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A route for polonium 210 production from alpha irradiated bismuth-209 target

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Summary

A method is proposed for production and purification of polonium-210 *via* the ^{209}Bi (α, 3n) ^{210}At nuclear reaction. Bombardment of a bismuth-209 target was performed with a 37 MeV alpha beam that conducts to the production of astatine-210 (8.1 hrs), which decays to polonium-210. It is further purified from the bismuth target matrix by employing liquid–liquid extraction method using tributyl phosphate (TBP) in *para*-xylene from 7 M hydrochloric acid. Back extraction of polonium-210 was performed by 9 M nitric acid. This method allows to purify a tracer amount of Po-210 (2.6 10^{-13} mol) from macroscopic amount of Bi (2.8 10^{-2} mol).

Keywords:

Polonium-210; bismuth; α irradiation; liquid-liquid extraction.

Introduction

Although Pierre and Marie Curie made the discovery of the element polonium (Po) more than a century ago, physical and chemical properties of this element and its complexes are still barely known. This can be explained by two main reasons: first, polonium has forty-two known isotopes with a mass ranging from 188 to 220, all radioactive [1]. Second, one of its isotopes, polonium-210 (Po-210) that occurs naturally in the uranium-238 decay is very rare in nature which impedes its studies. The equilibrium ratio of U to Po is 1.19 10^{10}, so that the Po concentration in uranium ores is less than 0.1 mg/ton [2]. Po-210 is a high-energy alpha emitter (E= 5.305 MeV) [3] with a radioactive half life of 138.376 days. It is known for being one of the most toxic radionuclides due to its high specific activity (1.66 10^{14} Bq/g) [4] It presents an internal radiation hazard due to its short range of alpha particles in biological tissues (40-50 μm) [5]. With the advent of cyclotrons and nuclear reactors and their intense fluxes, production of milligrams of Po-210 has become feasible. Bombardment of stable bismuth-209 (Bi-209) with intense neutron fluxes has been studied in nuclear reactors [6]. Bi-210 in its ground state Bi-210g (5.012 days) is produced by the Bi-209(n,γ)Bi-210 reactions and decays to ^{210}Po *via* β^- decay. The production cross-section of Bi-210 being about 15 mb, a Po-210 activity close to 4.10^3 GBq/kg of Bi-209 is obtained after 200 days of irradiation in a thermal neutron flux of 1.10^{14} n/cm^2.s [7]. Po-210 can also be produced in cyclotrons *via* the direct reaction Bi-209(α, t)Po-210 [8]. Another possibility is to produce polonium from the decay of At-210 obtained *via* the reaction Bi-209(α,3n)At-210[9]. This reaction is used in the present work as an easy way to produce Po-210 for radiochemistry studies. For this application, the production of Po-210 with good radionuclide and chemical purities is required.

The separation of polonium includes several steps: (i) dissolution of the irradiated target, (ii) Bi-Po separation and (iii) reconditioning.

The bismuth target will not dissolve easily in hydrochloric acid solution alone. In other hand, bismuth could be dissolve in nitric acid and no special problems are presented. A mixture of hydrochloric acid and nitric acid in a proportion of 1:4 and 3:4 will dissolve bismuth metal [10].

Many methods can be used for the separation of Po from Bi, such as electrodeposition,[11] solvent extraction and ion separation[12, 13]. For simplicity of setting up, liquid-liquid extraction has been chosen. Several extracting agents [14-33] were used for the extraction of Po from different aqueous layers (HCl, HBr, HNO_3, H_2SO_4, $HClO_4$, H_3PO_4 or lactic acid) into various organic layer (carbon tetrachloride, chloroform, xylene, toluene or cyclohexane) (Table.1). For all of the previously mentioned methods, the extraction yield of polonium is significant. However, some of the papers mention a poor extraction of Po/ Bi separation [17] while others indicate a good separation but without quantitative data. In addition, most of the studies report the separation polonium-210 tracer from bismuth-210 tracer [27].

As it is mentioned before, a prerequisite in the approach is to dissolve Bi target before Po extraction. Therefore, an extracting agent efficient in strong acidic conditions (HCl and HNO_3) is required. Among the different molecules proposed, TBP has been chosen in this work. The challenge of this work was to separate radiotracer quantity of polonium-210 from macroscopic quantities of bismuth using TBP. In order to optimize this purification method, several parameters were studied (solvent nature, acidity, aqueous medium, extraction time, TBP dilution, organic solvent) for defining the best conditions for polonium separation.

Materials and method

Chemicals

All solutions were prepared using Milli-Q water and all experiments were conducted at room temperature (22±3˚C). All the chemicals used were of analytical reagent grade.

Bismuth foils (chemical purity 99.999% content of Pb, Cu and Ag below 3, 2 and 2 ppm respectively, 25 x 25 mm^2, thickness of 1 mm), kapton Polyimide Film (75 µm , 25 x 25 mm^2), aluminium foil (chemical purity 99.999%, 25 x 25 mm^2, thickness of 1 mm), silver foil (chemical purity 99.999%, 25 x 25 mm^2, thickness of 0.25 mm) were purchased from Goodfellow and used as received. Po-210 (2 M HCl) and Bi-207 (1 M HCl) tracers were purchased from Eckert Ziegler and CERCA LEA, AREVA, France, respectively. Both are used after reconditioning and diluting into the appropriate medium.

Irradiation

The targets are bismuth foil (Bi-209) placed behind an aluminium foil in a capsule. The ARRONAX cyclotron is designed to deliver a fixed 67.4 MeV alpha beam. In order to decrease the energy of the alpha beam, a degrader system was used. This system is built up and placed along the alpha beam line (Figure 1).The target was placed in air 13 cm away from the end of the beam line which is closed by a kapton foil. The alpha beam used for irradiation is extracted at 67.4 MeV. Typical irradiation stands one hour with an average beam current of 0.2 µA. According to literature cross section data of α-particle induced reactions on Bi-209, an energy of 37 MeV was selected for optimal production of At-210 (Figure 2).[8, 34-35] An aluminum foil of 0.9 mm thick is used to degrade the incident energy .The target and its degrader foil are presented in Figure 1. The bismuth foil was ensured by an air cooling system during irradiation. After the end of irradiation, the target cell containing the aluminium and bismuth foils was left in a lead container for a week, allowing At-210 to decay into Po-210.

Batch experiments

Tubes of polypropylene were used for liquid/liquid batch experiment, due to the adsorption of polonium on glass [25] which causes difficulty in the accurate determination of polonium activity. Before polonium addition, the organic layer was pre-equilibrated with the aqueous solution, i.e. the composition does not change in presence of the organic layer.

2 mL of organic and aqueous solution were brought in contact. After equilibrium of the biphasic system, an aliquot of polonium (300 Bq) and/or bismuth (100 Bq) was added and the tubes were shaken for 90 min. This time proved to be sufficient to achieve distribution equilibrium of polonium/ bismuth between the phases.

After phase separation, an aliquot of the aqueous and organic phases were withdrawn to derive the percentage of extraction, %E, corresponding to:

$$\%E = \left(\frac{A_{org}}{A_{org} + A_{aq}}\right)\left(\frac{V_{aq}}{V_{org}}\right) \times 100 \qquad \text{eq.(1)}$$

where, A_{org} and A_{aq} define the polonium activities measured in the organic and aqueous phases, respectively. V_{org} and V_{aq} define the volume of organic and aqueous phases, respectively.

For the back-extraction, %BE is defined as:

$$\%BE = \left(\frac{A_{aq}}{A_{org} + A_{aq}}\right)\left(\frac{V_{org}}{V_{aq}}\right) \times 100 \qquad \text{eq.(2)}$$

Analytical Tools

The radionuclide composition was checked by gamma spectrometry. A gamma spectrometer from ORTEC, using a high purity germanium detector with 1.82 keV resolution at 1.33MeV, was used to quantify the production of gamma rays-emitting radionuclides, and more particularly At-210 and At-211, after Bi-209 irradiation [8, 36].

After the dissolution of the irradiated bismuth foil in 7 M HCl solution, an alpha spectrometer was used for the determination of polonium-210 activity. Sample for alpha spectrometry was prepared by spontaneous deposition of Po-210 on silver discs [37-39] in a water bath (80˚C) for two hours. The detector consists of a 24 mm ruggedized surface barrier detector connected to a 7401 VR Canberra spectrometer. Spectra are obtained by a TRUMP 8 K multichannel buffer card on PC. Efficiency of the detector is determined by a multi-alpha surface source with geometry identical to the Po-silver sample disks. (Figure 3)

For batch experiments, activity of the radionuclides (Po-210 or Bi-207) was determined by liquid scintillation counting using a Packard 2550 TR/AB Liquid Scintillation analyzer. The samples were prepared by mixing an aliquot of 0.2 mL of the solution to measure with 2 mL of 1 M HCl and 2.2 mL of ultimate gold AB scintillation cocktail. The measuring time was fixed at 1 hour. The quenching arising from the organic solvent was taken into account according to the following relation:

$$A = A_m\,(-9.10^{-5}\times Tsie^3 + 673\ 10^{-4}\times Tsie^2 - 17.26\times Tsie + 1565.8) \qquad \text{eq.(3)}$$

A_m being the activity measured by liquid scintillation and *Tsie*, an independent parameter from polonium analyses defined by the apparatus to determine the quenching parameter. Determination of stable bismuth-209 concentration was performed by Inductively Coupled Plasma Atomic Emission Spectroscopy (ICP-OES, Thermo Scientific iCAP 6500) with a plasma power of 1150 W. These analyses were done with a rate of 0.50 L/min auxiliary gas, 12.00 L/min gas cooling and 0.45 L/min for gas nebulizer. The wavelength (λ) used for bismuth analysis was 223.061 – 306.770 nm. Mass spectrometer - Electrospray (MS-ESI) was used to analyze the hydrolysis of tributyl phosphate in 8 M nitric acid solution. The analysis of phosphate ions in the final purified polonium solution was done by Ion-exchange chromatography ICS2500 with a conductivity detector. These analyses were done using AS18 (4 x 250 mm) column type with a 50 µL injection loop, 1 mL/ min flow and 120 mA current suppression.

Results and discussion

Production of Po-210

Using SRIM-2008 software [40], calculation shows that the bombardment of Bi-209 foil (1 mm) by an alpha particle beam with energy of 37.02 MeV is achieved by using a 0.9 mm aluminium foil. The energy of the delivered alpha beam arrived on the aluminium foil with a value of 64.5 MeV and an average current being of 0.2 µA for one hour. The stopping point inside the bismuth target was measured to be 0.314 mm at 37.02 MeV.

The produced activity can be determined numerically using the following formula:

$$A = \Phi_0 \frac{\rho}{A} N_a (1 - \exp(-\lambda t)) \int_{E_{\min}}^{E_{\max}} \frac{\sigma(E)}{\frac{dE}{dx}} dE \qquad \text{eq.(4)}$$

Where, ϕ_0, ρ, A, N_a, λ and σ(E) are the incident particle number (particle/s/cm^2), target density, target number of mass, Avogadro number, radioactive constant (s^{-1}) and production cross section, respectively.

$\frac{dE}{dx}$ is the energy loss calculated using the Bethe formulae:

$$-\frac{dE}{dx} = \frac{K.Z_p^2.Z.N_A}{\beta^2.A}\left[\ln(\frac{2.m_e.\gamma^2.\beta^2}{I}) - \beta^2\right] \qquad \text{eq.(5)}$$

K, m_e, Z_p, Z, I, γ and β correspond to a constant (equal to 0.307 MeV.cm^2), electron mass, projectile charge, target charge, ionization potential and relativistic coefficients associated to the projectile, respectively.

The theoretical activity calculated based on the irradiation parameters (0.2 µA, 1 hr) amounts to 6.34 MBq of At-210. The experimental results shows the production of 4.11 MBq of At-210 which corresponds to the production of almost 10 kBq of Po-210.

The analysis of the irradiated Bi-209 target by using gamma spectrometry (Figure 4), shows the production of At-210 and At-211 which decay to Bi-207 ($t_{1/2}$=32.9 y) and Bi-206 ($t_{1/2}$=6.423 d), respectively. No other γ ray emitting radionuclides were found. After bismuth target irradiation, the foil is kept 7 days in a lead container for the total decay of At-210 into Po-210.

Polonium / Bismuth batch experiments

The method for separation of polonium is essentially based on the work of Chen *et al*. [27], *i.e.* the extraction is performed in HCl whereas the back extraction is realized in nitric acid. However several optimization steps were studied in order to find the best conditions for achieving a high extraction yield of polonium-210 and a high selectivity for polonium extraction over bismuth.

These experiments were done with synthetic solutions containing either Po-210 or Bi-207. The influence of the solvent nature containing TBP (10%) was first studied from 6.9 M aqueous hydrochloric acid. Hexane, *para*-xylene, toluene, *ortho*-xylene, and chloroform were studied. Results show no effect of the investigated solvent on the extraction of Po with an approximate extraction percentage of 83.0 ± 6.0 %, except for chloroform for which a negligible extraction value is found. *Para*-xylene was chosen for the further studies.

The extraction of polonium from hydrochloric acid solution is strongly dependent on the acid concentration of the aqueous phase. Investigation of the extraction of Po and Bi as a function of hydrochloric concentration in 10 % TBP / para-xylene is presented in Figure 5. A good polonium extraction value of 89.0 ± 5.0 % was obtained at 0.17 M HCl. However, when the acidity increases, the extraction values decrease until it reaches a minimum value of 5.0 ± 0.3 % at 2 M HCl. After 2 M HCl, the extraction value of Po increases again and reaches its maximum value of 96.0 ± 4.7 % at 7 M HCl. For Bi, the extraction is weak in the acidic range studied with a maximum extraction value of 6.7 ± 0.5% for 0.17 M HCl. In conclusion, a good compromise (high extraction yield, good selectivity) would be to work at 0.1 M or 7 M HCl, with a preference for the latter condition.

Then, the influence of the concentration of tributyl phosphate in para-xylene from 7 M aqueous hydrochloric acid was performed (Figure 6). The extraction of Po is greater than 90 % whatever the concentration of TBP studied with a slight increase, as the TBP concentration increases. For Bi, the extraction is almost equal to zero for a TBP concentration below 1 M. Above 1 M, Bi extraction becomes significant with 25 % of Bi extracted at 3.5 M of TBP. To have a good compromise between extraction yields on selectivity, a concentration of 0.36 M in TBP (or 10 % in weight) was selected for the extraction step.

The following step is to back extract polonium to a new aqueous layer to be used for further studies. Investigation shows nitric acid to be the convenient aqueous layer for polonium back extraction [41]. Back extraction of Po in *para*-xylene is studied as function of HNO_3 concentration (Figure 7). Back extraction of polonium increases as the acidity increases and reaches a maximum value of 96.0 ± 4.0 % for a solution of 9 M HNO_3. This can be explained by the increasing competition between Po and HNO_3 for TBP [42]. The potential degradation of TBP can contribute to explain the important back-extraction. Indeed according to the literature, at high concentrations of nitric acid, TBP can be hydrolyzed like it occurs in reprocessing spent reactor fuels [43, 44]. To confirm the hydrolysis of TBP at high acidic nitric acid solution, the aqueous phase (8 M HNO_3) obtained after extraction was analyzed by mass spectrometer. Results show the presence of phosphoric acid, monobutyl phosphate and dibutyl phosphate which are the hydrolyzed products of tributyl phosphate. Thus, the concentration of 9 M nitric acid will be chosen for the purification process.

Finally, the idea was to test the protocol optimized for the polonium extraction and back-extraction to see the behavior of bismuth using the whole procedure. The final step was to confirm the absence of cold bismuth in the back extracted solution. As the analysis using ICP-

OES gives signals below the detection limit of ICP-OES (<1 ppb), absence of bismuth-209 can be guaranteed.

Target dissolution and Po/ Bi separation

In this part, we want to use the results previously presented in order to obtain the best conditions for the Po/ Bi separation from the irradiated target. The first step is the transfer all the polonium from the target into solution. The depth of penetration determined by SRIM-2008 software corresponded to 0.314 µm. To recover all produced Po-210, a total dissolution of the irradiated foil (thickness of 1 mm) is required. The easiest way was to dissolve directly the target in 0.1 M or 7 M HCl (see previous part). However the solubility of metallic Bi is not important in HCl. It was then decided to follow the work of Bhatki *et al.* [10] and to use 10 M HNO_3. The next step is then the reconditioning of the extraction medium after evaporation. In order to remove the trace quantities of nitric and nitrous acids, two evaporation cycles in 7 M HCl using IR-lamp are done. IR-lamp was used for acidic solution evaporation to minimize the loss of polonium due to its high volatility [45-47]. 8.0 ± 0.5 moles of nitric acid was used per mole of dissolved bismuth; the off gases appear to be nitric oxide and nitrogen dioxide [33].

Solubility tests showed that an HCl concentration lower than 3.5 M was sufficient for a complete dissolution of the residue. An attempt was done to decrease the HCl concentration to 0.1 M, after complete dissolution of the target, with the idea to precipitate bismuth while keeping polonium in solution. However, a strong decrease in polonium concentration was observed indicating a co-precipitation process in agreement. This was already evidenced by Holgye [48]. Therefore, it was decided to use 7 M HCl for the extraction step.

Alpha spectrometry analysis of the dissolved Bi foil shows only one peak at 5.3 MeV, which corresponds to the presence of Po-210 (Figure 3). Gamma spectrometry analysis indicates the presence of Bi-207 and Bi-206, produced from the decay of At-211 and At-210, respectively.

After the extraction of polonium in the presence of 10% TBP in *para*-xylene, the trace quantities of bismuth extracted in the organic phase were removed by washing two times with a new equilibrated layer of 7 M HCl. The affinity of Bi at this acid concentration is largely favorable for the aqueous phase. The extraction yield for Po was 96.0 ± 5.0 %.

The following step was to back extract polonium from the previously extracted organic phase to 9 M HNO_3.The percentage of polonium back extracted was 96.0± 4.0 % and the percentage of the total recovery yield of polonium in this methodology was 85.2 ± 4.5%. Bismuth was confirmed to be under the detection limit.

The final step was to evaporate the nitric acid and to recondition in the appropriate medium such as $HClO_4$ or HCl for the radiochemical studies. The phosphoric acid coming from the degradation of TBP (see previous part) was not observed in the reconditioned solution (Ionic chromatography, DL = 50 ppb). The complete methodology is depicted in Figure 8.

Conclusion

The bombardment of bismuth-209 target with a 37 MeV alpha particle beam conducts to production of astatine-210 (8.1 hrs) which decays to polonium-210. The produced radiotracer of polonium is purified from macroscopic quantities of bismuth by a wet methodology using liquid–liquid extraction. The procedure is presented in Figure 8. Results confirm that polonium can be extracted (96.0 ± 4.7%) to an organic phase containing 10% TBP in *para*-xylene from 7 M HCl while the bismuth nuclides remain in the aqueous phase. Moreover, the back-extraction of polonium from the organic phase can be performed by using a 9 M HNO_3 phase (96.0 ± 4.0%). The optimized process leads to a solution of Po-210 with a global recovery yield of 85.2 ± 4.5% with good radionuclide and chemical purities.

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Table and Figures

Table 1: Literature data of protocols for polonium-210 extraction using liquid-liquid extraction.

EXTRACTANT	Solvent	Aqueous phase	Po extraction (%)	Reference
Cupper-diethyldithiocarbamate	CHCl3	1.06 M HNO3	55	[17]
	CHCl3	0.07-0.72 M HNO3	65 -86	[17]
	CHCl3	0.03 M HNO3	60	[17]
Triuylamine	Xylene	7M HCl	91	[19]
Tributylphosphate 10%	ortho-xylene	7M HCl	85	[27]
Tributylphosphate 10%	Decalin	7-9 M HCl	98	[28]
Tributylphosphate 20%	Dibutyl ether	6M HCl	99	[29]
Dibutyl carbitol		3M HNO3	98	[33]

Figure 1: Schematic diagram for the irradiation setup of bismuth-209 foil by a 37 MeV alpha beam.

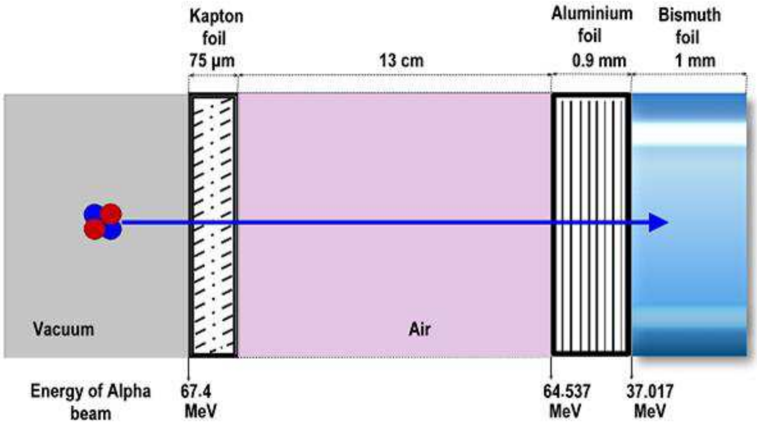

Figure 2: Direct production cross-section for At-211, At-210, and Po-210 by nuclear reaction of Bi-209(α,2n)At-211, Bi-209(α,3n)At-210 and Bi-209(α, t)Po-210, respectively. [8, 32]

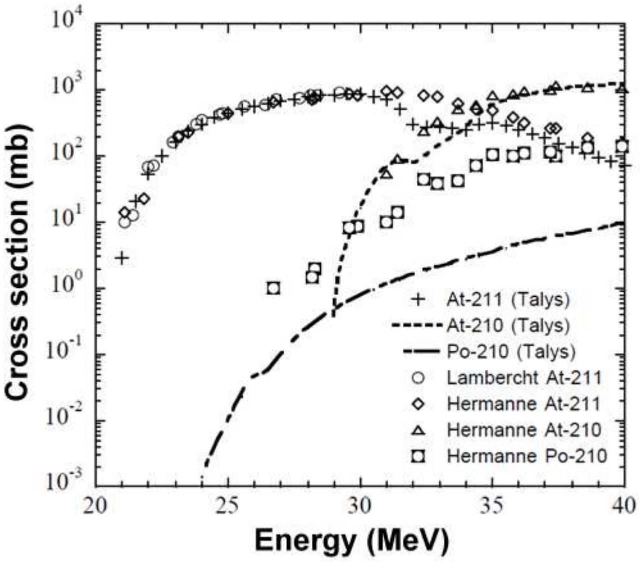

Figure 3: Alpha spectrum of the irradiated bismuth target after dissolution in 2 M HCl and spontaneous deposition on a silver disk.

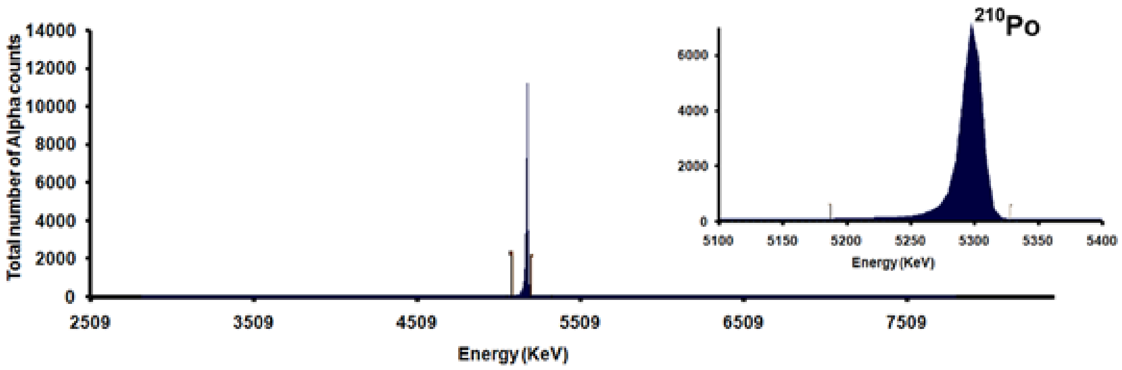

Figure 4: Gamma spectrum obtained for a Bi-209 foil after irradiation with a 37.0 MeV alpha beam with a current of 0.2 μA for 1 hour.

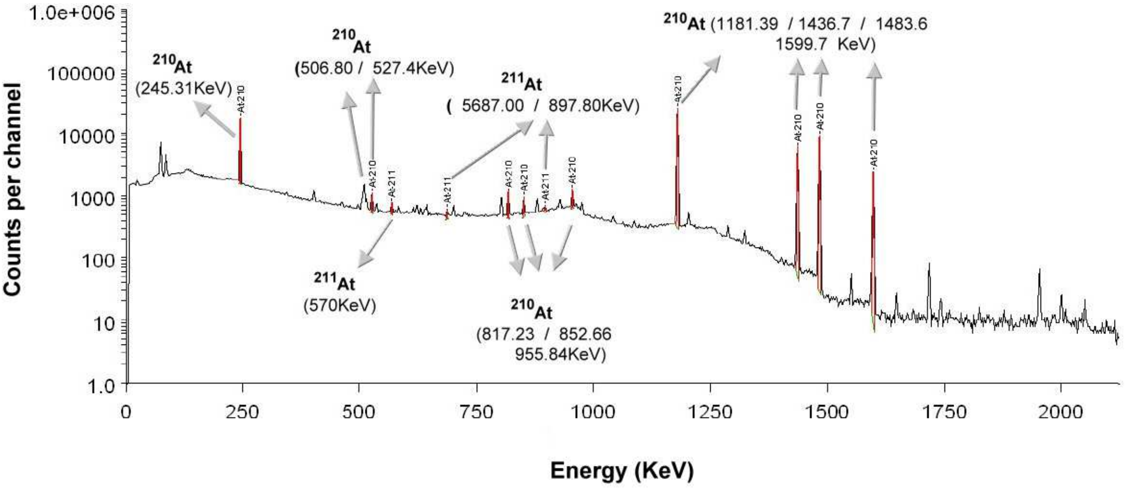

Figure 5: Effect of the nature and acidity of aqueous layer on the extraction of polonium-210 and bismuth-207 in 10% TBP- para-xylene. The batch activity used were 300 Bq and 100 Bq , for Po-210 and Bi, respectively.

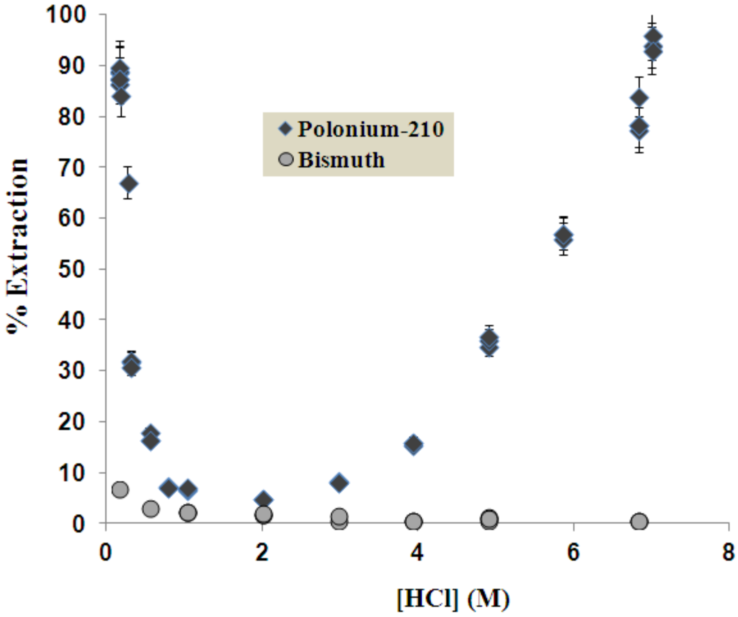

Figure 6: Effect of the concentration of tributyl phosphate on the extraction of polonium-210 and bismuth from aqueous layer of 7M HCl. The batch activity used were 300 Bq and 96.1 Bq , for Po-210 and Bi-207, respectively.

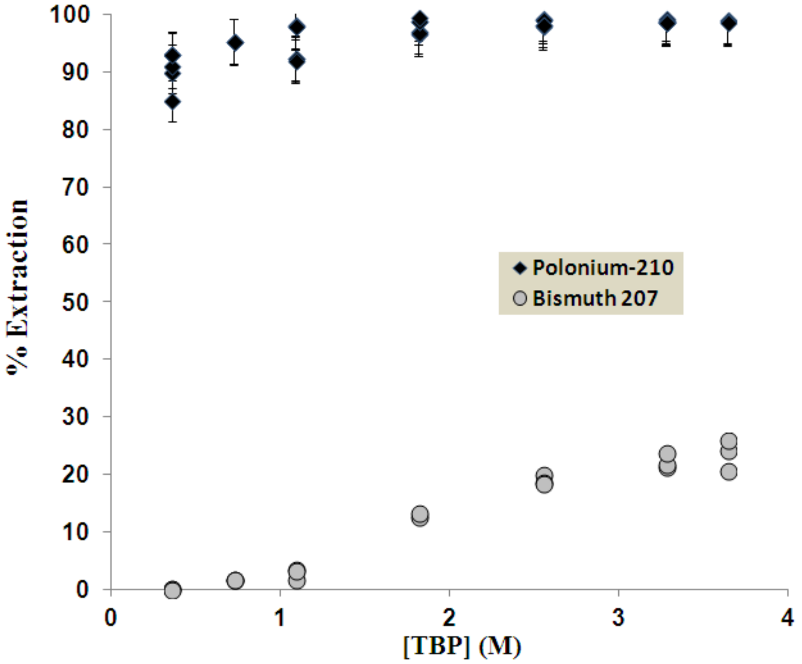

Figure 7: Effect of the concentration of nitric acid on the back-extraction of polonium-210 from organic layer (10%TBP-paraxylene). The batch polonium activity used was 288 Bq.

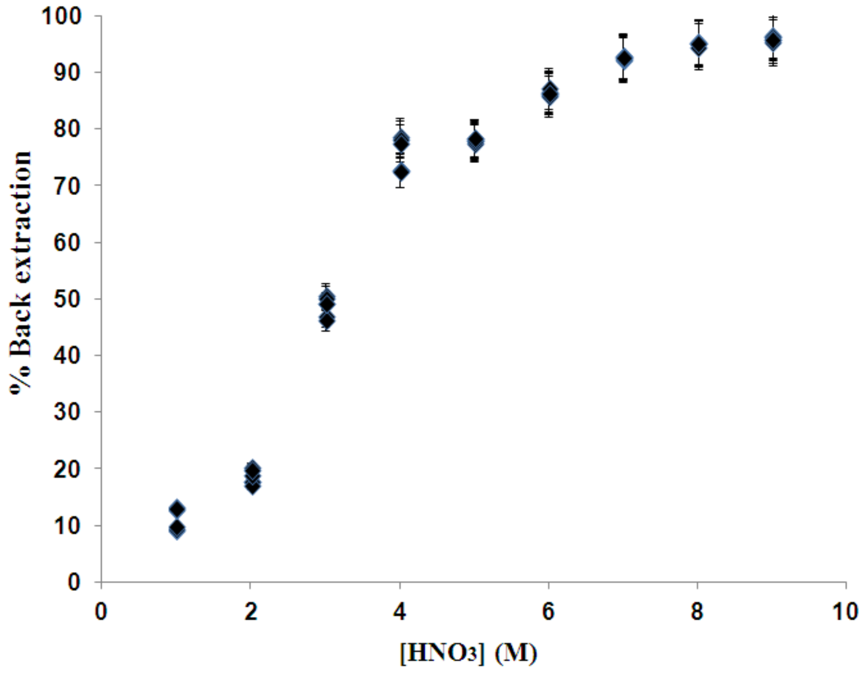

Figure 8 Methodology followed to recover Po-210 (2.6 10^{-13} mol) from the irradiated bismuth target (2.8 10^{-2} mol).