GAS ABSORPTION WITH CHEMICAL REACTION

by

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INTRODUCTION

Gas absorption is a unit operation in which the transfer of a soluble gas from a gas mixture into a liquid takes place. In most industrial applications of this operation, one of the most important considerations is the selection of a proper absorbent. The usual objective in the selection of an absorbent for scrubbing a gas is to find a liquid, or possible a solution, which has a very large capacity for absorbing the solute gas without building up an appreciable equilibrium back pressure. This can be accomplished readily by choosing a chemical with which the solute gas will react irreversibly. The mechanism of such mass transfer is one of physical absorption combined with the chemical reaction in the liquid phase.

Theories governing the absorption rate in the presence of a chemical reaction have been proposed by Hatta (5), Davis and Grandall (3) and Weber and Nilson (15). A theory based on the unsteady-state diffusion concept proposed by Higbie (6) has been developed for slow first-order reactions and for rapid second-order reactions taking place in the liquid-film. Some advances in the theory in this field have been made in recent years. However, the picture is still far from complete. It is not yet possible to make a general correlation between gas absorption with chemical reaction and gas absorption without chemical reaction. Each case must still be studied experimentally.

The purpose of this investigation was to study gas absorption with chemical reaction in a specially designed disc column.

It is a continuation of the previous work by Hwu (7). Two
working equations from which individual film coefficients may
be evaluated were developed by Hwu. The present work was con-
fined to the study of the absorption of ammonia into mono-ammonium
orthophosphate solutions and di-ammonium orthophosphate solutions
in order to further ascertain the gas-film characteristics of
this column. The absorption of chlorine into water was also
studied because of the unusual behavior of this system resulting
from the pronounced ionization of chlorine in water. By con-
tinuing the present work, absorption data for different systems
can be collected. It is hoped that this material will help in
amassing sufficient data to gain an understanding of the mecha-
nism by which transfer between gas and liquid phases occurs when
absorption is accompanied by irreversible chemical reaction in
the liquid phase, and thus to contribute to the development and
extension of the theory of gas absorption.

EXPERIMENTAL

Equipment

The main experimental equipment used in this experiment was
a disc column. This type of equipment was first reported by
Stephen and Morris (13). It possesses the advantages of both
packed and wetted wall columns and is particularly suitable for
the determination of liquid film coefficients for systems in-
volving chemical reactions. The construction and assembly of
the disc column at Kansas State College was reported by Hwu (7).

It consisted mainly of a Pyrex glass column with an inside diameter of 1/8 inch and a height of 34 inches. A series of 35 unglazed ceramic discs with a diameter of 1.5 cm. and a thickness of 0.48 cm. were mounted vertically and centrally in the column. They were threaded edgewise upon a Fiberglas cord and successive discs were maintained at right angles with each other. The principal constants of this disc column are listed in the Appendix.

Two fifty-gallon stainless-steel tanks were used as storage tanks. The phosphate solutions were prepared in batches. A certain amount of the chemicals was added to a definite quantity of distilled water and agitated for more than thirty minutes to make sure of complete solution.

The details of the column and the schematic arrangement of the experimental equipment are shown on Plates I and II respectively. The inlet liquid was pumped from the storage tanks into the liquid feeder at the top of the column. The liquid rate was controlled by a Fisher - Porter C-clamp Flowrater. The liquid flowed downward over the surface of the discs while the gas stream passed upward counter-currently. In the absorption of ammonia in ammonium phosphate solution, air-ammonia mixtures were used, while in the absorption of chlorine in water, both pure chlorine and air-chlorine mixtures were used. Air from the compressed air supply and ammonia (or chlorine) from a cylinder were metered by flowmeters and the combined gas streams were mixed by passing thoroughly before entering the column. Manometers were installed on the gas line to measure the pressure

of the gas and the pressure difference inside and outside the column.

The effluent liquid flowed down through the effluent passage and then went through a sampler. The sampler, as shown on Plate II, consisted of an overflow arrangement with two Pyrex tubes placed concentrically. The liquid sample was siphoned off with a rubber tube which was connected through a glass tee to a buret for measuring the volume of sample. The liquid was kept flowing continuously through the sampling system during operation.

Procedure

In starting, the air was first expelled from the effluent liquid passage by raising and lowering the sampler. Liquid was then introduced into the top of the column, and the gas was turned on slowly. A sudden increase in the gas rate would cause the gas to rush out of the column before the water head was built up in the liquid feeder to balance the gas pressure inside the column. Meanwhile, the sampler was placed at the proper position so that the liquid level was kept up to the neck of the long funnel inside the column. This was done in order to minimize the end effect of absorption.

In the absorption of chlorine in water, the first sample was taken thirty minutes after starting. The samples were then taken successively at a time interval of ten minutes until the analysis of the samples showed the chlorine absorbed was constant. In the ammonia-phosphate system, a total of four samples were taken. The first one was taken one hour after starting and the

EXPLANATION OF PLATE I

Close-up View of Disc Column

A. Pyrex tube

B. Ceramic discs

C. Supporting Fiberglas cord

D. Liquid feed jet

E. Long stem funnel

F. Gas inlet

G. Gas outlet

H. Liquid feed cylinder

I. Effluent to liquid sampler

J. Drain for liquid lost by splash

6
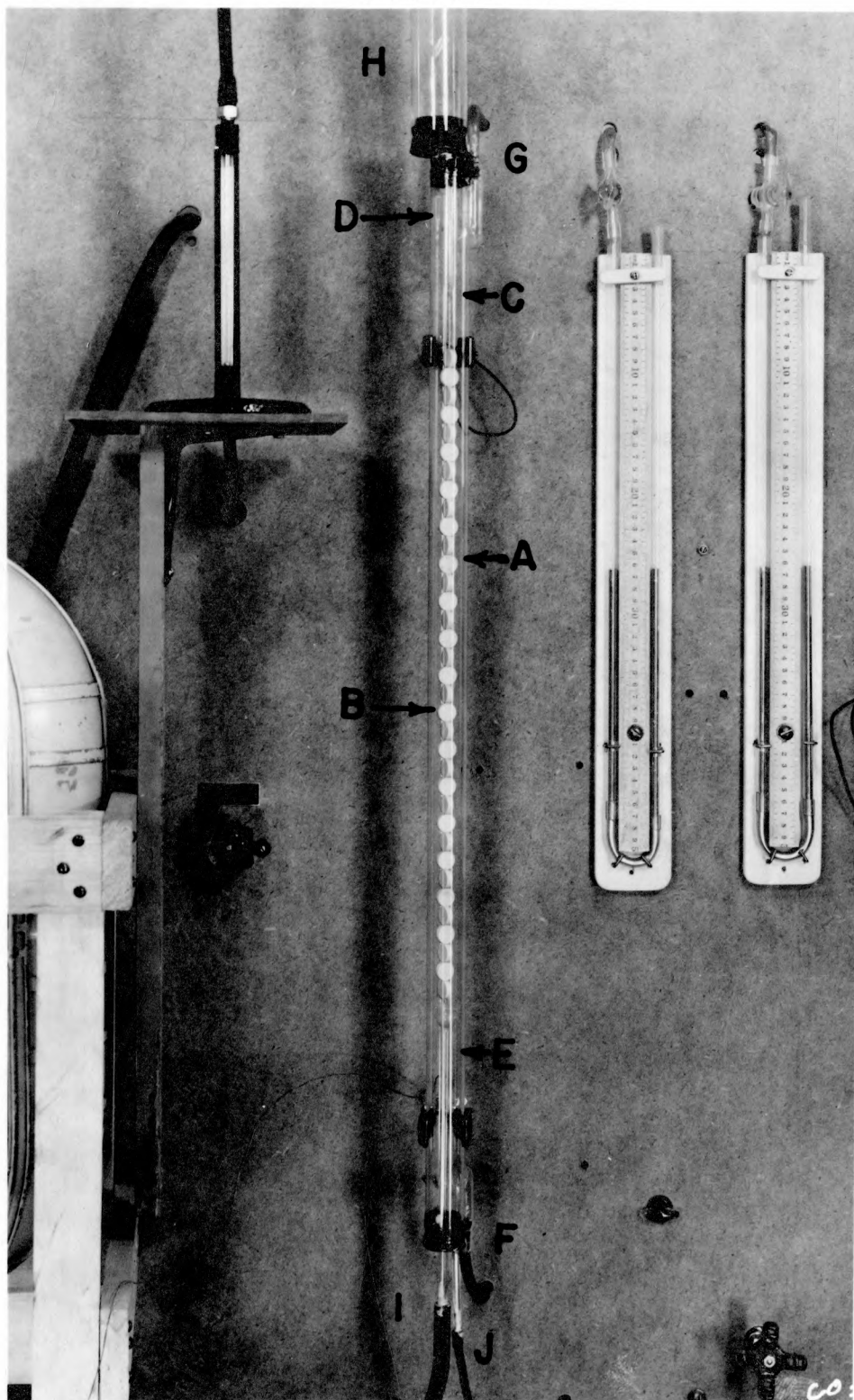
# PLATE I

EXPLANATION OF PLATE II

Schematic Diagram of Experiment Layout

$\quad$ A. $\quad$ Disc column

$\quad$ B. $\quad$ Liquid feed cylinder

$\quad$ C. $\quad$ Drain for liquid lost by splash

$\quad$ D. $\quad$ Liquid sampler

$\quad$ E. $\quad$ Air inlet

$\quad$ F. $\quad$ Solute gas cylinder

$\quad$ G. $\quad$ Air surge tank

$\quad$ H. $\quad$ Air filter

I_1, I_2. $\quad$ Pressure reducers

J_1, J_2. $\quad$ Needle valves

K_1, K_2, K_3. $\quad$ Rotameters

$\quad$ L. $\quad$ Gas mixer

M_1, M_2, M_3, M_4. $\quad$ Manometers

N_1, N_2. $\quad$ Gas sampling points

$\quad$ O. $\quad$ Liquid inlet

$\quad$ P. $\quad$ Centrifugal pump

Q_1, Q_2, Q_3. $\quad$ Thermometers

PLATE II
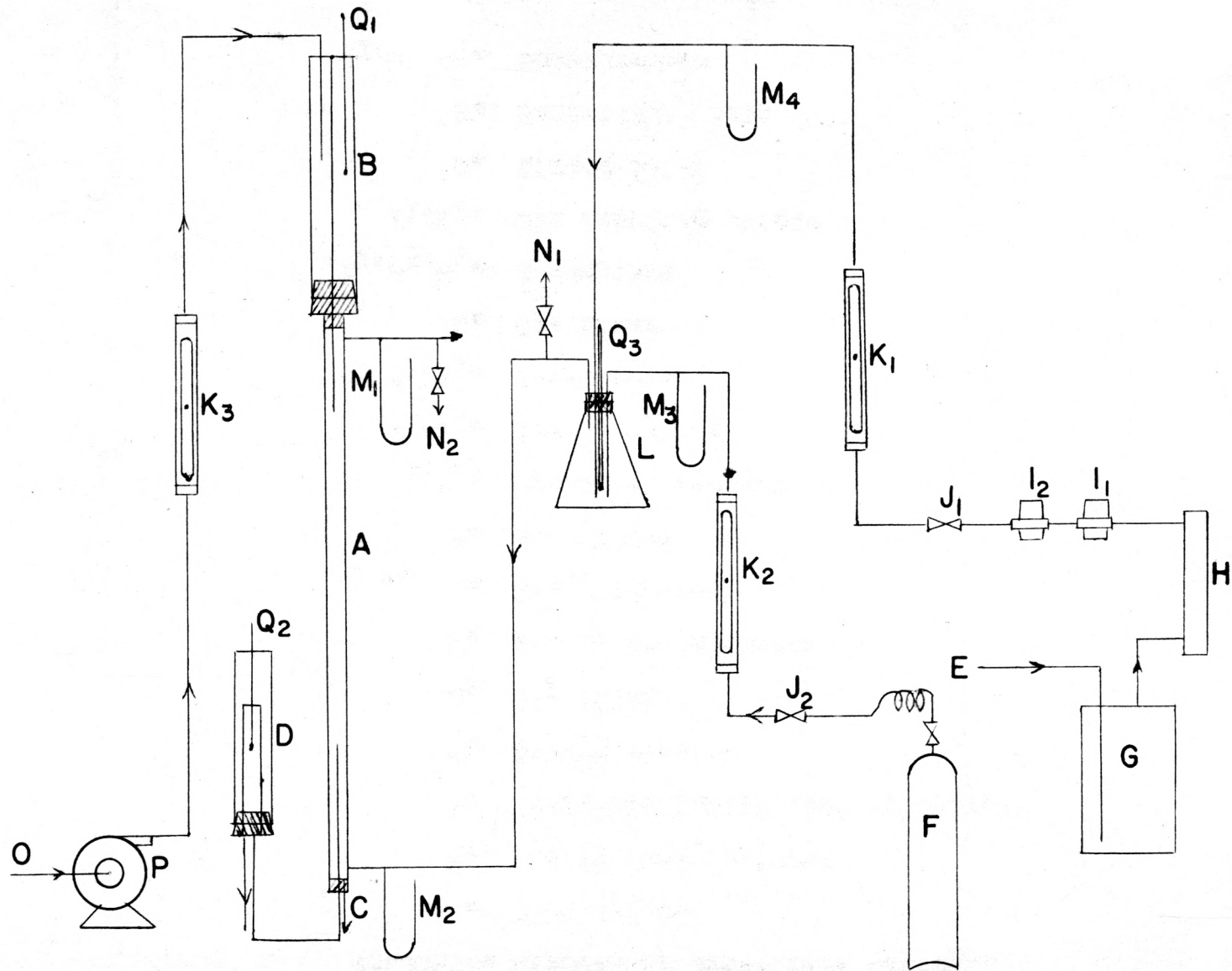
remaining were taken every ten minutes. The longer time for
this case is to make sure the system reached a steady state be-
cause the analysis required a longer time and the results were
not immediately available.

Chemical Analysis

Chlorine in Water. The chlorine absorbed in the water was
determined by the following method Perry (11). The determination
depends primarily upon the reaction:

$$Cl_2 + 2KI \rightleftharpoons 2KCl + I_2$$
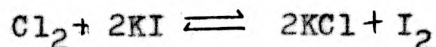
The iodine liberated by the chlorine is titrated with
$Na_2S_2O_3$ solution and the equivalent chlorine calculated.

The following procedures were used throughout the whole
experiment:

(A) Twenty ml of a tenth normal solution of KI were added
to a 250 ml Erlenmeyer flask.

(b) Ten to twenty ml of sample (depending on the experi-
mental conditions) were added to the flask. The delivery tip
of the buret was just above the surface of the iodine solution.
The flask was then closed and the contents were vigorously shaken.

(C) The sample was titrated with a standardized tenth nor-
mal solution of sodium thiosulfate. When the color of the solu-
tion became faint, a few drops of starch solution were added,
and titration was continued until the solution became color-
less.

Ammonia in Phosphate Solution. The amount of ammonia
absorbed was determined in the following way. The inlet liquid
was analyzed so that the content of phosphate and ammonia (ex-
pressed in terms of P_2O_5 and N_2 respectively) were known. The
analysis of the exit liquid gave the ammonia content after ab-
sorption. The difference between these two is the amount of am-
monia absorbed. All of the analyses were done by the Analytical
and Service Laboratory of the Chemistry Department of Kansas
State College.

The analysis of the ammonia content was done by the Kjeldahl
method described by Scott (11). Following is the procedure:

(A) Five ml of sample were placed in a Kjeldahl flask. A
few pieces of granulated zinc and 25 ml of K_2S and $Na_2S_2O_3$
solution were added, and the flask shaked.

(B) A sufficient amount of NaOH solution to make the solu-
tion strongly alkaline was added.

(C) The flask was connected to a condenser by means of a
Kjeldahl connecting bulb, taking care that the tip of the con-
denser extended below the surface of a standard acid solution
in a receiver. The contents were mixed by shaking, and the lib-
erated NH_3 distilled into standard acid. This was then titrated
with standard alkaline solution, using methyl red as the indi-
cator.

The analysis of the phosphate content was based on the
conventional method used for the analysis of fertilizer (9).
The procedures were as follows:

(A) Ten to thirty-five ml of sample were placed in a flask and neutralized with NH_4OH (using litmus paper), then made slightly acid with nitric acid.

(B) Fifteen to twenty ml of a 50 per cent solution of NH_4NO_3 were added.

(C) Thirty-five to fifty ml of ammonium molybdate solution were added, and the sample shaken for 30 minutes.

(D) The precipitate was filtered and washed.

(E) The precipitate was transferred back to the flask. An excess of standard alkaline solution was added and the excess was back titrated with standard HCl solution.

RESULTS AND DISCUSSION

The Absorption of Chlorine in Water

Summary of Experimental Results. A total of 21 runs were made for the absorption of chlorine in water. The effect of water rate and the effect of gas rate were studied. In the study of the effect of liquid rate, pure chlorine was used, while in the other cases, air-chlorine mixtures were used. The liquid rate was measured as the rate in pounds per hour per foot of perimeter of a tube with the same surface area per foot of length as the surface area of the ceramic discs per foot of length in the column. The liquid rate varied from 100 lbs per hour per foot of perimeter to 450 lbs per hour per foot of perimeter; and the temperature was within the range of 25° C. to 30° C. The data obtained are presented in Tables 1 and 2 in the appendix.

The overall coefficient was calculated from the equation:

$$K = \frac{W(C_2 - C_1)}{A \; \Delta C_{lm}} \tag{1}$$

$$\Delta C_{lm} = \frac{(C_{e1} - C_2) - (C_{e2} - C_1)}{\ln \dfrac{C_{e1} - C_2}{C_{e2} - C_1}} \tag{2}$$

Where W is the liquid rate in lbs. per hr. C_1, and C_2 are the concentrations of the inlet and outlet liquid respectively expressed in pounds of chlorine per pound of water. C_{e1} and C_{e2} are the interfacial equilibrium concentrations corresponding to the inlet and outlet gas stream respectively. The solubility data for chlorine in water was taken from Perry's Handbook (10). A is the absorption surface area, its numerical value was 0.22 square feet, based on dry packing. The units of K_L used are pounds of chlorine transferred per hour per square foot of absorption surface per unit concentration (expressed in pounds of chlorine per pound of water).

Because of the low solubility of chlorine in water, the resistance to the transfer of chlorine can be considered to be offered entirely by the liquid film. Therefore the observed overall coefficient, K_L can be regarded as the liquid film transfer coefficient k_L. These observed values were further corrected to a common temperature level of 20° C. by the following equation:

$$k_L \propto e^{nt} \tag{3}$$

Where e is the base of the natural system of logarithms

n is a constant

t is temperature, °C.

With t expressed in °C, Whitney and Vivian reported a value of 0.021 for the constant n, which is the value used in this work. This is fairly close to the value of 0.023 proposed by Sherwood and Holloway (12) for the general case.

Discussion of Experimental Results. The effects of gas rate and liquid rate upon the liquid film transfer coefficient, k_L were studied. The results may be summarized as follows:

Gas Rate: Runs 1 to 5 were carried out under conditions of constant liquid rate and constant gas composition, but with gas velocity varying from 1.96 to 4.21 ft./sec. (The free area for gas glow in the column was equivalent to the area of a tube with a diameter of 0.059 ft. This equivalent diameter was used for calculating gas velocities throughout this work.) The coefficients for these runs are given in Table 2. The results show as was expected, that gas rate has no appreciable effect on the value of the liquid film coefficient. Plate III is a plot of k_L versus various gas rate.

Liquid rate: The variation of liquid film transfer coefficient, k_L with different liquid rates is shown on Plate IV. The values of k_L have been corrected to 20° C. Values of k_L at various liquid rates are shown in Table 2. They are correlated in the following form.

$$k_L = 0.0163 \; \Gamma^{0.81} \tag{4}$$

where Γ is the liquid rate in pounds per hour per foot of perimeter.

Previous work on the absorption of chlorine in water was

done by Adams and Edmonds (1), by Vivian and Whitney (14) and
by Whitney and Vivian (17) (18). Their data indicated that the
liquid film coefficient varied as the 0.8 and 0.6 power of the
liquid rate respectively. The value of 0.81 obtained in this
work is close to that obtained by Adams and Edmonds, but much
higher than that of Whitney and Vivian. However, in view of the
unusually high sensitivity of this disc column to liquid rate as
shown by Hwu (7), this result is more comparable to that of
Vivian and Whitney (14).

Table 1. Effect of gas rate on k_L for the absorption of chlorine
in water. Liquid rate 31.4 lbs/hr. Temperature cor-
rected to 20° C.

Run No.	Gas Rate ft./sec.	Liquid film transfer coefficient k_L, lbs./(hr.) (sq.ft.) $\frac{(lbs.)}{cu.ft.}$
1	2.84	0.58
2	3.82	0.59
3	2.26	0.565
4	1.96	0.706
5	4.21	0.684

The numerical value of k_L is only 70 per cent of the calcu-
lated value based on the equation proposed by Hwu (7), which was
obtained from the absorption date of CO_2 in water. The departure
of the experimental values from the predicted values is shown on
Plate IV.

Vivian and Whitney (14) also found that the experimental
value of k_L is only 1/3 to 2/3 of that predicted from an equation
based on the data for the absorption of oxygen in water. An

Table 2. Effect of liquid rate on k_L for the absorption of
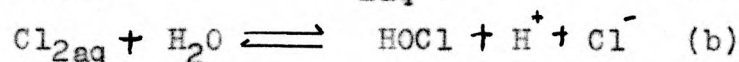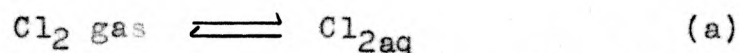chlorine in water. Temperature corrected to 20° C.

Run No.	Liquid Rate lbs./(hr.) (ft.)	Liquid Film Transfer Coefficient lbs./(hr.) (sq.ft.) $\left(\dfrac{\text{lbs.}}{\text{cu.ft.}}\right)$	
		k_L (1)	k_L (2)
1	244	1.2978	1.7889
2	195.3	0.9886	1.3694
3	300	1.0367	1.3598
4	163	0.6997	0.9345
5	285	1.0787	1.3736
6	180.3	0.6413	0.7963
7	133	0.6188	0.7656
8	103.9	0.4744	0.6007
9	346.6	1.4554	1.7934
10	423.6	1.6398	2.0162
11	434.6	1.7578	2.2937
12	344.1	1.5818	2.0867
13	408.7	1.75	2.2603
14	118.9	0.6498	0.8186
15	148	0.7704	0.958
16	103.9	0.5314	0.6878

(1) k_L calculated by using total chlorine concentration
     as driving force
(2) k_L calculated by using molecular chlorine concentration
     as driving force

explanation of the failure of the chlorine-water system to cor-
relate with the other systems was offered by Whitney and Vivian
as follows:

The main difference between the absorption of chlorine by
water and other physical gas absorptions is the hydrolyzing effect
when chlorine dissolves in water. The following chemical re-
actions take place:

$$Cl_2 \text{ gas} \rightleftharpoons Cl_{2aq} \qquad\qquad (a)$$

$$Cl_{2aq} + H_2O \rightleftharpoons HOCl + H^+ + Cl^- \qquad (b)$$

EXPLANATION OF PLATE III

Effect on Gas Rate on k_L for the absorption of
Chlorine in water

Ordinate: Liquid film transfer coefficient, k_L,

lbs./ (hr.) (sq.ft.) $\left(\dfrac{lbs.}{cu.ft.}\right)$

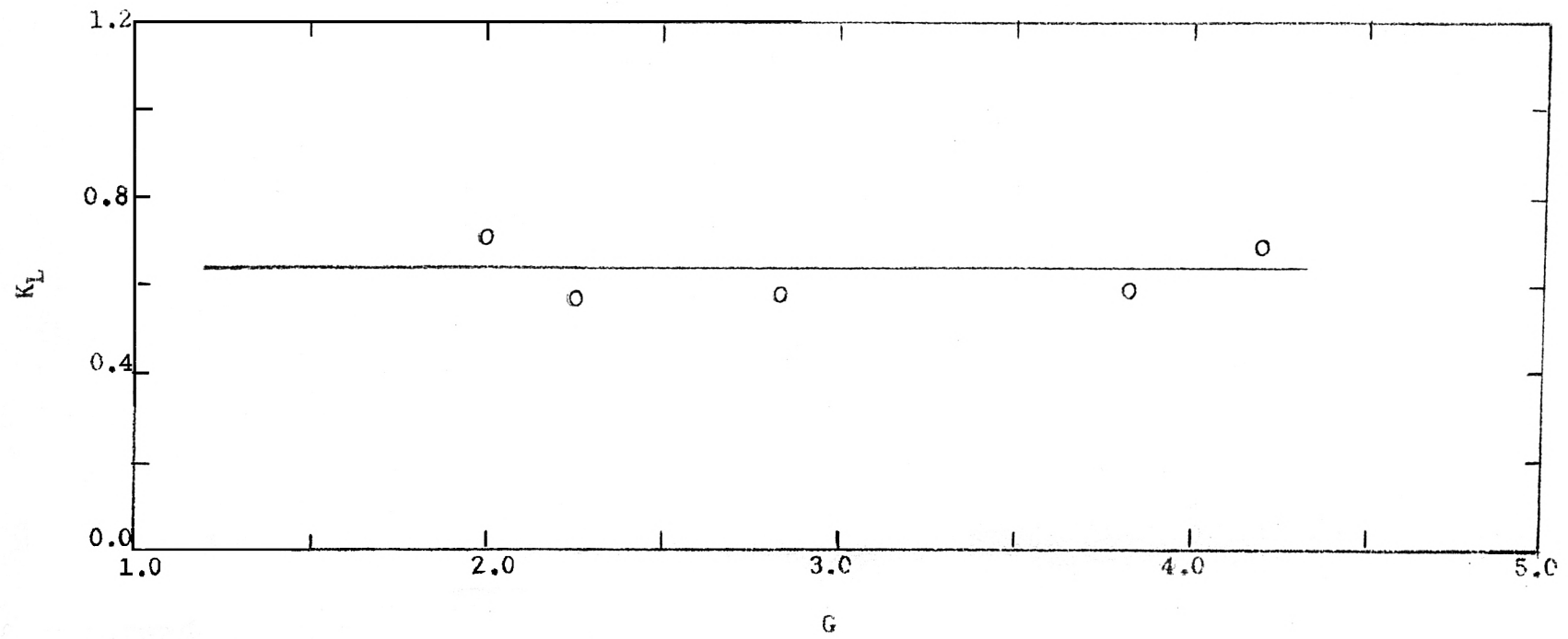Abscissa: Gas rate, G, ft./sec.

Experimental conditions

Liquid rate--------------31.4 lbs./hr.

Chlorine composition----10% Cl_2 in air

Temperature corrected to 20° C.

PLATE III

EXPLANATION OF PLATE IV

Effect of Liquid Rate on k_L for the Absorption of Chlorine in Water

Ordinate: Liquid film transfer coefficient, k_L, lbs./ (hr.) (sq. ft.) $\left(\dfrac{lbs.}{cu.ft.}\right)$

Abscissa: Liquid rate, Γ , lbs./ (hr.) (ft.)

Curve I: Predicted value of k_L from the data for the absorption of CO_2 in H_2O
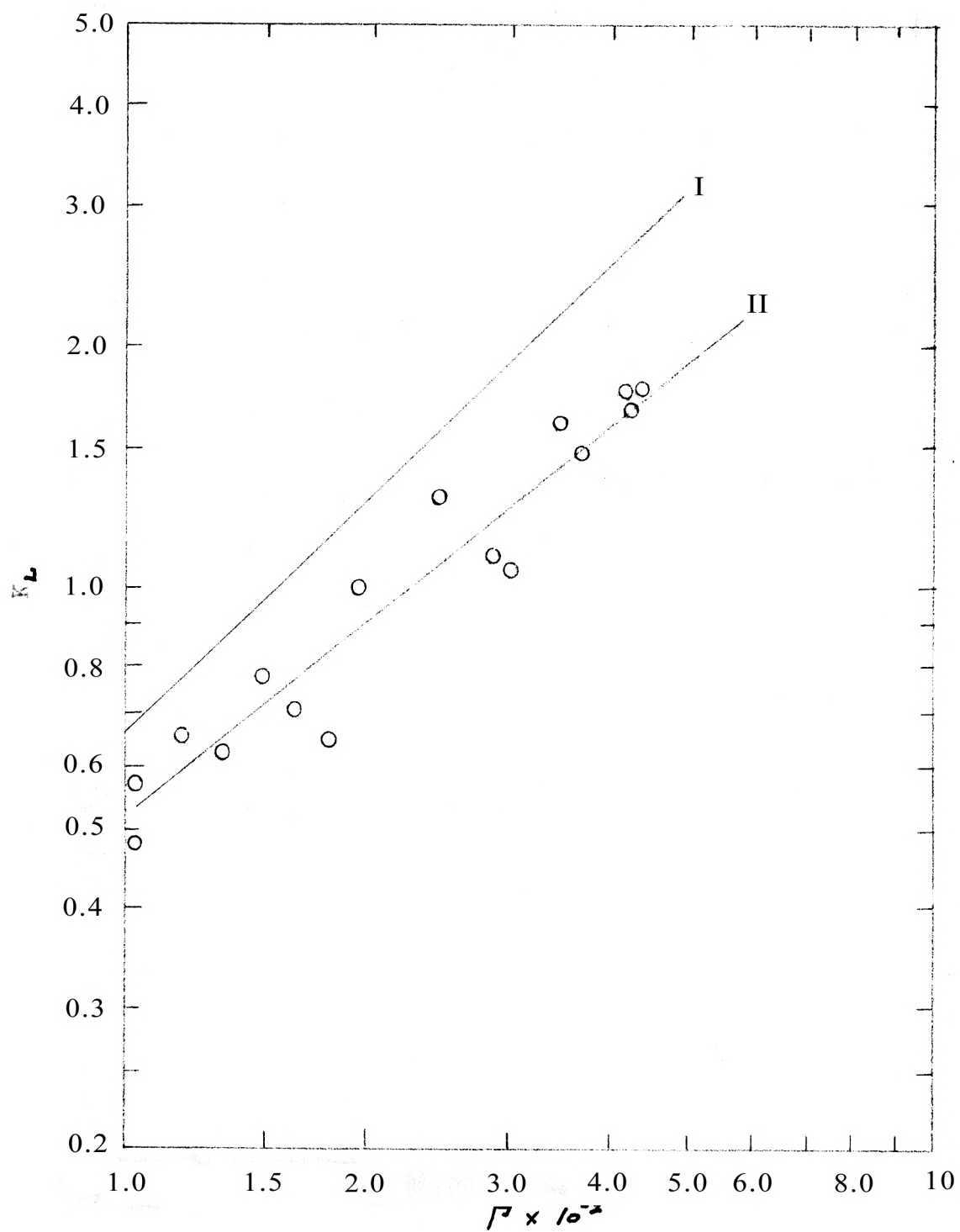
Curve II: Experimental values

Experimental conditions

        Chlorine composition-----pure chlorine

        Temperature corrected to 20° C.

PLATE Ⅳ

When reaction (b) reaches equilibrium, the dissolved chlorine is present in the water in three forms: unreacted molecular chlorine, Cl_2, hypochlorous acid, HOCl, and chloride ion. When physical equilibrium exists between the gas and liquid, and when, in addition, the second chemical reaction is at equilibrium, the following conditions apply according to the law of mass action.

For reaction (a)
$$C_{Cl_2} = \frac{p}{H} \tag{5}$$

where H is the Henry's law constant and p is the partial pressure of chlorine in the gas phase.

For reaction (b)
$$\frac{C_{HOCl}\ C_{Cl}\ C_{H}}{C_{Cl_2}} = K_e \tag{6}$$

If C represents the concentration of total dissolved chlorine, from the above expression, we have:

$$C = \frac{p}{H} + \frac{Ke\ p}{H}^{1/3} \tag{7}$$

The validity of this equation was proved by Whitney and Vivian (16).

It was pointed out by Whitney and Vivian that the rate of the physical reaction (a) at the interface is almost certain to be very large, though the chemical reaction (b) may be relatively slow. Thus, chlorine molecules may dissolve in the liquid and begin to diffuse through the liquid film before reaction (b) has an opportunity to occur. If this is true, the concentration of chlorine in the liquid at the interface will be that of dissolved but unreacted chlorine which is less than C, as shown before. If the reaction is so slow that the chlorine molecules penetrate a considerable distance into the liquid without reaction, the

true driving force available to produce diffusion is ΔC_{cl_2} (molecular chlorine) rather than ΔC (total chlorine). It is apparent that this driving force is smaller when the concentration is that of unreacted chlorine than when it is total chlorine. As a result the transfer coefficient based on the concentration of molecular chlorine is larger. This is called the pseudo transfer coefficient.

The driving force based on the molecular concentration was calculated through equation (2) using the molecular chlorine concentration instead of the total chlorine concentration. The interfacial equilibrium concentration was calculated by using equation (5), and the numerical value of H given in Perry's Handbook (10). The molecular chlorine concentration in the liquor was computed by subtracting the concentration of hydrolyzed chlorine, from the total chlorine concentration, which was determined by direct analysis. The chemical equilibrium constant, Ke, was obtained from Perry's Handbook (10). However, it was found that the hydrolyzed chlorine concentration is too small to be significant and the total chlorine concentration of the liquor was used as that of the molecular concentration. The only difference in the calculation of the two driving forces, then lies in the values of C_{e_1} and C_{e_2} used. For the calculation of the actual coefficients, these concentrations were taken from solubility data for total chlorine concentration at the existing chlorine partial pressure in the gas phases. For the calculation at the pseudo coefficients, these concentrations were calculated from equation (5) using values of H corresponding to molecular

chlorine only.

The effect of liquid rate on the pseudo coefficient is shown on Plate V. The following correlation was obtained for the pseudo coefficients:

$$k_L = 0.01173 \, \Gamma^{0.855} \tag{8}$$

As shown on Plates V and VI, the pseudo transfer coefficients are much closer to the values of the liquid film transfer coefficient calculated from the equation for the absorption of carbon dioxide in water.

From the foregoing discussion, it was concluded that in designing for the absorption of chlorine, if the values of k_L are to be predicted from equations based on other systems, such as CO_2, the molecular chlorine concentration should be used in the driving force term instead of total chlorine concentration.

The points on the plot concentration of chlorine in the outlet liquor versus water rate are scattered, but show a trend of decreasing with an increase of water rate. (Plate VII) When compared with results obtained from the same column previously, there is some inconsistency. So far, three different gases have been used for the study of gas absorption: ammonia, chlorine, and carbon dioxide.

(a) For the $NH_3 - H_2O$ system, the exit liquor concentration decreases as the water rate increases.

(b) For the Cl_2-H_2O system, the exit liquor concentration (total chlorine content) decreases as the water rate increases.

(c) For the $CO_2 - H_2O$ system, the exit liquor concentration increases as the water rate increases as reported by Hwu.

EXPLANATION OF PLATE V

Effect of Liquid Rate on the Pseudo Transfer
Coefficient of the Absorption of Chlorine in Water

Ordinate: Pseudo transfer coefficient, k_L,

                 lbs./ (hr.) (sq.ft.) $\left(\dfrac{lbs.}{cu.ft.}\right)$

Abscissa: Liquid rate, Γ, lbs./ (hr.) (ft.)

Curve I: Predicted value of k_L from the date of

                 the absorption of CO_2 in H_2O

Curve II: Pseudo transfer coefficient from the

                 experimental data.

Experimental conditions

                 Chlorine composition---------pure chlorine

                 Temperature corrected to 20° C.

PLATE Ⅴ
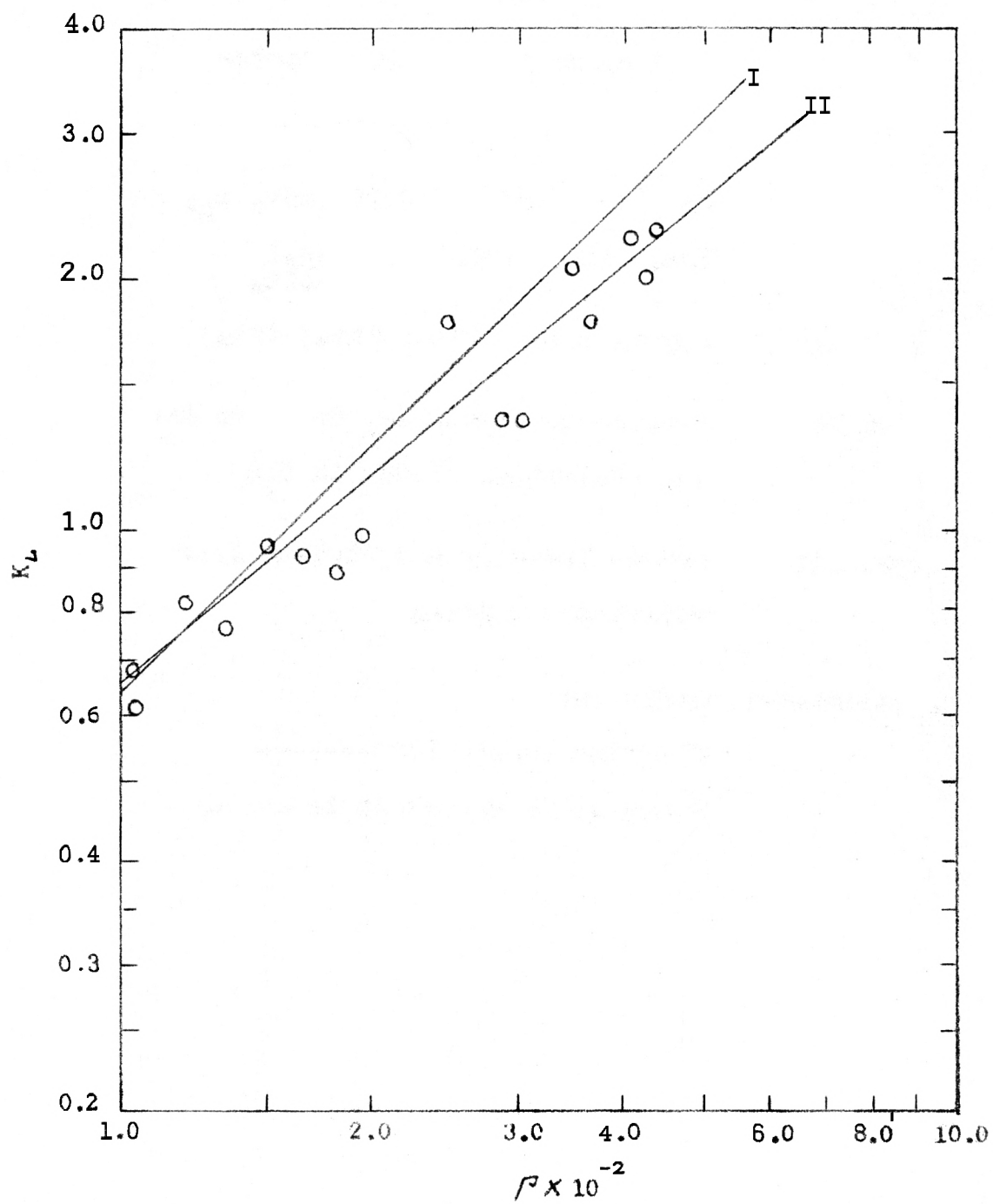
EXPLANATION OF PLATE VI

Comparison of Predicted, Experimental and Pseudo Liquid
Film Transfer Coefficients for the Absorption of Chlorine
in Water

Ordinate: Liquid Film Transfer coefficient, k_L,

                 lbs./ (hr.) (sq.ft.) $\frac{(lbs.)}{cu.ft.}$
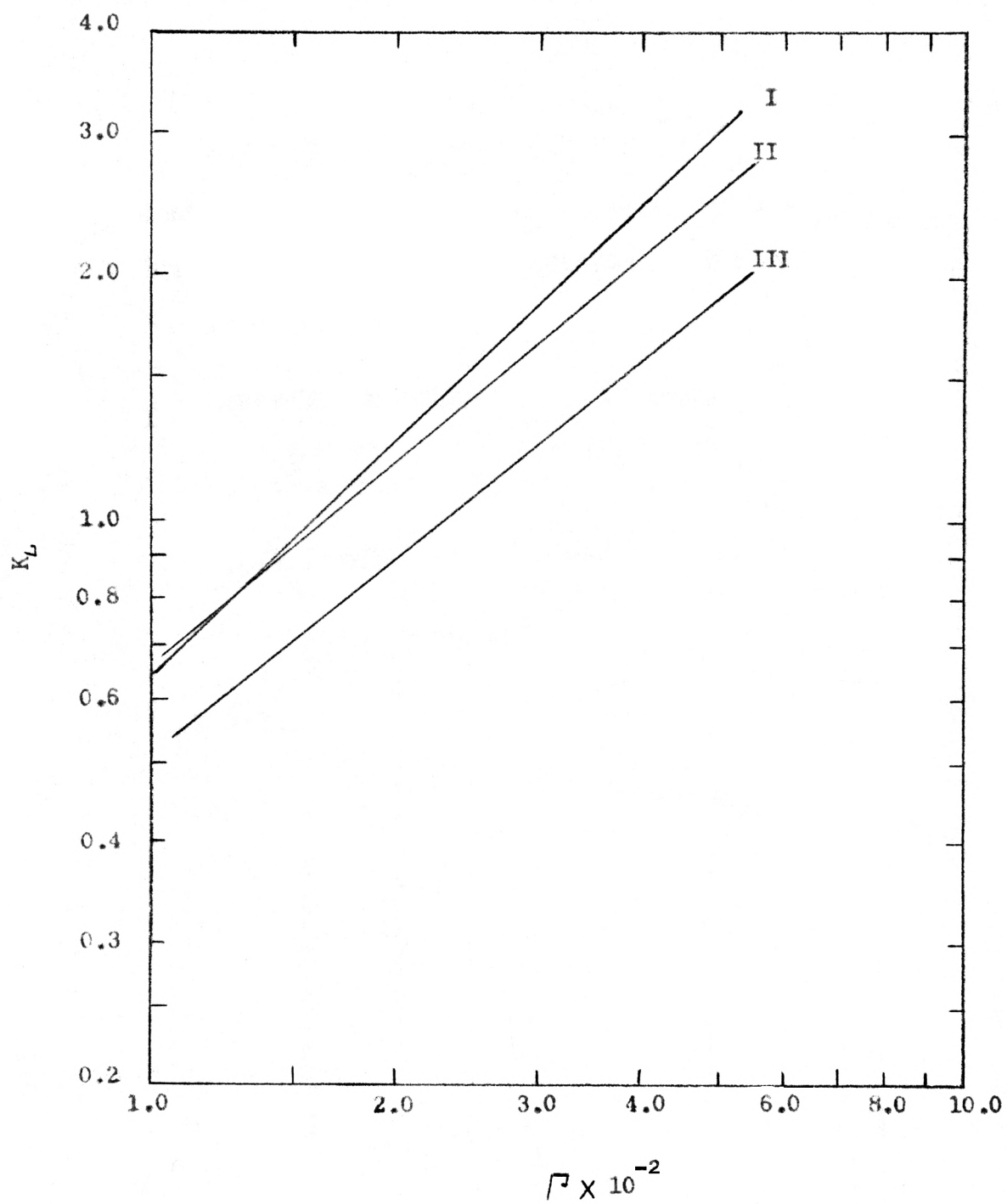
Abscissa: Liquid rate, Γ, lbs./ (hr.)-ft.)

Curve I: Predicted value of k_L

Curve II: Value of pseudo transfer coefficient

Curve III: Experimental value of k_L

PLATE VI

It was pointed out by Hwu that there are two opposing factors influencing the outlet liquor concentration, first the time of contact with the gas stream and second the degree of turbulence which affords the "mixing" effect below the liquid surface and provides fresh surfaces for absorption. In the case of the absorption of carbon dioxide in water, the latter factor plays a dominant role. It was also pointed out that the high smoothness and the poor wettability of the disc itself made some contribution to this situation.

Since both CO_2 and Cl_2 experiments were carried out in the same equipment and under the same operating conditions, and since in both systems the liquid-film controls, similar results were expected for the two systems. The explanations made by Hwu apparently fail in the case of chlorine.

Furthermore, the resistance to molecular diffusion is inversely proportional to the diffusion coefficient. These values are given in Perry's Handbook as follows:

Solute	Solvent	Diffusion Coefficient at 20° C.
CO_2	water	1.5
NH_3	water	1.76
Cl_2	water	1.22

Increasing the time of contact has the effect of increasing the amount of gas dissolved. However, if the contact time is short as compared with that required for reaching equilibrium, then the contact time has a greater influence on those gases with large diffusion coefficients. In other words, for gases with large diffusion coefficients, the exit liquor concentration

should increase to a greater extent with a given increment of contact time than for gases with small diffusion coefficient. Conversely for a given increment in water exit concentration would be conversely reduced to the greatest extent in the case of ammonia, among the three gases above, with carbon dioxide and chlorine following in order.

It is recognized that the degree of turbulence is proportional to the water rate. It was proposed by Danckwerts (2) that the rate of absorption is proportional to the number of fresh surfaces exposed to the gas. The rate of creation of fresh surface is, in turn, a function of turbulence and therefore of the rate of liquid flow from one disc to the next. Therefore, the exit liquor concentration should increase with the water rate.

Since the conditions for the absorption of chlorine in water in this work were the same as those for the absorption of carbon dioxide in water in the work of Hwu, and since both systems have the same characteristics, the effect of turbulence should be the same. However, the time of contact has a greater influence on the carbon dioxide-water system, than on the chlorine water system. If Hwu's explanation holds true, in the case of the absorption of chlorine in water, the outlet liquor concentration should decrease, not increase with the decrease of water rate, and this tendency should be more pronounced than that found in the carbon dioxide-water system.

Unfortunately, insufficient data are available to resolve this inconsistency. However, it may be pointed out that the exit

liquor concentrations in Hwu's work on both ammonia and carbon dioxide absorbed in water are widely scattered, and that while the latter shows a tendency to increase with the liquid rate, the slope can not be accurately determined. Hwu's data on ammonia and carbon dioxide in water, and the data for the chlorine water system are shown in Plates VII and VIII.

The Absorption of Ammonia in Phosphate Solutions

Summary of Experimental Results. Forty-four runs on the absorption of ammonia in phosphate solutions were made. (The data are presented in Tables 3 and 4 in the Appendix). Mono-ammonium orthophosphate and di-ammonium orthophosphate were used in concentrations up to 0.25M. Again the effects of gas rate and liquid rate were studied. The gas and liquid rates were in the same range as those used in the study on the absorption of ammonia in water. The liquid rate varied from 100 to 350 lbs./ (hr.) (ft. of perimeter), and the rate varied from 2 to 6 ft./sec. The ammonia concentration in the air stream was between 5% and 10%.

The overall transfer coefficient were calculated through the following equation:

$$K_G = \frac{NH_3 \text{ Absorbed}}{A \, \Delta P_{lm}} \qquad (9)$$

The amount of ammonia absorbed was determined by the difference of ammonia content in the liquid stream before and after going through the column. "A" is the absorption surface area,

EXPLANATION OF PLATE VII

Plot of Exit Liquor Concentration vs. Liquid Rate

Ordinate: Exit liquor concentration, C, lbs./ 100
             lbs. H_2O

Abscissa: Liquid rate, Γ ,   lbs./ (hr.) (ft.)

Legend:

    ⊖ Cl_2 - H_2O data

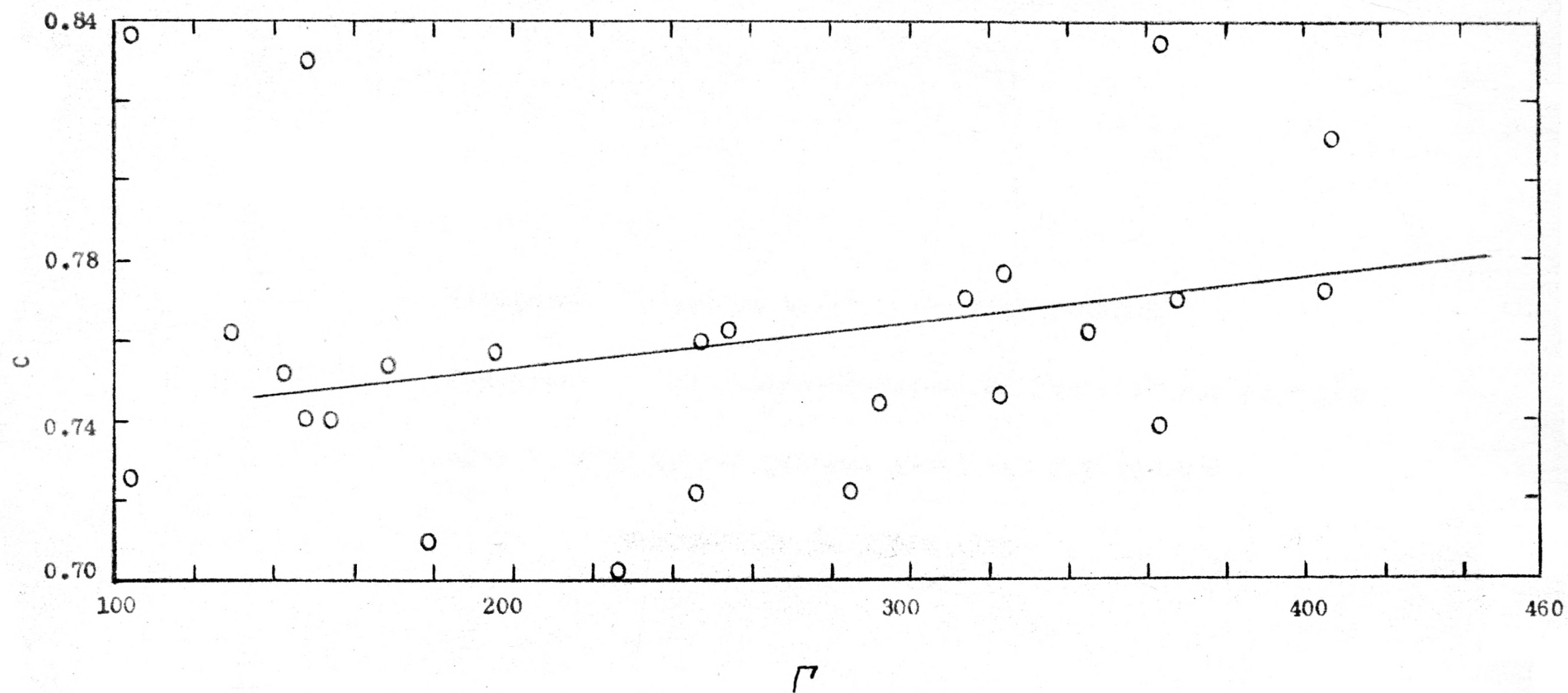   ○ NH_3 - H_2O data

PLATE VII
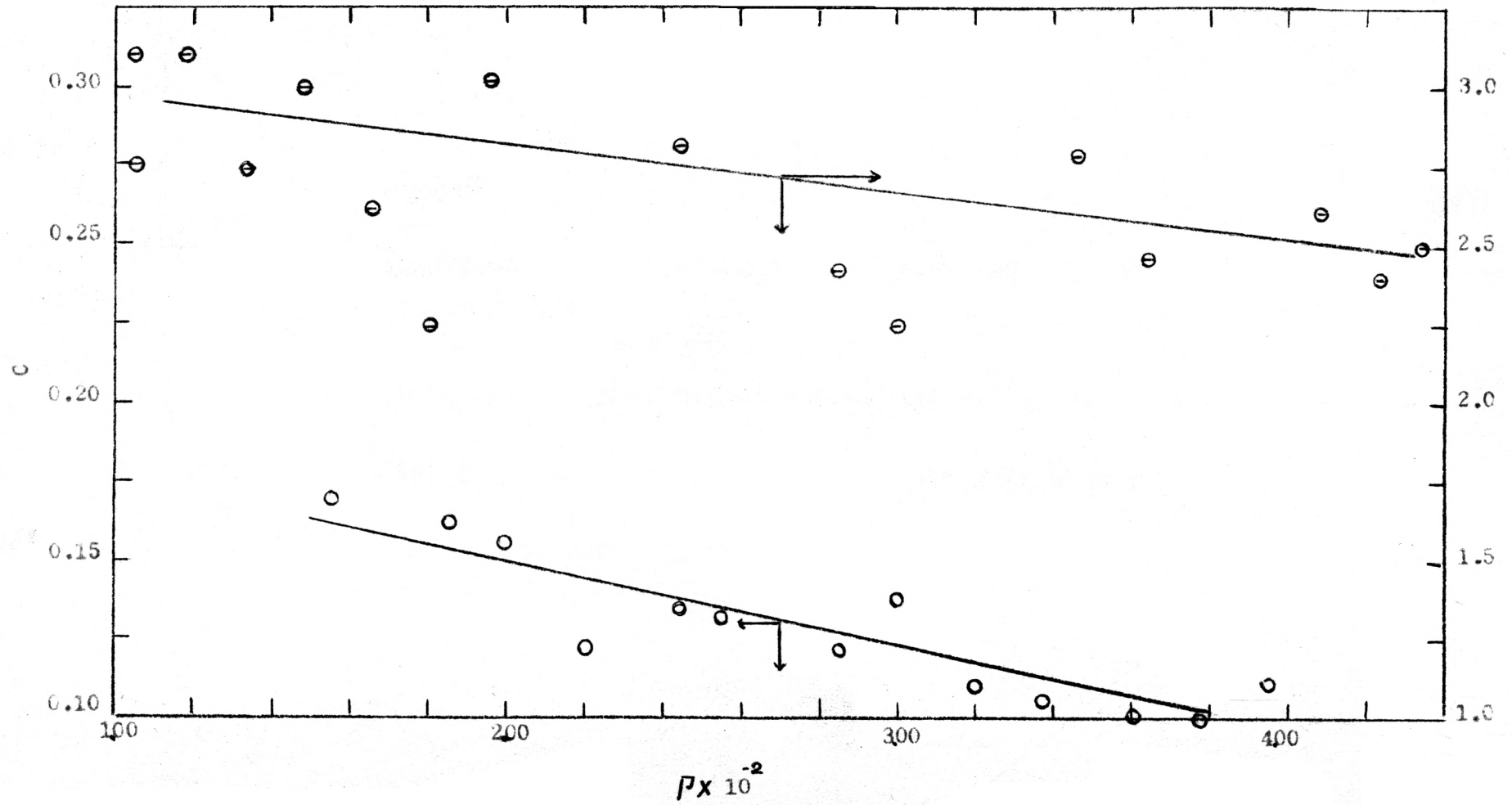
EXPLANATION OF PLATE VIII

Plot of Exit Carbon Dioxide Conc. vs. Liquid Rate

Ordinate: Exit concentration, C, lbs. CO_2/100 lbs. H_2O

Abscissa: Liquid rate, Γ , lbs./hr.-ft.

PLATE VIII

and for this column had a value of 0.22 sq. ft. on the dry basis. P_{lm} is the logrithmic mean driving force.

Since the solubility of ammonia in phosphate solution was not available, the evaluation of the exact mean driving force was impossible. However, it could be found for the two limiting cases: (a) assuming that there was no back pressure of ammonia such as would be the case in a strong acid solution, (b) assuming that the same relationship existed in the ammonia-phosphate system as in the ammonia-water system and that the same Henry's law constant applied.

Values of K_G based on these two assumptions were calculated and subsequently corrected to a standard temperature of 20° C. using the equation proposed by Molstad (8).

$$K_G \propto 24.3 - 0.175\, t \qquad (10)$$

where t is temperature, °C.

Discussion of Experimental Results. The primary usefulness of this specially designed column is in the study of the liquid film coefficients for various systems, both with and without chemical reaction. This should make it possible to find a general correlation for k_L and k'_L where k_L is the liquid film coefficient with chemical reaction and k'_L is that without chemical reaction. The ammonia-phosphate system was first studied to further ascertain the column characteristics with respect to the gas film resistance. This system has the advantage that the desired ratios of ammonium ion and the phosphate ion can be readily obtained. The overall gas film coefficient, K_G is related to the individual gas and liquid film

coefficients by the equation:

$$\frac{1}{K_G} = \frac{1}{k_G} \quad \frac{H}{k_L} \qquad (11)$$

$$\frac{H}{k_L} = \frac{1}{K_G} - \frac{1}{k_G} \qquad (11a)$$

Where K_G is the overall transfer coefficient, k_L and k_G are the individual coefficients of the liquid film and the gas film, respectively, and H is the Henry's law constant, defined by the equation:

$$p = HC \qquad (12)$$

Since k_G can be determined from the equations developed by Hwu (7) using ammonia and water and carbon dioxide and water and if K_G is determined experimentally, then the liquid film coefficient, k_L can be evaluated. However, in cases where K_g is close to the value of k_G, so that the gas film comprises the controlling resistance, the difference between these two reciprocals is very small. Therefore, a slight experimental error in the determination of K_g would result in a large error in k_L and make the correlation unreliable.

Another approach then is to compare the total transfer coefficient K_G instead of the film coefficients, k_G. Using the data for the absorption of ammonia into water reported by Hwu (7), the following correlation was obtained:

$$K_G \propto G^{0.391} \quad p^{0.377} \qquad (13)$$

Effect of Gas Rate: The numerical values of K_G were calculated by equation (9) for each of the two extremes for which the driving force could be evaluated. These values are given in

Tables 3 and 4 and are shown graphically in Plates IX to XII. For the di-ammonium phosphate system the numerical values of K_G based on the assumption of no ammonia back pressure lie below the corresponding values of K_g for ammonia absorbed in water. On the other hand, the values based on the assumption that the Henry's law constant for ammonia in water applies, lie above those of the ammonia-water system. In the mono-ammonium phosphate system the values of K_G based on both of these assumptions are greater than the corresponding values for the ammonia-water system. However, the slope of the lines appear to be nearly the same in all cases as that in the ammonia-water system. This is consistent with the absorption of ammonia in sulfuric acid as reported by Doherty and Johnson (4). In the absorption of ammonia in 3.5N sulfuric acid, their numerical values of K_G were 1.45-1.65 times those for the absorption of ammonia in water, but the slopes of the straight lines were the same.

The near constancy of the effect of gas velocity on the different systems can also be explained by Hatta's theory for absorption followed by a rapid, irreversible reaction in the liquid phase. Hatta (5) visualized a double film on the liquid side with the chemical reaction occurring at the boundary between them. Hatta further postulated that, the double liquid film disappears gradually as the concentration of the reacting solute in the liquid phase increases. In other words, the function of the reacting solute is only to reduce the resistance offered by the liquid side. The absorption of ammonia is recognized as one in which the gas film controls and the main

Table 3. Effect of gas velocity on K_G for the absorption of ammonia in mono-ammonium phosphate solution. Temperature corrected to 20° C.

Run No.	Gas Rate Ft./sec.	Liquid Concentration M./Liter	lbs./ (h4.) (sq.ft.) (atm.)	
			K_G (1)	K_G (2)
1	2.006	0.154	7.93	10.4
2	6.07	0.154	12.03	14.59
3	4.578	0.154	10.09	12.27
4	3.31	0.154	9.26	11.08
5	4.573	0.06	9.94	11.16
6	6.04	0.06	12.68	14.61
7	2.336	0.06	7.46	8.24
8	3.16	0.058	8.85	9.48
9	2.11	0.1	7.53	8.73
10	3.17	0.1	8.746	9.82
11	4.29	0.1	9.9	11.37

(1) K_G calculated by assuming that no back pressure of NH_3 existed over the solution.
(2) K calculated by assuming the Henry's law constant for NH_3-H_2O applies.

resistance to the transfer of solute gas is offered by the gas film; hence, the values of K_G do not vary much even with the presence of chemical reaction. The relationship between K_G and gas velocity, should, therefore, be almost the same in all cases.

Values of K_G for this system, (where the concentration of di-ammonium phosphate is high), calculated by using the same value of Henry's law constant as that in water, were unusually high. (Fig. 3) This indicates the inadequacy of the assumption. Since, according to this assumption, the mean driving force can be expressed by the following equation:

Table 4. Effect of gas velocity on K_G for the absorption of ammonia in di-ammonium phosphate solution. Liquid rate approximately 30 lbs./hr. Temperature corrected to 20° C.

Run No.	Gas Velocity ft./sec.	Liquid Concentration Moles/liter	Overall transfer coefficient lbs./(hr.) (sq.ft.) (atm.)		
			$K_G(1)$	$K_G(2)$	$K_G(3)$
5	4.24	0.1	7.14	8.99	7.14
6	6.057	0.1	9.57	12.34	9.89
7	4.926	0.1	8.54	11.24	8.82
8	2.171	0.1	6.51	8.19	6.64
9	2.171	0.262	6.25	12.93	6.35
10	3.296	0.262	7.77	13.94	7.88
11	6.06	0.267	11.95	25.28	12.41
12	4.93	0.267	9.71	16.01	9.98
13	2.546	0.267	5.67	8.04	5.76
14	2.171	0.09	6.61	8.17	6.79
15	3.174	0.09	8.46	10.11	8.69
16	4.3	0.09	10.01	11.89	10.26
17	5.27	0.09	9.96	11.71	10.23
18	2.171	0.069	6.27	7.2	6.38
19	6.07	0.069	11.05	12.7	11.31
20	3.31	0.069	7.75	8.77	7.94
21	4.564	0.069	8.86	10.29	9.07
22	2.17	0.024	6.79	7.06	6.93
23	2.85	0.024	7.87	8.35	8.03
24	4.3	0.024	9.94	10.63	10.25
25	5.27	0.024	10.4	11.08	10.82

(1) K_G calculated by assuming no back pressure of ammonia over the phosphate solution.
(2) K_G calculated by assuming the Henry's law constant for NH_3-H_2O applied.
(3) K_G calculated by assuming no concentration effect existed.

$$\Delta p_{lm} = \frac{(p_1-p_{e1}) - (p_2-p_{e2})}{\ln \frac{p_1-p_{e1}}{p_2-p_{e2}}} \qquad (14)$$

where p = partial pressure of the solute gas in the entering gas stream

EXPLANATION OF PLATE IX

Effect of Gas velocity on the Overall Transfer

Coefficient, K_G, for the Absorption of NH_3 in

$(NH_4)HPO_4$ solution.

Ordinate: Overall transfer coefficient, K_G,
               lbs./ (hr.) (sq.ft.) (atm.)

Abscissa: Gas velocity, V, ft./sec.

Fig. 1.   Solution concentration 0.069 M.

               Liquid rate 3/lbs. hr.

Fig. 2.   Solution concentration 0.024 M.

               Liquid rate 30 lbs./hr.

Legend:

        ▱ : Assuming no back pressure of NH_3 over
             the solution.

        O : Assuming the Henry's law constant for
             NH_3-H_2O applies.

        △ : Assuming no concentration effect.

Solid line: Data on the absorption of NH_3 in H_2O.

PLATE IX

Figure 1.

Figure 2.

EXPLANATION OF PLATE X

Effect of Gas velocity on the Overall Transfer
Coefficient, K_G, for the Absorption of NH_3 in
$(NH_4)_2HPO_4$ solution

Ordinate: Overall transfer coefficient, K_G,
                   lbs./ (hr.) (sq. ft.) (atm.)

Abscissa: Gas velocity, V, ft./sec.

Fig. 3.   Solution concentration 0.262 M.

                   Liquid rate 30 lbs./hr.

Fig. 4.   Solution concentration 0.09 M.

                   Liquid rate 31.4 lbs./hr.

Legend:

        ▱ : Assuming there is no back pressure of
             NH_3 over the solution.

        o : Assuming the Henry's law constant for
             NH_3-H_2O applies.

        △ : Assume no concentration effect.

Solid line: Data of the absorption of NH_3 in H_2O.
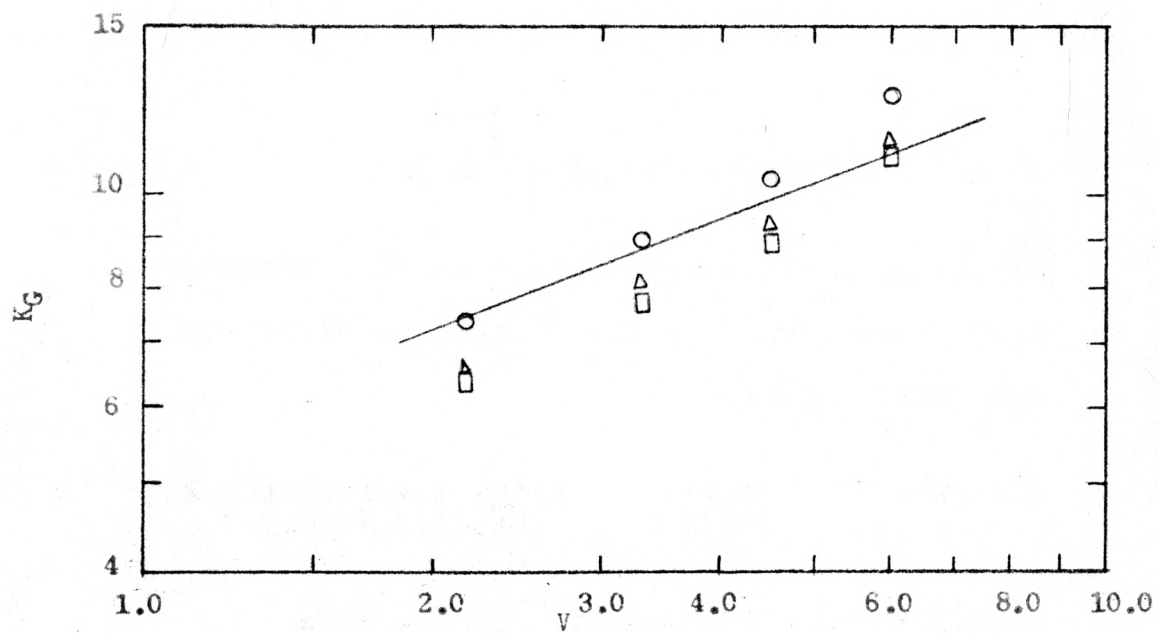
PLATE X

Figure 3.
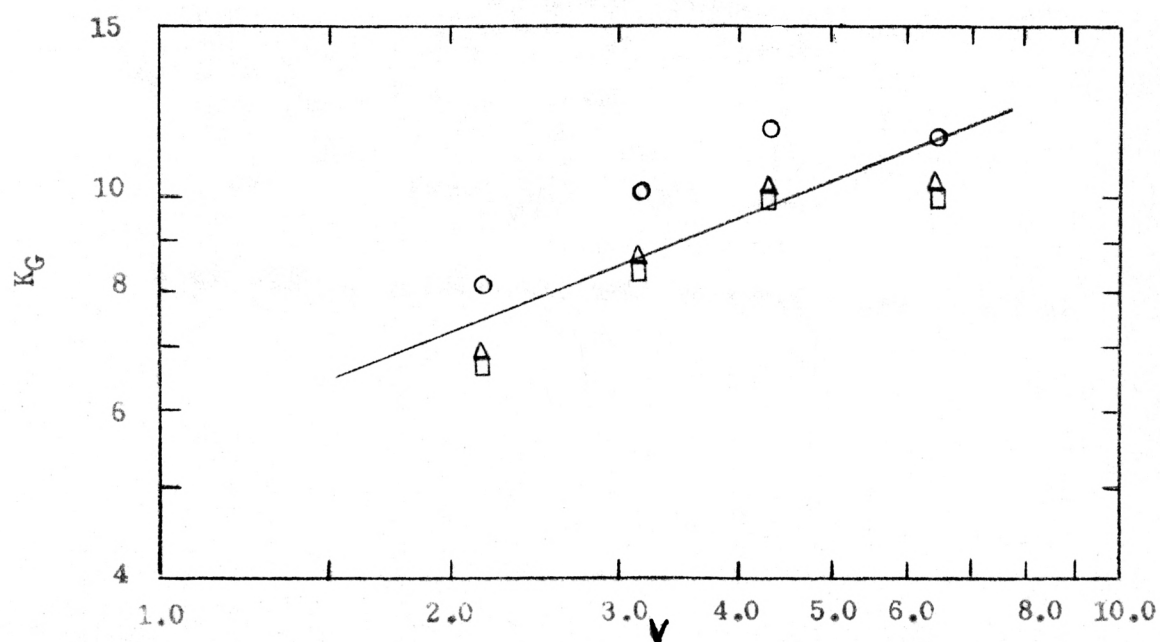
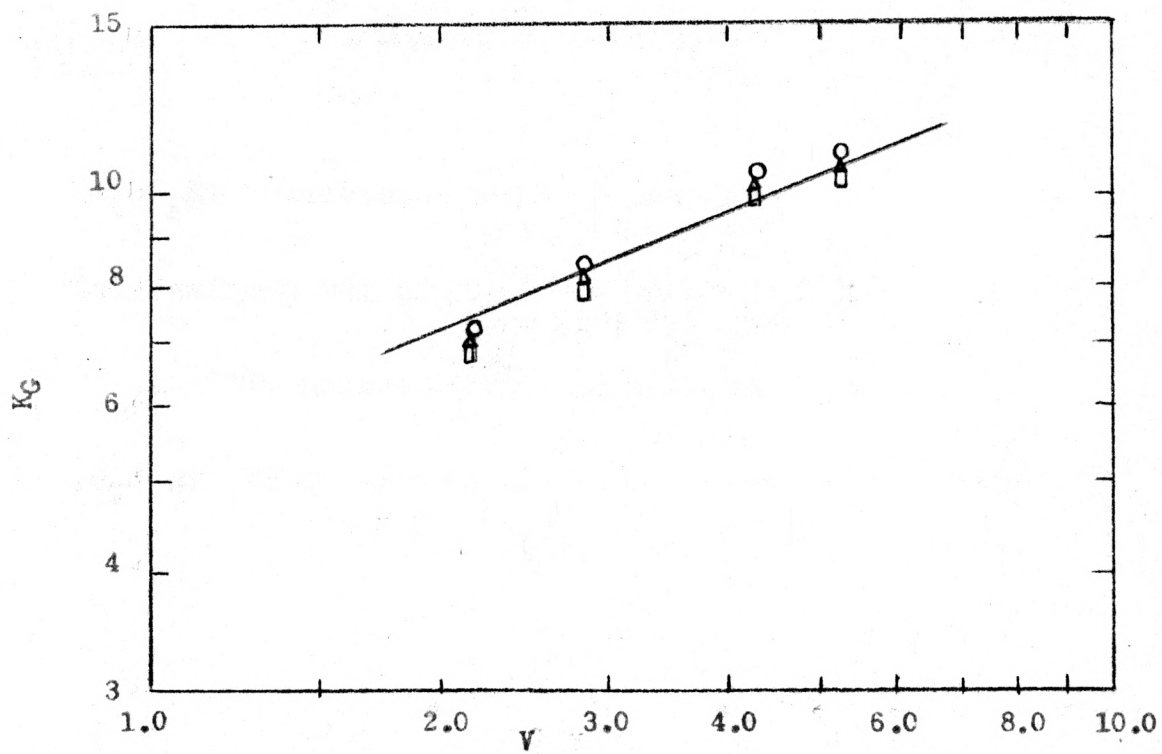Figure 4

EXPLANATION OF PLATE XI

Effect of Gas velocity on the Overall Transfer
Coefficient, K_G, for the Absorption of NH_3 in
$(NH_4)H_2PO_4$ Solution.

Ordinate: Overall transfer coefficient, K_G,
                   lbs./(hr.) (sq.ft.) (atm.)

Abscissa: Gas velocity, V, ft./sec.

Fig. 5.   Solution concentration 0.1 M

                   Liquid rate 30.4 lbs./hr.

Fig. 6.   Solution concentration 0.06 M.

                   Liquid rate 30.4 lbs./hr.

Legend:

          $\triangle$: Assuming no NH_3 back pressure over the
             solution.

          o: Assuming the Henry's law constant for
             NH_3-H_2O applies.

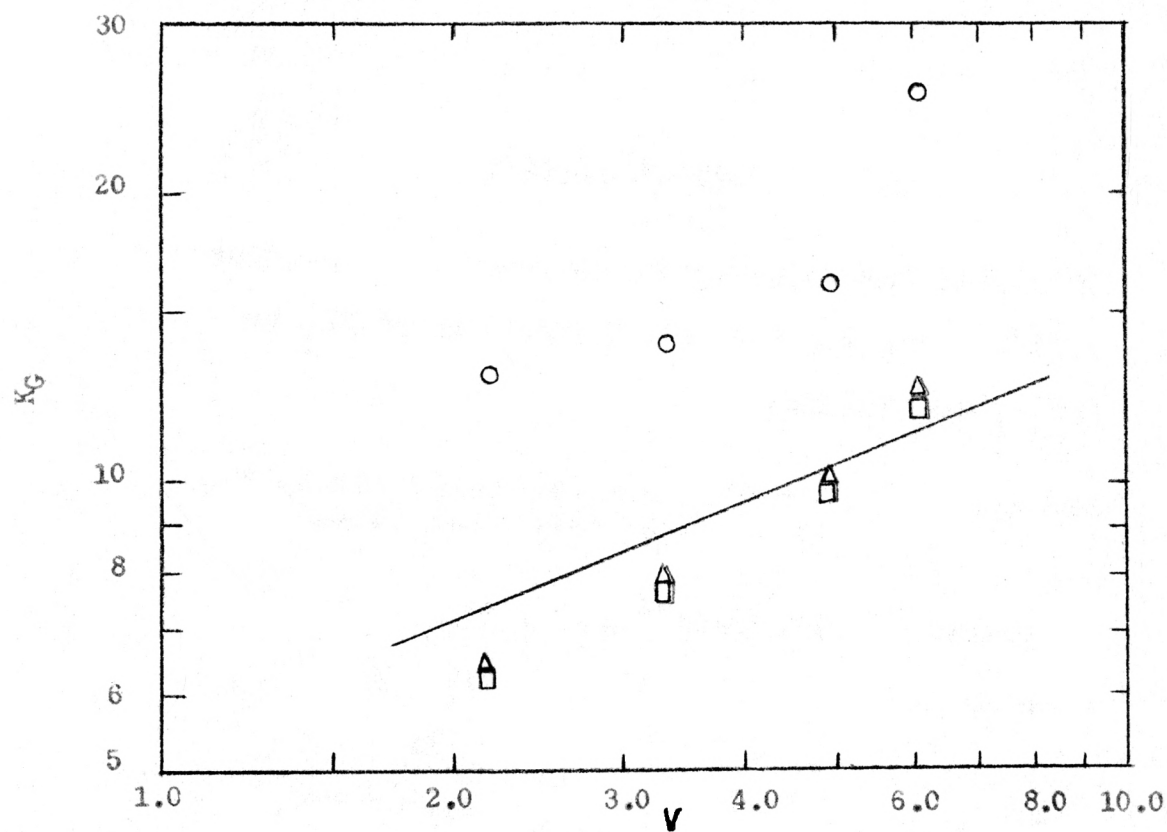Solid line: Data of the absorption of NH_3 in H_2O.
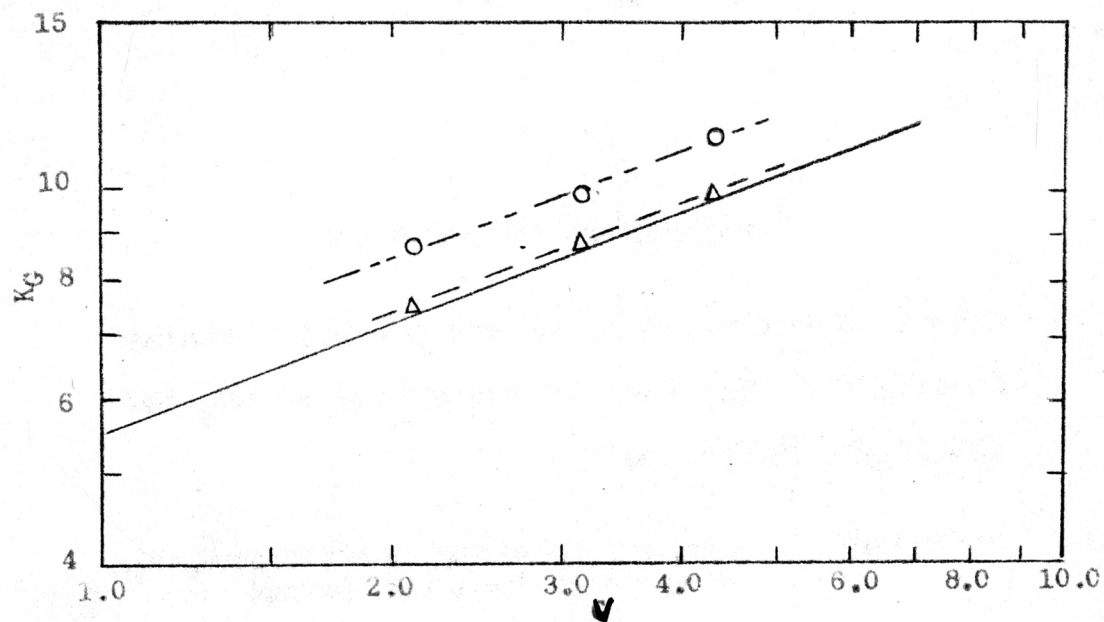
PLATE XI

Figure 5.
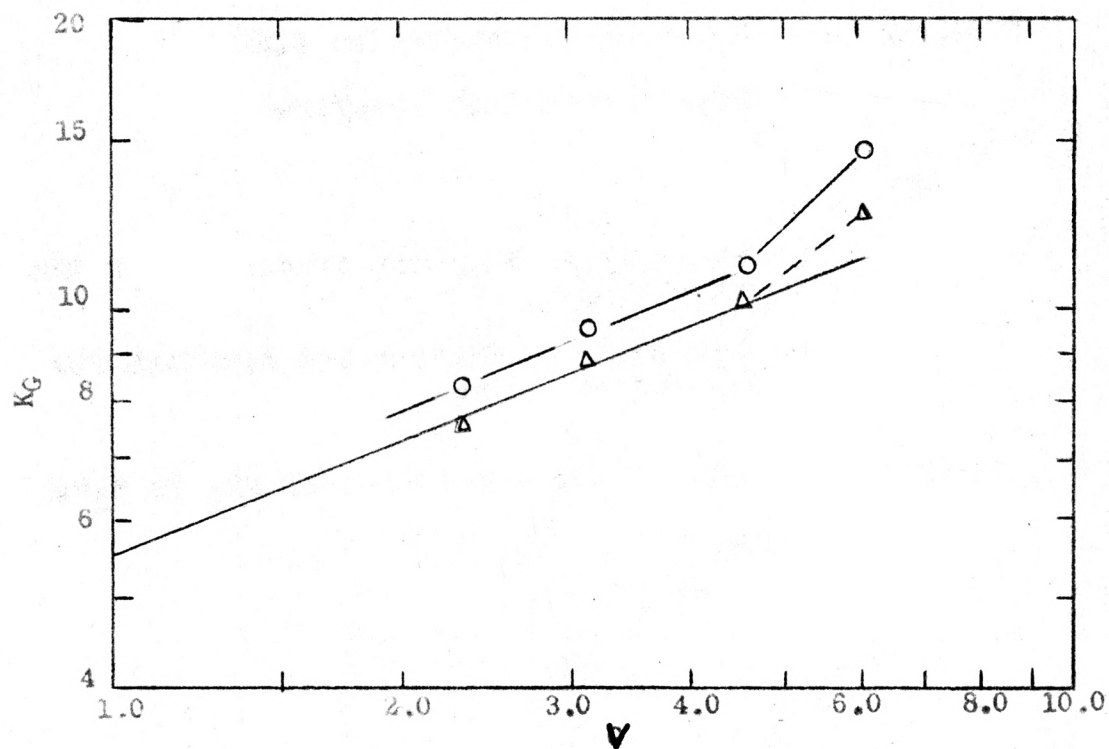
Figure 6.

EXPLANATION OF PLATE XII

Effect of Gas Rate on the Overall Transfer

Coefficient, K_G, for the Absorption of NH_3 in

$(NH_4) H_2 PO_4$.

Ordinate: Overall transfer coefficient, K_G,
               lbs./ (hr.) (sq.ft.) (atm.)

Abscissa: Gas velocity, V, ft./sec.

Solution concentration 0.154 M.

Liquid rate 30 lbs./hr.

Legend:

     △ : Assuming no back pressure of NH_3 over
          the solution.

     ○ : Assuming the Henry's law constant for
          NH_3-H_2O applies.

Solid line: Data of the absorption of NH_3 in H_2O.

PLATE XII
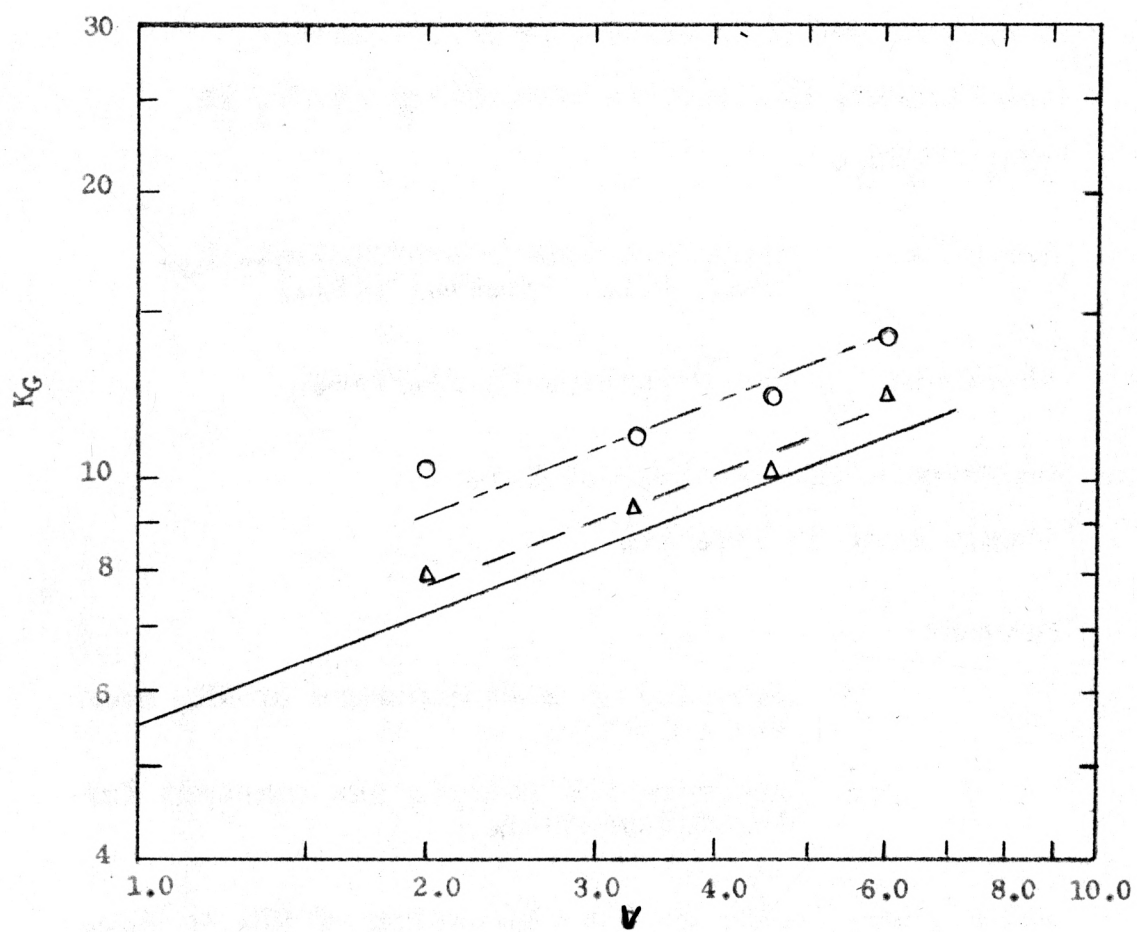
p_2 = partial pressure of the solute gas in the leaving gas stream.

p_{e1} = equilibrium partial pressure of the solute gas over the liquid stream, leaving.

p_{e2} = equilibrium partial pressure of the solute gas over the entering liquid stream.

p_{e1} and p_{e2} are expressed by the following equations (15) and (16):

$$p_{e1} = HC_2 \qquad (15)$$

$$p_{e2} = HC_1 \qquad (16)$$

C_1 and C_2 are the entering liquid concentration and outlet liquid concentration respectively. H is the Henry's law constant.

When the initial concentration of the liquid is high, p_{e2} is relatively large as compared with p_2. This makes the value of $\ln \dfrac{p_1-p_{e_1}}{p_2-p_{e_2}}$ large and the mean driving force $\triangle p_{lm}$ small. Since K_G is inversely proportional to the mean driving force, values of K_G calculated on this basis become unusually high.

Effect of Liquid Rate: Values of K_G calculated on both assumptions are given in Tables 5 and 6 and plotted versus liquid rate in Plates XIII and XIV. The slope of these straight lines appear to be nearly the same.

As may be expected from Hatta's theory, the numerical value of the ratio $K_G/K_{G(H_2O)}$ (where K_G is the overall transfer

coefficient for the absorption of ammonia in phosphate solution and $K_{G(H_2O)}$ is that for water) depends on the concentration of the liquid, the gas rate and the liquid rate. The liquid film resistance becomes small as the liquid rate increases. Since the only effect of the presence of chemical compounds is to reduce the resistance offered by the liquid film, the effect of concentration on the value of $K_G/K_{G(H_2O)}$ is less pronounced at the high liquid rate than that at low liquid rate. This observation was also made by Doherty and Johnson. The absence of this tendency in this experiment may be attributed to the fact that the liquid rate was not high enough.

Effect of Concentration: The data shown in Plates IX, X, XIV indicate that all values of K_G for the absorption of ammonia by di-ammonium phosphate solutions calculated on the assumption of no back pressure of ammonia are less than those for the absorption of ammonia in water, while those calculated using the Henry's law constant for the ammonia-water system are well above those for water. Since these represent the two extremes the true values of K_G should lie between these two limits. An assumption was made that the presence of di-ammonium phosphate has no effect at all. The absorption mechanism itself in this case is essentially the same as in the absorption of ammonia by water and the solubility of ammonia in the phosphate solution is the same as that in water. The values of K_G s calculated on this basis were in most cases, nearly the same as those for water. The values of K_G calculated this way are also given in Tables 5 and 6 and plotted against the gas velocity in Plates XV and XVI.

Table 5. Effect of Liquid rate on K_G for the absorption of ammonia in di-ammonium phosphate solution. Gas rate, 2.171 ft./sec. Temperature corrected to 20° C.

Run No.	Liquid Rate lbs./(hr.) (ft.)	Concentration Moles/liter	Overall transfer coefficient lbs./(hr.) (sq.ft.) (atm.)		
			K_G(1)	K_G(2)	K_G(3)
1	233	0.1	6.51	8.19	6.64
2	322	0.1	7.98	11.18	8.6
3	169	0.1	5.7	7.14	6.13
4	109	0.1	4.27	5.26	4.39

(1) K_G calculated by assuming no back pressure of ammonia over the phosphate solution.
(2) K_G calculated by assuming the Henry's law constant for NH_3-H_2O applies.
(3) K_G calculated by assuming no concentration effect existed.

Table 6. Effect of liquid rate on K_G for the absorption of ammonia in Monoammonium phosphate solution. Constant gas rate 3.34 ft./sec. Temperature corrected to 20° C.

Run No.	Liquid Rate lbs./(hr.) (ft.)	Concentration Moles/liter	Overall transfer coefficient lbs./(hr.) (sq.ft.) (atm.)	
			K_G(1)	K_G(2)
12	236	0.0563	8.03	9.28
13	331	0.0563	10.05	10.7
14	175	0.0563	7.76	8.35
15	114	0.0563	5.46	5.91
16	236	0.093	9.52	10.42
17	299	0.093	9.92	10.85
18	164.5	0.093	7.74	8.48
19	109.3	0.093	5.95	5.76

(1) K_G calculated by assuming no back pressure of ammonia over the phosphate solution.
(2) K_G calculated by assuming the Henry's law constant for NH_3-H_2O applies.

EXPLANATION OF PLATE XIII

Effect of Liquid Rate on the Overall Transfer

Coefficient for the absorption of NH_3 in

$(NH_4)H_2PO_4$ Solution.

Ordinate: Overall transfer coefficient, K_G,
                   lbs./(hr.) (sq.ft.) (atm.)

Abscissa: Liquid rate, Γ lbs./(hr.) (ft.)

Fig. 7.   Solution concentration 0:093 M.

                   Gas velocity 3.32 ft./sec.

Fig. 8.   Solution concentration 0.0563 M.

                   Gas velocity 3.32 ft./sec.

Legend:

     $\triangle$: Assuming no back pressure of NH_3 over
                   the solution.

     $\bigcirc$: Assuming the Henry's law constant for
                   NH_3-H_2O applies.

Solid Line: Data on the absorption of NH_3 in H_2O.

PLATE XIII
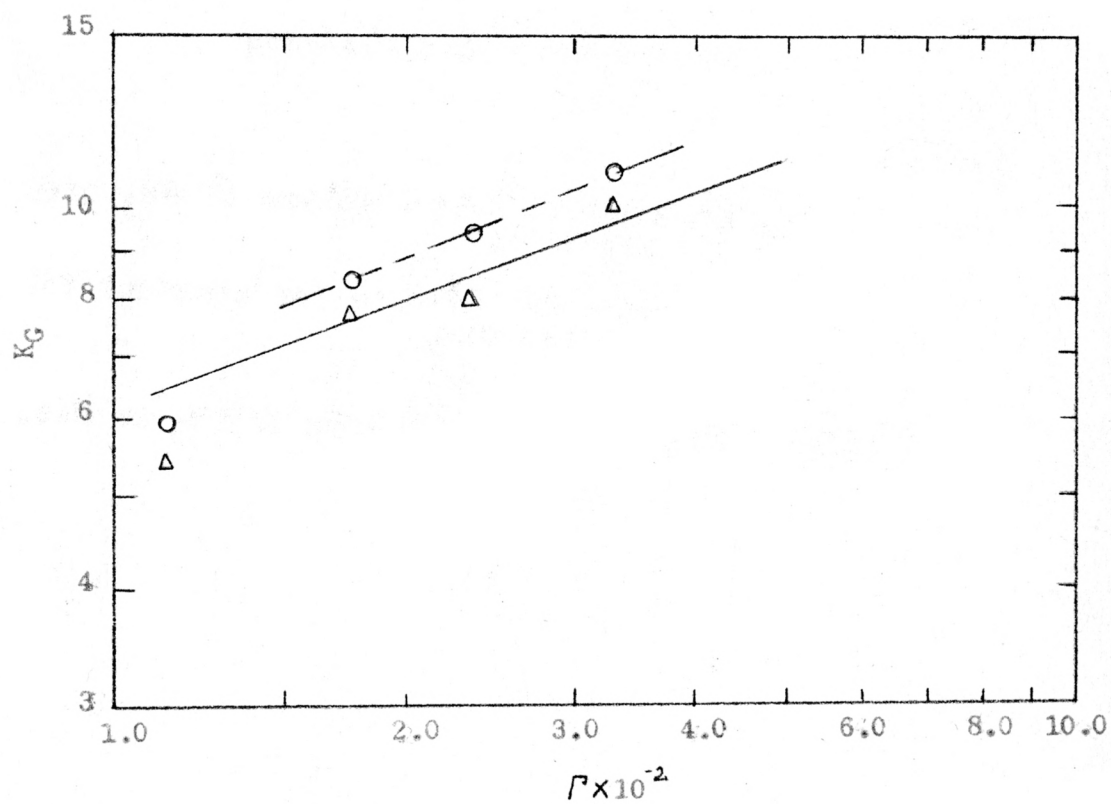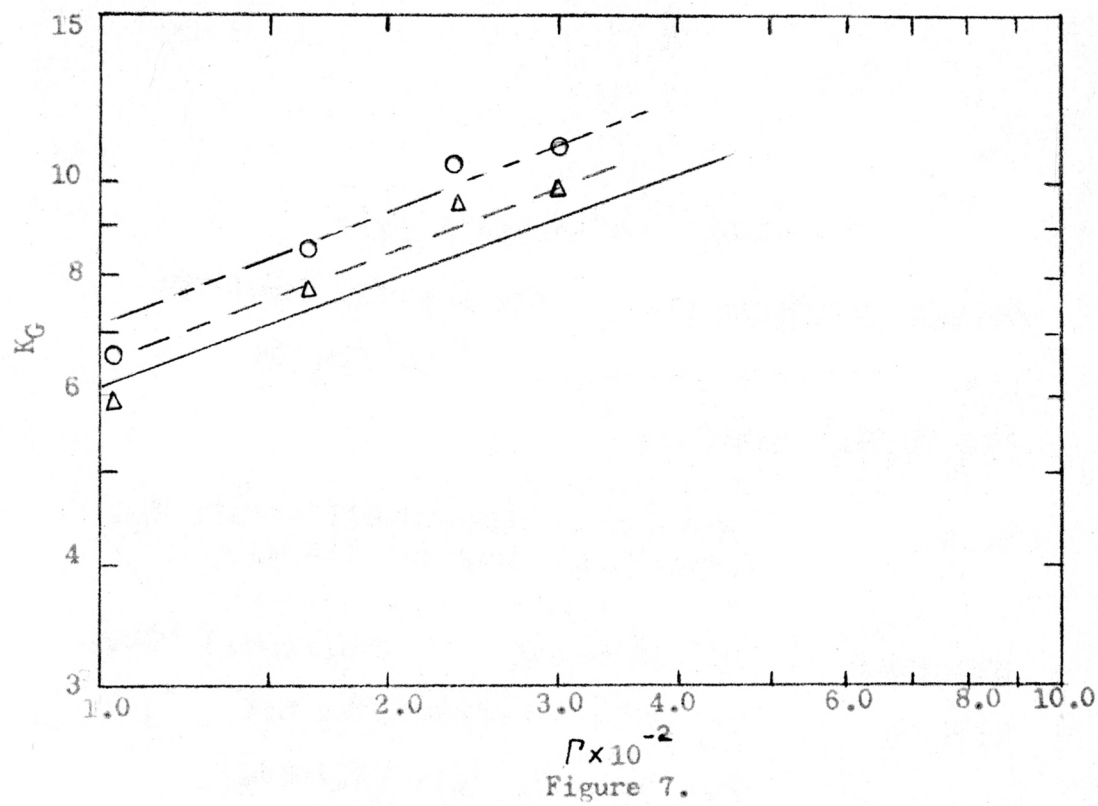
Figure 7.

Figure 8.

EXPLANATION OF PLATE XIV

Effect of Liquid Rate on the Overall Transfer
Coefficient for the Absorption of NH_3 in
$(NH_4)_2HPO_4$ Solution.

Ordinate: Overall transfer coefficient, K_G,
               lbs./ (hr.) (sq.ft.) (atm.)

Abscissa: Liquid rate, Γ, lbs./ (hr.) (ft.)

Solution concentration 0.1 M.

Gas velocity 3.32 ft./sec.

Legend:

        □ : Assuming there is no back pressure of
            NH_3 over the solution.

        △ : Assuming the Henry's law constant for
            NH_3-H_2O applies.

        O : Assuming no concentration effect.

Solid Line: Data of the absorption of NH_3 in H_2O.

PLATE **XIV**

$\Gamma \times 10^{-2}$
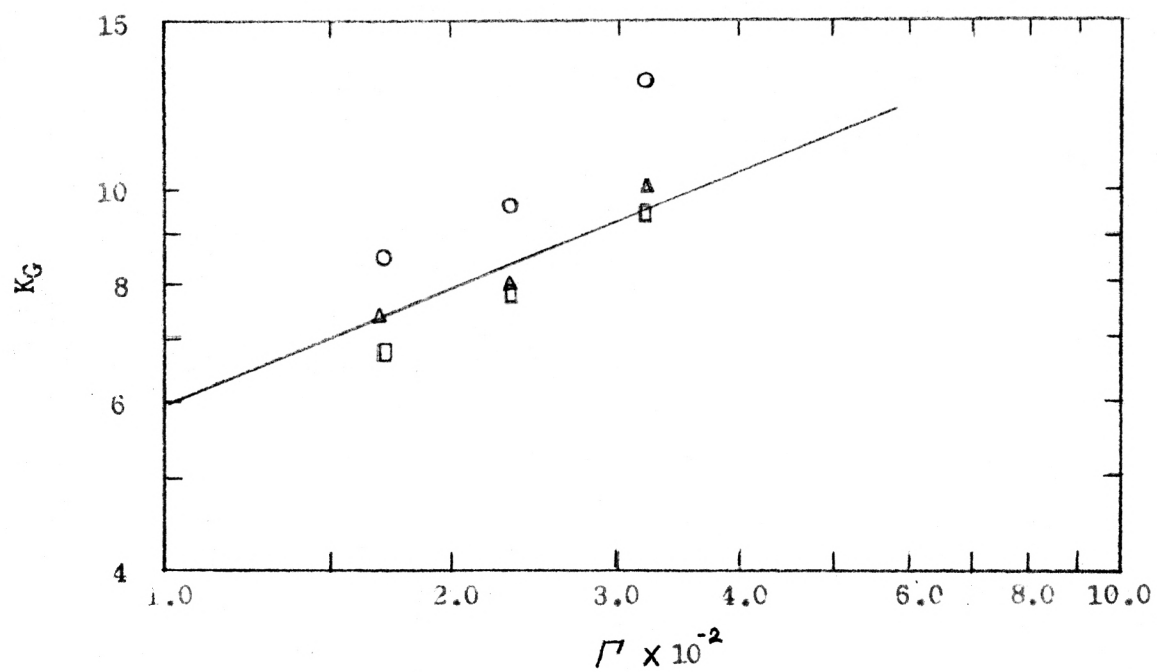
EXPLANATION OF PLATE XV

Plot of K_G versus gas velocity, V, at various

Concentration Levels for the Absorption of

Ammonia in $(NH_4)_2HPO_4$ Solution.

Ordinate: Overall transfer coefficient, K_G,
              lbs./ (hr.) (sq.ft.) (atm.)

Abscissa: Gas velocity, V, ft./sec.

Experimental conditions

    Liquid rate: 30 lbs/hļ.

    Temperature corrected to 20° C.

Legend:

        ⊖   0.262 M.

        ⊡   0.09 M.

        △   0.069 M.

        ○   0.024 M.

Remark: K_G calculated on the basis that there is no
          concentration effect.
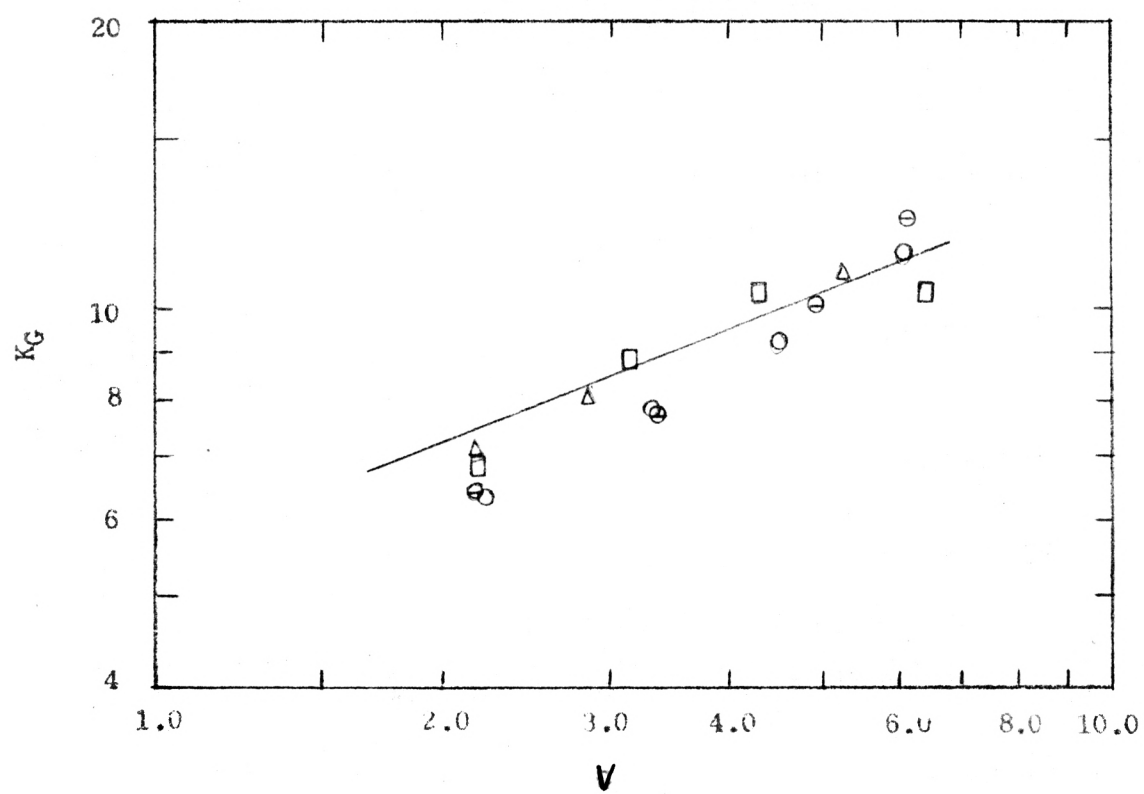
PLATE **XV**

EXPLANATION OF PLATE XVI

Effect on Concentration on the Overall Transfer
Coefficient, K_G, for the absorption of Ammonia in
$(NH_4)H_2PO_4$ Solution.

Ordinate: Overall transfer coefficient, K_G,
               lbs./ (hr.) (sq.ft.) (atm.)

Abscissa: Concentration C, M./liter

Liquid rate: 30 lbs./hr.

Gas velocity:

    Curve I   6.07 ft./sec.

    Curve II   4.3 ft./sec.

    Curve III 2.17 ft./sec.

Remark: K_G calculated on the assumption that there
         was no concentration effect.

PLATE XVI
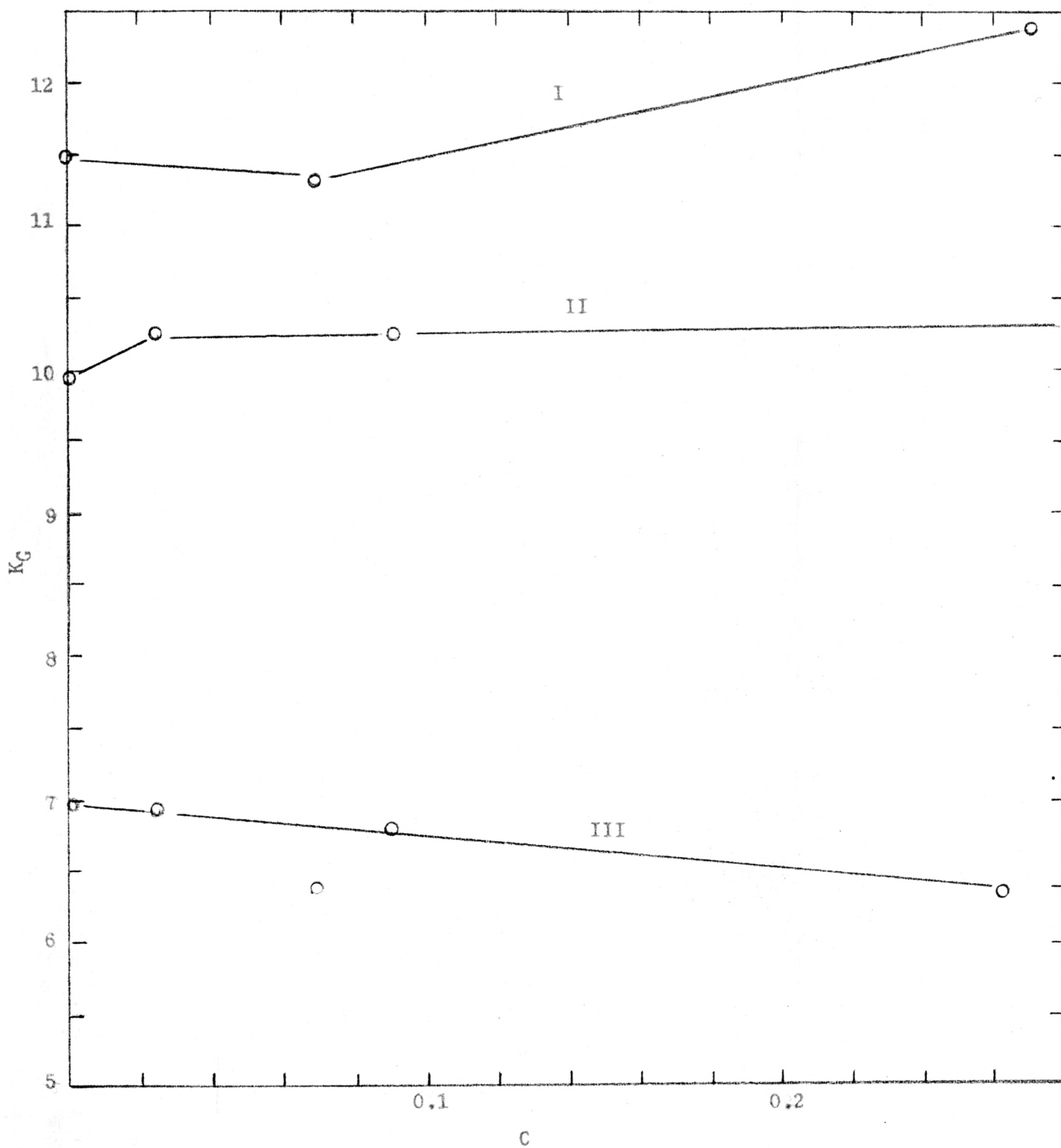
Apparently liquid concentration has no effect on the film coefficients determined in this way.

For the absorption of ammonia in mono-ammonium phosphate solutions, values of K_G calculated on both limiting conditions (a) and (b) as stated before, are greater than those for water in most cases. The effect of concentration on K_G and K_G is shown in Plates XVII and XVIII.

According to Hatta's theory, when the concentration is below a certain value, the relation between concentration and K_G is linear. Values of K_G calculated by using the solubility data of ammonia in water do show this tendency. However, K_G calculated on the assumption that there is no back pressure, does not show this characteristic.

The difference in behavior of these two similar compounds can be explained by the fact that the pH of the di-ammonium salt solution was about 7.5, which is similar to water, while the mono-ammonium salt solution was slightly acidic. The absorption of ammonia in water can be expressed as:

$$NH_3 \rightleftharpoons NH_{3aq}$$
$$NH_{3aq} + H_2O \rightleftharpoons NH_4OH \rightleftharpoons NH_4^+ + OH^-$$
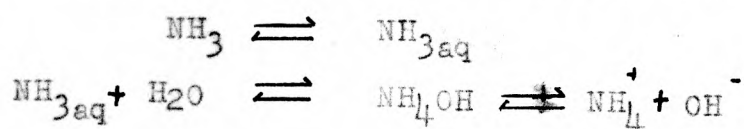
The first step is simply physical equilibrium, while the second one involves chemical reaction. When the liquid used for absorption is acidic, the equilibrium will tend to shift from left to right. This results from the neutralization between hydrogen ion and hydroxyl ion. The result is that more ammonia is absorbed.

EXPLANATION OF PLATE XVII

Effect of Concentration on the Overall Transfer
Coefficient, K_G, for the absorption of Ammonia
in $(NH_4)H_2PO_4$ Solution.

Ordinate: Overall transfer coefficient, K_G,
               lbs./ (hr.) (sq.ft.) (atm.)

Abscissa: Concentration, C, moles/liter

Liquid rate: 30 lbs./hr.

Gas velocity:

    Curve I   4.5 ft./sec.

    Curve II   3 ft./sec.

    Durve III   2.5 ft./sec.

Remark: K_G calculated using Henry's law constant
         for NH_3-H_2O.

PLATE **XVII**
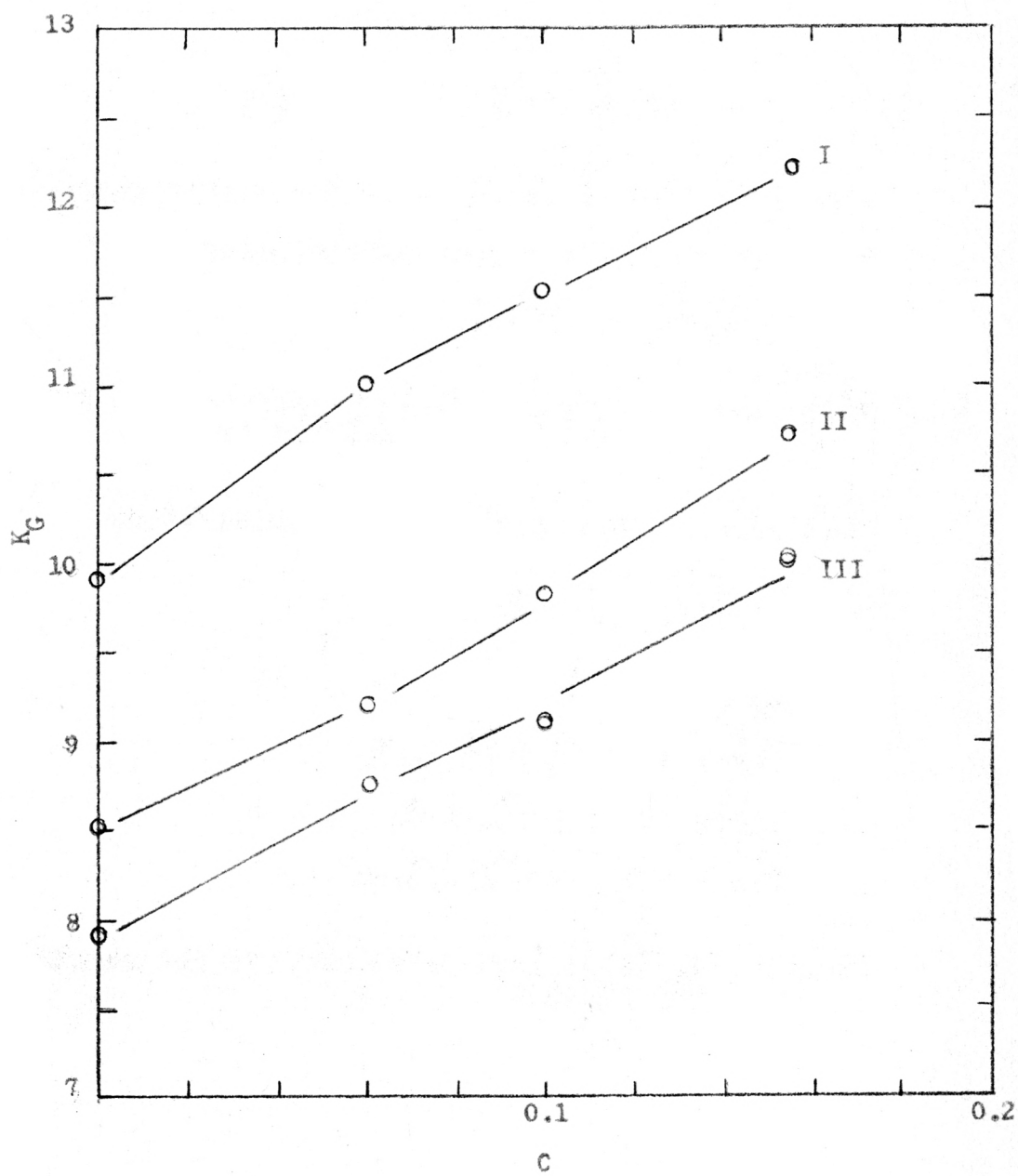
EXPLANATION OF PLATE XVIII

Effect of Concentration on the Overall Transfer
Coefficient, K_G, for the Absorption of Ammonia
in $(NH_4)H_2PO_4$ Solution.

Ordinate: Overall transfer coefficient, K_G,
                 lbs./ (hr.) (sq.ft.) (atm.)

Abscissa: Concentration, C, moles/liter

Liquid rate: 30 lbs./hr.

Gas velocity:

    Curve I   4.5 ft./sec.

    Curve II   3 ft./sec.

    Curve III   2 ft./sec.

Remark: K_G calculated on the assumption of not back
         pressure of NH_3 over the solution.

PLATE XVIII
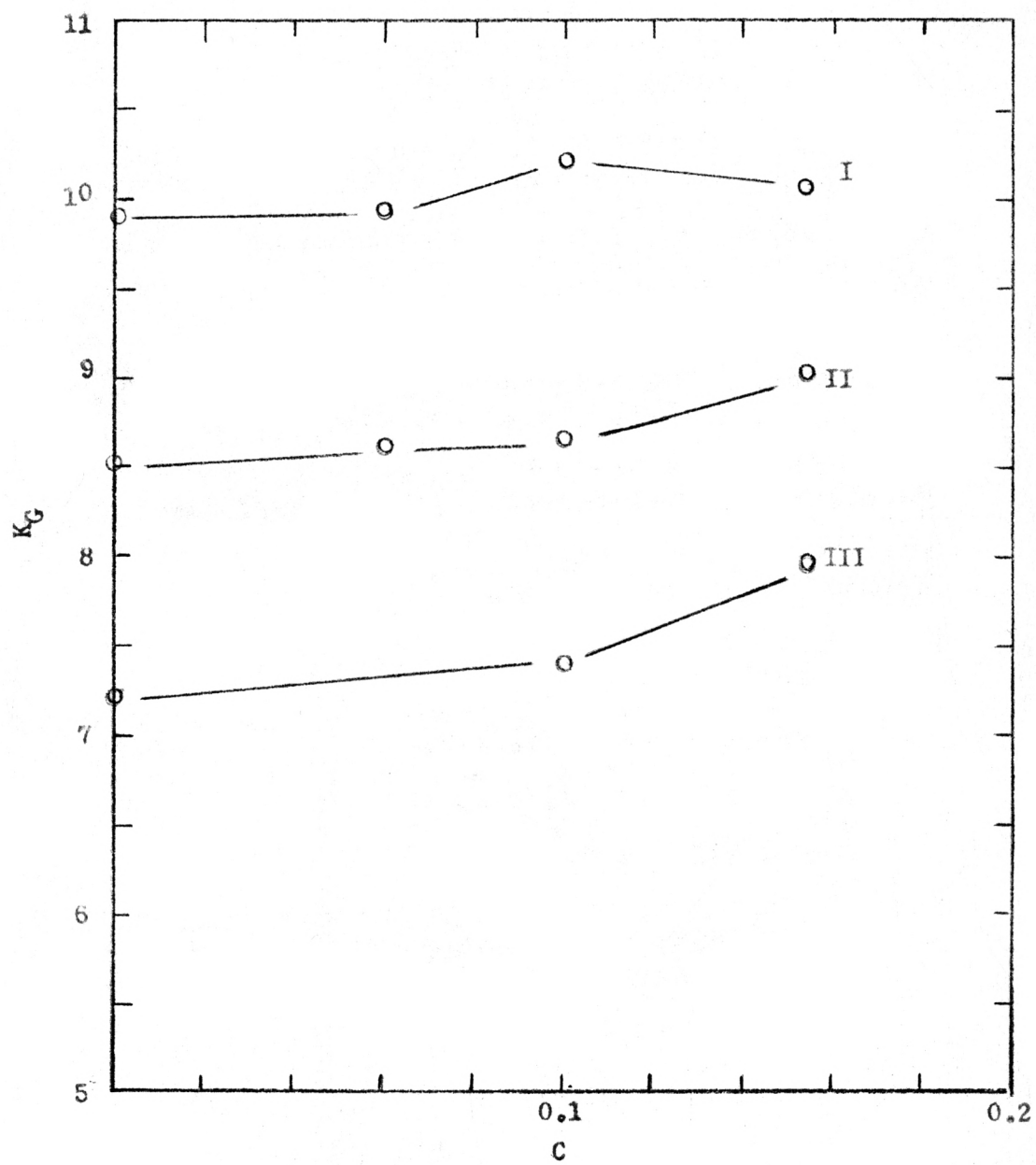
The actual mechanism is by no means so simple, because it
involves a series of equilibrium systems. However, this kind
of reaction can be considered as a neutralization reaction be-
tween an acid and a base. As far as neutralization is concerned,
the most important factors are the hydrogen ion concentration
and the hydroxyl ion concentration. The higher values of K_G
for the absorption of ammonia is mono-ammonium phosphate solu-
tion compared with those for water can be explained by the fact
that the mono-ammonium salt solution is slightly acidic. Along
this line of reasoning, it is expected that if some other sub-
stances like $Na(NH_4)HPO_4$ or $Na(NH_4)_2PO_4$ are used for the ab-
sorption of ammonia, the corresponding values of K_G should be
less than these for water. This prediction is shown in Plate
XIX.

CONCLUSIONS

The results of the present work and the conclusions derived
therefrom are summarized as follows:

1. The liquid film coefficient for the absorption of chlo-
rine in water was determined. It was shown that k_L varies with
0.81 power of the liquid rate. This exponent is lower than that
for the absorption of carbon dioxide in water. This is con-
sistent with the previous work of Vivian and Whitney (14). The
following empirical equation was obtained:

$$k_L = 0.01163 \, \Gamma^{0.81}$$

2. It was found that the numerical value of K_G for the

EXPLANATION OF PLATE XIX

Prediction of the Overall Transfer Coefficient, K_G,
for the absorption of NH_3 in different solutions

Ordinate: Overall transfer coefficient, K_G,
                 lbs./ (hr.) (sq.ft.) (atm.)

Abscissa: Gas velocity, G ft./sec.

Curve I $(NH_4)H_2PO_4$
Curve II $(NH_4)_2HPO_4$
Curve III $Na(NH_4)HPO_4$
Curve I Na_2HPO_4

65

PLATE XIX
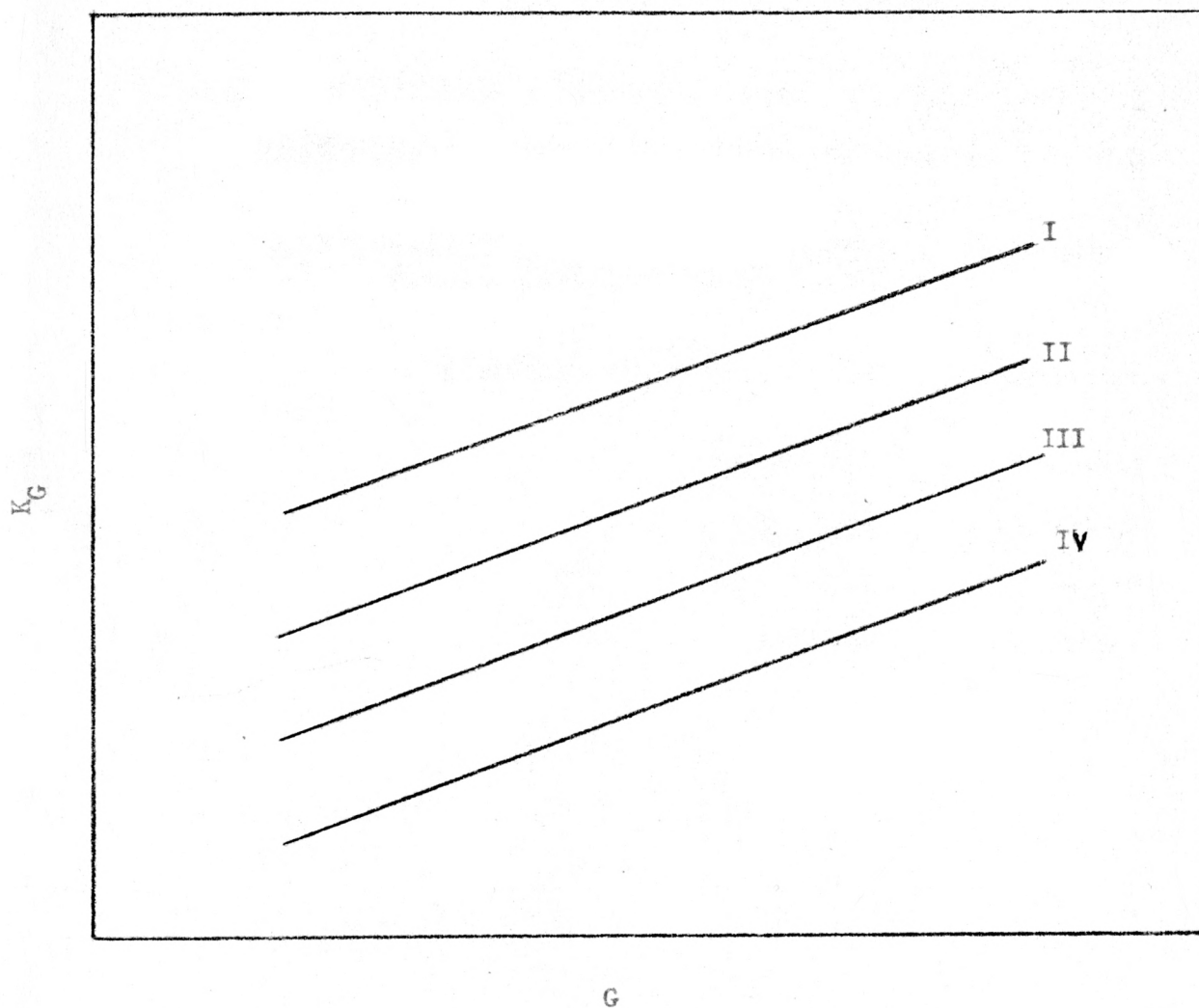
absorption of ammonia in di-ammonium orthophosphate solution
is essentially the same as that in water. No concentration ef-
fect was observed.

3. The absorption of ammonia in mono-ammonium phosphate
solution was also studied. The liquid rate effect and the gas
rate effect seem to be nearly the same as that for water. It
was also shown that K_G does increase with an increase in con-
centration.

ACKNOWLEDGMENTS

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APPENDIX

Table 1. Absorption of Chlorine into water at varied gas rate.

	Runs				
	1	2	3	4	5
Liquid rate					
W (lbs./hr.)	31.4	31.4	31.4	31.4	31.4
(lbs./hr.ft.)	247	247	247	247	247
Gas rate					
Q cu.ft./hr.	28	37.6	24.4	19.3	41.4
V (ft./sec.)	2.84	3.82	2.26	1.96	4.21
Liq. conc.(gram/liter)					
Inlet	0	0	0	0	0
Outlet	0.353	0.3693	0.3376	0.4104	0.397
Gas composition (lbs. Cl_2/lbs. air)					
Inlet	0.2756	0.2978	0.2679	0.2661	0.2568
Outlet	0.2697	0.2933	0.2615	0.2562	0.2524
Gas pressure (mm Hg)					
Inlet	735.4	735.9	735	727.3	730
Outlet	735.8	735.4	734.5	727	728.4
Liq. temp. ($^{\circ}$C.)					
Inlet	25.6	25.6	25.6	27.8	27.8
Outlet	26.8	25.9	26.1	29	29
k_L lbs./(hr.)(sq.ft.) ($\frac{lb.}{cu.ft.}$)	0.654	0.665	0.638	0.845	0.818
k_L (corrected to 20°C.)	0.58	0.59	0.565	0.706	0.684

Table 2. Liquid-film coefficient for the absorption of chlorine into water at varied liquid rate.

	Run 1	Run 2	Run 3	Run 4	Run 5	Run 6	Run 7	Run 8	Run 9	Run 10	Run 11	Run 12	Run 13	Run 14	Run 15	Run 16
Liq. rate																
W (lbs./hr.)	31	24.8	38.1	20.7	36.2	22.9	16.9	13.2	46.5	53.8	55.2	43.7	51.9	15.1	18.8	13.2
(lbs./(hr.)(ft.)	244	195.3	300	163	285	180.3	133	103.9	346.6	423.6	434.6	344.1	408.7	118.9	148	103.9
Liq. conc. (gram/liter																
Inlet	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
Outlet	2.82	3.02	2.244	2.621	2.4225	2.273	2.722	2.74	2.4625	2.403	2.502	2.70	2.61	3.098	2.988	3.078
Gas composition	Pure Cl_2	Pure Cl_2	Pure Cl_2	Pure Cl_2	Pure Cl_2	Pure Cl_2	Pure Cl_2	Pure Cl_2	Pure Cl_2	Pure Cl_2	Pure Cl_2	Pure Cl_2	Pure Cl_2	Pure Cl_2	Pure Cl_2	Pure Cl_2
Gas pressure (mm Hg)																
Inlet	738.6	738.7	738.8	738.8	737.9	738.1	738.2	734	734.1	734.4	734.7	735	733	733.4	733.1	734.9
Outlet	738	738	738.2	738	737.3	737.4	737.4	733.3	733.5	733.5	733.8	734.2	731.4	732.5	732.3	733.9
Liq. temp. (°C.)																
Inlet	23	23	23.5	23.5	25	25	25.2	24.6	25.5	25.5	23.5	23.5	24.5	25.5	25.5	26.5
Outlet	23	23.5	24	24.5	26.9	26.5	28	27.2	27.2	27.2	24.2	25.2	25.5	28.2	28.2	30
k_L (lbs./(sq.ft.)(hr.)($\frac{lb.}{cu.ft.}$)																
(I)*	1.381	1.052	1.1238	0.7505	1.2226	0.7268	0.7115	0.5377	1.6638	1.874	1.9055	1.7356	1.9432	0.7215	0.8883	0.6319
(II)**	1.904	1.4576	1.474	1.013	1.5568	0.9025	0.8803	0.6808	2.0502	2.3049	2.4864	2.2895	2.5098	0.909	1.1046	0.812
k_L (corrected to 20°C.)																
(I)*	1.2978	0.9886	1.0367	0.6997	1.0787	0.6413	0.6188	0.4744	1.4554	1.6398	1.7578	1.5818	1.75	0.6498	0.7704	0.5314
(II)**	1.7889	1.3694	1.3598	0.9345	1.3736	0.7963	0.7656	0.6007	1.7934	2.0162	2.2937	2.0867	2.2603	0.8186	0.958	0.6878

* k_L calculated by using total chlorine concentration as driving force.

** k_L calculated by using molecular chlorine concentration as driving force.

Table 3. Overall coefficient for the absorption of ammonia into diammonium hydrogen phosphate solution.

	Run 1	Run 2	Run 3	Run 4	Run 5	Run 6	Run 7	Run 8	Run 9	Run 10	Run 11	Run 12	Run 13	Run 14	Run 15	Run 16	Run 17	Run 18	Run 19	Run 20	Run 21	Run 22	Run 23	Run 24	Run 25
Liq. rate																									
W (lbs./hr.)	29.6	40.8	21.5	13.9	29.6	29.6	29.6	29.6	29.6	29.3	30	29.5	24.4	31.4	31.4	31.4	31.4	31.4	31.4	31.4	31.4	30	30.3	31.4	29
G (lbs./hr.ft.)	233	322	169	109	233	233	233	233	233	231	236	232	229	247	247	247	247	247	247	247	247	236	2.38	247	228
Gas rate																									
Q (cu.ft./hr.)	21.37	21.37	21.37	21.37	41.7	59.5	48.5	21.37	21.37	32.44	59.5	48.5	25	21.4	31.2	42.3	5.19	21.4	59.5	32.6	44.9	21.4	27	42.3	51.8
V (ft./sec.)	2.171	2.171	2.171	2.171	4.24	6.057	4.926	2.171	2.171	3.296	6.06	4.93	2.546	2.171	3.174	4.3	5.27	2.171	6.07	3.31	4.564	2.17	2.85	4.3	4.27
Liq. conc.																									
$(NH_4)H_2PO_4$ (M)	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.262	0.262	0.267	0.267	0.267	0.09	0.09	0.09	0.09	0.069	0.069	0.069	0.069	0.024	0.024	0.024	0.024
NH_3 (g N_2/100g sol'n)																									
Inlet	0.278	0.270	0.278	0.270	0.284	0.282	0.282	0.278	0.712	0.712	0.7	0.715	0.715	0.252	0.252	0.252	0.252	0.186	0.186	0.186	0.186	0.67	0.066	0.065	0.064
Outlet	0.368	0.347	0.394	0.43	0.2825	0.405	0.396	0.368	0.802	0.816	0.847	0.840	0.84	0.338	0.380	0.408	0.415	0.27	0.332	0.294	0.293	0.154	0.196	0.22	0.23
Gas composition (lbs.NH_3/lb.air)																									
Inlet	0.279	.0279	.0279	.0279	.02243	.02044	.0203	.0279	0.279	.0233	.02044	.0208	0.0383	0.0279	0.03036	0.0292	.0278	0.0279	.022	.026	.0298	.0279	0.034	0.292	.0279
Outlet	.0067	.00443	.01362	.01116	.01023	.01019	.00925	.0067	.0067	.0075	.00804	.0082	0.0132	0.0065	.00852	.00957	.01103	.00697	.00913	.0085	.0083	.00623	.0091	.00966	.0108
Gas pressure (mm Hg)																									
Inlet	732.5	732.5	732.6	732.5	737.8	740.9	739	732.5	734.7	735.9	737.8	735.8	735.3	745.7	746.5	747.9	749.5	728.1	733.5	729.2	730.9	739	739.7	741.3	743
Outlet	8.27	732.2	732.2	732.2	735.9	737	736.3	8.27	734.5	734.7	734.7	733.9	733.3	745.1	745.5	745.9	746.6	727.5	729.6	728	728.7	738.7	738.9	739.6	740.1
Liq. temp (°C.)																									
Inlet	24.8	24.8	25.9	27	27.3	27	27.0	24.8	23.0	23.8	25.6	24.1	24.9	25.5	25.5	26.5	26.5	23	21.8	23	23.6	25	2.52	25.1	25
Outlet	24.9	23.8	25.9	27	27.2	27	26.8	24.9	23.9	23.0	25.7	24.1	24.9	26.5	26.5	26.5	26	23	21.7	23.2	23.7	25	25.5	25.3	25.3
Kg (lbs./chr.)(sq.ft.)(atm)																									
(I)*	6.24	7.60	5.41	4.02	6.71	9.	8.05	6.24	6.01	7.52	11.39	9.34	5.44	6.28	8.04	9.5	9.46	6.11	10.89	7.55	8.59	6.5	7.51	9.5	9.99
(II)**	8.45	10.75	6.79	4.95	8.45	11.6	10.6	7.86	12.44	13.5	24.1	15.4	7.71	7.76	9.6	11.29	11.12	7.02	12.31	8.54	9.97	6.76	7.97	10.17	10.6
(III)***	6.37	8.75	5.89	4.15	6.71	9.3	8.32	6.37	6.11	7.86	11.83	9.6	5.54	6.45	8.25	9.74	9.71	6.22	11.14	7.73	8.79	6.64	7.67	9.8	10.3
Kg (corrected to 20° C.)																									
(I)*	6.51	7.98	5.7	4.27	7.14	9.57	8.54	6.51	6.25	7.77	11.95	9.71	5.67	6.61	8.46	10.01	9.46	6.27	11.05	7.75	8.86	6.79	7.87	9.94	10.4
(II)**	8.19	11.18	7.14	5.26	8.99	12.34	11.24	8.19	12.93	15.94	25.28	16.01	8.04	8.17	10.11	11.89	11.71	7.2	12.7	8.77	10.29	7.06	8.35	10.63	11.08
(III)***	6.64	8.6	6.13	4.39	7.14	9.89	8.82	6.64	6.35	7.88	12.41	8.98	5.76	6.79	8.69	10.26	10.23	6.38	11.31	7.94	9.07	6.93	8.03	10.25	10.82

* Assuming no back pressure of NH_3 over the solution.
** Assuming the Henry's law constant for NH_3-H_2O applies.
*** Assuming no concentration effect.

Table 4. Overall coefficients for the absorption of ammonia into mono ammonium dihydrogen phosphate solution.

	Run 1	Run 2	Run 3	Run 4	Run 5	Run 6	Run 7	Run 8	Run 9	Run 10	Run 11	Run 12	Run 13	Run 14	Run 15	Run 16	Run 17	Run 18	Run 19
Liq. rate																			
W (lbs./hr.)	30	30	30	30	30.4	30.4	30.4	30	30.4	30.4	30.4	30	42	22.25	14.44	30	38	22	13.88
(lbs./hr.ft.)	233	233	233	233	234	234	234	233	234	234	234	236	331	175	114	236	299	165.4	109.3
Gas rate																			
Q (cu.ft./hr.)	19.74	59.76	40.05	32.58	44.99	59.45	22.99	31.26	20.75	31.2	42.2	32.9	32.9	32.9	32.9	32.9	32.9	32.9	32.9
V (ft./sec.)	2.006	6.07	4.578	3.31	4.573	6.04	2.336	3.16	2.11	3.17	4.29	3.34	3.34	3.34	3.34	3.34	3.34	3.34	3.34
Liq. conc.																			
$(NH_4) H_2 PO_4$ (M)	0.154	0.154	0.154	0.154	0.06	0.06	0.06	0.058	0.1	0.1	0.1	0.0563	0.0563	0.0563	0.0563	0.093	0.093	0.093	0.093
NH_3 (N_2 gram/100 grams sol'n)																			
Inlet	0.24	0.24	0.247	0.247	0.104	0.104	0.104	0.078	0.160	0.166	0.166	0.08	0.08	0.08	0.08	0.134	0.134	0.132	0.136
Outlet	0.326	0.4	0.378	0.369	0.228	0.24	0.194	0.138	0.261	0.3	0.324	0.218	0.19	0.18	0.302	0.284	0.256	0.318	0.388
Gas composition (NH_3 lbs./lb. of air)																			
Inlet	.02619	.0221	.02274	.02608	.0227	.0186	0.02585	0.03089	.0284	.03046	.02942	0.0316	0.0316	0.0316	0.0316	0.0316	0.0316	0.0316	0.0316
Outlet	.00408	.00861	.00808	.00708	.00818	.00728	0.00573	0.00827	.00891	.0826	.01007	0.01011	0.00762	0.0107	0.015	0.0083	0.0075	0.0104	0.0134
Gas pressure (mm Hg)																			
Inlet	720.1	725.8	723.1	721.4	730.2	732.9	739.3	739.6	735.1	735.1	735.1	728.5	728.5	728.5	728.5	733.5	733.5	735.5	735.5
Outlet	719.8	721.9	721	720.3	727.9	729.2	738.2	738.7	734.6	734.6	734.6	727.5	727.5	728.5	728.5	733.3	732.3	732.3	732.3
Liq. temp. (° C.)																			
Inlet	24.2	24	24	24.2	28.7	29	25.8	25.8	26	26	26	24.2	24.5	26.3	28	22.2	22.3	23.2	24
Outlet	24	25	24.5	25.2	29	29.2	26	26.2	26.5	26.5	26.5	25	25.1	27.2	28.8	23.2	23.3	24.2	25
Kg (lbs./(hr.)(sq.ft.)(atm)																			
(I)*	7.659	11.57	9.73	8.9	9.2	11.7	7.09	8.37	7.16	8.41	9.25	9.69	9.64	7.33	5.07	8.99	9.69	7.406	5.7
(II)**	10.05	14.03	11.83	10.65	10.33	13.49	7.83	8.97	8.3	9.44	10.75	10.59	10.27	7.89	5.49	9.84	10.59	8.81	6.54
Kg (corrected to 20° C.)																			
(I)*	7.93	12.03	10.09	9.26	9.94	12.68	7.46	8.85	7.53	8.746	9.9	8.03	10.05	7.76	5.46	9.52	9.92	7.74	5.93
(II)**	10.4	14.59	12.27	11.16	11.16	14.61	8.24	9.48	8.73	9.82	11.37	9.28	10.7	8.35	5.91	10.42	10.85	8.48	5.76

* Assume no back pressure of NH_3 over the solution.

** Assume the Henry's law constant for NH_3 - H_2O applies.

Sample Calculation

A. Absorption of Chlorine in Water

Run No. 3

Observed data:

Liquid Rate
W = 38.1 lb./hr. or 16.86 l./hr.
= 300 lbs./hr.ft.
Liquid Concentration
Inlet X_1 = 0
Outlet X_2 = 2.224 gram Cl_2 per liter
Liquid Temperature
Inlet   23.5° C.
Outlet 24° C.
Atmospheric Pressure = 740.1 mm Hg
Room Temperature = 27° C.
Manometer Reading
Column Inlet 2.2 mm Hg
Column Outlet 1.6 mm Hg
Absorption Surface = 0.22 sq. ft.

Calculation:

Corrected Pressure = 740.1 - 27 (0.128) = 736.6 mm Hg
Partial Pressure of Chlorine
Inlet = 736.6 - 2.2 = 738.8 mm Hg = 0.972 atm
Outlet= 736.6 - 1.6 = 738.2 mm Hg = 0.971 atm
Interfacial Equilibrium Pressure
Inlet = 6.64 grams of Cl_2 per liter
Outlet = 6.55 grams of Cl_2 per liter
Mean Driving Force is calculated from the following equation:

$$\Delta C_{lm} = \frac{(6.64 - 2.244) - 6.55}{\ln \frac{6.64 - 2.244}{6.55}} = 5.4014 \; g/l$$

$$k_L = \frac{W(X_2 - X_1)}{A \; \Delta C_{lm}} = \frac{2.244 \times 16.86}{0.22 \times 5.4014} = 31.8 \; g/hr.\text{-}ft^2\text{-}\frac{g}{l} = 1.125 \; \frac{lbs/hr.\,ft^2}{lb/ft^3}$$

Since

$$k_L \propto e^{0.021t}$$

$$k_L[\text{corrected to } 20°] = 1.125 \times {}^{1.523}/_{1.65} = 1.0367$$

In the calculation of pseudo transfer coefficient, only the difference of molecule chlorine concentration can be counted as

driving force and molecule concentration of chlorine was calculated by the following equation:

$$C = H\phi$$

Values of C are given by Whitmey and Vivian

The interfacial equilibrium concentrations were found as follows:
    Inlet   5.342 grams of Cl_2 per liter
    Outlet   5.336 grams of Cl_2 per liter
Mean driving force

$$\Delta C_{lm} = \frac{(5.342 - 2.244) - 5.336}{\ln \frac{5.342 - 2.244}{5.336}} = 4.118$$

$$k_L = \frac{2.244 \times 16.86}{4.118 \times 0.22} = 40.9 \quad grams / (hr.)(sq.ft.)(\frac{g}{\ell})$$

$$= 1.45 \quad lbs / (hr.)(sq.ft.)(\frac{lbs}{cu.ft})$$

$$k_L (corrected \; to \; 20) = 1.3598$$

B. Absorption of Ammonia in Phosphate Solution

Run No. 9 in Table 3

Observed data:

    Absorption Surface = 0.22 sq.ft.
    Liquid rate = 29.6 lbs./hr. = 233 lbs./hr.ft.
    Gas rate
        Air rate 0.34 cu.ft./min. of G = 1.53 lbs./hr.
        NH_3 rate 0.0161 cu.ft./min.
    Liquid concentration
        $(NH_4)_2 HPO_4$ = 0.262 M./liter
        NH_3 (reported as grams N per 100 grams of solution)
            Inlet   0.712
            Outlet   0.802
    Mean liquid temperature = 24.5° C.
    Borometric pressure = 737 mm Hg
    Room temperature = 23° C.
    Manometer reading
        Column Inlet = 0.58 mm Hg
        Column Outlet = 0.3 mm Hg

Calculation:

Corrected pressure = 737 mm Hg - 23(0.1197) = 734.2 mm Hg

Inlet gas concentration $Y_1 = \dfrac{0.0161 \times 0.0442}{0.34 \times 0.075}$ = 0.0279 lb. of NH_3 per lb. of air

By material balance

$W (X_2 - X_1) = G (Y_1 - Y_2)$

$X_2 - X_1 = (0.802 - 0.712) \times \dfrac{17}{14} \times \dfrac{1}{1000}$
= 0.001093 lb. of NH_3 per lb. of water

$29.6 \times 0.001093 = 1.53 (0.0279 - Y_2)$

Y_2 = 0.0067 lb. of NH_3 per lb. of air

Partial pressure of NH_3 at inlet

$(734.2 + 0.5) \times \dfrac{0.0161}{0.34 + 0.0161} = 35.22$ mm Hg

Partial pressure of NH_3 at outlet

$(734.2 + 0.33) \times \dfrac{0.0067/17}{1/29 + 0.0067/17} = 8.3$ mm Hg

Mean driving force

(1) Assume there is no back pressure

$P_{1m} = \dfrac{35.22 - 8.3}{\ln \dfrac{35.22}{8.3}} = 16.83$ mm Hg

(II) Assume the solubility of ammonia in phosphate solution is the same as that in water

$\log \dfrac{P'}{c} = 4.699 - 3460/T,^{\circ} R$

where P' = equilibrium partial pressure of ammonia in atm
        C = concentration of ammonia in molality

now T = 24.5° C. or 536.1° R

$\log \dfrac{P'}{c} = 4.699 - \dfrac{3460}{536.1} = 2.244$

P' = 0.01754 C

Interfacial equilibrium pressure

$\quad$ Inlet $= 0.01754 \times 760 \times 0.802 \times \frac{1}{14} \times \frac{1}{100} \times 1000$

$\qquad = 7.64$ mm Hg

$\quad$ Outlet $= 0.01754 \times 760 \times 0.702 \times \frac{1}{14} \quad \frac{1}{100} \times 1000$

$\qquad = 6.78$ mm Hg

$$P_{lm} = \frac{(35.22 - 7.64) - (8.5 - 6.78)}{\ln \frac{35.22 - 7.64}{8.5 - 6.78}} \times \frac{1}{760}$$

$$= 0.01183 \text{ atm}$$

(III) Assume the presence of $(NH_4)_2 HPO_4$ has no effect

$\quad$ P = 0.01754 x C

$\quad$ Interfacial equilibrium pressure

$\qquad$ Inlet $= 0.01754 \times 760 \times (0.802) - 0.712) \times \frac{1}{14} \times$

$\qquad\qquad \frac{1}{100} \times 1000$

$\qquad\qquad = 0.86$ mm Hg

$\quad$ Outlet $= 0$

$$\Delta P_{lm} = \frac{(35.22 - 0.86) - 8.3}{\frac{35.22 - 0.86}{8.3}} = 18.34 \text{ mm Hg} = 0.0241$$

Overall transfer coefficient K_G were calculated by the following equation:

$$K_G = \frac{W(X_2 - X_1)}{A \quad \Delta P_{lm}}$$

For case I

$$K_G = \frac{29.6 \times 0.001093}{0.0245 \times 0.22} = 6.01 \text{ lbs./(hr.)(sq.ft.)(atm)}$$

For case II

$$K_G = \frac{29.6 \times 0.001093}{0.01183 \times 0.22} = 12.44 \text{ lbs./(hr.)(sq.ft.)(atm)}$$

For case III

$$K_G = \frac{29.6 \times 0.00109}{0.0241 \times 0.22} = 6.11 \text{ lbs./(hr.)(sq.ft.)(atm)}$$

Since

$$K_G \quad 24.3 - 0.175t$$

$$t = 24.5^\circ \text{ C.}$$

Take 20° C. as reference, the temperature correction factor

equals

For case   I $K_G = 6.25$ lbs./(hr.)(sq.ft.)(atm)
For case   II $K_G = 12.44$ lbs./(hr.)(sq.ft.)(atm)
For case   III $K_G = 6.35$ lbs./(hr.)(sq.ft.)(atm)

Principal Constants of the Disc Column

Number of discs	32
Diameter of the disc	1.5 cm (0.594 inch)
Thickness of the disc	0.48 cm (0.188 inch)
Diameter of the Pyrex column	1 1/8 inches
Mean perimeter for liquid flow	0.127 ft.
Equivalent diameter for gas flow	0.059 ft.
Absorption surface*	0.22 sq. ft.

*Dry basis.

Nomenclature

A = absorption surface area, sq. ft.

C = contraction of solute gas in liquor

C_e = interfacial equilibrium concentration of solute gas in liquid

ΔC_{lm} = logarithmic driving force expressed in concentration term

G = mass rate of air, lbs./hr.

H = Henry's law constant, defined by $\hbar = HC$

K_G = overall transfer coefficient lbs./(hr.)(sq.ft.)(atm)

k_G = gas film transfer coefficient lbs./(hr.)(sq.ft.)(atm)

K_L = overall transfer coefficient lbs./(hr.)(sq.ft.)$\left(\frac{lbs.}{cu.ft.}\right)$

k_L = liquid film transfer coefficient lbs./(hr.)(sq.ft.)$\left(\frac{lbs.}{cu.ft.}\right)$

p = partial pressure of solute gas in the gas stream

pe = interfacial equilibrium pressure

P_{12} = logarithmic driving force expressed in atm

V = gas velocity ft./sec.

X_1 = concentration of solute in inlet liquid lb./lb.

X_2 = concentration of solute in effluent lb./lb.

Y_1 = inlet gas concentration of lb./lb.

Y_2 = outlet gas concentration lb./lb.

W = liquid rate, lbs./hr.

GAS ABSORPTION WITH CHEMICAL REACTION

by

CHI TIEN

B. S., National Taiwan University, China, 1952

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AN ABSTRACT OF A THESIS

submitted in partial fulfillment of the

requirements for the degree

MASTER OF SCIENCE

Department of Chemical Engineering

KANSAS STATE COLLEGE
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The purpose of this thesis was to study the absorption of ammonia into mono-ammonium orthophosphate solution as well as diammonium orthophosphate solution to further ascertain the gas-film characteristics of the disc column, a specially designed new type of laboratory column, which can be used as a versatile research tool for the study of gas absorption with chemical reactions.

The absorption of chlorine in water was also studied because of the pronounced hydrolyzing effect of dissolved chlorine in water. For this reason, attempts to correlate the chlorine-water system with other similar systems in which the liquid film also is the controlling resistance such as carbon dioxide and water, have resulted in failure.

The absorption of chlorine in water was carried out at liquid rates varying from 100 to 450 lbs./(hr.)(ft.) and gas rates varying from 1.5 to 5 ft./sec. The temperature ranged from 25° C. to 30° C. The liquid film coefficients determined from the experimental data did not vary with the gas rate, and were correlated with the water rate by the following equations:

$$k_L = 0.01163 \, \Gamma^{0.81}$$ ------- by using the total chlorine concentration as driving force.

$$k_{L2} = 0.01173 \, \Gamma^{0.855}$$ ------- by using the molecular chlorine concentration as driving force.

The absorption of ammonia in phosphate solutions was carried out at liquid rates varying from 100 to 350 lbs./(hr.)(ft.) and

gas rates varying from 2 to 6 ft./sec. The concentration of the entering phosphate solution varied from 0.024 to 0.26 moles/liter. Since the solubility of ammonia in phosphate solutions is not available, two assumptions were made: (a) That no back pressure of ammonia over the phosphate solution existed. (b) That the same solubility relationship existed between the ammonia-water system and the ammonia-phosphate system. Values of the overall transfer coefficients, K_G, were calculated on both assumptions.

It was shown that there is no concentration effect for the absorption of ammonia in diammonium phosphate solutions. There is some increase of the numerical value of K_G with an increase of the solution concentration, but not to any great extent. This was expected because this system is one in which the gas film controls. This is also a verification of Hatta's theory for the absorption of gas in a liquid followed by a rapid irreversible reaction.