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Negative Rejection of Group Ib Metal Cyanide Complexes in the Hyperfiltration by Cellulose Acetate Membranes. Donnan Membrane Effect*

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Abstract

Anionic cyanide complexes of Group Ib family metal ions, $Cu^I(CN)_2^-$, $Ag^I(CN)_2^-$, and $Au^I(CN)_2^-$, showed negative rejection in hyperfiltration through cellulose acetate membranes when the feed solution contained such polyvalent anions as succinate, tartrate, aspartate, ethylenediaminetetraacetate, polyacrylate, polyvinylsulfonate, phosphate, and sulfate in a substantial concentration. The dicyanometal complex anions are essentially membrane permeable, while the polyvalent anions are essentially membrane impermeable. Both types of anions combined with the membrane permeable countercations form an ionic ternary system in which the Donnan membrane effect operates to enhance the diffusion of the permeable salt through the membrane, resulting in its enrichment in the permeate solution. By taking advantage of this phenomenon, a selective enrichment of dicyanoargentite in the permeate and its separation from the coexisting transition metals were achieved. Based on this principle, a new hyperfiltration-ultrafiltration technique was proposed for the ion-selective separation of salts.

INTRODUCTION

Desalting or separation of solutes from aqueous solutions by reverse osmotic permeation or hyperfiltration is now well established, and many

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basic data as well as practical information concerning this process have been collected and compiled (*1*, *2*). These studies have, naturally, been concerned mainly with the rejection characteristics of the solutes, and not with the selective permeation of some specific solute of interest which is dissolved in solution. It would be intriguing and also highly valuable from a practical point of view if selective separation of certain solutes in solution was attained by using a hyperfiltration membrane under hydraulic pressure.

With regard to the above notion, it is notable that Golomb in 1972 observed that in hyperfiltration treatment of citrate-buffered plating waste containing dicyanoaurite(I), the permeate contained more gold than the feed solution when cellulose acetate membranes were used (*3*). Later, Kamizawa et al. restudied the same system and found that in multi-ion solutions, such as gold plating rinse, a less permeable ion such as citrate improved the permeability of a more permeable ion of like charge such as $Au(CN)_2^-$ (*4*). In our present study the phenomenon of negative rejection of dicyanoaurite(I) is treated from a broader view, and its cause and origin are presented in a more explicit way. The phenomenon is extended to other metal cyano complexes as well as to other interfering salts. A new separation technique which takes advantage of such a phenomenon is also proposed.

EXPERIMENTAL

Membranes

The preparation of cellulose acetate membranes followed the method of Manjikian (5). Cellulose acetate (Eastman Kodak, acetyl content 39.8 ± 0.5%, ASTM viscosity 3 ± 1 s, 17 g) was dissolved in a mixture of formamide (27 g) and acetone (56 g), and cast to a 200 μm-thickness. The solvent was allowed to evaporate at 25°C for 30 s. The membrane was then treated with water at 0–4°C and finally annealed in water at 40–90°C. The membranes were tested for salt rejection and permeate fluxes, and the results were analyzed according to the theory of the solution diffusion model (*6*), which confirmed that the membranes prepared in the present study were typical of cellulose acetate membranes.

Reverse Osmosis

A batch-type hyperfiltration cell of 13.8 cm^2 effective membrane area and 300 mL feed capacity (Atsuryoku Kiki Engineering Co.) was used. The permeation experiments were ordinarily performed at 18–20 atm and 25°C. Before each permeation experiment the membrane was successively tested for water flux with pressurized pure water and for the salt rejection with pressurized 0.535% aqueous sodium chloride solution. After the permeation experiment the membrane was again tested in the same manner, and the absence of any damage or deterioration during the experiment was confirmed. A new membrane was used for each permeation experiment in order to avoid any irreducible absorption of solutes on the membrane or change in the membrane performance.

The permeation experiments were carried out with a 250-mL feed solution, and the permeate was fractionated to 30–60 mL on a fraction collector. The salt concentration in the fractionated permeate was determined conductometrically for single-metal salt solutions and atomic absorption spectrophotometrically for mixed-metal salt solutions. The sensitivity of atomic absorption photometry was often strongly affected by the coexisting salts, and the appropriate calibration line was constructed for each determination of the metal. The rejection (R, %) of the solute and the flux of the permeate solution (F, $\mathrm{Lh^{-1}m^{-2}}$) were calculated for each fractionated permeate. Equation (1) defines the rejection, where C_f and C_p are the concentrations of the solute in the feed and the permeate, respectively:

$$R = 100(C_f - C_p)/C_f \tag{1}$$

RESULTS AND DISCUSSION

Negative Rejection

The negative rejection of complex ions has been reported only for dicyanoaurite (*4*). However, it is quite reasonable to expect similar behavior in dicyanoargentite(I), $Ag(CN)_2^-$, since both complex anions are structurally and chemically alike. Thus the permeation behavior of dicyanoargentite ion through cellulose acetate membranes was studied in the presence of various added salts. The results are summarized in Table 1.

The following are observed in Table 1. a) Sodium dicyanoargentite is essentially more permeable through the cellulose acetate membrane than sodium chloride (Runs 1 and 2). Separate experiments further indicated that sodium dicyanoargentite was more permeable than sodium nitrate, which in turn was more permeable than sodium chloride. b) The effect of salts with membrane-impermeable cations is small (Runs 4 and 5). c) The salts with membrane-impermeable or less-permeable anions cause a marked decrease in the rejection of dicyanoargentite. The less permeable are the constituent

TABLE 1

Effect of Additives on the Rejection of $Ag(CN)_2^-$ [a]

Run	Additive	R_{Ag}[b] (%)	F_{soln}[c] ($Lh^{-1}m^{-2}$)	R_{NaCl}[d] (%)	F_{water}[e] ($Lh^{-1}m^{-2}$)
1	None	26	63	56	74
2		59	25	85	29
3	Potassium nitrate[f]	43	19	85	26
4	Ethylenediamine hydrochloride	23	32	76	35
5	Tetraethylammonium chloride	28	31	76	35
6	Acetic acid	4.9	21	92	21
7	Polyacrylic acid[g]	3.1	23	76	26
8	Succinic acid	−44	22	89	27
9	Tartaric acid	−57	27	87	29
10	Citric acid	−74	24	86	26
11	Ethylenediamine-tetraacetic acid	−65	22	93	24
12	Polyethylene glycol[h]	58	28	74	40

[a] Feed solution: 5×10^{-4} M $NaAg(CN)_2$, 0.01 M (1 M = 1 mol/dm^3) added salt, pH 7.0 ± 0.1. (Potassium hydroxide and hydrochloric acid were used when acids and bases were neutralized.) Applied pressure, 19 ± 0.5 atm; 25 °C. Sixty milliliters of the initial permeate were collected from the 250-mL feed.

[b] Rejection of $Ag(CN)_2^-$.

[c] Flux of permeate solution.

[d] Rejection of sodium chloride; see Experimental section.

[e] Flux of water as measured against pure water; see Experimental section.

[f] Concentration, 0.1 M.

[g] $\bar{n} = 2000$, 0.1 wt%, 0.014 M (monomer unit).

[h] $\overline{MW} = 20{,}000$, 0.5 wt%.

anions, the more pronounced is the effect (Runs 6–11). The rejection of dicyanoargentite can even be negative (Runs 8–11). d) The state of ion dissociation also seems important (Runs 6 and 7); the degree of proton dissociation is less for polyacrylic acid at pH 7.

The negative rejection of dicyanoargentite was studied further in detail for the citrate system. Figure 1 shows the relation between the negative rejection of dicyanoargentite (R_{Ag}) and the membrane performance as estimated by the rejection of sodium chloride (R_{NaCl}). As the value of R_{NaCl} increased, the flux of the permeate solution decreased monotonously as should be the case. The value of R_{Ag}, on the other hand, first decreased until R_{NaCl} reached around 85% and then increased as R_{NaCl} increased further, showing a maximum negative rejection with the membrane of R_{NaCl} around 85%. This behavior indicates that negative rejection or the abnormally enhanced

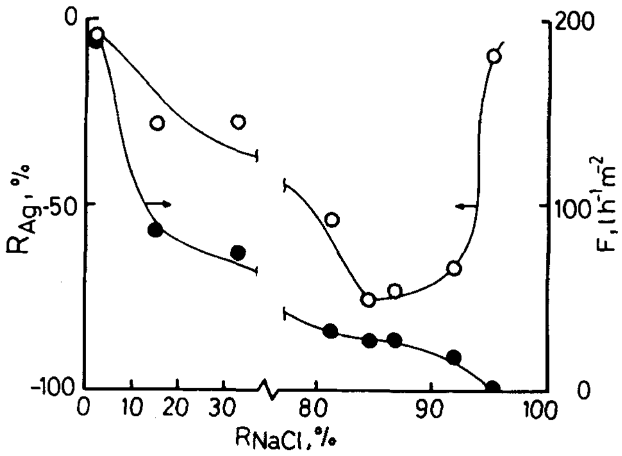

FIG. 1. Effect of membrane performance on the rejection of dicyanoargentite. R_{Ag}, rejection of $Ag(CN)_2^-$; R_{NaCl}, rejection of NaCl (see experimental section); F, flux of the permeate solution. 0.01 M potassium citrate (pH 7.0 ± 0.1), 5×10^{-4} M $NaAg(CN)_2$, 19 ± 0.5 atm. Sixty milliliters of the initial permeate were collected from the 250-mL feed.

permeation of dicyanoargentite is coupled with the flow of water through the membrane.

The effect of salt concentration on the negative rejection of dicyanoargentite was studied for citrate, succinate, and tartrate (Fig. 2). The effect in reducing the rejection of dicyanoargentite began at a salt concentration of around 10^{-4} M and gradually became remarkable with an increase in concentration. Citrate was more effective than succinate or tartrate in enhancing negative rejection. Citric acid is a tribasic acid and, under permeation conditions (pH 7.0), a considerable fraction of the acid exists as triply-charged conjugate anion, which should be more resistant to permeation through the membrane than the doubly-charged conjugate anions derived from either of the other two carboxylic acids.

The experiments in Fig. 2 were analyzed in terms of water and dicyanoargentite fluxes. The results are summarized in Fig. 3. The water fluxes decreased as the concentration of the added salts increased owing to the increased osmotic counterpressure. On the contrary, the dicyanoargentite flux increased, at least up to the 0.01 M salt concentration. This again indicates the importance of the concentration of the membrane-impermeable anions for the occurrence of substantially negative rejection of the complex anion.

The effect of applied hydraulic pressure was studied (Fig. 4). The added salts were rejected much more efficiently than sodium chloride at all

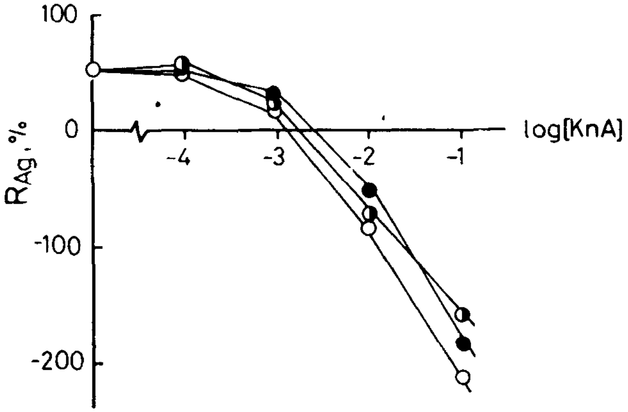

FIG. 2. Effect of the added salt concentration on the rejection of dicyanoargentite. Membrane: R_{NaCl} 85% (citrate ○), 89% (succinate, ●), 81% (tartrate, ◑). Feed: 5×10^{-4} *M* $NaAg(CN)_2$ with citrate, succinate, and tartrate (potassium salt, KnA, pH 7.0 ± 0.1) at 19 ± 0.5 atm. Sixty milliliters of the initial permeate were collected from the 250-mL feed.

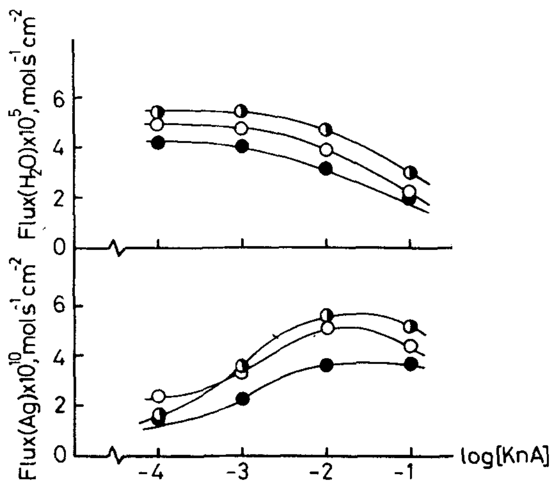

FIG. 3. Effect of the added salt concentration on the mole fluxes of water and dicyanoargentite. Conditions and keys are the same as those in Fig. 2.

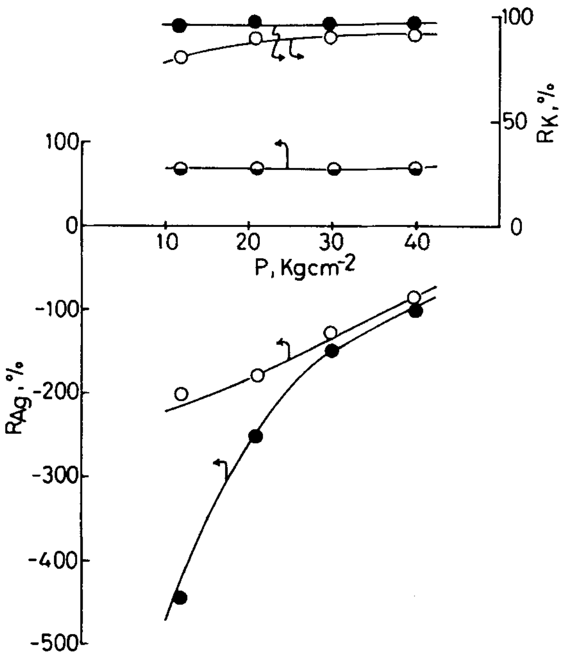

FIG. 4. Effect of applied pressure on the rejection of dicyanoargentite. Membrane: R_{NaCl} 95% (citrate, ○), 65% (succinate, ●), 86% (without added salt, ●). Feed: 5×10^{-4} *M* $NaAg(CN)_2$ with 0.1 *M* citrate and 0.1 *M* succinate (potassium salt, pH 7.0 ± 0.1) and without added salt at 19 ± 0.5 atm. Sixty milliliters of the initial permeate were collected from the 250-mL feed.

pressures used, and their rejections were increased as the pressure increased. A similar trend was observed with the rejection of dicyanoargentite in a much more pronounced manner. It was most certainly due to the dilution effect of the increased water flux under higher pressure.

The effect of pH of the feed solution was studied by using dicyanoaurite rather than dicyanoargentite, because the former was more stable under acidic conditions. Potassium succinate was used between pH 5 and 7. The results are shown in Fig. 5. Obviously, the decrease in the negative rejection of dicyanoaurite paralleled the decrease in the fraction of the dianionic species of succinate.

A combined use of nitrate and succinate resulted in the cancellation of the negative rejection of dicyanoargentite exhibited by the simple succinate system (Fig. 6).

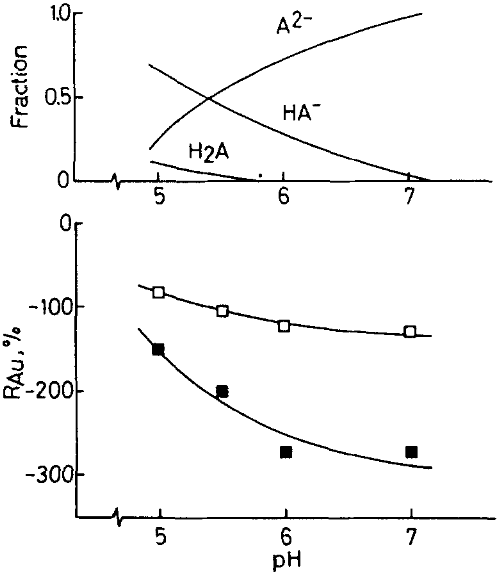

FIG. 5. Effect of pH of the feed solution on the rejection of dicyanoaurite(I), and the pH distribution of the ionic species of the added succinate. Membrane: R_{NaCl} 50% (□), 88% (■). Feed: 5×10^{-4} M $\mathrm{NaAu(CN)_2}$ with 0.1 M potassium succinate ($\mathrm{H_2A}$–KOH) at 19 ± 0.5 atm. Sixty milliliters of the initial permeate were collected from the 250-mL feed.

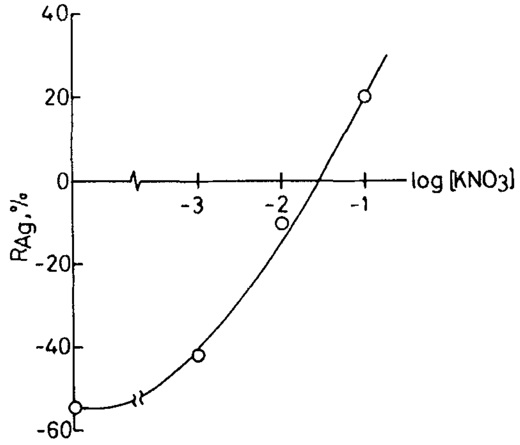

FIG. 6. Effect of nitrate concentration on the rejection of dicyanoargentite in the succinate system. Membrane: R_{NaCl} 74%. Feed: 5×10^{-4} M $\mathrm{NaAg(CN)_2}$ with 0.01 M potassium succinate (pH 7.0 ± 0.1) at 19 ± 0.5 atm. Sixty milliliters of the initial permeate were collected from the 250-mL feed.

Mechanism of Negative Rejection

All the experimental observation presented above are phenomenally in good accord with the idea of the Donnan-membrane effect in the hyperfiltration of a ternary system, which was theoretically and experimentally fully described for Na^+ (membrane-permeable cation)–Cl^- (permeable anion)–citrate (impermeable anion) system by Lonsdale and Pusch (*7*). In the treatment of a citrate-buffered gold plating rinse (*3*), K^+ and $Au(CN)_2^-$ replace Na^+ and Cl^-, respectively, in the ternary system. In the present study, $Ag(CN)_2^-$ or $Au(CN)_2^-$ replaces Cl^-, and the anionic component of the additives in Runs 6–11 in Table 1 substitutes the role of citrate in the Donnan-membrane effect.

Kamizawa et al. (*4*) reported that among the membranes tested (cellulose acetate, polybenzimidazolone, aromatic polyamide, composite membrane NS-200 with a coating of sulfonated polyfurfuryl alcohol over a microporous polysulfone support), only cellulose acetate membranes exhibited meaningful reduction in the rejection of $Au(CN)_2^-$ in the gold plating rinse. Lonsdale and Pusch (*7*) in their theoretical treatment clarified that a slight decrease in the rejection characteristics of the membrane toward a permeable ion causes, in the ternary system with an impermeable co-ion, a strong decrease in the rejection of the permeable salt, readily leading to the observable negative rejection. As is seen in Table 1, sodium dicyanoargentite is quite permeable through the cellulose acetate membrane than is sodium chloride. A special affinity of this type of complex salts toward cellulose acetate can be easily confirmed by their absorption on the cellulose acetate membranes or cellulose acetate powder. There is no doubt that the negative rejection of dicyanoaurite observed only with the cellulose acetate membrane (*4*) was merely an exaggerated manifestation of this affinity.

It was also reported that the modification of the cellulose acetate membrane with anionic or cationic dye did not destroy the low rejection characteristics of gold in the treatment of the plating rinse (*4*). This is in accord with the above idea that the rejection is essentially not due to the fixed charge in the membrane but due to the Donnan effect between the two aqueous solutions separated by a semipermeable membrane. In fact, the negative rejection of sodium dicyanoargentite could be experimentally fully simulated under unpressurized permeation conditions. A two-compartmented glass cell (*8*) was separated by a cellulose acetate membrane. A "feed" solution containing sodium dicyanoargentite and an appropriate salt was placed in one side (feed side) of the cell, and an equal volume of pure water was placed in the other (permeate side). The permeation of dicyanoargentite was monitored to give the time–concentration profiles shown in Fig. 7.

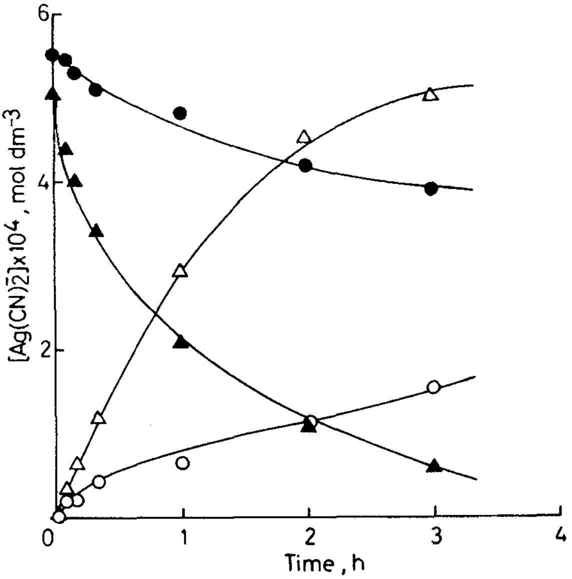

FIG. 7. Permeation of $Ag(CN)_2^-$ through cellulose acetate membrane under unpressurized conditions. Membrane: R_{NaCl} 90%. Feed side: 5×10^{-4} M $NaAg(CN)_2$. Permeate side: pure water. Concentrations of $Ag(CN)_2^-$ are in the feed (●, ▲) and in the permeate (○, △) sides, with (▲, △) and without (●, ○) the added 1 wt% polyacrylic acid ($\bar{n} = 2000$). The feed solution was neutralized to pH 7.0 ± 0.1 with KOH. See text for further experimental conditions.

Though a small correction for the dilution effect due to the osmotic water flux was not made, it is clear that dicyanoargentite is eventually concentrated in the permeate side. Similar results were obtained when citrate or succinate was used as the added salt.

Possible Application of Negative Rejection in Separation of Salts

In the consideration of a Donnan-membrane effect for possible application in separation chemistry, it would be important to know at the beginning, a) the range of commonly available "membrane-impermeable" ions, and b) to what extent a specific ion such as $Ag(CN)_2^-$ can be negatively rejected or enriched in the permeate under reasonable concentrations of the membrane-impermeable co-ions. Table 2 summarizes the results of such a study. A variety of organic and inorganic anions is usable, but from a practical point of view, polymer electrolytes such as polyacrylate and polyvinylsulfate should be most convenient because of their complete impermeability and the ease of recovery (see below).

For a possible separation of silver and gold from some transition metals in their cyano complexes, the rejection behavior of copper(I), nickel(II), and zinc(II) to cellulose acetate membranes was studied in the presence of cyanide ion. The results are summarized in Table 3. It is seen that the negative rejection is characteristic of the Group Ib family metal ions. The ionic size, charge, and extent of hydration would no doubt be contributing factors, but the details are yet to be explored. The following are also noteworthy: a) the rejection of the copper(I) species is dependent markedly on cyanide concentration, b) the cyanide complexes of nickel(II) and zinc(II) are rejected to the same extent as sodium chloride. As to the former point, it is to be further added that the rejection of copper(II) was also dependent on the cyanide concentration. In fact, cupric ion was quantitatively reduced to cuprous ion by the added cyanide ion. Thus, under conditions similar to those in Table 3, the rejection of copper(II) dropped from the initial 85% to −80% by the addition of a fourfold molar excess of cyanide ion. A further addition of cyanide then increased the rejection, approaching the value in Table 3, Run 4.

TABLE 2

Effect of Coexisting Salt on the Negative Rejection of $Ag(CN)_2$[a]

Run	Added salt	R_{Ag}[b] (%)	F_{soln}[b] ($Lh^{-1}m^{-2}$)	R_{NaCl}[b] (%)	F_{water}[b] ($Lh^{-1}m^{-2}$)
1	Potassium succinate	−195	13	89	27
2	Potassium tartrate	−159	20	81	35
3	Potassium citrate	−223	14	85	32
4	Potassium aspartate	−129	20	76	37
5	Potassium polyacrylate[c]	−82	21	76	37
6	Potassium phosphate	−111	21	74	40
7	Sodium sulfate	−105	24	76	37
8	Magnesium sulfate	11	25	76	37

[a] Feed: 5×10^{-4} M $NaAg(CN)_2$ and 0.1 M added salt at 19 ± 0.5 atm. The pH was adjusted at 7.0 ± 0.1 with KOH. Sixty milliliters of the initial permeate were collected from the 250-mL feed.

[b] Refer to Table 1 for definitions.

[c] $\bar{n} = 2000$, 0.5 wt% acid.

TABLE 3
Rejection of Metal Cyanide Complexes by Cellulose Acetate Membrane[a]

Run	Solute[b]	$T_{CN} \times 10^3$ [c] (M)	R_M[d] (%)	F[e] ($Lh^{-1}m^{-2}$)	R_{NaCl}[e] (%)	F[e] ($Lh^{-1}m^{-2}$)
1	$Au(CN)_2^-$	1	−227	13	88	24
2	$Ag(CN)_2^-$	1	−195	13	89	27
3	$Cu(CN)_2^-$	2	−64	17	83	34
4	$Cu(CN)_3^{2-}$	25	66	13	86	27
5	$Ni(CN)_4^{2-}$	25	83	15	84	31
6	$Zn(CN)_3^-$	25	80	18	83	34

[a] Feed: 5×10^{-4} M metal ion and 0.1 M potassium succinate (pH 7.0 ± 0.1) at 19 ± 0.5 atm.

[b] Representative composition of the complex species present in the feed solution (calculated by using the published stability constants of cyanide complexes; Ref. 9). For Runs 1 and 2, the metals are present solely in the ionic form indicated. For Run 3, copper(I) is in the approximately 1:1 mixture of $Cu(CN)_2^-$ and $Cu(CN)_3^{2-}$.

[c] Total concentration of CN^- ion including those added as potassium cyanide.

[d] Rejection of combined metal species.

[e] Refer to Table 1 for definitions.

The permeation of a mixed solution of cyanide complexes resulted in the selective enrichment of silver in the permeate (Table 4). This is in accord with the results in Table 3. A simple calculation shows that successive treatment of the permeate obtained in Table 4 by a similar procedure with renewed added citrate gives 60% recovery of silver, which is contaminated only by 2% of copper and 0.5% of zinc and nickel. This suggests a potential usability of the principle of the Donnan-membrane effect in the selective separation of ionic species by the hyperfiltration process. In order for such a process to be of practical value, the impermeable salts which affect the negative rejection ("activating salt") must be recycled and reused. This means that the concentrated feed solution after permeation must be depleted of the remainder of feed salts without losing the activating salt. Polymer electrolytes are the most suited for this purpose. The feed solution after permeation was subjected to ultrafiltration, where all the low-molecular-weight solutes are depleted. In fact, it was shown that by using potassium polyvinylsulfate as the activating salt, silver and gold were successfully concentrated in the permeate and separated from the other transition metals in the feed solution. The transition metals concentrated in the feed were then

TABLE 4
Permeation of a Mixed Metal Cyanide Solution[a]

Metal ion	$[\text{Metal}]_i \times 10^4$ [b] (M)	$[\text{Metal}]_f \times 10^4$ [b] (M)	Fraction of metal permeated[c] (%)
Ag(I)	4.4	1.9	77
Cu(I)	4.6	8.0	13
Z(II)	3.9	7.6	6.2
Ni(II)	4.3	8.2	6.8

[a] Membrane: R_{NaCl} 90%. Feed: $3.9\text{–}4.6 \times 10^{-4}$ M transition metal ion, 2.5×10^{-2} M KCN, 0.1 M potassium citrate (pH 7.0 ± 0.1). The feed solution (250 mL) was permeated to half of its volume under 19 ± 0.5 atm.

[b] Concentration of metal in the feed, before ($[\text{metal}]_i$) and after ($[\text{metal}]_f$) permeation.

[c] Percent fraction of the permeated metal to the total metal charged in the feed solution.

removed by filtration through an ordinary cellulose- or polysulfone-ultrafiltration membrane. The activating salt solution was then mixed with a new feed salt solution and subjected to hyperfiltration again. The details will be the subject of a separate publication.

It is finally emphasized that the negative rejection of salts, in principle, can be realized whenever *uncharged* membranes with ion-selective affinity and permeability are available. As an example with cellulose acetate membranes, picrate ion indicated an appreciable adsorption affinity and a permeability to the membrane. In accordance with this, picrate showed a strongly negative rejection under hyperfiltration conditions in the present study. In this respect, the prospect of the proposed separation technique is totally dependent on the development of such ion-selective, uncharged membranes in the future.

CONCLUSION

In hyperfiltration with cellulose acetate membranes, anionic cyanide complexes of Group Ib family metal ions, $M^I(CN)_2^-$ ($M = Cu^I$, Ag^I, Au^I), were negatively rejected or enriched in the permeate when the feed solution contained membrane-impermeable anions. The Donnan-membrane effect was responsible for this phenomenon. The phenomenon can, in principle, be used for practical purposes such as the selective separation of specific salts in solution from the remainder of the salts. The development of ion-selective uncharged membranes is the key for the success of this separation technique.

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