Separation Science and Technology
Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713708471

Selective Floatation of Inorganic Ions

W. Charewicz[a]; W. Walkowiak[a]
[a] TECHNICAL UNIVERSITY OF WROCĹW INSTITUTE OF INORGANIC CHEMISTRY AND METALLURGY OF RARE ELEMENTS, WROCŁW, POLAND

To cite this Article Charewicz, W. and Walkowiak, W.(1972) 'Selective Floatation of Inorganic Ions', Separation Science and Technology, 7: 6, 631 — 646
To link to this Article: DOI: 10.1080/00372367208057972
URL: http://dx.doi.org/10.1080/00372367208057972

SEPARATION SCIENCE, 7(6), pp. 631–646, 1972

Selective Floatation of Inorganic Ions

W. CHAREWICZ and W. WALKOWIAK

TECHNICAL UNIVERSITY OF WROCŁAW
INSTITUTE OF INORGANIC CHEMISTRY AND METALLURGY OF RARE ELEMENTS
WROCŁAW, POLAND

Summary

On the basis of both the literature data and the results of our own studies, some chemical and physicochemical aspects of selective flotation of inorganic ions was discussed. Classification of properties and events that determine the flotated ion separation, such as ionic charge, base–acid equilibria, complexation processes, solubility of ion-collector compounds, and ion affinity to surfactants have been described and exemplified.

INTRODUCTION

Development of modern methods of chemical metallurgy—especially hydrometallurgy—has always been and still is determined by improvement of technique for concentration and separation of solution components. Extraction and ion exchange are very important methods, widely applied in practice, also being very helpful research instruments. Theoretical and experimental studies on ion flotation, initiated by Sebba (*1*) and now developed broadly, show that this process of concentrating ions in aqueous solution is the most effective method of the ones developed up to now, especially for low concentration ranges (*1*, *2*).

The efficiency of a physicochemical method of concentration and separation is determined by the removal ratio, enrichment, and selectivity. Removal ratio and enrichment are the functions of technical parameters such as concentration, medium flow rate, and mixing;

selectivity is determined by physicochemical processes. In the case of ion exchange or extraction, selectivity of a system could be established from suitable equilibrium constants, of which a large number have already been found experimentally.

The problem of ion-flotation selectivity is more complicated because in this case it is necessary to use the gas–liquid, interfacial surface, formed during the process. The separated substances do not diffuse through the phase boundary and are not stably bonded there (*2*). Sebba (*1*) in his considerations on thermodynamics, treated the problem generally. According to him the derived sublation constant F is undoubtedly equivalent to the ion-exchange equilibrium constant or to the extraction equilibrium constant. Values for F (*1*) indicate, to a certain extent, its analogy to the solubility of the ion-collector compounds. They indicate also that F depends essentially on the charge of the examined ion. The basic assumption for the above considerations is the thermodynamical equilibrium between the solution and the interfacial surface. However, because there are no simple methods for obtaining reproducible values F, it has little practical application.

Up to now results of investigations on ion flotation do not give sufficient data to describe fully all the phenomena, occurring simultaneously with that process. Ion binding by the collectors and factors conditioning that process are involved. In the case of surface-inactive inorganic ions, one cannot state whether formation of the ion-collector compounds precedes the absorption of binding formed on the gas–bubble surface or whether the reverse is true. It is very likely, that in both cases some physicochemical factors, such as ion charge, acid–base equilibria, complexation processes, and physicochemical properties of the components of the solution subjected to flotation, regulate the observed resolving power of the flotation methods of concentration of ions. These factors determine the ion-collector affinity. Very often they determine the real process rate obtained under experimental conditions.

On the basis of our results and those obtained by others, the problem of the selective flotation of inorganic ions, as dependent on the factor mentioned, is discussed in the present paper.

Experiments were performed on the apparatus described previously (*3*) using radioisotopes ^{198}Au, ^{193}Pt, ^{186}Re, ^{111}Ag, ^{103}Pd, ^{99}Mo and ^{58}Co as analytical tracers. The course of the processes examined was recorded by α-radiation spectrometry; for isotopes ^{63}Ni and ^{32}P the scintillation mediated of β-radiation detection was applied.

TABLE 1

Selective Flotation of Ag^+ and $AuCl_4^-$ Ions with Cationic Surfactant $(C_{16}H_{33}(C_2H_5)_2NHCl)$ at a Concentration of 2×10^{-4} *M*

$[AuCl_4^-]$ (mole/liter)	$[Ag^+]$ (mole/liter)	$\frac{[AuCl_4^-]}{[Ag^+]}$	$D_{AuCl_4^-}$ (%)	D_{Ag^+} (%)	$\phi_{AuCl_4^-/Ag^+}$
1×10^{-5}	1×10^{-6}	10:1	85	6	82.1
1×10^{-6}	1×10^{-6}	1:1	90	10	80.1
1×10^{-6}	1×10^{-5}	1:10	86	14	38.8

ELECTRIC CHARGE OF ION

The rule of ion flotation justifies the selectively toward ions of opposite charge. Separation of cations from anions can be realized by using both cationic and anionic surfactants. In the first case the cations remain in solution; in the second, the anions. This is illustrated by the results of flotation of mixed solutions of $AgNO_3$ and $KAuCl_4$ toward the cationic surfactant $(R)_3NH^+$ (Table 1) and the anionic surfactant $R\text{-}SO_3^-$ (Table 2) at a constant gas flow rate (nitrogen), at pH = 3.5 ± 0.1, and at a temperature of 22° ± 2°C. Knowledge of the stability constant of the chlorauric ion (*4*) allows calculation of the contribution of Au^{3+} cations from the chloraurate solution concentration. It is a quite remarkable contribution, and, for gold concentrations equal to 1×10^{-4} and 1×10^{-6} *M*, it amounts to 0.97 and 35.5%, res-

TABLE 2

Selective Flotation of Ag^+ and $AuCl_4^-$ Ions with Anionic Surfactant $[C_{12}H_{25}C_6H_4SO_3H$ + sulfonated ricinoleic acid (3:2)] at a Concentration of 1×10^{-3} *M*

$[AuCl_4^-]$ (mole/liter)	$[Ag^+]$ (mole/liter)	$\frac{[AuCl_4^-]}{[Ag^+]}$	$D_{AuCl_4^-}$ (%)	D_{Ag^+} (%)	$\phi_{Ag^+/AuCl_4^-}$
1×10^{-6}	1×10^{-5}	1:10	8	83	53
1×10^{-6}	1×10^{-6}	1:1	15	83	49
1×10^{-5}	1×10^{-6}	10:1	4	83	27.5

pectively. This explains the removal ratio values (D) obtained for gold subjected to flotation with the anionic surfactant (Table 2).

Similarly, partial flotation of silver ions with the cationic collector (Table 1) can result from the formation of negatively charged micelles $(AgCl)Cl^-$, since, under the experimental conditions, used the concentration of silver and chloride ions exceeded the solubility value of AgCl. In such a selective flotation the ion distribution coefficient $\phi_{Ag^+/AuCl_4^-}$ depends mainly on the concentration ratio of separated metals and increases with increasing concentration ratio.

ACID-BASE EQUILIBRIA

The characteristics for a number of amphoteric, transition metal ion transformations in water solutions of their oxygen compounds, could be the basis for selective ion flotation. Depending on concentration and pH in solutions of the oxygen compounds of these metals, there appear a number of different forms, such as MeO^{n+} oxycations, undissociated H_nMeO_m acids, polyanions, and simple anions of the MeO_m^{n-} type. Typical transformations of this kind are observed in water solutions of molybdate salts, where, in sufficiently acidic medium, molybdenyle oxycations MoO_2^{2+} are found. With increase of pH, MoO_2^{2+} oxycations change into undissociated molecules of molybdenic acid (H_2MoO_4) and of polymolybdenic acids, and next, through polyoxoanions, into the simpler forms $HMoO_4^-$ and MoO_4^{2-}, which dominate at pH values above 6 (*5–8*). In the case of some transition metals (e.g., rhenium) the stability of simple forms of oxyanions is observed in a broad pH range. Application of the cationic surfactant to the flotation of diluted and acidified solutions containing the oxygen compounds of both amphoteric and acidic metals, could lead to the selective flotation of the latter, whereas the former metal will remain in solution in the neutral or oxycationic unflotated form. The results of experiments with selective flotation of solutions of Mo(VI) and Re(VII) with cationic surfactants of the $(R)_3NH^+$ and $(R)_4N^+$ types are presented in Figs. 1 and 2. Since it follows from the relationship obtained for the separation of rhenium from molybdenum, advantage was taken of the fact that in the pH range 1–2 in diluted solutions, Mo(VI) appears mainly in the form of undissociated compounds of the $Mo(OH)_6$ or H_2MoO_4 type or in the form of MoO_2^{2+} oxycations, which could not have been bonded by sur-

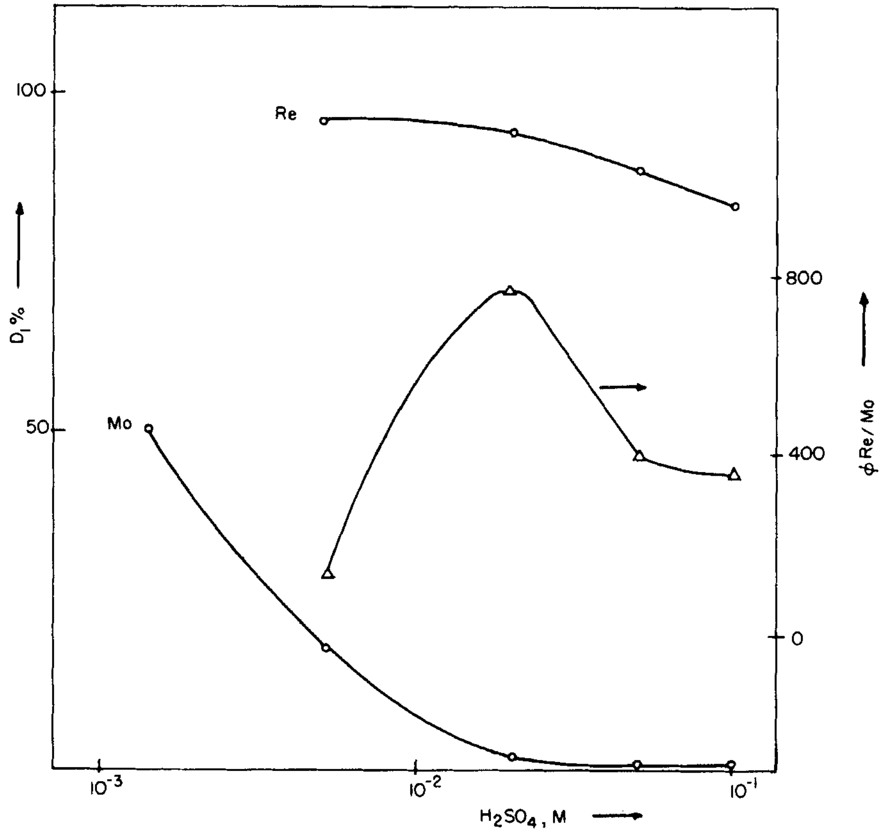


FIG. 1. Selective flotation of perrhenates and molybdates with diethylcetylammonium hydrochloride from sulfuric acid solutios. $[ReO_4^-] = [MoO_4^{2-}] = 2 \times 10^{-5}$ *M*; [DECAHCl] $= 2 \times 10^{-4}$ *M*; $N_2 = 28$ cm^3 min^{-1}; $t = 22° \pm 2°C$.

factants in the course of the experiment. Thus, Re(VII) appears in that range in the form of the stable ReO_4^- anions, originating from the strong perrhenic acid. Some lowering of the Re/Mo distribution coefficient, observed for surfactants of the $(R)_3NH^+$ type (Fig. 1) at higher sulfuric acid concentration, may result from the lowering of surfactant dissociation degree, particularly, since in the case of strongly basic surfactants of the $(R)_4N^+$ type such an effect did not appear.

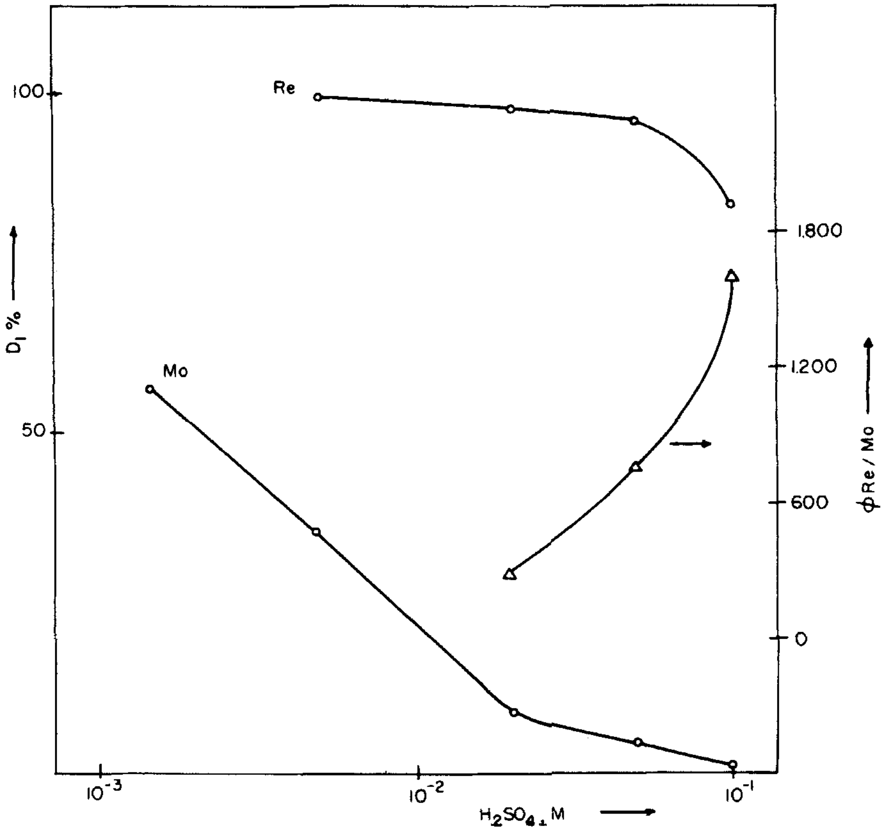


FIG. 2. Selective flotation of perrhenates and molybdates with dimethyllaurylbenzylammonium bromide from sulfuric acid solutions. $[ReO_4^-] = [MoO_4^{2-}] = 2 \times 10^{-5}$ *M*; $[DMLBABr] = 6 \times 10^{-4}$ *M*; $N_2 = 28$ cm³ min⁻¹; $t = 22° \pm 2°C$.

COMPLEX COMPOUNDS

The ionic structure of solution of complex compounds depends on stability, solubility, and other physicochemical properties of every component of the solution. Complexation processes affect the value and electric charge of the ion form of metals in a solution. Hence, an understanding of these processes could decide the success of the separation and the ion flotation kinetics. Conversion of cations into anion form, necessary because of the high activity of some cationic surfactants in a broad pH range, can be easily achieved due to the complexation processes of metal cations by simple ligands such as Cl^-, CN^-, CNS^-, or $S_2O_3^{2-}$.

The ability of complex binding with such ligands could be the basis for selective separation of metals, using the ion flotation method. Jacobelli-Turi and co-workers (*9*)—making use of the fact that Th(IV), unlike U(VI), does not form chloride complexes—they were able to separate these metals in the ion flotation with cationic surfacants of the type $(R)_4N^+$ in 8 *M* HCl medium. Similarly, the application of a cationic surfactant of the $(R)_4N^+$ type to the flotation of carbonate solutions of V(V) and U(VI) allowed the speration of these metals, based on the fact that uranium, unlike vanadium, under these conditions, easily forms the complex ions $UO_2(CO_3)_3^{4-}$ (*10*).

Stability of complex compounds also play an essential role in separation processes. In the case when with the same ligand the separated metals form two similar compounds that vary only in their stability, the more stable complex can be expected to arise at the lower ligand concentration. Karger *et al.* (*11*, *12*) by using the stability differences of Fe(III) and Hg(II) chloride complexes, have determined the selective flotation conditions for solutions of a mixture of these metals with a cationic surfacant of the $(R_4)N^+$ type in the presence of hydrochloric acid. The $HgCl_4^{2-}$ complex arises at an HCl concentration equal to about 0.5 *M*, whereas iron appears under these conditions in the form of a cation. The hydrochloric acid concentration necessary for tetrachloroferric acid formation amounts to about 8 *M*. Lusher and Sebba (*13*) have separated Al from Be by the ion flotation process of oxalate complexes of these metals with the cationic surfactant of the $R{-}NH_3^+$ type. The less stable $Be(C_2O_4)_2^{2-}$ complex decomposed under these experimental conditions and flotated rather slightly.

Stability of the metal ion compounds with surface-active chelating compounds can also lead to the selective flotation process. Jacobelli-Turi *et al.* (*14*), based upon the fact that the Zn^{2+} chelate complex with dodecylaminedipropionic acid is unstable at pH values above 12 (whereas the analogous Cu^{2+} compound is stable), have established the conditions for the selective ion flotation of these metals. Solubility is another property of the complex compounds which can lead to the above process. The low solubility of sodium fluoroaluminate allowed the separation of Al from Be in flotation process of their fluoride solutions with a cationic surfactant of the $R - NH_3^+$ type (*13*). The Na_3AlF_6

precipitate, in the form of negatively charged micelles, obtained in presence of excess fluoride ions, quickly rose to the foam layer with the cationic surfactant. It was typical for processes of this kind (*15*).

ION AFFINITY TO SURFACTANTS

Besides the factors discussed that determine the selectivity of the flotation methods of separation of surface-inactive inorganic ions from diluted aqueous solutions, the various physicochemical processes that regulate the real ion affinity to a surfacant are complicated and not satisfactorily explained. In such processes, under the same conditions, differences are observed in the rate or efficiency of the flotation, obtained for certain monomial ions in the flotated solution. The results of investigations carried out by Schonfeld *et al.* (*16*), who satisfactorily separated Sr^{2+} from Sm^{3+} in flotation of solutions containing aminopolycarboxylic acids, and studies performed by Jacobelli-Turi (*10*) on the foam separation of $UO_2(CO_3)_3^{4-}$ ions from VO_3^- ions, indicate the essential influence of the electric charge of separated ions on the observed affinity to the surfactants. Based an these studies as well as on flotation constants (*F*) determined by Sebba (*1*), it can be concluded that, in the case of anions, the affinity is proportional to the anion charge. This is an obvious conclusion from the point of view of physics and from the assumption that the ion-collector bonding process is electrostatic. In any case, it can be expected that such processes are much more complicated. For tetrahedral oxyanions of the AO_4^{n-} type, a higher affinity to the cationic surfactant should be expected in the case of higher charged anions at the same concentration and pH. Perrhenate and phosphate anions are good examples. Flotation of these ions with the cationic surfactant dimethyllaurylbenzylammonium bromide (DMBLABr) from alkaline aqueous solutions at a constant gas flow rate (nitrogen), a constant ionic strength of 0.001, and at a temperature of 22° ± 2°C is presented in Fig. 3.

It was calculated from dissociation constants of phosphoric acid (*17*) that, under experimental conditions, HPO_4^{2-} and PO_4^{3-} anions make-up 96 and 4%, respectively, of the total phosphate concentration. Under the same conditions, rhenium occurred only as 1− negative ReO_4^- anions. The results obtained (Fig. 3) are evidence of the higher affinity of the latter anions to the surfactants used as compared with HPO_4^{2-} anions. Moreover, the course of the flotation of examined ions indicates

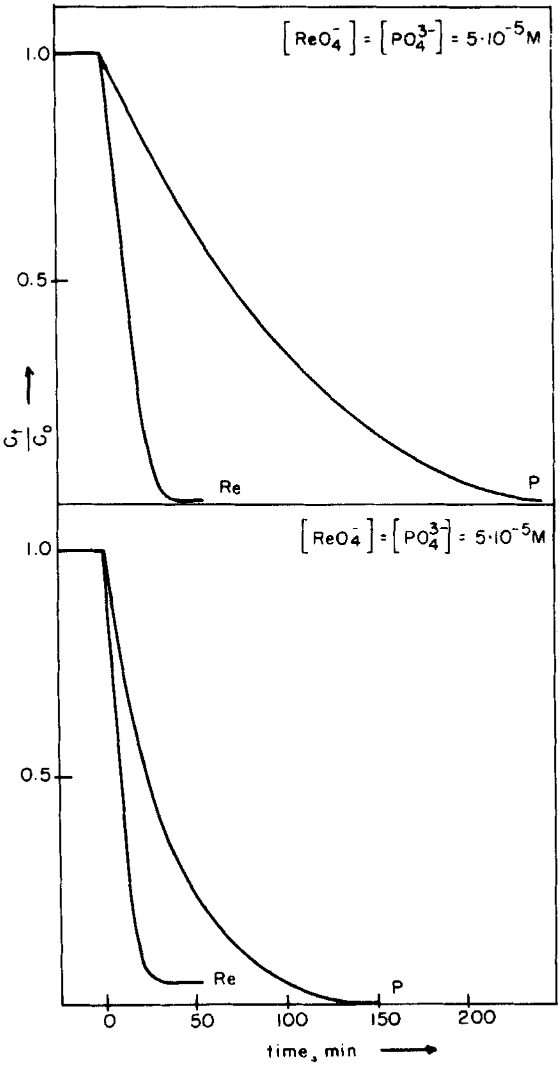
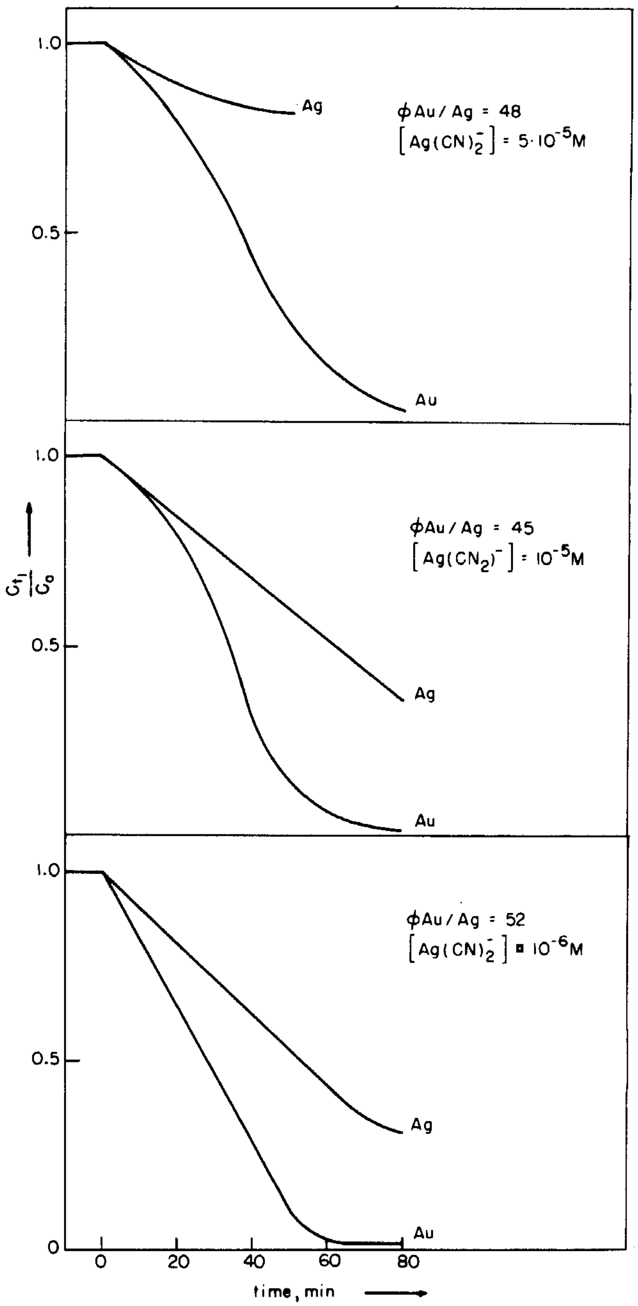


FIG. 3. Flotation of phosphates and perrhenates with the cationic surfactant DMLBABr versus concentration. [DMLBABr] = 8×10^{-4} M; pH = 11.2 ± 0.2; t = 22° ± 2°C. (C_t) Flotated ion concentration at time t (min); (C_0) initial concentration of flotated ion.


1.0
0.5
Ag
Au
ϕAu / Ag = 48
[Ag(CN)$_2^-$] = 5·10^{-5}M
1.0
0.5
C_t / C_o
Ag
Au
ϕAu / Ag = 45
[Ag(CN$_2$)$^-$] = 10^{-5}M
1.0
0.5
0
Ag
Au
ϕAu / Ag = 52
[Ag(CN)$_2^-$] = 10^{-6}M
0
20
40
60
80
time, min

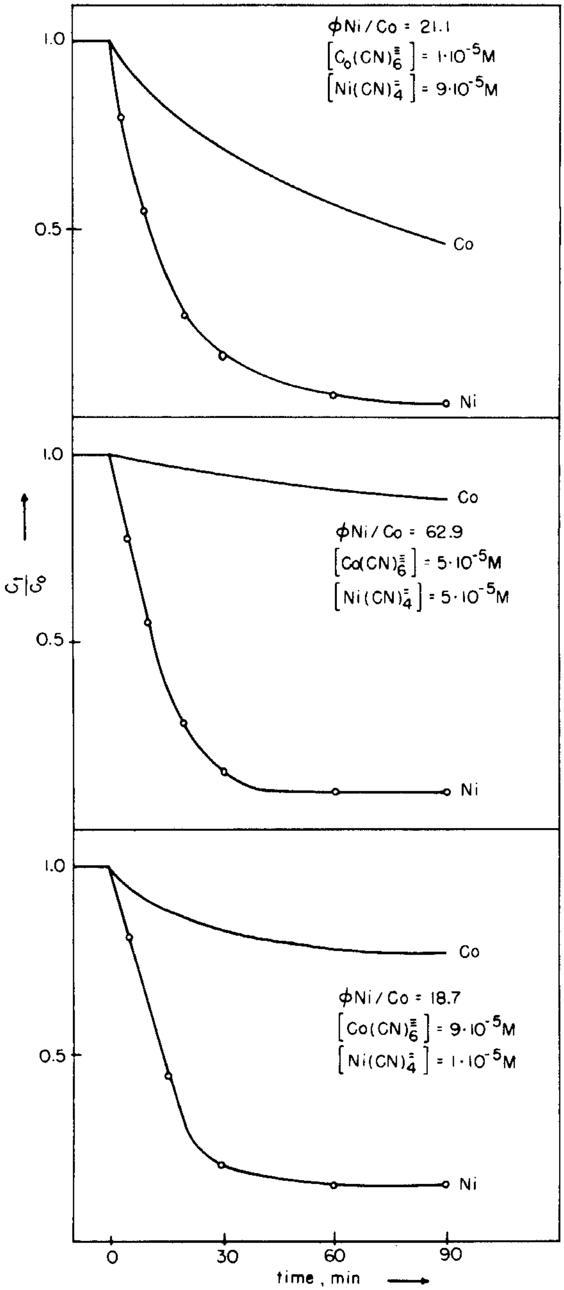
that, because if the essential difference in the rate of the process, separation of ions can be performed with high efficiency in a multistage ion flotation process with fractionated removal of the foam, the first fractions being enriched in rhenium.

In the case of monomial ions having similar structure and identical charged, the differences in their affinity to the surfactant, resulting from their physicochemical properties, make the selective flotation of a mixture, possible. Cyanide Ag(I) and Au(I) complexes, having identical linear structures, fit the above requirements. The results of flotation of these ion solutions with the cationic surfactant (DMBLABr) at stable concentration, gas flow rate (nitrogen), temperature, ionic strength, and pH are presented in the Fig. 4. (*18*). The greater affinity of cyanoaurate anion to the applied surfactant may result in this case from differences in stability or dimensions of the examined ions. The values obtained for the distribution coefficient indicate the possibility of selective isolation of gold from solutions containing a varying amounts of silver.

SOLUBILITY OF ION-COLLECTOR COMPOUNDS

Solubility of ion-collector compounds subjected to flotation with surfactants is another factor that can determine the ion flotation selectivity under certain conditions. Kinetics and mechanism of flotation of precipitates showing colloidal properties differ markedly from ion (*19*) flotation kinetics. Thus, in the case of the cyanide Co(III) and Ni(II) complexes, subjected to flotation with cetylpirydinium chloride (CPyCl)at constant gas flow rate, ionic strength, temperature, and pH, the essential differences in the process are observed (Fig. 5). Anions $Co(CN)_6^{3-}$ and $Ni(CN)_4^{2-}$ are stable complex ions (*4*) that differ in their charge and spatial structure, since the first has an octahaedral structure, and the second, a plain one. These factors, as well as the difference in number of coordinated ligands could be the reason that the cyanocobaltate anion—unlike $Ni(CN)_4^{2-}$—forms colloidal precipitate with the applied surfactant. The micelles without surface charge, because of the lack of excess free anions in solution, undergo slight flota-

FIG. 4. Flotation of $Au(CN)_2^-$ and $Ag(CN)_2^-$ with the cationic collector (DMLBABr) versus concentration. [DMLBABr] = 8 × 10^{-4} M; [$Au(CN)_2^-$] = 5 × 10 M; [CN^-] = 2 × 10^{-2} M; pH = 10; t = 21.5° ± 2°C.


φNi / Co = 21.1
[Co(CN)$_6^{\equiv}$] = 1·10^{-5}M
[Ni(CN)$_4^{=}$] = 9·10^{-5}M
1.0
0.5
Co
Ni
1.0
Co
φNi / Co = 62.9
[Co(CN)$_6^{\equiv}$] = 5·10^{-5}M
[Ni(CN)$_4^{=}$] = 5·10^{-5}M
0.5
Ni
C$_t$/C$_o$
1.0
Co
φNi / Co = 18.7
[Co(CN)$_6^{\equiv}$] = 9·10^{-5}M
[Ni(CN)$_4^{=}$] = 1·10^{-5}M
0.5
Ni
0
30
60
90
time, min

tion. This leads to essential separation of nickel from cobalt, even in the batch process.

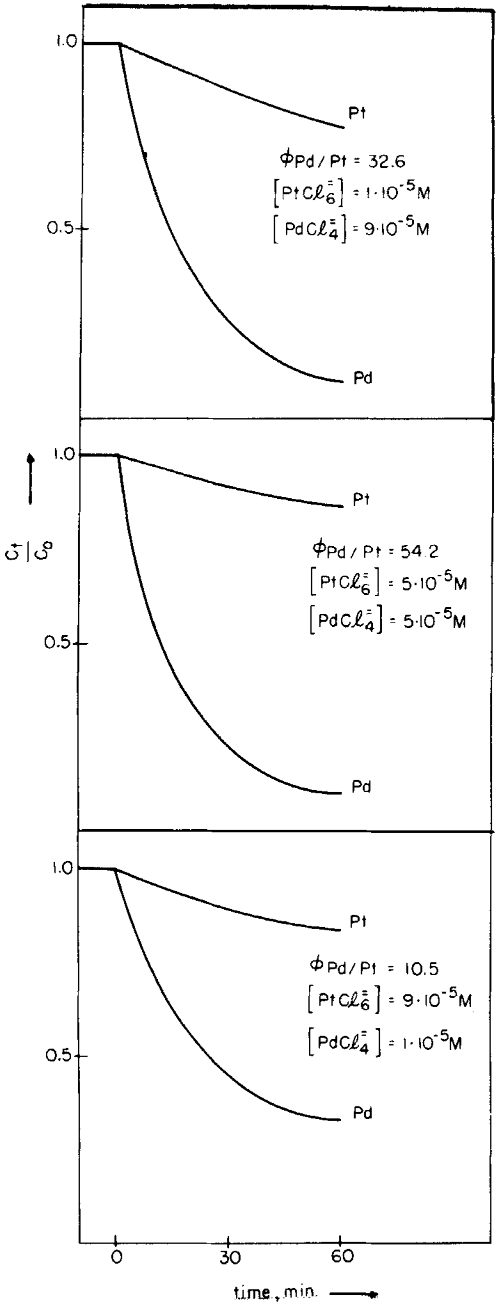
Selective batch flotation of the anion mixture is possible also in when the separated ions have the same charge but different structure and coordination number. The results of flotation of Pt(IV) and Pd(II) chloride complexes with the cationic surfactant dimethylbenzylcetylammonium chloride (DMBCACl) at constant gas flow rate, ionic strength, pH, and temperature, are shown in Fig. 6. Similarly as in the case of Ni(II) and Co(III) cyanate complexes, in the case of $PtCl_6^{2-}$ ions, formation of a colloidal precipitate with surfactant is observed. Under experimental conditions, it leads to the lowering of the platinum flotation efficiency. However, in contrast to the previous example, one deals here with anions of identical charge. It could be supposed, therefore, that the differences in the coordination number and in the structure of separated ions are the main cause of the phenomenon observed in both cases. The $PtCl_6^{2-}$ ion has octahaedral symmetry, whereas the $PdCl_4^{2-}$ ion is plain. The differences in solubility of ion-collector compounds, resulting from the properties of separated ions, could be the basis for selective flotation. Moreover, high separation coefficients can be obtained even in the batch process.

CONCLUSIONS

The material presented by us leads to the conclusion that it is very difficult to give a general and clear definition of factors determining the action selectivity of a particular surfactant toward various ions. The ionic structure of a solution undoubtedly plays an essential role in processes of selective ion flotation. The complexation processes, leading to a change in magnitude and regulating the kind of electric charge of an ion, are equally important.

By taking phosphorate, perrhenate oxyanions, and cyanoaurate and cyanosilver complexes as an example, it was shown, that in the case of anions having the similar spatial structure, the electric charge of an ion does not decide directly the affinity to the cationic surfacant and, thus, the flotation efficiency.

FIG. 5. Flotation of $[Co(CN)_6]^{3-}$ and $[Ni(CN)_4]^{2-}$ ions with the cationic surfactant CPyCl versus concentration. [CPyCl] = 5×10^{-4} *M*; pH = 8.3; $t = 21.5° \pm 1°C$.


1.0
0.5
Pt
Pd
$\phi_{Pd/Pt} = 32.6$
$[PtCl_6^{=}] = 1\cdot 10^{-5}M$
$[PdCl_4^{=}] = 9\cdot 10^{-5}M$
1.0
0.5
Pt
Pd
$\phi_{Pd/Pt} = 54.2$
$[PtCl_6^{=}] = 5\cdot 10^{-5}M$
$[PdCl_4^{=}] = 5\cdot 10^{-5}M$
C_t/C_o
1.0
0.5
Pt
Pd
$\phi_{Pd/Pt} = 10.5$
$[PtCl_6^{=}] = 9\cdot 10^{-5}M$
$[PdCl_4^{=}] = 1\cdot 10^{-5}M$
0
30
60
time, min.

The base–acid equilibria (characteristic for a number of amphoteric elements), leading to decay or even to the conversion of the electric charge, are also factors that determine the ionic structure of a solution and, thus, the ion-collector action selectivity. The position of these equilibria, resulting from differences in the strength of suitable oxygen acids, allows one to choose a pH range, so that only a fixed element of the mixture examined would be subjected to flotation.

The selectivity of a collector toward oppositely charged ions, based on assumption and confirmed by the example of chloroaurate and silver ions, has been proved. With the example of Pt(VI) and Pd(II) chloride complexes as well as Co(III) and Ni(II) cyanide, the essential role of the solubility of ion-collector compounds has been shown. It can be assumed that in these cases the coordination number and spatial structure of ions have the general meaning.

The properties and phenomena discussed above lead to two main kinetic effects: (*1*) decay or reduction of flotation of a particular component of the solution and (*2*) differences in the process rate in relation to several components. In the first case the separation of components can be achieved by the batch flotation process; in the latter case, the foam product in the faster flotated component can be enriched by fractionated removal of the concentrate.

REFERENCES

1. F. Sebba, *Ion Flotation*, Elsevier, Amsterdam, 1962; *A.I.Ch.E-Chem. Eng., Symp. Ser.*, *1*, 14 (1965).
2. W. Charewicz and J. Niemiec, *Wiadomości Chem.*, *20*, 693 (1966).
3. W. Charewicz and J. Niemiec, *Nukleonika*, *14*, 17 (1969).
4. *Stability Constants. Part II, Inorganic Ligands*, The Chemical Society, London, 1958.
5. J. Chojnacka, *Roczniki Chem.*, *39*, 161 (1965); Theory of the Structure of Complex Compounds, *Paper Symp., Wroclaw, Poland, 1962*, pp. 669 (1964).
6. I. B. Jacymirskij and I. J. Aleksiejeva, *Zh. Neorgan. Khim.*, *4*, 318 (1959); *ibid.*, *8*, 2513 (1963); *ibid.*, *12*, 1840 (1967).
7. A. Bartecki and D. Dembicka, *Roczniki Chem.*, *39*, 1783 (1965); *J. Inorg. Nucl. Chem.*, *29*, 2907 (1967).
8. Y. Sasaki, I. Lindquist, and L. G. Aillen, *J.Inorg.Nucl.Chem.*, *9*, 93 (1959).
9. C. Jacobelli-Turi, A. Barocas, and S. Terenzi, *Ind. Eng. Chem.*, *6*, 163 (1967).

FIG. 6. Flotation of $PtCl_6^{2-}$ and $PdCl_4^{2-}$ ions with the cationic surfactant DMBCACl versus concentration [DMBCACl] $= 2 \times 10^{-4}$ *M*; pH = 3.4; $t = 20.5° \pm 0.5°C$.

10. C. Jacobelli-Turi, S. Terenzi, and M. Palmera, *Ind. Eng. Chem.*, *6*, 161 (1967).

11. B. L. Karger, R. P. Poncha, and M. M. Miller, *Anal. Letters*, *1*, 437 (1968).

12. B. L. Karger and M. M. Miller, *Anal. Chim. Acta*, *48*, 273 (1969).

13. J. A. Lusher and F. Sebba, *J. Appl. Chem.*, *15*, 577 (1965); *16*, 129 (1966).

14. C. Jacobelli-Turi, M. Palmera, and A. Margani, *Ric. Sci.*, *36*, 1198 (1966).

15. W. Walkowiak and W. Charewicz, *Wiadomości Chem.*, *25*, 419 (1971).

16. E. Schonfeld, R. Sanford, G. Mazzella, D. Ghosh, and S. Mook, *U.S. At. Energy Comm. Rept.*, NYO-9577, July 1960.

17. D. Freiser and Q. Fernando, *Ionic Equilibria in Analytical Chemistry*, Wiley, New York, 1963.

18. W. Charewicz and T. Gendolla, *Chem. Stosowana*, *16*, 383 (1972).

19. W. Walkowiak, unpublished data (1971).

Received by editor March 16, 1972