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# Competitive Adsorption of Gold Cyanide and Organic Compounds onto Porous Adsorbents

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## ABSTRACT

Activated carbon has had a tremendous impact on the technology of gold recovery from leached liquors. Ion-exchange resins have been proposed as possible alternatives to carbon, while ion-exchange fibers and membranes were investigated recently as adsorbents for metal recovery in view of their fast rate of uptake. This paper deals with the competitive adsorption of organic compounds and gold cyanide onto activated carbon, ion-exchange resin, ion-exchange fiber, and membrane. Loadings of organic compounds were measured on gold equilibrated adsorbents and compared to loadings on virgin adsorbents. Both the kinetic and equilibrium parameters in a film/surface diffusion model were affected, which indicated a competitive type of mechanism between gold cyanide and organic compounds. A two-component Freundlich-type isotherm fitted the equilibrium for adsorption on carbon, membrane, and fiber. Furthermore, low molecular mass organic substances revealed a smaller inhibiting effect on gold adsorption than long-chain or high molecular mass organic compounds.

## INTRODUCTION

The use of activated carbon for the recovery of gold from cyanide-leached liquors has gained wide acceptance in the mining industry. In most plants the carbon-in-pulp (CIP) process is the preferred method for gold recovery. Recently, ion-exchange resins have been examined as possible alternatives to the CIP process. However, resins are less selective than carbons in the presence of base metals and are affected negatively by high ionic strength solutions (1). More recently, ion-exchange fibers and membranes have been investigated as possible adsorbents for gold in view of their relatively large surface areas and relatively small pores sizes.

Most leached slurries contain various organic substances which can influence the extraction performances of these adsorbents negatively. Numerous authors have observed that organic compounds inhibit the adsorption of gold cyanide by activated carbon (2–5). Petersen (6) showed that low organic loadings on carbon affected only the rate of gold adsorption whereas high organic loadings affected both the rate and equilibrium of gold adsorption. Ion-exchange resins, fibers, and membranes have also shown a reduction in the rate of gold adsorption in the presence of organic contaminants (5, 7, 8). However, all of these studies expressed the negative effect of organic compounds in terms of variations in kinetic and equilibrium parameters of a single species (gold cyanide).

A number of approaches have been published to describe the equilibrium of multisolute organic mixtures on activated carbon (9, 10). Van Deventer (11) investigated the competitive effect of one metal cyanide on the adsorption performances of other metal cyanides. Young et al. (12) studied the competitive adsorption of gold cyanide and potassium ethyl xanthate on activated carbon using a binary equilibrium approach. Although these authors claimed that small amounts of ethyl xanthate influenced the gold loading significantly, no conclusive evidence on the exact mechanism was supplied. This means that it was not clear whether the gold loading was affected by the poisoning of carbon with organic foulants or whether a competition for active sites on the carbon existed between gold cyanide and ethyl xanthate.

It is the objective of this paper to elucidate the mechanism for the simultaneous adsorption of gold cyanide and organic species onto porous adsorbents. Binary equilibrium expressions will be used to model this two-component system based on a film/surface diffusion approach. Furthermore, the sensitivity in kinetic and equilibrium parameters will be used as an indication of the behavior of gold and an organic species in a two-component adsorption system.

## THEORY

The equilibrium solid-phase concentration of a single component adsorption system can be described only in terms of the equilibrium liquid-phase concentration of the adsorbate if all other experimental conditions are kept constant. In multicomponent mixtures the solid-phase loading of a particular adsorbate is determined not only by its equilibrium liquid-phase concentration but also by the competitive effect of the other components in solution. The Freundlich isotherm fitted the experimental data for the adsorption of a single gold cyanide species onto activated carbon.

For a two-component system the equilibrium model equations are developed on the basis of two solutes with different competitive adsorption behaviors as described by (10)

$$Q_{s,1} = A_1C_{s,1}(C_{s,1} + B_{12}C_{s,2})^{n1-1} \tag{1}$$

for Component 1, while the isotherm for Component 2 is

$$Q_{s,2} = A_2C_{s,2}(C_{s,2} + B_{21}C_{s,1})^{n2-1} \tag{2}$$

Equations (1) and (2) can be written in the form

$$f(C_{s,1}) = \left[\frac{A_1C_{s,1}}{Q_{s,1}}\right]^{1/(1-n1)} - C_{s,1} = B_{12}C_{s,2} \tag{3}$$

and

$$f(C_{s,2}) = \left[\frac{A_2C_{s,2}}{Q_{s,2}}\right]^{1/(1-n2)} - C_{s,2} = B_{21}C_{s,1} \tag{4}$$

With $f(C_{s,1})$ or $f(C_{s,2})$ plotted against $C_{s,2}$ or $C_{s,1}$, respectively, the competition coefficients $B_{12}$ or $B_{21}$ can be obtained from the slopes of the curves. Equations (1) and (2) are based on the assumption that Species 1 and 2 both follow the Freundlich isotherm in a single solute solution.

A dual resistance model incorporating both external film diffusion and intraparticle surface diffusion was used to monitor the two-species adsorption. Although this type of model is by no means novel, it is necessary to state the following main assumptions:

- The carbon and resin particles can be treated as equivalent spheres, while the fiber can be treated as an infinite cylinder for modeling purposes.
- Isothermal conditions are assumed during adsorption.
- End effects are ignored for the fiber and membrane, and diffusion is assumed to occur in one dimension.
- Accumulation of gold cyanide and organic compounds within the pores of the adsorbents is negligible.
- It is assumed that the adsorption reaction on the carbon, and the ion exchange reaction on the resin, fiber, or membrane, occur instantaneously, so that equilibrium exists at the solid–liquid interface.

- The diffusivities of gold cyanide and organic compounds remain constant during a run and are independent of the position inside the adsorbents.

All mass balance equations developed for carbon, resin, and fiber will be presented in radial coordinates, while those for the membrane will be recast in Cartesian coordinates.

## Material Balance inside Adsorbent (Eqs. 5)

Activated Carbon and Resin:

$$\frac{\partial Q}{\partial t} = D\frac{\partial^2 Q}{\partial r^2} + \frac{2D}{r}\frac{\partial Q}{\partial r}$$

Membrane:

$$\frac{\partial Q}{\partial t} = D\frac{\partial^2 Q}{\partial x^2}$$

Fiber:

$$\frac{\partial Q}{\partial t} = D\frac{\partial^2 Q}{\partial r^2} + \frac{D}{r}\frac{\partial Q}{\partial r}$$

## Material Balance for the Liquid Phase in a Batch Reactor (Eqs. 6)

Activated Carbon and Resin:

$$\frac{dC}{dt} = \frac{k_f A \beta}{V}(C_s - C), \qquad \text{where } A = \frac{6M_c}{\rho_c d_c} \text{ (carbon)}$$

$$A = \frac{6\epsilon V_R}{d_r} \text{ (resin)}$$

Membrane:

$$\frac{dC}{dt} = \frac{4M_m k_f \beta}{\rho_m d_m V}(C_s - C)$$

Fiber:

$$\frac{dc}{dt} = \frac{4M_f k_f \beta}{\rho_f d_f V}(C_s - C)$$

### Boundary Condition: No Accumulation Occurs at External Surface of Adsorbent (Eqs. 7)

Activated Carbon, Resin, and Fiber:

$$\frac{\partial Q}{\partial r_{r=R}} = \frac{k_f\beta}{\Phi D}(C - C_s), \qquad \text{where } \Phi = \rho_c \text{ (carbon)}$$

$$\Phi = \frac{1}{\epsilon} \text{ (resin)}$$

$$\Phi = \rho_f \text{ (fiber)}$$

Membrane:

$$\left.\frac{\partial Q}{\partial x}\right|_{x=a} = \frac{k_f\beta}{\rho_m D}(C - C_s)$$

### Equilibrium Expressions to Relate $C_s$ and $Q$ (Curve H)

See Table 2, below, for values of constants.

The Freundlich expression fitted the equilibrium for the carbon, membrane, and fiber:

$$Q_e = AC_e^n$$

The Langmuir expression fitted the equilibrium for resin:

$$Q_e = \frac{K_1 C_e}{K_2 + C_e}$$

A computer program was developed for the numerical solution of the model. Equations (5) were transformed into ordinary differential equations by the use of average metal loadings in the pores. The value of $C_e$ is guessed, and the calculations of Eq. (5) are repeated with a fourth-order Runge–Kutta method until Eq. (7) is satisfied.

## EXPERIMENTAL

Potassium aurocyanide, obtained from Barry Colne and Co. as a crystalline salt, was used as an adsorbate. A range of organic compounds was used as depicted in Table 1. All reagents were of analytical grade, and distilled deionized water was used throughout.

Adsorption tests were performed in 1 L baffled reactors which were agitated mechanically. It was found that at a stirring speed of 120 rpm

TABLE 1
Different Organic Compounds

| Organic compound | Molecular mass (g/mol) | Average size (Å) |
|---|---|---|
| Ethanol | 46 | 1.2 |
| Sodium ethyl xanthate (SEX) | 160.28 | 54 |
| Potassium amyl xanthate (PAX) | 144 | 49 |
| Phenol | 94.11 | 4.9 |

the adsorbents were completely in suspension. Equilibrium tests were conducted in stirred vessels for a period of 3 weeks for carbon, and 2 days for resin, fiber, and membrane.

A Varian Techtron AA-1275 atomic absorption spectrophotometer was used for the analysis of gold cyanide in solution. The organic foulant concentrations, except for ethanol, were determined by measuring the ultraviolet absorbance on an LKB Ultrospec II E ultraviolet spectrophotometer. The ethanol concentrations were measured by gas chromatography. The pH of all solutions was monitored by using a Beckman Chem-Mate pH-meter.

The following adsorbents were used:

(a) A coconut-shell-activated carbon, Le Carbone G210 AS, with an average diameter of 1.4 mm and an apparent density of 838 $kg \cdot m^{-3}$. The pore size of the carbon varies between 8 to 250 Å.

(b) A macroporous ion-exchange resin, Duolite A161, with an average diameter of 0.8 mm and a wet-settled density of 700 $kg \cdot m^{-3}$.

(c) An ion-exchange membrane, Ionac 3475, cut into 1 × 1 cm squares, and treated by washing alternately with a 5% sodium hydroxide solution and a 5% sulfuric acid solution.

(d) Three types of ion-exchange fibers, namely
    (i) Polypropylene-based strong-base fiber, designated Fiber A.
    (ii) Polypropylene-based weak-base fiber, designated Fiber B.
    (iii) Actilex B 402 weak-base fiber, designated Fiber C.

The ion-exchange materials as indicated in (b) to (d) yielded average pore diameters between 60 and 200 Å. Only selected results will be presented in this paper, although most of the observations will be discussed qualitatively.

## RESULTS AND DISCUSSION

Although numerous researchers have observed that organic substances affect the adsorption of gold cyanide onto activated carbon adversely, no substantial evidence on the exact mechanism of poisoning of carbon has been provided as yet. Two schools of thought are evident from the literature: (a) the organic substances block the pores of carbon permanently, thus rendering them inaccessible to gold adsorption (3, 5), and (b) there is competitive adsorption between gold cyanide and organic substances for active sites on the carbon's surface (12). Furthermore, a lack of information exists on the behavior of other adsorbents such as ion-exchange resins, fibers, and membranes in the presence of organic compounds. Although the results presented here only used sodium ethyl xanthate (SEX) as the organic compound, similar results were obtained for the other organic compounds.

### Comparative Performance of Different Adsorbents

The adsorbents were pretreated with different organic compounds (Table 1) for 6 hours, rinsed with distilled water, and then contacted with a clear solution of gold cyanide. The average loading of organic compound on these adsorbents was 180 mg organic compound/g carbon, fiber, or membrane, or 128 mg organic compound/L of wet-settled resin. The inhibition of gold diffusion to the adsorbents was studied by measuring the liquid-phase concentration of gold cyanide. It was also essential to monitor any desorption of organic substances from the adsorbents into the solution. Ultraviolet spectrophotometry confirmed a loss of only 4% of organic compound loading from the adsorbents during the adsorption of gold cyanide. Hence, it is clear that organics were adsorbed fairly irreversibly.

Results presented in Fig. 1 show that both the rate of adsorption and the equilibrium loading were affected for the adsorption of gold cyanide onto activated carbon. The resin particles showed an influence in the rate of adsorption only, whereas, interestingly, both a kinetic and equilibrium influence were observed for membranes and fibers (see Figs. 2 and 3). Furthermore, the model parameters in Table 2 show a decrease in the value of the surface diffusivity $D$ if compared to the untreated adsorbent. Moreover, from Figs. 1, 2, and 3 it can be seen that the degree of inhibition of mass transfer of gold cyanide is larger for the carbon particles than for either the membrane or fiber. This is further manifested in the values of $D$ and $A$ in Table 2. It can be argued that the effective pore size of activated carbon is large in relation to those of the membrane and fiber. This would

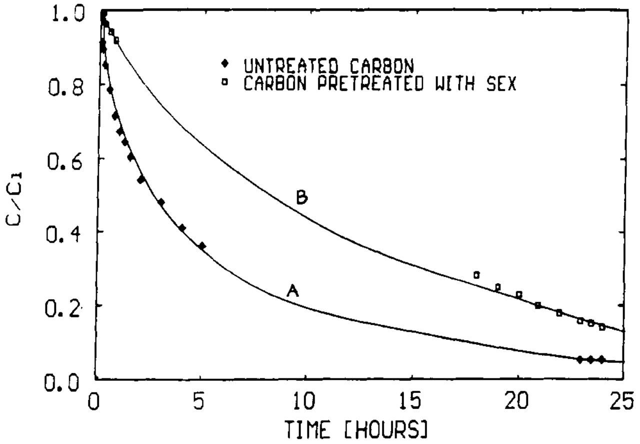

FIG. 1 Inhibited mass transfer of gold cyanide to activated carbon owing to fouling by 180 mg SEX/g carbon in a batch reactor. $C_i$ = 20 mg Au/L; $V$ = 1.0 L; $N$ = 250 rpm; $M_c$ = 1 g.

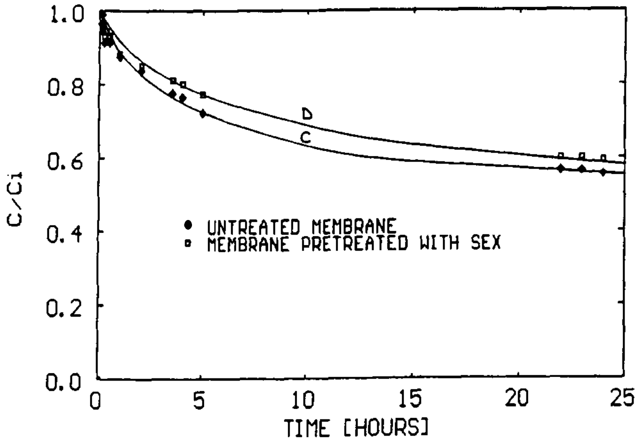

FIG. 2 Inhibited mass transfer of gold cyanide to ion-exchange membrane owing to fouling by 180 mg SEX/g membrane in a batch reactor. $C_i$ = 20 mg Au/L; $V$ = 1.0 L; $N$ = 250 rpm; $M_m$ = 0.2 g.

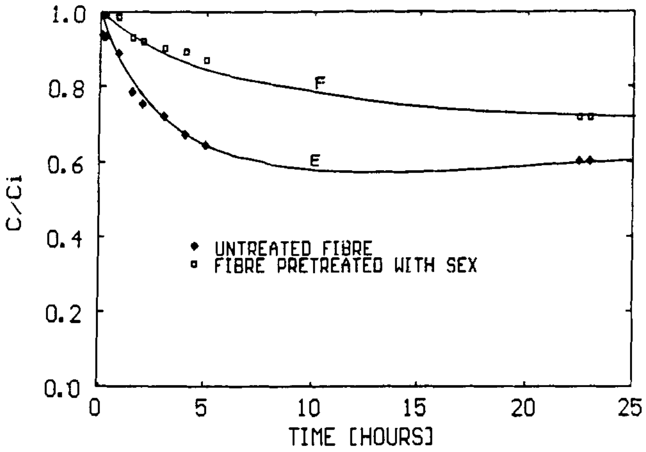

FIG. 3 Inhibited mass transfer of gold cyanide to ion-exchange fiber owing to fouling by 180 mg SEX/g fiber in a batch reactor. $C_i$ = 20 mg Au/L; $V$ = 1.0 L; $N$ = 250 rpm; $M_f$ = 0.5 g.

mean that an organic compound can penetrate a carbon pore much deeper, and hence can seal off the pore completely.

It is worth noting that the film transfer coefficient $k_f$ was insensitive toward the diffusion of gold cyanide into the adsorbents. This observation is to be expected in view of the fact that only the mass transfer inside the adsorbent pore will be affected, and not the effective mass transfer on

TABLE 2
Sensitivity of Mass Transfer Parameters for Gold Adsorption onto Different Adsorbents

| Curve | Adsorbent | Conditions | $k_f \times 10^6$ (m/s) | $D \times 10^{12}$ ($m^2$/s) | $A$ | $n$ |
|---|---|---|---|---|---|---|
| A | Carbon | Untreated | 3.74 | 4.3 | 29 | 0.50 |
| B | Carbon | Pretreated with SEX | 3.72 | 3.7 | 26 | 0.52 |
| C | Membrane | Untreated | 4.7 | 1.9 | 19 | 0.55 |
| D | Membrane | Pretreated with SEX | 4.7 | 1.7 | 16.5 | 0.55 |
| E | Fiber A | Untreated | 3.4 | 3.2 | 32 | 0.23 |
| F | Fiber A | Pretreated with SEX | 3.4 | 2.9 | 28 | 0.23 |
| — | Fiber B | Untreated | 2.4 | 2.8 | 21 | 0.21 |
| — | Fiber B | Pretreated with SEX | 2.4 | 2.6 | 19 | 0.21 |
| — | Fiber C | Untreated | 2.1 | 2.5 | 18 | 0.19 |
| — | Fiber C | Pretreated with SEX | 2.1 | 2.3 | 15.2 | 0.19 |

the outside or in solution which is linked directly to the mixing conditions. Therefore, the results in Figs. 1, 2, and 3 can be ascribed to either *permanent pore-blocking* or *competitive adsorption* between the organic compound and gold cyanide for active sites on the adsorbents' surface. Most researchers have based their modeling approach of gold adsorption in the presence of organic contaminants on competitive adsorption. However, large organic contaminants such as xanthates can diffuse into the adsorbents' pores and seal them off completely, thus restricting gold cyanide from entering the pores. Therefore, such an assumption is not particularly true when large organic compounds such as xanthates are present. The methodology in the next section explains how the present study differentiates between the two mechanisms.

## Competitive Adsorption

In these tests the adsorbents were pretreated with gold cyanide solution until equilibrium was reached, after which the adsorbents were subjected to a solution containing 20 ppm of gold and an organic compound. The equilibrium loadings of gold cyanide were 28.3 mg Au/g carbon, 19.8 mg Au/g membrane, 30.2 mg Au/g fiber, and 17.1 mg Au/L wet-settled resin. The effect of competitive adsorption was studied by measuring the concentration of organic compound in solution. The reason for introducing gold cyanide together with an organic compound was merely to maintain the equilibrium between gold cyanide on the carbon and that in the solution. This was important in view of the possible elution of gold from the carbon by organic species. Figure 4 indicates a clear difference in the adsorption profiles for the uptake of sodium ethyl xanthate (SEX) by activated carbon for the untreated and pretreated carbons. Similar results were obtained when phenol, ethanol and potassium amyl xanthate were used. The other adsorbents showed the same behavior, but the differences in their equilibrium values were less than for activated carbon, as illustrated in Table 3 and Fig. 5 for resin. Furthermore, the difference in equilibrium between the untreated and pretreated adsorbents was relatively sensitive toward the organic compound loading on the adsorbents. At high organic loadings the difference in equilibrium was more profound than at low organic loadings, especially for activated carbon. This observation was true for all the organic compounds and adsorbents, except for phenol which did not show any adsorption by the membrane and fiber. This can be ascribed to the size of the aromatic ring which seems to be larger than the pore size of the membrane and fiber.

It is clear that the effect of gold on the equilibrium of the adsorption of organic compounds cannot be explained in terms of *pore-blocking* (the

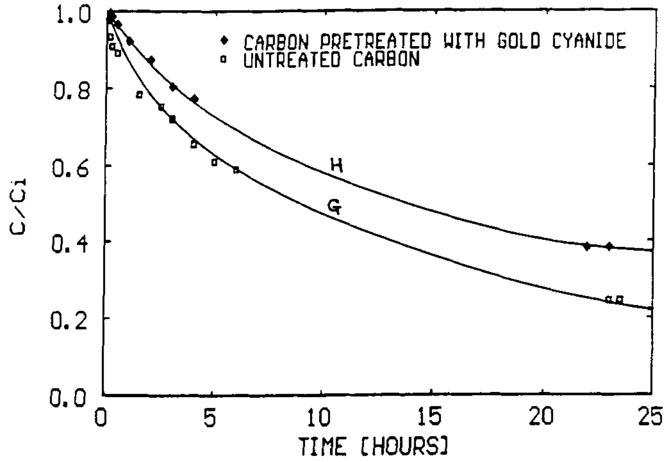

FIG. 4 Adsorption profile of SEX onto activated carbon pretreated with 19.8 mg gold cyanide/g carbon. $C_i$ = 150 mg SEX/L; $V$ = 1.0 L; $N$ = 250 rpm; $M_c$ = 1 g.

gold cyanide molecule is too small). The only possible explanation is that the organic species compete with gold cyanide for active sites on the adsorbents' surface. This would mean that the equilibrium interaction parameters between the organic species and gold cyanide become important. Figure 6 presents the results of the bi-solute isotherm for gold cyanide and SEX in linear form. From the linear relationship, the slope of the line, which represents the competition that SEX offers to the adsorption of gold cyanide, was determined by least-squares regression to be 0.633.

The equilibrium gold loading is then given by

$$Q_{e,Au} = 14.32C_{e,Au}(C_{e,Au} + 0.67C_{e,SEX})^{0.1975}$$

TABLE 3
Competition Factors for Different Organic/Gold Adsorption

| Organic compound | Competition factor ($B_{Org,Au}$) |
|---|---|
| Ethanol | 1.008 |
| Sodium ethyl xanthate | 1.264 |
| Potassium amyl xanthate | 1.236 |
| Phenol | 2.205 |

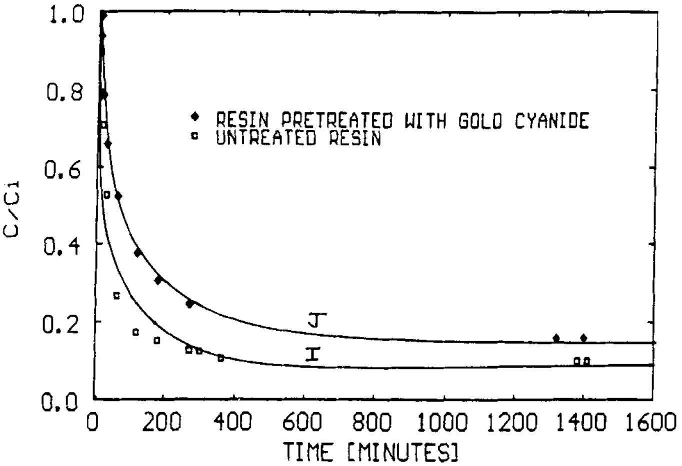

FIG. 5 Adsorption profile of SEX onto resin pretreated with 17.1 mg gold cyanide/L wet-settled resin. $C_i$ = 150 mg SEX/L; $V$ = 1.0 L; $N$ = 250 rpm; $V_R$ = 2 mL.

By using the slope of 1.2 from Fig. 7, the equilibrium SEX loading may be given as

$$Q_{e,SEX} = 67.42C_{e,SEX}(C_{e,SEX} + 1.2C_{e,Au})^{0.3250}$$

Sheindorf et al. (10) stated that the interaction parameter $B_{12}$ should be the reciprocal of $B_{21}$. The fact that this is clearly not the case here shows

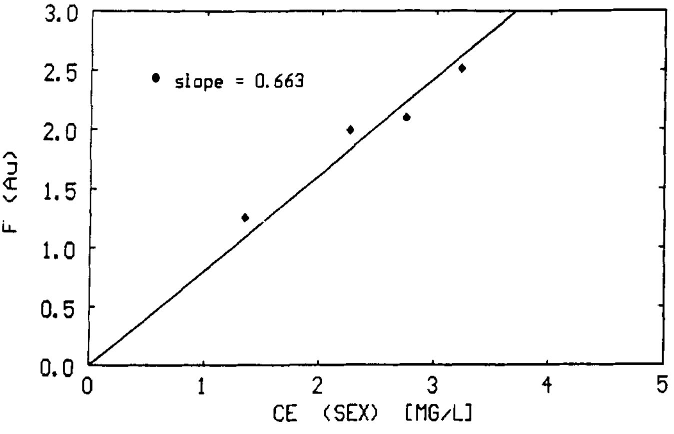

FIG. 6 Estimation of the competition factor $B_{21}$ for the competitive adsorption of gold cyanide and SEX onto activated carbon.

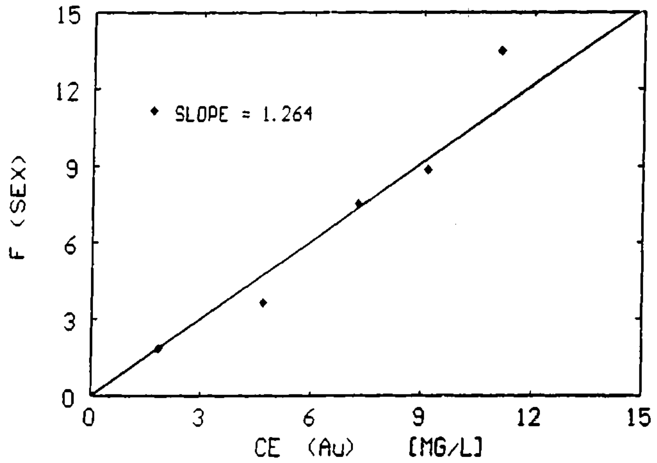

FIG. 7 Estimation of the competition factor $B_{12}$ for the competitive adsorption of gold cyanide and SEX onto activated carbon.

that these equations are merely convenient empirical expressions. It was observed that the competition that ethanol offers to the adsorption of gold cyanide was lower than what SEX, PAX, or phenol offer to gold adsorption. This means that the adsorption of gold cyanide is not affected so much by the presence of ethanol as it is in the presence of the other organic contaminants used in this study. In view of the small differences in the competition parameters for SEX, PAX, and phenol, the adsorption profiles using these organic compounds were fairly similar.

## Adsorption Tendency with Different Organic Compounds

As mentioned earlier, a wide range of organic compounds was used to investigate their effects on gold adsorption. The following categories were included: (a) *aromatic* substance (phenol); (b) *long-chain* organic compounds (sodium ethyl xanthate and potassium amyl xanthate); (c) *low molecular mass* (ethanol) and *high molecular mass* (sodium ethyl xanthate). In the tests conducted here, all the adsorbents were pretreated with organic compounds and exposed to a solution containing gold cyanide. We attempted to quantify the degree of inhibition of mass transfer of gold cyanide to the adsorbents by evaluating the intraparticle diffusion.

It should be mentioned that the organic loadings on the different adsorbents were fairly similar (180 mg/g adsorbent), so that the results in Fig. 8 can be compared directly on the basis of the specific characteristic of

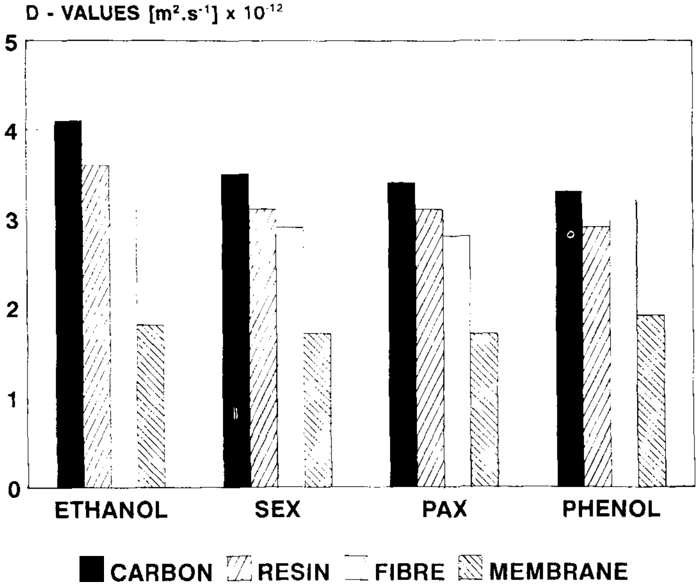

FIG. 8 Comparison of diffusivity for adsorbents pretreated with different organic compounds.

the organic compound. Moreover, these results indicate that the long-chain organic compound has a higher degree of inhibition of mass transfer of gold cyanide compared to the low molecular mass of organic substances (i.e., ethanol). It was difficult to distinguish clearly between the effects of aromatic substances (i.e., phenol) and the long-chain or high molecular mass organic compounds. However, it was interesting to note that the aromatic substances did not influence the adsorption profile of gold for the fiber or membrane. This can be attributed to their small pore diameters which do not allow the "bulky" aromatic to penetrate the porous matrix.

Although it seems that the low molecular mass organic substances have a smaller inhibiting effect on gold adsorption than the long-chain or high molecular mass organic compounds, it is difficult to quantify such a trend completely, based on results obtained in this study.

## CONCLUSIONS

High organic loadings (>180 mg/g organic adsorbent) influenced both the kinetics and equilibrium of gold adsorption for carbon, fiber, and membrane. This was ascribed to the competitive effect between gold cyanide and organic compounds by monitoring the organic compound profile onto gold equilibrated adsorbents. Furthermore, low molecular mass organic substances revealed a smaller inhibiting effect on gold adsorption than the long-chain or high molecular mass organic compounds. A two-component Freundlich-type isotherm fitted the equilibrium for adsorption on carbon, membrane, and fiber.

## NOMENCLATURE

| | |
|---|---|
| $2a$ | membrane thickness (m) |
| $\acute{A}$ | total external area of adsorbent particles in reactor ($m^2$) |
| $A$ | constant in Freundlich expression |
| $B$ | equilibrium interaction parameter |
| $C$ | solution phase concentration (mg Au/L) |
| $D$ | surface diffusion coefficient ($m^2/s$) |
| $d$ | diameter of particle (m) |
| $K_1$, $K_2$ | constants in Langmuir isotherm expression |
| $k_f$ | external film transfer coefficient (m/s) |
| $M$ | mass of adsorbent (kg) |
| $n$ | exponential parameter in Freundlich isotherm |
| $Q$ | metal loading on adsorbent (mg Au/g carbon, fiber or membrane) or resin (mg Au/L wet-settled resin) |
| $r$ | radial variable (m) |
| $R$ | radius of adsorbent particle (m) |
| $t$ | time variable (s) |
| $V$ | volume of liquid in the reactor ($m^3$) |
| $V_R$ | volume of wet-settled resin ($m^3$) |
| $x$ | distance coordinate (m) |

### *Greek Letters*

| | |
|---|---|
| $\beta$ | availability factor |
| $\rho$ | apparent density ($kg \cdot m^{-3}$) |
| $\phi$ | conversion factor for carbon ($kg \cdot m^{-3}$) or resin ($m^3$ of resin/$m^3$ of wet-settled resin) |
| $\epsilon$ | volumetric fraction |

### *Subscripts*

| | |
|---|---|
| c | carbon |
| e | equilibrium |

| | |
|---|---|
| f | fiber |
| $i$ | initial |
| m | membrane |
| r | resin |
| s | surface |

## ACKNOWLEDGMENT

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