EXPERIMENTAL AND ANALYTICAL
INVESTIGATION OF AMMONIA VAPOR
ABSORPTION INTO AMMONIA-WATER SOLUTION

A dissertation submitted in partial fulfillment of the
Requirement for the degree of

Doctor of Philosophy (Ph.D.)

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ACKNOWLEDGEMENT

{Serve Allah, and join not any partners with Him; and do good to parents} Quran 4-36.

I dedicate this study to my parents who have never grudged me anything that has contributed in a way or another to the realization of my aspirations of being a productive person.

I wish to express my sincere appreciation to my direct supervisor, Professor Masonari Monde, Monde’s wise foresight, scientific acumen, subtle encouragement, and insistence on precision have helped me focus my thoughts and separate the perceived from the actual. I owes a great deal to Associate Professor Yuichi Mitsutake and Lecturer Kenji Ishida, for their helpful advices and discussions. Also I would like to thank Mr. Kenichi Nakashima for his help with the workshop during the manufacturing of the experimental apparatuses.

“Those who teach me have my everlasting respect”, Arabic saying.

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SUMMARY

Absorption phenomenon of ammonia vapor into ammonia water solution has been investigated experimentally, by inserting superheated ammonia vapor into a test cell containing a stagnant pool of ammonia water solution. Before commencing the experiment, the pressure in the test cell corresponds to the equilibrium vapor of the ammonia-water system at room temperature. When the valve is opened, mechanical equilibrium is established quickly and the pressure in the test cell becomes equal to that of the ammonia vapor cylinder. The difference between the initial pressure in the vapor cylinder and the initial pressure in the test cell is found to have a major influence on the absorption rate. The main objective of this study is to investigate the effect of this initial pressure difference on the absorption rate of ammonia vapor. A correlation which gives the total absorbed mass of ammonia as a function of the initial concentration, the initial pressure difference and time is derived to estimate the mass absorption rate of ammonia vapor into ammonia-water solution. The absorbed mass at no pressure difference could be estimated from the absorbed mass at initial pressure difference. In addition the absorption process has been visualized by a Mach-Zehnder interferometer and the obtained fringes have been analyzed to get the concentration distribution together with that obtained by theoretical solution in which ammonia concentration at the interface changes with time. The obtained optical images allowed us to distinguish between two layers with different speeds of fringes propagation. The layer with slow propagation of fringes reflects pure mass diffusion with negligible heat effect especially after long time from starting the absorption, while the layer with fast propagation of fringes reflects a thermal controlled diffusion.
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NOMENCLATURE

\( A \) \hspace{1cm} \text{Coefficient of Eq.(11)}

\( a_{ij} \) \hspace{1cm} \text{Coefficient of Eqs. (8) and (9) (i=0, 1 and \( j \) = 0,1,2)}

\( b_{ij} \) \hspace{1cm} \text{Coefficient of Eq. (6-12) (i=0, 1,2)}

\( C \) \hspace{1cm} \text{Concentration of ammonia, \( kg_{NH_3} / kg_{sol} \)}

\( C_P \) \hspace{1cm} \text{Specific heat of the liquid, \( J/(kg \ K) \)}

\( D \) \hspace{1cm} \text{Mass diffusivity, \( m^2/s \)}

\( fr \) \hspace{1cm} \text{Fringe}

\( H \) \hspace{1cm} \text{Coefficient of Eqs. (6-7),}

\( k \) \hspace{1cm} \text{Thermal conductivity, \( (W/m.K) \)}

\( Le \) \hspace{1cm} \text{Lewis number}

\( m \) \hspace{1cm} \text{Mass, \( g \)}

\( m_0(t) \) \hspace{1cm} \text{Total absorbed mass when \( C_i=0.0 \ kg/kg, \ g/m^2 \)}

\( m_a(t) \) \hspace{1cm} \text{Mass absorption per unit area, \( g/m^2 \)}

\( n \) \hspace{1cm} \text{Refractive index}

\( r_{ij} \) \hspace{1cm} \text{Coefficients of equation (7-14), (\( j \) = 0,1,2)}

\( P \) \hspace{1cm} \text{Pressure, \( kPa \)}

\( P_{1i} \) \hspace{1cm} \text{Initial pressure of the solution in the test cell, \( kPa \)}

\( P_{2i} \) \hspace{1cm} \text{Initial pressure of the ammonia vapor, \( kPa \)}

\( S \) \hspace{1cm} \text{Cross section area of the test cell \( m^2 \)}

\( T_i \) \hspace{1cm} \text{Initial temperature, \( ^oC \)}

\( T_{jl}(t) \) \hspace{1cm} \text{Measured temperatures in liquid (\( j \) = 1,2), \( ^oC \)}

\( t \) \hspace{1cm} \text{Time, sec}

\( T_{int}(t) \) \hspace{1cm} \text{Interface estimated temperature, \( ^oC \)}

\( T_{jv}(t) \) \hspace{1cm} \text{Measured temperatures in vapor (\( j \) = 1,2,3), \( ^oC \)}
\( V \) Total volume of the vapor \( m^3 \)
\( v(t) \) Specific volume of the vapor, \( m^3/kg \)
\( z \) Depth from the interface, \( mm \)
\( z_{C} \) Depth propagated by concentration during diffusion, \( mm \)
\( z_{jL} \) Depth of thermocouple in liquid (\( j = 1,2 \)), \( mm \)
\( z_{jV} \) Depth of thermocouple in vapor (\( j = 1,2 \)), \( mm \)
\( z_{T} \) Depth propagated by temperature during diffusion, \( mm \)

**Greek symbols**

\( \Gamma(x) \) Gamma function
\( \Delta P_i \) Initial pressure difference, kPa
\( \alpha \) Thermal diffusivity, \( m^2/s \)
\( \lambda \) Wavelength
\( \rho \) Density, \( kg/m^3 \)
\( \sigma \) Relative standard deviation

**Sub/Superscripts**

\( a \) Absorbed
\( C \) Concentration
\( d \) Drop
\( f \) Final
\( g \) Gas
\( i \) Initial
\( int \) Interface
<table>
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<th>Symbol</th>
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<tr>
<td>$L$</td>
<td>Liquid</td>
</tr>
<tr>
<td>$Pred$</td>
<td>Predicted</td>
</tr>
<tr>
<td>$ref$</td>
<td>Reference</td>
</tr>
<tr>
<td>$V$</td>
<td>Vapor</td>
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Chapter 1

1.1 INTRODUCTION

The energy and environment scenario in the world is undergoing a rapid change. In the industrialized economy, energy is consumed in various forms, primarily heat, power and refrigeration. Currently each form of energy is generated separately. However, the concept of integrated energy solutions has become a key to energy conservation and optimization. As a result the concept of co-generation is being adapted increasingly in various forms, such as combined heat and power plants, and combined heat refrigeration and power plants.

Concerns about the depletion of the ozone layer and global warming have generated renewed interest in ammonia water absorption systems. Not only is the ammonia as working fluid benign with respect to ozone depletion, but also the ability to use waste heat or solar energy as the driving force for space conditioning or refrigeration leads to increased energy savings. Ammonia water mixture has many applications in industrial processes, geothermal systems and environmental systems. On power generation, Ocean Thermal Energy Convection (OTEC) which employs ammonia-water mixture as a working fluid also makes researchers to pay attention to ammonia. It is, therefore, of great interest to study the properties of ammonia-water and to investigate the absorption phenomenon of ammonia vapor into ammonia water mixture.
1.2 AMMONIA-WATER MIXTURE AND THE ENVIRONMENT

The first major environmental concern to strike the refrigeration-based industries was depletion of the ozone layer as the result of the emission of man-made chemicals into the atmosphere. The thinning of the ozone layer threatens human health through diseases such as skin cancer, eye cataracts and immune deficiency, affects flora and fauna, and also influences the planet’s climate. Ozone depletion is brought about by a number of chemicals known as ozone-depleting substances, the most notorious of which are the chlorofluorocarbons (CFCs). Recognition of the role of chlorine in stratospheric ozone depletion, and the determination that CFCs were the primary source of chlorine in the stratosphere, led to a series of intergovernmental agreements, beginning in 1985 with the Vienna convention on the protection of the ozone layer and the Montreal protocol on substances that deplete the ozone Layer in 1987. These were followed by the London, Copenhagen, Vienna, Montreal and Beijing Amendments of 1990, 1992, 1995, 1997 and 1999 [1]. Consequent upon the Montreal Protocol and subsequent measures try to ensure that the changeover to non-ozone depleting substances is achieved as quickly as possible. The second major environmental concern is climate change or global warming. This did not become a major area of attention until after the responses to ozone depletion had been initiated. Concerns on this issue are now beginning to complicate our handling of ozone depletion. Global warming arises because of the greenhouse effect.

To prevent the destruction of environment, natural working fluids have been getting a lot of attention in these days. Ammonia water mixture is an important substitute for refrigerants that contribute to global warming and the erosion of the ozone layer. Both ammonia and water are natural substances and without any major risk for the ozone layer and do not contribute to the greenhouse effect responsible for global warming. Through the Significant New Alternatives Policy (SNAP) program, the agency identified ammonia as an acceptable substitute to ozone depleting substances.
Chapter 1

INTRODUCTION

1.3 AMMONIA-WATER SOLUTION

1.3.1 Ammonia

Ammonia (NH₃) is a colorless, alkaline gas at ambient temperature and pressure, with a distinct pungent odor [2,3], and is highly soluble in water. A saturated aqueous solution of ammonia contains 45 percent ammonia by weight at 0°C and 30 percent at ordinary room temperatures. Dissolving into water, ammonia becomes ammonium hydroxide, NH₄OH, which is strongly basic and similar in chemical behavior to the hydroxides of the alkali metals. Ammonia was known to the ancients who derived both the name and the substance from sal ammoniac, which was produced at the Temple of Jupiter Ammon in Libya by the distillation of camel dung [4]. During the Middle Ages in Europe ammonia was obtained by heating the horns and hoofs of oxen and was called spirits of hartshorn. Free ammonia was obtained by the German alchemist Basil Valentine; its composition was determined by the French chemist Comte Claude Berthollet about 1777. In the 19th century the principal source of ammonia was the destructive distillation of coal; it was an important by-product of the manufacture of fuel gases. Today most ammonia is produced synthetically from hydrogen and nitrogen. Ammonia is an important refrigerant and is widely used in the chemical industries, especially in the manufacture of fertilizer, nitric acid, and explosives.

1.3.2 Properties of ammonia-water solution

Ammonia and water are highly polar substances, and have ability of hydrogen bonding. Despite many similarities they have considerably different normal boiling points and critical points. In Table 1.1 some properties of ammonia and water are presented.

<table>
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<tr>
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<th>Ammonia (NH₃)</th>
<th>Water (H₂O)</th>
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<tr>
<td>Molecular weight [kg/kmol]</td>
<td>17.03</td>
<td>18.0</td>
</tr>
<tr>
<td>Boiling point at 1.013 bar [K]</td>
<td>239.8</td>
<td>373.2</td>
</tr>
<tr>
<td>Freezing point at 1.013 bar [K]</td>
<td>195.4</td>
<td>273.2</td>
</tr>
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</table>
Ammonia and water form a non-azeotropic mixture. The difference between an azeotropic mixture and a non-azeotropic mixture is illustrated in Fig. 1.1, which shows temperature-composition diagrams for the two types of mixtures. In Fig. 1.1 the upper line represents the state of saturated vapor at the dew point temperature, which is where liquid starts to condense from the vapor. The lower line in the diagram is the saturated liquid line, where the mixture is at the bubble point temperature and vaporization starts to form vapor from the liquid mixture.

Thus, there is a two-phase mixture. For a pure substance, the bubble point and the dew point are identical. Therefore, for the pure compositions in the diagrams (i.e., x=0 and x=1), the saturation lines meet. For an azeotropic mixture, there is one composition, called the azeotropic composition, where the liquid and vapor in equilibrium have the same composition and temperature. For a mixture of this
composition, the boiling liquid forms vapor of the same composition and temperature as the liquid and the mixture behaves as a pure substance. For a nonazeotropic mixture, the temperature and composition continuously change during boiling, as illustrated by the points \( a \) to \( f \) in Fig. 1.1. When the mixture starts boiling at the bubble point temperature, given by the point \( a \), a vapor that is richer in the more volatile component is formed with a composition given by the point \( b \). When the mixture continues to be boiled, the temperature increases and the point \( c \) is reached, where the concentrations of the remaining liquid and of the vapor formed are given by the points \( d \) and \( e \), respectively. Eventually, the point \( f \) is reached, where the mixture is a saturated vapor at the dew point temperature and the concentration of the vapor is the same as the liquid concentration had at the beginning of the evaporation process.
1.4 AMMONIA ABSORPTION REFRIGERATOR CYCLE

The ammonia absorption refrigerator cycle differs from the vapor-compression cycle in the manner in which compression is achieved. In the absorption cycle the low pressure ammonia vapor is absorbed in water and the liquid solution is pumped to a high pressure by a liquid pump. Figure 1-2 shows a schematic arrangement of the essential elements of such a system.

![Diagram of the ammonia-absorption refrigeration cycle](image)

**Fig. 1.2** The ammonia-absorption refrigeration cycle

The low-pressure ammonia leaving the evaporator enters the absorber where it is absorbed in the diluted ammonia solution. This process takes place at a temperature slightly higher than that of the surroundings. Heat must be transferred to the surroundings during this process. The concentrated ammonia solution is then pumped through a heat exchanger to the generator where a higher pressure and temperature are maintained. Under these conditions ammonia vapor is driven from the solution as heat is transferred from a high-temperature source. The ammonia vapor goes to the
condenser where it is condensed, as in a vapor-compression system, and then to the expansion valve and evaporator. The diluted ammonia solution is returned to the absorber through the heat exchanger.

The distinctive feature of the absorption system is that very little mechanical work input is required because the pumping process involves a liquid. This follows from the fact that for a reversible steady-flow process with negligible changes in kinetic and potential energy, the work is equal to \(-\int vdP\) and the specific volume of the liquid is much less than the specific volume of the vapor. This cycle brings out the important principle that since the work in a reversible steady-flow process with negligible changes in kinetic and potential energy is \(-\int vdP\), a compression process should take place with the smallest possible specific volume [5].

1.5 AMMONIA ABSORPTION POWER CYCLE

The properties of ammonia and water are suitable for a power cycle working fluid mixture. For example, the substances are soluble in each other and easily separable. Ammonia and water have different boiling temperatures; thus, the ammonia-water mixture evaporates over a large temperature range, which is an advantage for power generation from a sensible heat source. The mixture has been proposed to substitute steam in a Rankine cycle. The most well known power cycle using an ammonia-water mixture as a working fluid is the Kalina cycle.

Figure 1.3 shows the configuration of the simplest possible Kalina cycle. The working fluid is vaporized and superheated in the vapor generator and then expanded through the turbine. The hot vapor leaving the turbine is cooled in the reheater, then; the working fluid is mixed with the ammonia lean liquid from the separator. The resulting stream, called the basic mixture, is condensed in the absorber. By lowering the mass fraction of ammonia of the working fluid before the absorber, the turbine can be expanded to a lower pressure. This results from the fact that a mixture with low mass fraction of ammonia has a lower condensing pressure than a mixture with high mass fraction of ammonia at the same temperature. After the absorber, the basic
mixture is pressurized and then split into two streams. A part of the streams is heated before it enters the separator. There, the partly vaporized stream is separated into a vapor enriched in ammonia and a liquid lean in ammonia. The enriched vapor is mixed with the other stream from the splitter after the absorber and the mass fraction of ammonia of the working mixture is restored. The working mixture is then condensed and pressurized before it enters the vapor generator.

![Ammonia-water cycle diagram](image)

**Fig. 1.3** The simplest possible ammonia-water cycle
The advantage of the ammonia–water cycle over the Rankine cycle is more pronounced when the heat source is in the form of sensible heat, in the temperature range of 400–600°C, and has a large temperature drop [6]. The main reason for the high power production of the ammonia–water cycle, compared to the Rankine cycle, is the boiling and condensing behavior of the ammonia–water mixture working fluid, which is a non-azeotropic mixture. As the composition of the working fluid changes when the mixture boils at a constant pressure, the temperature increases and similarly, the temperature decreases during condensation. A pure substance, like water, boils and condenses at a constant temperature, if the pressure is constant. This is illustrated in Fig. 1.4, which shows the temperature profiles of a bottoming cycle boiler [6]. The boiling behavior of the mixture makes it possible to decrease the irreversibilities in the boiler as the temperature profiles of the heat source and the working fluid are more closely matched than in the Rankine cycle. Through the variation of the ammonia concentration throughout the ammonia–water cycle, the temperature profiles in the other heat exchangers can also be closely matched.

![Fig. 1.4 Temperature’s profile of a bottoming cycle boiler](image-url)
Another reason for the higher power output of the ammonia–water cycle is the low boiling temperature of the ammonia–water mixture. For example, at atmospheric pressure, the boiling temperature of pure ammonia is 233°C and the boiling temperature of pure water is 100°C. The ammonia–water cycle can therefore use heat sources of low temperature that the Rankine cycle cannot use. The ammonia–water cycle also has a higher potential for internal heat recovery compared to the Rankine cycle.

1.6 REMARKS ON AMMONIA-WATER ABSORPTION

The ammonia-water fluid has a volatile absorbent, thus presenting both heat and mass transfer resistances across the respective temperature and concentration gradients in both the liquid and vapor phases. The highly non-ideal ammonia-water fluid pair releases a considerable amount of heat of absorption at the vapor-liquid interface that must be transferred across a liquid film into the coolant. Some of this heat released at the interface is also transferred to the vapor, depending on the local temperature differences. The coupled heat and mass transfer processes in both phases have presented challenges for analysis, modeling, experimental validation, and design. In addition, the fluid flow in the respective phases is rarely well defined. In ammonia water systems, the absorber, in which absorption takes place, is one of the major and critical components; its performance greatly affects the overall system performance [7], it is a combined heat and mass exchanger that absorbs the vapor phase of the absorbate (NH₃) in a liquid absorbent (H₂O) before pumping the solution to the high-pressure side of the cycle.
1.7 LITERATURE REVIEW

A brief review of the literature on absorption heat and mass transfer is provided in this section. Based on this review, the deficiencies in the understanding of absorption phenomena are pointed out, and the objectives for the current experimental work are formulated.

1.7.1 Bubble-type absorbers

Lee et al. [8] analyze experimentally the ammonia-water absorption process for falling film and bubble modes in a plate type absorber; they conclude in the plate type absorber, the mass transfer performance of the bubble mode was better than that of the falling film mode. Though more heat was generated in the bubble mode, the mount of transferred heat in the bubble mode was less than that in the falling film mode at the low ammonia gas flow rate. Merrill and Perez-Blanco [9] investigated increasing the interfacial area per unit volume of vapor and liquid mixing at the vapor-liquid interface by breaking the vapor up into small bubbles and injecting them into the liquid. Terasaka et al. [10] investigate the mechanism of gas absorption from a bubble containing soluble and insoluble components, a gaseous mixture of ammonia and nitrogen was bubbled into water and describes the mass transfer by separating it into three mass transfer resistances for the gas phase, interface and liquid phase. For the beginning of bubble formation, the resistance at the interface was important rather than the other resistances. They conclude that, by applying the mass transfer mechanism to the non-spherical bubble formation model, the bubble growth curves, the bubble shapes, the bubble volume at its detachment and the mass transfer rate from growing bubbles at an orifice were accurately estimated. For the case of ammonia absorption into water, 80–90% of the ammonia in the feed gas was dissolved during the bubble formation. Kang et al. (12) evaluated the heat and mass transfer resistances in both the liquid and vapor regions in a countercurrent ammonia-water bubble absorber composed of a plate heat exchanger with offset strip fin inserts. They concluded that the heat transfer resistance was dominant in the vapor phase,
while the mass transfer resistance dominated in the liquid phase. In addition, the interfacial mass transfer area and vapor hold up were found to be significant in determining the size of the absorber. A vertical-tube bubble absorber with co-current solution and vapor flow in an inner tube, and countercurrent coolant flow in the annulus, was modeled by Perez-Blanco [13]. Several simplified assumptions were made including: the absorption process was at steady state, the process occurred at a constant pressure, all bubbles had the same diameter and velocity at a given location along the absorber length, bubble breakup and coalescence were negligible, resistance to mass transfer in the bubble was negligible, and no direct heat transfer took place between the vapor and the coolant. They found that as the ammonia concentration within the bubble decreased to that at equilibrium, water transfer was in the opposite direction of ammonia transfer; however, when the ammonia concentration dropped below that at equilibrium, water and ammonia transfer were in the same direction.

1.7.2 Falling-Film absorption

Among the ammonia-water investigations pertinent to the subject project is the experimental study of a coiled-tube absorber Jeong S [14] in which dilute solution flowed over the outside of the tubes with the ammonia vapor flowing upward in the shell and coolant flowing within the tube. They conducted experiments with and without absorption and found that film heat transfer coefficients were lower for experiments with absorption than those without absorption, perhaps due to insufficient wetting caused by vapor shear. For laminar, steady flow at low Reynolds numbers with uniform wetting, Perez-Blanco [13] presented a simple 1-D model for the absorption process in a horizontal-tube, falling-film absorber, while accounting for water transport both into and out of the solution film. To avoid unrealistic excessively high mass transfer rates at the interface, he used a two-film model to decouple the interface concentrations of ammonia in the liquid and vapor. Potnis et al. [15] developed a computer program that simulated the GAX process with liquid-film absorption over a coiled fluted tube with countercurrent vapor flow, and convective desorption inside the fluted tube. Takuma et al. [16] analyzed condensation of ammonia-water mixtures on horizontal tube bundles using a heat and mass transfer
analogy and confirmed the predictions using measurements on a full-scale shell-and-tube condenser as well as a test apparatus using the Coherent Anti-Stokes Raman Spectroscopy technique for ammonia concentration measurement. They concluded that the accumulation of ammonia at the interface presents an important resistance to condensation. Attempts at obtaining compact ammonia-water absorber geometries include counter-current fluted-tube absorbers, Kang and Christensen [17]. A generalized design tool capable of modeling several components within an absorption system was presented by Kang et al. [18]. Goel, N., and Goswami [19] proposed a new design of a falling film absorber that could considerably reduce the absorber size. The concept forms a liquid film between the horizontal tubes by a flow guidance system formed by wrapping a mesh alternatively between the left and right sides of the adjacent tubes. The mesh can be made of a compatible material, such as aluminum, steel, glass fiber, or nylon. In addition to increasing the liquid-vapor interfacial area, the concept also enhances the film stability by preventing coalescence of droplets on the horizontal tubes.

1.7.3 Marangoni convection

The Marangoni convection stimulation is a practical method used by the thermal absorption technology in the past decades in order that the absorption processes are significantly improved. The literature devoted to this effect is rich, including theoretical and experimental studies on various working fluid-absorbent–surfactant combinations. Isvoranu et al. [20] explained the basic mechanism of the Marangoni effect, using the two-point theory (TPT) of mass and heat transfer. They concluded high surface tension gradient plays the most important role in Marangoni convection. TPT is a powerful tool in the refined qualitative/quantitative binary two-phase local interaction analysis. Kang et al. [21] investigated the enhancement of mass transfer rate by adding different kinds of alcohol as a surfactant and visualized Marangoni convection that is induced by adding a heat transfer additive, n-Octanol. They concluded that the absorption performance could be improved by increasing the heat transfer additive concentration. The absorption heat transfer was enhanced as high as 3.0–4.6 times by adding the heat transfer additive.
1.7.4 Researches with other points of focus

Ferrario et al. [22] simulated ammonia mass diffusivity and estimated the diffusion coefficient values within the range $2\sim5\times10^{-9}$ m$^2$/s at a temperature of 20 °C. Takuma et al. [16] analyzed condensation of ammonia-water mixtures on horizontal tube bundles using a heat and mass transfer analogy and confirmed the predictions using measurements on a full-scale shell-and-tube condenser as well as a test apparatus using the Coherent Anti-Stokes Raman Spectroscopy technique for ammonia concentration measurement. They concluded that the accumulation of ammonia at the interface presents a significant resistance to condensation. Data for boiling of the non-azeotropic binary mixture, water/ammonia, on a horizontal surface are presented by Arima et al. [23]. Issa et al. [24] studied absorption of ammonia vapor into ammonia water solutions at several ammonia mass fractions, $C_i$, with a constant pressure difference of $\Delta P_i = 50$ kPa. Issa et al. applied three different methods to measure the rate of mass absorption, the first one was based on vapor pressure drop, and the second one was estimated by converting the interface heat flux, and the third one involved an optical procedure to observe density change during heat and mass diffusions. They found that the estimation by the vapor pressure drop does not agree with that by interface heat flux, but agrees with the third procedure. Therefore, they recommended that the first method is more reliable. Issa et al. [24] proposed Eq. (1.1) to predict the absorbed mass of ammonia into ammonia solution for a range of $C_i = 0.0$ to 0.82 kg/kg for a fixed $\Delta P_i$ of 50 kPa.

$$m_{a,\text{pre}(t)} = (\sum_{j=0}^{N} a_{0j} t^{j/2}) + (\sum_{j=0}^{N} a_{1j} t^{j/2}) e^{-C_i/H}$$  \hspace{1cm} (1-1)

They recommended $k = 0.26$ and $N = 2$ in Eq. (1-1).
1.8 OBJECTIVES

Referring to section 1.7, some additional researches should be carried out to:
- Understand the absorption process supported by quantification of the heat and mass transfer resistances in the vapor and liquid phases and the assessment of their relative significances over a wide range of concentrations.
- Illustrate the liquid-vapor interface behavior and its properties during absorption process. Experiments with theory-based validation are necessary to address these needs.

The present research is of experimental type that is performed to analytically investigate the absorption phenomenon of ammonia into ammonia water solution. The effect of initial pressure difference and initial concentration on absorption process has been investigated by carrying experiments with different initial pressure difference between the test cell and the ammonia vapor cylinder. Absorption process has been visualized by a Mach-Zehnder interferometer, the visualized fringes show two layers with different speeds of fringes propagation. The layer with fast propagation represents pure thermal diffusion while the slow propagation layer is characterized by a superposition of mass and heat diffusions where heat effect is big at short time from starting the absorption and this effect decreases with the elapse of time. Therefore, the main objectives can be declared as:

1. Estimate the interface temperature and concentration during the absorption process.
2. Calculate the total absorbed mass of ammonia vapor from measured pressure and temperature.
3. Investigate the effect of pressure on the absorption rate.
4. Investigate the effect of initial concentration on the absorption process.
5. Derive a correlation which gives the total absorbed mass of ammonia as a function of initial concentration, initial pressure difference and time.
7. Obtain the concentration distribution in the liquid.
8. Distinguish between two layers with different speeds of fringes propagation.
Chapter 1

1.9 SCOPE OF THE DISSERTATION

This dissertation describes an experimental study on the absorption of ammonia vapor into ammonia water solution. It is divided into eight chapters:
Chapter 1 provides a literature survey on ammonia-water systems and present study with its objectives.
In Chapter 2, details of the experimental setup are presented. Experimental procedure, measurement ranges and uncertainties are also described in this chapter.
In Chapter 3, the measured temperature and pressure are shown, the effect of initial concentration and initial pressure difference on pressure drop are discussed. All data are presented and analyzed in various ways to understand the mechanism of absorption process.
In Chapter 4, the absorbed mass of ammonia vapor is estimated, and the effect of initial pressure difference and initial concentration on mass absorption are concluded.
In Chapter 5, the temperature and concentration of the interface are estimated by using Monde’s inverse heat conduction problem technique, IHCP.
In Chapter 6, developing an equation that represents the dependency of the total ammonia mass absorption upon ammonia initial mass fraction in the solution, pressure difference, and time t is discussed
In Chapter 7, the concentration distribution in the liquid is obtained by analyzing the visualized fringes.
In Chapter 8, Concluding remarks are enumerated.
Chapter 2

EXPERIMENT APPARATUS

2.1 GENERAL DESCRIPTION

In this experimental study, superheated ammonia vapor is allowed to flow into a stagnant pool of ammonia water solution with a certain mass fraction, $C_i$, to be absorbed. During the absorption, temperatures and pressure are recorded; meanwhile, the absorption process is visualized by using a Mach-Zehnder interferometer. Figure 2.1 shows schematic diagram of the experiment’s target.
Chapter 2

EXPERIMENT APPARATUS

2.2 EXPERIMENTAL SETUP

A photograph and schematic diagram of the experimental setup are shown in Figs. 2.2 and 2.3, respectively. The main components of the experimental apparatus are the test cell, where absorption takes place, the ammonia vapor cylinder, and the mixing cylinder. The main components, all connections and valves are made of stainless steel, and are thermally insulated; Teflon is used as a seal material.

Fig. 2.1 Schematic diagram of the experimental target
Fig. 2.2 A photograph of the experimental setup
2.3 EXPERIMENTAL APPARATUS

The components are:

- The test cell, where the absorption process takes place.
- The Ammonia vapor cylinder (AVC).
- Mixing Cylinder (MC).
- Mach-Zehnder interferometer.
- Measurement pressure and temperature tools.
- Water filtering unit.
- Circulating aspirator for vacuum.
- Personal Computer.
- Video cameras.

2.3.1 Test cell:

The test cell, where the absorption process takes place, is made of stainless steel 304 SUS and has an internal volume of 180,000 mm$^3$ (120×100×15 mm). Figure 2.4
shows a cross section in the test cell. The five thermocouples are installed downward; two in the liquid at positions $z_{1L}$ and $z_{2L}$ from the interface to measure temperatures $T_{1L}$ and $T_{2L}$ respectively, and the other three are at different points to measure the vapor temperature. In order to measure vapor pressure in the test cell during the experiment, a pressure gage and a semi-conductor transducer are installed.

The internal dimensions: 120x100x15 mm

*Fig. 2.4* Cross section in the test cell

To ensure uniform distribution of ammonia vapor all over the interface when flowing into the test cell, a 20-µm stainless steel mesh is installed at the top, besides making three inlets instead on only one by using a collector. The test cell is equipped with polished Pyrex sight glass with 30 mm sight diameter to allow the laser beam to transmit through for visualization, and to measure the position of the interface.
2.3.2 Ammonia vapor cylinder (AVC):

The ammonia vapor cylinder, with an internal volume of 1350 cm$^3$, is used to feed the test cell with superheated ammonia vapor all through an experiment. The cylinder AVC is made of 304 SUS with 94 mm internal diameter, 200 mm height, and a thickness of 4 mm. It is designed to hold as much pressure as 2 MPa. In order to measure temperature and pressure, a thermocouple and a semi-conductor transducer are installed.

2.3.3 Mixing cylinder (MC):

The function of mixing cylinder (MC) is to prepare ammonia water solution with a certain mass fraction and then inserted into test cell. The cylinder MC is made of 304 SUS with 94 mm internal diameter, 200 mm height, and a thickness of 4 mm. It is designed to hold as much pressure as 2 MPa. To record temperature a thermocouple is installed, in addition a pressure gage is installed for reading pressure. For observing the existence of liquid a sight glass is installed at the upper cover.

2.3.4 Mach-Zehnder interferometer

During the absorption process, Mach-Zehnder interferometer is used to determine the phase shift caused by simultaneous heat and mass transfer into the liquid. The incident beam of the Mach-Zehnder interferometer is split into two beams before transmitting through the test cell. One of the two beams transmits through the sight glasses of the test cell and is called the measured beam. The other beam passes through the air and is, therefore, called the reference beam. Both of the beams are united again at a beam splitter to incident finally on the CCD camera that is connected to the DV tape recorder. The condition under which fringes are formed is the optical path difference between the measured and the reference beams, where the optical path is defined as the geometrical distance multiplied by the refractive index $n$. Refractive index is a function of three parameters:

\[ n = f(C, T, \lambda) \]  

(2-1)

Where, $C$ is the concentration, $T$ is the temperature, and $\lambda$ is the wavelength.
2.3.5 Water filtering unit:

The water used in this experiment is filtered by YAMATO WG-221 water filtering unit, where three steps are achieved. First, water enters to the filter, second, an ion exchange process, third and last is the distillation process.

2.3.6 Circulating aspirator for vacuum:

The Circulating aspirator that used in this research is Circulating aspirator WJ-20, obtained from SIBATA. The benefit of using this unit is to vacuum Test cell, AVC and MPC before feeding Ammonia or the mixture.

2.3.7 Cameras:

In order to film the interference fringes which are formed during ammonia diffusion in the liquid, a CCD camera (NEC TI-324A) is used. To capture images of the interface position, SONY DCR-PC300 has been used. As a video recorder, another digital video camera is connected to the two cameras.

2.4 DATA ACQUISITION SYSTEM

During the experiment only the pressure and temperature are measured, at the test cell and AVC. Figure 2-5 shows the data acquisition system for both temperature and pressure. This system is based on converting electrical signal that comes from the measured medium to readable values through certain logic. The data sampling rate is 1 Hz, allows the measuring loop to record one measured value at each second.
2.4.1 Measuring temperature tools:

The thermocouple provides a simple and efficient means of measuring temperature because it produces a voltage which is a function of temperature. This voltage can be read using an analog to digital converter. Figure 2.6 shows temperature measuring loop. It is starts by the low voltage electrical signal that comes from the thermocouples. This signal, then, is amplified in a voltage amplifier to send to the AD converter that is installed in the personal computer. The software program, which is written in Basic language, converts the AD output to a data saved as binary file. Therefore, another program written in FORTRAN language is used to convert the unreadable data (HEXA) to readable temperatures (ASCII) in C° unit. The temperature measuring loop consists of:

1. Thermocouples: The maker is CHINO, type SK, class 2, material SUS316, and the diameter of the sheath is 1 mm.
2. Icebox, SAMOCUT, D1000, material SUS.
3. Voltage amplifier with 8 channels, NEC RMT CONT AMP AH1108
4. AD converter with 8 channels, CONTEC AD16-16U
5. Personal computer, NEC PC-9801
2.4.2 Measuring pressure tools:

Figure 2.7 shows pressure measuring loop which is started by sensing the pressure changes and reflecting these changes as a mechanical signal. This signal is converted to low voltage electrical signal within the transducer itself, which is connected to the measured medium enclosure. This signal then is amplified in a strain amplifier to send to the AD converter that is installed in the personal computer. The same software program that is used for the temperature is used to convert the AD output to unreadable data, which is converted to readable one in kPa unit.
Pressure measuring loop consists of:
1. Semi-conductor transducer: maker KYOWA, type PG-20HU, CAP. 20 Kgf/cm².
2. Strain amplifier: maker KYOWA, type DPM-711B.
3. The same personal computer of the temperature has been used for pressure loop.

Both voltage and strain amplifiers need calibration from the very beginning to establish gains and offsets. The reference value for the strain amplifier is the absolute atmospheric pressure.
2.5 EXPERIMENTAL PROCEDURE

2.5.1 Preparing ammonia water solution:

I. According to Gibbs phase rule, two properties at equilibrium condition of two-phase binary mixture should be known to obtain the others such as specific volume and enthalpy. These two properties are the desired ammonia mass fraction of the solution $C_i$ and the surrounding temperature $T_i$. The equilibrium pressure can be estimated by using PROPATH software [25].

II. The mixing cylinder and its connecting tubes with the water cylinder and the ammonia tank are evacuated completely.

III. A certain quantity of the pure water is inserted into the mixing cylinder. This quantity is determined by the specific volume of the desired solution.

IV. Ammonia is inserted slowly and gradually into the mixing cylinder while observing the pressure. During the insertion of ammonia into the mixing cylinder, some time is necessary to check whether the pressure inside the cylinder is close to the equilibrium pressure that is obtained from the known values of $C_i$ and $T_i$, [25]. When the pressure inside the mixing cylinder reaches the equilibrium pressure at the established temperature, the mixing process is completed.

2.5.2 Experimental steps:

The experiment is performed by the following steps:

1. The test cell and its connection tube with ammonia vapor cylinder are evacuated.

2. Ammonia water solution from the mixing cylinder is inserted into the test cell until it completely covers the thermocouple that measures $T_{1L}$ (see Fig.2.4).

3. After the equilibrium state is reached, the initial saturated vapor pressure of the solution in the test cell, $P_{1i}$, can be read through a semiconductor transducer, installed in the test cell with a distance of about 40 mm above the interface.

4. The designed initial vapor pressure, $P_{2i}$, which must be exerted in the ammonia vapor cylinder, is calculated as: $P_{2i} = P_{1i} + \Delta P_i$ [kPa].
5. The ammonia vapor cylinder is evacuated and then ammonia vapor is inserted into it until the designed initial pressure $P_{2i}$ is reached at the established initial temperature $T_i$.

6. When the initial temperature $T_i$ and the pressure difference $\Delta P_i$, are established, the computer is ready to record the data. The experiment can be started by opening the vapor inlet valve of the test cell, which connects the test cell and the ammonia vapor cylinder.

7. We open the inlet valve to allow superheated ammonia vapor to flow into the test cell. Meanwhile the personal computer records the pressure and the temperatures during the experiment.

It should be noted that the inlet valve is kept open throughout the experiment. Therefore, the total vapor volume is the sum of the vapor volume in the test cell, the volume of the ammonia vapor cylinder, and the volume of the tube connecting them.

Whenever operating with ammonia, extreme caution must be paid to avoid injury. When ammonia is being charged and discharged from the loop, the operator wears a full face gas mask with chemical cartridges approved for ammonia and methylamine.

2.6 UNCERTAINTIES OF MEASUREMENTS

The uncertainty in measurement is ± 1 % for the temperature, and ± 0.5 % for the pressure. The data sampling rate is 1 Hz, which allows the measuring loop to record one measured value for each second. It is worth mentioning that the sensitivity of the pressure transducer is 600 Pa with uncertainty of ± 3 Pa, and the response frequency is 2.5 kHz ± 0.25 kHz.

2.7 EXPERIMENTAL RANGE

The effect of initial pressure difference on the absorption process is investigated experimentally by testing six different initial pressure differences, $\Delta P_i = 50, 100, 150,$
200, 250 and 300 kPa for four different initial concentrations of ammonia solution. Fig. 2.8 shows the schematic of the initial pressure.

![Figure 2.8 Schematic of experiment's initial condition](image)

The initial concentration is ranging from 0 to 63%. It should be mentioned that the initial concentration 59% was investigated only under one initial pressure difference $\Delta P_i = 50$ kPa. The initial temperature was 20.7 to 25.4 °C. The experimental range is summarized in Table 2-1.

<table>
<thead>
<tr>
<th>$C_i$ [$kg_{NH_3}/kg_{H2O}$]</th>
<th>$\Delta P_i$ [kPa]</th>
<th>$T_i$ [°C]</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.0, 0.27, 0.46, 0.59, 0.63</td>
<td>50, 100, 150, 200, 250, 300</td>
<td>21 to 25</td>
</tr>
</tbody>
</table>

**Table 2.1** Initial condition of experiment
Chapter 3

MEASURED DATA AND ANALYSIS

During the experiment, vapor pressure and the temperature in liquid and vapor are measured. This chapter shows the measured temperature and pressure during the absorption process in the experiment.

3.1 MEASURED TEMPERATURE

In the test cell, five thermocouples are installed to measure temperatures in liquid and vapor phases, as is mentioned in the last chapter. Figure 3.1 shows the positions of the thermocouples, three thermocouples in the vapor and the other two in the liquid.

![Fig. 3.1 A photograph shows the positions of the thermocouples in the test cell](image)
3.1.1 Temperature in liquid

To illustrate clearly the behavior of the temperature in the liquid during absorption process, measured temperatures $T_{1L}$ and $T_{2L}$, at $z_{1L}$ and $z_{2L}$, in the case of $C_i = 0.59$ kg/kg, are individually shown in Fig. 3.2. The temperature $T_{1L}$ closer to the interface increases immediately after the inlet valve is opened to start the experiment. It attains a maximum value, 22.8 °C, within about 75 sec. Then, the temperature $T_{1L}$ gradually decreases.

\[ C_i = 0.59 \text{ kg/kg} \]
\[ \Delta P_i = 50 \text{ kPa} \]

**Fig. 3.2** Measured temperatures in liquid for the case $C_i = 0.59$ kg/kg

It was observed that, measured temperatures in the liquid for all the experiments have the same trend as shown in Fig. 3.2. Increasing the initial pressure difference $\Delta P_i$ increases the peak value of the temperature $T_{1L}$. Another remarkable notice is that, with increasing the initial mass fraction $C_i$, the peak value becomes smaller [26]. The reason behind the increasing of temperature $T_{1L}$ is due to the heat generation of reaction during the absorption of ammonia vapor into ammonia-water solution. As the number of free water molecules, which chemically reacts the ammonia as hydrogen
bonding, decreases from the interface the reaction rate becomes very slow, then the temperature $T_{1L}$ starts decreasing. On the other hand, the temperature $T_{2L}$ which is far from the surface, hardly changes with time, which means the heat does not penetrate in the depth of $z_{2L}$.

Figures 3.3 (a) through (d) show the measured temperatures in the liquid during the absorption process, for all the experiments.
Fig. 3.3 Measured temperatures in the liquid (continued)
Fig. 3.3 Measured temperatures in the liquid
3.1.2 Measured temperature in the vapor

Referring to Fig. 3.1, the temperatures, $T_{1v}$, $T_{2v}$, and $T_{3v}$ in the vapor are measured by three thermocouples at three different distances, $z_{1v}$, $z_{2v}$, and $z_{3v}$, respectively from the interface. Figures 3.4 (a) through (l) show the measured temperature in vapor at the test cell for several selected cases, and each figure shows the measured vapor temperatures at a particular initial pressure difference $\Delta P_i$. Referring to Fig. 3.3 (a), where $C_i = 0.0$ kg/kg and $\Delta P_i = 50$ kPa, the temperature, $T_{1v}$, suddenly increases, immediately after opening the inlet valve; within about 10 sec the thermocouple at $z_{1v}$ recorded 34$^\circ$C, and then it starts to decrease. Whereas in Fig. 3.3 (f), $C_i = 0.0$ kg/kg and $\Delta P_i$ is 300 kPa, the temperature, $T_{1v}$, within less than 10 sec, it reaches a maximum value of about 41$^\circ$C. Increasing the initial pressure difference increases the absorption heat. The measured temperatures, $T_{2v}$ and $T_{3v}$, show almost the same behavior as $T_{1v}$, but the maximum value is the highest at the temperature, $T_{1v}$, and the lowest for the temperature, $T_{3v}$, in consequence of temperature $T_{1v}$ is the closest to the interface at which the absorption heat is released, and the temperature, $T_{3v}$, is the most far from the interface. Temperature, $T_{1v}$, has a similar behavior of $T_{1L}$, since they are measured by closest thermocouples to the interface, the same observation of the temperature, $T_{1L}$, is noted for the temperature, $T_{1v}$, when ammonia mass fraction $C_i$ is 0.27, 0.46 and 0.63 kg/kg, the temperature, $T_{1v}$, increases followed by a drop. With increasing the initial mass fraction $C_i$ in the solution, the peak value of the temperature $T_{1v}$ becomes smaller. In the cases of $C_i = 0.46$ and 0.63 kg/kg, the temperature, $T_{1v}$, does not suffer a considerable increase and the measured temperatures, $T_{2v}$ and $T_{3v}$, are almost identical.

Ammonia vapor temperature is measured by four thermocouples, three thermocouples measure the vapor temperature in the test cell, and the other thermocouple measures the temperature in the ammonia vapor cylinder. The measured temperatures of the vapor in the test cell represent the temperatures at the points those are close to the interface and do not represent the temperature of the total vapor volume especially in the ammonia vapor cylinder which is about 1.5 m from the test cell. Furthermore, the vapor volume of the test cell is only 6 % of the total
vapor volume $V$. Therefore, we consider that the temperature in the ammonia vapor cylinder approximates a representative of the temperature for ammonia vapor, which is almost constant with time throughout the experiment.
Fig. 3.4 Measured temperatures in the vapor (continued)
Results and Discussion

Fig. 3.4 Measured temperatures in the vapor (continued)
Chapter 3

RESULTS AND DISCUSSION

Fig. 3.4 Measured temperatures in the vapor (continued)
Fig. 3.4 Measured temperatures in the vapor (continued)
Fig. 3.4 Measured temperatures in the vapor (continued)
Fig. 3.4 Measured temperatures in the vapor
3.2 MEASURED PRESSURE

Two semiconductor transducers are used to measure the vapor pressure during the absorption process $P_V(t)$. The first one is installed in the test cell, approximately 40 mm above the interface. The other semiconductor transducer is installed in the ammonia vapor cylinder. Fig. 3.5 shows the schematic diagram of the initial pressure, in Fig. 3.5 $P_{1i}$ denotes the initial pressure in the test cell and $P_{2i}$ denotes the initial pressure in the ammonia vapor cylinder pressure.

![Schematic diagram of the initial pressure difference](image)

**Fig. 3.5** Schematic of the initial pressure difference

When the valve is opened, mechanical equilibrium is established quickly and the pressure in the test cell becomes equal to that of the ammonia vapor cylinder. The pressure $P_{ref}$ is the first pressure reading, measured by the transducer that is installed in the test cell, which corresponds to the time immediately after the inlet valve is opened to start the experiment. Figures 3.6 (a) and (b) show the behavior of the vapor pressure in the test cell in the cases $C_i = 0.0$ and 0.27 kg/kg for all initial pressure difference $\Delta P_i$. 

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Fig. 3.6 Pressure change during absorption
The pressure \( P(t) \) decreases with time due to the absorption of ammonia vapor into the solution. The measured pressure at \( t = 1 \) sec shows that the ratio of \( P_{\text{ref}}/P_2 \) ranges from 0.9 to 0.98 depending on the value of \( \Delta P_i \) and the value of \( C_i \). When starting the experiment, ammonia vapor flows into the test cell resulting in a sudden increase of the pressure \( P_{1i} \) up to \( P_{\text{ref}} \) followed immediately by a drop due to the absorption process. The amount of pressure drop during absorption \( P_d(t) \) can be calculated from Eq. (3-1)

\[
P_d(t) = [P_{\text{ref}} - P_v(t)]
\]  

(3-1)

### 3.2.1 Effect of initial pressure difference on pressure drop during absorption

Figure 3.7 shows the effect of initial pressure difference on \( P_d(t) \) in the case of \( C_i = 0.27 \) kg/kg. After 600 sec from starting the absorption, the amount of pressure drop \( P_d(t) \) in the case of \( \Delta P_i = 50 \) kPa is about 13 kPa, while in the case of \( \Delta P_i = 300 \) kPa, \( P_d(t) \) is nearly 54 kPa. The large initial pressure difference results in the large drop of the vapor pressure. The increase in pressure on the interface increases the amount of absorbed ammonia molecules until the solution reaches equilibrium condition.
3.2.2 Effect of initial concentration on pressure drop during absorption

The experimental data demonstrate that the pressure drop $P_d(t)$ is dependent on the initial concentration. It is increased by decreasing the initial concentration and attained maximum value of 82 kPa for $C_i = 0.0$ kg/kg. The pressure $P_d$ is partially effected by time in the case of higher initial concentration, but this affect is worth mentioning for $C_i = 0.0$ kg/kg, it reveals sharp increase with time (0-400 sec), and then it turns to be constant. The high value of pressure drop for $C_i$ advocates the high rate of absorption for $C_i = 0.0$ kg/kg, as ammonia molecules, under high pressure, has great feasibility to transfer their energy to water molecules.
Fig. 3.8 Effect of initial concentration on pressure drop during absorption
4.1 ESTIMATION OF ABSORBED MASS OF AMMONIA VAPOR

The drop in ammonia vapor pressure and the released heat at the liquid-vapor interface indicate the absorption process, and the absorbed mass can be estimated. On the assumption that there is an instant equilibrium condition during the absorption process, two parameters of the vapor state are required to obtain the other parameters such as specific volume. The specific volume of vapor at any instant during the absorption process can be obtained from the measured vapor pressure $P_v(t)$ and the ammonia vapor temperature, using Tillner and Friend actual gas equation [27], vapor mass in the vapor chamber at any time during the absorption can be calculated as:

$$m_v(t) = \frac{V}{v(t)}$$

(4-1)

where $v(t)$ is the vapor specific volume at a time $t$, and $V = 1.57 \times 10^{-3}$ m$^3$ is the total vapor chamber which includes the volume of the vapor in the test cell, the volume of the ammonia vapor cylinder, and the volume of the connecting tube. The volume $V$ is constant since the total ammonia absorbed quantity is too small to cause noticeable rise in the liquid level of the test cell. Reference ammonia vapor mass $m_{refs}$ which exists in the vapor chamber just when absorption starts, can be calculated by Eq.(4-1) noting that $v(t)$ corresponds to the reference measured vapor pressure $P_{ref}$. Therefore,
total mass absorption of ammonia per unit area $m_a$ until time $t$ can be obtained by substituting the temporal vapor mass, from the initial ammonia vapor mass $m_{ref}$, as:

$$m_a(t) = \frac{|m_{ref} - m_v(t)|}{S}$$  \hspace{1cm} (4-2)

where $S = 1.5 \times 10^{-3} \text{ m}^2$ is the interface area.

### 4.1.1 Effect of initial concentration on mass absorption rate

Figure 4.1 shows the total absorbed mass per unit area that is estimated by Eq. (4-2) within the selected time of 600 sec from starting the absorption, and for different initial concentration $C_i$. It is noticeable that the total ammonia absorbed mass decreases dramatically with increasing ammonia initial mass fraction $C_i$. The same observation was noticed by Issa et al. [24]. The increase in the initial concentration of the solution decreases the ratio of free water molecules and, therefore, reduces the feasibility of energy transfer from high kinetic ammonia molecules to water molecules.

**Fig. 4.1** Effect of initial concentration on total absorbed mass
4.1.2 Effect of initial pressure difference on mass absorption rate

The effect of $\Delta P_i$ on absorption process is investigated from the calculated total absorbed mass. Figure 4.2 (a) through (d) show total absorbed mass for the six different values of $\Delta P_i$. In the case of $C_i = 0.0$ kg/kg, the initial pressure difference when $\Delta P_i = 50$ kPa, the total absorbed mass after 600 sec is about 227 g/m$^2$, whereas the total absorbed mass is around 913 g/m$^2$ at $\Delta P_i = 300$ kPa under same time span. Thus, increasing the pressure difference by a factor of six does not yield six times the absorbed mass for a given time. In the case of $C_i = 0.63$ kg/kg after 600 sec and at $\Delta P_i = 300$ kPa the total absorbed mass is only 117 g/m$^2$. The decrease in the absorbed mass with increasing initial concentration is due to the shortage of free water molecules.
Fig. 4.2 Effect of initial pressure difference on total absorbed mass (continued)
Fig. 4.2 Effect of initial pressure difference on total absorbed mass
Based on Henry’s law, the solubility of ammonia vapor affected by its partial pressure above the solution. As shown in Fig. 4.3, at equilibrium, before starting the experiment, the rate of escape of ammonia molecules from the solution equals to the rate of capture of ammonia molecules by the solution (a). After opening the inlet valve, the increase in pressure throws the system out of equilibrium (b); more ammonia molecules dissolve to equalize the rates of escape and system again attained equilibrium (c).

![Fig. 4.3 Schematic diagram of the solubility of ammonia molecules](image)
Estimation of Interface Conditions

5.1 Estimation of Interface Temperature

During ammonia absorption, heat is released at the interface and diffuses into the liquid and vapor, and hence, the closest temperature to the interface is always at the maximum due to the absorption heat released, as was shown in Figs. 3.2, 3.3 and 3.4 of Chapter 3. The released heat depends on the final concentration of the formed solution, \( C_f \). When 1 mole of ammonia vapor is absorbed into liquid water, the \( C_f \) of the formed solution is almost 0.0 mole fraction, and the reaction as follows:

\[
\text{NH}_3(g) + H_2O(l) \leftrightarrow \text{NH}_3(aq) + Q
\]  

(5-1)

where, \( Q = 34.18 \text{ kJ/mol} \) is the heat of solution which is the same of the standard solution enthalpy \( -\Delta H^\circ_{\text{sol}} \) at the temperature 298 K and the pressure 101.325 kPa. The reaction molar heat \( Q \) consists of two parts; the first one is the latent heat of condensation \( -\Delta H_{\text{cond}} \) that has the value of 23.35 kJ/mol at 101.325 kPa. The second part is the solution enthalpy \( -\Delta H_{\text{sol},0.0} \) of forming the solution that has a final mole fraction \( C_f \), which is almost 0.0. The sign “aq” stands for aqueous and indicates huge amount of the solvent, water. The thermal diffusion in the liquid can be stated as heat conduction in a semi-
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infinite body. The temperatures \( T_{1L} \) and \( T_{2L} \) are changing with time so that simple interpolation is not applicable. For a transient case, an inverse solution is needed, such as Monde’s inverse solution to the inverse heat conduction problem (IHCP) [28], to estimate the interface thermal condition from the measured temperatures \( T_{1L} \) and \( T_{2L} \). Figure 5.1 shows schematic heat release and the required parameters for implementing IHCP. The one-dimensional differential equation of the heat in a body is given as:

\[
\frac{\partial T}{\partial t} = \alpha \frac{\partial^2 T}{\partial z^2}
\]

(5-2)

where, \( \alpha \) is the thermal diffusivity of the body where diffusion occurs.

Fig. 5.1 Heat is released at the interface due to the absorption process
For certain initial and boundary conditions, the temperature distribution in a body can be found as a function of position and time. This is called the direct solution because the boundary condition is already known and the distribution of temperature inside the body is sought. In many practical applications, the temperature changes at the boundaries cannot be measured, such as the current experimental situation, but the temperature changes at a point inside the body can be easily measured. In such cases, inverse heat conduction method is needed to estimate the boundary conditions with the help of the measured temperature at that point. The interface temperature, denoted as $T_{int}$, can be estimated as shown in Figs. 5.2 (a), and (b).
Fig. 5.2 Estimated interface temperature

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5.2 ESTIMATION OF INTERFACE CONCENTRATION

The interface concentration is not equal to the concentration in the liquid, its value between the concentration of the vapor, which is nearly 100%, and the concentration of the liquid. However, the interface concentration, $C_{int}$, can be estimated if the following conditions are available:

- On the assumption, there is an instantaneous and local equilibrium condition at the interface.
- Interface pressure and temperature.

The interface pressure is almost equal to the vapor pressure, shown in chapter 3, and the interface temperature is already discussed in Section 5.1. Consequently, the interface concentration can be estimated by using Tillner and Friend actual gas equation [27]. Figure 5.3 shows the estimated interface concentration $C_{int}$. As can be seen, when initial concentration $C_i = 0.46$ and 0.63 kg/kg, the interface concentration is almost constant. In the case of $C_i = 0.27$ kg/kg, the interface concentration $C_{int}$ starts to increase sharply until it reaches the equilibrium condition within about 50 seconds from starting the absorption, and then it hardly changes with time. At the beginning of the absorption, the rate of reaction is high until the interface becomes almost saturated with the hydrated ammonia, when the reaction rate slows down, the interface concentration becomes almost constant.
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Fig. 5.3 Estimated interface concentration
The behavior of interface concentration when \( C_i = 0.0 \) kg/kg is noteworthy. The interface concentration \( C_{int} \) of the cases \( \Delta P_i = 50 \) and 100 kPa exhibits a similar tendency, \( C_{int} \) decreases rapidly compared to other initial pressures \( \Delta P_i \). The molecules of ammonia directly diffuse to the water phase due to abundance of free water molecules, as shown in Fig. 5.4 (a). When \( \Delta P_i = 150 \) and 200 kPa, the concentration \( C_{int} \) increases in a short time, about 20 sec, then drops slowly. Increasing the initial pressure forces ammonia molecules to penetrate into the solution, due to the retardation of mass diffusivity, ammonia molecules start to accumulate at the interface until the absorbed ammonia diffuses into a certain depth in the solution, (see Fig. 5.4 (b)). On the other hand, the interface concentration \( C_{int} \) of the cases \( \Delta P_i = 250 \) and 300 kPa are increasing within about 50 sec, then are hardly changing with time. They behave almost in a similar way as of \( C_{int} = 0.27 \) kg/kg.

![Schematic of the interface zone when \( C_i = 0.0 \) kg/kg](image)

**Fig. 5.4** Schematic of the interface zone when \( C_i = 0.0 \) kg/kg
Chapter 6

DEPENDENCY OF TOTAL ABSORBED MASS

6.1 DEPENDENCY OF MASS ABSORPTION

The four cases of solutions with different $C_i = 0.0, 0.27, 0.46$ and $0.63$ kg/kg are examined with six different pressure differences $\Delta P_i$. The results show that the obtained total absorbed mass of ammonia vapor, $m_a$, is a function of initial concentration $C_i$, initial pressure difference $\Delta P_i$, and time $t$, this relation can be expressed as the following:

$$m_a = f(\Delta P_i, C_i, t)$$  \hspace{1cm} (6-1)

Thus:

$$dm_a = \left( \frac{\partial m_a}{\partial t} \right)_{\Delta P_i, C_i} dt + \left( \frac{\partial m_a}{\partial C_i} \right)_{\Delta P_i} dC_i + \left( \frac{\partial m_a}{\partial \Delta P_i} \right)_{t, C_i} d\Delta P_i$$  \hspace{1cm} (6-2)

The first term in the right side of Eq. (6-2) represents the dependency of the total absorbed mass of ammonia $m_a$ upon time $t$ at a fixed initial concentration $C_i$ and an initial pressure difference $\Delta P_i$, and this term is illustrated in Figs. 4.2 (a) through (d) in Chapter 4. The second term in the right side of Eq. (6-2) represents the dependency of the total absorbed mass of ammonia $m_a$ upon $C_i$ at fixed initial pressure difference and time, is explained in Issa et al. [24]. On the other hand, the third term in the right side of Eqs. (6-1) and (6-2) describe the dependency of the total absorbed mass $m_a$...
upon initial pressure difference $\Delta P_i$ at fixed values of the initial mass fraction $C_i$ and time $t$. To illustrate the significance of the third term, several time steps were selected between 1 sec from starting and 600 sec at ending the experiment, a total of twenty six times. Figures 6.1 (a) through (d) for example show the total absorbed mass vs. $\Delta P_i$ at 5, 20, 50, and 100 sec.
Fig. 6.1 Total absorbed mass vs. initial pressure difference in the duration of absorption. (a) $C_i = 0.0 \text{ kg/kg}$ (continued)
Fig. 6.1 Total absorbed mass vs. initial pressure difference in the duration of absorption. (d) $C_i = 0.63 \text{ kg/kg}$
As can be observed from Figs 6.1 (a) through (d), a linear relationship is held for the total absorbed mass $m_a$ vs. $\Delta P_i$, and the straight lines in the figures are the linear fit of the absorbed mass $m_a$. The total absorbed mass of ammonia per unit area which obtained experimentally at $C_i = 0.0 \text{ kg/kg}$ is denoted by $m_0$. Thus, the approximation of $m_0$ as a function of $\Delta P_i$ at constant initial concentration is:

$$m_0 = a_0(t) + a_1(t)\Delta P_i$$  

(6-3)

where $a_0$ and $a_1$ are time variable coefficients and their approximations are given by Eqs. (6-4) and (6-5). The coefficients in these two equations were determined by the least-square method to obtain the best fit for all measured data in Fig. 6.1 (a). As shown in Figs 6.2 (a) and (b).
Fig. 6.2 Approximation of coefficients $a_0$ and $a_1$
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DEPENDENCY OF TOTAL ABSORBED MASS

When \( N \geq 2 \), the higher order terms almost have no effect on the result of Eqs. (6-4) and (6-5), and therefore, the value of \( N \) was selected to be 2.

\[
a_0(t) = \sum_{i=0}^{N} a_{0i} t^{i/2}, \quad [N=2]
\]

\[
a_1(t) = \sum_{i=0}^{N} a_{1i} t^{i/2}, \quad [N=2]
\]

where \( a_{0i} \) and \( a_{1i} \) are constants and their values are listed in Appendix I Table I.1. Hence Eq.(6-3) becomes:

\[
m_0 = \left( \sum_{i=0}^{N} a_{0i} t^{i/2} \right) + \left( \sum_{i=0}^{N} a_{1i} t^{i/2} \right) \Delta P_i
\]

It may be worth mentioning that the coefficients of \( a_{00} \) and \( a_{10} \) become \( a_{00} = a_{10} = 0 \), which correspond to \( m_0 = 0 \) at the start of absorption process.

The total absorbed mass \( m_a(\Delta P_i, C_i, t) \) obtained experimentally at the four different initial concentrations, is divided by \( m_0(\Delta P_i, t) \) to obtain the proportion of \( m_a \) to \( m_0 \) which is presented by the vertical axis in Fig. 6.3, where the horizontal axis presents the initial concentration \( C_i \).
Figure 6.3 shows that the proportion $m_a/m_0$ is presented by a curve which can be fitted by the following exponential decay.

$$\frac{m_a(\Delta P_i, C_i, t)}{m_0(\Delta P_i, t)} = e^{-C_i/H}$$  \hspace{1cm} (6-7)

where $H = 0.25$ is a constant. Combining Eqs. (6-6) and (6-7) gives;

$$m_a = \left[ \left( \sum_{i=0}^{N} a_{0i} t^{i/2} \right) + \left( \sum_{i=0}^{N} a_{1i} t^{i/2} \right) \Delta P_i \right] e^{-4C_i}$$  \hspace{1cm} (6-8)
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DEPENDENCY OF TOTAL ABSORBED MASS

Equation (6-8) allows us to calculate the total mass absorption of ammonia per unit area at any time up to 600 sec from starting the absorption and for any value of \( \Delta P \) and \( C_i \), under the current experimental conditions. It should be noticed that the value of \( H = 0.25 \) is almost identical to the \( H \) value, \( H=0.26 \), in Eq. (1-1) which is derived by Issa et al [24]. This fact seems to mean that the absorption independently occurs between initial pressure difference and initial concentration.

To evaluate the accuracy of the prediction by the derived correlation, Eq.(6-8), with the experimental data, relative standard deviation \( \sigma \) is generally used:

\[
\sigma = \sqrt{\frac{1}{N} \sum_{j=1}^{N} \left[ 1 - \left( \frac{m_a}{m_{a,pred}} \right)_j \right]^2}
\]

(6-9)

where \( m_{a,pred} \) is the total absorbed mass that is predicted by Eq.(6-8), \( m_a \) is the total absorbed mass of ammonia obtained experimentally, and \( N \) represents the number of the data until the considered time \( t \). Figure 6.4 (a) shows the relative standard deviation over the range of 0 to 120 sec, where Fig. 6.4 (b) shows the relative standard deviation of the data from 200 to 600 sec. The relative standard deviation for \( C_i = 0.63 \) kg/kg is removed from Figs. 6.4 (a) and (b) due to its high values with uncertainties compared with other initial concentrations. These large values are caused by the small amount of total absorbed mass at \( C_i = 0.63 \) kg/kg. However, based on Figs 6.4 (a) and (b), the Eq. (6-8) is recommended to predict the total absorbed mass of ammonia vapor until 200 sec from starting the absorption.
Fig. 6.4 Relative standard deviation of the predicted and experimentally obtained total absorbed mass of ammonia
On the other hand, the developed equation, Eq. (6-8), is compared with Eq. (1-1), and Fig. 6.5 shows the comparison for $C_i = 0.0$ and 0.27 kg/kg at $\Delta P_i = 50$ kPa. Both correlations are fairly good agreement, although the standard deviation of both equations is 12% over the range of 0 to 120 sec.

![Figure 6.5 Predicted value of $m_a$ by Eqs. (1-1) and (6-8)](image)

Fig. 6.5 Predicted value of $m_a$ by Eqs. (1-1) and (6-8)
6.2 FREE ABSORPTION AT $\Delta P_i = 0$ kPa

Absorption process also takes place under the condition of no pressure difference between the ammonia vapor cylinder and ammonia water solution in the test cell, whereas the driving force for the diffusion is the temperature and the concentration. This free absorption can be estimated by two ways, the first one is based on experimental result, by extending the linear relationship of the mass absorption to $\Delta P_i = 0$, the second one is based on theoretical estimation.

6.2.1 Free absorption estimated from the experimental result

As mentioned above, the total absorbed mass of ammonia per unit area was taken against the initial pressure difference $\Delta P_i$, at fifteen selected times between 1 and 600 sec. Figures 6.1 (a) through (d) show the value of absorbed mass $m_a$ against $\Delta P_i$ at four selected times, 5, 20, 50, and 100 sec. The total absorbed mass of ammonia is increasing proportionally with an increase in $\Delta P_i$, and it is characterized by a linear function of $\Delta P_i$. By extending the straight line for any selected time until it intersects with the vertical axis where $\Delta P_i = 0$ kPa, we would obtain an estimate of the total absorbed mass when there is no initial pressure difference, as shown in Figs 6.6 (a) and (b).
Fig. 6.6 The extended lines of total absorbed mass vs. initial pressure difference
The free absorptions in the cases of 0.0 and 0.27 kg/kg are shown in Fig. 6.7. In the case of $C_i = 0.46$ kg/kg, the absorbed mass during the free absorption becomes $m_a(0,t) = 0.72, 1.52, 3.15$ and 5.7 g/m$^2$ accordingly as $t = 5, 20, 50$ and 100 sec. It is noticeable that the free absorption for $C_i = 0.46$ kg/kg is much smaller than that of $C_i = 0.0$ and 0.27 kg/kg. In addition, for the $C_i = 0.63$ kg/kg, the absorbed mass almost becomes $m_a(0,t) = 0$ at any time. This means there is no free absorption in the case of $C_i = 0.63$ kg/kg.

![Fig. 6.7](image)

Fig. 6.7 The free absorption obtained in cases of $C_i = 0.0$ and 0.27 kg/kg

It can be noted that the low initial concentration produces a high level of free absorption. The driving force for the absorption process is only the initial concentration. The affinity between ammonia vapor and water is responsible for molecules of ammonia being absorbed into the solution and diffusion process occurs when ammonia molecules spontaneously move from the vapor at high concentration, into a solution at low concentration. In the case of high initial concentration, $C_i = 0.63$ kg/kg, there is no free absorption, as the initial concentration is not sufficient to cause the absorption process.
6.2.2 Theoretical estimation of free absorption

Total absorbed mass of ammonia under the condition of no pressure difference between the ammonia vapor and ammonia water solution, can be obtained theoretically from the one dimensional governing equation for diffusion process without any convective effect as:

$$\frac{\partial C}{\partial t} = D \frac{\partial^2 C}{\partial z^2}$$  \hspace{1cm} (6-10)

where $D$ is the mass diffusivity and $z$ is depth in the liquid, the total absorbed mass for a semi-infinite case can be given as:

$$\dot{m}_a = \int_0^t \rho \frac{\partial C}{\partial Z} \bigg|_{z=0} dt$$ \hspace{1cm} (6-11)

Hence,

$$\dot{m}_a = \rho_L \frac{2}{\sqrt{\pi}} \sqrt{Dt} (C_{\text{int}}(t) - C_i)$$  \hspace{1cm} (6-12)

where $\dot{m}_a$ is the rate of absorbed mass, $\rho_L$ is the density of the solution, the mass diffusivity $D$ has been found ranging from $2 \times 10^{-9}$ to $5 \times 10^{-9}$ m$^2$/s [22], $C_i$ is the initial concentration, and $C_{\text{int}}(t)$ is the concentration on the interface when $\Delta P_i = 0$ kPa.

6.2.2.1 Interface concentration at $\Delta P_i = 0$ kPa

The interface concentration at $\Delta P_i = 0$ kPa is required to solve Eq. (6-12), It can be obtained from the estimated interface concentration. In Chapter 5 the estimated interface concentration was discussed, and Figure 5.3 shows the estimated interface concentration in the cases of $C_i = 0, 0.27, 0.46$, and $0.63$ kg/kg for all initial pressure differences $\Delta P_i$. 

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In the case of \( C_i = 0.27 \text{ kg/kg} \), the interface concentration starts to increase sharply within 50 seconds, and then it reaches equilibrium. The interface concentration before the equilibrium stage can be expressed by equation (6-13). The reason why a half polynomial function of time is chosen is that the fundamental solution can be constituted by a half polynomial one as can be seen in Ref. [32].

\[
C_{\text{int}}(t) = b_0 + b_1 t^{1/2} + b_2 t
\]

(6-13)

where \( b_0 \), \( b_1 \) and \( b_2 \) are constants, can be calculated by differentiating Eq. (6-13) with respect to time, given initial conditions are \( t = 50 \) sec and \( C_{\text{int}} = 0.3 \text{ kg/kg} \). The values of constants \( b_0 \), \( b_1 \) and \( b_2 \) are shown in Appendix I, Table I.2.

In the equilibrium stage, after 50 sec from starting the experiment, the interface concentrations up on \( \Delta P_i \) kg/kg for 50, 98, and 130 sec are shown in Fig 6.8. The data is presented by a linear equation, the interface concentration at \( \Delta P_i = 0 \text{ kPa} \) can be obtained by extending the straight lines to \( \Delta P_i = 0 \text{ kPa} \) as shown in Fig. 6.8.

![Equilibrium interface concentration at \( \Delta P_i = 0 \text{ kPa} \) when \( C_i = 0.27 \text{ kg/kg} \)](image)

**Fig. 6.8** Equilibrium interface concentration at \( \Delta P_i = 0 \text{ kPa} \) when \( C_i = 0.27 \text{ kg/kg} \)
In the case of $C_i = 0.27$ kg/kg, the equilibrium concentration at $\Delta P_i = 0$ kPa is 0.3 kg/kg. However, Fig. 6.9 shows the profile of the interface concentration at $\Delta P_i = 0$ kPa in the case of $C_i = 0.27$ kg/kg.

![Graph showing interface concentration at $\Delta P_i = 0$ kPa for $C_i = 0.27$ kg/kg](image)

**Fig. 6.9** Interface concentration of $C_i = 0.27$ kg/kg at $\Delta P_i = 0$ kPa

According to Fig. 5.3, the interface concentrations when $C_i = 0.46$ and 0.63 kg/kg hardly change with time. The interface concentrations vs. $\Delta P_i$ in the cases of $C_i = 0.46$ and 0.63 kg/kg at 10, 50 and 100 sec, are shown in Figs. 6.10 (a) and (b), respectively. The interface concentrations at $\Delta P_i = 0$ kPa, which obtained by extending the fitted lines to the vertical axis, is found to be 0.463 kg/kg roughly larger than the initial concentration in $C_i = 0.46$ kg/kg. In the case of $C_i = 0.63$ kg/kg the interface concentrations at $\Delta P_i = 0$ kPa was 0.625 kg/kg, this abnormal value will be neglected, as the interface concentrations at $\Delta P_i = 0$ kPa supposed to be larger or equal to the initial concentration when $C_i = 0.63$ kg/kg.
Fig. 6.10 Interface concentration when $\Delta P_i = 0$ kPa of $C_i = 0.46$ and 0.63 kg/kg
Chapter 6

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The tendency of interface concentration when $C_i = 0.0$ kg/kg for different $\Delta P_i$ does not follow a unique shape. Therefore, it is difficult to obtain the interface concentration at $\Delta P_i = 0$ kPa.

However, by substituting the obtained interface concentration at $\Delta P_i = 0$ kPa in Eq. (6-11), the free absorption can be obtained. In the case of $C_i = 0.63$ kg/kg there is a negligibly small free absorption, as a result of that, the interface concentration at $\Delta P_i = 0$ kPa is almost equal to the initial concentration. Whereas, the free absorption when $C_i = 0.46$ kg/kg is insignificant compared to the case $C_i = 0.27$ kg/kg. Figure 6.11 shows the free absorbed mass calculated by the two methods for the cases of $C_i = 0.27$ kg/kg. As can be seen in Fig. 6.11, the free absorption obtained experimentally has a fairly good agreement with the theoretical one when $D = 5 \times 10^{-9}$ m$^2$/s. The large value of mass diffusion $D$ makes them closer.

![Figure 6.11](image)

**Fig. 6.11** The free absorption obtained experimentally and theoretically, in cases of $C_i=0.27$ kg/kg
Chapter 7

INVESTIGATION OF HEAT AND MASS DIFFUSION

7.1 MACH-ZEHNDER INTERFEROMETER

In this experiment a Mach-Zehnder interferometer is implemented to visualize the penetration of ammonia vapor into ammonia water solution. Figure 7.1 illustrates a schematic of the Mach-Zehnder interferometer.

![Schematic of Mach-Zehnder interferometer](image)

**Fig. 7.1** Schematic of Mach-Zehnder interferometer

The Mach-Zehnder interferometer is a particularly simple device for demonstrating interference by division of amplitude. A light beam is first split into two parts by a beamsplitter and then recombined by a second beamsplitter.
Depending on the relative phase acquired by the beam along the two paths the second beamsplitter will reflect the beam with efficiency between 0 and 100%. The operation of a Mach-Zehnder interferometer is often used as an example in quantum mechanics because it shows a clear path-choice problem [33]. Fig. 7.2 shows a photograph of the optical system of the experiment. Preparation of Mach-Zehnder interferometer performed by following the next steps:

- Adjust the mirrors 1 and 2 so that the two beams are superimposed at the reflective surface of the beamsplitter.
- In order to aim for two plane waves nearly parallel, a lens in the interferometer output is placed, and the second beamsplitter is adjusted, so both beams are superimposed in the focal plane of the lens. There should be no shear between the two beams.
- Steps 1 and 2 are repeated until there is no shear between the beams.
- Adjust the second beamsplitter to get interference fringes.

In this experiment, the He-Ne laser beam with a diameter of about 1 mm is emitted from the laser unit then transmits through a half-wave plate, which has the function of making the beam linearly polarized. Then the beam’s direction is changed 90° by using a mirror, so that it incidents on a quarter-wave plate, which has the function of circularly polarized. Then the beam passes through a beam expander which releases it with a diameter that expands with distance, through a mirror that changes its direction 90°, the beam then transmits through a collimator lens which allows the beam out of it with a fixed diameter, more than 30 mm. This beam is then split into two beams by a beam splitter before transmitting into the test cell. One of the two beams transmits through the sight glass of the test cell and is called the measured beam. The other beam continues its path through the air and is, therefore, called the reference beam. Both of the beams are united again at a beam splitter to incident finally on a CCD camera to record the produced fringes. Due to the sensitivity of Mach-Zehnder interferometer, it is mounted with the test cell on a special optical table with vibration isolation system.
Fig. 7.2 A photograph of the optical measuring system
7.2 OPTICAL PATH DIFFERENCE (OPD)

Optical path length (OPL) is the product of the geometric length of the path light follows through the system, and the index of refraction of the medium through which it propagates. A difference in optical path length between two paths is often called the optical path difference (OPD). Optical path length is important because it determines the phase of the laser and governs interference and diffraction of light as it propagates. The optical path difference OPD is given as:

$$OPD = n_m L_m - n_{ref} L_{ref}$$  \hspace{1cm} (7-1)

where $L_m$ and $L_{ref}$ are the geometrical distances traveled by the measured and the reference beams, respectively, and $n_m$ and $n_{ref}$ are the refractive indices of the measured and the reference beams, respectively. A series of bright and dark parallel fringes will be observed, if an optical path difference is generated. This pattern of bright and dark fringes is described mathematically as:

$$2m \lambda = 0 \text{, for bright fringes} \hspace{1cm} (7-2)$$

$$2(m+1) \lambda = 0 \text{, for dark fringes} \hspace{1cm} (7-3)$$

where, $m$ is the order of the fringe. Initially, homogeneous mixture exists in the test cell and, therefore, only bright image is observed. This condition is described mathematically as:

$$OPD = (n_0 L_L + 2n_g L_g) - n_{ref} (L_L + 2L_g) = (2m) \frac{\lambda}{2} \hspace{1cm} (7-4)$$

where, $n_0$ is the refractive index of the liquid mixture at the initial condition, $n_g$ is the refractive index of the sight glass, and $L_L$ and $L_g$ are, respectively, the lengths of the solution and the sight glass that the laser beam perpendicularly passes through. For a condition 1 at which one fringe is formed is:

$$OPD = (n_1 L_L + 2n_g L_g) - n_{ref} (L_L + 2L_g) = 2(m+1) \frac{\lambda}{2} \hspace{1cm} (7-5)$$

The change in the refractive index $\Delta n$, which corresponds to the formation of one fringe in the visualized image, can be obtained by subtracting Eq. (7-4) from Eq. (7-5):
$n_1 = n_0 + (1) \frac{\lambda}{L_L} \Rightarrow \Delta n = n_1 - n_0 = \frac{\lambda}{L_L}$  \hspace{1cm} (7-6)

The wavelength of the Red He-Ne laser is $\lambda = 632.8 \times 10^{-6}$ mm and the length of the liquid that the laser beam perpendicularly transmits through is $L_L = 15$ mm, the inner width of the test cell. By substituting the values of $\lambda$ and $L_L$ in Eq. (7-6) we have:

$$\Delta n = \frac{\lambda}{L_L} = \frac{632.8 \times 10^{-6}}{15} = 4.2186 \times 10^{-5}$$  \hspace{1cm} (7-7)

The value that is given by Eq. (7-7) indicates the change in the refractive index $\Delta n$ that corresponds to the forming of one fringe. Refractive index $n$ is a function of the concentration of the solution $C$ through which light transmits, the temperature $T$, and the wavelength of the light $\lambda$:

$$n = f(C, T, \lambda)$$  \hspace{1cm} (7-8)

Since laser light is used in this technique, the wavelength $\lambda$ can be considered constant, hence:

$$n = f(C, T)$$  \hspace{1cm} (7-9)

As Eq. (7-9) states, refractive index $n$ is a function of concentration $C$ and temperature $T$, each of which has its own contribution in the formation and the distribution of the fringes. The total differential of $n$ can be expressed as:

$$n = f(C, T) \Rightarrow dn = \frac{\partial n}{\partial C} dC + \frac{\partial n}{\partial T} dT$$  \hspace{1cm} (7-10)

During the diffusion process, concentration $C$ and temperature $T$ are functions of depth $z$ and time $t$. Thus, at a fixed time $t$, Eq. (7-10) can be written as:

$$n = f(C, T) \Rightarrow dn = \frac{\partial n}{\partial C} \frac{dC}{dz} dz + \frac{\partial n}{\partial T} \frac{dT}{dz} dz$$  \hspace{1cm} (7-11)
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The terms $dC/dz$ and $dT/dz$ represent the concentration and the temperature distributions in the liquid at a fixed time $t$, respectively. The dependency of refractive index $n$ upon concentration $C$ and the temperature $T$, represented by the terms $\partial n/\partial C$ and $\partial n/\partial T$, are needed to implement Eq. (7-11). Figure 7.3 shows the dependency of the refractive index $n$ of ammonia water mixtures upon ammonia mass fraction $C$ at a fixed temperature of 24 °C [34]. The dependency of the refractive index $n$ upon temperature $T$ is not available. However, with the help of Fig. 7.3 the change in the refractive index $\Delta n$, which is caused by a concentration change $\Delta C$, is tentatively calculated at value of ammonia mass fraction $C_i = 0.59$ kg/kg. By the calculated value along with the value provided by Eq. (7-7), the concentration change, $C_o$, which is resulted from one fringe formed in the visualized image, can be calculated as follows:

$$\frac{\Delta C}{f} = C_o$$  \hspace{1cm} (7-12)

For the selected case of $C_i = 0.59$ kg/kg, the value of $C_o$ is found to be $1.52 \times 10^{-3}$ kg/kg. This value is considered as a reference to which the analysis is made.

![Fig. 7.3 Refractive index of ammonia solution at 297 k [34]](image)

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7.3 CONCENTRATION DISTRIBUTION

7.3.1 Theoretical solution

The one-dimensional governing equation for the diffusion with initial and boundary conditions is given as:

\[ \frac{\partial C}{\partial t} = D \frac{\partial^2 C}{\partial z^2} \]  

(7-13)

where, \( D \) is the mass diffusivity. Before starting the experiment, the mass fraction of ammonia solution \( C_i \) is constant. The boundary condition is changing with time during the absorption process. The mass fraction at the interface \( C_{\text{int}}(t) \), which is explained in Chapter 5, is time variable. An approximation function is needed to analytically solve Eq. (7-13), and the following polynomial square root of time has been adopted:

\[ C_{\text{int}}(t) = \sum_{i=0}^{N} r_i t^{1/2}, \quad N = 2 \]  

(7-14)

The number of terms, \( N \), in Eq. (7-14) should be limited as small as possible to avoid complexity in the solution. Fig. 7.4 shows the approximation of \( C_{\text{int}}(t) \) within the time interval 10-600 sec.
By using the boundary condition which is described by Eq. (7-14), the Eq. (7-13) can be solved as presented in Appendix II, Eq. (II-1). The mass diffusivity $D$ is required to obtain the theoretical concentration distribution. The experimental work of Kojima et al. [35] showed $D$ values in the range of $3.3 \sim 4 \times 10^{-9}$ m$^2$/s in mixtures of ammonia mole fraction ranging from 0.35 to 0.7. The molecular dynamic simulation work of Ferrario et al. [22] provided $D$ values in the range of $2 \sim 5 \times 10^{-9}$ m$^2$/s for the complete range of ammonia mole fraction. Figures 7.5 shows the concentration distribution in the case of $C_i = 0.59$ kg/kg, obtained by the theoretical solution, Eq. (II-1) in Appendix II, at 40 and 360 sec.
7.3.2 Fringes analysis

Before starting the experiment, the solution in the test cell is homogeneous and, consequently, there are no fringes in the visualized images, as can be seen in the left side of Fig. 7.6. When the experiment is started, the ammonia vapor flows into the test cell where it reaches the interface and undergoes a chemical reaction with water molecules through which a new species is formed and, simultaneously, heat is released. This leads to changes in the density of the liquid mixture due to the changes in the mass fraction $C$ and the temperature $T$. Therefore, the refractive index of the liquid changes as Eq. (7.9) indicates and, necessarily, optical path differences, $OPD$, are generated as Eq. (7.1) states, which result in forming fringes that propagate in the liquid with time.
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Initial condition  
Homogenous solution  

Fringes distribution  
t = 40 sec  

Fringes distribution  
360 sec  

Fig. 7.6 Parallel and horizontal fringes apparent in the liquid after starting the experiment

Figure 7.6 shows the visualized fringes at the initial condition, and two different times, 40 and 360 sec from starting the experiment. The lowest bright area in the liquid, where mass fraction has the initial value of $C_i$, is assigned by $fr_0$. Each depth corresponds to the middle of the bright area between each two successive dark fringes, is a depth of fringe, so that, the fringes from $fr_0$ are $fr_1, fr_2, fr_3...$ up to the closest fringe to the interface. The fringes distribution can be analyzed to obtain the concentration distribution with the help of Eq. (7-12). The concentration at any fringe $fr_j$, is given as:

$$C = C_j + \sum_{j=0}^{N} jC_o$$  \hspace{1cm} (7-15)

where, $j$ is the counter of the fringes starting from 0, $N$ represents the total number of fringes at the considered time, and $C$ is the concentration at the fringe. The value of $C_o$ is calculated by Eq. (7-12), it is includes the effect of concentration upon the formation and the propagation of the fringes, and the effect of temperature. Figure 7.7 shows the concentration distribution, obtained by fringes analysis, in the case of $C_i$=0.59 kg/kg, after 40 and 360 sec from starting the experiment.
7.3.3 Simultaneous mass and heat transfer

Figure 7.7 shows the concentration distribution by the fringe analysis, it is high mass diffusivity compared with that obtained by the theoretical solution, which shown in Fig. 7.6. This indicates the heat greatly influences the forming of the fringes within short time from starting the absorption, \( t = 40 \text{ sec} \). In this experimental work, the mass and heat diffuse simultaneously in the solution and, as it is known, thermal diffusivity in ammonia water mixtures is in the order of \( 10^{-7} \text{ m}^2/\text{s} \), while mass diffusivity is in the order of \( 10^{-9} \text{ m}^2/\text{s} \), which means that thermal diffusivity is 100 times larger than mass diffusivity. In other words, the penetration depth of thermal diffusion is nearly 10 times larger than that of mass diffusion since the mathematical solution is composed of the square root of the diffusivity. Based on this fact, the fringes distribution can be analyzed by relying on the square root of Lewis number:

\[
\sqrt{Le} = \sqrt{\frac{\alpha}{D}}
\]  

(7-16)
Where, $D$ is the mass diffusivities in the liquid, and $\alpha$ is the thermal diffusivity, which can be calculated from the following equation:

$$\alpha = \frac{k}{\rho_L C_p} \quad (7-17)$$

where, $k$ is the thermal conductivity, $\rho_L$ is the density and $c_p$ is the specific heat of the solution. In the case of $C_i = 0.59$ kg/kg, thermal diffusivity is found to be $1.51 \times 10^{-7}$ m$^2$/s. By substituting the value of thermal diffusivity $\alpha$ and the two selected values of mass diffusivity $D$, $2 \times 10^{-9}$ and $5 \times 10^{-9}$ m$^2$/s, in Eq. (7-16) we have the values 8.7 and 5.5, respectively. The calculated value 8.7 means that the depth propagated by temperature is 8.7 times larger than that propagated by concentration when mass diffusivity is $D = 2 \times 10^{-9}$ m$^2$/s, and it is 5.5 times larger when $D = 5 \times 10^{-9}$ m$^2$/s. The difference between the speed of propagate mass diffusivity and thermal diffusivity denotes that fringes will be divided into two layers and the boundary between them is the position at which mass diffusion reaches at a time $t$ that is given by:

$$z_C = 3.64\sqrt{Dt} \quad (7-18)$$

As a result, the layer in which mass diffusion contributes in the forming of the fringes becomes $z \leq z_C$ starting from the interface. In this layer, mass and heat superpose during the diffusion process. The layer that is beneath the boundary line $z > z_C$, is characterized with pure thermal diffusion where mass does not penetrate it. To determine the depth that is propagated by temperature, $z_T$ until the considered time, we use the following equation which is derived from the basic solution of diffusion problem:

$$z_T = 3.64\sqrt{\alpha t} \quad (7-19)$$
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Theoretical $D = 2 \times 10^{-9} \text{ m}^2/\text{s}$

Theoretical $D = 5 \times 10^{-9} \text{ m}^2/\text{s}$

Fringes, $(Le)^{0.5} = 8.7$

Fringes, $(Le)^{0.5} = 5.5$

$C_i = 0.59 \text{ kg/kg}$

$t = 40 \text{ sec}$

$C [ \text{ kg/kg}]$

Depth, $z \ [\text{ mm}]$

(a)

$C [ \text{ kg/kg}]$

Depth, $z \ [\text{ mm}]$

$C_i = 0.59 \text{ kg/kg}$

$t = 360 \text{ sec}$

(b)

Fig. 7.8 Concentration distribution in the liquid
Figure 7.8 (a) shows the concentration distributions after 40 sec from starting the experiment, \( z_T \) is found to be 8.94 mm, beneath which initial temperature exists and thus, the depth \( z_T \) is referred to first fringe appears from starting the experiment. The depth that is propagated by concentration, \( z_C \), can be obtained from Eq. (7-18). The so obtained depths of the concentration propagation are 1.03 and 1.62 mm at \( D = 2 \times 10^{-9} \) and \( 5 \times 10^{-9} \) m\(^2\)/s, respectively. The two values of \( z_C \) are represented by the two horizontal dot lines when \( D = 2 \times 10^{-9} \). The interpretation of these two lines is that each of them represents a boundary between two layers. The layer that is formed by heat diffusion and the layer that formed by simultaneous mass and heat diffusion, as can be seen in Fig. 7.9.

**Fig. 7.9** Simultaneous mass and heat diffusion in the liquid
Mathematically, the first term in the right side of Eq. (7-11), $dC/dz$, is 0 which indicates that only temperature is responsible for the forming of the fringes in this fast layer. On the other hand, the layer that is above the boundary line which is formed by concentration and temperature superpose during the diffusion process and each of them has its own contribution in the forming and the propagation of the fringes. In other words, none of the two terms in the right side of Eq. (7-11), $dC/dz$ and $dT/dz$, is 0. Heat effect on the fringes is big at short time from starting the absorption, 40 sec, in consequence of that, there is no agreement between the concentration distribution obtained theoretically and that by fringe analysis.

Figure 7.9 (b) shows the concentration distributions after relatively long time from starting the absorption, $t = 360$ sec, that are obtained by the theoretical solution and by the fringe analysis. Clearly there is no agreement between the concentration distribution obtained by fringe analysis and that obtained by the theoretical solution. This indicates that heat still influences the fringes even after relatively long time from starting the absorption, $t = 360$ sec. Therefore, based on the square root of Lewis number, Eq. (7-16), the position of the boundary line between the two distinguished layers can be determined, the depth that is propagated by temperature is $Z_T = 26.84$ mm. Thus, by implementing Eq. (7-18) the depths propagated by concentration $z_c$ can be calculated, which are found to be 3.09 and 4.88 mm corresponding to the two values of mass diffusivity $D=2\times10^{-9}$ and $5\times10^{-9}$ m$^2$/s, respectively. These two values of $z_c$ are shown by the horizontal dot line when $D=2\times10^{-9}$ in Fig. 7.8 (b).
Chapter 8

CONCLUSIONS

An experimental study about the absorption of ammonia vapor into ammonia water solution has been carried as presented in this dissertation. In this study, the effect of initial pressure difference on absorption process has been investigated by carrying experiments with different initial pressure difference and initial concentration. Absorption process was visualized by Mach-Zehnder interferometer, and the obtained fringes were analyzed to get the concentration distribution together with that obtained by the theoretical solution in which ammonia concentration at the interface was not constant with time. With respect to the declared objectives in Chapter 1 and to the measurements and the analysis results, the following conclusions are adopted:

1. By using the Monde’s inverse solution, IHCP, the liquid-vapor interface temperature and concentration could be estimated.

2. The behavior of interface concentration, $C_{int}$, when $C_i = 0.0$ kg/kg for the cases $\Delta P_i = 250$ and 300 kPa, exhibits a similar tendency of $C_i = 0.27$ kg/kg. The concentration $C_{int}$ is increasing within about 50 sec and then it is hardly changes with time.

3. Total absorbed mass of ammonia vapor per unit area dramatically increases with decreasing initial concentration.
4. Total absorbed mass of ammonia linearly increases with increasing the initial pressure difference between the test cell and the ammonia vapor cylinder.

5. A correlation was derived to calculate the total absorbed mass of ammonia per unit area from the start of the absorption process until 600 sec for any value of the initial pressure difference and the initial concentration under the current experimental conditions (see table 2.1).

6. Absorption process occurs under no initial pressure difference and the absorbed mass could be estimated.

7. The free absorbed mass calculated by experimental method for the cases of $C_i = 0.27$ kg/kg has an agreement with the theoretical one when $D = 5 \times 10^{-9}$ m$^2$/s.

8. The optical observation of the diffusion process, when $C_i = 0.59$ kg/kg, shows two layers. The first fringes in appearance constitute a layer of pure thermal diffusion. The following fringes constitute a layer that is characterized with superposition of mass and heat diffusions.

9. At short times from starting the absorption, heat greatly affects the fringes propagation but with the lapse of time, heat effect becomes gradually small and, therefore, mass diffusion dominates at the slow fringes propagation layer.

10. In the case of $C_i = 0.59$ kg/kg and after 360 sec from starting the absorption, the concentration distribution obtained by the fringe analysis agrees with that obtained by the theoretical solution when mass diffusivity $D = 5 \times 10^{-9}$ m$^2$/s where heat has a small effect on the fringes distribution at the considered time.
REFERENCES


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REFERENCES


**APPENDICES**

**APPENDIX I**

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**Table I.1** Constants of Eqs. (6-4) and (6-5)

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<th>$b_0$</th>
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**Table I.2** Constants of Eq. (6-12)
Solution of Eq. (7-13), which is related to Chapter 7, at the following specified
Initial and boundary conditions:
I.C: \( t = 0, \ C = C_i = \text{const} \).
B.C: \( z = 0, \ C = C_{\text{int}} = r_0 + r_1 t^{0.5} + r_2 t \)
\( z = \infty, \ C = C_i = \text{const} \).
The solution of Eq. (7-13) is given as:

\[
C = (r_0 - C_i) \text{erfc} \left( \frac{z}{2\sqrt{Dt}} \right) + r_i \Gamma(1.5) \left[ \frac{2\sqrt{t}}{\sqrt{\pi}} e^{-\frac{z^2}{4Dt}} - \frac{z}{\sqrt{D}} \text{erfc} \left( \frac{z}{2\sqrt{Dt}} \right) \right] + \\
\frac{r_2 t}{2D} \left( 1 + \frac{z^2}{2Dt} \right) \text{erfc} \left( \frac{z}{2\sqrt{Dt}} \right) - \frac{z}{\sqrt{\pi Dt}} e^{-\frac{z^2}{4Dt}} \right) + C_i
\]

(Eq. II.1)

The coefficients \( r_j \) are listed in Table II.1

<table>
<thead>
<tr>
<th>Coefficients ( r_j )</th>
<th>Time interval [sec]</th>
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Table II.1 Coefficients \( r_j \) of Eq. (7-13)