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**Spectrophotometric Determination of Aluminum (III) Using the Color
Reaction between *o*-Hydroxyhydroquinonephthalein and
Aluminum (III) in the Presence of a Combination
of Surfactants, and Its Application¹⁾**

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The color reaction between *o*-hydroxyhydroquinonephthalein (Qnph) and aluminum(III) was examined in the presence of various surfactants alone or in combination (cationic, anionic, nonionic and amphoteric surfactants) in weakly acidic media. A simple, rapid and sensitive spectrophotometric determination of aluminum(III) (0–2.0 μg of aluminum(III) in a final volume of 10 ml, at 555 nm) was developed by using the Qnph-aluminum(III) complex in a mixed micelle system of zephiramine (tetradecyldimethylbenzylammonium chloride) as a cationic surfactant and Brij 35 (poly(oxyethylene) dodecylether) as a nonionic surfactant. This proposed method (apparent molar absorptivity of $2.1 \times 10^5 \text{ l} \cdot \text{mol}^{-1} \cdot \text{cm}^{-1}$) showed good reproducibility, and the influence of foreign ions was reduced to 1/4–1/10 of that in the previous method with Qnph alone in weakly basic media.

The method was used for the determination of aluminum(III) in pharmaceutical preparations (aspirin aluminum). Further, since aluminum may leach out of aluminum packaging of the kind widely used for instant foods, this proposed method was applied to the assay of aluminum(III) in biological specimens (calf serum and human urine), and in leaching solution used on aluminum foil. The recovery was good, and it was found that the leaching solution contained significant amounts of aluminum(III).

Keywords—spectrophotometry; *o*-hydroxyhydroquinonephthalein; aluminum (III); surfactant; color reaction; zephiramine Brij 35 combination; aluminum leaching; aluminum foil

Aluminum is widely used in the manufacture of domestic utensils such as pans and kettles, and its usage for food packaging or wrapping (as aluminum foil) and in cans or bags for juice, beer, alcoholic products, *etc.* is increasing rapidly.

Recently, there has been increasing interest in the amounts of aluminum(III) in biological samples because of the probable relationship between “dementia” and aluminum(III).^{2,3)} Thus, a highly sensitive, selective and simple method for the determination of aluminum(III)

in biological and environmental samples is required.

Generally, flameless atomic absorption analysis using a graphite atomizer or inductively coupled plasma atomic emission spectroscopy (ICP-AES) have been used, but these methods lack simplicity in comparison with spectrophotometry. Many methods for the spectrophotometric determination of aluminum(III) with oxine, aluminone, chromazurol S, xylenol orange (XO), alizarin S, *o*-hydroxyhydroquinonephthalein (Qnph), *etc.* have been reported,^{4–13)}

but most of them are complicated and/or not sufficiently sensitive. On the other hand, color reaction system involving various dyes such as XO, pyrogallol red, pyrocatechol violet, pyrogallolphthalein and Qnph, various metal ions and surfactant [cationic—hexadecyltrimethylammonium chloride (HTAC), zephiramine (tetradecyl dimethylbenzylammonium chloride, Zp); nonionic—Brij 35 (poly(oxyethylene) dodecylether); anionic—sodium dodecyl sulfate (SDS); amphoteric surfactant—*N*-lauroylsarcosine(LS)] have been discussed, and sensitive and simple spectrophotometric determinations of various metal ions using these color reaction systems in micellar media have been proposed.^{14–21)} The combination of two or three surfactants in these color reaction systems or spectrophotometric determinations may be superior to the use of a single surfactant.^{22–27)}

In the present work, firstly, the coloring reaction systems based on Qnph as a xanthene dye, aluminum(III) and various surfactants alone or in combination in weakly acidic media were systematically examined. Based on the results, a simple, rapid and highly sensitive spectrophotometric determination of aluminum(III) with Qnph is proposed in the presence of Zp (a cationic surfactant) and Brij 35 (a nonionic surfactant) as a mixed micelle system in weakly acidic media. This method was applied to the determination of micro amounts of aluminum(III) in biological samples such as calf serum or human urine, and further to the assay of aluminum leached from aluminum foil into leaching solutions. The application of the method to assay of pharmaceutical preparations containing aluminum is also discussed.

Experimental

Apparatus and Reagents—Absorption spectra and absorbance were measured on Shimadzu UV-

200, 240 and 160 recording spectrophotometers with 1.0-cm silica cells. Hitachi-Horiba F-7 AD and M-5 glass electrode pH meters were used for the pH measurements. Values obtained with the three spectrophotometers and with the two pH meters were identical. An ICP atomic emission spectrophotometer (Shimadzu ICPS-1000 V) was used for the determination of aluminum(III) in biological and pharmaceutical samples.

A $1.0 \times 10^{-3} \text{ M}$ ($= \text{mol} \cdot \text{dm}^{-3}$) solution of Qnph was prepared according to the previous reports.^{13,21)} A stock solution of aluminum(III) ($1.0 \times 10^{-2} \text{ M}$) was prepared according to the previous method¹³⁾ by dissolving aluminum (99.999%, Mitsuwa Chemicals) in concentrated-hydrochloric acid, and a working solution of aluminum(III) ($5.0 \times 10^{-4} \text{ M}$) was made by dilution of the stock solution. A $1.0 \times 10^{-2} \text{ M}$ Zp solution was prepared by dissolving Zp (Dojindo Lab.) in water. A 1/15 M sodium dihydrogen phosphate-dipotassium hydrogen phosphate solution (Sørensen buffer) was used for the pH adjustment.

All other reagents and materials were of analytical grade and were used without further purification.

Doubly distilled or deionized water was used.

Standard Procedure—To a series of solutions containing 0–2.0 μg of aluminum(III), 1.0 ml of a $1.0 \times 10^{-2} \text{ M}$ Zp solution, 0.5 ml of a 2.0% Brij 35 solution, 2.0 ml of a buffer (pH 5.5) solution and 0.75 ml of a $1.0 \times 10^{-3} \text{ M}$ Qnph solution were added, mixed and diluted to volume in a 10-ml calibrated flask. The mixture was heated at 60°C for 20 min, and cooled in water for 10 min (solution A) together with reference solution (solution B). The absorbance of solution A was measured at 555 nm against solution B.

Results and Discussion

Color Reaction in the Presence of Surfactant

Table I shows the results obtained with various surfactants alone or in combination, at room temperature or at 60°C.

In the color reaction systems between Qnph and aluminum(III) at room temperature, the presence of a cationic surfactant such as *N*-

TABLE I. Effects of Surfactants on the Color Reaction between Qnph and Aluminum (III)

Cation	Surfactants			$\lambda_{\max}$ (nm)	Absorbance at $\lambda_{\max}$	
	Nonion	Anion	Amphoter		at 60°C	at room temp.
Zp	Brij 35	—	—	555	0.650	0.450
—	—	—	—	515	^a	0.130
—	Brij 35	—	—	515	0.170	0.130
HTAC	Brij 35	—	—	555	0.460	0.320
HTAC or Zp	—	—	—	555	^a	0.330
HTAC+Zp	—	—	—	555	^a	0.330
—	—	SDS	—	520	^a	0.158
—	—	—	LS ^b	520	^a	0.065
—	Brij 35	—	LS	520	0.065	0.065
—	Brij 35	SDS	—	518	0.218	0.160
—	Tween+Brij 35	—	—	515	0.158	0.130

Al (III), $0.8\mu\text{g}/10\text{ ml}$; Qnph, $7.5\times 10^{-5}\text{ M}$; cationic surfactant, $1.0\times 10^{-3}\text{ M}$; nonionic, anionic and amphoteric surfactant, 0.5 ml of 2.0% surfactant solution/ 10 ml ; pH 5.5. Reference, reagent blank.

^a) Unstable and/or imperfect micelles. ^b) *N*-lauroylsarcosine.

hexadecylpyridinium chloride (HPC) was most effective, but the use of a nonionic surfactant such as Brij 35 or an anionic surfactant such as SDS alone was not effective. On the other hand, in the color reaction systems between Qnph and aluminum(III) at about 60°C , the combination of Zp as a cationic surfactant and Brij 35 as a nonionic surfactant was most effective (in this case, the complex may be Qnph-aluminum(III)-Zp ternary complex).

Figure I shows the visible-region absorption spectra of Qnph-aluminum(III) solution (solution A') and Qnph solution (solution B') in the presence of Brij 35 alone, and those of Qnph-aluminum(III) solution (solution A) and Qnph solution (solution B) in the coexistence of Zp and Brij 35 (mixed surfactant system) at pH 5.5.

Although the Qnph-aluminum(III) solution in the presence of Zp alone, even at a concentration as large as the critical micelle concentration (cmc , $3.7\times 10^{-4}\text{ M}$), formed a precipitate of ternary complex in water at about 60°C , the complex was completely solubilized in the water phase in the coexistence of a nonionic surfactant such as Brij 35, and the solution was very stable. Moreover, the molar

Fig. 1. Absorption Spectra of Qnph and Qnph-aluminum (III) Solutions in the Presence of Brij 35 Alone, and in the Presence of Zp and Brij 35 (Mixed Surfactants) at pH 5.5

Qnph, $2.0\times 10^{-5}\text{ M}$; aluminum (III), $2.0\times 10^{-5}\text{ M}$; Brij 35, 0.5 ml of 2.0% Brij 35 soln./ 10 ml ; Zp, $1.0\times 10^{-3}\text{ M}$; reference, water.

- : Qnph solution in the presence of Brij 35 (solution B').
-x..... : Qnph-aluminum (III) solution in the presence of Brij 35 (solution A').
- : Qnph solution in the presence of Zp and Brij 35 (solution B).
- x——— : Qnph-aluminum (III) solution in the presence of Zp and Brij 35 (solution A).

Fig. 2. Molar Ratio of Qnph to Aluminum(III) in the Ternary Complex Examined by the Continuous Variation Method in the Presence of Zp and Brij 35

[Qnph+aluminum(III)]= 5.0×10^{-5} M; pH, 5.5; Zp, 1.0×10^{-3} M; Brij 35, 0.5 ml of 2.0% Brij 35 soln./10 ml; reference, reagent blank.

ratio of Qnph to aluminum(III) in the color reaction system was found to be 2:1 by the continuous-variation and molar-ratio methods, irrespective of the presence of Brij 35 or Zp alone, or both. Figure 2 shows typical results for the combination of surfactants.

The sharp color reaction in the mixed micelle system may be based on an effect similar to that in anionic and nonionic mixed micelles.²⁸⁾

Generally, for spectrophotometric determinations of metal ions based on the formation of a sparingly soluble precipitate, solvent extraction or preconcentration by using a membrane filter²⁹⁾ has been used, but these methods lack simplicity and rapidity. Therefore, we attempted to develop a simple and sensitive spectrophotometric determination of aluminum(III) in the water phase by using a micelle-solubilized ternary complex.

As shown in Fig. 1, the ternary complex between Qnph and aluminum(III) in a mixed micelle system gave a larger absorbance at

555 nm, with better reproducibility, than the Qnph-aluminum(III) binary complex solution in the presence of Brij 35 or Tween 20 alone. The magnitude of the difference of absorbances at 555 nm between solutions A and B was constant and proportional to the concentration of aluminum(III). Accordingly, all further work was carried out in a mixed micelle system of cationic surfactant plus nonionic surfactant.

Conditions for the Spectrophotometry of Aluminum(III)

The maximum and almost constant difference of absorbance between solutions B and A was obtained in the pH range of 5.0–7.8 using 1.0–3.0 ml of a $1/15$ M phosphate buffer solution (Sørensen buffer) for pH adjustments, as shown in Fig. 3.

Among various cationic surfactants examined, Zp was found to be the best in the presence of Brij 35. The maximum and constant absorbance intensity could be obtained with 0.5–3.0 ml of a 1.0×10^{-2} M Zp solution in the final volume of 10 ml. Further, as shown in Table II, the use of 2.0% Brij 35 solution (0.2–1.0 ml/10 ml) as a nonionic surfactant was found

Fig. 3. Effect of pH on the Color Reaction between Qnph and Aluminum(III)

Aluminum(III), $0.8 \mu\text{g}/10 \text{ ml}$; Qnph, 7.5×10^{-5} M; Zp, 1.0×10^{-3} M; Brij 35, 0.5 ml of 2.0% Brij 35 soln./10 ml; reference, reagent blank (solution B).

TABLE II. Effect of Cationic Surfactant and Nonionic Surfactant on the Color Reaction between Qnph and Aluminum (III)

Surfactants		$\lambda_{\max}$ (nm)	Absorbance at $\lambda_{\max}$	
			at 60°C	at room temp.
Zp	+Brij 35	555	0.650	0.450
Zp or HTAC	—	555	^{b)}	0.330
—	Brij 35	515	0.170	0.130
HTAC	+Brij 35	555	0.460	0.320
HPC	+Brij 35	555	0.430	0.305
HPB ^{a)}	+Brij 35	555	0.410	0.305
Zp	+Tween 20	555	0.488	0.340
Zp	+PVA	553	0.435	0.310
Zp	+PVP	555	0.488	0.340
Zp	+HTAC	555	^{b)}	0.330

Al (III), 0.8 $\mu\text{g}/10\text{ ml}$; Qnph, $7.5 \times 10^{-5}\text{ M}$; cationic surfactant, $1.0 \times 10^{-3}\text{ M}$; nonionic surfactant, 0.5 ml of 2.0% nonionic surfactant solution/10 ml; pH 5.5. Reference, reagent blank.

a) hexadecylpyridinium bromide.

b) Unstable and/or imperfect micelles.

to be the best.

In this system, the nonionic surfactant may act as an auxiliary dispersant of the Qnph-aluminum(III)-Zp ternary complex.

The effect of Qnph concentration was examined by measuring the absorbance at 555 nm with 0.8 μg of aluminum(III) in a final volume of 10 ml. The maximum and almost constant absorbance was obtained with 0.3 to 1.2 ml of $1.0 \times 10^{-3}\text{ M}$ Qnph solution.

At room temperature, more than 90 min was required for completion of the color reaction. Though the Qnph-aluminum(III)-Zp ternary complex without Brij 35 was precipitated by heating at 60°C, the ternary complex solution

in the presence of Brij 35 was very stable, and the results showed good reproducibility. The effects of time and temperature were examined by heating solutions A and B for 10–60 min at various temperatures. The maximum and constant absorbance was obtained by heating these solutions at 60°C for 20 min, and then cooling in water for 10 min.

The calibration curve was linear in the range of 0–2.0 $\mu\text{g}/10\text{ ml}$ of aluminum(III) with a Sandell sensitivity of 0.00013 $\mu\text{g}/\text{cm}^2$ aluminum(III). The apparent molar absorptivity was calculated to be $2.1 \times 10^5\text{ l} \cdot \text{mol}^{-1} \cdot \text{cm}^{-1}$.

The reproducibility for 1.0 $\mu\text{g}/10\text{ ml}$ of aluminum(III) (5 experiments) was good at 0.4%. In Table III, these results are shown together with those obtained by the method reported previously.¹³⁾

As summarized in Table IV, small amounts (less than about 5-fold) of iron(III), copper(II), zinc(II), bismuth(III), tin(IV), thorium(IV) and zirconium(IV) caused positive errors, and cobalt(II), phosphate and sulfide ions caused negative errors. On the other hand, the coexistence of nickel(II), mercury(II), cadmium(II), citrate, tartrate, salicylate, cyanide ions and creatinine in 10–50-fold excess over aluminum(III) did not interfere. Large amounts of magnesium(II), calcium(II), carbonate, fluoride, chloride ions, urea, and triethanolamine did not interfere. The effects of iron(III), copper(II), cobalt(II), and zinc(II) could be depressed by addition of potassium cyanide solu-

TABLE III. Comparison of Analytical Results Obtained by the Proposed and Previous Methods for Spectrophotometric Determinations of Aluminum (III)

Method	pH range	Temp. (°C)	Surfactants	Calibration ($\mu\text{g}/10\text{ ml}$)	Molecular interference by foreign ions	
					extinction coefficient	relative to previous method
Proposed method	5.0–7.8 (5.5)	60	Brij 35+Zp	0–2.0	2.1×10^5	1/4–1/10
Previous method ¹³⁾	8.2–9.2 (8.8)	15–25	—	0–3.2	1.4×10^5	1

TABLE IV. Effect of Foreign Ions on Determination of Aluminum (III) by the Proposed Method

Foreign substance	Added		Mole ratio foreign ^{a)/} Al (III)	A ₅₅₅
	as	μg/10 ml		
-	-	-	-	0.795
Bi(III)	Nitrate	4.2	1/2	0.801
		8.4	1	0.904
Fe(III)	Sulfate	4.5	2	0.800
		11.2	5	0.957
Sn(IV)	Sulfate	5.9	1	0.810
Th(IV)	Nitrate	9.3	1	0.799
		18.6	2	0.900
Zr(IV)	Nitrate	0.7	1/5	0.790
Cu(II)	Nitrate	0.5	1/5	0.800
Ni(II)	Nitrate	23.5	10	0.798
		47.0	20	0.800
Co(II)	Nitrate	23.6	10	0.782
		47.1	20	0.770
Zn(II)	Nitrate	13.1	5	0.800
		26.2	10	0.940
Cd(II)	Nitrate	13.2	3	0.798
Hg(II)	Nitrate	16.0	2	0.795
Mg(II)	Nitrate	91.1	100	0.798
CN ⁻	Potassium	13.0	10	0.790
Cl ⁻	Sodium	2193.8	1000	0.795
F ⁻	Sodium	72.0	100	0.795
C ₂ O ₄ ²⁻	Sodium	0.4	1/10	0.735
SO ₃ ²⁻	Sodium	4.0	1	0.780
CO ₃ ²⁻	Sodium	1125.0	500	0.795
H ₂ PO ₄ ⁻	Potassium	3.6	1	0.795
		17.8	5	0.715
Citrate	Sodium	2700.0	250	0.795
Tartrate	Sodium	218.4	50	0.788
Salicylate	Sodium	258.7	50	0.795
Urea		2252.3	1000	0.795
Creatinine		252.8	50	0.795
Albumin		10.0	-	0.782
Triethanolamine		2797.5	500	0.795

Al (III) taken, 1.0 μg/10 ml; Zp, 1.0 × 10⁻³ M; Qnph, 7.5 × 10⁻⁵ M; Brij 35, 0.5 ml of 2.0% Brij 35 soln./10 ml; pH 5.5. Reference, reagent blank.

a) Foreign substance.

tion as a masking agent. The influence of foreign ions on the proposed method was 1/4—1/10 of that in the previous method⁽¹³⁾ using the Qnph-aluminum(III) binary complex at pH 8.8.

Application

The procedure was applied to the assay of aluminum(III) in aluminum acetylsalicylate,

and the experimental results were in good agreement with the values measured by the JP X method⁽³⁰⁾ and ICP-AES. The recoveries of aluminum(III) added to the pharmaceutical preparations were satisfactory by the proposed method, being about 99.6%. The recoveries of aluminum(III) added to calf serum (Nakarai Chemical Co., Ltd.) and human urine were also examined. Calf serum (5.0 ml) spiked with up to 80 μg of aluminum(III) was mixed with 1.0 ml of 1.0 × 10⁻¹ M aqueous sodium fluoride and 10.0 ml of 7.5% trichloroacetic acid as a deproteinizing agent, and the mixture was centrifuged, neutralized with sodium hydroxide solution, and diluted with an equal volume of water. As a sample, 1.0 ml of this diluted solution or 1.0—2.0 ml of human urine spiked with up to 2.0 μg of aluminum(III) was taken. The aluminum(III) content was determined according to the standard procedure. Although the recovery of aluminum(III) added to calf serum was unsatisfactory in the absence of sodium fluoride (recovery=47.0%, *n*=5), it was considered that the binding of aluminum(III) to protein was prevented by the addition of sodium fluoride solution. The results were as follows: recovery=100.01%, coefficient of variation (C.V.)=0.7% for serum, and recovery=100.03%, C.V.=0.5% for urine (*n*=5).

In order to examine the occurrence of leaching of aluminum from simple packages of aluminum foil or sheet, samples of aluminum foil were allowed to stand in aqueous leaching solutions at various pH values at room temperature for 30 min, 60 min, or 48 h, or in the boiling solutions for 30 min, or 60 min. The results (Table VI) indicate that leaching of aluminum may occur under every-day conditions and may represent an important source of human intake of aluminum.

Although further investigation is necessary,

TABLE V. Determination of Aluminum (III) in Pharmaceutical and Biological Samples

Samples	Amounts of aluminum (III)					Recovery of proposed method	
	Calcd. (%)	Found ^{a)}					
		Method					
		proposed (%)	previous ¹³⁾ (%)	JP X (%)	ICP-AES ^{b)} (%)	Average ^{a)} (%)	C.V. (%)
Aluminum							
acetylsalicylate	6.0—7.0	6.98	6.85	6.82	6.95	99.58	0.9
Prepared sample ^{c)}	6.00	6.24	6.15	6.19	6.12	99.63	0.8
Serum ^{d)}						100.01	0.7
Human urine ^{e)}						100.03	0.5

a) Average of 5 determinations. b) Wavelength, 2575.1 Å. c) Acetylsalicylate-aluminum mixture. d) 1.0 ml of a 1.0×10^{-3} M aluminum (III) solution was taken in 5.0 ml of calf serum. e) 1.0 μ g of aluminum (III) was taken in 2.0 ml of human urine.

TABLE VI. Results of Assay of Aluminum (III) in Leaching Solution

Water	Leaching			Found, aluminum (III)			Recovery ^{a)}	
	pH	Heating Temp.	Time (min)	Proposed method		ICP-AES ^{b)} (ppm)	-	masking ^{c)}
				- (ppm)	masking ^{c)} (ppm)			
Tap water	6.8	Boiling	30	1.3	1.3	-	94.12	95.38
	6.8	Boiling	60	7.0	7.0	-	93.21	94.55
Borate buffer ^{d)}	3.4	Boiling	30	45.4	-	44.9	99.51	-
water	3.4	Boiling	60	92.0	-	91.1	100.13	-
Citrate ^{e)}	3.3	Ambient	30	0.5	-	-	100.05	-
water	3.3	Ambient	2880	0.9	-	-	101.98	-
	3.3	Boiling	30	11.4	-	11.0	101.40	-
Deionized	6.6	Boiling	30	-	-	-	100.55	-
water	6.6	Boiling	60	-	-	-	100.23	-
Ammonia buffer ^{f)}	11.3	Boiling	30	18.1	-	17.6	94.71	-
water	11.3	Boiling	60	45.6	-	44.7	92.06	-

a) Recovery test: aluminum (III) taken, 0.3 μ g/ml. b) Wavelength, 2575.1 Å. c) Masking agent: KCN solution. d) 100 ml of 1/10 M Borate-HCl buffer. e) Citric acid, added 2.1 μ g/ml. f) 100 ml of 1/10 M NH_4Cl - NH_4OH buffer. Sample solution: 5 g of aluminum foil was leached with 100 ml of water under various conditions.

the proposed method may be applied for a simple and rapid spectrophotometric determination or detection test of aluminum(III) in various samples, such as canned juice, canned beer, foods packaged with aluminum sheet or foil, and environmental and biological samples.

Conclusion

The color reaction between aluminum(III) and Qnph in the presence of surfactants alone or in combination (mixed micelle system) was examined in weakly acidic media.

Although the Qnph-aluminum(III)-Zp ternary complex solution at 60°C was unstable

and precipitated in the absence of nonionic surfactants, this ternary complex was stable in the presence of Brij 35, as a nonionic surfactant, and showed high and constant absorbance.

By using this Qnph-aluminum(III)-Zp ternary complex in the presence of a Zp and Brij 35 mixed micelle system, a simple, rapid and highly sensitive spectrophotometric determination of aluminum(III) was established. The calibration curve was linear in the concentration range of 0—2.0 μ g/10 ml for aluminum(III) at pH 5.5 with an apparent molar absorptivity of $2.1 \times 10^5 \text{ l} \cdot \text{mol}^{-1} \cdot \text{cm}^{-1}$. The

sensitivity was about 2–10 times greater than that of the reported methods^{2,3,6,31–33} using aluminone ($\epsilon=1.4 \times 10^4$), XO ($\epsilon=2.1 \times 10^4$), or chromazurol S-HTAC ($\epsilon=1.1 \times 10^5$), and the proposed method showed good reproducibility (0.4% for 1.0 μg of aluminum(III)).

The proposed method was then applied to the assay of aluminum(III) in biological samples such as calf serum, human urine and pharmaceutical samples, and the recoveries were shown to be satisfactory. Moreover, it was found that significant amounts of alumi-

num(III) were leached from aluminum foil into aqueous leaching solutions of various pH values within 30 min at room temperature or on boiling.

In conclusion, the above-mentioned method is simple, selective and highly sensitive, and should be useful for detection or determination of aluminum(III) in various samples. We consider that particular attention should be paid to establishing the possible significance of human intake of aluminum leached into food and drink from various kinds of aluminum packaging (including cans) and utensils.

References

- 1) This paper is Part LXIV of a series entitled "Application of Xanthene Derivatives for Analytical Chemistry," Part LXIII, *Chem. Pharm. Bull.*, **35**, 5004 (1987). A part of this work was presented at the 35th Annual Meeting of the Analytical Chemistry Society of Japan, Okayama, October, 1986.
- 2) D.R. Roccer, S.S. Frishan, and A.J. Dalton, *Science*, **180**, 511 (1973).
- 3) K. Tsunoda, *Bunseki*, **1985**, 119.
- 4) K. Motoshima, *Nippon Kagaku Kaishi*, **76**, 903 (1955).
- 5) E.B. Sandell, "Colorimetric Determination of Traces of Metal," 3rd ed., Interscience, New York, 1959, p.575.
- 6) Y. Shijo and T. Takeuchi, *Bunseki Kagaku*, **17**, 61 (1968).
- 7) M. Otomo, *Bull. Chem. Soc. Jpn.*, **36**, 809 (1963).
- 8) H. Onishi "Photometric Determination of Traces of Metals," Part IIA, John Wiley and Sons, New York, 1986, p.13.
- 9) C.L. Barton, *Anal. Chem.*, **20**, 1068 (1948).
- 10) U.T. Hill, *Anal. Chem.*, **28**, 1419 (1956).
- 11) N.A. Agrinskaya, *Zavod. Lab.*, **23**, 279 (1957).
- 12) N. Jean, *Anal. Chim. Acta*, **10**, 526 (1954).
- 13) Y. Fujita, I. Mori, and T. Enoki, *Bunseki Kagaku*, **24**, 253 (1975).
- 14) I. Mori, Y. Fujita, K. Fujita, Y. Koshiyama, H. Kawabe, T. Tanaka, and R. Munakata, *Bull. Chem. Soc. Jpn.*, **59**, 958 (1986).
- 15) Y. Shijo and T. Takeuchi, *Bunseki Kagaku*, **22**, 1341 (1973).
- 16) E.A. Morgan, E.S. Roshikskaya, and N.A. Vlosolv, *Zh. Anal. Khim.*, **30**, 1384 (1975).
- 17) K. Ueno, *Bunseki Kagaku*, **20**, 736 (1971).
- 18) B.W. Bailey, J.E. Chester, R.M. Dagnal, and T.S. West, *Talanta*, **15**, 1359 (1968).
- 19) D. Thorburn Burns, and D. Dadgaov, *Analyst* (London), **105**, 751 (1980).
- 20) T. Yotsuyanagi, H. Hoshino, and S. Igarashi, *Bunseki*, **1985**, 496.
- 21) I. Mori, Y. Fujita, K. Fujita, T. Tanaka, Y. Koshiyama, and H. Kawabe, *Chem. Pharm. Bull.*, **34**, 4836 (1986).
- 22) D. Wang, J. Chen, J. Bai, and Q. Pan, *Huaxue Shiji*, **6**, 139 (1984); *idem*, *Anal. Abstr.*, **47**, 6 B 110 (1985).
- 23) Xu. Quinbeng, *Huaxue Shiji*, **4**, 214 (1982); *idem*, *Anal. Abstr.*, **45**, 6 B 146 (1983).
- 24) Q. Xu, Y. Peng, and S. Lin, *Fenxi Huaxue*, **11**, 485 (1983); *idem*, *Anal. Abstr.*, **45**, 6 B 146 (1983).
- 25) S. Zheng, *Fenxi Huaxue*, **11**, 684 (1983); *idem*, *Anal. Abstr.*, **46**, 9 B 146 (1984).
- 26) Y. Zheng and S. Zhon, *Fenxi Huaxue*, **11**, 210 (1983); *idem*, *Anal. Abstr.*, **46**, 3 B 53 (1984).
- 27) Wu. Quianfeng, *Talanta*, **32**, 507 (1985).

-
- 28) M. Abe, T. Kakihara, H. Uchiyama, and K. Ogino, *Yukagaku*, **36**, 135 (1987).
 - 29) I. Mori, Y. Fujita, H. Kawabe, and K. Fujita, *Chem. Pharm. Bull.*, **34**, 902 (1986).
 - 30) "Commentary of the Japanese Pharmacopoeia, X" C-56, Hirokawa Publishing Co., Tokyo, 1981.
 - 31) M. Otomo, *Bull. Chem. Soc. Jpn.*, **36**, 809 (1963).
 - 32) J.A. Corbett and B.D. Guerin, *Analyst* (London), **91**, 490 (1966).
 - 33) P. Pkalus, *Anal. Chim. Acta*, **22**, 57 (1965).