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# Use of Chemical Equilibrium Data to Calculate Iodox Column Operations

J. C. Mailen

OAK RIDGE NATIONAL LABORATORY
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J. C. Mailen

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## NOMENCLATURE

$C_1$ = $I_2$ concentration in inlet gas, mole per liter

$C_2$ = $I_2$ concentration in outlet gas, mole per liter

$C_3$ = $I_2$ concentration in inlet liquid, assumed negligible

$C_4$ = $I_2$ concentration in outlet liquid, mole per liter

$C_M$ = concentration of component M in acid phase, mole per liter (ion charge indications are deleted)

DF = decontamination factor for $I_2$, DF = $C_1/C_2$

$F_G$ = liquid flow rate, liter per min

$k_3$ = pseudo first order reaction rate constant, $dC_I/dt = - k_3C_I$

$K_1$ = equilibrium constant, $K_1 = C_I^2C_{N_2O_4}/C_{I_2}$

$K_3$ = equilibrium constant, $K_3 = C_I/C_{I_2}$

t = time, min

$t_i$ = initial time

$V_{LH}$ = liquid holdup on stage, liters

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## ABSTRACT

In an operation whose chemical mechanism is complicated, such as Iodox, prediction of the operation with changes in operating parameters can be difficult. In this report correlations of the decontamination factors of Iodox for iodine and $CH_3I$ as functions of chemical equilibria will be presented. Different equilibria appear to be involved where nitrogen oxides are absent in the gas stream and where considerable quantities of nitrogen oxides are present. Two separate correlations will be developed to cover these two cases.

## INTRODUCTION

Oak Ridge National Laboratory is engaged in the development of nuclear fuel reprocessing schemes for Liquid Metal Fast Breeder Reactors (LMFBR). One of the major problems of concern with this fuel is retention of radioiodine. One promising technique for the retention of radioiodine (the Iodox Process[1-5]) is to trap the iodine in hyperazeotropic nitric acid at 25°C in which it is ultimately oxidized to the pentavalent form. W. S. Groenier and B. A. Hannaford have previously correlated packed column data[6,7] for trapping iodine species in hyperazeotropic nitric acid using a mass transfer model. They have also examined bubble-cap data[7] for methyl iodide and $I_2$ trapping and concluded that stage efficiencies ranged from 40 to 83%. In this paper we have examined additional bubble-cap column data for trapping $I_2$ and $CH_3I$ in hyperazeotropic nitric acid; nearly equilibrium stages were found.

This paper describes models which use gas-liquid distribution, chemical equilibrium, and kinetic data to correlate and predict the performance of bubble-cap contactors using the Iodox process. The effect of $NO_2$ in the gas phase on iodine trapping is calculated.

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## IODINE TRAPPING IN THE ABSENCE OF NITROGEN OXIDES

When the equilibrium between iodine species and nitrogen oxides in nitric acid is measured at long equilibration times, two equilibria can be determined. These are:

$$I_2 + 4HNO_3 \underset{\leftarrow}{\overset{K_1}{\rightarrow}} 2I^+ + 2NO_3^- + 2H_2O + N_2O_4 \qquad (1)$$

and

$$I^+ + 3HNO_3 + NO_3^- \underset{\leftarrow}{\overset{K_2}{\rightarrow}} IO_3^- + H^+ + 2N_2O_4 + H_2O. \qquad (2)$$

Values of these equilibrium constants have been determined.[4] However, when the initial few seconds of the reaction of iodine with nitric acid are examined,[5] a definite leveling off in the disappearance of $I_2$ is seen, as shown in Fig. 1, which does not correspond in $I_2$ lost and $N_2O_4$ formed to either of these equilibria. The data from these kinetic plots was obtained as $I_2$ concentration vs time. $N_2O_4$ and $I^+$ concentrations were obtained by assuming all $I_2$ which had disappeared was converted to $I^+$ with the appearance of one $N_2O_4$ per $I_2$ consumed. In many of the runs, nitrite was added before the iodine; in these cases the $N_2O_4$ corresponded approximately to the initially added nitrite. The data from these experiments were fitted to a constant of the form:

$$K_3 = \frac{(C_I)^a (C_{N_2O_4})^b}{C_{I_2}} . \qquad (3)$$

The empirical values of a and b determined from the data were

$a = 1.24$

$b = 0.13.$

Using the nearest integral values,

$$K_3 = C_I/C_{I_2}. \qquad (4)$$

The values of $K_3$ determined as a function of nitric acid concentration are shown in Fig. 2. Also shown are values of $K_3$ determined from bubble-cap column and gas-liquid bubbler tests. The model for these determinations will be developed in the following section.

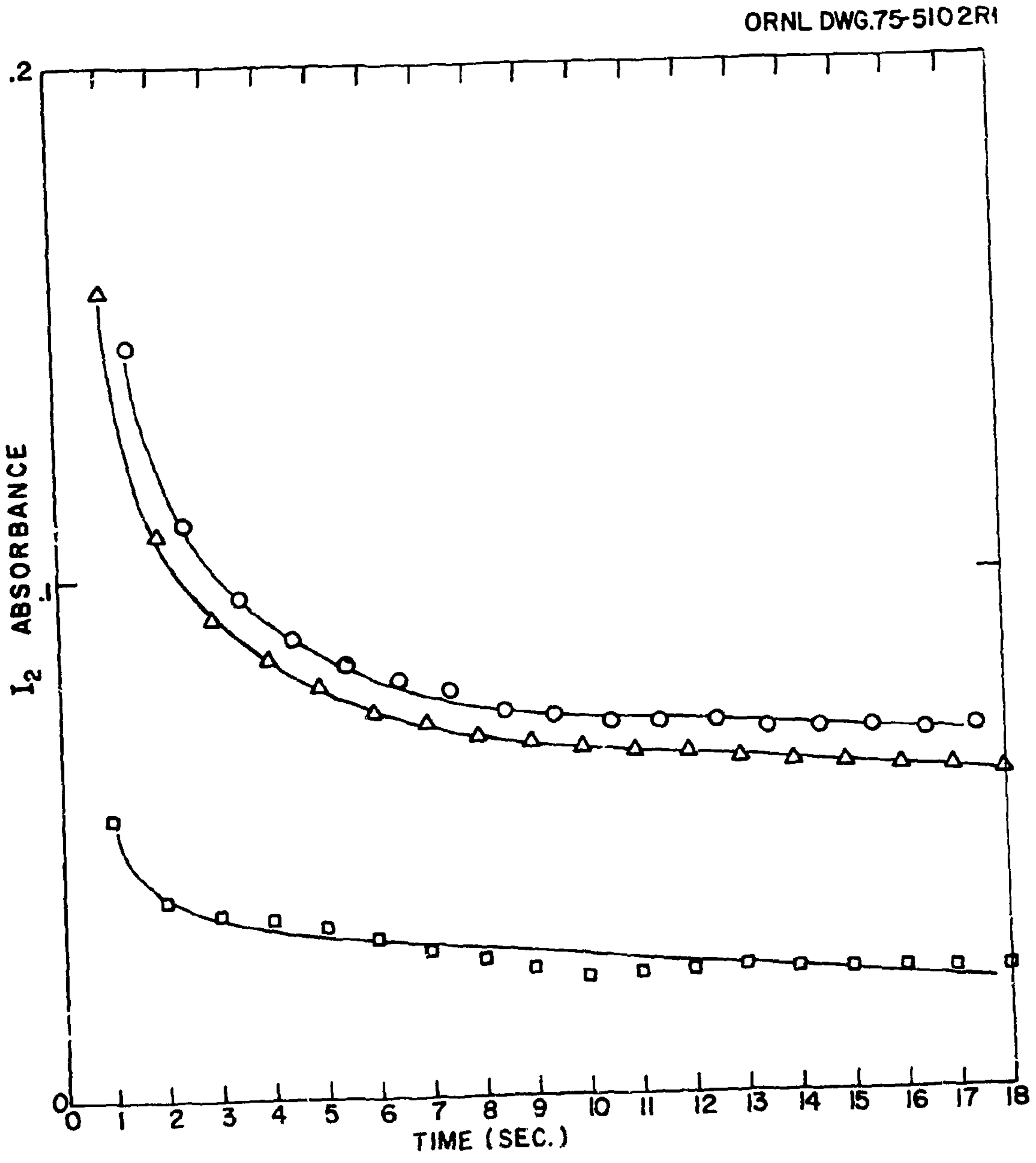

Fig. 1. Reaction of $I_2$ with 19.6 $\underline{M}$ nitric acid; initial $I_2$ concentrations of 0.005 $\underline{M}$ (upper two curves) and 0.0025 $\underline{M}$ (lower curve). The molar absorptivity of $I_2$ in nitric acid (5) is 692 $mole^{-1}cm^{-1}$.

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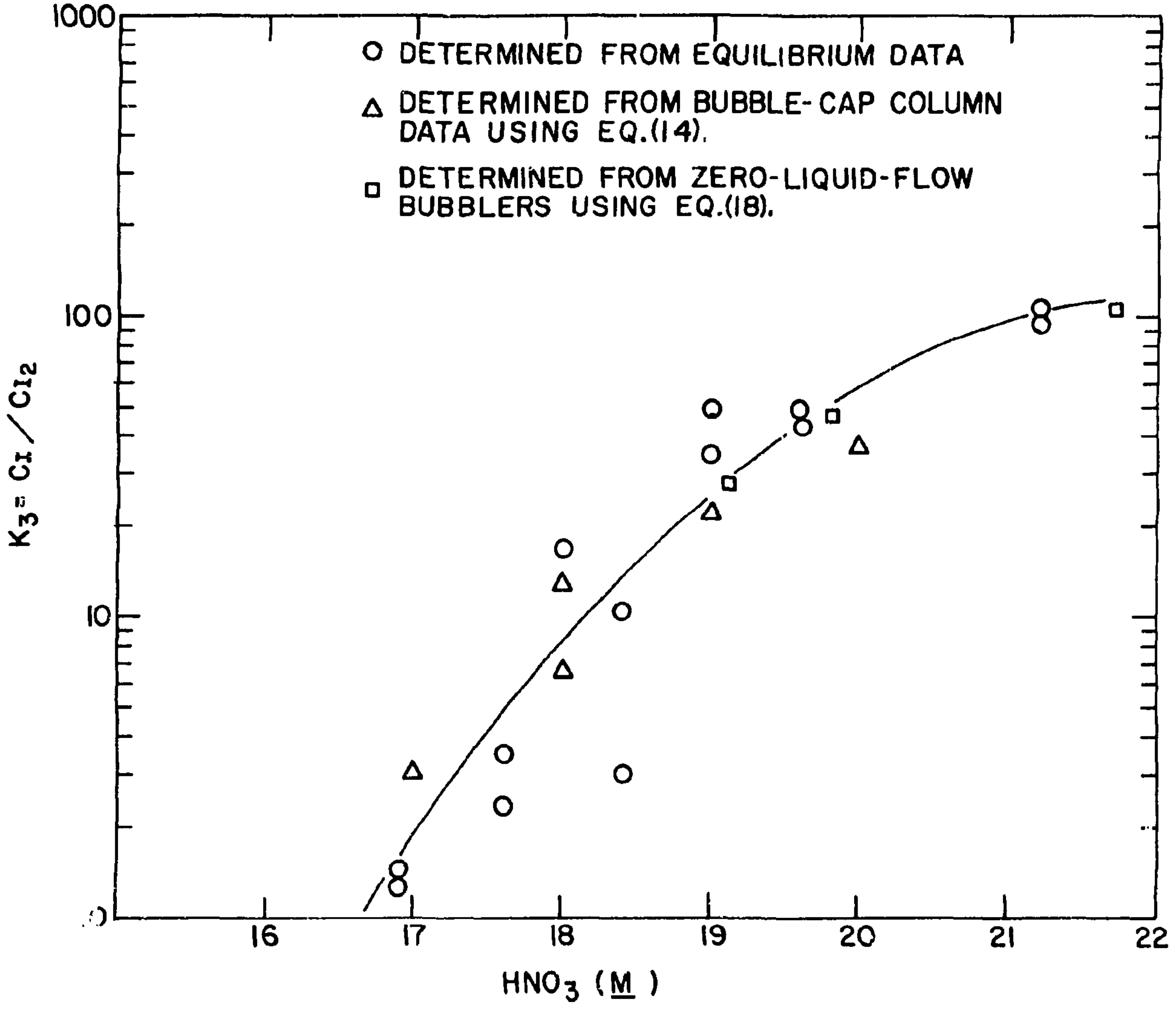

Fig. 2. $K_3$ vs acid concentration.

## MODEL DEVELOPMENT

The assumed model of an equilibrium stage is shown in Fig. 3. All symbols are defined in the nomenclature. The assumptions to be used in the development are:

1. $I_2$ is the only volatile iodine species.
2. Iodine concentration $C_3$ is negligible [relatively high decontamination factor (DF) per stage].
3. Equilibrium $K_3$ is obeyed.
4. Consumption of $I^+$ to yield $IO_3^-$ obeys the pseudo-first-order kinetic equation:

$$\frac{dC_I}{dt} = - k_3 C_I . \tag{5}$$

Values of $k_3$ can be calculated from[5]

$$\ln k_3 = - 45.25 + 13.69 \ln C_{HNO_3} , \tag{6}$$

where $k_3$ is in $sec^{-1}$ and $C_{HNO_3}$ is molarity. Values of $K_3$ are taken from Fig. 2. The distribution of $I_2$ between air and nitric acid has been determined by Crouse and Howerton[8] for up to 15 $\underline{M}$ $HNO_3$. A linear extrapolation of their data indicates that between 19 and 20 $\underline{M}$ $HNO_3$ the distribution ($K_G^L$) is about 77.5. Figure 4 shows a semi-log plot of their data. In Fig. 3 for the equilibrium stage,

$$K_G^L = C_4/C_2 . \tag{7}$$

The moles of $I_2$ transferred to the liquid is

$F_G(C_1 - C_2)$.

The moles of $I_2$ leaving in the liquid is $F_L C_4$. The moles of $I^+$ leaving in the liquid is $F_L C_I$ and the moles of $IO_3^-$ leaving in the liquid is $F_L C_{IO_3}$. A total iodine balance around the liquid phase gives

$$F_G(C_1 - C_2) = F_L C_{I_2} + \frac{F_L C_I}{2} + \frac{F_L C_{IO_3}}{2} \tag{8}$$

at equilibrium. Also, at equilibrium the rate of formation of $IO_3^-$ in solution is equal to its rate of outflow

$$\frac{dC_{IO_3}}{dt} = k_3 C_I V_{LH} . \tag{9}$$

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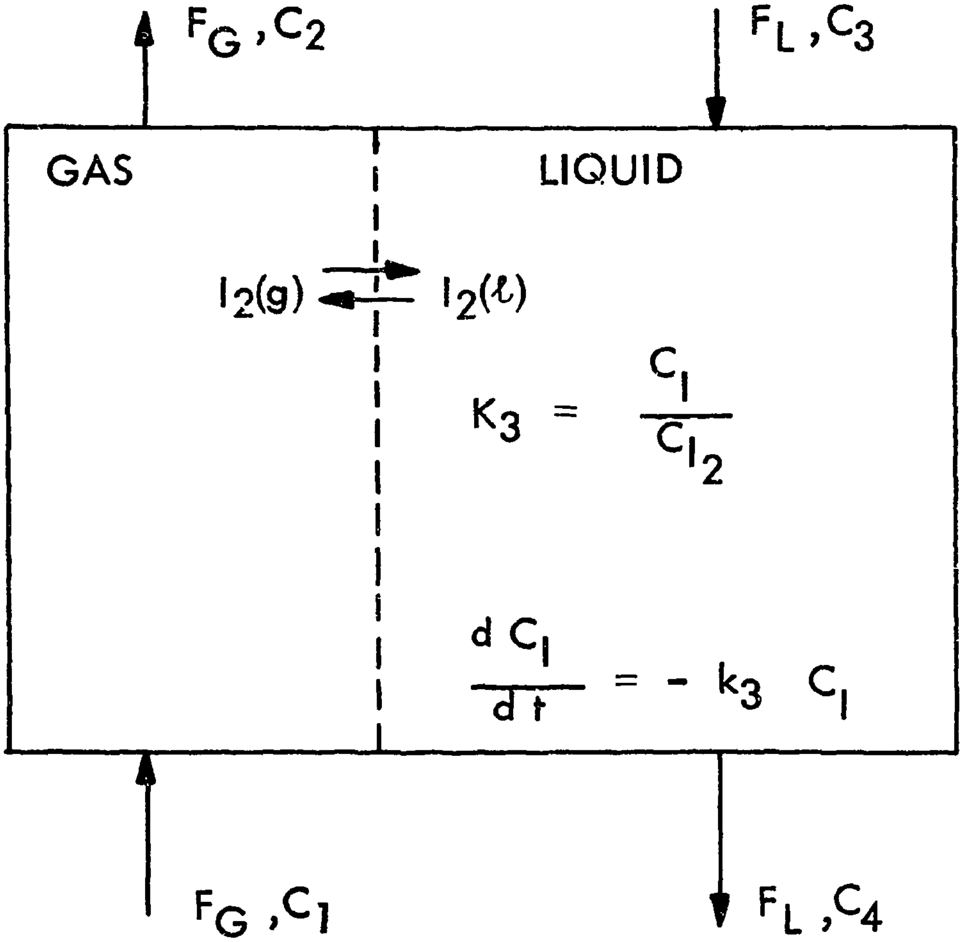

Fig. 3. Equilibrium stage for $I_2$ removal from gas without $NO_2$.

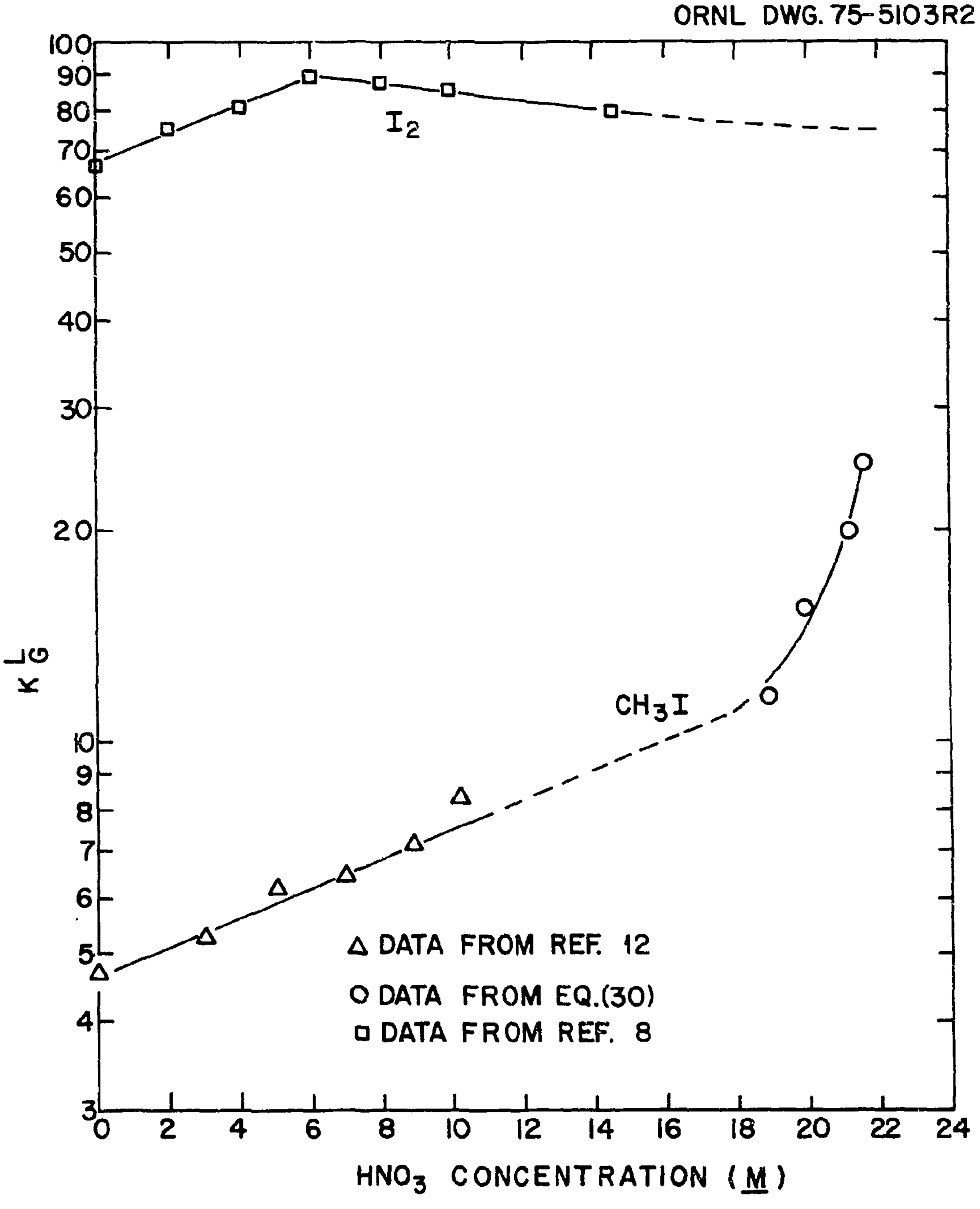

Fig. 4. Liquid-vapor distribution coefficients for $CH_3I$ and $I_2$.

Substituting Eq. (9) into (8) and noting that $C_{I_2} = C_4$ gives

$$F_G(C_1 - C_2) = F_L C_4 + \frac{F_L C_I}{2} + \frac{V_{LH} k_3 C_I}{2} . \tag{10}$$

Solving for $C_I$ gives

$$\frac{2[(C_1 - C_2)F_G/F_L - C_4]}{1 + V_{LH} k_3 F_L} = C_I . \tag{11}$$

Substituting Eq. (11) into (4) gives

$$K_3 = \frac{2[(C_1 - C_2)F_G/F_L - C_4]}{C_4(1 + V_{LH}\, k_3/F_L)} . \tag{12}$$

Substituting Eq. (7) into (12) to eliminate $C_4$ gives

$$K_3 = \frac{2[(C_1 - C_2)F_G/F_L - C_2 K_G^L]}{C_2 K_G^L (1 + V_{LH} k_3/F_L)} . \tag{13}$$

By defining the decontamination factor as $DF = C_1/C_2$, substituting and rearranging gives

$$DF - 1 = \frac{K_3 F_L K_G^L (1 + V_{LH} k_3/F_L)}{2F_G} + \frac{F_L K_G^L}{F_G} . \tag{14}$$

## TESTING OF MODEL

The DFs for $I_2$ have been determined by G. I. Cathers and W. E. Shockley[3] by using a 1-in.-diam bubble-cap column. The values determined at 25°C along with the experimental conditions are given in Table 1. Values of DF calculated by Eq. (14) are given in column 6 and are seen to agree well with those found experimentally. Reversing the procedure, the DFs from the bubble-cap column runs were used to calculate values of $K_3$, which are shown in Fig. 2.

## MODEL FOR SCRUBBER WITH A STATIC LIQUID PHASE

The balance around the liquid phase with no liquid flow is

$$F_G(C_1 - C_2) = V_{LH} \left(\frac{dC_{I_2}}{dt} + \frac{1}{2}\frac{dC_I}{dt} + \frac{1}{2}\frac{dC_{IO_3}}{dt}\right), \tag{15}$$

since

Table 1. Iodine absorption tests using a 1-in. diam bubble-cap column with one bubble cap per stage[a]

| $HNO_3$ conc. (M) | $K_G^L$ | $K_3$ | $k_3$ ($min^{-1}$) | DF | |
|---|---|---|---|---|---|
| | | | | Expt | Eq. (14) |
| 17 | 80 | 1.9 | 0.0936 | 1.49 | 1.37 |
| 18 | 79 | 8.5 | 0.2046 | 2.85 | 2.29 |
| 19 | 78 | 25 | 0.429 | 5.39 | 5.81 |
| 20 | 77 | 57 | 0.864 | 11.70 | 17.6 |
| 18 | 79 | 8.5 | 0.2046 | 4.1 | 4.9 |

[a] Temperature was 25°C; liquid holdup was 0.03 liter/stage; liquid flow rate was 0.01 liter/min; and gas flow rate was 4.8 liter/min except in last case where gas flow rate was 1.6 liter/min.

Table 2. Tests of models using scrubber with static liquid phase. Iodine results.

| | 19.1 M | | 19.8 M | |
|---|---|---|---|---|
| | Expt DF | Calc. DF | Expt DF | Calc. DF |
| Zero $NO_2$ | 12.1 | 11.9 | 31.7 | 32.5 |
| 0.5% $NO_2$ | - | 11.0 | 20.2 | 30.2 |
| 1% $NO_2$ | 4.3 | 3.5 | 11.6 | 9.5 |
| 2% $NO_2$ | 2.7 | 2.1 | 8.4, 7.0 | 3.8 |
| 3% $NO_2$ | 2.2 | 1.6 | 3.8 | 2.2 |

$$\frac{dC_{IO_3}}{dt} = k_3C_I$$

$$F_G(C_1 - C_2) = \frac{V_{LH}}{2}\left(2\frac{dC_{I_2}}{dt} + \frac{dC_I}{dt} + k_3C_I\right). \tag{16}$$

In this system both $dC_{I_2}/dt$ and $dC_I/dt$ are small since continued conversion of $I^+$ to $IO_3^-$ occurs. The differential quantities are appreciable only where low DF's occur, a condition which we will not examine. Leaving out the differentials, we obtain

$$F_G(C_1 - C_2) = \frac{V_{LH}k_3C_I}{2}. \tag{17}$$

Examining Eq. (10) and (17), we see that if the terms involving $F_L$ are set equal to zero in Eq. (10), we obtain Eq. (17). Thus the DF for a scrubber having no liquid flow is found from Eq. (14) by setting $F_L = 0$. This is as follows:

$$DF - 1 = \frac{K_3V_{LH}\ k_3K_G^L}{2F_G}\ . \tag{18}$$

Table 2 gives experimental and calculated [Eq. (18)] values for DF for $I_2$ for a bubbler with no liquid flow (line marked zero $NO_2$). Excellent agreement is found. The values of $K_3$ determined from the experimental DFs are plotted on Fig. 2.

## IODINE TRAPPING FROM GAS STREAMS CONTAINING $NO_2$

When considerable amounts of nitrogen oxides are present in the gas stream, reaction 1 should become important. The equilibrium constant values for reaction 1 indicate that very little $I_2$ will be present in the liquid phase in greater than 19 $\underline{M}$ $HNO_3$. For material balances, the $I_2$ in the liquid phase will be neglected. Other assumptions are:

1. $I_2$ is the only volatile species.
2. Iodine concentration $C_3$ is negligible.
3. The equilibrium in Eq. (1) is obeyed.
4. Conversion of $I^+$ to $IO_3^-$ is negligible.

The diagram of the stage is the same as Fig. 3 except equilibrium $K_1$ replaces $K_3$ and the conversion of $I^+$ to $IO_3^-$ is neglected.

The equilibrium constant for reaction 1 can be written as

$$K_1 = C_I^2 C_{N_2O_4}/C_{I_2}. \tag{19}$$

An iodine balance around the liquid phase gives

$$F_G(C_1 - C_2) - \frac{F_L C_I}{2} = V_{LH}\left(\frac{dC_{I_2}}{dt} + \frac{1}{2}\frac{dC_I}{dt}\right). \tag{20}$$

At equilibrium,

$$\frac{dC_{I_2}}{dt} = \frac{dC_I}{dt} = 0,$$

and

$$F_G(C_1 - C_2) = \frac{F_L C_I}{2}. \tag{21}$$

Substituting Eqs. (19) and (7) to eliminate $C_2$ gives

$$F_G\left(C_1 - \frac{C_I^2 C_{N_2O_4}}{K_1 K_G^L}\right) = \frac{F_L C_I}{2}. \tag{22}$$

Rearranging gives the quadratic form,

$$\frac{F_G C_{N_2O_4}}{K_1 K_G^L} C_I^2 + \frac{F_L C_I}{2} - F_G C_1 = 0. \tag{23}$$

Equation (23) can be solved for $C_I$, and $C_I F_L/2$ is the iodine removed from the gas stream. The DF can then be calculated.

For a vessel in which there is no liquid flow, the material balance at time (t) is

$$F_G(C_1 - C_2) = V_{LH}\left(\frac{dC_{I_2}}{dt} + \frac{1}{2}\frac{dC_I}{dt}\right). \tag{24}$$

For most cases, $dC_{I_2}/dt$ can be neglected leading to

$$F_G(C_1 - C_2) = \frac{V_{LH}}{2}\frac{dC_I}{dt}. \tag{25}$$

Substituting Eqs. (19) and (7) to eliminate $C_2$ gives

$$F_G(C_1 - C_I^2 C_{N_2O_4}/K_1 K_G^L) = \frac{V_{LH}}{2}\frac{dC_I}{dt}. \tag{26}$$

Rearranging Eq. (26) gives

$$\frac{dC_I}{dt} = \frac{2F_G C_1}{V_{LH}} - \frac{2F_G C_{N_2O_4}}{V_{LH} K_1 K_G^L} C_I^2. \tag{27}$$

This equation can be solved for $C_I$ as a function of time in a scrubber with a static liquid phase. It should be noted that conversion of $I^+$ to $IO_3^-$ has been neglected; hence, somewhat low values of iodine trapped in solution are expected.

If such a scrubber is sparged with air containing $NO_2$ and no iodine, the equation for $C_I$ as a function of time can be shown to be

$$\frac{1}{C_I} - \frac{1}{C_{I,i}} = (t - t_i)\left(\frac{2F_G C_{N_2O_4}}{V_{LH} K_1 K_G^L}\right). \tag{28}$$

## EXPERIMENTAL RESULTS USING A SCRUBBER WITH A STATIC LIQUID PHASE

A series of experimental runs were made with a scrubber having a static liquid phase to determine the effect of $NO_2$ on the scrubbing efficiency for $I_2$. The constants required for use in the equations which have been developed are listed in Table 3. The distribution of $NO_2$ from air to nitric acid was determined by Mailen and Howerton.[9] The literature values of the polymerization of $NO_2$ in the gas phase were used.[10]

Values of iodine DF were calculated for the cases in the absence of $NO_2$ by using Eq. (18). Cases with $NO_2$ were calculated by using Eqs. (27) and (28). About 2 min of sparging with gas without $I_2$ was performed in most cases; loss of $I_2$ from the solution was calculated to be small in all tests. Calculated and experimental results are given in Table 2.

From the calculated versus experimental results we see that Eq. (18) for DF in the absence of $NO_2$ gives an excellent prediction; the predicted DFs [by Eqs. (27) and(28)] in the presence of 1% or more $NO_2$ are lower than the experimental values. This may be due to neglecting the formation of $IO_3^-$ in the liquid phase. When below 1% $NO_2$, DFs predicted by integration of Eq. (27) are much higher than the experimental values. This may be due to the difficulty of accurately determining a small difference in two large numbers (the loss from the stage is found as the difference between the iodine fed and that trapped) or it may indicate a basic error in the assumed mechanism. Assuming the mechanism used is reasonably accurate and recognizing that for high DFs it will predict on the high side,

Table 3. Values of parameters for developed equations.

| | 19.1 $\underline{M}$ $HNO_3$ | 19.8 $\underline{M}$ $HNO_3$ |
|---|---|---|
| $F_G$ | 1.35 | 1.35 |
| $V_{LH}$ | 0.03 | 0.03 |
| $C_1$ | $1.79 \times 10^{-6}$ | $1.79 \times 10^{-6}$ |
| $K_1$ | $2.23 \times 10^{-3}$ | $9.5 \times 10^{-3}$ |
| $C_{N_2O_4}$ (0.5% $NO_2$) | $6.27 \times 10^{-3}$ | $9.42 \times 10^{-3}$ |
| $C_{N_2O_4}$ (1% $NO_2$) | 0.0214 | 0.0321 |
| $C_{N_2O_4}$ (2% $NO_2$) | 0.0665 | 0.0998 |
| $C_{N_2O_4}$ (3% $NO_2$) | 0.1221 | 0.1834 |
| $K_G^L$ | 77.5 | 77.5 |
| $k_3$ | 0.461 | 0.755 |
| $K_3$ | 27.5 | 50.0 |

it is of interest to examine the operation of a bubble-cap column. Two cases for a bubble-cap column are given in Table 4. The conditions are the same except case 1 is for 1% $NO_2$ in the gas and case 2 is for 2% $NO_2$. According to these calculations the effect of nitrogen oxides on Iodox will be to require more stages in the column for the same overall DF. However, the reduction of the DF is less as one goes up the column due to the decrease of iodine in the gas phase. To maintain the same equilibrium, the DF increases as total iodine decreases since the $I^+$ concentration in the liquid phase is to the second power while the $I_2$ concentration in the liquid phase (which is in equilibrium with $I_2$ in the gas phase) is to the first power.

It is recommended that tests in an operating bubble-cap column be made to confirm that the presence of $NO_2$ gives the predicted effects since the calculations rely on an assumed mechanism developed to fit bubblers with a static liquid phase and the mechanism could be in error.

Table 4. Calculated DFs of Iodox bubble-cap column. Gas feed to stage 1 contains $10^{-5}$ $\underline{M}$ $I_2$.

| Stage No. | Case 1[a] | Case 2[b] |
|---|---|---|
| 1 | 4.6 | 2.5 |
| 2 | 14.8 | 4.04 |
| 3 | 201[c] | 11.6 |
| 4 | | 105[c] |

[a] Case 1, 1% $NO_2$, 19.8 $\underline{M}$ $HNO_3$, $F_L = 3 \times 10^{-3}$ liter/min, $F_G = 1.35$ liter/min.

[b] Case 2, 2% $NO_2$, 19.8 $\underline{M}$ $HNO_3$.

[c] By using Eq. (14) for operation in the absence of $NO_2$, we know that the maximum DF is 37.

## CORRELATION OF METHYL IODIDE REMOVAL DATA

It has been shown that the first observed reaction of methyl iodide with concentrated nitric acid is as given below:[7]

$$CH_3I + 2HNO_3 \rightarrow CH_3NO_3 + 1/2\ I_2 + 1/2\ N_2O_4 + H_2O. \qquad (29)$$

This reaction is more rapid than the subsequent reaction of $I_2$ with nitric acid.[11] From these observations, it would be presumed that the reaction of methyl iodide with nitric acid would be similar to that of $I_2$ but perhaps somewhat less efficient due to the lower distribution of methyl iodide from air to nitric acid. This has been found to be the case with DFs for methyl iodide for vapor-liquid contactors commonly being about factors of 2 to 3 less than those for $I_2$.

Since the vapor-liquid distributions for $CH_3I$ are not known for concentrated acid, it is assumed that they can be calcualted using the following equation for a gas-liquid bubbler with no liquid flow:

$$DF - 1 = \frac{K_3 V_{LH} k_3 K_G^L}{F_G} . \qquad (30)$$

The results are shown on Fig. 4 along with the literature values of the liquid-vapor equilibria for $I_2$ and $CH_3I$.[12] The values of $K_G^L$ as determined by Eq. (30) and from the literature can be placed on a smooth curve. This may be an indication that other models similar to those developed for $I_2$ will work for $CH_3I$. These are:

bubble-cap column, zero $NO_2$,

where

$$DF - 1 = \frac{K_3F_LK_G^L\,(1 + V_{LH}k_3/F_L)}{F_G} + \frac{F_LK_G^L}{F_G}\,. \tag{31}$$

Bubble-cap column, $NO_2$ in gas,

where

$$\frac{F_GC_{N_2O_4}}{2K_1K_G^L}\,C_I^2 + F_LC_I - F_GC_1 = 0. \tag{32}$$

Zero liquid flow scrubber, $NO_2$ in gas,

where

$$\frac{dC_I}{dt} = \frac{F_GC_1}{V_{LH}} - \frac{F_GC_{N_2O_4}}{2K_1K_G^LV_{LH}}\,C_I^2. \tag{33}$$

In Eqs. (32) and (33), $C_1$ is the concentration of $CH_3I$ in the inlet gas.

Table 5 gives calculated and experimental values of DF for $CH_3I$ using a zero-liquid-flow bubbler when $NO_2$ is present (Eq. 33). The calculated values are about as good as this model gave for $I_2$ trapping.

Figure 5 compares the calculated values of DF for $CH_3I$ in bubble-cap column runs with the experimental values. The agreement is not as good as that obtained for $I_2$, but does give predicted values generally within ± 50% of experimental determinations.

## CONCLUSIONS

Chemical equilibrium data can be used to correlate and predict the DFs for both $I_2$ and methyl iodide for bubble-cap columns and gas-liquid bubblers. Correlations in the presence of $NO_2$ are much poorer than those in the absence of $NO_2$. This may reflect inadequacies in the model for trapping in the presence of $NO_2$. The calculated effect of $NO_2$ in the gas on the performance of Iodox is to decrease the DF in the lower few stages. The upper stages are probably not greatly affected.

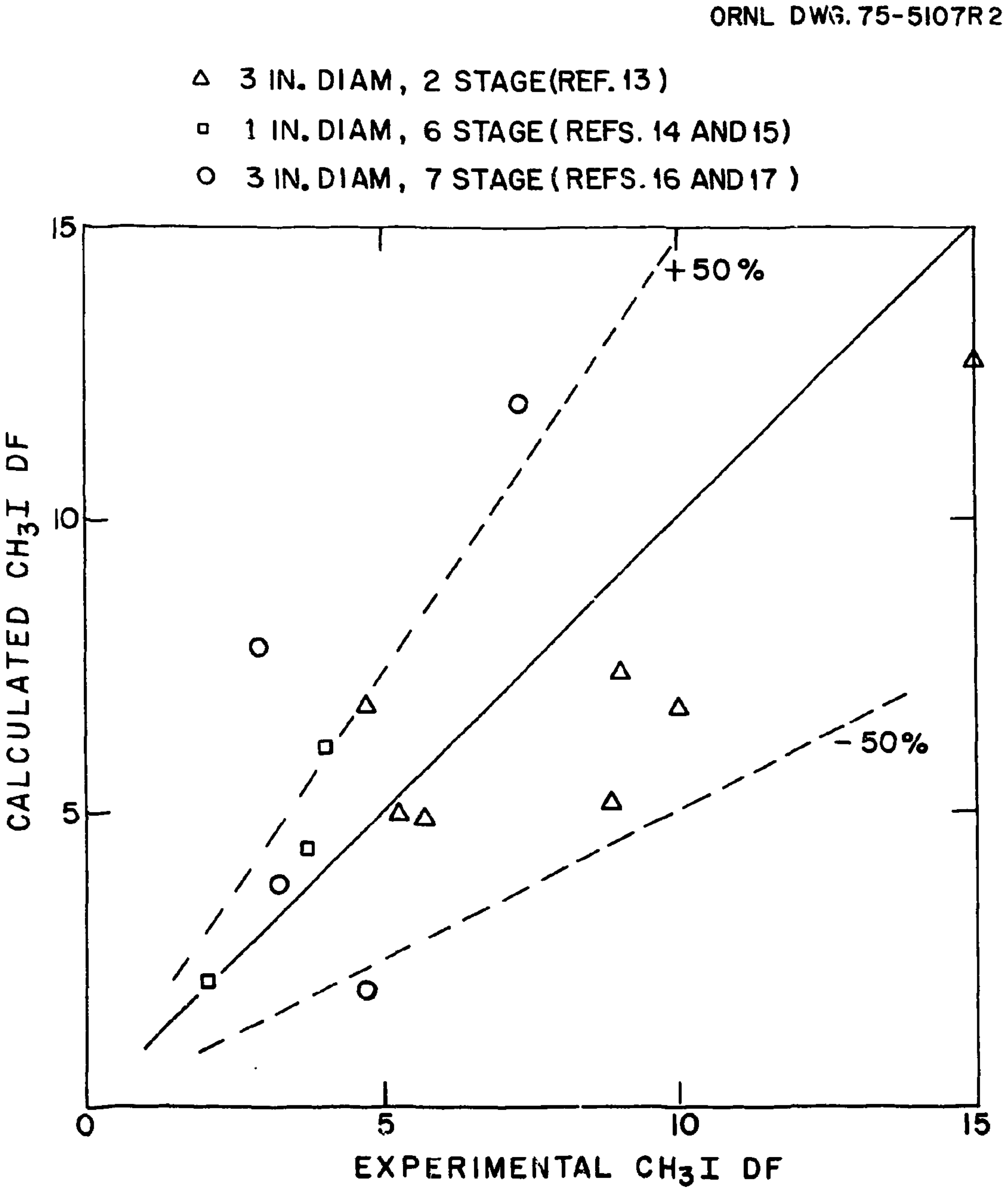

Fig. 5. Experimental Iodox DFs for $CH_3I$ vs DFs calculated by Eq. (31). Data for three bubble-cap columns in the absence of $NO_2$.

Table 5. Test of scrubber model for $CH_3I$. Acid was 19.8 M.

| | Expt. DF | Calc. DF |
|---|---|---|
| 1% $NO_2$ | 4.59 | 6.98 |
| 2% $NO_2$ | 2.71 | 3.01 |
| 3% $NO_2$ | 2.19 | 2.12 |

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