Optimization of auto-deposition for Po-210 in environmental sample

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1. Introduction

Monitoring polonium-210 in effluent and natural waters requires a method which is reliable, rapid and sensitive. The most widely used techniques for determination of Po-210 are the spontaneous deposition of polonium on to silver from weakly acidic solutions [1,2]. This method is subject to interference from oxidants, organic and inorganic materials. The presence of interfering ions makes the procedure less reliable both by reducing the deposition yield of polonium and increasing the thickness of the deposited layer. Therefore, for obtaining reliable activity concentration of Po-210 in water, it is necessary to purify polonium from interference elements and nuclides before plating.

Recently, highly selective chemical methods of Po-210 like extraction chromatography with Sr-resin in environmental samples have been developed [3,4]. With Sr Spec resin, the chemical recoveries of Po-210 in environmental samples are so changeable that they are sometimes obtained to be less than 60 %. Therefore, it is desirable to check the parameters affecting the recovery of polonium such as pH, time, volume and temperature in the deposition solution for obtaining reliable data of Po-210 analytical method. In this study, the chemical yield was investigated in simulated solution on different deposition conditions and the determination method of Po-210 modified deposition step was applied to tap water sample.

2. Experiments and Results

In a series of experiments, polonium was deposited on silver plate with changing pH (-1.0 – 5.0), time (0.5 h – 3 h), volume (5 mL – 25 mL) and temperature (30 °C – 90 °C). The plating experiments were performed twice. After completion of deposition, the silver disc was removed, washed with distilled water and dried at room temperature. The planchette was then measured by alpha spectrometry. To compensate for chemical recovery, Po-209 (30 mBq), as a yield, were added into 2.5 L tap water sample. MnO_2 precipitation was obtained in ammonia solution pH 9, after adding $KMnO_4$ and $MnCl_2$. The precipitate was dissolved with 2 M HCl (1% H_2O_2) and Po was purified by Sr Spec resin method. After pH adjustment, the purified Po fraction was auto deposition and polonium was measured by α-spectrometry.

In chemical aspects, several parameters such as pH, reducing agent (ascorbic acid) and complexing agent (sodium citrate) are influenced on spontaneous deposition of polonium in the large volume of water sample. Among factors affecting the recovery of polonium, pH is a main parameter in the deposition of polonium for purified polonium sample after chemical separation. Most of polonium was deposited on the silver plate at acidic pH (0.0 - 2.0) zone. Over pH 3.0, polonium is easily adsorbed onto the surface of beaker and reduced recovery. Also, below pH -0.5, the silver was etched, which reduced recovery and decreased the α-peak resolution.

In physical aspects, the theoretical deposition rate of polonium can be expressed as

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

where dN/dt is the rate of deposition; K_1 and K_2 are constants; T is the absolute temperature; A is the active area; V is the volume of solution, ω is the angular velocity of rotation of the disc; N_0 and $N_s$are the concentrations of polonium in the bulk liquid and at the disc surface respectively [5]. This equation indicates that optimum plating conditions would require high temperature and angular velocity and small solution volume. In this study, the recoveries of Po-209 from the simulated solution were measured as a function of time, volume and temperature with/without stirring. About 99 % of polonium was deposited on the silver plate over 1.5 h. With deposition times longer than 3 h, the surface of

silver plate may be etched from acidic solution and reduces recovery. At a stationary silver disc, the recovery of Po-209 from the solution at 90 °C was just 60 % after 1.5 h. With rotation for 1.5 h, recovery of Po-209 was nearly 100 % at 90 °C. Also, with increasing temperature, the recovery of Po-209 was increased. The improvement in the deposition at higher temperatures with rotation has been explained by the removal of ozone, which coated on the surface of silver disc as silver oxide or peroxide, formed from dissolved oxygen in the solution.

With extraction chromatography with Sr-resin, the activity concentrations of Po-210 in tap water of Vienna was measured to be in the range of 1.50 mBq/L to 3.96 mBq/L with a mean value of 2.18 ± 1.06 mBq/L. The chemical yields of Po-210 in tap water samples with extraction chromatography *method were presented in Table 1. The chemical yields for the deposition solution adjusted to pH 2* were lower than those for the deposition solution adjusted to pH 0. These means the deposition solution adjusted to pH 2 may be not enough to dissolve the evaporated residue, which was contained polonium and crown ether, passed through Sr-resin. However, the deposition solution adjusted to pH 0 can dissolve easily the residue, so that the deposition solution is transferred completely to deposition apparatus. Also, the addition of reducing agent increased the chemical yield by 7 - 11 %. However, there were not found to be significant effect on increasing the chemical yield after adding complexing agent, which can facilitate the deposition of Po, though it was reported that the chemical yield of Po was increased with adding complexing reagent on the spontaneous deposition of Po in the presence of large quantities of foreign ions.

3. Summary

The deposition conditions for plating polonium have been optimized with deposition parameters such as pH, volume and temperature of the deposition and deposition time. In the tap water, the chemical yields of polonium forthe deposition solution adjusted to pH 0 were higher than those for the deposition solution adjusted to pH 2. This modified auto-deposition method made it possible to obtain reliable data of activity concentration of Po-210.

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Table 1. Chemical yields of Po-210 in tap water

Chemical method	Deposition method	Activity concentration of Po-210 (mBq/L)	Chemical yield (%)
Solvent extraction	Classical (pH 2)	1.42± 0.16	93.2 ± 4.5
	Optimized (pH 0)	1.67 ± 0.14	92.2 ± 5.1
Extraction chromatography	Classical (pH 2)	1.93 ± 0.22	60.1 ± 3.9
	Optimized (pH 0)	1.62 ± 0.15	90.3 ± 4.2