

**INTERNATIONAL UNION OF PURE
AND APPLIED CHEMISTRY**

ANALYTICAL CHEMISTRY DIVISION

COMMISSION ON MICROCHEMICAL TECHNIQUES AND TRACE ANALYSIS

**GENERAL ASPECTS OF TRACE
ANALYTICAL METHODS—III.**

CONTAMINATION IN TRACE ANALYSIS

Prepared for publication by

A. MIZUIKE

Nagoya University, Japan

and

M. PINTA

Office de la Recherche Scientifique et
Technique Outre-Mer, Bondy, France

PERGAMON PRESS

OXFORD · NEW YORK · PARIS · FRANKFURT

GENERAL ASPECTS OF TRACE ANALYTICAL METHODS.
III - CONTAMINATION IN TRACE ANALYSIS.

Project Leaders : Atsushi Mizuike* and M. Pinta**

* Faculty of Engineering, Nagoya University, Chikusa-ku, Nagoya 464, Japan. ** Laboratoire de Spectrographie, Office de la Recherche Scientifique et Technique Outre-Mer, 70-74 Route d'Aulnay, F-93140 Bondy, France

Abstract - The present status of the contamination problem in trace analysis has been reviewed with the aid of a questionnaire and by consultation of the literature.

INTRODUCTION

During chemical analysis, appreciable amounts of the elements or compounds being determined are often introduced into the sample from various external sources. These sources include the laboratory atmosphere, the reagents and the apparatus used, and the analyst performing the analysis. This phenomenon, which is simply called "contamination", is one of the most troublesome problems in trace analysis at or below the low ppm level, because it vitiates the reliability of analytical results.

It is common practice to subtract a blank value from the analytical values. However, when the blank value is comparable to, or larger than, the amount of the element being determined, this method is unsatisfactory, because (1) a result based on the difference between two signals of about the same size is generally inaccurate, and (2) the contamination is variable except for that due to reagents. To obtain accurate analytical results, a blank value should be much lower, say at least by one order of magnitude, than the amount of the element being determined. Therefore, in trace analysis, every effort is made to reduce contamination. Contamination and loss of the trace element may accidentally compensate each other. The loss should be separately evaluated by the radioactive tracer technique.

The project "Contamination in Trace Analysis" was proposed and accepted at the Commission meeting in Munich (August 1973). Its object was to review the present status of the contamination problem with the aid of a questionnaire and by consultation of the literature. Copies of the questionnaire were sent to approximately 230 laboratories throughout the world, and replies were received during the period December 1974 to November 1975 from 96 laboratories dealing with trace analysis at the low ppm level or lower (see Table I). Almost all of them (93 laboratories) replied that they had encountered contamination problems. The stages where contamination occurred are summarized in Table II, which shows that the decomposition step is most critical in this respect.

AIRBORNE CONTAMINATION

The laboratory atmosphere usually contains various kinds of inorganic and organic gases as well as aerosols (solid and liquid particulate matter) which may contaminate the sample. These contaminants come from natural and man-made sources outside the laboratory and from reagents, apparatus, analysts, and everything in the laboratory. Accordingly, kinds and quantities of the contaminants depend on the circumstances. In principle, any element may be present. For example, non-conditioned air contained about 200 μg dust per m^3 and the following elements were found in the dust: Ca 10 %, Si 5 %, Fe 3 %, Al 1.5 %, Na 1.5 %, Mg 1 %, K 1 %, Cu 0.5 %, Mn 0.5 %, Tl 0.2 %, traces of Ge, Pb, Ba, As, etc. (1). In another chemical laboratory the dust contained the following metals (in %): Ag 0.15, Bi 0.07, Ca 2, Cu 0.5, Fe 2, K 2, Mg 2, Mn 0.1, Na 2, Ni 0.01, Pb 0.15, Si 2, Sn 0.02, V 0.03, Ti 0.2, and Zn 0.5 (2). The lead contamination due to automotive exhaust is notorious.

Table I. Trace analytical laboratories that returned the questionnaire.
(Total = 96. Number of laboratories in parentheses.)

Geographical distribution	Austria (1) Belgium (1) Canada (2) France (3) Germany (12) Hungary (2) Israel (1) Italy (1) Japan (35) Spain (2) Switzerland (1) UK (2) USA (26) USSR (1) Yugoslavia (1) Unidentified (5)
Purpose of analysis	Pollution (53) Materials research (33) Geochemical (22) Industrial (13) Medical (11)
Materials analyzed	Waters (57) Rocks, soils, and minerals (36) Industrial products (26) Metals and alloys (23) Biological media (23) Air (20) Plant materials (12)
Trace elements determined	Ag (5) Al (8) As (19) Au (1) B (7) Ba (3) Be (4) Bi (7) Br (2) C (10) Ca (11) Cd (41) Cl (7) Co (15) Cr (24) Cs (1) Cu (35) F (5) Fe (20) Ga (2) H (6) Hg (30) I (5) In (1) K (5) Li (1) Mg (5) Mn (20) Mo (6) N (9) Na (5) Ni (17) O (5) P (2) Pb (43) Pd (1) Rb (6) S (9) Sb (7) Se (7) Si (2) Sn (2) Sr (6) Ta (1) Te (1) Th (5) Ti (3) Tl (3) U (6) V (9) Zn (36) Rare earths (4) Many (13)
Determination methods used	Atomic absorption and fluorescence spectrometry (68) Spectrophotometry (33) Polarography and stripping voltammetry (21) Emission spectroscopy (17) Activation analysis (16) X-ray fluorescence (14) Mass spectrometry (13) Fluorimetry (9) Electrochemical Methods (9) Chromatography (7) Gas analysis (6) Flame photometry (5)
Concentration techniques used	Solvent extraction (63) Ion exchange (30) Precipitation and coprecipitation (23) Evaporation (15) Electrolysis (6) Miscellaneous (8) None (14)

Table II. Stages where contamination occurred.

Stages	Number of laboratories (Total = 96)	%
Collection of sample	44	46
Storage of sample	44	46
Comminution	17	18
Sieving	10	10
Filtration	17	18
Desiccation	11	11
Decomposition of sample	62	65
Separation	53	55
Instrumental measurement	31	32

Ammonia, hydrochloric acid, and mercury vapor often exist in the air around laboratories. According to the completed questionnaires, airborne contaminants as listed in Table III have been observed in various laboratories. Other airborne contaminants reported in the literature include Au, Br, F, Ga, In, Mo, P, and Sb (3).

To minimize airborne contamination, various facilities and apparatus are being used in many laboratories as shown in Table IV.

"Clean rooms" and "clean hoods" (4-6) are flushed with particulate-free air filtered through high-efficiency particulate air (HEPA) filters. The combined use of laminar airflow (especially vertical) and positive-pressure principles is very effective. Thus the class 100 cleanliness level [less than 100 particles per cubic foot (1 cubic ft \approx 28 liter) of air larger than 0.5 μm and no particle larger than 5 μm] can be attained. Particles smaller

Table III. Airborne contaminants.

Contaminants	Number of laboratories	Concentration in air or fallout
Al	4	1×10^{-7} g/m ³
As	1	
B	2	1×10^{-7} g/(50 cm ² ·day), 1×10^{-7} g/(100 cm ² ·day)
C	2	
Ca	8	1×10^{-7} g/m ³ , 1×10^{-6} g/(100 cm ² ·day)
Cd	4	6×10^{-9} g/(120 cm ² ·6 days)
Cl	4	1×10^{-7} g/(10 cm ² ·h)
Co	1	
Cr	3	
Cu	4	1×10^{-8} g/(120 cm ² ·6 days) (in clean hood)
Fe	14	1×10^{-7} g/m ³ , 1×10^{-6} g/(100 cm ² ·day), 4×10^{-7} g/(100 cm ² ·day), 4×10^{-8} g/(120 cm ² ·6 days), 1×10^{-6} g/(100 cm ² ·day)
Hg	4	$(1 - 5) \times 10^{-9}$ g/(cm ² ·day)
Mn	2	
Na	4	1×10^{-7} g/m ³ , 4×10^{-7} g/(100 cm ² ·day)
Ni	1	
Pb	14	1×10^{-7} g/(100 cm ² ·day), 1×10^{-7} g/(m ² ·day), 4×10^{-12} g/(25 cm ² ·h) (in evaporation chamber), 6×10^{-8} g/(100 cm ² ·day), 8×10^{-9} g/(120 cm ² ·6 days), 1×10^{-7} g/(100 cm ² ·day)
S	4	
Se	1	
Si	7	1×10^{-7} g/m ³ , 3×10^{-8} g/(100 cm ² ·h), 1×10^{-7} g/(100 cm ² ·day)
Sn	1	
Ti	3	
Zn	3	
NH ₃	2	2×10^{-6} g/day
SO ₂	1	
HCHO	1	

than 0.3 μ m as well as inorganic and organic gases are not removed by HEPA filters. Activated charcoal filters and scrubbers are effective to remove these contaminants. Materials used for ceilings, walls, floors, and fixtures in clean rooms should be properly selected to prevent contamination due to corrosion, flaking, and abrasion. Special garments for analysts and frequent cleaning of the rooms are also required to maintain the clean environment. The relative humidity must be kept above 30 % to preclude electrostatic charges, which may cause serious problems in particulate attraction on apparatus.

Glove boxes, evaporation chambers (7) flushed with purified air or nitrogen,

Table IV. Facilities and apparatus employed for minimizing airborne contamination.

Facilities or apparatus	Number of laboratories (Total = 96)	%
Laminar-flow "clean room"	17	18
Nonlaminar-flow "clean room"	20	21
Laminar-flow "clean hood or bench"	24	25
Glove box	23	24
Small chamber supplied with purified air, nitrogen, etc. for evaporation, decomposition of sample, etc.	30	31
Hermetically sealed vessel for decomposition of sample	37	39

Table V. Reduction of airborne contamination by use of special facilities and apparatus.

Experiment	Analytical results			
	In ordinary laboratory		In clean room, etc.	
Analysis of particulates in air (10)	Fe	0.2	$\mu\text{g}/\text{m}^3$	In clean room
	Cu	0.02	"	Fe 0.001 $\mu\text{g}/\text{m}^3$
	Pb	0.4	"	Cu 0.002 "
	Cd	0.002	"	Pb 0.0002 "
				In class 100 clean hood
				Fe 0.0009 $\mu\text{g}/\text{m}^3$
				Cu 0.007 "
				Pb 0.0003 "
			Cd 0.0002 "	
Evaporation of 500 ml of 6M hydrochloric acid in a Teflon beaker over a period of 8 days (11)	Pb	4.07, 2.32	μg	In evaporation chamber
				Pb 1.13 μg
				In clean air laboratory
				Pb 0.44 μg
Preparation of anhydrous sodium carbonate from purified 25 % sodium carbonate solution (Cr<0.007, Cu 0.010, Fe 0.010, Ni<0.010 ppm in sodium carbonate) (5)	Cr	0.037	ppm	In class 100 clean hood
	Cu	0.024	"	Cr 0.004 ppm
	Fe	0.080	"	Cu 0.012 "
	Ni	0.029	"	Fe 0.020 "
				Ni 0.010 "
Analysis of nitric acid (12)	Al	0.050	ppm	All operations hermetized
	Fe	0.030	"	Al 0.001 ppm
	Ca	0.050	"	Fe 0.008 "
	Cu	0.005	"	Ca 0.003 "
	Mg	0.030	"	Cu <0.0001 "
	Mn	0.0006	"	Mg 0.005 "
	Ni	0.0007	"	Mn 0.0001 "
				Ni <0.0001 "

and hermetically sealed decomposition vessels (8, 9) are less expensive, but often very useful to reduce airborne contamination.

The effectiveness of these facilities and apparatus is shown in Table V.

CONTAMINATION DUE TO REAGENTS

Impurities present in the reagents used may become serious sources of contamination (13, 14), especially when mineral acids and salts are employed in large quantities in the decomposition of solid samples and in separations. Even if this kind of contamination is relatively reproducible, it is essential to use minimum amounts of reagents and high-purity material to obtain reliable results in trace analysis. Many high-purity reagents are now commercially available. However, they still contain appreciable amounts of impurities, which may not be tolerable in certain cases. It is almost impossible to reduce all impurities in a reagent down to undetectable levels. Total concentrations of metallic impurities are generally at the sub-ppm level in very carefully purified acids. In addition, reagents are contaminated during storage by the material of which the container is made.

According to the completed questionnaires, 66 laboratories (69 %) are not fully satisfied with purity of commercial analytical reagents. For the above and some other reasons (Commercial high-purity reagents are expensive, may not be easily available in some areas, etc.), mineral acids and other reagents as well as water are purified or prepared in analytical laboratories as shown in Table VI. The purification method used depends on the nature of the reagent and the impurities to be removed.

Distillation is widely used for the purification of water, mineral acids, and organic solvents. Conventional or boiling distillation is simple and rapid, but significant contamination of the distillate is caused by entrainment of liquid particulates in the vapor stream formed during bubble rupture and by creeping of the unrectified liquid. To overcome these difficulties, two

Table VI. Purification and preparation of high-purity reagents.

Reagents	Methods	Number of laboratories
HCl	Distillation	12
	Isothermal distillation	8
	Sub-boiling distillation	6
	Electrolysis	3
	Solvent extraction	1
	Ion exchange	1
HNO ₃	Distillation	14
	Isothermal distillation	3
	Sub-boiling distillation	6
H ₂ SO ₄	Distillation	3
	Sub-boiling distillation	1
	HF-treatment	1
HClO ₄	Distillation	3
	Sub-boiling distillation	3
HF	Distillation	3
	Isothermal distillation	2
	Sub-boiling distillation	5
NH ₃ (Aq)	Isothermal distillation	4
	Dissolution of NH ₃ gas in water	1
Salts	Solvent extraction	7
	Ion exchange	4
	Recrystallization	4
	Electrolysis	3
	Acid plus alkali	1
Organic solvents	Distillation	7
	Back-extraction	1

techniques have been developed. First is isothermal or isopiestic distillation (15, 16), in which the reagent-grade reagent and high-purity water are placed separately in a closed vessel such as a desiccator and the vapor is allowed to diffuse into the water under isothermal conditions. This technique is useful for the preparation of small quantities of high-purity hydrochloric acid, hydrofluoric acid, ammonia, etc. The second technique is sub-boiling distillation (17-19), in which infrared radiators vaporize the surface without boiling the liquid in a silica or Teflon still. High-purity hydrochloric acid (31 %), nitric acid (70%), perchloric acid (70 %), sulfuric acid (96 %), hydrofluoric acid (48 %), and water are obtained by this technique with production rates of 300-4000 ml per 24 h. The sum of the common impurity elements (Pb, Tl, Ba, Te, Sn, In, Cd, Ag, Sr, Se, Zn, Cu, Ni, Fe, Cr, Ca, K, Mg, and Na) in the purified acids ranged from 0.0023 ppm in nitric acid to 0.027 ppm in sulfuric acid (17).

Other useful purification techniques include solvent extraction, ion exchange, electrolysis, and recrystallization (7, 20). Sometimes, chemical reactions are used to prepare high-purity reagents from other pure reagents. Thus high-purity hydrochloric, hydrobromic, and hydrofluoric acids are prepared by saturating pure water with the pure gaseous compounds, high-purity sodium and potassium hydroxides by conversion of pure sodium and potassium chlorides with an OH-form anion-exchange resin, and high-purity salts from pure acids and alkalis.

A clean environment and proper selection of the material of vessels and other apparatus used in the preparation and storage are essential in order to obtain high-purity reagents. Thus, in the analysis of the "Trace Elements in Glass" Standard Reference Materials, the U.S. National Bureau of Standards workers were able to lower the lead blank from 330 ± 250 ng to 2 ± 1 ng and the silver blank from 970 ± 500 ng to 3 ± 2 ng by using high-purity acids in clean rooms, over a period of 4 years (6).

CONTAMINATION DUE TO APPARATUS

Under certain conditions, the surfaces of containers and other apparatus which are in direct contact with the samples are attacked to some extent and contaminate the samples. Also, strong adsorption or adhesion of elements on the surfaces causes contamination of later samples from previous ones. High temperatures, high pressures, and prolonged contact may increase the contamination. Therefore, decomposition of samples, comminution, prolonged storage of sample and reagent solutions are dangerous steps from this viewpoint. On the other hand, there is relatively little fear of contamination due to containers or forceps used for treating dry solid samples at room temperature.

Table VII lists the contamination due to apparatus observed in the laboratories which returned the questionnaire. Examples of quantitative data from the literature are given in Table VIII (21). The best results were obtained with poly(fluorocarbon) vessels.

Table VII. Contamination due to laboratory ware.
(Number of laboratories in parentheses.)

Material	Contaminants							
Glass	Al (2)	As (3)	B (6)	Ca (5)	Cd (1)	Co (1)	Cu (1)	
	Fe (2)	Hg (2)	K (2)	Na (5)	Pb (6)	Sb (2)	Si (3)	
		Zn (3)						
Silica	Co (1)	Cr (1)	Cs (1)	Cu (1)	Fe (1)	Na (1)		
	Pb (1)	Sb (1)	Sc (1)					
Polyethylene	B (1)	Cd (1)	Co (1)	Cu (3)	Fe (2)	Hg (2)	Na (1)	
	Pb (3)	Sc (1)	Zn (3)	Solid particles (1)				
Polypropylene	As (1)	Cd (2)	Fe (2)	Na (1)	Pb (1)	Zn (2)		
Poly(fluorocarbon)	Cd (1)	Cu (2)	F (1)	Fe (2)	Hg (1)	K (1)	Pb (2)	
		Si (1)						
Rubber		Zn (3)						

Table VIII. Results of spectrochemical analysis of HF, HCl, and HNO₃ after evaporation in poly(fluorocarbon), platinum, and silica dishes.

Acid	Material	Elements determined (x10 ⁻³ ppm)										
		Al	Fe	Ca	Cu	Mg	Mn	Ni	Pb	Ti	Cr	Sn
HF	Poly(fluorocarbon)	3	3	1	<0.04	<3	0.1	<0.4	<0.1	0.1	<0.4	not detected
	Platinum	10	10	10	0.4	10	0.2	0.8	0.5	1	0.5	not detected
HCl	Poly(fluorocarbon)	<4	3	5	0.2	3	0.1	not detected	<0.4	not detected	not detected	not detected
	Platinum	2	2	10	1	6	0.2	0.6	<0.4	0.4	traces	<0.4
	Silica	10	10	60	1	10	0.4	2	0.5	2	0.6	0.4
HNO ₃	Poly(fluorocarbon)	2	8	4	≤0.01	7	0.1	not detected	not detected	traces	not detected	not detected
	Platinum	20	20	30	0.4	20	0.6	traces	1	0.8	not detected	not detected
	Silica	20	20	60	0.1	20	0.6	not detected	1	0.3	not detected	not detected

Since contamination of this kind is quite difficult to estimate because of its poor reproducibility, much attention has to be paid to the selection of the apparatus materials, the history of the apparatus, and the cleaning of the apparatus. Poly(fluorocarbon), polyethylene (made by the high pressure process), silica, and platinum are most suitable for container materials in trace analysis because of their purity and chemical inertness. It must be remembered that these commercial materials often contain various impurities at the ppm levels. Impurities in plastics, silica, glass and other materials used in laboratories have been reported (13, 14, 20, 22-25).

Numerous fine particles (0.5 mm or smaller) containing Fe, Ti, Mg, Ca, Si, Zn, Al, Cr, Ni, Cu, and Mn have been found in various samples of poly(fluorocarbon), polyethylene, and polypropylene (22, 26). These particulates are effectively removed by vigorous prolonged cleaning with hot hydrochloric and nitric acids or their mixtures (26). Recently, it has been reported that the mercury contamination of sea water samples stored in polyethylene containers may be caused by leaching of mercury from container surfaces or by passage of mercury vapor from the ambient air through the container wall into the solution or may originate from both sources (27). The diffusion of mercury vapor through polyethylene foils (thickness 0.3 mm) was confirmed in another laboratory.

Table IX summarizes the agents used in various laboratories for cleaning containers and other apparatus. For glass, silica, and plastic ware, nitric acid, hydrochloric acid, and detergents are most frequently used at various concentrations and temperatures. Classic chromic acid cleaning solution is still popular because of its cleaning power, but it must be kept in mind that appreciable amounts of chromium tenaciously remain on glass surfaces even after careful rinsing with water. Thiers (20) recommended the use of a one-to-one mixture of concentrated sulfuric acid and nitric acid instead of chromic acid cleaning solution. Glass and silica ware can also be effectively cleaned with a dilute solution of hydrofluoric acid and its mixtures with other mineral acids (14, 24, 25). However, this procedure may produce freshly roughened surfaces from which impurities can be more readily leached. Any cleaning procedure should be followed by copious rinsing with purified water. The application of an ultrasonic field accelerates the cleaning. To avoid airborne contamination, apparatus should be cleaned immediately before use or stored under cover after cleaning.

Filters, mortars, and sieves may often become sources of serious contamination (14, 20). Therefore, whenever possible, filtration, grinding, and sieving should be avoided.

Table IX. Cleaning agents for laboratory ware.
(Number of laboratories in parentheses.)

Materials	Cleaning agents				
Glass and silica	HNO ₃ (44)	HCl (23)	Detergent (18)		
	Chromic acid cleaning solution (18)				
	HCl-HNO ₃ mixture (10)		Acetone (7)	Methanol (5)	
	HF (3)	H ₂ SO ₄ -HNO ₃ mixture (3)		H ₂ SO ₄ (1)	
	HClO ₄ (1)	HF-HCl mixture (1)	HF-HNO ₃ mixture (1)		
	Ethanol (1)				
Plastics	HNO ₃ (30)	HCl (23)	Detergent (12)	HF (4)	
	HCl-HNO ₃ mixture (4)		Ethanol (3)		
	Acetone (3)	H ₂ SO ₄ -HNO ₃ mixture (3)		Methanol (2)	
	HClO ₄ (1)	Chromic acid cleaning solution (1)			
	HF-HNO ₃ mixture (1)		HF-HClO ₄ mixture (1)		
Platinum	Pyrosulfate (7)	HNO ₃ (5)	HCl (5)	HF (1)	
	HClO ₄ (1)	HF-HNO ₃ mixture (1)			

CONTAMINATION DUE TO THE ANALYST

Careless manipulations cause serious contamination. Touching the surfaces of solid samples or apparatus (which will come in direct contact with samples) with fingers can cause contamination with such elements as Cl, Na, Ca, Cd, and Pb. Thus the use of plastic gloves is essential in the determination of traces of chlorine. Cosmetics, medications, watches, and jewelry are other sources of contamination. Cross-contamination from other work in the laboratory sometimes occurs.

Acknowledgment - We are grateful to those who completed the questionnaire and to the authors of the literature cited.

REFERENCES

1. G. Ehrlich, R. Gerbatsch, K. Jatsch, and H. Scholze, Reinstoffe in Wissenschaft und Technik, Berlin (1963).
2. M. Pinta, Dosage des éléments à l'état de traces dans les roches et les autres substances minérales naturelles, Éditions du Centre National de la Recherche Scientifique, Paris (1970) p. 25.
3. G. Tölg, Talanta, **19**, 1489 (1972).
4. J. A. Paulhamus, Ultrapurity: Methods and Techniques (M. Zief and R. Speights, eds.), Marcel Dekker, New York (1972) p. 255.
5. M. Zief and A. G. Neshor, Environmental Science and Technology, **8**, 677 (1974).
6. T. J. Murphy, Accuracy in Trace Analysis: Sampling, Sample Handling, and Analysis, NBS Special Publication 422, Vol. I (P. D. LaFleur, ed.) (1976) p. 509.
7. A. Mizuike, Techniques of Metals Research, Vol. 3 Part 1 (R. F. Bunshah, ed.), Interscience, New York (1970) p. 25.
8. J. W. Mitchell, Anal. Chem., **45**, 492A (1973).
9. G. Tölg, Talanta, **21**, 327 (1974).
10. E. J. Maienthal, U.S. National Bureau of Standards Technical Note 545 (J. K. Taylor, ed.) (1970) p. 53-54.
11. T. J. Chow and C. R. McKinney, Anal. Chem., **30**, 1499 (1958).
12. L. S. Vasilevskaya, through J. Minczewski, Trace Characterization — Chemical and Physical, NBS Monograph 100 (W. W. Meinke and B. F. Scribner, eds.) (1967) p. 385.
13. D. E. Robertson, Anal. Chem., **40**, 1067 (1968).
14. D. E. Robertson, Ultrapurity: Methods and Techniques (M. Zief and R. Speights, eds.), Marcel Dekker, New York (1972) p. 207.
15. H. Irving and J. J. Cox, Analyst, **83**, 526 (1958).
16. W. Kwestroo and J. Visser, Analyst, **90**, 297 (1965).
17. E. C. Kuehner, R. Alvarez, P. J. Paulsen, and T. J. Murphy, Anal. Chem., **44**, 2050 (1972).
18. J. M. Mattinson, Anal. Chem., **44**, 1715 (1972).
19. K. Little and J. D. Brooks, Anal. Chem., **46**, 1343 (1974).
20. R. E. Thiers, Methods of Biochemical Analysis, Vol. V (D. Glick, ed.), Interscience, New York (1957) p. 273.
21. L. S. Vasilevskaya, Analysis of High-Purity Materials (I. P. Alimarin, ed.), Israel Program for Scientific Translations, Jerusalem (1968) p. 13.
22. E. C. Kuehner and D. H. Freeman, Purification of Inorganic and Organic Materials (M. Zief, ed.), Marcel Dekker, New York (1969) p. 297.
23. O. G. Koch and G. A. Koch-Dedic, Handbuch der Spurenanalyse, Teil 1, Springer, Berlin (1974) p. 82.
24. P. B. Adams, Ultrapurity: Methods and Techniques (M. Zief and R. Speights, eds.), Marcel Dekker, New York (1972) p. 293.
25. G. Hetherington and L. W. Bell, Ultrapurity: Methods and Techniques (M. Zief and R. Speights, eds.), Marcel Dekker, New York (1972) p. 353.
26. T. Kuroha, Bunseki Kagaku, **21**, 502, 506 (1972).
27. M. H. Bothner and D. E. Robertson, Anal. Chem., **47**, 592 (1975).