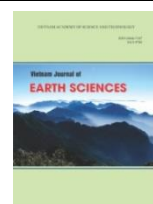




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Improvements of ^{210}Po determination method in thermal water samples

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ABSTRACT

Determination of naturally radionuclides have been known well as an important topic in environmental study in recently. One of the most toxic radioisotope in nature, a daughter product of ^{238}U decay chain is ^{210}Po (polonium). The improvement and optimizations methods for determination of this attractive isotope are still presenting so far. In this paper, a new improved method was elaborated for ^{210}Po determination in thermal water sample. In the proposed method, analytical optimization of spontaneous/auto deposition does not use Teflon cup, magnetic stirring or any preparing equipment/item only normal glass and a side of square silver. In addition, the optimization was neglected with absent of purification of polonium (Liquid-liquid extraction methods/Ion exchange chromatography/Extraction chromatographic separations). The outcome of optimal procedure were simplify, less time consuming, great reduction of costs with chemical recovery >80% and could apply for any liquid environmental samples.

Keywords: Auto-deposition; ^{210}Po in thermal water; NORM; Polonium; ^{210}Po determination method.

1. Introduction

Polonium (^{210}Po) is a naturally occurring radioactive isotope discovered by Maria Skłodowska - Curie (1898). There were 43 isotopes of polonium, but only three isotopes were found to have half-lives more significant than one day (^{208}Po (2.98 years), ^{209}Po (125.2 years), and ^{210}Po). ^{210}Po is one natural isotope of polonium and identified as having the largest half-life (183.4 days). The ^{210}Po

originates from the natural decay chain ^{238}U . It is the progeny of ^{222}Rn , a noble gas radioactive isotope which highly mobile and easy to release and transfer in the atmosphere, plants, aquifer, fish, and creatures of the environment (Lindsey et al., 1996; Inácio et al., 2017; Keramati et al., 2018; Ramola et al., 2008). ^{210}Po in the atmosphere is derived from soil, surrounding geological formations, volcanoes, dust, forest fires, and fossil fuels (Matthews., 2007; Figgins., 1961; Persson., 1970; Poet., 1972; Jia., 2001; Baskaran.,

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2011). ^{210}Po in the air fall onto ground surface and transfer to plants (Pietrzak-Flis et al., 1995; Duong et al., 2021; Marciulioniene et al., 2015; Vandenhove et al., 2009). ^{210}Po exists in water absorbed by suspended particles and enters fish and creatures (Çatal et al., 2012; Mishra et al., 2009; Van-Hao et al., 2020; Carvalho, F. P et al., 1994 and 2018). In aquifers, ^{210}Po is derived from aquifers formations, sedimentary and parent isotopes (Balistrieri et al. 1995; Carvalho et al., 2017; Seiler et al., 2011; Zhong et al., 2020; Thakur et al., 2020; Chaudhuri et al., 2010; Szabo et al., 2012; Roba et al., 2012). ^{210}Po can enter the human body (IAEA et al., 2009; Martin et al., 2004; Momoshima et al., 2002; Al-Masri et al., 2004; Dubey et al., 2015; Chen, J. et al., 2016). Therefore, the ^{210}Po was interested and studied by many scientists around the world (Santos et al., 1990; Clayton et al., 1995; Shaheed et al., 1997; Stepnowski et al., 2000; Jia et al., 2000; García-Orellana et al., 2002; Vesterbacka et al., 2005; Narayana et al., 2010; Prabhath et al., 2015; Sethy et al., 2015; Ababneh et al., 2018; Van et al., 2020; Zhong et al., 2020; Behbehani et al., 2020). Due to the complex chemistry properties (Ram et al., 2019; Thakur et al., 2020), there were different methods to determine of ^{210}Po in environmental samples: ^{210}Po measurement techniques have been studied, described, and applied by many other scientists (Jia et al., 2001, 2003; Biggin et al., 2002; Meli MA et al., 2013; Kavitha E et al., 2017; Kim G. et al., 2005; Harada K et al., 1989; Skwarzec B et al., 2009; Skwarzec B et al., 2001; Skwarzec B et al., 2003; Boryło A et al., 2013; Turtiainen T et al., 2011; Guérin N et al., 2015; Vajda N et al., 1997; Matthews et al., 2007). However, the main disadvantages of these methods needed a highly complex separation process, time-consuming, and relatively high cost. In recent years, the improved and optimized determination

methods of ^{210}Po were still studied (Uesugi et al., 2010; Rigaud et al., 2013; Dubey et al., 2015; Szarlowicz et al., 2019; Prusińska et al., 2021). Uesugi improved the ^{210}Po determination method for the high deposition yield. The chemical recovery was enhanced to 75% (Uesugi et al., 2010); Rigaud improved the procedure by use of ion-exchange resin for ^{210}Po removal after the initial plating step, the outcome of method determine ^{210}Po within a 6% (relative uncertainty) (Rigaud et al., 2013); Dubey improved deposition step and the new deposition the recovery was 78%–94% (Dubey et al., 2015); Szarlowicz minimized with 0.1–0.2 g in weight without using hydrofluoric acid for digestion of sediment sample and minimized amounts of reagents with a yield of radiochemical procedure >90% in total (Szarlowicz et al., 2019); Prusińska improved of silica digestion or optimized of electrode material with chemical efficiency >85% (Prusińska et al., 2021).

Vietnam has abundant thermal water with above 400 natural sources and thermal water boreholes. Thermal water sources have well-known uses for spas, balneotherapy, medical treatment, health-improving effects on the human organism, the development of tourism, and fulfilling the mineral water demand in Vietnam and around the world. In thermal water, the presence of ^{210}Po is believed to originate from the decay of parent isotopes inside the aquifer through solid-liquid interactions (Nguyen Dinh et al., 2021; Harada et al., 1989; Seiler et al., 2011; Carvalho et al., 2017). ^{210}Po was determined to be firmly bound to the aquifer surface (Balistrieri et al., 1995; Seiler et al., 2011). The ^{210}Po are usually readily removed from groundwater by co-precipitation with Fe hydroxides, Mn oxides, colloids, and sulfides when adsorbed to the solid surface in aquifer systems (Seiler et al., 2011; Zhong et al., 2020; Thakur et al., 2020). Therefore, the

presence of ^{210}Po in groundwater is mainly detected at low concentrations and less than 40 mBq/L (Harada et al., 1989; Hess et al., 1985; Bontto et al., 2009). For low activity environmental samples, a difficult determination needs to be deal with the MDA of the method.

During the reaching of underground thermal water flow to the near ground surface, the potential for anions and cations led to high total mineralization (Nguyen Dinh et al., 2021). For the methods of ^{210}Po determination, a chemical separation process using a Dowex 100-200 mesh anion column was used. High mineralization content in the sample will lead to contamination of Dowex columns and must be replaced after each user who needs time and high cost consuming.

The determination of low ^{210}Po activity concentrations in various environmental samples was based on a primary procedure and required sensitive and reliable methods (Jia et al., 2007; Dubey et al., 2015). The measurement techniques were described by Fleer and Bacon (1984). Matthews (2007), IAEA (2009), and Thakur (2020) re-introduced and reviewed procedures for the determination of ^{210}Po in various water and environmental samples (Matthews et al., 2007; IAEA., 2009; Thakur et al., 2020). After chemical dissolution and separation, ^{210}Po can be deposited directly on silver disc from dilute HNO_3/HCl solution as an auto-deposition process. To prepare alpha samples required preparation of flat and homogeneous disk. This process was usually carried out using a Teflon beaker shaped like a vortex bottom thread cylinder or electrode with the particular way so that ^{210}Po was absorbed only a side of the silver disc (IAEA., 2009; Dubey et al., 2015). This step is quite complicated and time-consuming; avoid ^{210}Po was absorbed by both sides of the disk to outcome high recovery (Dubey et al., 2015). In this study, we elaborated a new method to improve and optimize the technical depositing

of ^{210}Po on the side of silver discs by using a Leo Tape adhesive tape. This technique showed a convenient and significant reduction of costs and time-consuming. In addition, the chemical recovery was guaranteed and ensured the chemical separation of ^{210}Po with high recovery >80%. The application of this method will be expressed for different environments.

2. Thermal water samples

The thermal water samples were collected from three mines at Kim Boi, Hoa Binh, Tien Lang, Hai Phong and Kenh Ga, Ninh Binh provinces which were reported low activity and high TSD in our unpublished data. The collected water samples were hosted in various limestone formations (Van-Hao et al., 2020). Each water sample was taken with a 20 liters in plastic can and acidified to pH ~ 2 to prevent polonium from being hydrolyzed and absorbed to the surface of the plastic can wall (Thakur et al., 2020). Because of the low ^{210}Po activity in study sample, the using volume was performed with at less 3 liters of each thermal water.

3. Experimental

3.1. Conventional method

3.1.1. Separation and purification of polonium

* Ion exchange chromatography

An ion-exchange chromatography method is a common laboratory analytical technique to separate and isolate substances in a composite (Fig. 1). The polonium separation technique is performed with solution samples such as HCl (0.05–12 M) or HNO_3 (0.8–5 M). The strongly retained in anion exchange resins were used Dowex - exchange resins. 1, Dowex-2, IR-120, AG 50 W \times 8 and AG MP50 and Bio-Rad AG1- \times 4 (Figgins P, 1961; Kmak KN et al., 2017; Strelow F, 1988). This procedure is usually time-consuming (prolonged flow rate). Some situations could take up to a few days, which did not apply to many applications.

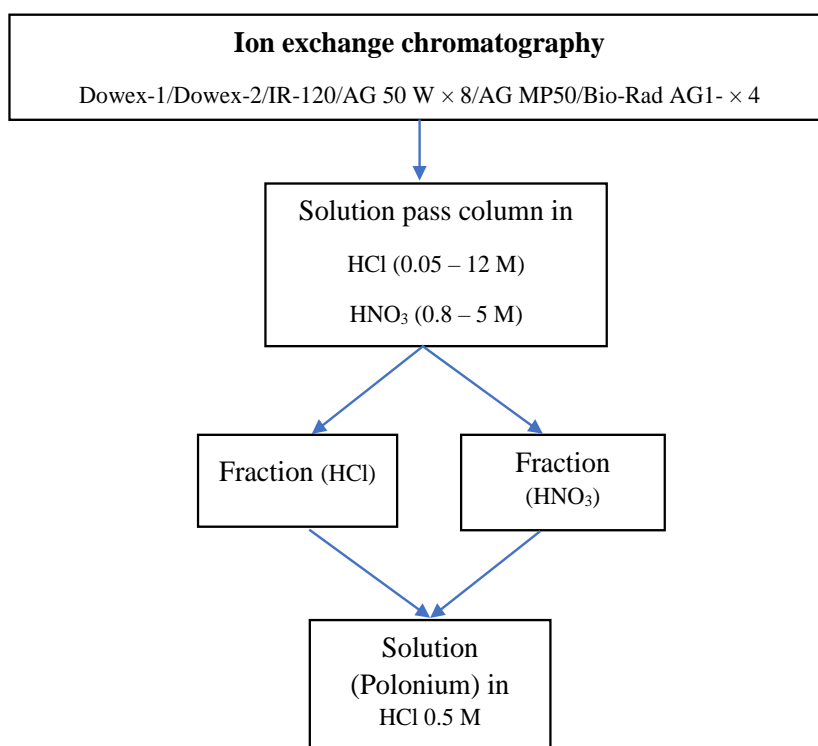


Figure 1. Flow diagram of ^{210}Po ion exchange chromatography (conventional method)

** Liquid-liquid extraction methods*

Liquid-liquid extraction methods were used for separating Po from Pb and Bi. This technique uses several types of organic extracting such as isopropyl ether, diisopropyl ketone, methyl isobutyl ketone, tributyl phosphate, methyl isobutyl ketone, diethylammonium diethyl dithiocarbamate, diethylammonium diethyldithiocarbamate, thenyltrifluoroacetone, tri-n-octylphosphine oxide (Jia., 2004; Jia., 2008; Kim., 2009; Lee., 2010). The limitation of this method is that the process is complicated and requires many extraction steps and during the separation process, many mixed radioactive wastes were generated (Thakur et al., 2020).

** Extraction chromatographic separations*

The extraction chromatography (EC) method used resins to retain polonium, which was reported with high selectivity and faster polonium exchange kinetics than anion exchange. A few EC methods used different

resins such as di-t-butyl cyclohexane-18-crown-6 in 1-octanol/ iso-decanol, N'-tetraethyl diglycolamide, Amberchrom CG-71 (Horwitz., 1992, 1994; Vajda., 1997; IAEA., 2006). However, this method is a high cost of conducting with environmental samples.

3.1.2. Source preparation

The source preparation step by auto deposition was the most common way for ^{210}Po determination by alpha-particle spectrometry (Fig. 2). The IAEA (2009) reported that polonium separating efficiency from other alpha-emitting radionuclides and matrix elements which may not have been completely isolated during the chemical separation step (IAEA., 2009). The step required flat and homogeneous alpha sources (silver disk) where polonium was spontaneously absorbed in dilute acid solution. The equipment primarily for this process is a constructed Teflon cup and

magnetic stirring so that ^{210}Po is deposited on one side of the silver disc. The other ways were electro-deposition and Micro-precipitation methods. Polonium auto-deposition was usually remained at a temperature range from 70 to 100°C during 2–5 hours (IAEA., 2009; Thakur et al., 2020). The long time consuming has also been reported by (Oliveira., 2006; Carvalho., 2010; Carvalho., 2011; Bagnall., 1990). During this step, usually add a small amount of ascorbic acid, hydroxylamine hydrochloride, and sodium citrate to limit the influence of Fe^{3+} , Ca^{2+} , Mg^{2+} ions; to effect of polonium deposition and to get high spectrum resolution (Figgins., 1961; Smith., 1984; IAEA., 2009). There were affecting factors of the efficiency,

including pH, temperature, sample volume, and time (Guérin., 2015; Lee., 2010; Porntepkasemsan., 2011; Bagnall., 1990; Thakur. et al., 2020). The other factor such as the disc material was also one of the issues that control the performance of the process. Although silver discs were considered to be the most commonly used (IAEA., 2009), however, polonium automatic deposition can also occur on low-cost copper, stainless steel, or nickel discs but they took lower chemical recovery under the same conditions (Skwarzec., 2001; Thakur et al., 2020). The polonium chemical recovery efficiency had been reviewed with a range from 70 to 98% for environmental samples (Thakur et al., 2020).

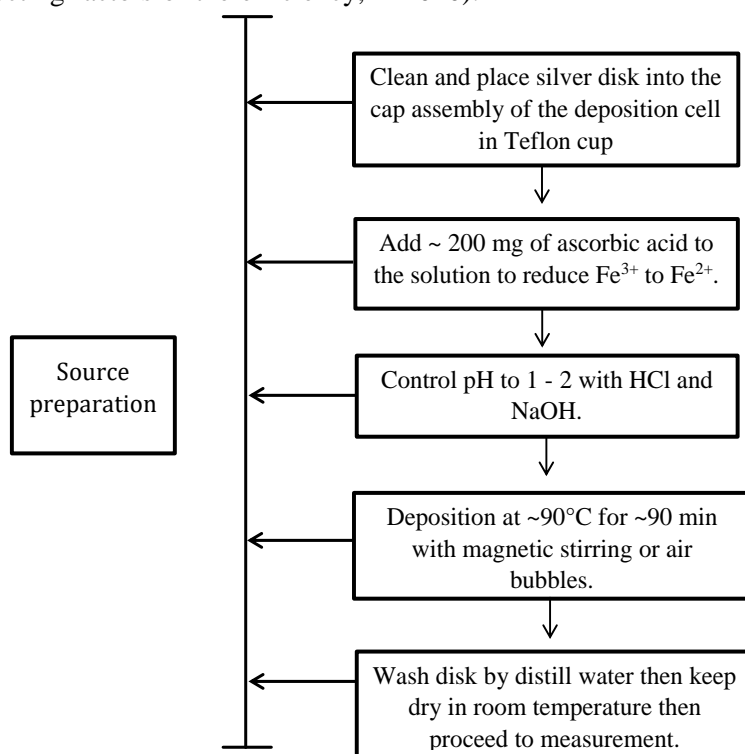


Figure 2. Flow diagram of ^{210}Po Source preparation (conventional method)

For the electro-deposition and micro-precipitation methods, the electro-deposition process directly depletes ^{210}Po from an acidic solution on various electrodes, including carbon, Cu, Ag, Ti, Pd, and Ni

(Figgins et al., 1961). The results recorded the highest efficiency at the Ni electrode. Micro-precipitation methods are effective for processing large batches of samples with less time than auto-absorption methods. However,

the limitation of that method is the lower recovery efficiency, require high technical, and need more items (Thakur et al., 2020). In this study, we propose a new procedure in ^{210}Po deposition, which proves to be effective in a simplified way, less time-consuming, significant reduction of costs, but the efficiency of process performance is still guaranteed.

3.2. Improved method

In the above section, the improving information was introduced in brief. When the ^{210}Po activity levels in the environmental sample are low but in high TDS, the issues will be the enhancing method with guarantee chemical recovery efficiency, less consuming time with the step of separation and purification of polonium and reduced cost. In this paper, we report the re-elaborating process for two main steps: separation and purification of polonium and source preparation. For the separation and purification of polonium, the single side of the silver disk and standard glass cup was used, which will reduce contamination deposits and cost when compared with the other ways. Teflon cup was used a silver disk in the bottom and magnetic stirring. The conventional procedure, automatic deposition on the silver disk, was performed in a cylindrical Teflon beaker. The Teflon structure must ensure that the ^{210}Po was only absorbed on one side of the silver disc. This type of cup was prone to degradation due to high temperature for a long time, leading to the absorption solution flow down the back of the silver disc as a limited procedure. The source preparation step was simplified without using a constructed Teflon cup, magnetic stirring, or electrodeposition. The method used only standard glass cups with a similar chemical procedure and materials but optimization without using other items and steps. The silver disk is cut with easy shape as square 1cm of size in the unit and 0.2-0.4mm in thickness. The disk with a side for

spontaneous deposit of polonium was prepared by Leo tape to cover one side of the disk. The effect of this step was tested for different temperatures from 50 to 125°C, time lapsing from 2-10 hours, and different pH (from 2-5) of the solution also. In addition, the outcome guarantee for this step was presented by the experiment result in the next section. This step showed that the effect was convenient, cheaper, and time-consuming, but the chemical recovery was still guaranteed.

3.3. Radioactivity measurement and calculation

3.3.1. Instrumentation

The ORTEC Alpha-Ensemble-4 spectrometer with Alpha-vision software and ALPHA-DUO-M1 - 450 mm² area detectors was used. The alpha spectrophotometer system measures alpha particles in the energy range from 0 to 10 MeV, energy resolution ≤ 20 keV (FWHM) with a detector-to-source spacing equal to the detector diameter, display resolution 3 nA, detector efficiency 22% is achievable with closest detector-to-source spacing (1 cm square silver disk), background ≤ 1 count/hour based with above 3 MeV. The Alpha Vision software was applied for the analysis alpha spectrometer. The detection limit of the equipment was 0.5 mBq and followed the bellow equation.

3.3.2. Activity calculation

The ^{210}Po activity in the thermal water samples is calculated using Formula 1:

$$C_{210} = (A_{209} \cdot \frac{I_{210}^{sp} - I_{210}^{bg}}{I_{209}^{st} - I_{209}^{bg}}) / V \quad (1)$$

C_{210} - the ^{210}Po activity in thermal water sample [mBq/L];

A_{209} - tracer ^{209}Po activity added to the sample [mBq];

I_{210}^{sp} , I_{210}^{bg} , and I_{209}^{sp} , I_{209}^{bg} are the count rates (counts/min) in the ^{210}Po and ^{209}Po peaks for the studied and background samples respectively;

V - Volume of the thermal water sample [L].

$$MDA\left(\frac{Bq}{kg}\right) = \frac{4.66\sqrt{B}}{(T \times \% \alpha \times \% \eta \times W)} \quad (2)$$

Where 4.66 is factor, B is background counts, T is counting time (s), $\% \alpha$ is alpha mission probability, $\% \eta$ is absolute efficiency,

W is sample volume (L).

The MDA of the method is 0.5×10^{-3} Bq (estimation for ^{210}Po (3σ confidence level) and 3 L of water sample at a counting time of a day (84 600 s). The MDA of the instrument was effected by system background, the size and geometry of sample and counting time.

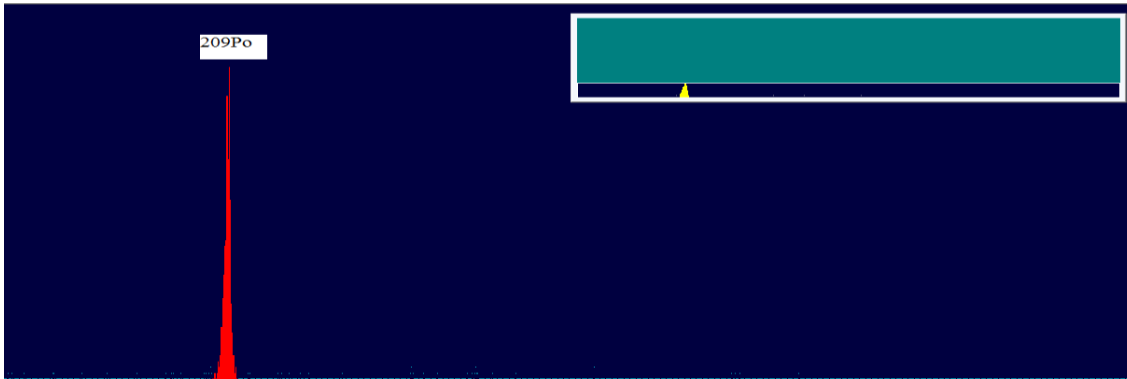


Figure 3. Alpha spectrum of ^{209}Po tracer

3.3.3. Quality assurance

The quality control was done using the reference materials ^{209}Po . The recovery rates of this tracer were up to 90%. The measurement time of each sample is chosen to account for the count rate uncertainty at the ^{209}Po peaks to be below the expected number (in test method, less than 5%).

The alpha spectrum of the ^{209}Po tracer shown in Fig. 3 with only a tracer peak offered the excellent result of the method's polonium separation and purification and source preparation. The contamination after subtracting the background count was less than 0.5%.

4. Discussions

Figure 4 showed the spectrum with well polonium separation and purification results and source preparation for the study Hai Phong thermal water sample. The background and reagent contamination was significantly less than the counting signal of ^{210}Po and ^{209}Po . Therefore, it can note here that the polonium separation and purification and

source preparation were completed, the recovery of the ^{209}Po marker by the improved method was also guaranteed similar to the conventional method (Table 1).

Both conventional and improved analytical methods determined the ^{210}Po activity in the thermal water samples and the ^{209}Po tracer (Tables 1, 2). The results showed that the difference between the two analytical methods was insignificant. Specifically, for the conventional method, the recovery rate of ^{209}Po tracer with three duplicated samples of Hai Phong, Kim Boi, and Kenh Ga thermal waters were 89, 85, and 91%, respectively. Meanwhile, the values were 80, 85, and 87% for the improved method, respectively. The difference between the two procedures for ^{210}Po activity determination was recorded as less than 5.4%. This was the expectation for this improving procedure. The overall difference ranges from 1.1–8.2%. The tracer with a low activity of 50.1 mBq was analyzed by both methods, the difference between the two methods being recorded as 1.1%. When the tracer activity was increased to 350.1 mBq, the difference was determined at 5.7%.

This study neglected the polonium separation and purification by column chromatography with Dowex 1x8 anion resin 100-200 mesh. However, the recovery efficiency is still guaranteed at > 80%. The silver disc was used for automatic deposition without a Teflon beaker and magnetic stirring, using an ordinary heat-resistant glass beaker, Leo tape. The method did not change the effect of the recovery efficiency. This improvement still ensures that ^{210}Po was deposited to only a side of the silver disc. The adhesive tape (Leo tape) was recorded its good heat resistance in dilute acid conditions and for a long time-lapse. Optimization of the ^{210}Po determination process in thermal water samples by eliminating separation and purification process by column chromatography with Dowex 1x8, 100-200

mesh anion resins and replacing deposition items of source preparation process which was significantly reducing the time and cost per analysis, meanwhile, the recovery performance is still guaranteed.

Note here that we do not deny the effectiveness of previous conventional methods in determining ^{210}Po . However, emergency response to radiation safety situations and some poor labs of developing countries is significant in today's conditions. The improving method effective with less time and cost is essential for any labs and scientists. In addition, this method has also been demonstrated on environmental samples with low ^{210}Po activity, high TSD, and a good separation that enhances the method's detection ability. Of course, this method is also used for regular or high ^{210}Po activities or TSD of environmental samples.

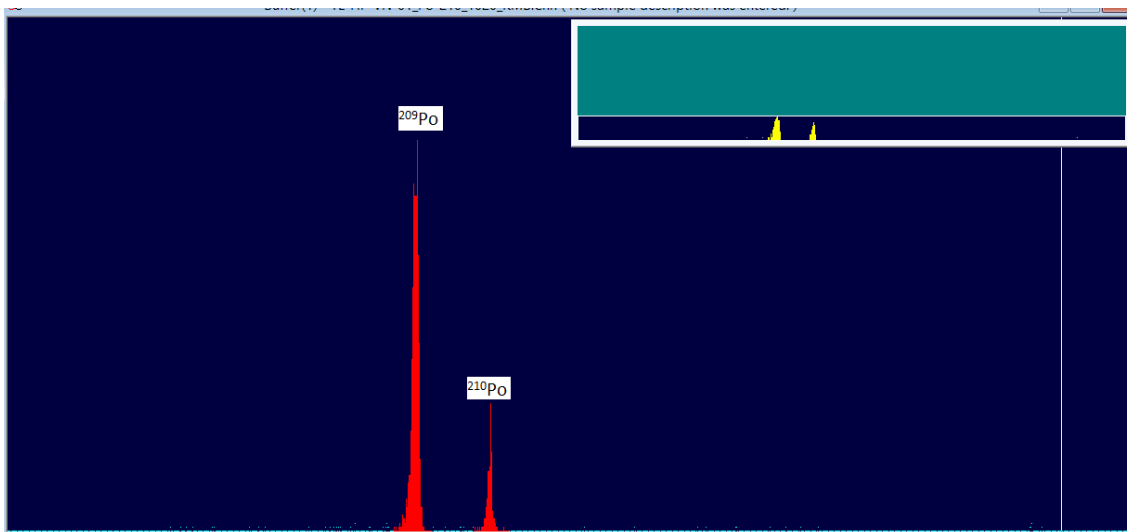


Figure 4. Spectrum with high peak resolution of Hai Phong thermal water sample

Table 1. ^{210}Po activity in thermal water samples

Sample name (n=2)	Volume (ml)	^{210}Po (mBq/l) (conventional method)	Tracer Recovery %	^{210}Po (mBq/l) (improved method)	Tracer Recovery %	^{210}Po (mBq/l) difference between two methods (%)
Hai Phong	3000	1.81 ± 0.19	89	1.78 ± 0.21	80	1.7
Kim Boi	3000	1.06 ± 0.06	85	1.02 ± 0.05	85	4.0
Kenh Ga	3000	8.56 ± 0.52	91	8.12 ± 0.63	87	5.4

Table 2. ^{209}Po tracer in certified material

Tracer no (n=2)	Activity (mBq)	^{209}Po recovery (mBq/l) (conventional method)	Recovery %	^{209}Po recovery (mBq/l) (improved method)	Recovery %	^{209}Po (mBq/l) difference between two methods (%)
^{209}Po -1	50.1	42.6 ± 3.8	85	43.1 ± 4.1	86	1.1
^{209}Po -2	149.9	133 ± 10	89	144 ± 8.6	96	8.2
^{209}Po -3	250.2	228 ± 15	91	210 ± 17	84	7.8
^{209}Po -4	350.1	308 ± 19	88	291 ± 20	83	5.7

Table 3 showed the comparison of ^{210}Po chemical recovery between the conventional and this study method which concerned the auto-deposition (spontaneous) method in environmental samples. The improving method has higher chemical recovery than some of the previous techniques (Matthews et al., 2007; Ham et al., 1997; Carvalho et al.,

2017; Gwynn et al., 2013; Lin & Wu, 2009; Prabhath et al., 2015) but it was lower than others (Sekudewicz & Gąsiorowski, 2019; Jia, Guogang, et al., 2003). The difference could be related to the volume of the sample, acid solution concentration, redox agents, deposition temperature and time, and detailed steps.

Table 3. ^{210}Po recovery comparison between methods for the auto-deposition in environmental samples

Disk type	Solution	Redox agents	Deposition temp. and time	^{210}Po recovery (%)	References
Silver	0.01M HCl	Citrate $\text{NH}_2\text{OH}\cdot\text{HCl}$	85°C, 3 h	20-95	Matthews et al., 2007
Silver	0.01M HCl	$\text{NH}_2\text{OH}\cdot\text{HCl}$	85°C, 3-4 h	67-84	Ham et al., 1997
Silver	0.5M HCl	Hydrazine	70-80°C	60-80	Carvalho et al., 2017
Silver	-	$\text{NH}_2\text{OH}\cdot\text{HCl}$	90°C, 3 h	70-80	Gwynn et al., 2013
Silver	0.5M HCl	Ascorbic acid	95°C, 3h	75-90	Lin & Wu, 2009
Silver	0.5M HCl	Ascorbic acid	90°C, 3h	15-98	Prabhath et al., 2015
Silver	0.3M HCl	Ascorbic acid	90°C, 1.5 h	~ 100	Sekudewicz & Gąsiorowski, 2019
Silver	-	-	85-90°C, 4h	96	Jia et al., 2003
Silver	0.5M HCl	Ascorbic acid	80°C, 2-4h	>80	This study method

5. Recommended radiochemical procedure

There were common five main steps for ^{210}Po determination in environmental samples, which consists of a-digestion; b-evaporation and co-precipitation; c- separation and purification; d- source preparation; e-measurement:

(a) For the environmental sample in solid state, the digestion step was required. The digestion techniques were classified mainly in two ways including (1) acid digestion (Lin et al., 2009; Cunha et al., 2001; Swift et al., 1998; Ham et al., 1997; Martin et al., 1998; Cho et al., 2016; Prabhath et al., 2015;

Sreejith et al., 2014; Jia et al., 2018; Chen et al., 2001; Szarłowicz et al., 2018; Kılıç, 2014, 2018; Belivermis et al., 2019; Sanchez-Cabeza et al et al., 1998; Planinšek et al., 2013; Takizawa et al., 2000; Beals et al., 1989; Sadi et al., 2016) and (2) fusion method (Jia et al., 2007; Sadi et al., 2016; Maxwell et al., 2019). Acid digestion has been limited to the effect of ^{210}Po volatilization under different temperature conditions (Martin et al., 1969; Heyraud et al., 1979; Jia et al., 2007; Mabuchi et al., 1963; Henricsson et al., 2011; Seiner et al., 2014). Meanwhile, the fusion method was presented to be a more active

decomposition technique in completely dissolving various solid samples in the medium than acid digestion;

(b) The samples will be pre-concentrated after the decomposition process (evaporation and co-precipitation). This step need to perform for high volume sample or sample need to reduce volume. Polonium co-precipitation was an important step to improve the MDA and sensitivity of the method. Hydroxy iron ($\text{Fe}(\text{OH})_3$) and oxit manganese (MnO_2) were the most frequently used reagents for polonium co-precipitation (Martin., 1998; Kim., 2009; Lee., 2010; Lee., 2014; Fonollosa., 2015; Sekudewicz., 2019; Jia., 2001). There were some ways to co-precipitate polonium such as a Te carrier from an SnCl_2 solution (Rushing., 1966), bismuth phosphate (Holgye., 2007; Maxwell., 2013), and by cobalt ammonium pyrrolidine dithiocarbamate (Co-APDC) chelate (Wildgust., 1998; Gasco., 2002), calcium phosphate (Haridasan., 2001) and calcium carbonate (Nozaki., 1973).

(c) For separation and purification of polonium, there were three common ways which used include (1) liquid-liquid extraction methods (Figgins et al., 1961; Kim et al., 2009; Lee et al., 2010; Jia et al., 2004, 2008; Martin et al., 1992; Hampson et al., 1973; Chen et al., 2001); (2) Ion exchange chromatography (Kmak et al., 2017; Strelow et al., 1988) and (3) extraction chromatographic separations (Horwitz et al., 1992; Vajda et al., 1997; IAEA., 2006; Horwitz et al., 1994; Meli et al., 2013; Maxwell et al., 2019). The liquid-liquid extraction technique separated Po from Pb and Bi, which is essential in separating ^{210}Po in environmental samples. The ion-exchange chromatography and chromatographic extraction techniques were commonly used to separate and isolate polonium.

(d) For the ^{210}Po sample (source), preparation includes (1) spontaneous

deposition from an acidic solution, heating on a metal plate (Oliveira et al., 2006; Carvalho et al., 2010, 2011); (2) the ^{210}Po source preparation process was carried out by other methods such as electrodeposition (Figgins et al., 1961) and (3) micro-precipitation methods (Maxwell et al., 2013; Guérin et al., 2014; Guérin et al., 2013; Song et al., 2017). These methods became an excellent deposition technique to reduce the time and limit the temperature influence on the process for different samples (Thakur et al., 2020).

(e) Measurement of ^{210}Po was performed by three main methods including (1) alpha spectrometry; (2) photon electron rejecting liquid alpha spectroscopy (PERALS) (Véronneau et al., 2000; Landstetter et al., 2014; Case et al., 1982) and (3) Liquid scintillation counter (LSC) (Véronneau et al., 2000; Matthews et al., 2007). Alpha spectrometry was the most common method for the ^{210}Po determination in environmental and biological samples using silicon surface barriers or PIPS detectors. The PERALS method was high sensitivity, speed of sample preparation allows to combine liquid-liquid extraction separation with alpha activity measurement in a water-immiscible scintillator. The LSC was also a novel and effective technique for the ^{210}Po determination in environmental samples. The alpha spectroscopy is effective in the accurate determination of alpha and ^{210}Po emission isotopes (IAEA., 2009).

In this study, following stages (Fig. 5), the recommended radiochemical procedure includes sample preparation, co-precipitate, precipitate separation, source preparation, and measurement. The first step (Sample preparation) with 3 liters of each sample was to add HCL (5M) to avoid the lost chemical then add about 100mBq of ^{209}Po tracer. The co-precipitation of polonium with MnO_2 under pH >9 conditions was added of KMnO_4 and MnCl_2 (Skwarzec, 1997). Adjust pH >9

was used with ammonia (NH₃) before precipitation with MnO₂. To optimize the co-precipitation with MnO₂, the sample solution was mixed by magnetic stirring for about 30 minutes at a temperature of ~ 80°C. The obtained sample was kept overnight before precipitate separation. The next step of precipitate separation was begun by separating and washing the residue from the liquid and distilled water. The obtained residue was dissolved in HCl 5-8M and evaporated to dryness. Then the sample was dissolved with 30 ml HCL (0.5 M). This solution was added ascorbic acid to reduce Fe³⁺ to Fe²⁺ because Fe³⁺ in the sample interferes with spontaneous deposition of polonium. In the source preparation step, the ²¹⁰Po in solution was automatically deposited on a side of the silver disc for 3-4 hours at a

temperature of ~80°C. In this study, the silver disk used Leo tape to secede the liquid with a side of the silver disk. The obtained sample was washed with distilled water then keep dry at room temperature. The sample was ready for PIPS alpha spectrometry measurement in the final step. Simultaneously with the study sample, a blank background sample (distilled water) was also prepared and measured to account for the background derived from the chemical reagents, electronic noise, or alpha particles emitted from the chamber. The ready sample was measured using an ORTEC Alpha-Ensemble-4 spectrometer. Quality control was performed using the reference ²⁰⁹Po tracer. The measurement time of each sample was chosen to get the uncertainty in measuring the counting rate at the peaks of ²¹⁰Po and ²⁰⁹Po less than 10%, respectively.

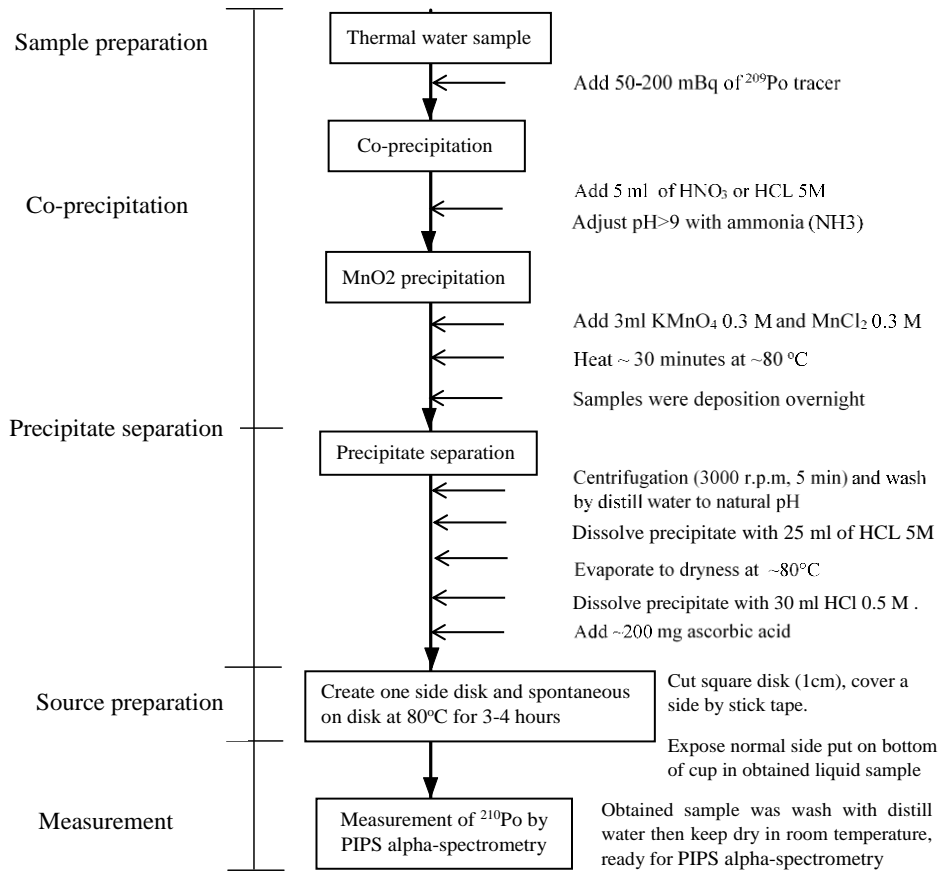


Figure 5. Recommendation for ²¹⁰Po determination procedure in thermal water

6. Conclusions

In this study, the analytical procedure of a new improved method was elaborated for ^{210}Po determination in thermal water sample. The method procedure has been optimized by eliminating the separation and purification of polonium step in the way without using a chromatographic column with Dowex 1×8 100-200 mesh anion resin for Po separation and simplified spontaneous deposition step do not use of the Teflon beaker or electrodeposition. The optimized step was used with a single silver disc and regular glass cups. The result showed that the ^{210}Po determination improving method was elaborated more simplified, less time-consuming, and more significant reduction of costs with chemical recovery >80%. Therein the chemical recovery was insignificant compared to conventional and previous methods. The re-elaborated method could apply for any liquid environmental samples in particular and environmental samples in general.

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