



## Physico-Chemical Aspects of Solubility of Reactive Gases in Water

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The solubility *versus* partial pressure data for various reactive gases in water has been described. The contribution of chemical reaction in solubility of SO<sub>2</sub>, NH<sub>3</sub>, Cl<sub>2</sub> and ClO<sub>2</sub> in water has been highlighted. Chlorine dioxide showed only Henry type solubility and existed in the gaseous form even in the aqueous phase. On the contrary, NH<sub>3</sub>, Cl<sub>2</sub> and SO<sub>2</sub> showed appreciable contribution of solubility due to chemical reaction and existed in the ionized form in the aqueous phase. An empirical relation having three adjustable parameters (a, b, and h) have been derived to discuss the physical and chemical terms of the total solubility in water. The Henry's coefficient (H) of SO<sub>2</sub>, NH<sub>3</sub>, Cl<sub>2</sub> and ClO<sub>2</sub> has been evaluated at various temperatures and the temperature dependence of Henry's coefficient has been discussed using a four parameter equation. The enthalpy of dissolution ( $\Delta H$ ) for various gases is evaluated and compared with the literature values.

**Keywords:** Solubility, Gas, Henry's coefficient, Empirical relation, Enthalpy of dissolution.

### INTRODUCTION

Gas absorption is one of the main techniques used in pollution control devices for the removal of gaseous pollutants from the industrial gas streams. In absorption processes, the gaseous pollutant is brought into close contact with the liquid to facilitate diffusion across the gas-liquid interface. Once the pollutant enters the liquid phase, it can simply dissolve, or it can react with other chemicals present in the liquid phase. In case of simple dissolution, there may be a definite limit to mass transfer. Once the pollutant in the liquid phase has reached its solubility limit, there is no net transfer of pollutant across the gas-liquid interface. Henry's law can be used to determine the solubility limit of absorption. If the liquid phase contains some chemical with which the dissolved gaseous pollutant can react, then equilibrium may never be established (a condition that is usually beneficial for pollutant absorption).

The concentration of CO<sub>2</sub>, SO<sub>2</sub>, NO<sub>x</sub> and other pollutants in the atmosphere has increased steadily with the increasing energy demands by modern civilization. Thermal power plants in India are emitting huge amount of SO<sub>2</sub> due to combustion of high ash content coal. The numbers of coal-fired power plants in India are increasing at a fast pace and are creating a serious problem of SO<sub>2</sub> emission. Due to stringent regulations in the recent years, the abatement of noxious gases from the industrial gases has become the burgeoning need of the hour. The most attractive method for removal of poisonous gases is

the absorption method. Therefore, the solubility data for various gases are highly essential not only in understanding mass transfer phenomena but also in improving the design and performance of the absorber.

A number of experimental studies have been reported in the literature for rate of SO<sub>2</sub> absorption into water<sup>1-7</sup>. Research on SO<sub>2</sub> absorption into aqueous solutions has been focused on chemical reactions taking place in the liquid phase as liquid phase resistance frequently dominates the absorption process<sup>8-12</sup>. One of the important factors of SO<sub>2</sub> absorption is that it dissociates instantaneously and reversibly to form H<sub>3</sub>O<sup>+</sup> and HSO<sub>3</sub><sup>-</sup> in an aqueous phase and this dissociation reaction makes the absorption rate much faster than physical absorption rate. Several authors<sup>1,8,11</sup> reported SO<sub>2</sub> absorption into water as well as in aqueous solutions of H<sub>2</sub>SO<sub>4</sub> and NaHSO<sub>4</sub> and found that the presence of H<sub>2</sub>SO<sub>4</sub> or NaHSO<sub>4</sub> prevented SO<sub>2</sub> dissociation, resulting in a lower absorption rate than that in pure water. Ammonia is yet another reactive gas which has a number of industrial applications. Numerous studies on solubility of ammonia in water and aqueous solutions are available in literature<sup>13-16</sup>.

Both chlorine and chlorine dioxide are very effective disinfectant. Accurate solubility data of chlorine and chlorine dioxide has important applications in water treatment in the design of chlorine supply systems and of chlorine dioxide generators. Aqueous solution of chlorine dioxide has recently been used in removal of SO<sub>2</sub> and NO<sub>x</sub> from the flue gas<sup>17-19</sup>. In

water treatment applications, chlorine often used in gaseous form and must be transferred from the gas phase into aqueous solution. Accurate knowledge of the equilibrium concentration of molecular chlorine at the air-water interface is thus essential to calculate the mass-transfer driving force. Several fundamental studies were carried out to establish the solubility of chlorine gas in water<sup>20</sup> and in many aqueous solutions<sup>21-24</sup>.

Though abundant studies have been carried out on solubility of reactive gases in water but no tangible model is available to correlate the data. The solubility of non-reactive gases in water can be explained by Henry's law, where the solubility vs. partial pressure plot should yield a linear relation. This law holds good only for very dilute solutions of non-reactive gases and practically it deviates from linearity for most of the reactive gases. The objective of the study is to derive a mathematical relation to describe the solubility of real gases over a wide range of composition and temperatures. It will not only help in the analysis of industrial mass transfer processes but also in the reactor design and modeling of absorption processes. To derive this relation, we selected various gases like SO<sub>2</sub>, NH<sub>3</sub>, ClO<sub>2</sub> and Cl<sub>2</sub> and their solubility data has been taken from the literature<sup>25</sup>.

### Henry law: validity and limitations

When a gas comes in contact with a liquid, it tends to dissolve in the liquid. At equilibrium, the concentration of gas in the liquid is determined by its partial pressure in the gas and its solubility in the liquid. This relationship is described as Henry's law, which basically states that the amount of a slightly soluble gas dissolved in a liquid is proportional to its partial pressure<sup>26,27</sup>. This law is a function of the temperature, type and nature of liquid. In industry, Henry's law thus can be used to determine the condition and type of liquid that will maximize the removal efficiency of the noxious gases.

For dilute solutions of non-reactive gas, the most common form of Henry's law is:

$$x = hp \quad (1)$$

where 'x' is the solubility of the gas (in terms of mole fraction), 'h' is the solubility coefficients (Pa<sup>-1</sup>) and 'p' is the partial pressure of the gas (Pa). The solubility coefficient is a measure of how easily the gas dissolves in the liquid. The solubility vs. equilibrium partial pressure curve is a straight line and the reciprocal of the slope of the straight line is taken as the Henry law's constant (H). Henry's law constant provides a general indication of the differences in the solubility of gases.

Henry's law can be used to predict solubility only when the equilibrium line is straight, which is the case when solution is very dilute and gas is non-reactive. Henry's law does not apply at high concentrations for most of the gases and at low-to-moderate concentrations for the gases which react or dissociate upon entering the liquid phase. Due to chemical reaction or dissociation of reactive gases in water, the solubility relationship is curved. Therefore, the solubility of a gas, which reacts or dissociates in water must be determined empirically.

The solubility of a gaseous pollutant in a particular liquid at a given temperature limits the quantity of the pollutant that can be absorbed in a given amount of the liquid. As stated earlier, once the gaseous pollutant has reached its solubility

limit, mass transfer backward from the liquid phase to the gas phase matches the rate of mass transfer from the gas phase to the liquid phase. However, this equilibrium limit can be removed by converting the dissolved gas into the chemical form that cannot diffuse out of the liquid. In general, alkaline/acidic absorbents can be added to buffer and neutralize the H<sup>+</sup>/OH<sup>-</sup> ions resulting from acid/alkaline gases in the scrubbing solution. Accordingly, the gas cannot diffuse out of solution. It is possible to continue dissolving the gas into the liquid phase as long as there are enough reactants in the absorber. This is the common situation in various types of air pollution control systems dealing with the removal of acid gases from the industrial gas streams.

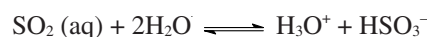
### Equilibrium processes for various gas-water systems

When a gas is dissolved in water, there exists equilibrium at the gas-liquid interface between unreacted gas in liquid phase and that in gaseous phase (described by Henry's law). A part of dissolved gas may react with water and subsequently dissociate into ionic species, thereby changing the pH and ionic strength of the medium. The change in pH and ionic strength does not affect the Henry's law constant but certainly affects the equilibria in liquid phase. For example, the wet scrubbing method for flue gas desulphurization use alkaline solution for abatement of SO<sub>2</sub>, which shifts the equilibrium processes in liquid phase to forward direction and facilitates the SO<sub>2</sub> absorption. However, reverse will be true for the alkaline gas like NH<sub>3</sub>. Therefore, for precise analysis of the performance of the absorber, it is necessary to investigate the effect of pH, ionic strength and nature of the absorbent on the chemical contribution to overall sorption of gas in water.

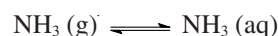
The equilibria in various gas-water systems can be described by the following equations:



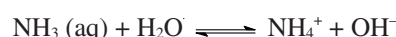
At gas-liquid interface (dissolution) (2)



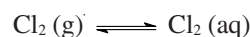
In liquid phase (ionization) (3)



At gas-liquid interface(dissolution) (4)



In liquid phase (ionization) (5)



At gas-liquid interface (dissolution) (6)



In liquid phase (ionization) (7)

The corresponding equilibrium constants in liquid phase can be written as follows:

$$K_3 = \frac{[\text{HSO}_3^-][\text{H}_3\text{O}^+]}{[\text{SO}_2(\text{aq})]} \quad (8)$$

$$K_5 = \frac{[\text{NH}_4^+][\text{OH}^-]}{[\text{NH}_3(\text{aq})]} \quad (9)$$

$$K_7 = \frac{[\text{HOCl}][\text{H}_3\text{O}^+][\text{Cl}^-]}{[\text{Cl}_2(\text{aq})]} \quad (10)$$

The ionization constants for above equations have been investigated by several workers. The hydrolysis of sulfur dioxide is relatively rapid compared to that of ammonia and chlorine. Chlorine dioxide does not hydrolyze. The values of ionization constant for eqns. 3, 5 and 7 were found to be 0.0142 mol/L<sup>28</sup>, 1.6 × 10<sup>-5</sup> mol/L<sup>29</sup> and 4.5 × 10<sup>-4</sup> mol<sup>2</sup>/L<sup>24</sup> respectively. The enthalpy of dissolution ( $\Delta H$ ) of for eqns. 2, 4 and 6 can be calculated by plotting  $\ln H$  vs.  $1/T$ . As the equilibrium constant ( $K$ ) is a function of temperature, so the overall enthalpy of reaction in liquid phase for eqn. 3, 5 and 7 can be calculated by plotting  $\ln K$  vs.  $1/T$ .

### Mathematical model for the solubility of reactive gases in liquid

It is known that some of the gases may react with water, partially ionize and change the pH, ionic strength and conductance of water. Therefore, it is postulated that the absorption of a gas in water is sum of two terms *i.e.* the physical absorption and chemical absorption. The physical absorption is exclusively governed by the Henry's law. However chemical absorption yields a saturation type curve as a result of the chemical reaction of gas with water. The contribution of both terms is dependent on the nature of the gas.

It is postulated that three adjustable parameters *viz.* a, b and h can describe the solubility of a gas where the parameter 'h' is termed as the solubility coefficient. However the term 'a' and 'b' describe the solubility due to chemical reaction between gas and water. It has been observed that solubility vs. partial pressure plot for several gases deviates significantly from linearity. There is slight increase at low partial pressure and then a straight line with a constant slope is obtained. This slope is considered equal to 'h', the reciprocal of which provides the value of Henry's law constant (H).

The overall solubility thus can be expressed in terms of following two equations:

$$x_p = hp \quad \text{Physical absorption} \quad (11)$$

$$x_c = \frac{ap}{b+p} \quad \text{Chemical absorption} \quad (12)$$

where  $x_p$  and  $x_c$  represents physical and chemical contribution to the overall solubility respectively (in terms of mole fraction), p is the partial pressure of the gas (Pa), h is the solubility coefficient (Pa<sup>-1</sup>), a and b are adjustable temperature dependent constants.

Combining eqns. 11 and 12, the total solubility can be expressed as:

$$x_T = hp + \frac{ap}{b+p} \quad (13)$$

This empirical relation is based upon the dual absorption phenomenon. First term on right hand side of eqn. 13 gives contribution of dissolution of gas in water in the unreacted form and second term deals with the solubility as a result of chemical interaction of gas with water. When the solubility vs. partial pressure data is fitted in the eqn. 13, it yielded the three unknown parameters *i.e.* a, b and h. These parameters are listed in Table-1 at various temperatures along with standard deviation in solubility ( $\sigma_x$ ). The term 'a' corresponds to the

TABLE-1  
ADJUSTABLE PARAMETERS OF EQN. 13 FOR VARIOUS GAS-WATER SYSTEMS AT DIFFERENT TEMPERATURES ALONG WITH STANDARD DEVIATION IN SOLUBILITY ( $\sigma_x$ )

System	Temp. (K)	a	b × 10 <sup>-3</sup> (Pa)	h × 10 <sup>7</sup> (Pa <sup>-1</sup> )	$\sigma_x \times 10^3$
NH <sub>3</sub> -H <sub>2</sub> O	283.15	0.3762	18.299	9.9661	2.1
	293.15	0.3691	29.157	7.1916	0.7
	303.15	0.3794	48.052	4.7006	0.4
	313.15	0.4004	77.586	2.6493	1.5
SO <sub>2</sub> -H <sub>2</sub> O	283.15	0.0029	10.232	3.5832	0.13
	293.15	0.0017	7.7658	2.6142	0.08
	303.15	0.0014	8.6912	1.9093	0.07
	313.15	0.0009	7.7379	1.4363	0.04
Cl <sub>2</sub> -H <sub>2</sub> O	283.15	0.0003	2.0755	0.21644	0.02
	293.15	0.0003	2.4598	0.15488	0.02
	303.15	0.0003	2.4633	0.11716	0.02
	313.15	0.0003	2.4640	0.09157	0.02
ClO <sub>2</sub> -H <sub>2</sub> O	283.15	0.0000	0.0000	3.3410	0.02
	293.15	0.0000	0.0000	2.3106	0.01
	303.15	0.0000	0.0000	1.6551	0.01
	313.15	0.0000	0.0000	1.1868	0.01

maximum solubility of a gas as a result of chemical reaction of gas with water. The magnitude of parameter a, b and h for various gas-water systems varied as:  $a_{\text{NH}_3} > a_{\text{SO}_2} > a_{\text{Cl}_2} > b_{\text{NH}_3} > b_{\text{SO}_2} > b_{\text{Cl}_2}$  and  $h_{\text{NH}_3} > h_{\text{SO}_2} > h_{\text{ClO}_2} > h_{\text{Cl}_2}$ , respectively. The solubility vs. partial pressure data of SO<sub>2</sub>, NH<sub>3</sub>, ClO<sub>2</sub> and Cl<sub>2</sub> gas is shown in Figs. 1-4 at four different temperatures.

### Temperature dependence of the Henry's law constant (H)

The solubility of gases decreases with the increasing temperature. When the temperature of a system changes, the Henry's law constant also changes. This is why, it is preferred to name it Henry's coefficient instead of Henry's law constant. Henry's coefficient, H, exhibits a van't Hoff equation-type dependency on temperature<sup>11</sup>. The differential form of the relation to assess the temperature dependence of Henry coefficient is expressed as<sup>30</sup>:

$$\frac{d \ln H}{d(1/T)} = -\frac{\Delta H}{R} \quad (14)$$

The above relation can be expressed in the linear form as follows:

$$\ln H = -\frac{\Delta H}{RT} + \ln Z \quad (15)$$

where Z is a constant (Pa),  $\Delta H$  is the enthalpy of the dissolution of gas in liquid (J mol<sup>-1</sup>), R is the gas constant (J K<sup>-1</sup> mol<sup>-1</sup>). The Henry's coefficient can be calculated at any other temperature, knowing its value at one temperature by rearranging the eqn. 15 in the following form:

$$\ln \frac{H_2}{H_1} = \frac{\Delta H}{R} \left( \frac{1}{T_1} - \frac{1}{T_2} \right) \quad (16)$$

or

$$H_2 = H_1 \exp \left\{ \frac{\Delta H}{R} \left( \frac{1}{T_1} - \frac{1}{T_2} \right) \right\} \quad (17)$$

where H<sub>1</sub> and H<sub>2</sub> are the Henry's coefficients at temperature T<sub>1</sub> and T<sub>2</sub> respectively. From eqn. 15, the plot of  $\ln H$  vs.  $1/T$

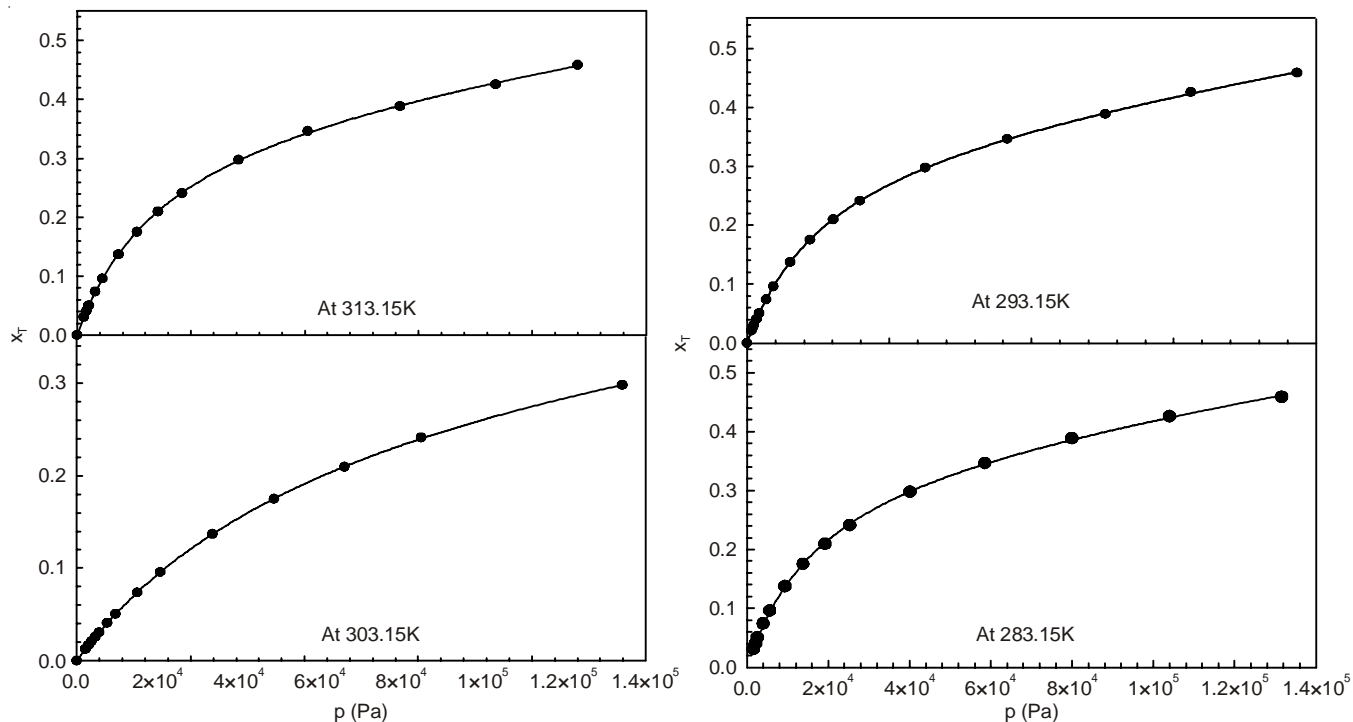


Fig. 1. Solubility vs. partial pressure plots for ammonia-water system at various temperatures. The symbols and the line represent the measured values and curve fitting of eqn. 13, respectively

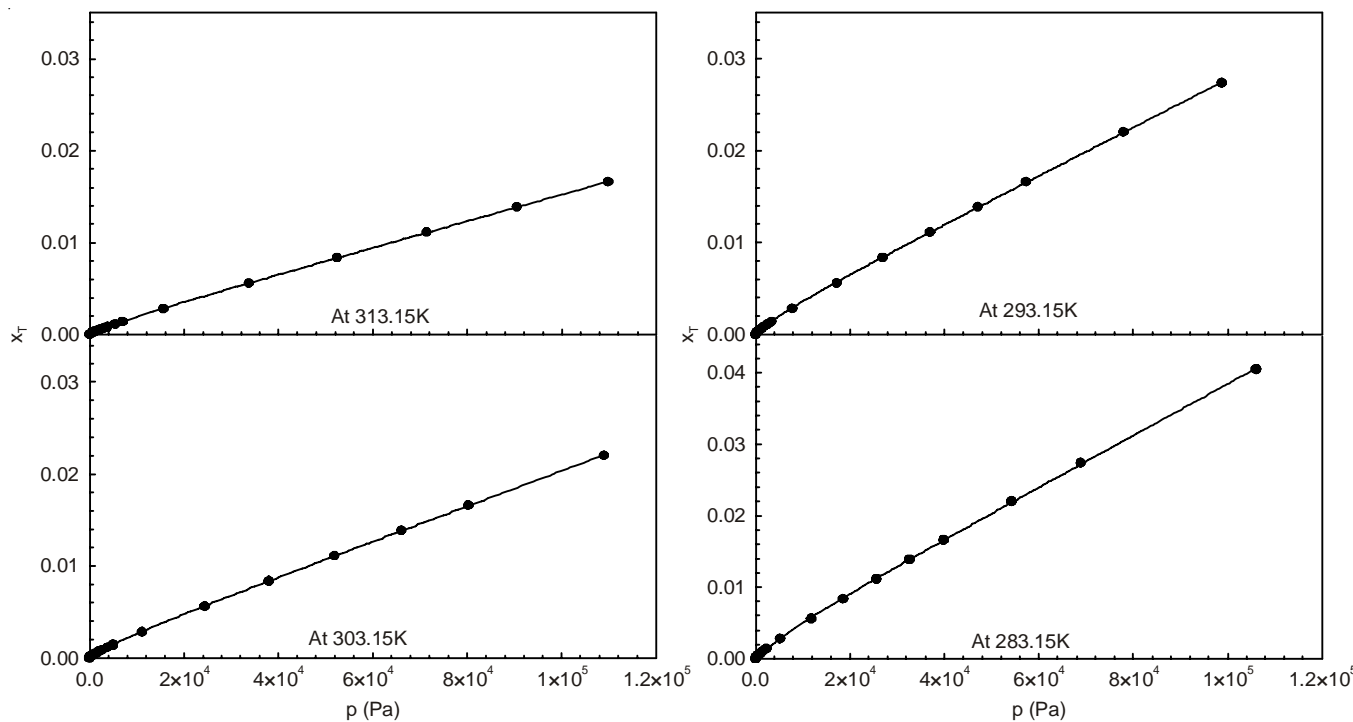


Fig. 2. Solubility vs. partial pressure plots for sulfur dioxide-water system at various temperatures. The symbols and line represent the measured values and curve fitting of eqn. 13, respectively

should be linear. It is indeed being observed in almost all the cases. The equations for straight lines [with square of regression coefficient ( $R^2$ )] for gas-water various systems are obtained as follows:

$$\ln H = -\frac{3884.02}{T} + 27.465$$

$$R^2 = 0.9760 \text{ (ammonia-water)} \quad (18)$$

$$\ln H = -\frac{2710.37}{T} + 24.410$$

$$R^2 = 0.9998 \text{ (sulfur dioxide-water)} \quad (19)$$

$$\ln H = -\frac{2539.20}{T} + 26.629$$

$$R^2 = 0.9984 \text{ (chlorine-water)} \quad (20)$$

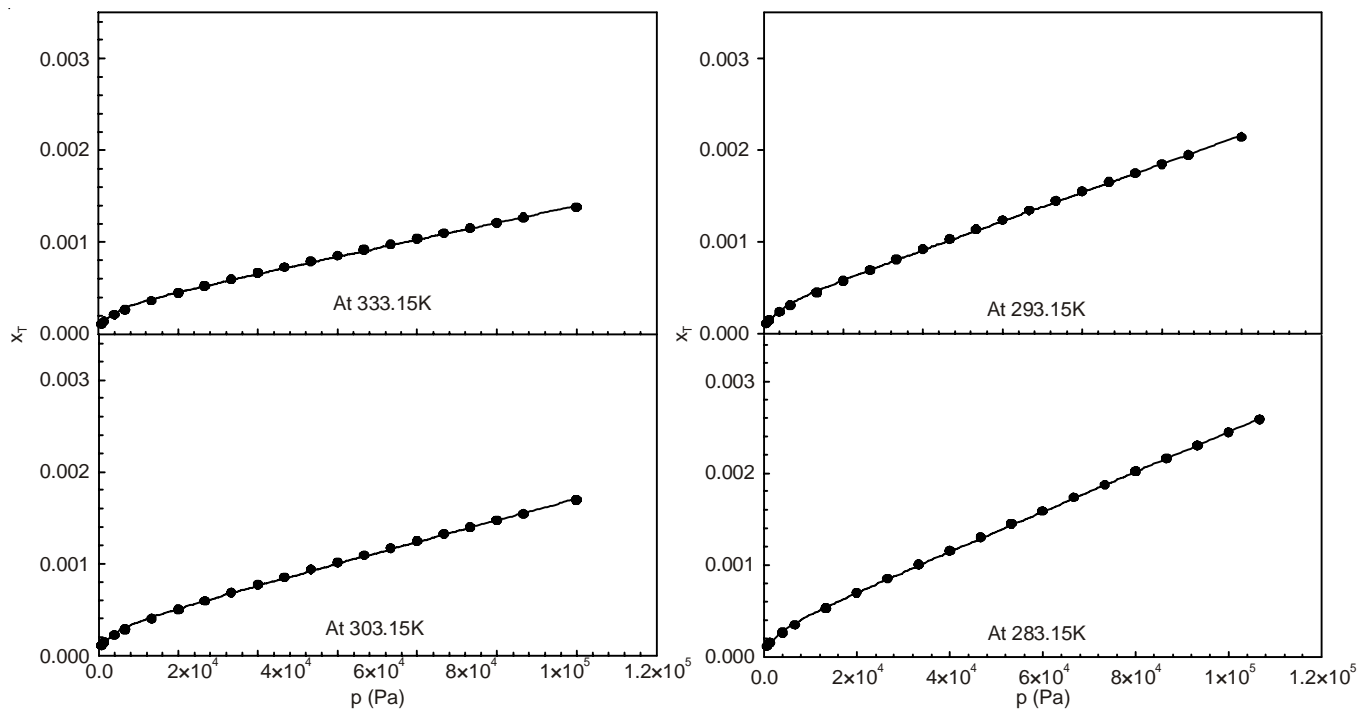


Fig. 3. Solubility vs. partial pressure plots for chlorine-water system at various temperatures. The symbols and line represent the measured values and curve fitting of eqn. 13, respectively

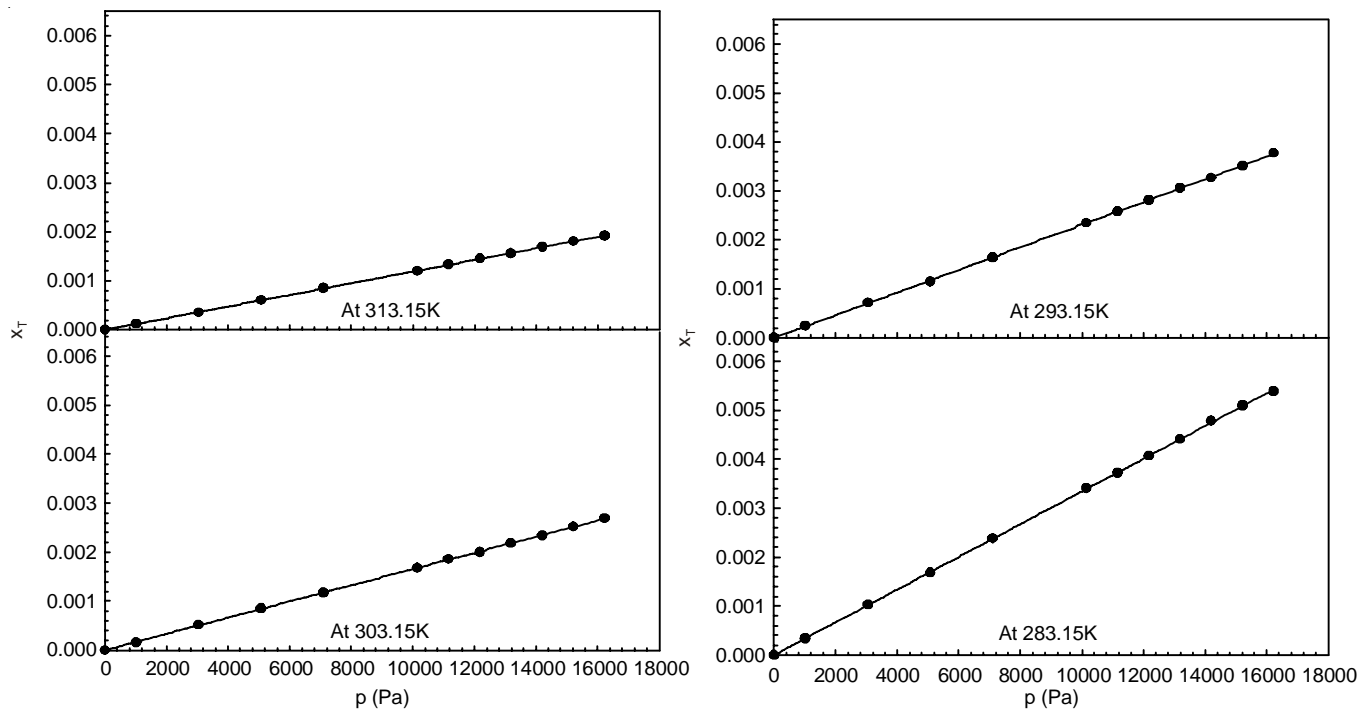


Fig. 4. Solubility vs. partial pressure plots for chlorine dioxide-water system at various temperatures. The symbols and line represent the measured values and curve fitting of eqn. 13, respectively

$$\ln H = -\frac{3049.13}{T} + 25.680$$

$$R^2 = 0.9999 \text{ (chlorine dioxide-water)} \quad (21)$$

The  $\ln H$  vs.  $1/T$  plots for various systems are shown in Fig. 5. The pre-exponential factor ( $Z$ ) and enthalpy of the dissolution ( $\Delta H$ ) of various gas-water systems were calculated from the intercepts and the slopes of the respective straight

lines and are presented in Table-2 along with Henry's coefficients at 25 °C.

Zhang *et al.*<sup>11</sup> measured the solubility of  $\text{SO}_2$  in the concentrated aqueous sulfuric acid. The presence of sulfuric acid increased the  $\text{H}^+$  ion concentration and thus suppressed the hydrolysis of dissolved  $\text{SO}_2$ . Thus, solubility of  $\text{SO}_2$  in concentrated aqueous sulfuric acid is totally governed by Henry's law. The enthalpy of the dissolution ( $\Delta H$ ) of  $\text{SO}_2$  in concentrated



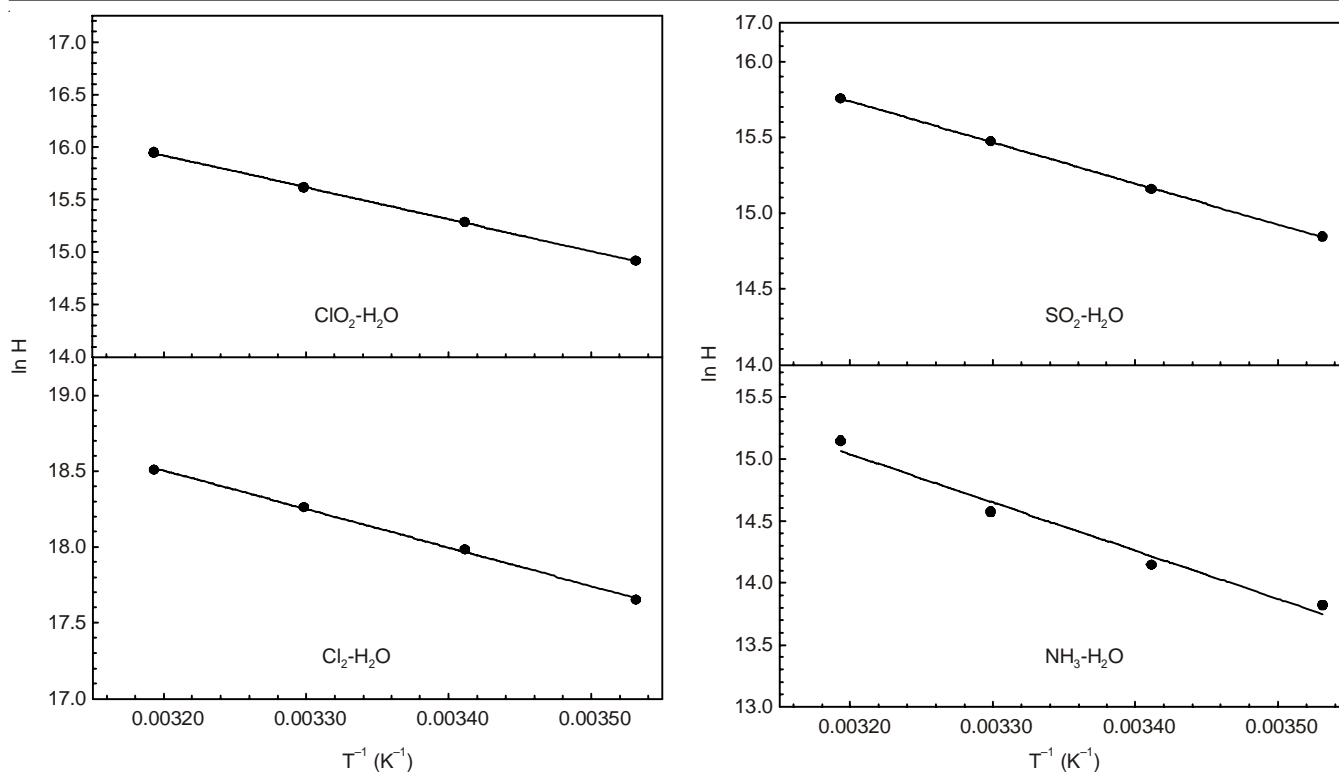


Fig. 5.  $\ln H$  vs.  $1/T$  plots for various gas-water systems. The symbols and line represent the measured values and curve fitting of eqn. 15, respectively

aqueous sulfuric acid was found  $-20.95 \text{ kJ mol}^{-1}$ . On the contrary, enthalpy of the dissolution of  $\text{SO}_2$  in water is little higher *i.e.*  $-22.53 \text{ kJ mol}^{-1}$  (Table-2). The difference is certainly the enthalpy of hydrolysis of dissolved  $\text{SO}_2$  in water.

TABLE-2  
PARAMETERS OF EQN. 15 FOR VARIOUS GAS-WATER SYSTEMS ALONG WITH HENRY'S COEFFICIENT AND ENTHALPY OF THE DISSOLUTION ( $\Delta H$ ) OF GAS IN LIQUID AT 298.15 K

System	$Z \times 10^{-10}$ (Pa)	$H \times 10^6$ (Pa)		$\Delta H$ ( $\text{kJ mol}^{-1}$ )	
		Calcd.	Lit. <sup>*</sup>	Calcd.	Lit. <sup>*</sup>
$\text{NH}_3\text{-H}_2\text{O}$	84.7028	1.863	0.562 <sup>31</sup> 0.1005 <sup>14</sup>	-32.29	-28.29 <sup>13</sup> -34.02 <sup>14</sup> -30.50 <sup>33</sup>
$\text{SO}_2\text{-H}_2\text{O}$	3.9914	4.499	4.082 <sup>28</sup> 4.625 <sup>6</sup> 3.208 <sup>4</sup>	-22.53	-25.19 <sup>28</sup> -25.70 <sup>6</sup> -23.80 <sup>4</sup>
$\text{Cl}_2\text{-H}_2\text{O}$	36.7137	73.476	61.797 <sup>31</sup> 65.390 <sup>32</sup>	-21.11	-19.75 <sup>23</sup>
$\text{ClO}_2\text{-H}_2\text{O}$	14.2129	5.143	5.624 <sup>31,33</sup>	-25.35	-27.83 <sup>31</sup>

<sup>\*</sup>The units of  $H$  and  $\Delta H$  reported in literature have been changed into Pa and  $\text{kJ mol}^{-1}$  respectively.

When solubility data of a reactive gas is measured at very high partial pressure and in the wide range of temperature, then a slight non-linearity in  $\ln H$  vs.  $1/T$  plot is detected. It reflects some fallacy in the relation between  $\ln H$  and  $T$  as expressed in eqn. 15. Henry's law assumes that enthalpy of solution ( $\Delta H$ ) is a constant, independent of temperature. In fact,  $\Delta H$  varies with temperature. Therefore non-linearity of solubility vs. partial pressure curve may be attributed to the dependence of enthalpy of solution on the temperature. To

overcome this inconsistency, another modified expression thus has been proposed to relate the Henry's coefficient with temperature *i.e.*<sup>7</sup>:

$$\ln H = A + B/T + CT + D \ln T \quad (22)$$

where  $A$ ,  $B$ ,  $C$  and  $D$  are the adjustable parameters. These parameters were calculated by fitting the data in the eqn. 22 and have been recorded in Table-3 along with relative deviation in Henry's coefficient. The relative deviation is calculated as follows:

$$\text{Relative deviation (\%)} = \left( \frac{H_{(\text{exp})} - H_{(\text{calc})}}{H_{(\text{exp})}} \right) \times 100 \quad (23)$$

The  $\ln H$  vs.  $T$  plots for various gas-water systems are shown in Fig. 6. The Henry's coefficient calculated from eqn. 22 agreed well with experimental values with relative deviation of about 1 % in all cases except  $\text{NH}_3$ .

TABLE-3  
ADJUSTABLE PARAMETERS OF EQN. 22 FOR VARIOUS GAS-WATER SYSTEMS ALONG WITH RELATIVE DEVIATION (r.d.) IN HENRY'S COEFFICIENT

System	A	B	C	D	r.d. in $H$ (%)
$\text{NH}_3\text{-H}_2\text{O}$	-43.998	14611.777	0.247	-11.264	8.1
$\text{SO}_2\text{-H}_2\text{O}$	19.725	-2006.113	0.008	-0.011	0.2
$\text{Cl}_2\text{-H}_2\text{O}$	35.223	-5486.238	-0.041	2.390	1.1
$\text{ClO}_2\text{-H}_2\text{O}$	21.529	-2413.943	0.007	-0.026	1.0

## Conclusion

The solubility of  $\text{SO}_2$ ,  $\text{NH}_3$ ,  $\text{Cl}_2$  and  $\text{ClO}_2$  gases has been evaluated as a function of partial pressure and it was found

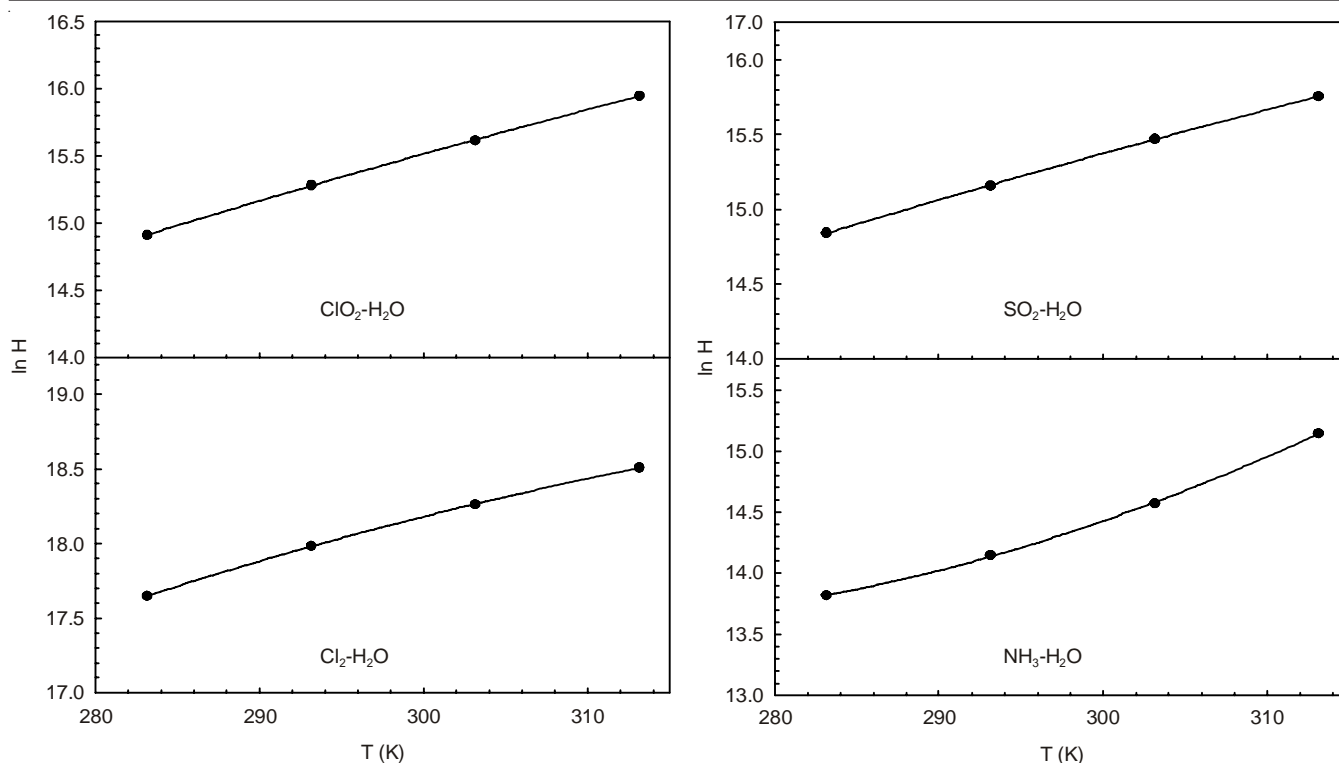


Fig. 6.  $\ln H$  vs. temperature plots for various gas-water systems. The symbols and line represent the measured values and curve fitting of eqn. 22, respectively

that chemical solubility has appreciable contribution in case of gases, which react with water to form ionic species. An empirical relation having three adjustable parameters (a, b and h) have been suggested to discuss the physical and chemical terms of the total solubility in water. This correlation would prove a more reliable tool in predicting the accurate solubility of reactive gases in water. The Henry's coefficient (H) of reactive gases has been evaluated at various temperatures and temperature dependence of Henry coefficient has been discussed in terms of a new mathematical equation. This relation makes the estimation and extrapolation of Henry's coefficient and solubility more accurately at higher temperatures and pressures.

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