Evaluation of Bunsen Reaction Kinetics in Iodine-sulfur Thermochemical Process for Hydrogen Production

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Abstract. *There are many thermochemical processes; one of the potential processes is iodine-sulfur (IS) thermochemical process which involves three reaction steps. Bunsen reaction is the first reaction. Kinetic rate data of Bunsen reaction is essential for design and scale up of I-S process. Experiments have been carried out in stirred glass reactor to determine kinetic rate data for Bunsen reaction. The aim of the work is to evaluate intrinsic kinetic rate constant of Bunsen reaction. Mass transfer coefficient is evaluated experimentally for water-sulfur dioxide system. Stirring speed is varied up to 360 rpm to obtain a regime in which mass transfer is independent of absorption rate. Mass transfer coefficients are evaluated in the range of 8.7×10^{-6} m/s to 1.5×10^{-5} m/s at different stirring speeds. Mass transfer coefficients obtained experimentally. These coefficients are compared with published literature and found to be in good agreement. Bunsen reaction has been carried out in a stirred glass reactor and obtained reaction kinetic rate constant as 1.8 second^{-1}. Hatta number and Infinite enhancement factors are calculated from the experimentally obtained kinetic rate constant and mass transfer coefficient and found to be 3.6 and 18 respectively. From the calculated values of Hatta number and Infinite enhancement factors, the regime of chemical reaction is found to be in the fast pseudo first order reaction regime.*

Keywords: Hydrogen production, iodine-sulfur process, Bunsen reaction, stirred glass reactor, reaction kinetics

Introduction

The need for alternative, environmentally friendly energy carriers is becoming increasingly urgent as awareness of the impacts of climate change increases. Hydrogen is one of the most attractive fuels to meet the need for clean energy, especially for the transportation sector. However, most hydrogen is currently produced from fossil fuels, which ultimately contribute to greenhouse gas emissions.

The water splitting process using renewable primary energy sources offers a sustainable solution for large-scale hydrogen production. One promising method is the thermochemical cycle, in which water is split into hydrogen and oxygen through a series of chemical reactions. The energy required for this process can be obtained from clean energy sources such as nuclear energy or solar energy, making it an environmentally friendly option.

The iodine-sulfur (I-S) thermochemical cycle is one of the most potential processes for hydrogen production due to its high thermodynamic efficiency. This process consists of three main stages,
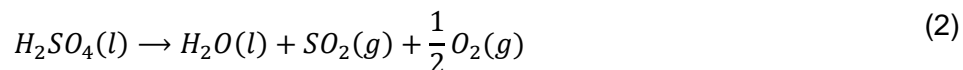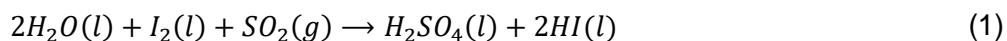
with the Bunsen reaction as the first step that determines the overall efficiency of the process. Evaluation of kinetic data and mass transfer coefficients of the Bunsen reaction is crucial for the design and scale-up of the I-S process.

In this study, experiments were conducted to evaluate the intrinsic kinetic rate constant and mass transfer coefficient of the Bunsen reaction. This study aims to provide an in-depth understanding of the kinetic and mass transfer characteristics of the Bunsen reaction, and to ensure that the results are in accordance with the published literature. With this information, the development of the I-S process for large-scale hydrogen production is expected to be carried out more effectively. The aim of the work is to evaluate intrinsic kinetic rate constant of Bunsen reaction.

Theoretical Background

The energy required can be provided by clean enery sources such as nuclear energy or by solar energy. Studies are carried out for various hydrogen production methods comparing the economic, environmental and social impacts [1]. There are many thermochemical processes, one of the highly researched process is iodine-sulfur thermochemical process [2] consisting of three chemical reaction steps which is shown below :

$$2H_2O(l) + I_2(l) + SO_2(g) \longrightarrow H_2SO_4(l) + 2HI(l) \tag{1}$$

$$H_2SO_4(l) \longrightarrow H_2O(l) + SO_2(g) + \frac{1}{2}O_2(g) \tag{2}$$

$$2HI(l) \longrightarrow I_2(g) + H_2(g) \tag{3}$$

Water, sulfur dioxide and iodine react to form hydriodic and sulfuric acids in reaction (1) which is called as Bunsen reaction. Excess water and iodine makes the reaction (1) feasible and makes the two products into immiscible phases. The produced HI_x (mixture of HI, I_2 and H_2O) and H_2SO_4 phases are separated. Subsequently sent to the concentration units [3]. The H_2SO_4 phase is concentrated and then catalytically decomposed into oxygen and sulfur dioxide [4], [5], as mentioned in reaction (2). Experimental and modelling studies on sulfur trioxide decomposition using solar concentrator are carried out [6], [7]. The sulfur dioxide and water are recycled to the Bunsen section after removing heat. The HIx phase is sent to the hydrogen iodide (HI) decomposition section, where the HI is separated from the mixture by distillation, and catalytically decomposed into hydrogen and iodine [8]–[10], as shown in reaction (3). Membranes are also used for HI decomposition to separate hydrogen [11].Studies on closed loop I-S process is carried for feasibility of operation [12].

Bunsen reaction is a gas-liquid reaction system in which mass transfer of sulfur dioxide in the liquid reactants is improved by the chemical reaction [13]. Thus the Bunsen reaction progress depends on chemical reaction resistances as well as mass transfer resistances. Kinetics date for a gas-liquid reaction is required for design and scale up of reactor. Bunsen reaction studies have been carried out to find the best operation conditions focusing on product yield and phase separation [13]–[16]. Studies of side reactions in Bunsen product mixture is carried out at various concentrations and temperatures [17]. Cross contaminations in Bunsen product phases are also studied [18]. Reaction kinetic study to evaluate kinetic parameters of Bunsen reaction has been studied [16], [19]–[23]. Kinetic studies in a semi batch reactor is studied by varying feed flow rates and reactant concentrations, but effects of mass transfer are not described and reaction is carried out at ambient pressure conditions [19].

Experiments are conducted in semi batch reactor to determine the overall reaction rate of Bunsen reaction [16]. Reaction kinetics of the electrochemical Bunsen reaction is studied but applicability of data for conventional reactor design is limited [20]. Using solvents, reaction kinetics has been studied however reaction mechanism will be entirely different without solvent [21]. Bunsen reaction kinetics studies have carried out using stirred cell reactor. By measuring pressure profiles in the experiment, reaction parameters are obtained [22], [24].

Absorption rates of SO_2 in HI_x solution (gas-liquid reaction) should be in a fast pseudo-first-order reaction regime to correlate kinetic absorption rates with experimentally determined absorption rates. In order to calculate intrinsic reaction kinetic parameters from concentration measurements, it is required to drive the reaction in fast reaction regime. The present research work has targeted to achieve this regime of gas-liquid reaction. Zhu Q. et al. has reported reaction rate constant parameters (frequency factor and activation energy) [19]. From the rate constant equation in the study, the rate constant is obtained as 0.13 second^{-1} at 30 ºC. The rate constant value is lower than that is obtained in this research work. This is probably because of less intense agitation provided during experimentation by Zhu Q. et al. The stirring rate is at low rpm in the study (for the same volume as this research work). The stirring rate may not be adequate to overcome the mass transfer resistances in this gas-liquid Bunsen reaction. Further the study has considered higher water content in the initial reaction solution.

Zhang Y. et al. has qualitatively studied kinetics of Bunsen reaction. Kinetic rate constant is not calculated out in the study [25] Zhou. et al. initially has studied reaction kinetics without iodine content. Followed by Bunsen reaction in the subsequent study [22], [26]. The rate constant obtained in the study is 1.9 second^{-1}, this is close to the value obtained in this research (1.8 second^{-1}). However, the study did not calculate the Hatta number and infinite Enhancement factor. These two factors are required to identify the regime of gas-liquid reaction.

Ying Z. et al. has studied the kinetics of Electro-chemical based Bunsen reaction [20]. The study has obtained rate equation parameters for anode and cathode reactions and the kinetics data obtained cannot be directly compared to conventional gas-liquid reaction. Ying Z. et al. has studied the Bunsen reaction by considering recycle HI stream as one of the components in the reaction mixture [23]. The rate constant obtained in the study is 0.32 second^{-1}. The water concentration is more in the study. The water concentration has the role in dilution of the solvent in which reaction occurs. The dilution causes decrease in effective collusion between the active species. Probably this could be reason for lesser value of rate constant, which is obtained from the study. Further, the study did not identify the regime in which the gas-liquid reaction is operating. However, absorption rates of SO_2 in HI_x solution should be in fast pseudo first order regime in order to correlate kineticabsorption rates with experimentally determined absorption rates [27]. In order to ensure the reaction regime to be in fast pseudo first order several experiments have been conducted in this study and obtained the kinetic rate constant. Several different types of equipment are developed to measure the rate of absorption in gasliquid reactions. These include wetted wall column, laminar jet reactor and a stirred reactor [28].

There are various experimental techniques to evaluate kinetic rate data. By having prior knowledge of interfacial area and mass transfer coefficient, reaction kinetics rates can be studied from the experimental values. The stirred glass reactor is used to obtain reaction kinetics data owing to its ease of operation and no liquid phase analysis is required. Hence it is adopted in this study. This reactor can be operated under isothermal conditions. Liquid phase and gas phase are

operated in batch mode in a stirred glass reactor. During the experiments gas absorption process is monitored by recording the pressure decrease in the reactor with time. Stirred glass reactor is a cylindrical shape with a stirring device. The composition and temperature of the reaction is homogenous in the reactor. The stirrer is the turbine type with four blades. The gas and liquid phases are agitated by two stirrers which rotate in opposite direction.

Among the various models available to describe mass transfer in the presence of chemical reaction film theory is widely applied because of its simplicity [29]. In film theory, mass transfer is by molecular diffusion through a stagnant layer of thickness x, while the bulk of liquid and gas is well mixed. The main assumption in film theory is that the whole resistance to absorption is located in the film layer of thickness x. Concentrations outside the film is uniform. The absorption takes place under steady state conditions. Gas absorption into liquid can be depicted from **Figure 1**. As pure gas is used so gas side resistance is neglected.
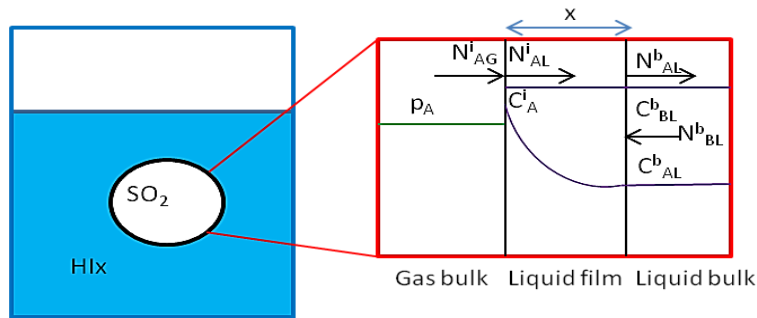
Figure 1. Mechanism of gas transfer into the liquid bulk

N_{AG}^i is the rate of molar A transfer per unit area from gas to interface. N_{AL}^i, N_{AL}^b is the flux of A in the film and liquid bulk respectively, N_{BL}^b is the flux of B entering liquid film; C_A^i, C_{AL}^b are the concentration of A at the interface and liquid bulk. p_A is the partial pressure of A, C_{BL}^b is the liquid phase concentration of B.

The *SO₂* absorption rate can be calculated from the experimental data by the mass balance in the reactor gas phase according to:

$$N_{AL}^i = -\frac{V}{RTA'}\frac{dp_A}{dt} = k_{AL}(C_A^i - C_{AL}^b) \tag{4}$$

Where V is the gas phase volume in the reactor, p_A is the partial pressure of *SO₂*, T is the temperature, R is the gas constant and A' is the gas liquid interfacial area, k_{AL} is the mass transfer coefficient of *SO₂* in the liquid phase, *t* is time. In Eq. (3), the ideal gas phase behaviour is applied. Measurement of absorption rates of *SO₂* into water gives the physical absorption behaviour. Mass transfer coefficient can be evaluated. Henry's law is used to obtain interface concentration.

From Eq. (4) and applying henry's law with C_{AL}^b (bulk *SO₂* liquid concentration) ~0, the following equation is deduced.

$$\frac{dp_a}{dt} = -\frac{RTA'}{V}k_{AL}\frac{p_a}{H} \tag{5}$$

H is the henry's constant. Eq. (6) is obtained by integrating Eq. (5) from initial pressure $p_{a,o}$ to p_a in time t.

$$\frac{p_a}{p_{a,o}} = exp\left(-\frac{RTA'k_{AL}}{VH}\,t\right)$$
(6)

Experimental pressure data can be used to obtain mass transfer coefficient k_{AL} from Eq. (6) by least square technique.

The absorption rate of SO_2 into an aqueous solution when chemical reaction takes place in the liquid phase with negligible concentration of A in the liquid bulk can be represented by the following equation with

$$N_{AL}^i = Ek_{AL}C_A^i$$
(7)

E is the enhancement factor. It describes the influence of a chemical reaction on the mass transfer rate.

Bunsen reaction can be represented in the form of gas-liquid reaction with A as SO_2 gas and B as liquid reactant here it is H_2O dissolved in HI_x solution which includes I_2. Non volatility of liquid solution is considered.

$$A(g) + v\,B(aq) \xrightarrow{k} C(aq)$$
(8)

The enhancement factor depends on the Hatta number, M and the enhancement factor of an infinitely fast reaction, E_∞. M gives the comparison of the reaction rate with the diffusion rate and is given by

$$M = \frac{\sqrt{k\,D_{AL}C_{BL}^b}}{k_{AL}}$$
(9)

k is the rate constant, D_{AL} is diffusion coefficient of A in liquid phase.

$$E_\infty = 1 + \frac{D_{BL}C_{BL}^b}{vD_{AL}C_{AL}^i}$$
(10)

Where D_{BL} diffusivity of B is in liquid phase, v is stoichiometric coefficient in Eq. (7). E_∞ represents the maximum enhancement that can occur in mass transfer because of reaction. When the Hatta number (M) is far less than the infinite enhancement factor (E_∞), it suggests that the concentration of H_2O is same in the liquid bulk and liquid film. The reaction in the liquid film is dependent on the concentration of SO_2 alone. Hence the reaction can be considered to be pseudo first order with respect to SO_2 in the liquid film.

With $Ha > 2$, the reaction is said to be fast in the liquid film [17].

$$2 < Ha \ll E_\infty$$
(11)

Inequality Eq. (11) represents the case of fast pseudo first order reaction in the liquid film. In this case enhancement factor is given by Eq. (12).

$$E = \frac{M}{tanh\,(M)} \tag{12}$$

For $Ha > 3$, it can be seen that:

$$tanh\,(M) \sim 1 \tag{13}$$

For this case, by introducing Eq. (4), (12) & (13) into Eq. (7), results in

$$N_{AL}^i = \frac{\sqrt{k`D_{AL}}\,p_A}{H} \tag{14}$$

$k`$ is the overall reaction rate constant. $k` = k\,C_{BL}^b$. By satisfying Eq. (11) & (13) the reaction of SO_2 with H_2O in HI_x solution can be considered to occur in the fast pseudo first order reaction regime.

Similarly, from Eq. (4), (7) and (14) the following relation can be derived:

$$\frac{dp_a}{dt} = -\frac{RTA'\sqrt{k`D_{AL}}}{V}\frac{p_a}{H} \tag{15}$$

Eq. (16) is obtained by integrating equation (15) from initial pressure $p_{a,o}$ to p_a in time interval t.

$$\frac{p_a}{p_{a,o}} = exp\left(-\frac{RTA'}{VH}\sqrt{k`D_{AL}}\,t\right) \tag{16}$$

Eq. (16) is used to obtain reaction rate constant from experimental data. This estimation is accomplished with the help of the least-square technique.

$$\ln H = A" + \frac{B}{T} + C*\ln T \tag{17}$$

Eq. (17) is the empirical correlation for Henry's law constant for SO_2 reaction system applicable for the range of operating conditions in which $A"$, B and C are the parameters obtained from [30]. Diffusion constant is estimated from wilke and Chang correlation [31].

Materials and Methods

Experimental schematic is shown in **Figure 2**. The setup consists of a stirred glass reactor with liquid hold up of 200 ml and 1200 ml vapour volume. The temperature is maintained by water circulating through the jackets. Temperatures are monitored using 'K' type thermocouples (M/s General Instruments). The liquid molar ratio of HI_x (I_2/H_2O) is 1/1.57/5.32. The temperatures and pressures of study are from 30 ºC to 50 ºC and up to 0.5 bar (g) respectively, varying stirring speed from 0 to 360 rpm. SO_2 gas is charged in the glass reactor from gas cylinder using mass flow controller (M/s Bronkhorst) operation. Pressure is monitored using a pressure transmitter (M/s Siemens) with accuracy of % ± 0.5 measured value. Stirred glass reactor is shown in **Figure**

3. Initially liquid is loaded and desired temperature is obtained through jacket heating then the gas is introduced into the reactor in a very short time i.e < 3 seconds and stirring is started.
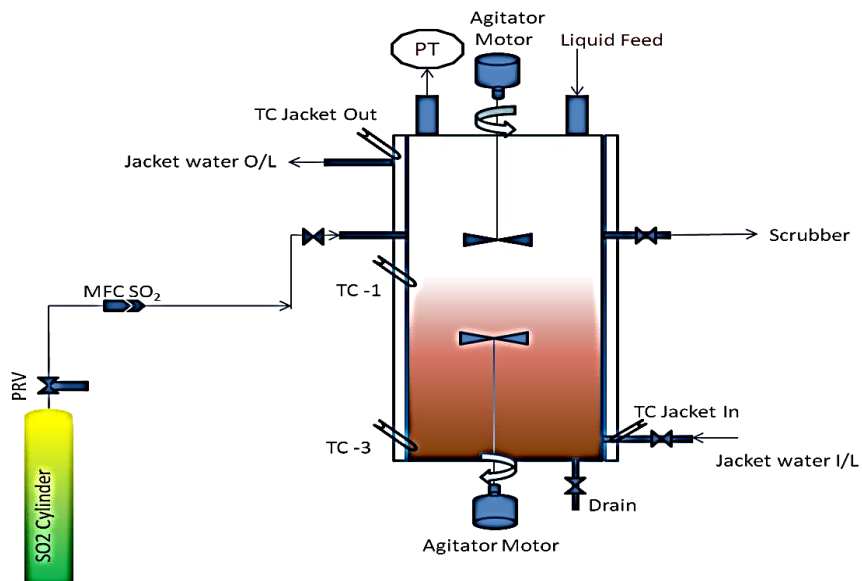
Figure 2. Schematic of experimental setup
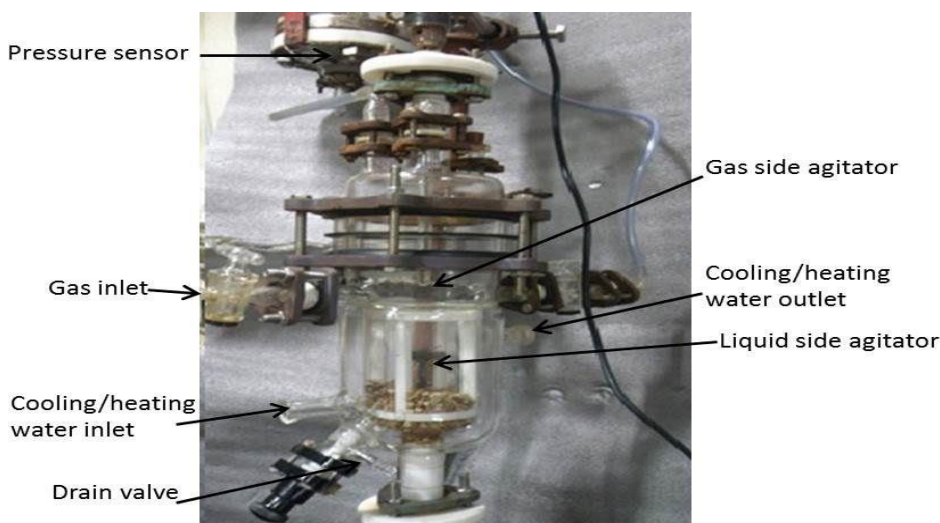
Figure 3. Glass stirred cell reactor

Initially experiments are carried out with sulphur dioxide dissolved in H_2O system and evaluated mass transfer coefficient at different stirring speeds. As stirring speed is increased rate of absorption increased and beyond certain stirring speed the mass transfer coefficient will not vary significantly and the absorption process of sulphur dioxide in reaction media (HI_x) is entirely governed by chemical reaction as given in Eq. (14). In all the experiments pure SO_2 gas is used

as feed and therefore gas side mass transfer resistance is neglected. Hence, initial experiments are carried out only to evaluate the liquid side mass transfer coefficient by varying stirring speeds from 0 rpm to 360 rpm. Later experiments are carried out with HI_x liquid phase and similar procedure is followed. Experiments are repeated to obtain the kinetic rate constant as described in Eq. (14).

Results and Discussion

Experiments have been performed with H_2O-SO_2 and HI-I_2-H_2O-SO_2 system at stirring speeds between 0-360 rpm. Varying temperatures from 30 ºC to 50 ºC and pressures up to 0.5 bar (g).

Effect of stirring speed on absorption rates

Figure 4 represents Sulphur dioxide pressure decrease rate at different stirring speeds. Increase in stirring speed has increased the rate of absorption of Sulphur dioxide which have been deduced from fall in pressure. The rate of pressure decrease has marginally changed when the stirring speed is increased from 0 rpm to 180 rpm. But the rate of pressure decrease has changed drastically from 240 rpm onwards and stabilized near 360 rpm. Mass transfer coefficient is obtained by regression and fitting parameter is obtained for Eq. (6).
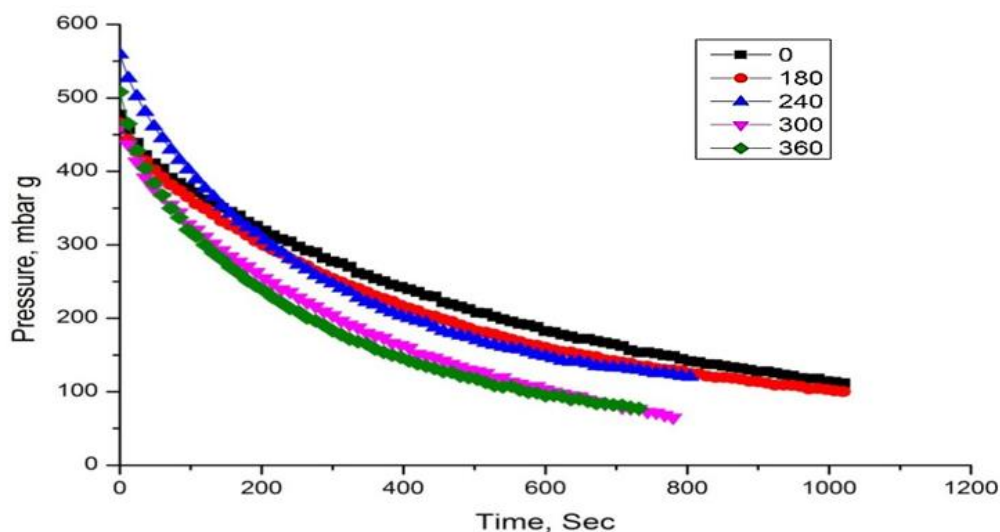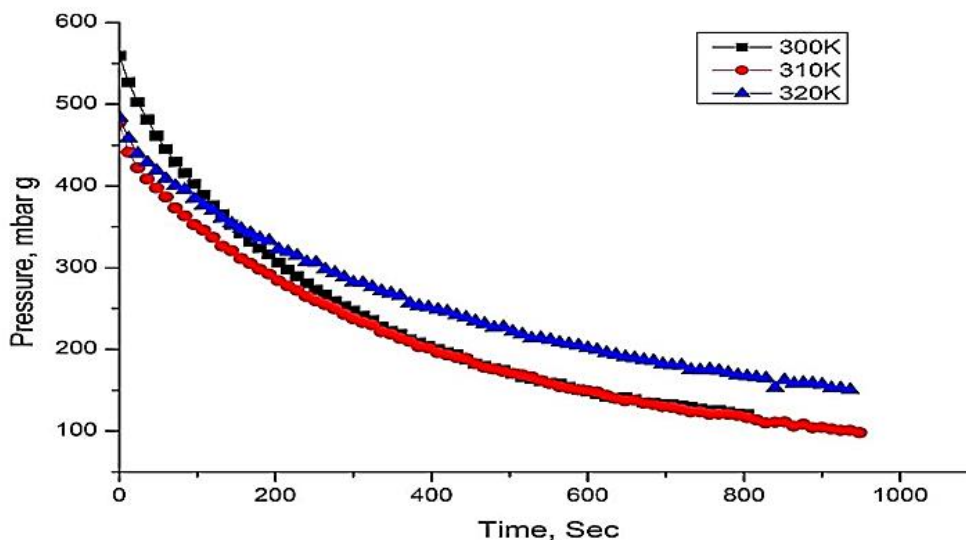

Figure 4. Rate of pressure variation profiles at various agitation speeds and 300 K for sulphur dioxide in water system

Effect of temperature on absorption rates

Figure 5 represents sulphur dioxide pressure decrease rate at different temperatures. As the temperature is increased there is a reduction in rate of pressure decrease. This is because of the fact that increase in temperature leads to decrease in solubility, which in turn decreases the absorption rates.

Figure 5. Rate of pressure variation profiles at different temperatures and agitation speed of 240 rpm for sulphur dioxide in water system

Effect of stirring speed on liquid side mass transfer coefficient

Eq. (6) is used to evaluate mass transfer coefficient which is the only unknown kAL parameter, by regression. Mass transfer coefficients obtained are plotted in **Figure 6**. The fitting parameters are good which can be seen from R^2 values given in **Table 1**. From the plot, it can be concluded that, beyond 300 rpm the stirring speeds, the mass transfer coefficient has little effect. Hence the effect of mass transfer can be neglected beyond 300 rpm in kinetic rate experiments in hydroiodic acid, iodine, water and sulphur dioxide.
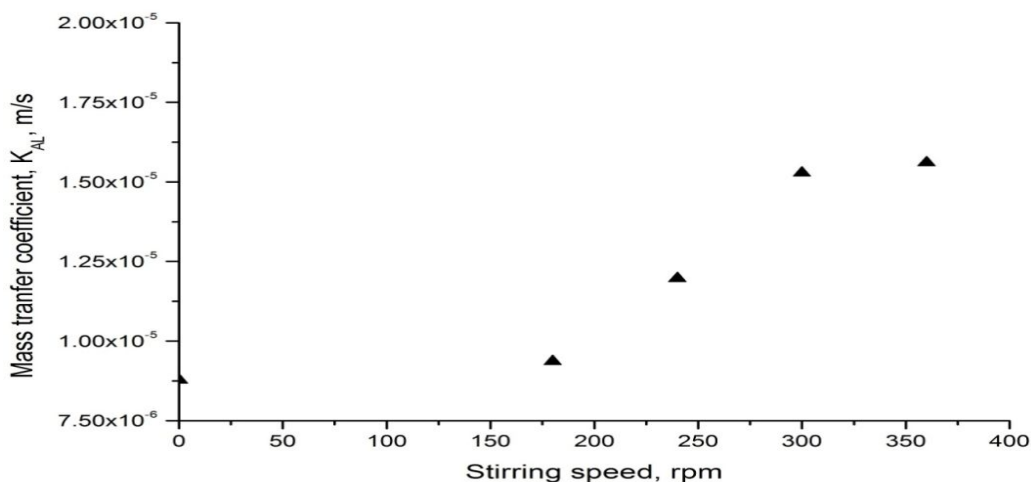
Figure 6. Variation of mass transfer coefficient with agitation speed at 300 K for sulphur dioxide in water system

Table 1. R^2 fitting values for different experiments to obtain mass transfer coefficient values

Experiment No.	Agitation speed (rpm)	Fitting parameter in Eq. (18)	R^2 fitting value
1	0	0.00135	0.9978
2	180	0.00144	0.9990
3	240	0.00184	0.9989
4	300	0.00235	0.9974
5	360	0.00240	0.9948

The experimentally obtained mass transfer coefficient for H_2O-SO_2 system has been compared with published literature correlation as given in Eq. (18).

$$Sh = 0.7082\ Re^{0.66}Sc^{0.11} \tag{18}$$

The plot is shown in **Figure 7**. Sh is Sherwood number which represents the ratio between mass transfer by convection and mass transfer by diffusion. Re is Reynolds number which represents the ratio of inertial to viscous forces. Sc is Schmidt number; it represents the ratio of momentum diffusivity (kinematic viscosity) and mass diffusivity. The experimentally predicted mass transfer coefficients are slightly higher than that obtained from Eq. (18). The average accuracy associated with Eq. (18) is ±11.8% in the 80-250 rpm as gven in [27], which is for CO_2-H_2O system. Upto 240 rpm the deviation in correlated and regressed mass transfer coefficient is within the average accuracy of Eq. (18). Beyond 240 rpm the variation between correlated and regressed mass transfer coefficient is more than ±11.8% which is under predicted Eq. (18). This is because SO2 is having higher solubility than CO_2 in H_2O. The stirring effects do not enhance the mass transfer coefficient after 300 rpm and can be seen in **Figure 7**.
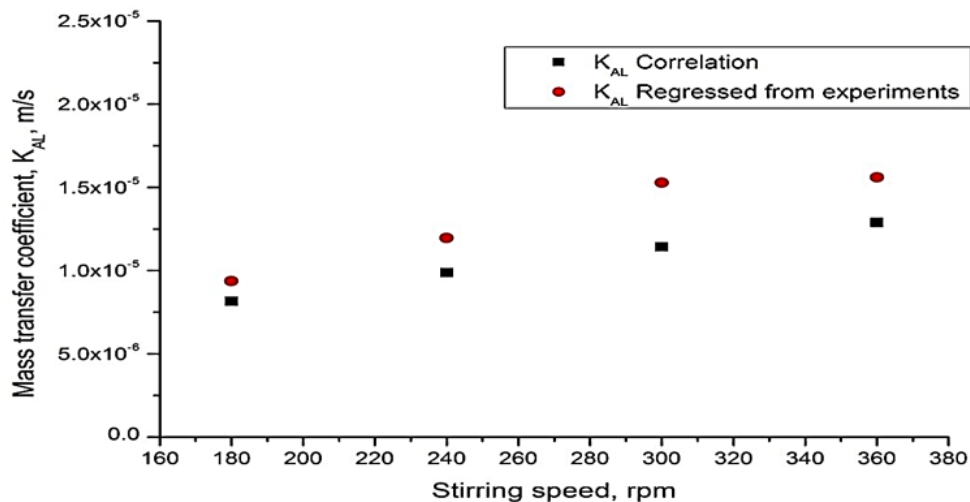
Figure 7. Variation of mass transfer coefficient with agitation speed at 300 K for H_2O-SO_2 system obtained from correlation and experimental regression

Pressure profile for hydroiodic acid, iodine, water and sulphur dioxide *system*

Figure 8 is the rate of pressure decrease for HI-I_2-H_2O-SO_2 system at 300 K and 300 rpm. The stirring speed for experiment in HI-I_2-H_2O-SO_2 system is selected based on the SO_2-H_2O system experiments, where mass transfer has little effect on reaction. Eq. (16) is used to evaluate overall kinetic rate constant, $k`$ which is the only unknown parameter, using **Figure 8** by regression.
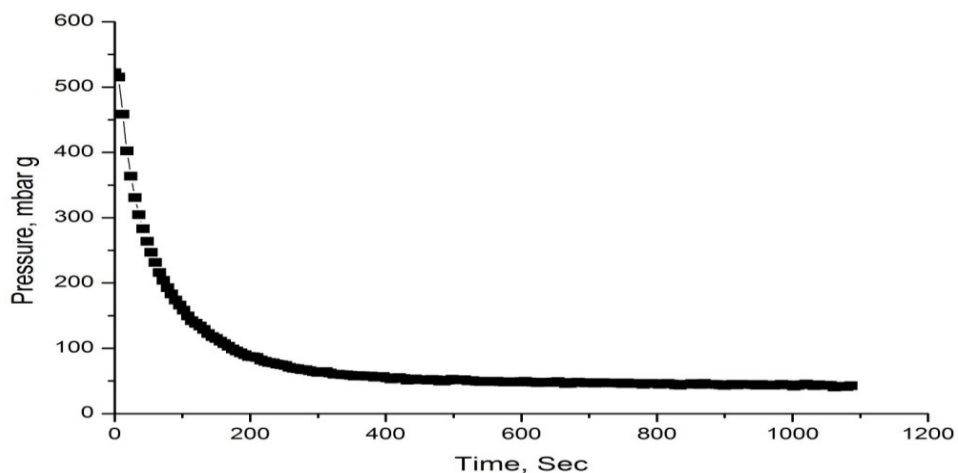
Figure 8. Rate of pressure variation at 300 K and 300 rpm system for HI-I_2-H_2O-SO_2 system

In order to ensure the chemical reaction regime is in fast pseudo first order regime, Eq. (11) and Eq. (13) needs to be satisfied. From the experimental analysis and Eq. (16), overall rate constant, $k`$ is obtained as 1.8 second^{-1}. From Eq. (9) Hatta number is 3.6. From Eq. (10) E_∞ is 18. Hence Eq. (11) and Eq. (13) are satisfied. The applicability of Eq. (14) is justified for the evaluation of overall kinetic rate constant.

Conclusions

Experiments have been performed in stirred glass reactor to evaluate mass transfer coefficient and reaction kinetic rate constant for Bunsen reaction. Experiments to evaluate mass transfer coefficient in water-sulfur dioxide system is carried out by varying stirring speed and temperature and obtained the same in the range of 8.7×10^{-6} m/s to 1.5×10^{-5} m/s . To evaluate kinetic rate constant for Bunsen reaction experiments are to be carried out such that mass transfer should be independent of stirring rates. Mass transfer coefficient values initially increased with stirring speeds of up to 300 rpm and remained almost constant at higher rpms, implying that the stirring speeds of more than 300 rpm do not greatly enhance the mass transfer rate. Beyond 300 rpm absorption of SO_2 in to the HI_x medium is governed by kinetics and evaluation of fitting parameter gives the value of kinetic rate constant. Overall kinetic rate constant is evaluated as 1.8 second^{-1} at 300 K. Hatta number and infinite enhancement factor is evaluated from the experimentally obtained kinetic rate constant as 3.6 and 18 respectively. From the calculated values of Hatta number and Infinite enhancement factors, the regime of chemical reaction is found to be in the fast pseudo first order reaction regime.

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