

Extraction spectrophotometric determination of aluminum in dialysis concentrates with 3,5-ditertbutylsalicylfluorone and ionic liquid 1-butyl-3-trimethylsilylimidazolium hexafluorophosphate

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Abstract

The paper describes a highly sensitive and selective extraction spectrophotometric method for determination of aluminum in dialysis concentrates with new reagent 3,5-ditertbutylsalicylfluorone abbreviated as DTBSF, in which the ionic liquid 1-butyl-3-trimethylsilylimidazolium hexafluorophosphate abbreviated as [C₄tmsim][PF₆] was used as novel medium for liquid/liquid extraction of aluminum(III). Under optimal condition, DTBSF reacted with aluminum(III) to form a neutral Al–DTBSF complex rapidly, the complex was then extracted into the [C₄tmsim][PF₆] phase, the absorbance of the complex in ionic liquid at 542 nm was recorded and used to determine aluminum(III). The apparent molar absorptivity of the complex and detection limit were found to be $3.52 \times 10^6 \text{ l mol}^{-1} \text{ cm}^{-1}$ and $0.06 \mu\text{g/l}$, respectively. The absorbance of the complex at 542 nm increases linearly with the concentration of aluminum(III) up to $3 \mu\text{g}$ of aluminum(III) in 250 ml of aqueous solution. The interference study show the determination of aluminum is free from interferences of almost all positive and negative ions found in dialysis concentrate samples. The determination of aluminum in dialysis concentrates were carried out by the present method and electrothermal atomic absorption spectrometry (GTAAS). The results were satisfactorily comparable so that the applicability of the proposed method with the ionic liquid system was also investigated. Moreover, the synthesis of and conditions for the formation and extraction of Al–DTBSF complex were investigated as well. © 2006 Elsevier B.V. All rights reserved.

Keywords: Aluminum; Dialysis concentrate; 3,5-Ditertbutylsalicylfluorone; Extraction spectrophotometry; 1-Butyl-3-trimethylsilylimidazolium hexafluorophosphate

1. Introduction

The dialysis fluids are prepared from dialysis concentrates that are mixed to pure water to assure the osmotic equilibrium of the patient during the hemodialysis session. Aluminum is present as contaminant in these fluids, it is able to diffuse through the dialysis membrane and penetrate into the blood stream of the patient. The contamination level in these cases depend strongly on the quality of the water and the dialysis concentrate used in the dialysis fluid preparation. Hence, a simple and sensitive method is of primary importance for the determination of aluminum concentration in dialysis concentrates. Analytical methods used for the determination of trace levels of aluminum include mainly electrothermal atomic absorption

spectrometry (GTAAS) [1,2], chemiluminescence analysis [3], fluorescence method [4–6], inductively coupled plasma optical emission spectrometry [7], electrochemical methods [8–10] and spectrophotometry [11,12]. These methods are difficult to directly determine aluminum in dialysis concentrates due to their disadvantages in sensitivity and selectivity. Recent years, many sample separation and preconcentration procedures based on the solvent extraction were frequently applied to the determination of aluminum. The solvent extraction can effectively decrease the detection limit and eliminate matrix interference, these coupling above methods have been used to determination of aluminum in dialysis concentrates. However, the use of classical extraction method requires large amounts of high purity organic solvents for the extraction, this may also result environmental and safety problems due to volatilization of the solvents.

Ionic liquids have no detectable vapor pressure and are relatively thermal stable. So, there is no loss of solvent through

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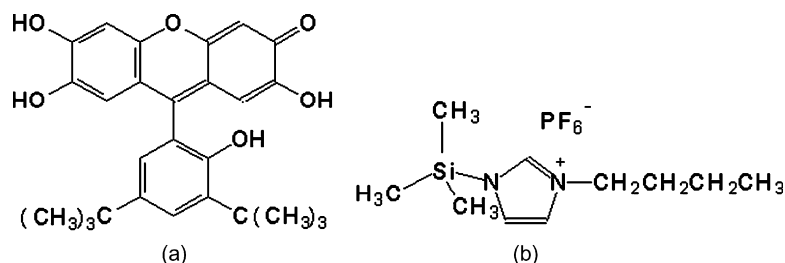


Fig. 1. Molecular structures of DTBSF (a) and [C₄tmsim][PF₆] (b).

evaporation with ionic liquids, hence many environmental and safety problems associated with organic solvents are avoided. Recently, the applications of ionic liquid in analytical chemistry have also started to receive attention [13–16], in which a typical ionic liquid 1-butyl-3-methylimidazolium hexafluorophosphate abbreviated as [C₄mim][PF₆] was often studied as extraction medium. In this study, we attempt to use the [C₄mim][PF₆] as solvent for the extraction spectrophotometric determination of aluminum in a relatively large dialysis concentrate sample (250 ml of dialysis concentrate), in which phenylfluorone was employed to form neutral complex. It was found the extraction efficiency of the complex is very low due to their relatively high water solubility [17] when the volume of [C₄mim][PF₆] was smaller than 25 ml. Moreover, the ionic liquid containing the reagent or the complex was not very transparent and the absorbance is difficult to be directly measured on spectrophotometer. In order to resolve the problem, a new reagent 3,5-ditertbutylsalicylfluorone abbreviated as DTBSF and ionic liquid 1-butyl-3-trimethylsilylimidazolium hexafluorophosphate abbreviated as [C₄tmsim][PF₆] (shown in Fig. 1) were designed and synthesized in the laboratory [18]. Basing the reaction of DTBSF with aluminum(III) and the extraction of Al–DTBSF complex with the [C₄tmsim][PF₆], a highly sensitive and selective extraction spectrophotometric method was developed for the determination of ultra-trace aluminum in dialysis concentrates. Proposed method offers the advantages of rapidity, simplicity and high sensitivity without the need for potentially toxic organic solvents.

2. Experimental

2.1. Apparatus

A UV-240 spectrophotometer (Shimadzu, Tokyo) with a 1-cm glass cell was used for recording the UV spectra or measuring the absorbance. Perkin-Elmer 2380 atomic absorption spectrometer including the autosampler equipped with Zeeman background correction (Varian, USA), and aluminum hollow cathode lamp radiation source (Varsal company, Peking, China) were used. The operating conditions of aluminum hollow-cathode were those recommended by the manufacturer (wavelength 309.3 nm, spectral band width 1.03 nm, lamp current 10 mA). A volume of 10 μl of the final solution was injected into the graphite tube by the autosampler. All pH value were measured on a pH-29A meter (Shanghai Second Analytical Instrument Factory).

2.2. Reagents and solution preparations

All reagents were of analytical reagent grade and purchased from Shanghai Chemical Company (Shanghai, China). Ultra pure water was used throughout the experiment and all glassware used was soaked with 10% (v/v) nitric acid for 1 day and rinsed with ultra pure water before use. A stock solution of 1.0 mg/ml aluminum(III) was prepared by dissolving appropriate amounts of analytical reagent grade Al(NO₃)₃ in 0.1 mol/l HNO₃. Working solutions of 2.0 μg/ml were prepared immediately before use. A 0.4 g/l of DTBSF solution was prepared by dissolving appropriate amounts of DTBSF in 95% ethanol. The ionic liquid [C₄tmsim][PF₆] was synthesized using a similar procedure [18]. To simplify the reaction procedure, a mixing reagent consisting of equal volume of 0.4 g/l DTBSF solution, 4.0 mol/l ammonium acetate and 1.0% (w/v) cetyltrimethyl ammonium bromide solution (CTMAB) was used, and this solution will be termed as the solution A in the subsequent experiment.

2.3. Synthesis of 3,5-ditertbutylsalicylfluorone

Acetic anhydride (20 g) and 2.0 ml of concentrated H₂SO₄ were transferred into 500 ml beaker, benzo-1,4-diquinone (7.0 g) was added stirring drop whilst maintaining a reaction temperature between 40 and 50 °C, before depositing the beaker overnight. After added 300 ml of cool water to the beaker, the precipitate was filtered and washed with cool water, then dried at 70 °C. Crude 1,2,4-trihydroxybenzene acetic ester was purified by re-crystallizing it with methanol, 16 g of 1,2,4-trihydroxybenzene acetic ester was obtained. 1,2,4-trihydroxybenzene acetic ester (16 g), 150 ml of 50% (v/v) ethanol, 4.0 ml of concentrated H₂SO₄ and 3,5-ditertbutylsalicylaldehyde (7.4 g) were transferred into 500 ml of flask, then, filled with nitrogen gas and warmed at 75 °C on a water-bath for 20 min to dissolve the mixture. The flask was sealed up and stood for 2 weeks under room temperature (<30 °C), the precipitate was filtered and dried at 80 °C in a vacuum system, 20 g of crude product were obtained. One gram of crude product was dissolved in 20 ml of 60% (v/v) ethanol containing 1.0 ml of concentrated H₂SO₄; then, the precipitate was filtered and washed with hot water three times. 0.6 g of pure product was obtained. Its structure was verified by IR, ¹H and ¹³C NMR spectrometry and the elemental analysis results also confirmed its purity.

Table 1
Composition of the dialysis concentrates according to the manufacturers

	Dialysis concentrate				
	I	II	III	IV	V
Sodium chloride	211	178	220	256	266
Potassium chloride	5.2	5.9	3.8	4.4	3.6
Calcium chloride	9.2	8.4	8.7	9.3	9.1
Magnesium chloride	3.8	4.1	3.2	2.5	5.3
Acetic acid	10.6	11.6	12.8	12.1	–
Glucose	–	–	–	35.9	–
Lactate	–	–	–	–	16.2

2.4. Measurement extraction efficiency of the complex

The 250 ml of aqueous standards containing 3.0 µg of standard aluminum solution and 5.0 mg of calcium(II) and 5.0 ml of the solution A were transferred into 500 ml separatory funnel. The solution was mixed well, the complex was extracted into the ionic liquid phase from aqueous solution by addition of 7.5 ml of the [C₄tmsim][PF₆]. The upper aqueous phase was taken out and the concentration of aluminum that was left in the aqueous phase was determined by GTAAS. The extraction efficiency of the complex (E_c) was calculated by $E_c (\%) = [(C_0 - C_1)/C_0] \times 100$, where C_0 and C_1 are the concentration of aluminum in aqueous phase before and after extraction, respectively.

2.5. Analytical procedure

Aluminum was analyzed in five kinds of dialysis concentrate samples that were routinely employed by the hospital in China. These include acidic dialysis concentrates with acetate and without glucose; acidic dialysis concentrates with acetate and glucose; acidic dialysis concentrates with lactate. The highest concentrations for each individual component in the dialysis concentrates, according to the respective manufacturers, are described in Table 1. Acidic dialysis concentrate was neutralized to pH 7 with sodium hydroxide solution prior to the determination of samples.

The 250 ml aqueous standards containing standard aluminum(III) and 5.0 mg of calcium(II) or real dialysis concentrate samples and 5.0 ml of the solution A were added to 500 ml separatory funnel, respectively. The solution was mixed well and deposited for 2 min, the complex was then extracted into the ionic liquid phase with 7.5 ml of the [C₄tmsim][PF₆]. Then the ionic liquid phase was collected the absorbance of the complex in ionic liquid at 542 nm was measured in a 1-cm cell against corresponding reagent blank. A reagent blank was prepared using a similar manner but without aluminum.

3. Results and discussion

3.1. The formation and characteristics of Al–DTBSF complex

The reaction of the derivatives of phenylfluorone with aluminum(III) can be carried out in a weak acid or base medium

in previous work [19], thus the effect of various buffer solutions that pH value are between 4 and 11 on the reaction was tested. Experiment showed that the reaction has the highest sensitivity in ammonium chloride–sodium acetate buffer solution. When the pH value of the ammonium chloride–sodium acetate buffer solution was between 5 and 10, the absorbance of the complex remained almost constant. Therefore an ammonium acetate solution (pH 7, without adjusting acidity) was adopted in this work. The reaction of the reagent with aluminum(III) give a very low sensitivity in the absence of surfactant. The effect of surfactant on the reaction was examined closely to increase the sensitivity. It was found different types of surfactant exhibited different effects on the absorbance of the complex. Anionic surfactant sodium dodecyl sulfate cannot improve the sensitivity obviously, and non-ionic surfactants such as TritonX-100 and Tween-80 and the cationic surfactants such as CTMAB enhanced the absorbance of the system remarkably. Among these, CTMAB gives the best sensitivity and was chosen. Under room temperature (25 °C), the reaction of DTBSF with aluminum(III) was very slow and required strongly using a heating procedure to shorten the reaction time. It was found, however, that some metal ion such as calcium(II) and magnesium(II) could catalyze the reaction obviously. When 5.0 mg of calcium(II) was added to the system, the reaction complete immediately at room temperature. Therefore a simple procedure, i.e. addition of 5.0 mg of calcium(II) to the reaction system was adopted in the present work. Moreover, the effect of amount of various reagents on the absorbance of the complex was also investigated, respectively. The results suggested that 5.0 ml of the solution A was optimum and chosen for subsequent experiments.

Under optimal condition, Al–DTBSF complex could form rapidly and stabilize for at least 24 h. The composition ratio of Al–DTBSF complex in the aqueous solution obtained using Job's method of continuous variation and the slope-ratio method was 1:3 (Al:DTBSF). The absorption spectra of the complex in aqueous and ionic liquid phase were shown in Fig. 2, respectively. From Fig. 2, it can be seen absorption spectra of the complex in aqueous solution and the ionic liquid phase have a very similar shape, this show the extraction does not influence on the complex's composition. It was also observed the stability of the complex in ionic liquid is better than that in the aqueous solution, the absorbance of the complex in the ionic liquid can remain stable for at least 48 h under temperature lower than 80 °C. Furthermore, the ionic liquid phase containing Al–DTBSF complex or DTBSF are very transparent enough to apply to photometric analysis.

3.2. The effect of amounts of ionic liquid on the extraction

Once the solution of Al–DTBSF complexes was mixed with ionic liquid [C₄tmsim][PF₆], Al–DTBSF was removed from aqueous phase and the characteristic absorption peak of Al–DTBSF complex in aqueous phase disappeared. It was found that amounts of the ionic liquid remarkably affect on the extraction efficiency of the complex. With the increase of the volume of the ionic liquid, the extraction efficiency increased rapidly at first and then leveled off after the volume of ionic liquid was

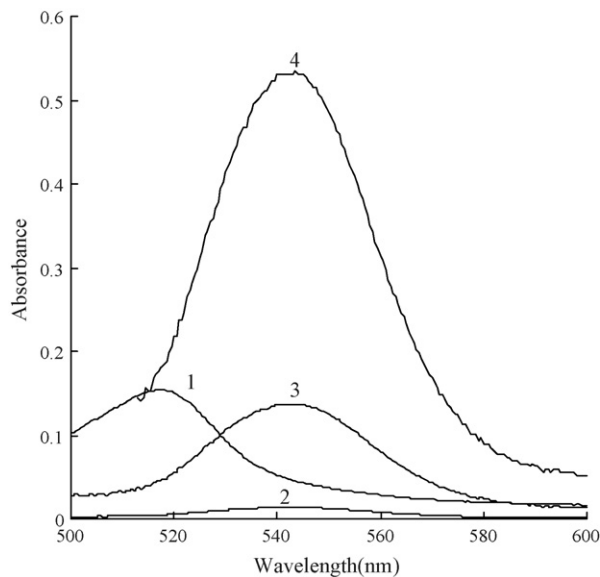


Fig. 2. Absorption spectra of the reagent aqueous solution against water (1) and the complex in aqueous phase (2 and 3) and in the ionic liquid $[C_4tmsim][PF_6]$ (4) against corresponding reagent blank. Aluminum concentrations are $4.0 \mu\text{g/l}$ for the curves 1 and 4, and $40.0 \mu\text{g/l}$ for the curve 3 in the aqueous system, respectively. Moreover, the curves 1–3 were measured by using a similar procedure described in Section 2.5 but without the extraction. The curve 4 was measured by using a same procedure as described in Section 2.5.

greater than 5.0 ml. Thus, an addition of 7.5 ml the ionic liquid is recommended.

To test advantage of the extraction system using new ionic liquid $[C_4tmsim][PF_6]$, the effect of classical ionic liquid $[C_4mim][PF_6]$ on the extraction efficiencies was also investigated by using a similar procedure, in which 5 min of centrifuging separation at 3000 rpm was used to accelerate the separation of the ionic liquid from aqueous phase. The results indicated the extraction efficiency is obviously lower than that of ionic liquid $[C_4tmsim][PF_6]$ when its amounts is less than 25 ml. This was due to relatively high water-solubility of the $[C_4mim][PF_6]$ [17]. High water-solubility results in the loss of the ionic liquid during the extraction process and decrease the extraction efficiencies of the complex. However, new ionic liquid $[C_4tmsim][PF_6]$ showed excellent performance for liquid/liquid extraction of metal complex due to good hydrophobic property and high surface tension [18].

3.3. Calibration graph, sensitivity and precision

The calibration graph was constructed from measurement obtained according to the analytical procedure. Beer's law was obeyed over the range from near the detection limits up to $3 \mu\text{g}$ of aluminum(III) in 250 ml aqueous solution. The apparent molar absorptivity of the complex was calculated from the slope of the calibration graph to be $3.52 \times 10^6 \text{ l mol}^{-1} \text{ cm}^{-1}$. The 1.645σ limit of detection ($n = 10$) for aluminum in real sample was $0.06 \mu\text{g/l}$ with a 1.2% of the relative standard deviation ($n = 5$) at the $4.0 \mu\text{g/l}$ sample solution [20]. The linear regression equation was calculated using the least squares method,

Table 2
Tolerated limits of foreign ions

Foreign ions	Tolerated limit (g/l)
Sodium chloride, potassium chloride, ammonium(I)	320
Calcium(II), magnesium(II)	16
Barium(II), zinc(II)	2
Manganese(II)	0.8
Cobalt(II), nickel(II), strontium(II), cadmium(II)	0.4
Chromium(III)	0.2
Mercury(II), lead(II)	0.04
Iron(III), tin(IV), molybdenum(VI), tungsten(VI)	0.0004
F^- , Br^- , SO_4^{2-} , CO_3^{2-} , NO_3^- , PO_4^{3-} , acetate	40
Glucose, lactate	80

which was found to be $A = 0.52C + 0.003$ (the regression coefficient is 0.9996), where A is the absorbance of the complex at 542 nm and C is the amount of aluminum (μg) in 250 ml of aqueous solution. According to the procedure, another calibration graph, constructed on various aluminum concentrations plus 80 g of sodium chloride were almost the same as that discussed, in which linear regression equation was $A = 0.52C + 0.004$ with regression coefficient of 0.9998. This indicates highly concentrated sodium chloride in dialysis concentrate has no effect on the results.

3.4. Interferences study

Dialysis concentrates contains many metal and non-metal ions, these will interfere with the proposed methods for the determination of aluminum appearing in the literature. In this work, more than twenty kinds of coexisting ions were investigated by adding a known concentration of each ion to a solution, the complex was then extracted into the ionic liquid using 7.5 ml of the $[C_4tmsim][PF_6]$ and the absorbance was determined at 542 nm, respectively. The tolerated limit of single ion was the concentration that caused an absorbance error less than $\pm 5\%$ in the determination of $3 \mu\text{g}$ of aluminum in 250 ml aqueous solution are given in Table 2. These results clearly indicate that all metal ions studied can be tolerated in considerable amounts. The anion is assumed to compete with DTBSF for complex formation and may lead to interference. The results indicated that the interference of anions is negligible. This is due to large formation constant of Al–DTBSF complex. As the dialysis concentrate contain very high concentration of sodium chloride, it may be interfere with the reaction and following the extraction of the complex, we also examined the effect of sodium chloride on the formation and extraction of Al–DTBSF. It was found that the reaction of aluminum(III) with DTBSF can carry out in the saturated sodium chloride solution, and its absorbance and extraction efficiencies of the complex almost remain unchanged. Thus, proposed method can be applied to determination of trace aluminum in dialysis concentrates.

3.5. Analytical application

Both proposed method and ETAAS described by Milacic and Benedik [1] were applied to determination of aluminum

Table 3
Determination of aluminum in the dialysis concentrates

Sample	Spiked ($\mu\text{g/l}$) ^a	Measured ($\mu\text{g/l}$) ^a		
		Proposed method	ETAAS method	Recovery (%)
Dialysis concentrate I	0.0	3.4 ± 0.20 ($F = 1.23, t = 2.07$)	3.3 ± 0.19	
	3.0	6.5 ± 0.20		103
	6.0	9.2 ± 0.17		97
Dialysis concentrate II	0.0	6.2 ± 0.33 ($F = 3.31, t = 1.79$)	6.5 ± 0.60	
	6.0	12.3 ± 0.24		102
	12.0	18.1 ± 0.33		99.2
Dialysis concentrate III	0.0	5.1 ± 0.16 ($F = 4, t = 2.46$)	4.9 ± 0.32	
	5.0	10.0 ± 0.17		98
	10.0	15.2 ± 0.31		101
Dialysis concentrate IV	0.0	4.7 ± 0.16 ($F = 2.11, t = 2.45$)	4.5 ± 0.11	
	5.0	9.6 ± 0.11		98
	10.0	15.0 ± 0.19		103
Dialysis concentrate V	0.0	5.9 ± 0.53 ($F = 5.46, t = 0.37$)	6.0 ± 0.81	
	6.0	12.1 ± 0.53		103
	12.0	17.9 ± 0.49		100

^a $X \pm (st/\sqrt{n})$ ($n = 5$); the t - and F -values refer to comparison of the proposed method with the atomic absorption spectroscopy method. Theoretical values at 95% confidence limits: $F = 6.39, t = 2.78$.

in dialysis concentrates, the results were listed in Table 3. The results obtained by proposed method were compared with those of the ETAAS using the t -test for accuracy and the F -value for the assessment of precision for five degrees of freedom and a 95% confidence level. The calculated values did not exceed the corresponding theoretical value, this indicated insignificant differences between the results. Since there is no certified reference material for dialysis concentrates, the percentage recovery was also employed for calculation of the method accuracy, the percentage recoveries are between 97% and 103%, in which an about two or three times addition amounts of the initial concentration of the aluminum in the samples were used for standard addition method to test the validity of proposed method and all. These results demonstrated the applicability of the proposed method to determine aluminum in various types of the dialysis concentrates.

3.6. Reuse of ionic liquid

The feasibility of the reuse of ionic liquid $[\text{C}_4\text{tmsim}][\text{PF}_6]$ was studied. As Al–DTBSF complex was formed in a neutral medium, strong acid can easily decompose the complex in aqueous solution. Basing on the fact, various mineral acids such as nitric acid, sulfuric acid and hydrochloric acid were introduced into the ionic liquid containing Al–DTBSF complex to strip aluminum(III). It was found all strong acid could decomposed the complex to aluminum(III) and transferred aluminum into water phase rapidly. In order to test recycling ionic liquid as solvent for liquid/liquid extraction of Al–DTBSF complex, the ionic liquid phase was mixed with equal volume of 0.1 mol/l nitric acid to remove aluminum(III) from the ionic liquid. Then, the ionic liquid was recycled for reuse. This extraction and stripping process were repeated six cycles, in which ETAAS was employed for measure the concentration of aluminum in acidic aqueous

phase after each stripping process. Average extraction efficiency of 99.7% was obtained, the result showed the $[\text{C}_4\text{tmsim}][\text{PF}_6]$ may be reuse to extract Al–DTBSF complex.

4. Conclusion

Proposed method is very simple, selective and sensitive, it can be applied to routinely determination of aluminum at $\mu\text{g/l}$ levels in various complex samples such as commercial dialysis concentrates and environmental water.

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