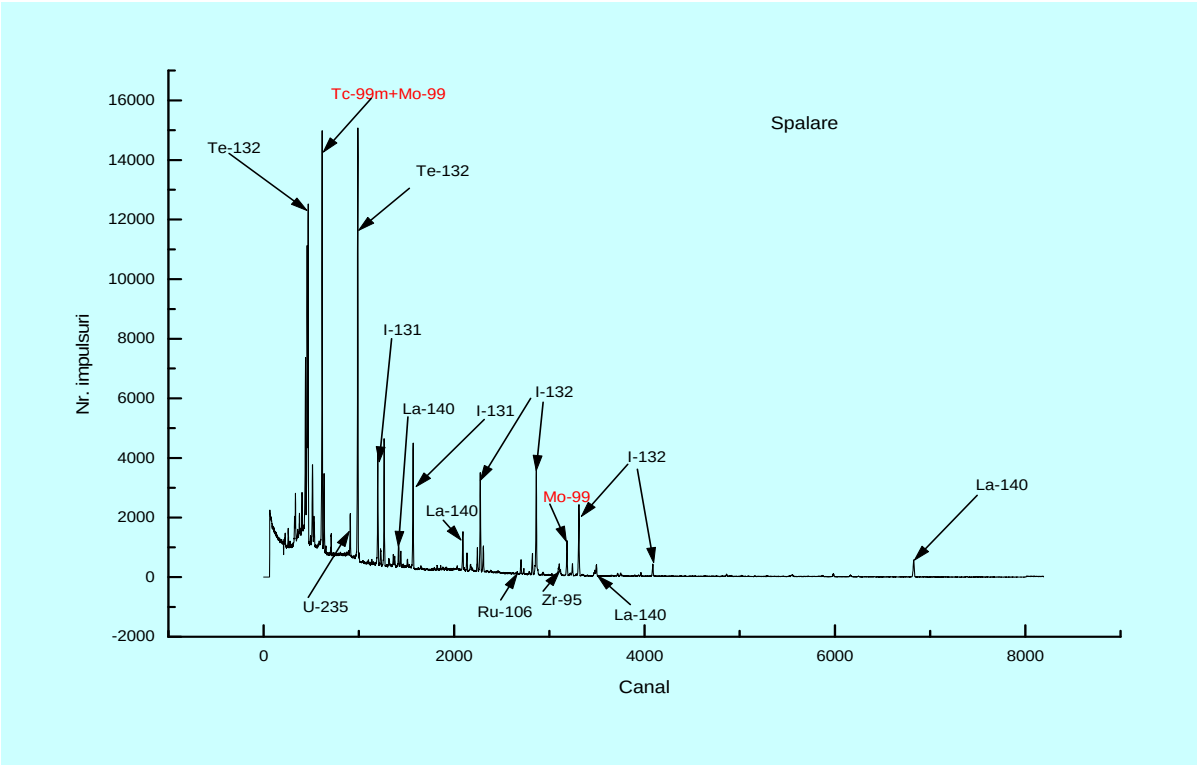

Figure 3. The spectre of the washing solution

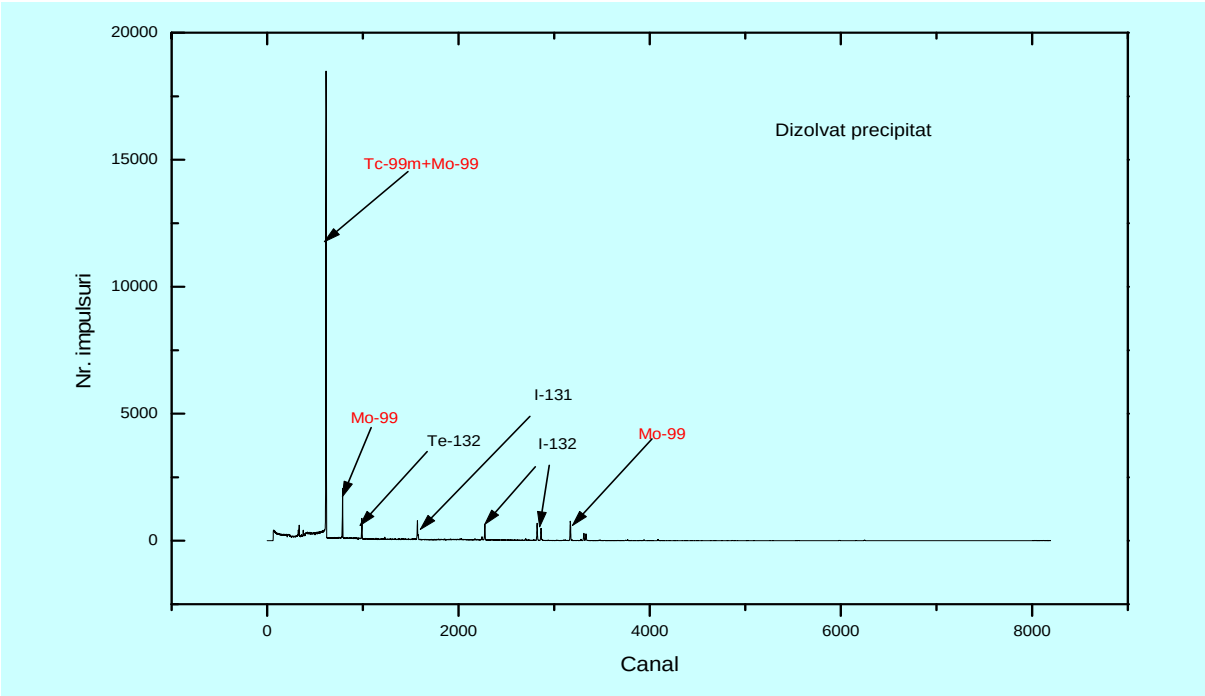

Figure 4. The spectre of the solution for the dissolution of the Mo- αBO precipitate

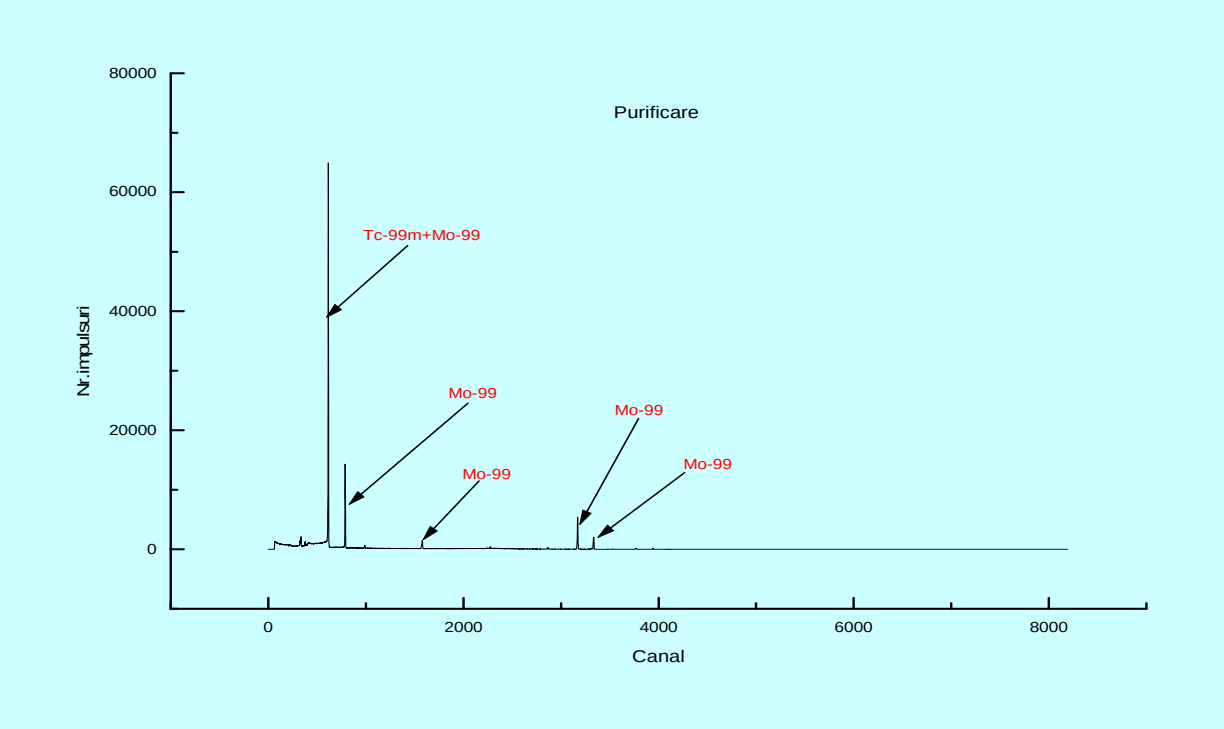

Figure 5. The spectre of the purified Mo 99 solution