

# RECOVERY OF $^{131}\text{I}$ FROM FISSION $^{99}\text{Mo}$ PRODUCTION PROCESS BY DISTILLATION METHOD

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## ABSTRACT

The results of one of the recovery stages of  $^{131}\text{I}$  as a byproduct of the  $^{99}\text{Mo}$  fission production process are presented in this work. A modified wet-distillation apparatus was used for iodide purification and some work parameters were tested. The assays included iodide anion eluent, concentration of  $\text{H}_2\text{SO}_4$ ,  $\text{H}_2\text{O}_2$ , the catalyst effect of  $\text{MoO}_3$ , temperature and time process. The results of eluting anions demonstrated lower recovery of  $^{131}\text{I}$  using nitrate salt. The sulfate solution and its respective acid ( $\text{H}_2\text{SO}_4$ ) at concentration 1.6 M, presented better results. Hydrogen peroxide was used as a main oxidant of the ions iodide, the experiments realized with 6 mL of  $\text{H}_2\text{O}_2$  3% showed higher recovery than assays prepared with  $\text{H}_2\text{O}_2$  higher and lower amount. Molybdenum oxide was evaluated as catalyst of iodine in solution; however, the results did not demonstrate an advantage in relation to its absence. In addition, the recovery of iodide was delayed at initial periods of the process in presence of the catalyst, leading to an increase on time process. Temperature decrease of distillation solution from 80 to 50 °C showed advantageous since the recovery yields were similar beside a lower wet drag to trap solution. Time distillation was carried out at least four hours with sampling each hour; the best times were between 1 and 2 hours. Shorter periods of distillation avoid dilution of the capture solution on the further recovery process. Several parameters were studied and it was possible to set up system that provided  $^{131}\text{I}$  recovery around 50% yield.

## 1. INTRODUCTION

The radioisotope  $^{131}\text{I}$  is one of most important for treatment of thyroid gland cancer and it is widely used in nuclear medicine for diagnosis and therapy of several diseases. Nowadays its production can be realized through two methods involving nuclear reactions. One of them involves the irradiation of  $^{235}\text{U}$  and the recovery of  $^{131}\text{I}$  from the fission products produced and the other includes the extraction of  $^{131}\text{I}$  from irradiated tellurium. Natural or enriched metallic uranium alloy can be used as a target for irradiation in the first method cited. Usually enriched uranium is employed in the form of uranium-aluminum alloy for  $^{131}\text{I}$  production in large scale [1]. The processing technology of  $^{235}\text{U}$  fission products is highly sophisticated, expensive and generates large amounts of mixed radioactive wastes. The extraction and purification of  $^{131}\text{I}$  produced by fission can be achieved in different ways. There are reports in the literature using solvent extraction, distillation methods, platinum sorption process, ion exchange separation, charcoal column chromatography, adsorption on a silver coated alumina column, among others [2, 3]. Brazil is not self-sufficient in the production of  $^{131}\text{I}$  and others radioisotopes generated by the fission of  $^{235}\text{U}$ , being hardly dependent of international producers. In order to solve this problem of dependence, currently, the country develops the Brazilian Multipurpose Reactor (RMB) project that will provide enough iodine-131 and molybdenum-99 (which generates  $^{99\text{m}}\text{Tc}$  for nuclear medicine). Besides providing  $^{131}\text{I}$  and  $^{99}\text{Mo}$ , this reactor will supply all needs of radioisotopes production in Brazil for medical and

Further researches about the process of separation and product of  $^{235}\text{U}$  fission are needed. This work is part of the KIMB research project to separate and purify the fission  $^{131}\text{I}$  as a byproduct of the  $^{99}\text{Mo}$  production process, the results of one of the recovery stages of  $^{131}\text{I}$  studies are being presented here. A modified wet-distillation apparatus for the iodide purification was used and some work parameters as the role of a catalyst in the process, the behavior of different iodide anion eluents, volumes of oxidant of the ions iodide, process temperatures and various acid concentrations were investigated [4].

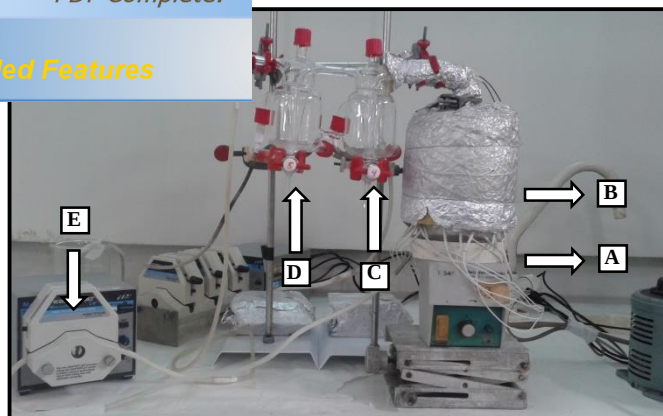
## 2. EXPERIMENTAL

### 2.1. Materials and Equipments

A  $1 \text{ mol L}^{-1}$  sodium sulfate solution, a  $1 \text{ mol L}^{-1}$  potassium nitrate solution and a  $1 \text{ g L}^{-1}$  potassium iodide solution were prepared by dissolving respective salts (Merck) in distilled water. Different concentrations of sulfuric acid were prepared by dilution as well as  $3.2 \text{ mol L}^{-1}$  nitric acid and  $0.1 \text{ mol L}^{-1}$  sodium hydroxide. Hydrogen peroxide 3% (Rioquímica) and molybdenum oxide (Merck) were used. The  $^{131}\text{I}$  samples used as a tracer were provided by the Center of Radiopharmacy, IPEN-CNEN/SP (Nuclear and Energy Research Institute). The instrument used throughout the work consisted of a gamma ray spectrometer HPGe (hyperpure germanium) connected to a multichannel analyzer (Canberra) with detector at 364 Kev.

### 2.2. Distillation System

The distillation system included a shaking hot plate where a flask containing the solution to be distilled was covered by a heated quartz bell jar, this hot stage was linked to two cooled washing flask for the uptake of iodine, a peristaltic pump provided the air and iodine drag, with reentry of iodine into the system on eventual exhaust (**Figure 1**). Simulating an iodide extraction solution on a chromatographic column, initial solution was composed by  $\text{Na}_2\text{SO}_4$  and KI,  $\text{H}_2\text{SO}_4$ ,  $\text{H}_2\text{O}_2$ , and  $^{131}\text{I}$  (as a radiotracer) were added. Iodine vapors were trapped at  $0.1 \text{ mol L}^{-1}$  NaOH solutions and samples were collected for recovery yield calculations after predetermined time intervals. Several assays were realized in this system evaluating iodide anion eluent,  $\text{H}_2\text{SO}_4$  concentration,  $\text{H}_2\text{O}_2$  concentration, catalyst effect of  $\text{MoO}_3$ , temperature and process time.



**Figure 1: Distillation System (A= Flask containing distilling solution; B= Hot Quartz bell jar; C= Flask for iodine uptake (recovery); D=Flask for iodine trap (safety); E= Peristaltic pump).**

### 2.1.1. MoO<sub>3</sub> Catalyst Effect

The catalyst effect of MoO<sub>3</sub> was evaluated in a system containing 20 mL of Na<sub>2</sub>SO<sub>4</sub> (1 mol L<sup>-1</sup>), 0.5 mL of KI (1 g L<sup>-1</sup>), 13 mL of H<sub>2</sub>SO<sub>4</sub> (1.6 mol L<sup>-1</sup>), 6 mL of H<sub>2</sub>O<sub>2</sub> 3% and <sup>131</sup>I as a radiotracer. Three assays were prepared using 0.1 g of molybdenum oxide dissolved in 0.5 mL of sodium hydroxide and six assays without the presence of oxide in the same conditions. The assays were performed during 2 hours at 80 °C. Additionally, it was studied the behavior of molybdenum oxide for an extended period of 4 hours, the assays were performed in triplicate containing or not molybdenum oxide in the same conditions described previously.

### 2.1.2. Iodide Anion Eluent

Nitrate and sulfate anions were studied with no catalyst. The experiments using nitrate were carried out with 0.5 mL of KI (1g L<sup>-1</sup>), 6 mL of H<sub>2</sub>O<sub>2</sub> (3%) and <sup>131</sup>I as a radiotracer. The amount of Na<sub>2</sub>SO<sub>4</sub> (1 mol L<sup>-1</sup>) or KNO<sub>3</sub> (1 mol L<sup>-1</sup>) established was 20 mL and H<sub>2</sub>SO<sub>4</sub> (1.6 mol L<sup>-1</sup>) or HNO<sub>3</sub> (3.2 mol L<sup>-1</sup>) 13.5 mL each. Initially KI solution was added into the flask of distillation with sulfate or nitrate solution, H<sub>2</sub>SO<sub>4</sub> or HNO<sub>3</sub>, H<sub>2</sub>O<sub>2</sub> and <sup>131</sup>I as a radiotracer were added. The distillation process was performed during 2 hours at 80 °C.

### 2.1.3. H<sub>2</sub>O<sub>2</sub> Concentration

Hydrogen peroxide was used as the main oxidant of the iodide ions. Three different H<sub>2</sub>O<sub>2</sub> amount were evaluated (3.0, 6.0 and 12.0 mL) under the same conditions described previously with no catalyst. Na<sub>2</sub>SO<sub>4</sub> (1 mol L<sup>-1</sup>), KI, H<sub>2</sub>SO<sub>4</sub> (1.6 mol L<sup>-1</sup>) and <sup>131</sup>I as radiotracer were included. The distillation process was performed during 2 hours at 80 °C.

Two temperatures were investigated during the distillation process, 80 °C and 50 °C. The assays were realized in triplicate during 4 hours. 20 mL of Na<sub>2</sub>SO<sub>4</sub> (1 mol L<sup>-1</sup>), 0.5 mL of KI (1 g L<sup>-1</sup>), 13.5 mL of H<sub>2</sub>SO<sub>4</sub> (1.6 mol L<sup>-1</sup>), 6 mL de H<sub>2</sub>O<sub>2</sub> (3%) and <sup>131</sup>I as the radiotracer were added without the presence of catalyst.

### 2.1.5. H<sub>2</sub>SO<sub>4</sub> Concentration

Four different concentrations of H<sub>2</sub>SO<sub>4</sub> were studied: 0.8, 1.6, 6.4 and 12.8 mol L<sup>-1</sup>, in the absence of the catalyst. The assays were realized in duplicate using 20 mL of Na<sub>2</sub>SO<sub>4</sub> (1 mol L<sup>-1</sup>), 0.5 mL of KI (1 g L<sup>-1</sup>), 13.5 mL of H<sub>2</sub>SO<sub>4</sub>, 6 mL de H<sub>2</sub>O<sub>2</sub> (3%) and <sup>131</sup>I as the radiotracer. The temperature used in the distillation process was 50 °C during 4 hours.

### 2.3. Analysis

The samples of iodine vapors trapped at NaOH solution were collected at predetermined time intervals and were analyzed by gamma ray spectrometer HPGe (hyperpure germanium) connected to a multichannel analyzer (Canberra) with the detector at 364 Kev. The areas of detection peaks correspond to the counts at selected energy channel. Iodine recovery calculation was made considering reference and recovery solution count values and respective amounts. Likewise, the percentages of safety and final solutions were calculated.

## 3. RESULTS

### 3.1. MoO<sub>3</sub> Catalyst Effect

The results generated in the experiments comparing presence and absence of catalyst are illustrated in **Table 1** and in **Figure 2** and **3** that show assays realized during two and four hours, respectively. In the assays realized during two hours it was observed value of iodine recovery  $46.22 \pm 6.75\%$  in the presence of catalyst and in the absence  $54.98 \pm 10.36\%$ . Considering the standard deviations values, with and no catalyst were similar indicating that the addition of molybdenum oxide did not promote a greater recovery of iodine. In the assays realized during four hours it was noticed that average percentage of iodine recovery in the presence of MoO<sub>3</sub> was  $53.05 \pm 1.18\%$  and in the absence  $51.93 \pm 1.28\%$  demonstrating again that the catalyst did not promote significant gains. Additionally, it was observed that the recovery of iodide was delayed in the initial periods of the process in the presence of the catalyst. In the assays realized in 2 hours it was verified that in the presence of catalyst the iodine recovery percentages at times 30, 60, 90 and 120 minutes were, respectively,  $3.45 \pm 0.01\%$ ,  $5.09 \pm 0.59\%$ ,  $32.68 \pm 5.00\%$  and  $45.57 \pm 9.42\%$  whereas in the absence of catalyst the respective percentages were  $19.82 \pm 5.35\%$ ,  $44.20 \pm 7.09\%$ ,  $49.23 \pm 9.57\%$  and  $53.92 \pm 10.78\%$ . These results indicate the presence of catalyst leads to increase time process.

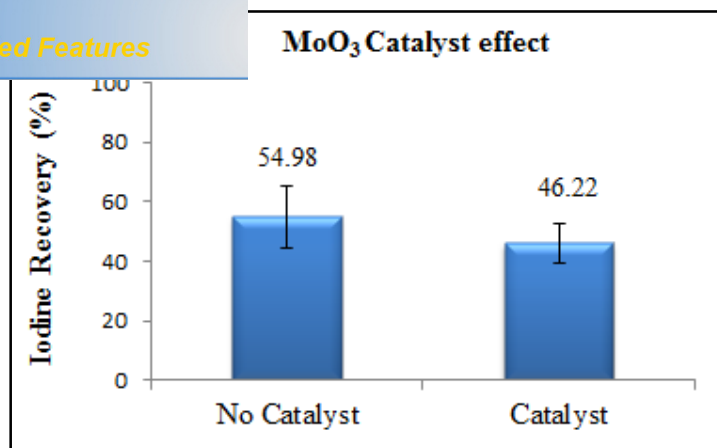


Figure 2: Iodine recovery in assays realized during 2 hours containing or not a catalyst

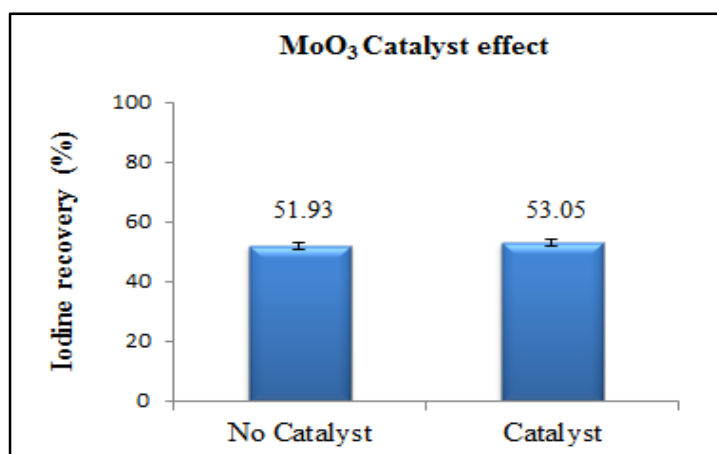


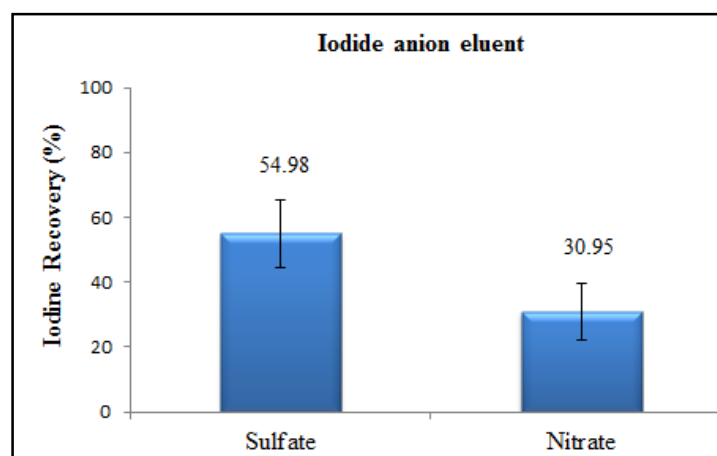
Figure 3: Iodine recovery in assays realized during 4 hours containing or not a catalyst

Table 1: Iodine recovery in different assays performed during two hours

| Catalyst<br>MoO <sub>3</sub> | ID<br>(%)     | Anion<br>Eluent | ID<br>(%)     | H <sub>2</sub> O <sub>2</sub><br>3.0%<br>(mL) | ID<br>(%)    | T<br>(°C) | ID<br>(%)    | H <sub>2</sub> SO <sub>4</sub><br>(mol L <sup>-1</sup> ) | ID<br>(%)     |
|------------------------------|---------------|-----------------|---------------|-----------------------------------------------|--------------|-----------|--------------|----------------------------------------------------------|---------------|
| Presence                     | 46.22 ± 6.75  | Nitrate         | 30.95 ± 8.65  | 3.0                                           | 48.61 ± 3.21 | 50        | 51.60 ± 3.76 | 0.8                                                      | 37.11 ± 5.34  |
| Absence                      | 54.98 ± 10.36 | Sulfate         | 54.98 ± 10.37 | 6.0                                           | 51.17 ± 2.37 | 80        | 46.23 ± 2.44 | 1.6                                                      | 48.76 ± 10.04 |
|                              |               |                 |               | 12.0                                          | 43.55 ± 9.48 |           |              | 6.4                                                      | 40.23 ± 2.88  |
|                              |               |                 |               |                                               |              |           |              | 12.8                                                     | 24.11 ± 6.62  |

ID (Iodine Recovery %); T (Temperature)

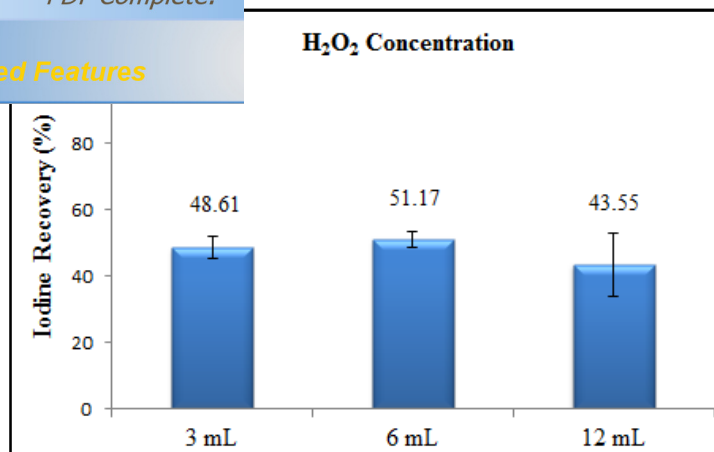
The results of assays involving the anions sulfate and nitrate realized during two hours with no catalyst are illustrated in the **Figure 4**. It was observed that the mean value of iodine recovery percentage in nitrate medium was  $30.95 \pm 8.65$  % and in sulfate medium was  $54.98 \pm 10.37$  % (**Table 1**). The nitrate medium showed an iodine recovery of 24.03%, lower than the sulfate medium considering the standard deviations obtained, probably due to the greater number of non-volatile iodine species formed in the presence of this ion [5]. Therefore, it was concluded that sulfate medium was more suitable on the process of iodine recovery.



**Figure 4: Comparison of iodine recovery on sulfate and nitrate medium**

### 3.3. H<sub>2</sub>O<sub>2</sub> Concentration

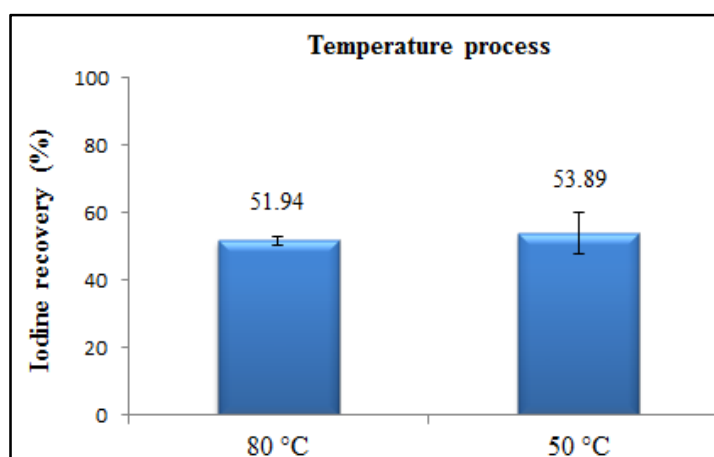
The results of iodine recovery in three different concentrations of hydrogen peroxide are demonstrated in the **Figure 5**. It was observed that using 3 mL of H<sub>2</sub>O<sub>2</sub> the iodine recovery was  $48.61 \pm 3.21$ % and using 6 mL of H<sub>2</sub>O<sub>2</sub> the recovery increase to  $51.17 \pm 2.37$ % and adding 12 mL of peroxide it was  $43.55 \pm 9.48$ % of iodine recovery (**Table 1**). The experiments realized with 6 mL of H<sub>2</sub>O<sub>2</sub> 3% showed better recovery than those using larger and lower amount, however, seeing the standard deviations the iodine recovery was quite similar in all the assays, indicating that adding more hydroxide peroxide did not provide a higher recovery of iodine.



**Figure 5: Iodine recovery in different H<sub>2</sub>O<sub>2</sub> concentration**

### 3.4. Temperature and Process Time

**Figure 6** demonstrates the iodine recovery percentage in assays realized during four hours at 50 °C and 80 °C. It was observed that at 80 °C and 50 °C the iodine amount recovered was, respectively,  $51.94 \pm 1.28\%$  and  $53.89 \pm 6.23\%$ . These values were similar to each other indicating low influence of temperature in the iodine recovery; however, the temperature decrease demonstrated advantage on the final recovery volume since there was wet lower drag to trap solution at 50 °C. The average of iodine recovery volume in the assays performed at 50 °C was  $7.7 \pm 1.8$  mL whereas at 80 °C was  $16.8 \pm 0.6$  mL.



**Figure 6: Iodine recovery in different temperatures**

All experiments were realized in the period of four hours picking sample up each hour. It was noticed that the best times were between 1 and 2 hours. In the assays realized at 80 °C, the iodine recovery after 1 and 2 hours was, respectively,  $29.35 \pm 4.76\%$  and  $46.23 \pm 2.44\%$ .

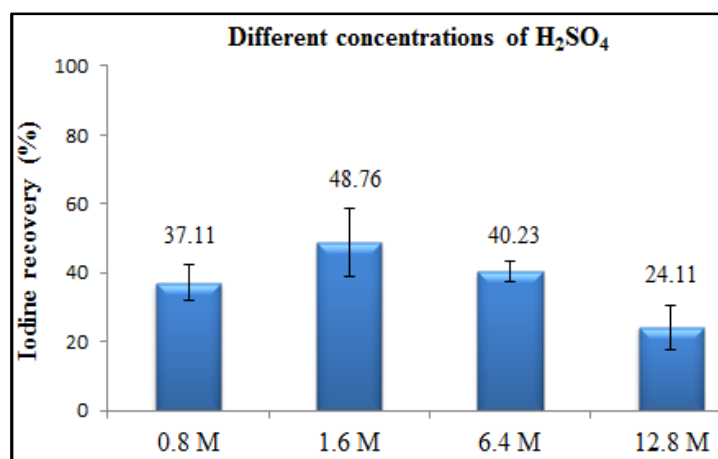


ight increase of recovery  $52.54 \pm 2.03\%$ . At the end of  
 The same behavior was observed in the assays realized

at  $50\text{ }^{\circ}\text{C}$ ; the two first times presented the most significant increase of iodine recovery. The values of recovery at 1 and 2 hours of the experiment were, respectively,  $48.36 \pm 3.02\%$  and  $51.60 \pm 3.76\%$ . After three and four hours of experiments, the iodine recovery remained practically constant generating values of, respectively,  $52.80 \pm 4.93\%$  and  $53.89 \pm 6.23\%$ .

### 3.5. $\text{H}_2\text{SO}_4$ Concentration

**Figure 7** illustrates the iodine recovery percentages in different concentrations of  $\text{H}_2\text{SO}_4$ , the obtained values in 0.8, 1.6, 6.4 and  $12.8\text{ mol L}^{-1}$  were  $37.11 \pm 5.34\%$ ,  $48.76 \pm 10.04\%$ ,  $40.23 \pm 2.88\%$  and  $24.11 \pm 6.62\%$ , respectively (**Table 1**). The highest concentration of acid promoted the lowest value of iodine recovery being, therefore, unnecessary to use high concentrations of acid. The range of concentration from 0.8 to  $6.4\text{ mol L}^{-1}$  presented similar results of recovery, so  $1.6\text{ mol L}^{-1}$  can be suitable concentration for iodine recovery.



**Figure 7: Iodine recovery in different concentrations of  $\text{H}_2\text{SO}_4$**

## 4. CONCLUSIONS

Several parameters were studied in the  $^{131}\text{I}$  recovery process, as a byproduct of the  $^{99}\text{Mo}$  production process by fission. Molybdenum oxide was evaluated as a catalyst for iodine in solution; however, the results did not demonstrate an advantage in relation to its absence. It was observed that in the presence of the catalyst the iodine recovery was lower than its absence. Additionally, the recovery of iodide was delayed in the initial periods of the process in the presence of the catalyst, leading to increase the time process. Nitrate and sulfate were studied as iodide anion eluents and the results demonstrated a lower recovery of  $^{131}\text{I}$  using nitrate salt. Hydrogen peroxide used as the oxidant of the iodide indicated that the iodine recovery was similar in all assays demonstrating that adding more hydrogen peroxide did not provide a higher recovery of iodine. The reduction of the distillation temperature showed advantageous since the recovery yields were similar beside a lower wet drag to trap solution. Additionally, it was evaluated the distillation time verifying that the best times for iodine



Shorter periods of distillation avoid dilution of the trap ss. Finally, acid concentration tests indicating that best results were found at moderate concentration. This study can contribute to provide set up conditions to  $^{131}\text{I}$  recovery system reaching around 50% yield.

## ACKNOWLEDGMENTS

The authors thank the Center of Radiopharmacy IPEN-CNEN/SP, and CNPq (National Council for Scientific and Technological Development).

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