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A review of radio chemical analysis and estimation of ^{210}Po in soil matrices

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ABSTRACT

The naturally occurring radionuclide ^{210}Po , arising from the uranium–radium decay series, provides a considerable contribution to the radiation exposure to humans. Polonium is analyzed for a variety of purposes, including for radiological impact assessment or as a tracer of environmental processes. Losses of polonium may occur at temperatures above 100 °C, depending on conditions, requiring particular care in sample preparation and treatment. There has been little development regarding analysis of polonium in environmental samples since 1960 as radiochemical analysis of polonium is quite straight forward due to easy of source preparation through auto-deposition on to metal surfaces. In this paper a brief review of estimation of polonium in the soil samples have given emphasis. Copyright © 2015, The Egyptian Society of Radiation Sciences and Applications. Production and hosting by Elsevier B.V. This is an open access article under the CC BY-NC-ND license (<http://creativecommons.org/licenses/by-nc-nd/4.0/>).

1. Introduction

^{210}Po is a naturally occurring alpha emitter with a half-life of 138.3 days. It was discovered by Pierre and Marie Curie in 1898 while working on uranium and thorium minerals. It is a silvery gray metal with soft enough to be scratched easily. Polonium occurs widely in the terrestrial environment and as an alpha emitter. Most of the isotopes of polonium are short lived (Table 1) and their behavior is dictated by the fact that they decay at the point of production and have little chance to exhibit different behavior to that of the parent nuclide. So it is the ^{210}Po ($T_{1/2}$: 138 days) assumes importance in the environmental and radiological health point of view. ^{210}Po can be

found at elevated concentrations at some distance from sites of release due to the extreme mobility of the noble radon gas, ^{222}Rn (Al-Masri et al, 2010). The rapid decay of ^{222}Rn ($T_{1/2}$: 3.82 days) in the atmosphere generates ^{210}Po and ^{210}Pb , which are rapidly adsorbed by aerosols and returned to earth surface (Eisenbud, 1987; Ivanovich & Harmon, 1992). ^{210}Po in environment may originate either as a product of the radioactive decay of radionuclide of ^{238}U series present in the soil (supported) or the result of the precipitation of radon decay products from the atmosphere (unsupported). Other natural sources of ^{210}Po include volcanic eruptions, fires, fossil-fuel burning, migration of sea–salt, and resuspension of soil dust (Jia et al., 2001; Poet, Moore, & Martell, 1972). Because of this widespread distribution it also enters food chains through

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Table 1 – The half-lives and main alpha-energies of ²⁰⁸Po, ²⁰⁹Po and ²¹⁰Po [Table of Isotopes].

Nuclide	Half-life	Decay mode	Energy (keV)	Abundance (%)
²⁰⁸ Po	2.897y	α	5110	99
			4220	2.4E-4
²⁰⁹ Po	102y	α	4877	99.26
			4617	0.48
²¹⁰ Po	138.4d	α	5304	100

direct uptake, ingestion or inhalation (Matthews, Kim, & Martin, 2007). Extensive studies were carried out to measure the activity concentration of polonium in soil, water, air, sediment, rainwater, human and non human biota, food etc. (Haridasan, Paul, & Desai, 2001, Jha, Tripathi, Sethy, Sahoo, & Puranik, 2013; Sethy et al, 2010 & 2014, Narayana & Rajsekhar, 2010). The behavior of U-238 series radionuclides including ²¹⁰Po in soils has been reviewed by Thorne and Mitchell (2011). For a detailed review of U-series chemistry in soils, two publications by the IAEA (1999, 2004) provide a good starting point that is complementary to Thorne and Mitchell (2011). Very little is known about the soil chemistry of polonium but it is expected to adsorb onto soil particles, such as mineral colloids and humic substances. The ²¹⁰Po is considered to be in equilibrium with ²¹⁰Pb, suggesting that the ²¹⁰Pb in the soil is the main source of ²¹⁰Po irreversibly adsorbed on clay and organic colloids in the soil (Parfenov, 1974). The activity concentration levels of ²¹⁰Po in various soils reported are in the range of 20–240 Bq.kg⁻¹ (Al-Arifi et al., 2006; Hasanen, 1977; Karunakara et al., 2000; Myrick, Berven, & Haywood, 1983). The chemical species of polonium in soil solution may control the soil retention and plant uptake. The physicochemical properties of the soil interacting with the polonium oxide may determine the chemical species (Hansen, 1970). Upon entry into the plant the chemical form may influence the availability of the isotope if ingested by animals. The radiological health hazards of ²¹⁰Po on agricultural soils are associated with soil retention and plant uptake. The study of polonium in soil is necessary to develop the data required for estimation of the hazards from ²¹⁰Po on agricultural soils. The radiochemical analysis of ²¹⁰Po divides into three steps which the first step is the processing of sample and second is purification using radiochemical separation, liquid–liquid chromatography, solid phase extraction or ion exchange resin. The final step is source preparation in order to measure the ²¹⁰Po concentration activity in the alpha spectrometer which spontaneous or auto deposition is employed.

2. Sample processing

Environmental samples such as soil commonly employ wet acid digestion methods. In the wet-ashing methods, mixtures of acids (HCl, HF, HNO₃ and HClO₄) in different proportions are used, with varying heating temperatures and digestion times to break up the matrix and provide complete or partial access to the radionuclide (IAEA, 2009). The acid is used to oxidize the organic material present and hydrofluoric acid to destroy the

silicates (Jeffery, Bassett, Mendham, & Denney, 1989). When analyzing for ²¹⁰Po it is important that the dry sample should not be heated above 150 °C to avoid loss of polonium and low recovery. However, high temperature wet digestion using heating to fumes with perchloric acid do not results in significant polonium loss (Martin & Hancock, 1992). It is better to avoid perchloric acid during processing as it spiked the temperature due to exothermic reaction and polonium is a fairly volatile element (IAEA, 1998–2014). The addition of water (double deionized) may (0–5 mL) improve the solubility of minerals and prevent temperature spikes due to exothermic reactions (USEPA, 1996). Wet digestion using nitric acid alone found to be incomplete recovery of polonium (Card & Bell, 1985) when compared with total dissolution method. Initial treatment with 4N HNO₃ followed by combination of HNO₃ + HClO₄ (Narayana & Rajashekar, 2010) can be used for digestion of soil and sediment sample. Vaaramaa, Aro, Solatie, and Lehto (2010) digested boreal forest soil first with HNO₃ and then with a mixture of HNO₃ and HCL (1:3) and finally with concentrated HCL. Fusion technique is one of the rapid and convenient methods for digestion of soil but various studies propose not to use fusion technique for sample digestion as it results in substantial loss of polonium (Akbar, Hutton, & Prescott, 1985; Lowson & Short, 1986). Digestion of sample using pressure vessels (“bombs”) with varying temperature and time have been used (Case & McDowell, 1982; Haridasan et al., 2001; Narita, Harada, & Burnett, 1989). Jia et al. (2001) observed satisfactory recovery for ²¹⁰Po by carrying wet digestion at atmospheric pressure and 250 °C. The acid digestion method are usually time consuming and have a risk of external and cross contamination. In recent years microwave assisted temperature controlled wet digestion (Koide & Bruland, 1975; Lawrie, Desmond, Spence, Anderson, & Edmondson, 2000) found to be more convenient and time saving. Microwave assisted wet digestion was observed to be produced more satisfactory result for recovery of polonium and lead from soil and sediment samples (Beals, 1989) as both element believed to be surface adsorbed and in alpha damaged lattice sites. Jia and Torri (2007) have devised a new method for digestion of soil and rock sample containing refractory matrices. They fused the refractory sample with Na₂CO₃ + Na₂O₂ at 600° C for pre-treatment and then ²¹⁰Pb and ²¹⁰Po were can be sequentially leached out at 200–250° C temperature with HNO₃ + HF, HClO₄ and HCL.

2.1. Radiochemical separation/purification

Radiochemical separation of is a step required when the activity concentration of ²¹⁰Po is expected to be low in environmental samples. The presence of competing ions and interfering radionuclides reduces the deposition yield of Po and increasing the thickness of the deposited layer. Therefore it is necessary to purify the Po from the interfering elements and nuclides. There are various methods of separation of Po with varying degree of recoveries. For obtaining a reliable activity concentration of ²¹⁰Po, highly selective chemical methods such as solvent extraction (Martin & Hancock, 2004; Hampson & Tennant, 1973) ion exchange (Figgins, 1961; Johnsson, 1976; Reischmann, Trautmann, & Herrmann, 1984)

and extraction chromatography (Ordóñez-Regil & Iturbe, 1993) are desirable.

2.2. Ion exchange

The speed, convenience and selectivity of ion exchange separation have made this one of the preferred method for radiochemical separation. HCl preconditioned anion exchange resins are successfully employed to separate Pb and Bi and from the radium solution (Figgins, 1961; Nikitin & Katykhin, 1964). Bismuth is eluted by concentrated HCl and polonium is eluted by concentrated HNO₃ (Atwood, 2010). Johnson (1976) studied the performances of different resins such as Pb Resins, TEtraValent Actinides Resins (TEVA) and Sr Resins for selective separation of ²¹⁰Po. The Sr-Spec resin is used for isolation of polonium isotopes from environmental samples (El Afifi & Borai, 2006). The extractant is a crown ether (1 m bis-4,4'(5'-t-butyl-cyclohexano-18,6-crown ether) dissolved in 1-octanol (Lee, Lee, Song, Kim, & Martin, 2010). The structure of the extractant is shown in Fig. 1.

2.3. Solvent extraction

Polonium separation and purification may be achieved through solvent extraction. Po has four expected oxidation states (-2, +2, +4, and +6), but only the +4 is stable in solutions. In HCl solutions, Po is assumed to be PoCl₆²⁻ (Martin & Hancock, 1992). Based on this chemistry, there are a wide variety of extractants used in the literature (Bagnall & Robertson, 1957; Barbano & Rigali, 1967; Danon & Zamith, 1956; Schulz & Richardson, 1968) that yield excellent results for separating Po from Pb and Bi, as well as other radionuclides. These include diethylammonium diethyl dithiocarbamate (DDTC), diethyl dithiocarbamate (DDC), Tri-isooctylamine (TIOA), and tricyclo phosphine oxide (TOPO) in toluene (Matheny, 2011). Several studies Lee et al. (2010), Martin and Hancock (1992) and Kim, Lee, and Martin (2009) have reported use of diethylammonium diethyl dithiocarbamate (DDTC) in CH₂Cl₂ for the extraction of Po from aqueous

solution in recent studies. Solvent extraction study on the hydrolysis of tracer concentration of polonium (IV) in nitrate solutions (Hataye, Suganuma, & Sakata, 1981), α-hydroxyisobutyric acid solutions (Johansson, Malmbeck, Wierczinski, & Skarnemark, 2001) and tri-n-butyl Phosphate (Chen, 2013) have been also reported. Hampson and Tennant (1973) separated polonium from environmental matrices by extracting with tri-n-octylphosphine oxide-n-heptane-nitric acid-sodium nitrate system and stripping into ammonium carbonate solution and electrodeposition.

2.4. Extraction chromatography

Extraction chromatography has been applied for the separation of Po in early studies of development of radiochemistry. Frlerson and Jones (1951) used a mixture of butanol, pyridine, concentrated hydrochloric acid, acetic acid and water produced a good separation of a Bi and Po.

Polonium also separated using paper chromatographic techniques from a mixture of Bi, Po, Te and Se in a mixture of HF and methyl ethyl ketone (Crouthamel, Heinrich, & Gatrousis, 1958). Separation of polonium species in nitrate (Suganuma et al., 2009) and HCl (El Afifi & Borai, 2006) solutions by paper chromatography techniques provides useful information about purification and quantification of polonium.

3. Estimation of ²¹⁰Po

3.1. Spontaneous deposition

Marckwald (1905) observed that Po can be deposited on Ag disc from a dilute solution of HNO₃ or HCl. A black film appears during the deposition on the Ag disc and this may consist of either an oxide or peroxide (Figs, 1961). Curie (1925) reported spontaneous deposition of Polonium on Ag from acetic acid solutions and the depositions improved in the presence of small amounts of HCl. Spontaneous deposition of Polonium is more efficient (>99%) in 0.5N HCl medium at 97 °C than HNO₃,

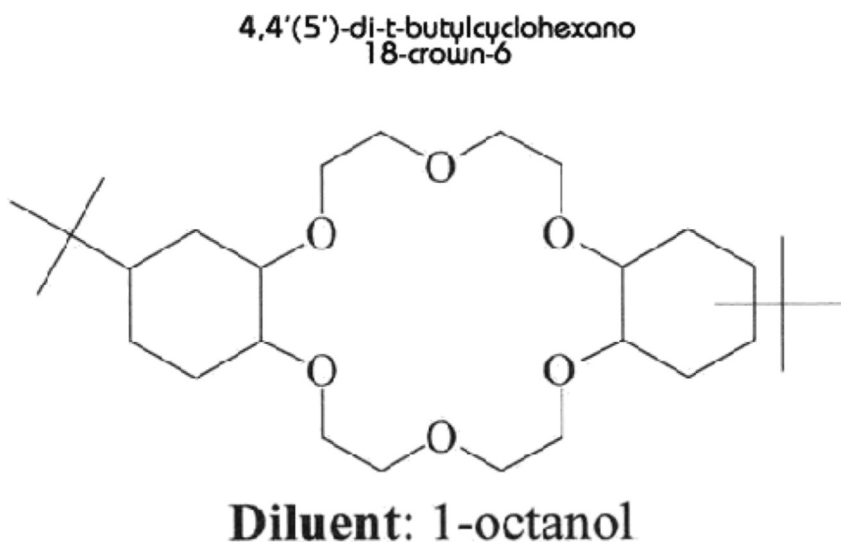


Fig. 1 – The Sr-resin structure. http://www.eichrom.com/products/info/sr_resin.cfm.

H₂SO₄ or acetic acid although deposition from HNO₃ reported to give less surface oxidation of the Ag disc (Curie and Joliot, 1931). With repeated use of fresh Ag surface the polonium can be scavenged from a dilute HCl solution with a recovery of 90% (Figs, 1961). The interference in the spontaneous deposition of polonium is due to presence of Fe³⁺, Au, Hg, Pt, or Te. These impurities inhibit the deposition process and must be removed either by reduction with hydrazine (Haissinsky, 1936a, 1936b), acerbic acid or SO₂ (Figs, 1961). The spontaneous deposition is simpler than electro-deposition, but it has the disadvantage that the electrode potential is fixed by the selection of the metal. As the deposition of potential of Po is close to that of Bi, presence of ²¹⁰Bi may produced an incorrect result if there is a delay in deposition and counting (Martin & Hancock., 1992). Flynn (1968) recommended using a suitable amount of Bi carrier to prevent the co-deposition of ²¹⁰Bi in the deposition surface. The spontaneous deposition of polonium on Ag using dilute HCl medium in presence of Bi carrier and acerbic acid/hydroxylamine hydrochloride produces a better yield at 70–80 °C with a deposition time of 2 h (Martin & Hancock., 1992). Number of other metal surface such as Cu, Ni, Bi, Pt and Pd can be used for spontaneous deposition of polonium with varying degree of recoveries and merits. The deposition efficiency of Po on different metals has been studied in details by Porntepkasemsan, Srisuksawad, and O-manee (2011). The deposition efficiency of Po towards these metals is silver > nickel > copper at the optimum conditions, 0.3 M HCl, at 800°C and 5 h of plating time. The theoretical deposition rate of the Po on cathode surface can be expressed as (Smith & Hamilton, 1984):

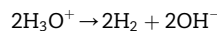
$$dN/dt = K_1(A\omega^{0.5}/V)(N_0 - N_s)\exp(K_2T)$$

Where,

- dN/dt is the rate of deposition;
- K₁ and K₂ are constants; T is the absolute temperature;
- A is the active area;
- V is the volume of solution,
- ω is the angular velocity of a rotation of a disc;
- N₀ and N_s are the concentrations of Po in the bulk liquid and at the disc surface respectively. This equation suggests that the optimum plating conditions would require a high temperature and high angular velocity and a small solution volume.

3.2. Electro deposition

Electrodeposition is a commonly used method in the preparation of sources in spectroscopic measurement as it is simple and produces a thin layer of the desired element. The mechanism of the electro-deposition process is governed by Hansen's theory of electrodeposition of lanthanides and actinides hydroxides at low current densities. A high concentration of hydroxyl ions adjacent to the cathode surface is required to precipitate hydroxides from very low mass concentration of the radionuclides in the electrolyte. This is called the "hydroxyl layer". The reaction to produce this layer on the cathode is the removal of hydrogen ions supplied by the electrolyte or by the dissociation of water:



The electrolysis of polonium was studied by Frederic Joliot (1930) in his doctoral research and spontaneous deposition on silver foils is still nowadays the preferred choice of procedure to prepare polonium sources (Crespo, 2012). The electrodeposition of the polonium depends upon the pH of the electrolyte and the presence of salts (Haissinsky, 1936a, 1936b). Polonium in trace quantities can be cathodically deposited on different electrodes with a suitable current density from a variety of electrolytes. (Meyer & Schweidler, 1906, Paneth & Hevesy, 1913; Broda & Wright, 1946; Miura, Kawabe, & Kirita, 2000). Kihara, Yoshida, and Matsui (1988) have studied electro deposition of polonium and the interaction between deposited polonium and based electrode material. Different radiation sources of alpha emitting nuclides have been prepared by electrodeposition from organic or aqueous electrolytes (Broda, 1950; Rodden, 1950) in very early years. Crowen, Hodge, and Folsom (1977), reported direct electrodeposition of polonium on carbon electrode from acidic solution with an efficiency of 40–85%. Electrodeposition using dilute HF solution produces excellent recovery (Figgins, 1961) and reasonable separation from Bi but there is no record of this procedure having been used in large scale separation of polonium (Atwood, 2010). Separation of polonium from uranium containing minerals and its electrodeposition on stainless steel disk (Ordoñez-Regil & Iturbe, 1989) produced encouraging results. Vesterbacka and Ikaheimonen (2005) have studied the effects of the deposition time, deposition conditions and the interferences by competing ions in the deposition of ²¹⁰Po. In summary, the electro deposition of ²¹⁰Po is performed from a dilute acid solution to which some complexing agents or carriers are added to prevent interference by competing ions.

4. Measurement

The deposited polonium on silver disk has to be quantified in order to find out the activity concentration of polonium in environmental matrices. This can be carried out by direct alpha counting of the disk in a ZnS(Ag) based detector or by alpha spectrometry. The alpha source counting technique with ZnS (Ag) detectors has been well documented and described for the determination of alphas derived from the U and Th series isotopes in various environmental and archeological matrices (Tite & Waive, 1962, Hansenm & Stout, 1968; Burcham, 1973; Bell, 1979; Liritzis, Y. & Galloway, R. B, 1982; Liritzis, I. & Galloway, R. B, 1982; Akber, Hutton, & Prescott, 1983; Aitken, 1985; Huxtable, Aitken, & Weber, 1972; Liritzis & Kokkoris, 1992; Xanthakis, Liritzis, & Galloway, 1992. The silver disk can be directly counted on both sides in a ZnS(Ag) detector based alpha counting set up (Carfi & Lonati, 1966, Hameed, Shaeed, & Somasunaram, 1997; Jha et al., 2013; Sethy et al., 2014; Rani, Jeevanram, & Kannan, 2014). The counting set up is calibrated against an electroplated alpha source of uranium (Jha et al., 2013) or plutonium (Carfi & Lonati, 1966). The average counting efficiency of the counter was 25 ± 3%. Average background of the counting set up was

1.3×10^{-3} counts per second. The ZnS(Ag) based alpha counting set up can be calibrated by using ^{209}Po tracer (Carfi & Lonati, 1966) or standard ^{210}Po solution made from high grade uranium ore (Suriyanarayanan et al., 2008, Jha et al., 2013, Sethy et al., 2014). The ^{210}Po activity concentration is calculated by counting the sample source for an appropriate time. The counting time depends on the sample and blank count rates and also the detection limit and decision threshold required (IAEA, 2009). For measurement and differencing isotopes, alpha spectrometric technique is followed. Alpha spectroscopy is considered as the best method of measuring polonium isotopes (IAEA, Po-Factsheet). The alpha energies of polonium isotopes (Table 1) are having narrow differences which need maximum resolution of peaks. Hamilton and Smith (1986) and Garcia-Torano (2006) have been given details reviews of for the determination of polonium isotopes by alpha spectrometry.

5. Conclusion

The extensive studies carried out over the years provide alternatives choices to the researchers for radiochemical analysis and determination of polonium in various environmental samples. Depending upon the need and requirement such as nature and activity level of the sample, detection level, cost effectiveness etc, a particular methodology can be chosen. Processing of sample using conventional wet digestion using different combination of acid is more cost effective than microwave assisted wet digestion. Spontaneous deposition of polonium followed by alpha counting in a ZnS(Ag) detector provides a cheaper alternative to alpha spectrometry. However for isotopic measurement alpha spectrometry is necessary. The flow diagram of radiochemical estimation of polonium in soil samples can be presented as

- Step 1: Sample Processing
- Step 2: Purification
- Step 3: Electrodeposition or Spontaneous deposition from acidic medium
- Step 4: Alpha counting/Alpha spectrometry

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