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STUDY OF NEW ROUTES FOR PURIFICATION OF FISSION ^{99}Mo

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ABSTRACT

^{99m}Tc is the most widely applied medical radioisotope in the world, especially for cancer diagnosis procedures. It is provided by 99Mo radioactive decay, which is one of the fission products of irradiated uranium by nuclear reactors. At main production centers, ^{99}Mo chemical processing line up different steps to separate it from other fission products, depending on features of targets or local requirements. In this work, two new routes (MR1 and MR2) for ^{99}Mo purification are presented, called MR1 and MR2, can be set up in the Brazilian Multipurpose Reactor project (RMB). In the first studied route, ^{99}Mo purification initiated by eluting the solution through a Dowex 1x8 resin chromatography column, followed by a second step using a Chelex resin column and finally, the process was finished using an alumina column. For a second route, the first step is also performed with a Dowex 1x8 column, followed by a second separation step using alumina column, and lastly, a sublimation process was carried out in a tubular oven under programmed conditions. The final yield for the MR1 route has reached 84.4% and the overall time process about 7 hours, by contrast, MR2 route came to 75.3% in 9 hours.

1. INTRODUCTION

Currently ^{99m}Tc is the main radioisotope used in nuclear medicine worldwide due to its pure gamma decay of low energy (140 keV) and short half-life (6.02 hours). It is commonly used linked to other molecules known as markers, which have affinity for some specific organ or tissue [1, 2]. In the world, more than 22 million medical procedures are performed annually in diagnostic imaging tests using ^{99m}Tc [3].

^{99m}Tc can be provided in ^{99}Mo/^{99m}Tc generators, for medical centers through all country [4], [5]. Therefore, ^{99}Mo is the most important precursor for maintaining global supply of ^{99m}Tc. Fission ^{99}Mo production starts with the irradiation of ^{235}U targets (UAl$_x$/Al) in a nuclear reactor, followed by suitable chemical processing, where ^{99}Mo is separated and purified. After aliquotting, lots are shipped to the generators manufacturing units [3].

After the shortages occurred in 2008-2009, when some producers' reactors of the world were disabled, several countries started planning new capabilities to produce ^{99}Mo. Particularly in Brazil, it was created the Brazilian Multipurpose Reactor project (RMB), such as an action to hold the fission radioisotopes local demand. The project predicts a construction of nuclear reactor, chemical processing facilities and research laboratories [6, 7].

Radiochemical purification process is an important stage on ^{99}Mo production chain due, thin separation from the others fission radionuclides produced by uranium decay. According to main production processes (e.g. KSA and ROMOL-99), solution from target dissolution, with fission radionuclides include ^{99}Mo, receive a chemical treatment in several steps. The choice of purification methods depends on adopted process, but in general, these methods are based on chromatographic columns and sublimation of ^{99}Mo [8-10].

In this work, two routes of ^{99}Mo purification were studied in order to contribute to set up production process on RMB project facilities. These routes were based on two worldwide stablished processes, KSA and ROMOL-99. The Fig. 1 presents a flow chart of the reference processes and the studied routes (MR1 and MR2).

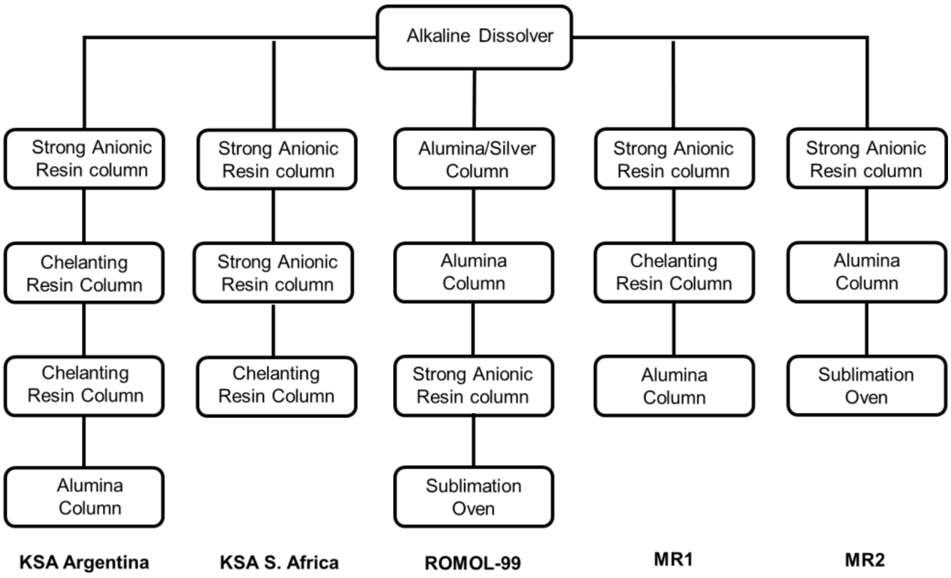

Figure 1: MR1 and MR2 routes of ^{99}Mo purification.

2. EXPERIMENTAL

The first studied route, MR1, was performed employing chromatographic columns filled witch strong anionic resin, chelating resin and inorganic ion exchanger alumina, respectively. The second route, MR2, was carried out including a sublimation step besides chromatographic columns. As showed in Figure 1, both purification routes presented the chromatography using a filled column with Dowex as a first step, and the following steps were performed differently.

The routes were tested using a previously prepared solution, which contains different elements with known concentrations. This solution represented a real solution from the alkaline dissolution of uranium targets and it was denominated load solution, which was composed of NaOH: 1 mol·L^{-1}, Al: 25 g·L^{-1}, Mo: 5 mg·L^{-1}, I: 1 mg·L^{-1}, Ru: 5 mg·L^{-1}, Te: 2 mg·L^{-1} e Zr: 7 mg·L^{-1}, and countless mass of ^{99}Mo, ^{131}I, ^{103}Ru, ^{121}Te and ^{95}Zr. Each test was carried out using 1,5 L of load solution, in duplicate.

The used material for experimental arrangement included hoses, valves and peristaltic pumps for controlling of volume and flow directions of radioactive solutions, considering the real dimensions of the purification process.

2.1. MR1 Route

For the ion-exchange chromatography of MR1 route's first step, glass columns 13 x 2.5 cm, filled with 35 g of Dowex 1x8 anionic resin without preconditioning were used. The columns were percolated with 1.5 L of load solution at flow rate 40 $mL\cdot min^{-1}$. After that, two bed washes were performed; the first one was done with 500 mL of 1 $mol\cdot L^{-1}$ NaOH solution at 20 mL min^{-1} and then washed with 500 ml of distilled water at 20 $mL\cdot min^{-1}$. The elutions of ^{99}Mo were made with 1.0 $mol\cdot L^{-1}$ Na_2SO_4 solutions at 3 $mL\cdot min^{-1}$.

The ^{99}Mo solution (210 mL) eluted from the Dowex 1x8 column were added with complexing agents, 140 mL of 6 $mol\cdot L^{-1}$ H_2SO_4, 18 mL of 6 $mol\cdot L^{-1}$ NH_4SCN, 14 mL of 0.5 $mol\cdot L^{-1}$ KI and 35 mL of 0.8 mol L^{-1} Na_2SO_3. The mixtures were allowed to stand for 15 minutes for preferential formation of the $[Mo(SCN)_6]^-$ complex.

After that, the charge solutions were percolated in the glass column (10 cm long by 1 cm diameter) filled with 7 g of resin Chelex previously conditioned with 50 ml of complexing solution. The column flowing from 5 $ml\cdot min^{-1}$ downwards and was washed with 50 ml of complexing solution. The ^{99}Mo elution was done with 1 $mol\cdot L^{-1}$ NaOH solution at a flow rate of 1 mL min^{-1}.

As a third step, glass column 10 cm long by 1 cm diameter filled with 10 g of acid alumina without preconditioning was used. The alkaline solutions eluted from the previous column were previously acidified with 1 $mol\cdot L^{-1}$ HNO_3 solution totaling approximately 80 mL, the percolation was conducted at 3 $mL\cdot min^{-1}$ flow downstream. The column was washed with 50 mL of distilled water, and then with 50 mL of 0.01 $mol\cdot L^{-1}$ NaOH solution. The elution was done with 1 $mol\cdot L^{-1}$ NaOH solution at 1 $mL\cdot min^{-1}$ flow rate.

2.2. MR2 Route

In MR2 route also was applied ion-exchange chromatography, including the thermic separation by sublimation. At first, the purification was started using a chromatographic column filled with Dowex 1x8 resin, and the alumina exchanger column was adopted in the followed step.

Both columns (with Dowex and alumina, respectively) were carried out based on the same method described previously, for the MR1 route, however, instead of 1.0 mol L^{-1} NaOH as eluting solution, it was used 1.0 mol L^{-1} $KHCO_3$ in the first column (Dowex) and 0.1 mol L^{-1} NH_4OH in the second one (alumina).

Before the sublimation step, the sample was pre-treated by evaporation. The eluted solution from chromatographic steps was heated at 80 ºC in a heating plate, for removing of excessed water and reducing of volume until about 5 mL. Then, this volume was transferred to a small platinum crucible, and heated until dryness.

After cooling, the crucible with ^{99}Mo solid residue was placed inside quartz tube, which one was fixed inside of the controlled programming oven. The oven's design was projected especially for these experiments. The set-up used was: sublimation temperature 850 ºC; heating rate: 15 ºC·min^{-1}; isotherm time 10 min; air flow inside the tube of 5 L h^{-1}.
Lastly, the molybdenum oxide, with ^{99}Mo, was carefully recovered by washes of internal part of tube, with 10 mL of 1 mol L^{-1} NaOH.

2.3. Measures of Radioactive Activities

All experiments were carried-out at 25 ºC and, in the end of each step of MR1 and MR2 routes, 10 mL of solutions (from loading, eluting and washing) were collected, for measure of activities and step efficiency calculations.

The measurements of the radioactive activities of the samples studied were performed using a gamma spectrometer with coaxial hyperpure Germanium detector (HPGe) model GX 1518, coupled with a Canberra Inc. multi-channel acquisition system.

The main emission peaks of the radioisotopes involved in the purification route tests were selected for the counts: ^{99}Mo 739 keV, ^{103}Ru 497 keV, ^{121}Te 573 keV and ^{95}Zr 756 keV. The yield rates of the experiments were calculated by equation 1:

$$R = \left(\frac{A_f}{A_i}\right) \times 100 \tag{1}$$

Where R is the elution, distillation or sublimation yield of the given radioisotope, A_f and A_i are two respective final and initial areas of the principal peaks of radioactive activities at a given acquisition time.

The result adopted for each study of the purification stages corresponds to the arithmetic averages of representative experiments among the others.

In the complete radioisotope purification route tests, total yields were obtained by successively multiplying the yield means of the steps involved by the equation 2:

$$R_t = R_1 \cdot R_2 \cdot R_{(n)} \tag{2}$$

Where R_t is the total recovery yield of a given radioisotope and R (1, 2, …, n) correspond to the arithmetic means of the yields in the purification steps of the tested routes.

3. RESULTS AND DISCUSSION

As mentioned, the first step of the two routes was ion-exchange chromatography using a column filled with Dowex 1x8 resin. At this step, molybdenum-99 present in solution as molybdate ions $\left(\mathrm{MoO}_2^{-4}\right)$ was retained in the column, while aluminum ions and other contaminants produced via alkaline dissolution of ^{235}U targets, in significant concentrations, remained in the solution, which was called "effluent". The retained ^{99}Mo was recovered by elution with Na_2SO_4 solution.

The second step, the routes presented different techniques for the purification of ^{99}Mo. In the MR1 route, a selective chelating column was used that retained the Mo−SCN, obtaining the best separation results between ^{99}Mo and the contaminants. On the other hand, in the route MR2, the major decontamination was carried out in the third step, in the process of sublimation. In both routes, the alumina column played auxiliary role to concentrate molybdenum-99.

3.1. MR1 Route

3.1.1. Dowex 1x8 column

Table 1 are presented the final yields in percentage of collected samples from elution, effluent and washes solutions, obtained after first step, using Dowex resin and Na_2SO_4 eluting solution. The choice of the Na_2SO_4 solution as the ^{99}Mo eluent was taken due to the next complexation step, because it does not interfere with the ^{99}Mo complexation reactions [11].

Table 1: Elution yields of ^{99}Mo and contaminants ^{131}I, ^{103}Ru, ^{121}Te and ^{95}Zr with the Dowex resin in MR1 route

Experiments	^{99}Mo (%)	^{131}I (%)	^{103}Ru (%)	^{121}Te (%)	^{95}Zr (%)
Efluent 1	0.30	1.70	92.6	82.7	68.8
Efluent 2	0.40	< DL	72.4	78.7	86.3
Average	0.35 (± 0.05)	–	82.5 (± 10.1)	80.7 (± 2.00)	77.5 (± 8,75)
Wash 1 OH^-	0.10	0.10	0.50	2.30	1.80
Wash 2 OH^-	< LD	< DL	3.50	2.10	1.80
Average	–	–	2.00 (± 1.50)	2.20 (± 0.10)	1.80 (± 0,00)
Wash 1 H_2O	< LD	< DL	< DL	< DL	< DL
Wash 2 H_2O	< LD	< DL	0.10	0.50	< DL
Média	–	–	–	–	–
Elution 1	99.0	< DL	0.40	0,90	9.10
Elution 2	98.4	0.76	2.99	4.10	5.50
Average	98.7 (± 0.30)	0.76	1.69 (± 1.29)	2.50 (± 1,60)	7.30 (± 1.85)

< DL = under detection limit

The analysis of results indicated a low ^{99}Mo concentration in effluent samples and high retention in the resin. The ^{99}Mo recovery was greater than 90% after the final elution, confirmed by the low loss observed after the percolation of load solution and washes. For other contaminants, the totals of elimination (effluent + washes) were about 79.3% (^{95}Zr), 82.9% (^{121}Te) and 84.5% (^{103}Ru). Due to the low concentrations of the contaminants in some samples, their radioactivity measurements presented the results lower than the limit of detection (<LD), in the measurement conditions employed

The Dowex 1x8 resin was very useful for significant separation of the Mo-99 from the other fission elements present in the initial process solution, according to obtained results.

3.1.2. Chelex 100 column

In the second step of MR1 route, the complexation of ^{99}Mo in the presence of thiocyanate ions and the chelating chromatography were performed using the eluted solution from Dowex column. The obtained results in this process are shown in Table 2.

Table 2: Elution yields of ^{99}Mo with the Chelex resin

Experiments	^{99}Mo (%)
Efluent 1	< DL
Efluent 2	< DL
Average	–
Wash 1	2.30
Wash 2	1.82
Average	2.06 (± 0.24)
Elution 1	96.2
Elution 2	98.4
Average	97.3 (± 1.10)

< DL = under detection limit

As the previous step, molybdenum recovery was also elevated, presenting elution average higher than 97% and ^{99}Mo loss about 2%, during the washes. The analysis of contaminants indicated radioactivities lower than the limit of detection, therefore their values were not presented.

3.1.3 Alumina column

The alumina column was used to reduce the concentrations of the remaining contaminants, as well as the elimination of possible residues from hoses, connections and resins. The capacity to concentrate the ^{99}Mo in the final eluted solution also was observed in this step. The obtained values of yield, in percentage, are listed in Table 3.

The recovery of ^{99}Mo, in this step, was approximately 88% on average, presenting lower result than the previous columns, however the samples purities improved, once the monitored contaminants showed radioactivities under the limit of detection.

Table 3: Elution yields of ^{99}Mo with the alumina resin in the MR1 route.

Experiments	^{99}Mo (%)
Efluent 1	0.90
Efluent 2	0.30
Average	0.60 (± 0.30)
Wash 1 OH^-	0.30
Wash 2 OH^-	0.40
Average	0.35 (± 0.05)
Wash 1 H_2O.	0.50
Wash 2 H_2O	0.20
Average	0.35 (± 0.15)
Elution 1	89.1
Elution 2	86.7
Average	87.9 (± 1.20)

The results of total route, in the end of all tested steps, and the final yield considering the consecutive losses of the process are presented in Table 4. These values were obtained by multiplying the yields in the three process steps.

Table 4: Total yield of MR1 route, in percentage

Step 1 (%)	Step 2 (%)	Step 3 (%)	Total (%)
98.7 (± 0.30)	97.3 (± 1.10)	87.9 (± 1.20)	84.4 (± 2.36)

The final yield of ^{99}Mo in the MR1 route was above 80%, besides the high purity demonstrated by the low radioactivities of contaminants.

The developed arrangement presented advantages in comparison with the original process [12], which one, two redundant chelating columns were used. The total process time required to complete all steps and material transfers, was approximately 7 hours.

3.2. MR2 Route

3.2.1. Dowex 1x8 column

Despite the Dowex resin had been used in the first step of both routes, the eluting solution used in each case was distinct. In MR2 route, $KHCO_3$ solution was used for ^{99}Mo recovery after percolation in Dowex resin. The potassium bicarbonate solution was chosen to the recovery of molybdenum considering the next step of purification this route, because the sulfate as eluent could interfere in the ^{99}Mo retention step in the alumina, increasing the loss in the effluent. [11]. The results for these experiments are shown in Table 5.

Table 5: Elution yields of ^{99}Mo and contaminants with the Dowex resin in the MR2 route.

Experiments	**^{99}Mo (%)**	**^{131}I (%)**	**^{103}Ru (%)**	**^{121}Te (%)**	**^{95}Zr (%)**
Efluent 1	0.10	0.20	85.1	87.3	70.5
Efluent 2	1.80	0.10	72.4	65.6	64.5
Average	0.95 (± 0.85)	0.15 (± 0.05)	78.7 (± 6.35)	76.4 (± 10.8)	67.5 (± 3.00)
Wash 1 OH^-	< DL	<DL	1.10	3.20	0.40
Wash 2 OH^-	1.20	<DL	1.84	4.60	0.87
Average	-	-	1.47 (± 0.37)	3.90 (± 0.70)	0.63 (± 0.23)
Wash 1 H_2O	< DL	<DL	0.10	< DL	0.20
Wash 2 H_2O	< DL	<DL	0.02	< DL	0.07
Média	-	-	0.06 (± 0.04)	-	0.13 (± 0.06)
Elution 1	97.4	0.20	0.30	< DL	3.50
Elution 2	96.4	0.30	2.20	< DL	7.19
Average	**96.9 (± 0.50)**	0.25 (± 0.05)	1.25 (± 0.95)	-	5.34 (± 1.84)

< DL = under detection limit

The concentration of ^{99}Mo and contaminants in the effluent, wash and elution samples were similar to the obtained values from the same column in the route MR1. However, there was a lower elimination of contaminants in the effluent and a smaller concentration of ^{121}Te in the eluted solution.

3.2.2. Alumina column

The aluminum oxide column on this route had the function of purifying ^{99}Mo after its initial separation of the contaminants, in addition to the preparation for the next step of the route (sublimation).

The acidification of solutions eluted from the previous column before the percolations in the columns of alumina, besides adjusting the pH for this column also favors the volatilization of part of the residuals of ^{131}I still present in the solution. The values obtained in the performed tests are presented in Table 6.

Table 6: Elution yields of ^{99}Mo and contaminants with the alumina resin in the MR2 route.

Experiments	**^{99}Mo (%)**	**^{103}Ru (%)**	**^{95}Zr (%)**
Efluent 1	0.90	2.58	4.60
Efluent 2	1.30	4.10	1.60
Average	1.10 (± 0.20)	3.34 (± 0.76)	3,10 (± 1,50)
Wash 1 H_2O	0.60	< DL	< DL
Wash 2 H_2O	1.20	< DL	< DL
Average	0.90 (± 0.30)	-	-
Wash 1 OH^-	0.20	< DL	< DL
Wash 2 OH^-	0.00	< DL	< DL
Average	0.10 (± 0.10)	-	-
Elution 1	90.2	< DL	< DL
Elution 2	82.3	< DL	< DL
Average	86.2 (± 3.95)	-	-

< DL = under detection limit

In this column small losses of Mo-99, approximately 1%, were observed in the percolation and washing steps. Small amounts of the Ru-103 and Zr-95 contaminants in the effluents were also observed.

In the following steps, the presence of the contaminants was not detected in the ^{99}Mo elutions, reaffirming the high purification capacity of this column at this position of the route.

3.2.3. Mo-99 sublimation

The ammonium hydroxide solution was chosen as the eluent for ^{99}Mo in the previous step of this route, due to the interference of the sodium ions in the next step, sublimation. The elution with NaOH solution would produce, by thermal decomposition, the sodium oxide together with the molybdenum oxide, thus making difficult the exit of ^{99}Mo at its sublimation temperature [13]. Table 7 are shown the yields of the ^{99}Mo sublimation process.

Table 7: Yields of sublimation of ^{99}Mo in tubular furnace

Experiment	**^{99}Mo (%)**	**Residue ^{99}Mo (%)**
1	91.5	5.02
2	88.7	4.30
Average	90.1 (± 1.40)	4.66 (± 0.36)

The recovery rate of ^{99}Mo was above 90% on average. Although the sample preparation step was carefully followed, the apparatus used certainly allowed losses during the evaporations. The remaining residues in the crucible presented values around 4%.

These values could be improved by carrying out further tests, adjusting the working conditions in the kiln and developing specific glassware for the preparation of samples in which they could contribute to the reduction of material losses.

Table 8 are shown the averages obtained in the steps of this route and the final yield, considering the consecutive losses of the process.

Table 8: Total yield of ^{99}Mo in the MR2 route, in percentage

Step 1 (%)	Step 2 (%)	Step 3 (%)	Total (%)
96,9 (± 0,50)	86,2 (± 3,95)	90,1 (± 1,40)	75,3 (± 5,00)

The total time demanded in the process steps and material transfers was approximately 9 hours. This route had a final yield of 75.3% and all the contaminants presented activities below the limit of detection, thus indicating a high degree of purification of 99Mo.

This route is similar to the ROMOL-99 process but with the inversion of Dowex 1x8 and alumina columns it was possible to bypass a delicate step of the reference process, which involves acidifying the solution to a narrow pH range (0.3 a 0.5) difficult to perform given the working conditions and the activities of the solutions. In addition, the initial 99Mo alkaline solution in ammonium hydroxide gave easier decomposition than the ammonium carbonate solution used in the ROMOL-99 process [9].

3.3. Comparative of Processes

The final yields of ^{99}Mo purification routes are shown in Table 9.

Table 9 – Final Process Yields

Purification route	MR1	MR2
Final Yield (%)	**84,4 (± 2,36)**	75,2 (± 5,00)

Among the routes tested, the MR1 route presented the highest final purification yield of ^{99}Mo (84.4 %) and with a high degree of decontamination. The results indicated that after the initial ^{99}Mo separation on anionic resin column, only two more chromatographic columns were sufficient for satisfactory purification of this radioisotope. This result resembles the process used in a South African producer center that also employs three chromatographic columns in its process and has a yield of approximately 85 % [14].

The process time of the MR1 route was also lower than MR2 route because, despite the additional reductions of chromatographic columns, the sublimation technique required a longer time. In addition, the operating conditions of the chelating column may possibly still be modified and simplified by changing the chemical conditions of the solution-charge so that the molybdate ion can be fixed directly to the resin, and the complexation step may be dispensed.

The MR2 route also showed promising recovery and purification yields, yet operational enhancements in the sublimation step are still necessary in order to reduce the operating time, such as higher heating rates in the initial periods of the furnace heating ramp and the installation of a forced cooling system. Such measures would certainly reduce the overall process time. The inversion in the elution direction of the alumina column and the development of a more suitable concentrator system could also contribute to the improvement in the efficiency of the MR2 route.

The difficulties of measuring the activities of the contaminating radioisotopes could perhaps be bypassed by increasing the irradiation times of the starting salts or by reducing the masses of their respective carriers. In parallel, known quantities of the respective loaders and tracers could be added to the samples, such measure would function as an internal standard addition technique, increasing its activities and reducing measurement errors, eliminating the problems of increasing differences in activities at each stage of the process.

CONCLUSIONS

In an analogous way to the variations of the KSA process adopted in Argentina and South Africa, whose alkaline dissolution of the targets is also employed, the ion exchange technique using anionic resin column, as an initial process step, was also applied here. The use of Dowex 1x8 resin in the first column of the process offered both the effective elimination capacity of most of the contaminants present in the solution-charge as well as the possibility of selective elution of the ^{99}Mo radioisotope.

Additionally, this process configuration avoids the need for pre-acidification of the solution-load, as occurs in processes such as ROMOL-99, which represents an extra difficulty of the process given the precipitation of the high aluminum load of the solution under these conditions.

Among the routes tested, the MR1 route presented the highest final recovery efficiency of ^{99}Mo with a high degree of decontamination and a shorter process time compared to the MR2 route. The results indicate the possibility of implementing a short and efficient production process using only three chromatographic columns, similar to the process used in South Africa.

The results also provide subsidies for further purification tests on pilot cells, targeting the production at RMB facilities.

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