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Ion Flotation of Dichromate with a Cationic Surfactant

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An experimental investigation is presented of the flotation of dichromate ion from aqueous solution with a cationic surfactant, ethylheaxadecyldimethylammonium (EHDA) bromide. Dichromate forms a colloidal complex with EHDA ion in a molar ratio of approximately 1 : 2. Excess surfactant causes the formation of a stable foam to which the complex is adsorbed, providing the separation of dichromate from aqueous solution. Batch flotation studies were conducted utilizing approximately 2-liter solutions containing from 10 to 200 mg. of dichromate ion (4.8 to 96.3 mg. of hevavalent chromium), with masses of EHDA-Br ranging from 400 to 800 mg. The effects of mass of surfactant added, dichromate concentration, pH, and possible interfering anions were determined. A comparison is made with pure surfactant solutions, and the mechanism of the process is discussed.

Most efficient operation is achieved with a molar feed ratio of EHDA ion to dichromate ion from 2.1 to 3.0, and removal ratios ranging from 80 to 95% are obtained. Reduction of the residual dichromate below 10% of the feed concentration requires additional surfactant and produces excessive foam, which could be overcome by modifications in operating conditions.

Ion flotation (foam separation) has been utilized by chemists and chemical engineers for the removal and separation of inorganic and organic cations and anions from aqueous solution. The process involves the addition of a surface-active agent to the solution; a complex between the anion or cation and the surfactant is formed, or the mechanism involves the electrostatic attraction between the surfactant ion and the ion to be concentrated. The complex may be floated to the surface of the solution by means of gas bubbles to the interfaces of which it is adsorbed and a froth may be formed. If the complex is insoluble in water and a scum is formed at the solution-froth interface, the process may be termed ion flotation. The insoluble complex may be removed from the surface of the solution as a froth if low gas flow rates are employed; or it may be carried into a tall column of foam if high gas flow rates are employed. The latter mode of operation provides more effective removal of excess quantities of the surface-active agent. The mechanism of the process involves the flotation of colloidal size particulates. An extensive discussion and review of applications have been presented by Sebba (*8*). If the complex is soluble and further enrichment is obtained in the foam phase, or if the process involves electrostatic ion attraction, it may be termed foam separation. A review of foam separation, including an excellent discussion on metal ions, has been presented by Rubin and Gaden (*5*). The most extensive engineering application has been made to the concentration of cations from radioactive waste solutions (*4, 6, 7*).

Hexavalent chromium in the form of chromate and/or dichromate is found in varying quantities in a multiplicity of industrial wastes. Most are wastewaters from metal finishing industries concerned with the cleaning of metals or with the coating of them with chromium. Chromic acid is used in plating plants for several purposes, including plating solutions, anodizing solutions, chromating solutions, and paint pretreatment solutions. Although the con-

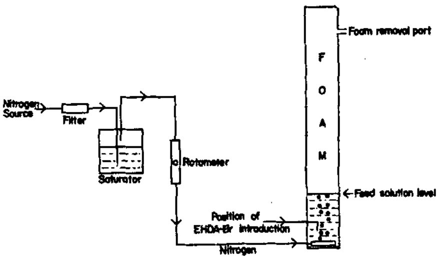

Fig. 1. Schematic diagram of experimental apparatus for batch studies.

centrated solutions are disposed of infrequently, the rinses following the baths are continually discharged to rivers or sewers. Since many states require effluent chromium concentrations not to exceed 2.0 mg./liter, and since the U. S. Public Health Service drinking water standard is 0.05 mg./liter, extensive in-plant treatment is provided by the plating industries. The recovery of chromium into concentrated streams is a benefit resulting from the waste treatment processes. The most commonly used technique is the reduction of the hexavalent form to trivalent chromium with such reducing agents as iron sulfate, sulfur dioxide, and sodium bisulfate at *p*H 3. After reduction is complete, alkali is added to neutralize the acid and to precipitate the trivalent chromium (3). The concentrations of the waste streams treated may vary from 1,000 to 10 mg./liter (as Cr^{6+}). New methods are constantly being sought to improve the economics of the recovery process.

The objective of this study is the determination of the feasibility of batch, multistage, ion flotation for the treatment of aqueous chromate wastes, using a cationic surfactant that would form an insoluble complex with dichromate ion. A brief preliminary investigation has been conducted, but it involved acid solutions only and determined solely the effect of addition of the surfactant (2). In this investigation the effects of dichromate concentration, surfactant concentration, *p*H, and interfering, mono-, di-, and trivalent anions are determined. Comparisons are made with pure surfactant solutions and the mechanism of the process is discussed.

EXPERIMENTAL

A schematic diagram of the experimental apparatus is presented in Figure 1. All of the experiments were conducted in a cylindrical, Lucite column, 10 cm. in diameter and 105 cm. in height. Nitrogen gas was saturated with water, metered with a calibrated rotameter, and passed through triple, sintered glass diffusers of 50 μ porosity. The gas flow was 4,250 ml./min. at 25°C. and 1 atm. For each experiment, from 1 to 20 ml. of a 0.0462 molar solution of potassium dichromate were added to approximately 1.930 liters of distilled water. The solution contained from 10 to 200 mg. of $Cr_2O_7^{2-}$ or from 4.8 to 96.3 mg. of Cr^{6+}. All chromate concentrations in this study are given in terms of the dichromate ion. The *p*H of the solution was adjusted with hydrochloric acid or sodium hydroxide, and for some of the experiments varying concentrations of sodium chloride, sodium sulfate, and sodium orthophosphate ($Na_3PO_4 \cdot 12\,H_2O$) were added.

The solution was placed in the column and the nitrogen flow was begun. Varying quantities of a 10 gm./liter (0.0264 molar) solution of ethylhexadecyldimethylammonium bromide, a cationic surfactant, were added intermittently through a tube 12 cm. above the base of the column and approximately 14 cm. below the level of the feed solution. Additions containing from 50 to 800 mg. were made, the number of adds varied from 1 to 16, and the resulting total masses of EHDA-Br per volume of solution foamed varied from 202 to 396 mg./liter. The total volume of the solution being foamed ranged from 1.980 to 2.020 liters and the concentrations of dichromate ion varied from 5.1 to 99 mg./liter of total feed. After each addition, nitrogen bubbles were passed through the solution until all foam or froth formation ceased; then the next add was made. Thus, during the course of an individual experiment an intermittent or pulsed feed of EHDA-Br was provided, with an additional quantity of surfactant added after the ion flotation (and froth formation) resulting from the previous quantity had terminated. This procedure was continued until the total number of adds had been made. Foam rose in the column and was collected at a port located 74.5 cm. above the initial feed solution level. Temperature was maintained within 24° to 26°C.

At the end of each experiment the residual solution volume was measured, the concentration of dichromate ion in the residual solution was determined colorimetrically with diphenylcarbazide (9), and the concentration of EHDA-Br was determined by a two-phase titration technique (*1*). The analysis for dichromate was accurate to within ± 5% and for EMDA-Br to within ± 1 mg./liter. Random analyses of the collapsed foam were made for material balance verification and to determine if the dichromate were chemically reduced.

Upon addition of the EHDA-Br to the potassium dichromate solution, a distinct cloudiness appeared, indicating the formation of fine colloidal particulates or macromolecules consisting of complexed EHDA and dichromate. Information on the size and nature of the particulates has been reported previously (*2*). It was established that the particulates, which were crystalline in nature and which appeared in the foam phase as minute orange crystals, contained 1.93 mg. mole of the monovalent quaternary ammonium ion (EHDA) for each g. mole of the divalent dichromate ion or 0.965 g. equivalent per g. equivalent. The probable formula of the complex is

$$[((C_{16}H_{33})(C_2H_5)(CH_3)_2N)_2\,Cr_2O_7]_M$$

THE EFFECT OF SURFACTANT CONCENTRATION

The first series of experiments involved solutions containing approximately 50 mg./liter of dichromate and masses of EHDA-Br per volume of solution foamed ranging from 202 to 396 mg./liter. From two to sixteen adds of EHDA-Br were made in dosages ranging from 200 to 50 mg. The *p*H of each feed solution was adjusted to 3.6 with hydrochloric acid. Foam concentrations were calculated from the following material balances:

$$V_i = V_r + V_f \qquad (1)$$

$$z_i V_i = z_r V_r + z_f V_f \qquad (2)$$

$$n_t = x_r V_r + x_f V_f \qquad (3)$$

The dependent variables considered are the residual concentration of surfactant x_r, the residual concentration of dichromate z_r, the volume of collapsed foam produced V_f, and the dichromate enrichment ratio z_f/z_r.

The residual concentration of EHDA-Br was rather insensitive to n_t/V_i, the mass of EHDA-Br per volume of solution foamed, varying from 6.5 to 15.5 mg./liter. This would be expected for fixed operating conditions and column geometry, since each experiment was not terminated until all foaming had ceased, and the residual surfactant concentration should be reduced to approximately the same value each time. The concentration of dichromate in the residual solution was found to decrease exponentially with n_t/V_i, as indicated by the following equation.

$$z_r = 240\,(\exp)^{-0.0184(n_t/V_i)} \quad z_i = 50\text{ mg./liter} \qquad (4)$$

This decrease is caused by the larger volume of foam that provides a larger carrying medium for the complex and by more complete complexing of the dichromate ion in the presence of greater excesses of EHDA ion. The ratio of residual dichromate to feed dichromate z_r/z_i ranged from 0.005 to 0.198.

Results for foam volumes and dichromate enrichment ratios are presented in Figure 2. The volume of collapsed foam produced increases linearly with n_t/V_i, the mass per volume of solution foamed. The amount of foam ap-

Table 1. Effect of 50 mg./liter Dichromate upon EHDA-Br Solutions ($pH = 3.6$)

n_t/V_i, mg./liter	mg. EHDA-Br/ addition	EHDA-Br			EHDA-Br + $K_2Cr_2O_7$		
		V_f, liter	x_r, mg./liter	x_f, mg./liter	v_f, liter	x_r, mg./liter	x_f, mg./liter
202	50	0.915	15.5	274	0.060	15.5	6173
202	100	1.268	14.5	308	0.050	14	7460
202	200	1.425	13.5	421	0.050	15	7420
300	100	1.570	16.0	378	0.560	6.5	1055

pears to be directly proportional to the amount of free (noncomplexed) surfactant present in the solution. The four values of n_t/V_i correspond to net molar feed ratios of EHDA ion to dichromate of 2.28, 2.86, 3.42, and 4.56. And since a ratio of about 2.0 is all that is required for almost complete (85 to 90%) complexing of the dichromate (excess EHDA ion is needed for complete complexing), the increases in n_t/V_i bring about direct increases in the concentration of free surfactant ion. Thus, a fraction of the EHDA-Br is used as a collector to form the complex, while a remaining fraction acts as the frother to provide a stable foam for the removal of the complex. Maximum enrichment ratios for dichromate are obtained with maximum n_t/V_i, but the quantity of foam necessary to obtain such enrichment is excessive and the optimum mass per volume of solution foamed is approximately 200 mg./liter.

Comparison of the data in Figure 2 at constant n_t/V_i shows that the foam volume decreases as the size of the add decreases and the number of adds increases. As the size of the add decreases foam is produced from a more dilute solution, since free surfactant and complex are removed by foaming before the next add is made, and less foam should result. Improvements in the enrichment ratios are obtained by decreasing the size of the add from 200 or 100 mg. to 50 mg. The number of adds was not increased beyond sixteen for $n_t/V_i = 396$ mg./liter (beyond twelve for $n_t/V_i = 300$ mg./liter, etc.), since the effects of the number of adds upon x_r and z_r became insignificant as the largest number of adds was approached. Batch operation was employed throughout this study and the addition of EHDA-Br in multiple dosages provides a batch analog to multicolumn, continuous flow operation.

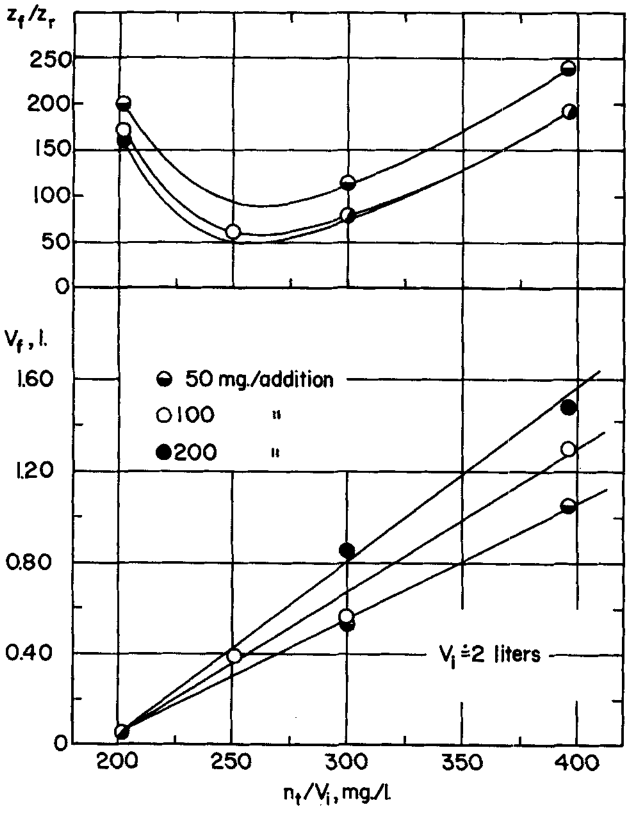

Fig. 2. Relations between V_f and z_f/z_r and n_t/V_i for z_i = 50 mg./liter.

Table 1 provides a comparison between experiments with feed solutions containing 50 mg./liter dichromate and experiments with feed solutions containing no dichromate, for two values of n_t/V_i. Much larger foam volumes are obtained with pure surfactant solutions, reflecting the higher concentrations of noncomplexed, free surfactant. The effect is less extreme at $n_t/V_i = 300$ mg./liter, since the ratio of free surfactant to total surfactant (in the dichromate solutions) is greater than at $n_t/V_i = 202$ mg./liter. The complex appears to have no positive effect on foam stability, and in fact may cause some foam breakage. The presence of dichromate brings about a 3- to 20-fold increase in the concentration of surfactant in the foam.

THE EFFECT OF DICHROMATE CONCENTRATION

The second series of experiments was conducted using solutions containing concentrations of dichromate ranging from 5.1 to 99 mg./liter, and the total masses of EHDA-Br added per volume of solution foamed were 202 mg./liter in two adds of 200 mg. each, 300 mg./liter in three adds of 200 mg., and 396 mg./liter in four adds of 200 mg. The pH was again adjusted to 3.6. In all of these experiments the concentration of EHDA-Br in the residual solution ranged in a random manner from 8 to 15 mg./liter. The residual dichromate concentration z_r was found to increase as a power function of the feed dichromate concentration z_i, as indicated by the following relations obtained from the data:

$$z_r = 5.13\ (10^{-5})\ z_i^{3.17} \quad (\text{for } n_t/V_i = 202 \text{ mg./liter}) \qquad (5)$$

$$z_r = 2.40\ (10^{-6})\ z_i^{3.42} \quad (\text{for } n_t/V_i = 300 \text{ mg./liter}) \qquad (6)$$

$$z_r = 1.12\ (10^{-9})\ z_i^{4.03} \quad (\text{for } n_t/V_i = 396 \text{ mg./liter}) \qquad (7)$$

The increases of z_r with z_i are brought about by two factors: (1) as more dichromate is added, less foam is produced and less complex is carried into the foam stream; (2) the ratio of surfactant to dichromate is decreased and a smaller percentage of the dichromate is complexed.

Figure 3 indicates the variation of the collapsed foam volume and of the dichromate enrichment ratio. The volume of foam decreases approximately linearly (at constant n_t/V_i) as the feed concentration of dichromate is increased, since the amount of free surfactant is decreased. For these studies, removal ratios of dichromate, z_fV_f/z_iV_i, and of EHDA-Br, x_fV_f/n_t (mass of material separated into the foam per mass fed) are presented in Figure 4, related to the net molar ratio of EHDA ion to dichromate ion in the feed (including all surfactant adds) with parameters of n_t/V_i. An increase in the net molar feed ratio at constant n_t/V_i causes a more complete complexing

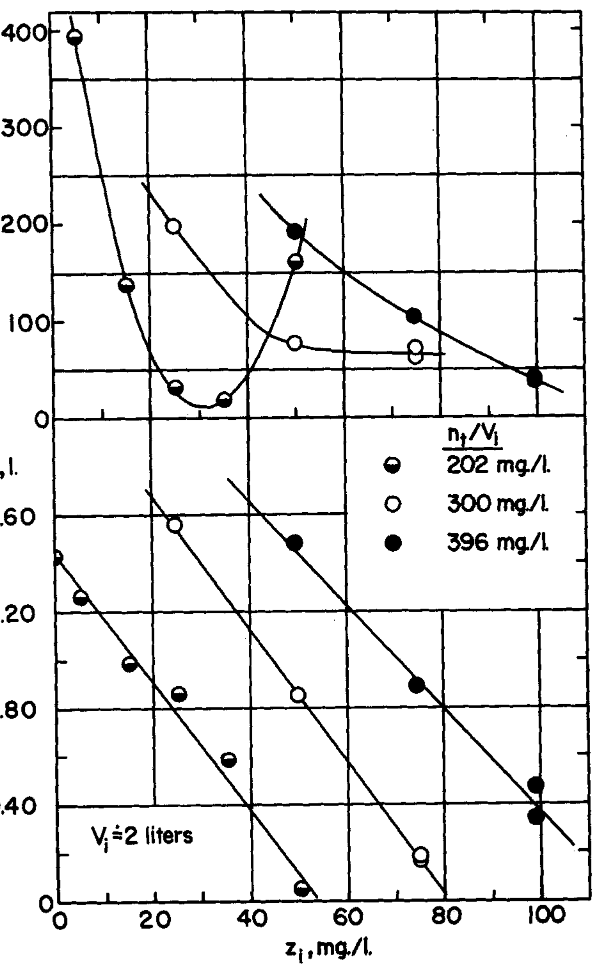

Fig. 3. Relations between V_f and z_f/z_r and z_i for three values of n_t/V_i.

of the dichromate and provides more free surfactant and thus more foam to carry the complex from solution. Virtually complete removal of dichromate is obtained at high values of the net molar feed ratio. At constant n_t'/V_iz_i', increasing the feed concentration of dichromate, and thus of surfactant, provides more free surfactant, more foam, and better removal. The removal ratios for EHDA-Br are greater than those for dichromate at low n_t'/V_iz_i' but are less at high n_t'/V_iz_i'. This is due to more complete complexing of the dichromate, assuming the removals of free surfactant and the complex are the same; or to the fact that the removal of free EHDA-Br, which is included in the EHDA-Br removal ratio but not in the dichromate removal ratio, is a weaker function of free EHDA-Br in the feed solution and thus of foam volume, than is the removal of the complex.

THE EFFECT OF *p*H, CHLORIDE, SULFATE, AND PHOSPHATE

The first two series of experiments indicated the feasibility of the flotation of acid chromate wastes. A final set of data was obtained to determine the influence of *p*H upon the process and to establish any interference that other anions might have upon the removal of dichromate. For these experiments the feed solutions contained 50 mg./liter of dichromate ion; 600 mg. of EHDA-Br were added in three doses. Hydrogen ion had a significant influence upon foam volume but produced rather slight changes in the concentration of dichromate in the residual solution. Decreasing the *p*H from 5.0, that of the distilled water solution of dichromate, to 1.3 caused an approximate doubling of V_f (from 0.850 to 1.560 liter), but z_r remained about constant, varying from 0.9 to 2.5 mg./liter in a random manner. Elevation of the *p*H to 6.6 by adding a small amount of sodium hydroxide caused a doubling in V_f, which then remained constant with further additions up to *p*H 12.0. No interference with dichromate removal was observed until above *p*H 10, when z_r was elevated from 0.5 to 4.5 mg./liter. For efficient operation over the entire *p*H range, the excessive increases in foam volumes would have to be overcome by modifications in operating conditions. The residual concentration of EHDA-Br was virtually independent of *p*H, ranging from 3 to 11 mg./liter over the *p*H range, 1.3 to 12.0.

Figure 5 presents the influence of chloride, sulfate, and phosphate (and monohydrogenphosphate) upon the residual concentration of dichromate. The experiments with chloride were conducted at two different values of *p*H. The *p*H's of the solutions containing phosphate ranged from 10.8 to 6.3, due to hydrolysis of the salt; the monohydrogenphosphate ion was in predominance. All of the anions interfere with the removal of dichromate, with the effect being related to the charge of the ion. High concentrations of phosphate in a chromate waste would seriously impair the ion flotation process. However, since there were only 0.46 meq./liter of dichromate present in the feed solutions, concentrations of any of the anions of the same order of magnitude would cause virtually no interference. The presence of low concentrations of chloride and sulfate provided increases in foam volume from 0.840 to 1.100 liter; further increases in concentration had very little effect. A low concentration of phosphate (0.26 meq./liter) doubled the foam volume and higher concentrations had no additional effect. Three experiments were conducted with 0.26, 0.79, and 1.58 meq./liter of phosphate and with the *p*H controlled at 3.6 with hydrochloric acid. V_f was increased from 0.850 to 1.170 liter and z_r from 1.5 to 4.0 mg./liter. Thus, dihydrogenphosphate behaved similarly to chloride and sulfate. Monohydrogenphosphate and phosphate appeared to cause the undesirable increases in V_f and z_r.

All of these experiments showed no significant varia-

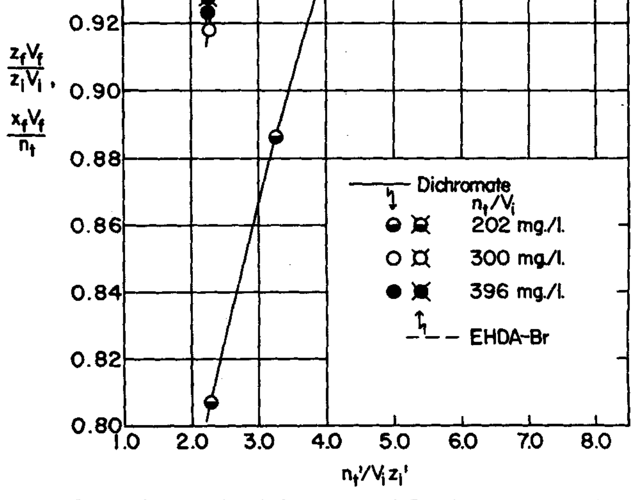

Fig. 4. Removal ratios for dichromate and EHDA-Br vs. molar feed ratios.

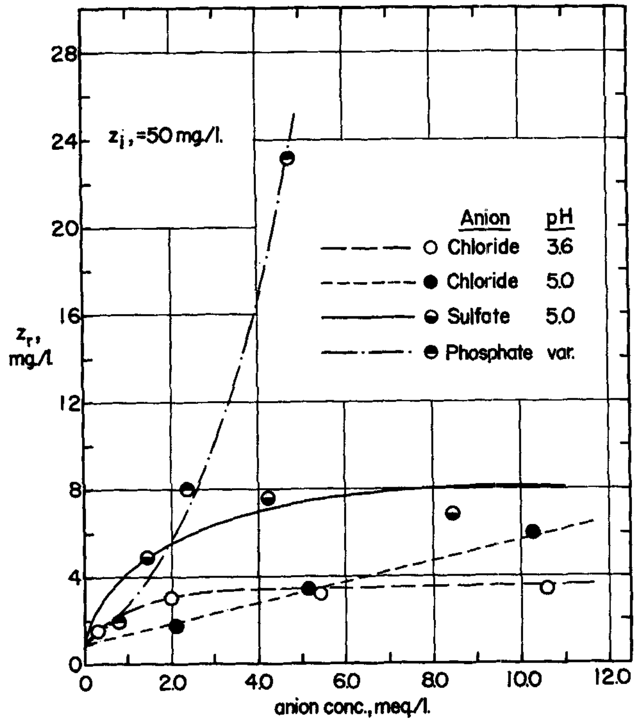

Fig. 5. Effect of anions upon ion flotation of dichromate.

tions in the concentration of surfactant in the residual solution. x_r ranged from 3 to 10 mg./liter. In all cases, the residual concentrations of chloride and sulfate were the same as the feed concentrations. The residual concentrations of phosphate were 10 to 20% lower than the feed concentrations, indicating some flotation.

CONCLUSIONS

For the batch, multistage, ion flotation of dichromate with a cationic surfactant, using fixed operating conditions and foam column geometry, the following conclusions may be made:

1. The contacting of dichromate ion with EHDA ion in aqueous solution brings about the formation of a colloidal complex containing approximately two moles of EHDA per mole of $Cr_2O_7^{2-}$. For complete complexing of the dichromate an excess of surfactant is required; the noncomplexed surfactant produces a stable foam at the gas-liquid interfaces of which the complex is preferentially adsorbed.
2. Based upon the criteria of low volume of collapsed foam and low concentration of dichromate in the residual solution, most efficient operation is achieved with a net molar EHDA/dichromate ratio of from 2.1 to 3.0. Removal ratios ranging from 80 to 95% are obtained under these conditions, depending upon the feed concentration of dichromate. Reduction of the residual dichromate below 10% of the feed concentration requires large quantities of EHDA-Br and produces an excessive quantity of dilute foam. However, significant improvements could certainly be made by changing such operating variables as gas rate, bubble size, and temperature.
3. At a constant feed concentration of dichromate, increases in the mass of free surfactant per volume of solution foamed effect linear increases in foam volume and exponential decreases in the concentration of dichromate in the residual solution. At a constant mass of surfactant per volume of solution foamed, foam volume decreases as a linear function of and the residual dichromate increases as a power function of the feed concentration of dichromate.
4. Variations in *p*H from 3.6 have little effect upon the residual dichromate, but create excessive foam volumes, which again could be limited by changes in operating variables. Concentrations of mono- and divalent anions of the same order as dichromate cause virtually no interference. Concentrations of divalent anions ten times or more greater than dichromate produce excessive foam and interfere with dichromate flotation.
5. Compared to the foam separation of solutions of pure EHDA-Br, dichromate has an extreme effect, decreasing the foam volume and most significantly improving the enrichment. In the flotation studies the residual concentrations of EHDA-Br were reduced to approximately 3% of the mass per volume of solution foamed.
6. The ion flotation of dichromate, for the recovery of chromium and for the treatment of industrial wastewaters, has sufficient promise to merit further study, particularly including continuous flow operation.

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NOTATION

n_t = total mass of surfactant added stepwise to feed, mg.
n_t' = total moles of surfactant added stepwise to feed, mg. mole
$n_t'/V_i z_i'$ = Net molar feed ratio of surfactant to dichromate
V_f = volume of collapsed foam, liter
V_i = volume of feed solution (including EHDA-Br), liter
V_r = volume of residual solution, liter
x_f = concentration of surfactant in the foam, mg./liter
$x_f V_f / n_t$ = surfactant removal ratio
x_r = concentration of surfactant in the residual solution, mg./liter
z_f = concentration of dichromate ion ($Cr_2O_7^{2-}$) in the foam, mg./liter
$z_f V_f / z_i V_i$ = dichromate removal ratio
z_f / z_r = dichromate enrichment ratio
z_i = concentration of dichromate in the total feed, mg./liter
z_i' = molar concentration of dichromate in the total feed, mg. mole/liter
z_r = concentration of dichromate in the residual solution, mg./liter

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