Determination of trace uranium (VI) using its self-assembly with a tetradebate–monodentate ditopic ligand by resonance light scattering

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ABSTRACT

We describe here a resonance light scattering (RLS) method for uranium (VI) detection by using phosphorylethanol-amido-salophen (PAS) as optical probe. PAS is a tetradebate–monodentate ditopic ligand in which the tetradebate and monodentate ligands are salophen moiety and phosphate group, respectively. PAS can chelate uranyl with its salophen moiety. The chelated uranyl can connect phosphate group in another PAS through coordination reaction. This causes the self-assembly of PAS with uranyl to form a metallo-supramolecular polymer, resulting in a production of strong RLS signal. The RLS method was established based on the self-assemble. The RLS intensity is linearly related to the concentration of uranium (VI) in the 0.8–32 ng mL\(^{-1}\) range, with a detection limit of 0.24 ng mL\(^{-1}\) detection limit under optimal conditions. The method was successfully applied to determine uranium (VI) in environmental water samples with the recoveries between 97.1% and 102.6%.

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1. Introduction

Uranium is the most important source in nuclear energy generation. Identification and isolation of uranium from various resources are of great significance due to the worldwide demand. Uranium is also a highly toxic metal element with the severe damage to human health and environment. Therefore, it is of importance to determine uranium in water and environment. Several analytical methods have been applied to determine uranium, including gamma ray absorption [1], atomic emission spectrometry [2], inductively coupled plasma-mass spectrometry (ICP-MS) [3], neutron activation analysis [4], surface-enhanced Raman spectroscopy [5], fission track registration technique [6], fluorimetry/laser fluorimetry [7,8], cathodic stripping voltammetry [9] and kinetic phosphorescence [10]. However, these methods often are unsuitable for the routine analysis because they need high running cost with expensive equipments. Although spectrophotometry [11,12] is not costly, it often needs pre-concentration procedure for the determination of trace uranium. Therefore, it is significant to study new analytical methods for the routine determination of uranium.

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Ditopic ligands are a type of important ligands. They can self-assemble with metal ions to form various metallo-supramolecular polymers (MSPs). MSPs have various and tunable properties and can be used in many fields by utilising these properties, such as they can be used to determine various target analytes by utilising their spectroscopic and electrochemical properties [13–16], and can be used to separate substances by utilising the self-assembly properties. Up to date, most of ditopic ligands used to assemble MSPs contain two same ligands. The two same ligands usually are tridentate ligands or bidentate ligands since the coordination numbers of metal ion usually are six or four. The ditopic ligands containing two different ligands also can be used to assemble MSPs. However, the study on this type of ditopic ligands and the corresponding MSPs as well as their application have less been reported.

Resonance light scattering (RLS) methods are a type of spectroscopic analytical methods [17]. They are based on the assembly or aggregation of the molecules and/or particles which result in the enlargement of particle size. The assembly and aggregation reactions include the self-assembly of ions [18–21], the aggregation of nanoparticles [22–25], the combination of antigens with antibodies [26], and the binding of small molecules with DNA and proteins [27–30]. RLS methods have the advantages of simplicity, rapidness and high sensitivity, and have been used to determine various target analytes, including ions [19,20,24], organic and biological molecules [18,21–23], medicines [27–30], as well as antigens and antibodies [25,26]. The self-assembly of ditopic ligands with metal ions to form MSPs also causes the enlargement of particle size and can result in an enhanced RLS signal. Therefore, a type of RLS analytical methods can be developed based on this self-assembly.

Uranyl ion is a linear cation. It can react with ligands to form planar complexes with typical coordination number of five. Salophen and its derivatives are Schiff base type tetradeinate ligands. They can chelate uranyl to form stable uranyl–salophen complexes. In these complexes chelated uranyl leaves one site open for the coordination with another monodentate ligand. Previous studies have demonstrated that uranyl–salophen complex can bind monodentate ligand phosphate group with strong affinity and high selectivity to form stable phosphate–uranyl–salophen sandwich-type structure [31,32]. Hence uranium has been separated and determined by utilising the formation of the sandwich-type structure [33,34].

In recent years, based on the formation of phosphate–uranyl–salophen sandwich-type structure and the further formation of MSPs, a series of RLS methods for the determination of uranium, fluoride, ATP and fructose 1,6-bisphosphate have been developed [35–38]. These RLS methods have the advantages of high sensitivity, good selectivity and low cost. However, these RLS methods need two different ditopic ligands or a tritopic ligand for the formation of MSP, which results in the need of multi-step operations or a complicated reaction for the determination. Therefore, it is significative to develop new simpler RLS methods in which the formation of MSP needs only one ditopic ligand and one-step reaction for the determination of uranium and other target analytes.

Herein, we aimed to prepare a ditopic ligand that contains a tetradeinate ligand and a monodentate ligand so as to assemble a MSP of uranyl for the determination of uranium by RLS method. The tetradeinate and monodentate ligands are salophen moiety and phosphate group, respectively. This tetradeinate–monodentate ditopic
ligand is phosphorylethanol-amido-salophen (PAS). It can be synthesised by utilising the condensation reaction of carboxylated salophen with phosphorylethanolamine. We anticipated that PAS can self-assemble with uranium (VI) or uranyl cation to form a MSP of uranium (VI), resulting in a production of strong RLS signal. Thus, a new RLS method for the direct determination of uranium (VI) can be established. The method should have the advantages of simple operation and high sensitivity because it uses only one ditopic ligand and one-step operation for the formation of MSP, and RLS methods are highly sensitive.

2. Experimental

2.1. Chemicals and apparatus

Salicylaldehyde, 3,4-diaminobenzoic acid, 1-ethyl-3-(3-dimethyl aminopropyl) carbodiimide (EDC) and N-hydroxysuccinimide (NHS) were purchased from Aladding Chemical reagent Co., Ltd. (Shanghai, China). Phosphorylethanolamine was purchased from Sigma-Aldrich (Oakville, ON, USA). Uranyl nitrate hexahydrate was obtained from Hubei Chushengwei Chemical Co., Ltd. (Wuhan, China). The used chemicals including other salts, acids and bases were of analytical grade. All aqueous solutions were prepared in doubly distilled water.

The standard stock solution of uranium (10.00 mg mL\(^{-1}\)) was prepared by dissolving 2.123 g uranyl nitrate hexahydrate into water with the total volume of 100 mL. The concentration of the standard stock solution was verified by Davies and Gray method using iron (II) sulphate as reductant and potassium dichromate as titrant \[39\]. The working standard solutions were prepared by diluting the standard stock solution with water.

RLS spectra were recorded with a Hitachi F-4500 spectrofluorometer.

2.2. Synthesis of carboxylated salophen

Carboxylated salophen was synthesised and characterised following literature procedure \[40\]. Briefly, 2.28 g of 3,4-diaminobenzoic acid and 3.66 g of salicylaldehyde were dissolved in 75 mL of anhydrous ethanol. The mixture was stirred for 90 min under reflux, then cooled to room temperature. The solid mass was filtered, and the product, N, N-bis (salicylidene)-4-carboxy-o-phenylenediamine (carboxylated salophen), was recrystallised from ethanol.

2.3. Synthesis of phosphorylethanol-amido-salophen

PAS was synthesised by utilising the condensation reaction of carboxylated salophen with phosphorylethanolamine. Carboxylated salophen (0.18 g), 500 \(\mu\)L of 2 mol L\(^{-1}\) EDC and 2 mL of 0.5 mol L\(^{-1}\) NHS were dissolved in 50 mL of 95% ethanol. The mixture was incubated for 24 h under stir for the activation of carboxyl. Then 15 mL of 95% ethanol containing 71 mg of phosphorylethanolamine was added and the mixture was incubated for another 24 h for the condensation reaction. After that, most of the solvent was removed through evaporation under reduced pressure. The solid mass was filtered,
recrystallised in ethanol and dried under vacuum to afford PAS (0.19 g, 78.5% yield). The synthesis reaction and molecular structure of PAS are shown in Figure 1.

The prepared product was characterised by elemental analysis, $^1$H NMR and ESI-MS. Elemental analyses were carried out with a Thermo Finnigan EA 1112 elemental analyser. $^1$H NMR spectra were obtained from a Bruker 300-MHz NMR spectrometer. ESI-MS were obtained from a Finnigan LCQ electrospray ionisation mass spectrometer. For elemental analysis of $C_{23}H_{22}N_3O_7P$, calculated: C 57.16, H 4.56, N 8.70; found: C 56.84 ± 0.68, H 4.48 ± 0.12, N 8.83 ± 0.17. For $^1$H NMR (300 MHz, DMSO-d6), δ: 13.05 (s, 2H, OH), 11.99 (s, 2H, O = P–O–H), 8.92 (s, 2H, N = C–H), 8.03–8.13 (t, 1H, N–H), 7.02–7.85 (m, 11H, ArH), 4.01–4.13 (t, 2H, CH$_2$), 3.37–3.48 (m, 2H, CH$_2$) (see Figure S1 in Supplementary Material). For ESI-MS, m/z = 482 (M–H$^+$). The UV absorption spectrum of PAS in water shows that the strong absorption bands are at 216 nm, 250 nm and 327 nm, respectively (see Figure S2 in Supplementary Material).

2.4. Procedure of uranium detection

One millilitre of 10.0 μmol L$^{-1}$ PAS solution and 1.0 mL of pH 7.0 Tris-HCl buffer were added into a 10 mL volumetric flask. Then 5.0 mL of uranium (VI) calibration solution or sample solution and 2 mL of 5.0 mmol L$^{-1}$ diethylenetriamine penta-acetic acid (DTPA) was added. The total solution volume was fixed at 10 mL with water. The solution was mixed thoroughly and was then incubated at room temperature for 25 min. After that, the RLS spectra of the solution were recorded on the spectrofluorometer. A calibration curve of the RLS intensity versus uranium (VI) concentration was plotted. The procedure of detecting uranium is also illustrated in Figure 1.
3. Results and discussion

3.1. RLS spectral characteristics

Figure 2 shows the RLS spectra of 1.0 μmol L⁻¹ PAS solution in the presence of different concentrations of uranium (VI). It can be seen that the RLS intensity of PAS solution is weak in the absence of uranium. The RLS intensity of uranium (VI) solution is also weak in the absence of PAS. When uranium coexists with PAS, there is a distinct enhancement in the RLS intensity, which indicates that a molecular self-assembly reaction has occurred. PAS is a tetradentate–monodentate ditopic ligand. When a uranyl ion is chelated by the tetradeutate ligand salophen moiety in a PAS molecule, the chelated uranyl can combine further a monodentate ligand phosphate group in another PAS molecule. Thus, the self-assembled reaction of PAS with uranyl to form MSP can occur through the coordination reaction of uranyl with salophen moiety and phosphate group, resulting in the production of strong RLS signal. The maximum scattering wavelength in the RLS spectra is at 368 nm. We found that at this wavelength, there is a linear relationship between the enhanced RLS intensity and the uranium concentration under a certain condition. Therefore, a RLS method for the determination of uranium (VI) can be developed by using the self-assembled reaction of PAS with uranyl to form MSP. Thus, the RLS intensity at 368 nm was selected for the determination of uranium.

3.2. Optimisation of reaction conditions

This RLS assay for uranium (VI) is based on the enhancement in the RLS intensity caused by the self-assembled reaction of PAS with uranyl. The main factors influencing the self-assembled reaction are PAS concentration, media pH, reaction time and temperature. To make this method reproducible and highly stable, we have investigated the effect of these parameters.

Figure 2. RLS spectra of (a) 32 ng mL⁻¹ uranium (VI) solution without PAS, (b)–(g) 1.0 μmol L⁻¹ PAS solution containing (b) 0.0, (c) 4.0, (d) 8.0, (e) 16.0, (f) 24.0 and (g) 32.0 ng mL⁻¹ uranium (VI). The solution acidity is at pH 7.0.
The effect of PAS concentration on the RLS intensity was investigated by using 24 ng mL\(^{-1}\) of uranium (VI) at pH 7.0 for the test. The results shown in Figure 3 demonstrate that in the absence of uranium (VI), the RLS intensity of PAS solution is very low and hardly change with the increase of PAS concentration. While in the presence of uranium (VI), the enhanced RLS intensity is dependent on the concentration of PAS. When PAS concentration is less than 0.3 nmol mL\(^{-1}\), the RLS intensity of the solution is low and becomes stronger gradually with the increase of PAS concentration. When PAS concentration is higher than 0.3 nmol mL\(^{-1}\), the RLS intensity keeps constant and reaches the maximum, indicating that uranium (VI) has reacted completely with PAS. Thus, 1.0 nmol mL\(^{-1}\) of PAS was selected for the determination experiment.

The effect of media acidity or pH was investigated by using 24 ng mL\(^{-1}\) of uranium (VI) and 1.0 nmol mL\(^{-1}\) of PAS for the test. The results are shown as an inset in Figure 3. It can be seen that in the presence of uranium (VI), the RLS intensity of PAS solution increases at first and then decreases with the increase of pH, and reaches maximum in the pH range of 6.5–7.5. The results indicate that the self-assembly reaction needs to be carried out in a neutral solution. The reason maybe that in acidic solution, the phosphate group and salophen moiety in PAS are protonated, resulting in the reduction of their binding affinity to uranyl. While in basic solution, the binding affinity of uranyl to phosphate group and salophen moiety in PAS is also reduced due to the combination of uranyl with hydroxyl ion. Thus, we used Tris-HCl as buffer to control the solution acidity at pH 7.0 for the self-assembly reaction.

The effect of reaction time shown in Figure 4 demonstrates that when the reaction time is more than 20 min, the RLS intensity reaches the maximum and keeps constant. Thus, the selected reaction time was 25 min. The effect of reaction temperature shown as an inset in Figure 4 demonstrate that in the temperature range of 20–30°C, the RLS intensity reaches the maximum. Thus, the selected reaction temperature was room temperature 25°C.
3.3. Analytical parameters for the determination of uranium (VI)

Under optimal conditions the analytical figures of merit for the determination of uranium (VI) was tested with different amounts of uranium (VI) according to the general procedure. The calibration curve demonstrates that there is a good linear relationship between RLS intensity ($I_{RLS}$) and uranium (VI) concentration ($c$) in the range of 0.8–32.0 ng mL$^{-1}$. The linear regression equation is $I_{RLS} = 13.6c$ (ng mL$^{-1}$) +19.4 with a correlation coefficient of $r^2 = 0.9984$. The detection limit of the method was determined to be 0.24 ng mL$^{-1}$ calculated from three times the standard deviation through 11 parallel blank tests. The relative standard deviation of six parallel determinations for 2.0 and 32.0 ng mL$^{-1}$ uranium (VI) were 2.7% and 2.2%, respectively.

Table 1 shows the comparison of the analytical linear range and detection limit of our method with that of other available methods used for the determination of uranium in aqueous solution. It can be seen that the detection limit of our method is lower than that of arsenazo-III spectrophotometry. The analytical linear range and detection limit of our method are approach that of laser fluorimetry and cathodic stripping voltammetry. Although the analytical range and detection limit of kinetic phosphorescence are wider and lower than that of our method, this method demands strict conditions for the determination.

Table 1. Comparison of methods used for detecting uranium in aqueous solution.

| Method                        | Analytical linear range (ng mL$^{-1}$) | Detection limit (ng mL$^{-1}$) | Reference |
|-------------------------------|----------------------------------------|-------------------------------|-----------|
| Arsenazo-III spectrophotometry| 20 ~ 2000                              | 20                            | [12]      |
| Laser fluorimetry             | 0 ~ 5                                  | 0.5                           | [8]       |
| Cathodic stripping voltammetry| 0.5 ~ 3.5                              | 0.5                           | [9]       |
| Kinetic phosphorescence       | 0.05 ~ 1000                            | 0.012                         | [10]      |
| This method                   | 0.8 ~ 32                               | 0.24                          |           |
3.4. Influences of coexistent ions

The effect of several coexistent ions on the determination of 4 ng mL$^{-1}$ uranium (VI) was investigated. The effect of each ion was examined individually by adding a relatively high concentration of this ion into the uranium (VI) solution. When the error is less than 5%, it can be considered that there is no interference in the determination. The results demonstrate that over 500 times Cl$^{-}$, NO$_3^-$, SO$_4^{2-}$, and CH$_3$COO$^-$, 200 times of Ca$^{2+}$, Mg$^{2+}$ and Mn$^{2+}$, 100 times of Zn$^{2+}$, Co$^{2+}$, Ni$^{2+}$ and Cr$^{3+}$, and 50 times of Ba$^{2+}$, Al$^{3+}$ and La$^{3+}$ and VO$_2^+$ do not interfere with the determination, but 50 times of Fe$^{3+}$ and Cu$^{2+}$ have interference. In order to increase further the selectivity of this method, we used DTPA as masking agent for eliminating the interference. DTPA has extremely high formation constants for binding many metal ions and has a much lower formation constant for binding uranium (VI). Therefore, DTPA is an excellent masking agent for eliminating the interference of these metal ions in the detection of uranium (VI) [41]. The results demonstrate that when 1.0 mmol L$^{-1}$ DTPA is present, over 500 times of Zn$^{2+}$, Co$^{2+}$, Ni$^{2+}$, Cr$^{3+}$, Ba$^{2+}$, La$^{3+}$, Al$^{3+}$, VO$_2^+$. Fe$^{3+}$ and Cu$^{2+}$ have no interference. These results demonstrate that the present method has good selectivity towards uranium (VI) when DTPA is present.

3.5. Analysis of real samples

In order to test the validation of this method for real samples, we collected three uranium slag heap drainages, two river water samples and two distilled water samples for the analysis. Each uranium slag heap drainage was diluted with water to a 100 times dilution sample and was then analysed by this method. The river water samples and distilled water samples were analysed directly analysed by this method. The recovery experiment of adding standard uranium (VI) concentrations was carried out for evaluating the applicability and reliability of this method. The samples were also analysed with the control experiment of ICP-MS method for comparison. All results are summarised in Table 2. It can be observed that the recoveries vary from 97.1% to 102.6%. It also can be observed that the analytical results obtained from our method show good agreement with that obtained from ICP-MS method. These results demonstrate that the new method developed in this study can be successfully applied to real samples.

Table 2. Analytical results of uranium in water samples ($n = 6$).

| Sample | Found$^b$ (ng mL$^{-1}$) | Added (ng mL$^{-1}$) | Total found$^b$ (ng mL$^{-1}$) | Recovery (%) | Found$^b$ (ng mL$^{-1}$) |
|--------|--------------------------|----------------------|-------------------------------|--------------|--------------------------|
| 1      | 4.85 ± 0.12              | 5.00                 | 9.92 ± 0.19                   | 101.4        | 4.84 ± 0.09              |
| 2      | 4.41 ± 0.14              | 4.50                 | 9.03 ± 0.30                   | 102.6        | 4.49 ± 0.13              |
| 3      | 4.69 ± 0.14              | 4.50                 | 9.06 ± 0.13                   | 97.1         | 4.72 ± 0.15              |
| 4      | 7.92 ± 0.09              | 4.50                 | 12.37 ± 0.06                  | 98.9         | 8.01 ± 0.06              |
| 5      | 3.57 ± 0.21              | 5.00                 | 8.61 ± 0.19                   | 100.8        | 3.63 ± 0.19              |
| 6      | –                       | 5.00                 | 4.97 ± 0.22                   | 99.4         | –                        |
| 7      | –                       | 8.00                 | 8.04 ± 0.15                   | 100.5        | –                        |

$^a$Samples 1, 2 and 3 are 100 times dilution samples of uranium slag heap drainages. Samples 4 and 5 are river water samples. Samples 6 and 7 are distilled water samples.

$^b$Mean ± SD, were mean is mean value of six measurements, SD is standard deviation.
4. Conclusions

In this study, we synthesised tetradentate–monodentate ditopic ligand PAS and developed a new RLS method for the determination of uranium (VI) by utilising the self-assembly reaction of PAS with uranium (VI) to form MSP. The method is simple and convenient with only one operation step. The RLS method based on the self-assembly reaction using only one ditopic ligand and only one operation step may open new strategy for the determination of targets.

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Disclosure statement

No potential conflict of interest was reported by the authors.

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