93. R. Sh. Nigmatullin, The Oscillographic Method as Applied to Polarography on Solid Electrodes. Author's abstract of dissertation [in Russian] (Kazan University, 1953).
94. I. I. Tsapiv, Zavod. laboratoriya, 21, 286 (1955); Zhurn. analit. khim., 11, 63 (1956); Nauchnye zapiski L'vovskogo politekhnicheskogo inst., 22, 29 (1956).
95. A. V. Gordievskii and Yu. A. Zotov, Zavod. laboratoriya, 23, 922 (1957).
96. I. Shain and A. L. Grittenden, Anal. Chem., 26, 281 (1954).
97. Ya. P. Gokhshtein, A. F. Volkov, S. V. Kuz'min, and V. A. Yanchevskii, Zavod. laboratoriya, 25, 1008 (1959).
98. I. N. Sheiko, A. V. Gorodyskii, and M. I. Bykova, Zhurn. neorgan. khim., 6, 2341 (1961).
99. C. J. Hills and J. E. Oxley, J. analyt. Chem., 173, 5 (1960).
100. T. Mine, J. Electrochem. Soc. Japan, Overseas Ed., 28, E51 (1960).
101. D. Papoušek, Collection, 20, 251 (1955).
102. K. Micka, Chem. listy, 55, 474 (1961).
103. E. M. Skobets and V. I. Shapoval, Zavod. laboratoriya, 26, 278 (1960); Ukrainskii khim. zhurn., 26, 446 (1960); Dokl. AN UkrSSR, 1421 (1960).
104. V. Leont'ev and N. Fedotov, Zavod. laboratoriya, 26, 276 (1960).
105. O. K. Kudra, Izvestiya Kievskogo politekhnicheskogo instituta, 15, 189 (1959).
106. M. Dolcetta and P. Lanza, Atti. Acad. naz. Lincei. Rend. Cl. sci. fis., mat. e natur., 14, 642 (1953).
107. D. E. Walker, R. N. Adams, and A. L. Juliard, Anal. chem., 32, 1526 (1960); D. E. Smith and W. H. Reinmuth, Anal. Chem., 32, 1892 (1960).
108. M. M. Nicholson, Anal. Chem., 32, 1058 (1960).
109. R. Need, Z. analyt. Chem., 180, 161 (1961).
110. Yu. K. Delimarskii, Uspekhi khimii, 23, 766 (1954).
111. S. Voshizawa, T. Nishi, A. Hashino, and M. Itonoshe, Bull. Engng. Res. Inst. Kyoto Univ., 16, 78 (1959).
112. S. Voshizawa, Rev. Polarogr., 9, 1 (1961).
113. Yu. K. Delimarskii and B. F. Markov, The Electrochemistry of Molten Salts [in Russian] (Metallurgizdat, 1960).
114. N. G. Chovnyk and V. V. Vashchenko, Zhurn. fiz. khim., 35, 580 (1961).
115. G. J. Hills and K. E. Johnson, Proc. Second Intern. Congr. Polarogr. (Cambridge, 1959), Vol. 3; Ed. Longmuir Ian S., Pergamon Press, 1960, p. 974.
116. Yu. S. Lyalikov, Zavod. laboratoriya, 17, 909 (1951).
117. Yu. S. Lyalikov, Zhurn. analit. khim., 8, 38 (1953).
118. K. M. Kalabalina and Yu. K. Delimarskii, Ukrainskii khimicheskii zhurn., 24, 152 (1958); 27, 270 (1961); Dokl. AN UkrSSR, 562 (1957).
119. Yu. K. Delimarskii and T. N. Kaptsova, Zhurn. neorg. khim., 3, 2751 (1958).
120. M. Rey, G. Donner, and M. Baileul, Metallurgia ital., 52, 537 (1960).

THE PULSE POLAROGRAPHIC METHOD OF ANALYSIS
(REVIEW)

V. I. Bodyu, I. V. Kozlova, and Yu. S. Lyalikov

Translated from Zavodskaya Laboratoriya, Vol. 28, No. 9,
pp. 1042-1047, September, 1962

Pulse polarography is one of the new developments in polarographic analysis. This expression [1] is taken to mean polarography using a pulsating polarization voltage.

Pulse polarography is more sensitive; it can be used to increase the degree of separation of neighboring polarographic waves and to reduce the interfering action of certain impurities. The possibility of determining concentrations as low as 10^{-7} mole/liter, and also ions with a low half-wave potential difference has led to the extensive use of this method abroad, especially in Japan.

Kaplan [1] was the first to review a large number of foreign papers dealing with this development. Articles by Humm [2] and other authors [3] dealt with the theory, practice, and apparatus for pulse polarography.

A paper by Reinmuth and Smith [3] criticizes the conclusions of Bauer and other authors on problems in the theory of pulse polarography. An article by Isikhasi Masayoshi reviewed papers for 1959 [4].

A number of papers and monographs [5,6] have described general principles of polarography with the application of an alternating voltage, and also have studied in detail the advantages of this method over ordinary polarography [7-10]. Nurnberg and Stackelberg have written two long articles [11, 12]. They give a detailed description of the apparatus, method, and theory of pulse polarographic curves. Special attention is paid to measurements of reaction kinetics.

Schmidt et al. [13], studied the effect of ohmic resistance and capacity of the double layer on the current of the peak in alternating current polarography. They studied the relationship between the maximum frequency ω and maximum current strength $\underline{i}$. They calculated their values for an equivalent circuit made up of resistances and capacitors connected in series. They studied the relationship between $\underline{i}$ and ω for reversible and irreversible processes.

Bauer [14] studied the height and slope of the peaks at low and high frequencies. The application of an alternating voltage to the cell produces an alternating current with a large number of harmonics.

Bauer et al. [15, 16], used the highest harmonics instead of the fundamental harmonics. This made it possible to avoid interference due to the presence of an electrical double layer, to reduce the value of the residual current, and increase the sensitivity of the determination. The measurement of harmonics made it possible to obtain separate peaks for mixtures of ions which could not be analyzed by means of ordinary alternating current polarography.

The polarization of a mercury electrode by a sine alternating current was dealt with by Micka et al. [17, 18]. The author allowed for the influence of the electrode capacitance when deriving a number of theoretical formulas. He showed that the phase shift between the potential and current is a function of the capacity and concentration of the depolarizer. The theoretical data were checked for the case of the pulse polarography of thallium and lead.

Tanaka, Kodama, and other authors [19, 20] studied a fast-dropping capillary and maxima of the second kind in classical polarography and in pulse polarography. In pulse polarography they found no effect due to mixing, causing maxima of the second kind.

Matsuda [21] used a flowing electrode at low amplitudes of the alternating current.

Bauer [22] derives an equation for an alternating current polarographic wave for low amplitudes of the alternating component. He showed that the maximum on the pulse polarogram does not coincide with the half-wave potential on an ordinary polarogram. The difference between them depends on the difference in the diffusion coefficients of the oxidized and reduced forms.

The article by Sturm [23] compared the boundaries of applicability of the classical polarographic and square-wave method for reversible and irreversible reactions. For the square-wave method he found an increase in sensitivity for reversible reactions by one order, and for irreversible actions by two orders. For the square-wave method the limiting ratio of concentrations of the ion to be determined and the ion which is reduced at a less negative potential is 10^2-10^3 instead of 50-100 for ordinary polarographic analysis.

Alternating current polarography was also used for tensimetric investigations of adsorption processes [24].

Bauer and Elving [25] studied methods for the calculation and accuracy of data for the pulse polarographic method of analysis.

A number of Soviet articles have appeared in the last few years describing various types of polarographs.

The Tsfasman monograph [6] emphasizes the theory of pulse polarography with an applied sine voltage. The principles are given for the design of pulse polarographs, and capacitance current cutoffs. The article gave a circuit for an automatic pulse polarographic concentration measurer [26], adopted for batch production.

Tsfasman and Bryksin have published a block circuit of a new model of an alternating current polarograph – the vector polarograph [27] – and have given its analytical data. The reliable limit for the determination of cadmium is $5 \cdot 10^{-7}$ mole; for a mixture of cadmium and copper, the resolving capacity is 1:2000.

Kaplan and Ulanovskii gave a detailed description of a visual pulse polarograph [28]. Gorelkinskii, Grinman, and Kozlov [29], and after them Bodyu, Senkevich, and Lyalikov [30], published circuits for pulse polarographs using a tube phase detector instead of a mechanical detector as the capacitance current cutoff. This ensured good sensitivity and operating stability in the pulse polarograph. A number of pulse polarographs were described in the article by Schmidt [31].

The use of a low amplitude of the alternating current and an electronic phase-sensitive detector in pulse polarography was described in [32]; this made it possible to separate the charging current from the Faraday current. The authors obtained a sensitivity of 10^{-8} mole/liter on a stationary mercury electrode. A number of other alternating current polarograph designs and attachments for them have also been described [33-36].

Barker has reported on his most recent discoveries in the field of square-wave polarography [37, 38] – pulse and radiofrequency pulse polarographs. In the pulse polarograph one square pulse lasting 0.04 sec is applied at a certain period in the life of the drop. A long pulse leads to an increase in sensitivity, especially for irreversible processes.

The sensitivity of the square-wave polarograph is limited by capillary noise due to the penetration of the solution into the capillary; in pulse polarography this interference is much less serious. In the radiofrequency polarograph the electrode is polarized by an alternating voltage with a frequency of a few MHz. At this frequency the effect of the capacitance current is less; a higher sensitivity can therefore be obtained by increasing the electrode surface in the microcell. The radiofrequency polarograph is suitable for irreversible processes of any degree.

Milner [39] described the use of a square-wave polarograph and a pulse polarograph in analytical chemistry.

Yasumori [40] described a new polarographic apparatus using square pulses. The potential of the square-wave and the gating signal are given by means of a time circuit based on a dekatron. The described instrument can be used to determine as low as $2 \cdot 10^{-7}$ mole of Cd^{2+}.

There are about 50 papers in the periodic literature dealing with the pulse polarography of inorganic ions. Japanese investigators are working very intensively in the pulse polarography of inorganic substances (almost three-quarters of the total number of published papers).

We should not overlook the comparatively small number of articles on the pulse polarography of inorganic substances published in the Soviet periodic literature.

Bauer [41] has published a long review article dealing with the determination of inorganic ions. He gives potentials of peaks with various supporting electrolytes for more than 25 cations and anions; he describes the conditions for the separation of peaks with similar reduction potentials; the clearest separation is achieved for low alternating components – as low as 5-10 mV.

Several papers on pulse polarography deal with ions which have been thoroughly studied from the polarographic point of view.

The Japanese investigators Itsuki, Nagao, Tajima, et al. [42-45], dealt with the pulse polarographic determination of zinc in ores. They described the determination of lead in ores, zinc, and iron in aluminum alloys. They studied the influence of accompanying elements and oxygen and noted the considerable reduction in time for the analysis. In aluminum alloys zinc and copper were determined on one pulse polarogram with a hydrochloric acid supporting electrolyte, iron was extracted by methyl isobutyl ketone, and after reextraction it was determined using an alkaline supporting electrolyte in the presence of citric salts.

Hideko Shirai reported on interesting investigations in the pulse polarographic behavior of zinc [46]. The author detected three waves on the pulse polarograms. One of them corresponded to the reduction of the zinc ion, the second wave was detected in the presence of oxygen and corresponded to the reduction of zinc hydroxide or the complex particle $[Zn(NH_3)_2](OH)_2$ in ammonia solutions; the third wave, appearing in ammonia solutions at pH values above 7.3 in the presence of an excess of ammonium salts, corresponded to the reduction of the $[Zn(NH_3)_3]^{2+}$ ion. Several articles deal with the determination of copper in various materials and study their behavior in pulse polarographic analysis.

Tajima, Kurobe et al. [47, 48], described the simultaneous determination of copper and lead in iron and steel. The determination was conducted with a hydrochloric acid supporting electrolyte after the oxygen had been removed by hydrogen. Lead, up to 5% chromium, and antimony do not interfere with the determination. In the determination of trace amounts (10^{-3} to 10^{-4}%), iron is extracted by methyl isobutyl ketone.

Itsuki and Kaji [49, 50] described the determination of copper in tin. They used perchloric acid as the supporting electrolyte and studied the influence of the electrode dimensions, the temperature of foreign ions, and other factors. This method of determination is very rapid.

Yasumori and co-workers [51-53] determined as low as 0.01 mg/liter of copper in sulfuric acid. Using a sulfuric acid supporting electrolyte at less than 5 M concentration, distinct copper waves were obtained; oxygen and sulfur dioxide were removed by nitrogen because they interfered with the determination. The best results were obtained at temperatures of about 50°.

Takahashi and Shirai [54] studied the pulse polarographic peaks of copper using a supporting electrolyte of caustic alkali. Three reduction peaks were obtained: The first corresponded to the reduction of $Cu(OH)_2$, the second and third corresponded to the reduction of the complex $[Cu(OH)_4]^{2-}$. Kaplan studied the pulse polarographic determination of lead, zinc, copper, and indium in mineral raw materials [55].

At the sixth conference of workers in plant and industrial laboratories of Kazakhstan and Central Asia, Lysenko, Lisitsyna, and Vishnevetskaya reported on the practical application of pulse polarography in the analysis of nonferrous and rare metals [56]. The authors developed methods for the polarographic determination of copper, indium, thallium, and other metals in metallic lead, high-purity zinc, ores, and benefication tailings. These methods can determine concentrations as low as 10^{-6}%. Pats, Tsfasman, and Semochkin [57] also reported on the determination of the same metals in various products of nonferrous metallurgy.

A number of articles by foreign authors have dealt with the determination of these elements. Takahashi [58], Shirai [59], Itsuki and other authors [60, 61], have proposed the pulse polarographic determination of bismuth and lead in silver. These metals are separated by coprecipitation with ferric hydroxide, the interfering influence of trivalent iron is removed by reducing it to the divalent state.

A number of articles have dealt with the pulse polarographic determination of trace quantities of elements.

Breyer [62], Milner [63, 64], and other authors [65] have described the pulse polarographic determination of uranium using a supporting electrolyte of perchloric acid and mineral acids in the presence of tartrates. Studies have been made of the dependence of the peak height on the concentration and the interfering action of other ions. In an article on the determination of uranium in sea water, Milner suggests concentrating the uranium by extracting it with di-2-ethyl-hexyl phosphoric acid. The accuracy of the determination is ±0.4%.

Analysts have paid less attention to the pulse polarographic determination of other metals.

One paper dealt with the pulse polarographic determination of lanthanides [66].

In a number of articles, Yasumori and co-workers described the pulse polarographic determination of the chlorine ion [67], iron in the presence of various complex formers [68], and vanadium [69], using a sulfuric acid supporting electrolyte. In the first of these papers the authors studied the peak of the chlorine ion and the copper ion. With a 10 M sulfuric acid supporting electrolyte the chlorine peak was superimposed on the copper peak, the height of the total peak being proportional to the chlorine ion concentration. By studying the behavior of the iron peak, the authors detected an increase in the peak with increase in the oxalic acid concentration (complex former) and a reduction in it with increase in the sulfuric acid concentration. Using a supporting electrolyte of 0.36 M sulfuric and 1 M oxalic acid, the peak potential was −0.2 V and its height was proportional to the concentration of trivalent iron. With this supporting electrolyte, copper did not interfere with the determination.

The authors determined vanadium with mercury and platinum electrodes. This is one of the few papers on the pulse polarographic use of solid electrodes. With a mercury electrode the peaks are vague and poorly reproducible; the position of the peaks depends on the sulfuric acid concentration. The authors recommend waves obtained with a platinum electrode. The sensitivity of the method is 0.1 mg/liter of vanadium.

In analytical chemistry it is often necessary to deal with high concentrations of trivalent iron and to determine other ions with ferric iron as the supporting electrolyte. An article by Kaplan [70] studies the effect of high iron concentrations on the pulse polarographic peaks of indium. Several articles deal with the determination of antimony [71], nickel [72, 73], and complex compounds of copper with mannitol [74].

Sato [75] studied more than 20 organic compounds with a pulse polarograph using various solutions as the supporting electrolyte and various pH values. For many compounds he found a linear relationship between the height of the peak and the concentration. He investigated picric acid, aromatic ketones, nitrobenzene, etc. For each of the compounds he gave the conditions for obtaining the clearest peaks.

Breyer and co-workers [76] described the pulse polarographic determination of riboflavin in various biological materials. The determination was conducted with various supporting electrolytes, depending on the concentration. For concentrations above 10^{-6} M they suggested an acetate buffer with pH = 4.7; for lower concentrations, a borate buffer with pH = 9.5. The accuracy in determining concentrations above 10^{-6} M was about 2%, for lower concentrations about 10%.

Takahashi and Shirai [77] have demonstrated the possibility of the pulse polarographic determination of nitrophenol isomers. In contrast to the polarographic determination in pulse polarography, the nitrophenol waves are not distorted by kinetic currents. With an alkaline supporting electrolyte, p-nitrophenol gives the following peaks: the first peak at −0.91 V, the second peak at −0.45 V, m-nitrophenol gives peaks at −0.82 and −1.24 V, respectively, o-nitrophenol gives peaks at −0.88 and −1.01 V, respectively. There is a linear relationship between the concentration and the heights. The fairly large difference in the potentials of the second peaks makes it possible to differentiate between the isomers of nitrophenols and their mixture.

Schwabe and Jering [78] studied the possibility of determining certain organic substances from their tensimetric peaks.

The authors studied sulphanylic acid, urotropine, various alcohols, and other compounds. Under certain conditions these compounds are adsorbed on a mercury electrode and give clear desorption waves.

A report by Lyalikov, Bodyu, and Sister [79] demonstrated the possibility of the pulse polarographic determination of a number of aromatic aldehydes with a sensitivity of 10^{-6} mole/liter. They demonstrated the possibility of determining small amounts of vanillin both from the peak on the pulse polarogram and from the reduction in the peak of 2,4-dinitrophenylhydrazine. The method is used to determine vanillin in cognacs and cognac spirits. These authors suggested a method of pulse polarographic titration by the neutralization method, based on the fact that the tensimetric peaks of cyclohexane, methyl red, and other surface-active substances change strongly with change in the pH of the solution. They are measured during titration by the neutralization method; the potential shift of the peak or the change in its height clearly indicates the equivalence point of the titration.

A study was made of the effect of dissolved oxygen in buffer and nonbuffer solutions on aldehydes and other organic compounds [33].

Koyama [80] suggested a method for determining small amounts of plutonium using square-wave polarography. He used a supporting electrolyte of hydrochloric and nitric acids; the potential of the oxidation peak was +0.71 −0.66 V.

All modifications of the pulse-polarographic method considerably extend the possibilities of polarographic investigations. The main trends in the further development of this method are now being outlined in the periodic literature. The first line of development will be the determination of trace contents of metals (10^{-7} to 10^{-6} mole/liter).

The advantages of pulse polarography show up especially clearly in the determination of mixtures of ions with similar half-wave potentials and in the determination of ions with more negative half-wave potentials in the presence of large amounts of ions with more positive half-wave potentials. Pulse polarography is very promising in the study of organic compounds. As well as the fact that all advantages mentioned for inorganic ions also apply to organic substances, this method can be used to detect tensimetric adsorption and desorption peaks of organic substances which are not reduced on the mercury cathode. These peaks can be used for analytical purposes. Unfortunately, the pulse polarography of organic substances is still a long way behind the pulse polarography of inorganic ions.

Because of its high sensitivity and specificity, the pulse polarographic method can be used extensively when studying a number of theoretical problems – the composition of complexes in various media, the solubility of precipitates, etc.

There are undoubted advantages to be gained from the development of pulse polarography in combination with other polarographic methods – pulse polarography on a stationary mercury drop, pulse polarography on solid electrodes, and the pulse polarography of melts.

LITERATURE CITED

1. B. Ya. Kaplan, Zavod. laboratoriya, 23, 7, 771 (1957).
2. D. Humm, Analyt. Chem., 32, 5, 137 (1960).
3. W. H. Reinmuth and D. E. Smith, Analyt. Chem., 33, 7, 964 (1961).
4. Isikhasi Masayoshi and Idzutsu Kimisuke, Chemistry (Japan), 15, 2, 165 (1960).
5. Yu. K. Delimarskii, and D. V. Gorodyskii, Electrode Processes and Methods of Investigation in Polarography [in Russian] (Izd. AN UkrSSR, Kiev, 1960).
6. S. B. Tsfasman, Electronic Polarographs [in Russian] (Metallurgizdat, 1960).
7. Itsuki Kamihico, Japan Analyst, 7, 4, 253 (1954).
8. H. Schmidt, Z. analyt. chem., 173, 1, 73 (1960).
9. H. H. Bauer, J. Electroanalyt. Chem., 1, 363 (1960); Chem. Zbl., 46, 16584 (1961).
10. M. von Stackelberg and H. Schmidt, Angew. Chem., 71, 508 (1959); Chem. Zbl., 20, 6599 (1960).
11. H. W. Nurnberg and M. von Stackelberg, J. Electroanalyt. Chem., 2, 3, 181 (1961).
12. H. W. Nurnberg and M. von Stackelberg, J. Electroanalyt. Chem., 2, 5, 350 (1961).
13. H. Schmidt and M. von Stackelberg, J. Electroanalyt. Chem., 1, 2, 133 (1959).
14. H. H. Bauer, J. Electroanalyt. Chem., 2, 1, 66 (1961).
15. H. H. Bauer, J. Electroanalyt. Chem., 1, 3, 256 (1960).
16. D. E. Smith and W. H. Reinmuth, Analyt. Chem., 33, 482 (1961); Chem. Zbl., 48, 17313 (1961).
17. K. Micka, Collect. czechoslov. chem. Communs., 24, 3708 (1959); Chem. Zbl., 50, 16665 (1960).
18. K. Micka, Z. Phis. Chem. (DDR), 206, No. 5-6, 345 (1957).
19. N. Tanaka, R. Tamamushi, and M. Kodama, Analyt. Chim. Acta, 20, 573 (1959); Chem. Zbl., 12, 3781 (1960).
20. N. Tanaka and M. Kodama, J. Chem. Soc. Japan, 79, 3, 409 (1958).
21. H. Matsuda, Z. Electrochem., 62, 9, 977 (1958).
22. H. H. Bauer, J. Electroanalyt. Chem., 1, 1, 2 (1959).
23. F. von Sturm, Z. Analyt. Chem., 173, 1, 11 (1960).
24. H. H. Bauer and P. J. Elving, Austral. J. Chem., 12, 3, 343 (1959).
25. H. H. Bauer and P. J. Elving, Austral. J. Chem., 12, 3, 335 (1959).
26. S. B. Tsfasman, I. E. Bryksin, and B. S. Bruk, Zavod. laboratoriya, 24, 11, 1409 (1958).
27. S. B. Tsfasman and I. E. Bryksin, Zavod. laboratoriya, 26, 9, 1064 (1960).
28. B. Ya. Kaplan and E. N. Ulanovskii, Zavod. laboratoriya, 25, 11, 1414 (1958).
29. Yu. V. Gorelkinskii, I. G. Grinman, and G. S. Kozlov, Zavod. laboratoriya, 26, 9, 1141 (1960).
30. V. I. Bodyu, V. V. Senkevich, and Yu. S. Lyalikov, Express Bulletin of the State Scientific and Technical Committee in the Council of Ministers of the Moldavian SSR, October, 1961.
31. H. Schmidt, Z. Instrumentkunde, 67, 12, 301 (1959).
32. D. E. Smith and W. H. Reinmuth, Analyt. Chem., 32, 13, 1892 (1960).
33. W. F. Head, Anal. Chim. Acta, 23, 297 (1960); Chem. Abstr., 21, 22135 (1960).
34. Takeo Takahashi and Eiyi Niki, Talanta, 1, 3, 245 (1958).
35. M. Ishibashi, T. Fujinaga, and A. Saito, J. Chem. Soc. Japan, 81, 5, 753 (1960).
36. M. Ishibashi, T. Fujinaga, and A. Saito, Collect. Czechoslov. Chem. Communs., 25, 12, 3387 (1960).
37. G. C. Barker, Anal. Chim. Acta, 18, 118 (1958).
38. G. C. Barker and A. W. Gardner, Z. analyt. Chemie, 173, 79-83 (1960).
39. G. W. C. Milner, Transactions of the Society of Instrument Technology (1959), p. 72.
40. Y. Yasumori, Japan Analyst, 8, 6, 361 (1959).
41. H. H. Bauer, J. Electroanalyt. Chem., 1, 5, 363 (1959/60).
42. K. Itsuki and M. Nagao, Japan Analyst, 9, 10, 836 (1960).
43. K. Itsuki and K. Nishino, Japan Analyst, 9, 12, 1062 (1960).
44. Senda Mitsugi, Senda Mitsuko, and Tachi Isamu, J. Elektrochem. Soc. Japan, 27, No. 1-3, E21, E24 (1959).
45. N. Tajima and M. Kurobe, Japan Analyst, 10, 2, 146 (1961).
46. Hideco Shirai, J. Chem. Soc. Japan, Pure Chem. Sec., 80, 609 (1959); Chem. Zbl., 45, 14959 (1960).
47. Ohiko Kammori and Mitsuyoshi Ikeda, J. Japan Inst. Metals, 23, 100 (1959); Chem. Zbl., 3, 956 (1961).

48. N. Tajima and M. Kurobe, Japan Analyst., 9, 10, 801 (1960).
49. K. Itsuki and Kaji Teru, Japan Analyst., 8, 9, 568 (1959).
50. K. Itsuki and Kaji Teru, Japan Analyst., 8, 11, 703 (1959).
51. Y. Yasumori and T. Eguchi, Japan Analyst., 9, 2, 127 (1960).
52. S. Hirano, Y. Yasumori, and T. Eguchi, Annual Rept. Engng. Res. Inst. Fac. Engng. Univ. Tokyo, 18, 2, 37 (1960).
53. Y. Yasumori, Chem. Abstr., 55, 16, 15209 (1961).
54. T. Takahashi and H. Shirai, J. Electroanalyt. Chem., 1, 5, 408 (1959/60).
55. B. Ya. Kaplan, Zavod. laboratoriya, 25, 1168 (1959).
56. V. I. Lysenko, É. V. Lisitsyna, and É. M. Vishnevetskaya, Theses of Reports to the Sixth Conference of Industrial Laboratory Workers of Kazakhstan and Central Asia [in Russian] (Alma-Ata, 1961).
57. R. G. Pats, S. B. Tsfasman, and T. V. Semochkin, Theses of Reports to the Sixth Conference of Industrial Laboratory Workers of Kazakhstan and Central Asia [in Russian] (Alma-Ata, 1961).
58. T. Takahashi and H. Shirai, Talanta, 5, 3/41, 193 (1960).
59. H. Shirai, Japan Analyst, 9, 3, 206 (1960).
60. K. Itsuki, B. Kobayashi, and K. Nishino, Japan Analyst., 8, 12, 804 (1959).
61. K. Itsuki and M. Nagao, Japan Analyst, 8, 2, 800 (1959).
62. R. Breyer and J. R. Beevers, J. Elektroanalyt. Chem., 1, 5, 345 (1960).
63. G. W. C. Milner, J. D. Wilson, J. A. Barnett, and A. A. Smales, J. Electroanalyt. Chem., 2, 1, 25 (1961).
64. G. W. C. Milner and J. H. Nunn, Anal. Chim. Acta, 21, 266 (1959); Chem. Zbl., 1, 240 (1960).
65. E. Goldblatt, J. South. African Inst. Mining Metallurg., 60, 292 (1960); Chem. Zbl., 50, 18068 (1961).
66. Iinzo Hashimoto, J. Japan Chem., 14, 240 (1960); Chem. Zbl., 3, 751 (1961).
67. Y. Yasumori and T. Eguchi, Japan Analyst, 9, 8, 680 (1960); Chem. Abstr., 19, 18447 (1961).
68. Y. Yasumori and T. Eguchi, Japan Analyst, 9, 12, 1049 (1960); Chem. Abstr., 21, 20779 (1961).
69. Y. Yasumori, T. Eguchi, and M. Nishiyama, RZhKh, 5D80 (1962).
70. B. Ya. Kaplan, Zavod. laboratoriya, 25, 6, 669 (1959).
71. K. Itsuki and M. Nagao, Japan Analyst, 12, 824 (1959).
72. Shirai Hideko, J. Chem. Soc. Japan, 81, 8, 1248 (1960).
73. Shirai Hideko, J. Chem. Soc. Japan, Pure Chem. Sec., 82, 3, 339 (1961).
74. T. Takahashi and H. Shirai, J. Chem. Soc. Japan., Indust. Chem. Sec., 64, 3, 515 (1961).
75. Hisashi Sato and Eisei Shikenjo Hôkoku,
76. B. Breyer and T. Biegler, J. Electroanalyt. Chem., 1, 6, 453 (1959/60).
77. T. Takahashi and H. Shirai, Talanta, 8, 4, 177 (1961).
78. K. Schwabe and H. Jering, Z. analyt. Chem., 173, 1, 36 (1960); Chem. Zbl., 33, 11826 (1961).
79. Yu. S. Lyalikov, V. I. Bodyu, and Yu. D. Sister, Theses of Reports to the Sixth Conference of Industrial Laboratory Workers of Kazakhstan and Central Asia [in Russian] (Alma-Ata, 1961).
80. K. Koyama, Analyt. Chem., 32, 4, 523 (1960).

All abbreviations of periodicals in the above bibliography are letter-by-letter transliterations of the abbreviations as given in the original Russian journal. *Some or all of this periodical literature may well be available in English translation.* A complete list of the cover-to-cover English translations appears at the back of this issue.
