# SANDIA REPORT

# The Solubility of Gases in NaCl Brine and a Critical Evaluation of Available Data

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# THE SOLUBILITY OF GASES IN NaCl BRINE

# AND A CRITICAL EVALUATION

# OF AVAILABLE DATA


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# ABSTRACT

Knowledge of the solubility of gases in pure water and sodium chloride brine is required for the evaluation of gas-liquid behavior associated with numerous processes in the chemical, biological, and geological sciences and in chemical engineering. Modelling of multiphase flow and transport processes used to simulate fluid behavior in geological media requires suitable and accurate gas solubility data, especially when applied to potential nuclear waste repositories such as WIPP. This report contains a critical evaluation of data available in the literature and provides compiled data sets for the solubility of hydrogen, nitrogen, methane, and carbon dioxide gases in pure water and in sodium chloride brine. Mole fraction gas solubilities are presented as functions of temperature and pressure. To evaluate the influence of electrolyte content of the aqueous solution upon the gas solubility, Sechenov coefficients are provided for several concentrated sodium chloride solutions. An estimate of the solubility of hydrogen in 5 N sodium chloride brine as a function of pressure exhibits a solubility at 40 MPa that is one fifth the value observed for pure water at the same pressure.

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# 1 INTRODUCTION

## 1.1 Motivation

Quantitative investigations of the solubility of gases in liquids have been reported in the literature as far back as the early nineteenth century. Even today, with improved experimental techniques and theories, studies of gas solubility in a variety of complex chemical systems are quite common in the literature. Because of the fundamental aspect of the interaction of gas with liquids, it is not surprising that the solubility of gases in liquids is of widespread interest to researchers in numerous scientific and engineering fields. The significance of water and aqueous solutions as solvents for gases has been of foremost concern. The ability of aqueous solutions to absorb gases is of fundamental interest in numerous biological and ecological processes. Chemical engineers rely considerably upon gas solubility studies for the design of industrial equipment for two phase systems. And, the solubility of gases in natural aqueous solutions is of vital concern in many of the sub-fields of earth science.

Gas solubility data are required for evaluating the behavior of complex fluids in geologic media. Petroleum reservoir engineers strive for the efficient recovery of valuable hydrocarbons from the gas saturated fluid reservoirs. Modelling of multiphase flow and transport of these fluids, so critical for optimizing hydrocarbon recovery, requires the input of gas solubility data. Mediation of contaminants in

groundwater and soils also rely on the modelling of multiphase flow with the incorporation of gas solubilities. Understanding the partitioning of gas between the aqueous solution and the vapor phase as a function of temperature, pressure, and composition is crucial for the accurate evaluation of multiphase flow and transport in geologic media (Parker, 1989).

## 1.2 Gas Concerns at WIPP

Recent scientific concerns for the safe isolation of nuclear wastes in geologic media have reemphasized the significance of gases in modifying the transport of contaminated waste beyond the bounds of a repository. The role of gas generation and its effect upon two phase flow is being investigated by researchers associated with the Waste Isolation Pilot Plant (WIPP) of southeastern New Mexico (see Lappin and Hunter, 1989). Accurate values of the solubility of several gases are required for the realistic modelling of two phase flow in the repository horizon of the bedded salt deposits of the Salado Formation.

Decomposition of waste material at WIPP by microbial activity and the anoxic corrosion of metallic drums will generate primarily hydrogen and carbon dioxide gases (Brush and Anderson, 1989). Nitrogen, methane, carbon dioxide, and some oxygen have been analyzed in samples from drift boreholes through the Salado Formation (D'Appalonia, 1985). Norman and Bernhardt (1982) examined fluid inclusions from the Salado Formation and determined that the associated vapor phase was comprised principally of nitrogen and carbon dioxide. It is therefore important that accurate

solubility data be obtained for these gases in aqueous brine to support the predictive capabilities of two phase flow and transport models.

## 1.3 Organization of Gas Solubility Topics

The primary intent of this paper is to provide a comprehensive review of the solubility of gases in brine including a critical evaluation of available solubility data from the literature. For this purpose, a discussion of background material relating to the thermodynamics and nomenclature of gas solubility is presented in the next section. Also included is a discussion of the role of electrolyte effects upon gas solubility and a summary of the experimental methods used to derive solubility values.

Gas solubility values are presented in the following four sections with the solubility data for the pure water solvent at 0.101325 MPa pressure (1 atm) presented first in Section 3, followed by the data for pure water solvent at elevated pressures in Section 4. Sections 5 and 6 provide the gas solubility data, including Sechenov coefficients, for brine at 0.101325 MPa and at elevated pressures, respectively. Concentrated brine compositions are represented by sodium chloride solutions ranging up to 5 N (molar) solutions (25 weight percent NaCl). Each of the sections includes a discussion of the sources of data and the methods used to compile and curve fit the data. Solubility data for hydrogen, nitrogen, methane, and carbon dioxide are presented in this fashion.

An attempt has been made in this compilation to be as up to date as possible with all pertinent gas solubility studies represented. Nonetheless, in spite of computer searches of the chemical and geological literature, some solubility data may have been overlooked. Extra effort has been made to include the gas data appropriate for application to the two phase transport models for simulation of the WIPP repository. Where the particular temperature, pressure, or compositional ranges have not been adequately covered for a particular gas, other related solubility data proximate to those conditions are presented. Section 7 of this paper presents an approach for estimating the pressure dependence of hydrogen gas solubility in sodium chloride brine.

## 2 THEORETICAL BACKGROUND AND NOMENCLATURE OF GAS SOLUBILITY

### 2.1 Theory

The thermodynamics that dictates the solubility of gases in liquids, either pure water, organic solvents, or concentrated brines, is based on the difference between the free energy of a gas molecule existing in the vapor phase and in the solution phase. Technically, this approach is known as vapor-liquid equilibria, and is commonly referred to as the solubility of a gas. A thermodynamics framework provides a simple, yet elegant, approach for examining not only simple binary component systems but the more complex multi-component gas or solvent systems. Although details of the thermodynamics of gas solubility are beyond the scope of this paper, the rudiments and nomenclature are presented in the following sub-sections. Battino and Clever (1966), Wilhelm et al. (1977), and Wilhelm (1982) provide excellent reviews and discussions of the thermodynamic background of gas solubility.

### 2.2 Units and Notations

The solubility of gases in liquids is of considerable interest to a wide range of scientific and engineering researchers. Because of this multi-disciplinary need of such data there is a wide variety of quantities and units that are observed in the literature for reporting gas solubility. This fact often leads to considerable confusion in

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comparing and evaluating the appropriateness of any particular study. Units for a particular expression of gas solubility are often chosen for convenience rather than by convention based upon previous studies. This confusing choice is usually based upon the particular experimental technique that is employed or the eventual application for which the data is to be utilized. It is therefore not surprising that gas solubility values are often reported in units of standard cubic feet per barrel or pounds per liter of solvent.

This paper, in order to be internally consistent and to conform with the most recent experimental studies, presents all solubility data in terms of mole fraction of the gaseous component in the liquid phase. Interconversion of solubility data to mole fraction units often involved the digitization of poorly resolved graphs when tabulated data were lacking. Numerous computer algorithms were used to reduce the data to units of mole fraction. The final figures which summarize the data provide gas solubility along the ordinate of each plot. The low or ambient pressure data are plotted with mole fraction times a factor of $10^5$ while the high pressure data (up to pressures of 200 MPa) are presented times a factor of $10^3$. This convention reflects the roughly two orders of magnitude general increase of gas solubility at the reported elevated pressures.

The intensive properties, temperature and pressure, are also repor d in the literature with a wide range of units. This paper presents all pressures ' units of pascals, usually as MPa for the elevated pressure studies. One atmosphere of pressure is presented as

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0.101325 MPa whereas one bar of pressure is 0.1 MPa. Temperatures are presented in units of absolute temperature, that is, in Kelvin.

## 2.3  Methods of Expressing Gas Solubility

### 2.3.1  General

As noted above, the multi-disciplinary need for gas solubility data has created a literature base providing numerous methods for expressing the solubility of a gas in a liquid. Generally, these different methods are easily interconverted from one form to another. However, assumptions such as ideal gas behavior or solution density are often required. Because several studies provide only limited experimental details and occasionally fail to provide the rough data, it is often impossible to provide all gas solubility data in terms of mole fraction. A short discussion of the principle methods is presented below.

The primary method of reporting the solubility of a gas in liquid for a binary system is that of mole fraction x:

$$x = \frac{n_g}{n_g + n_l} \tag{1}$$

where $n_g$ and $n_l$ are respectively the number of moles of gas dissolved in the liquid and the number of moles of liquid. If the masses of soluble gas $W_g$ and liquid $W_l$ are obtained by experiment, then the mole fraction solubility is obtained by the following equation:

$$x = \frac{W_g/M_g}{W_g/M_g + W_1/M_1} \quad \quad (2)$$

where $M_g$ and $M_1$ are respectively the molecular weights of the dissolved gas and the liquid. Occasionally, the mole ratio $N$ is utilized to define gas solubility in a binary system:

$$N = \frac{n_g}{n_1} \quad \quad (3)$$

Typically $n_g$ is much less than $n_1$ (for example, at low pressures) and therefore $N$ is roughly equal to $x$.

A mass basis approach for reporting gas solubilities is usually defined in terms of weight percent:

$$Wt \ \% = \frac{100 \ W_g}{W_g + W_1} \quad \quad (4)$$

Additional mass-based methods such as molal solubility (moles of dissolved gas per kilogram of solvent) and weight solubility (moles of dissolved gas per gram of solvent) are also used. A molar solubility (moles of dissolved gas per liter of solution; normality) is occasionally observed.

Another common method of reporting gas solubility is the Bunsen coefficient $\alpha$ which is defined as the volume of gas reduced to a temperature of 273.15 K and a pressure of 1 atmosphere which is absorbed by a unit volume of solvent at a selected temperature and a partial

pressure of 1 atmosphere. The Bunsen coefficient as defined is somewhat inconvenient to use if the gas is nonideal and molar volumes or densities are not available. However, if the gas is assumed to be ideal, the Bunsen coefficient can be stated as follows:

$$\alpha = \frac{V_g}{V_1} \frac{273.15}{T} \tag{5}$$

where $V_g$ is the volume of gas absorbed and $V_1$ is the original volume of the solvent.

The Ostwald coefficient L is defined as the ratio of the volume of the gas dissolved to the volume of the dissolving liquid:

$$L = \frac{V_g}{V_1} \tag{6}$$

The volumes are to measured at the same temperature. If the gas is ideal and only a small amount of gas is absorbed by the liquid, the Ostwald coefficient can be considered to be independent of the partial pressure of the gas.

Other volume-based solubilities are found in the literature. The Kuenen coefficient is the volume of gas reduced to 273.15 K and 1 atmosphere that is dissolved by one gram of solvent with a partial pressure of the gas at 1 atmosphere. Other absorption coefficients are expressed as the volume of gas reduced to 273.15 K and 1 atmosphere that

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is dissolved per unit volume of liquid with a total pressure of 1 atmosphere.

The final method of expressing gas solubility to be discussed here is the Henry's Law constant. It is usually presented as:

$$K_H = \frac{P_g}{x} \qquad (7)$$

where $K_H$ is the Henry's Law constant, $P_g$ is the partial pressure of the gas, and x is the mole fraction solubility of the gas. It is important to recognize that Henry's Law is a limiting law that is formally defined as:

$$K_H = \lim_{x_i \to 0} \left( \frac{f_i}{x_i} \right) \qquad (8)$$

where $f_i$ is the fugacity of the gas with i denoting the particular gas in a multicomponent system. This thermodynamic definition of Henry's Law states that the fugacity of a gas component above a solution is linearly related to the mole fraction concentration of the gas in the solution by the Henry's Law constant as the gas solubility approaches zero. Therefore, it applies to only very dilute solutions and would be appropriate for describing the gas solubility of most gas-water systems. Gas fugacity and partial pressure are often interchangably used at low pressures. Other forms of Henry's Law exist in which gas solubility terms other than mole fraction are used. The fugacity or presure parameter is also often replaced with some other concentration term. It

is therefore important that Henry's Law values be properly defined and that units of all terms be ascertained in order to properly use the Henry's Law solubility.

Battino et al. (1981) presents a detailed discussion of all the common interconverstion methods for gas solubility expressions and provides several helpful conversion formulae. Most of the above solubility expressions are presented for the case of a binary system, that is, the absorption of a pure gas by a single component liquid. Slight modifications to these equations must be made in order to account for more complex solvents such as aqueous brines that are the concern of this study. All mole fraction solubilities presented in this paper are calculated by taking into account all system components.

To unambiguously report gas solubilities no matter which of the above methods is used, it is required that the temperature, partial pressure, and total pressure of the measurement be reported. It is also necessary that details of the experimental apparatus, procedure, and sources of error be discussed. The purity of the gas and liquid used in the experiment should be noted. Impurities associated with the gas are usually the largest source of error. Improper degassing of previously dissolved gases from the solvent prior to reequilibration with the desired gas is another serious error often associated with the solubility experiments.

## 2.3.2 Electrolyte Effects

In order to properly evaluate the role of sodium chloride in modifying the solubility of a gas in aqueous solutions it is important that the concept of "salting-out" and the effect of electrolytes upon solubility be introduced. Salting-out refers to the addition of an electrolyte, or salt, to a liquid, causing the previously dissolved gas to exsolve. This response results from the change in the activity coefficient of the dissolved gas as modified by the added electrolyte.

The activity coefficient of a dissolved gas is a function of all aqueous species present in solution and for a given pressure and temperature can be represented by the following equation (Battino et al., 1981):

$$\log \gamma_i = \sum_m \sum_n k_{mn} \, C_s^n \, C_i^m \tag{9}$$

This equation provides the common logarithm of the activity coefficient of a gas species $\gamma_i$ as a power series of the electrolyte concentration $C_s$ and the solute gas concentration $C_i$ with the interaction parameter $k_{mn}$. For the case of low solubilities when solute interactions are negligible, equation (9) can be restated using just the linear terms:

$$\log \gamma_i = k_s C_s + k_i C_i \tag{10}$$

where $k_s$ is the salt effect parameter, commonly referred to as the Sechenov (or Setschenow) coefficient, and $k_i$ is the solute-solute gas

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interaction parameter. Because the dissolved gas activity is identical in the pure solvent and the electrolyte solution for a given partial pressure of the gas, the following relations hold:

$$a_i = \gamma_i S_i = \gamma_i^o S_i^o \qquad (11)$$

where the activity of the dissolved gas is $a_i$, and $S_i$ and $S_i^o$ are respectively the gas solubility in the electrolyte solution and in the pure solvent, along with their corresponding $\gamma$ terms. Note that solubilities are introduced into this formalism using the general notation of $S_i$ to represent any particular solubility expression, including mole fraction $x_i$. By evaluating the ratio of activity coefficients for the dissolved gas in electrolyte solution to pure solvent using equation (10), the following relations result:

$$\log \frac{\gamma_i}{\gamma_i^o} = \log \frac{S_i^o}{S_i} = k_s C_s + k_i (C_i - C_i^o) \qquad (12)$$

where $C_i^o$ represents the concentration of dissolved gas in the pure solvent. By assuming that the concentration difference for the dissolved gas in the different solvents is negligible relative to the first term (appropriate for gases), this equation reduces to the more familiar empirical Sechenov equation:

$$k_s = \frac{1}{C_s} \log \frac{S_i^o}{S_i} \qquad (13)$$

The form of this equation implies that electrolytes which salt-out, or decrease the gas solubility, will have positive Sechenov coefficients while those that salt-in, or increase the gas solubility, will have negative values. Morrison (1952), Ellis (1959), and Malinin (1978) provide discussions of the complex nature of these phenomena and how they relate to the specific interactions among ions, solute molecules, and solvent molecules. Long and McDevit (1952) discusses the thermodynamic theory of these interactions in evaluating the activity coefficients of the dissolved gas species.

Because of the assumptions required in deriving equation (13), observed gas solubilities do not generally follow the simple form of the Sechenov equation. Gas solute-electrolyte interactions do occur, especially for the case of concentrated electrolyte solutions and brines. Therefore, one can expect to see $k_s$ values derived from equation (13) varying as a function of electrolyte concentration. Nonetheless, the Sechenov equation is used widely as an empirical relation and provides a quantitative measure of the changes in gas solubility as effected by electrolytes.

There is no widely accepted standard for the units of the terms in the Sechenov relation. In keeping with the convention of this paper, the $S_i$ solubility values will be presented in terms of mole fraction. The electrolyte concentration is given as a molar quantity (moles per liter of solution or $moles/dm^3$). The Sechenov coefficients will therefore have units of $dm^3/mole$ and will be denoted as $k_{scx}$. The subscript scx refers to the Sechenov coefficient (s) using molar

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electrolyte concentration (c) and mole fraction gas solubilities (x). Numerous other combinations of units have been reported in the literature for the Sechenov coefficients. For example, $k_{smc}$ represents the Sechenov coefficient (s) based upon units of molal electrolyte concentration (m) and molar gas solubilities (c). Clever (1982) discusses the confusion generated by this practice and provides convenient conversion formula for the purpose of comparing salting-out effects.

Sechenov coefficients presented in the literature were used to generate mole fraction gas solubility curves for the electrolyte solutions. This process requires appropriate gas solubility data for the pure liquid, or water, solvent. Note that electrolyte concentrations presented in molar amounts, either for evaluation of $k_s$ or for calculation of molar gas solubility, usually assume complete dissociation of the electrolyte.

## 2.4 Temperature and Pressure Dependence of Gas Solubility

Of interest to researchers involved in the investigation of gas behavior in a variety of applications, ranging from petroleum reservoir engineering to two phase flow modelling of nuclear waste repositories, is the evaluation of gas solubility as a function of either temperature or pressure or both. Given the available solubility data sets, it is appropriate to fit these data to suitable empirical curves. The temperature dependence of the gas solubility is most critical to the thermodynamicist in that the curve fits to the observed solubility data

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provide useful thermodynamic function information.  Wilhelm et al.
(1977) and Battino et al. (1981) suggest the following form for the
temperature dependence of the solubility:

$$\ln x \; = \; A_0 \; + \; \frac{A_1}{\tau} \; + \; A_2 \ln \tau \; + \; A_3 \tau \qquad (14)$$

where x is the mole fraction solubility, $\tau$ is the temperature in units
of 100 K, and $A_i$ are the fitting coefficients.  In this formalism it is
possible to derive the free energy, enthalpy, entropy, and heat capacity
changes for the transfer of the gas from the vapor phase at a partial
pressure of 0.101325 MPa to the liquid phase (see Battino et al., 1981).
The inclusion of the fourth term in the curve fitting will depend upon
the overall precision and total number of data points.  It is often
ignored in the curve fitting process.

In order to combine the effects of both temperature and pressure
upon the mole fraction solubility Clever and Young (1987) recommends the
use of the following relation:

$$\ln x \; = \; B_0 \; + \; \frac{B_1}{\tau} \; + \; B_2 \ln \tau \; + \; B_3 \ln P \; + \; B_4 P \qquad (15)$$

where P is the partial pressure of the gas and the $B_i$ are the fitting
coefficients.  Again, the highest order term can be ignored if no
significant improvement in the precision of the fit occurs.  Equation
(15) can be simplified for an isothermal system by combining the fitting

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coefficients to produce an empirical fitting relation that relates the gas solubility to partial pressure:

$$\ln x = D_0 + D_1 \ln P + D_2 P \qquad (16)$$

$D_i$ are the fitting coefficients and again the highest order (third) term is optional. The coefficients are related to the difference between the partial molar volume of gas in solution and at infinite dilution (Michels et al., 1936; Wilhelm et al., 1977).

## 2.5  Solubility of Gas Mixtures

Very little research has been performed in evaluating the interaction of gas species when collectively absorbed into solution from a multicomponent vapor. Wilhelm et al. (1977) suggests that mixtures of gases behave more or less independently when dissolved in liquid water. Benson and Parker (1961) conclude from their precision solubility measurements that nitrogen, argon, and oxygen dissolve in water and sea water in proportion to their partial pressures. An example of the solubility of air, that is, nitrogen, oxygen, and inert gases, in water is presented in Glasstone and Lewis (1960). The pure gas solubilities of these gases multiplied by their partial pressures can be used to calculate the solubility of air in water to within one percent of the observed value (see Wilhelm et al., 1977). Being within the limits of Henry's Law behavior, it appears that most gases when dissolved in water or aqueous solution exhibit very weak interactions with the other gas

components in solution.  These gases essentially behave independent of each other while in solution.


## 2.6  Experimental Methods

Although numerous experimental techniques and procedures exist for measuring the solubility of a gas in a liquid, the methods all share the common approach of determining the amount of gas that has equilibrated with a known amount of solution.  This method usually involves either a physical determination of the gas amount where pressure, volume, and temperature values are measured, or a chemical determination where specific gas components are directly analyzed in solution.  Battino and Clever (1966) and Wilhelm et al. (1977) provide excellent reviews of the experimental techniques that have been utilized and discuss their varying degrees of precision.

The most common and simple method of deriving solubility values involves a manometric-volumetric approach whereby volumes of equilibrated gas are derived by examination of the gas-saturated solution.  Alternatively, the equilibrated solution can be extracted and then used to evaluate the pressure, volume, and temperature of liquid and gas.  Direct measurements of the quantities of the components in the gas and liquid phase can also be performed using a variety of other techniques.  Chemical, mass spectrometer, gas and liquid chromotography and other analytical means are often used to measure the amount of gas in solution.

The manometric-volumetric approaches have been used almost exclusively for measurement of the gas solubility values presented in this paper. The determination of nitrogen, hydrogen, methane, and carbon dioxide solubilities is usually amenable to one of the manometric-volumetric approaches. A unique modification of the these more traditional methods is that used by McGee et al. (1981) in their measurement of methane solubility in sodium chloride brines. The experiment involved the determination of the intersection of the liquid-vapor curve with the one phase liquid curve in pressure-temperature space for measured quantities of methane and brine. The derived pressure and temperature represent the equilibrium values for the solubility of the measured amount of methane in the fixed volume of brine. The results of the study by McGee et al. (1981) are presented in Section 6.3.

# 3  GAS SOLUBILITY IN PURE WATER

## 3.1  Introductory Remarks

An extensive compilation of the gas solubility data from the literature for hydrogen, nitrogen, methane, and carbon dioxide are presented in this and the next three major sections.  Smooth curves are presented in the figures for the mole fraction solubility, Henry's Law constant, and Sechenov coefficients for these gases as functions of temperature and pressure.  Each of the curves, unless stated otherwise, represents a regression of the available experimental data to a third or fourth order polynomial.  The data were processed in this manner for convenience of graphing although a similar fit using equation (14) or equation (16) could have been presented.

## 3.2  Hydrogen

A critical evaluation of the 0.101325 MPa solubility of hydrogen in pure water is provided by Young (1981) in which 75 data points from ten different studies (including Morrison and Billet, 1952; Crozier and Yamamoto, 1974) were used to produce a regression curve fit to the temperature dependence equation given by equation (14).  Figure 1, which includes this curve plus the smooth curve fits for nitrogen, methane, and carbon dioxide, and Figure 2 provide the results of the regression analysis.  The fitted curve for hydrogen solubility displays a distinct minimum at 327 K.  Figure 2 also provides the corresponding Henry's Law

SOLUBILITY IN PURE WATER
at 0.101325 MPa partial pressure
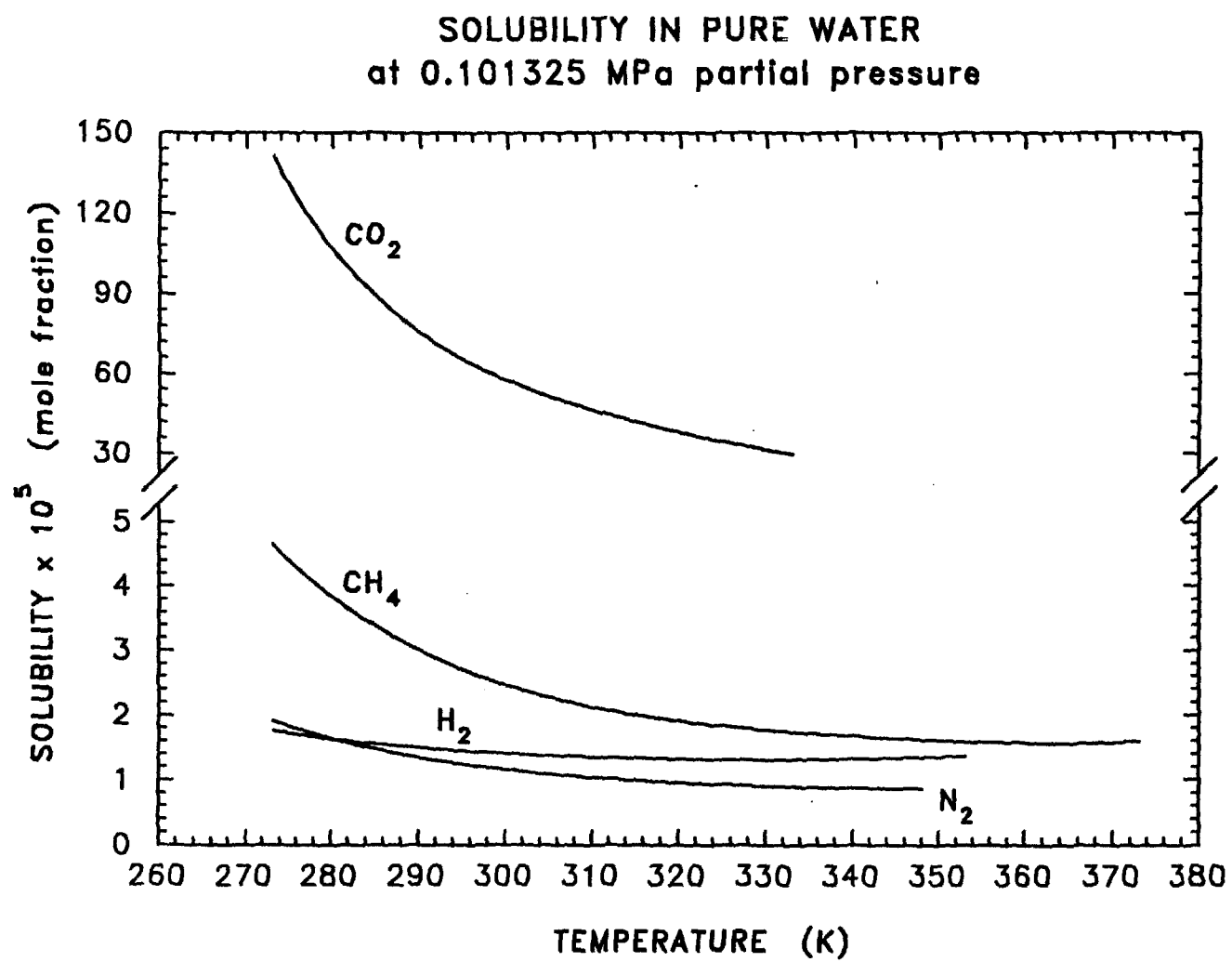
Figure 1

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HYDROGEN SOLUBILITY IN PURE WATER
at 0.101325 MPa partial pressure
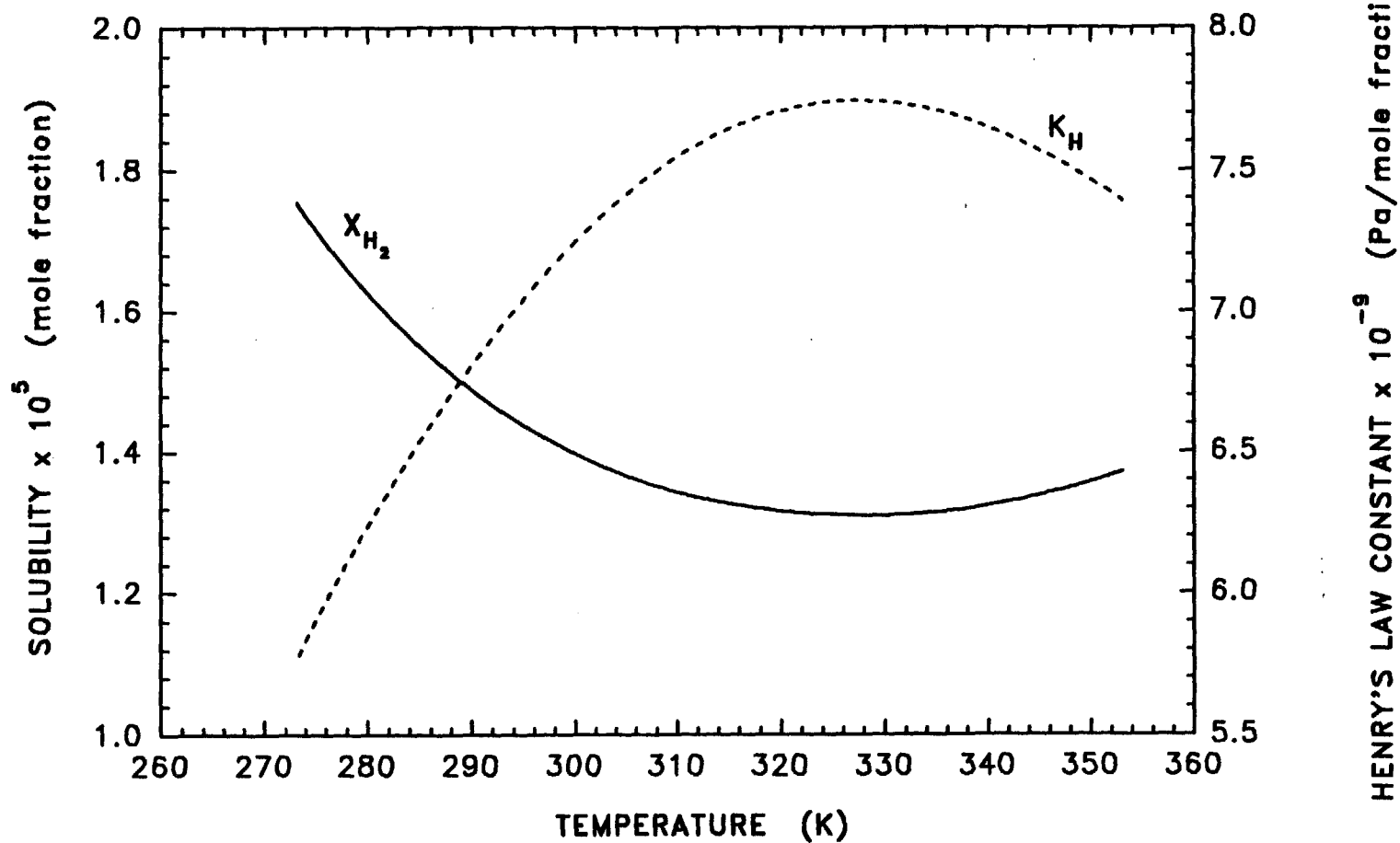
Figure 2

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constants, in units of Pa/mole fraction, assuming a hydrogen partial pressure of 0.101325 MPa.

## 3.3 Nitrogen

The solubility of nitrogen in pure water at 0.101325 MPa is evaluated by Battino (1982). The temperature dependence of the solubility is presented in Figures 1 and 3. A smooth curve fit was generated using 74 solubility values from nine different studies (including Morrison and Billet, 1952). The data exhibit a trend towards a minimum at the high temperatures. Henry's Law values derived from these data are also presented in Figure 3. Nitrogen and hydrogen exhibit very similar mole fraction solubility values for the temperature range of 275-350 K (Figure 1).

## 3.4 Methane

Clever and Young (1987) provide a critical evaluation of methane solubility in pure water at 0.101325 MPa. The study by Rettich et al. (1981) is recommended as the most precise and suitable evaluation of methane solubility of over thirty three studies. These data were fit to a smoothing equation and plotted in Figures 1 and 4. A minimum in the solubility curve appears at approximately 362 K. Henry's Law constants are also provided in Figure 4. Methane displays a greater mole fraction solubility than hydrogen and nitrogen for the given temperatures and which is more than two-fold greater at the low temperature end (Figure 1).

NITROGEN SOLUBILITY IN PURE WATER
at 0.101325 MPa partial pressure

Figure 3

# METHANE SOLUBILITY IN PURE WATER
## at 0.101325 MPa partial pressure
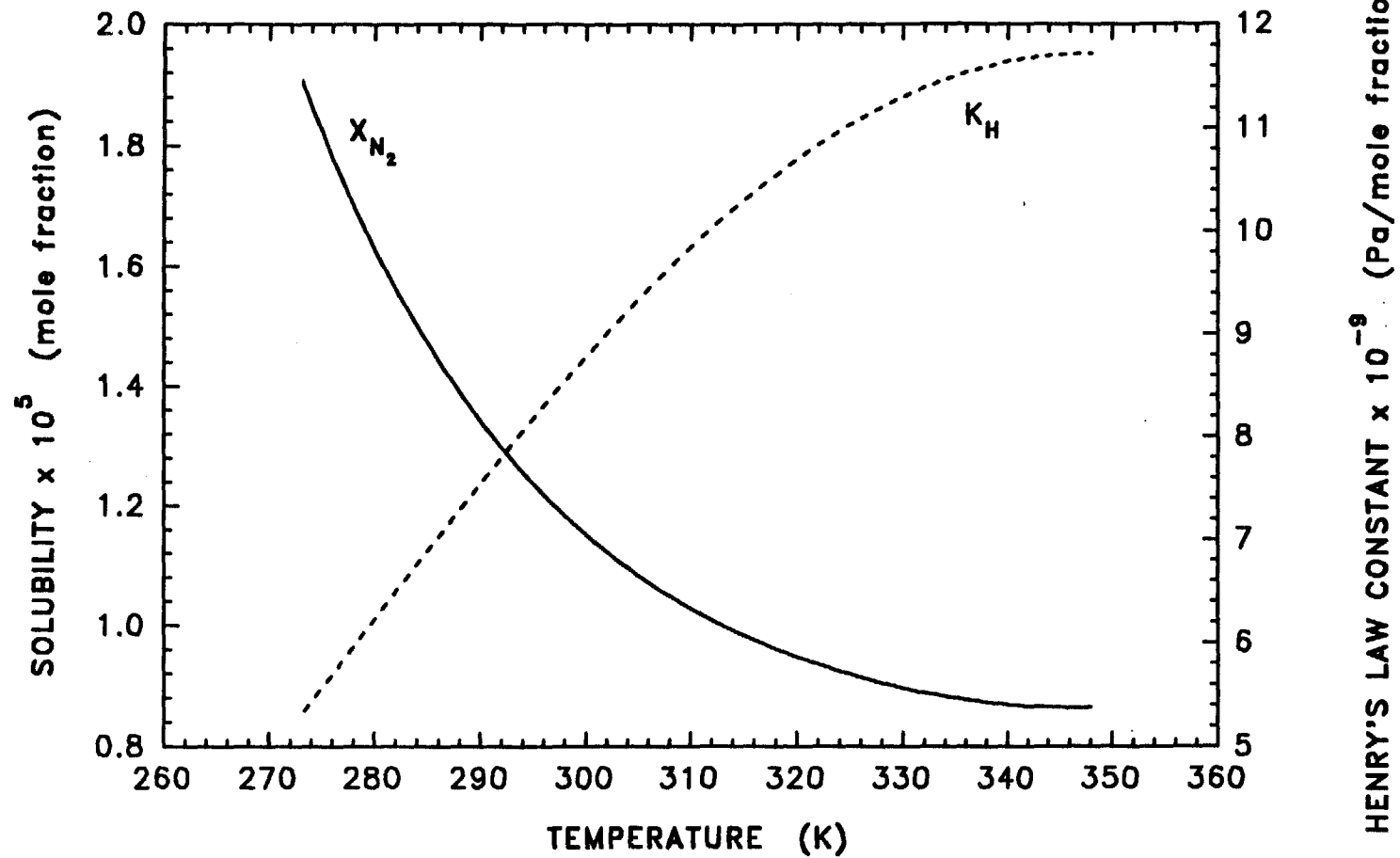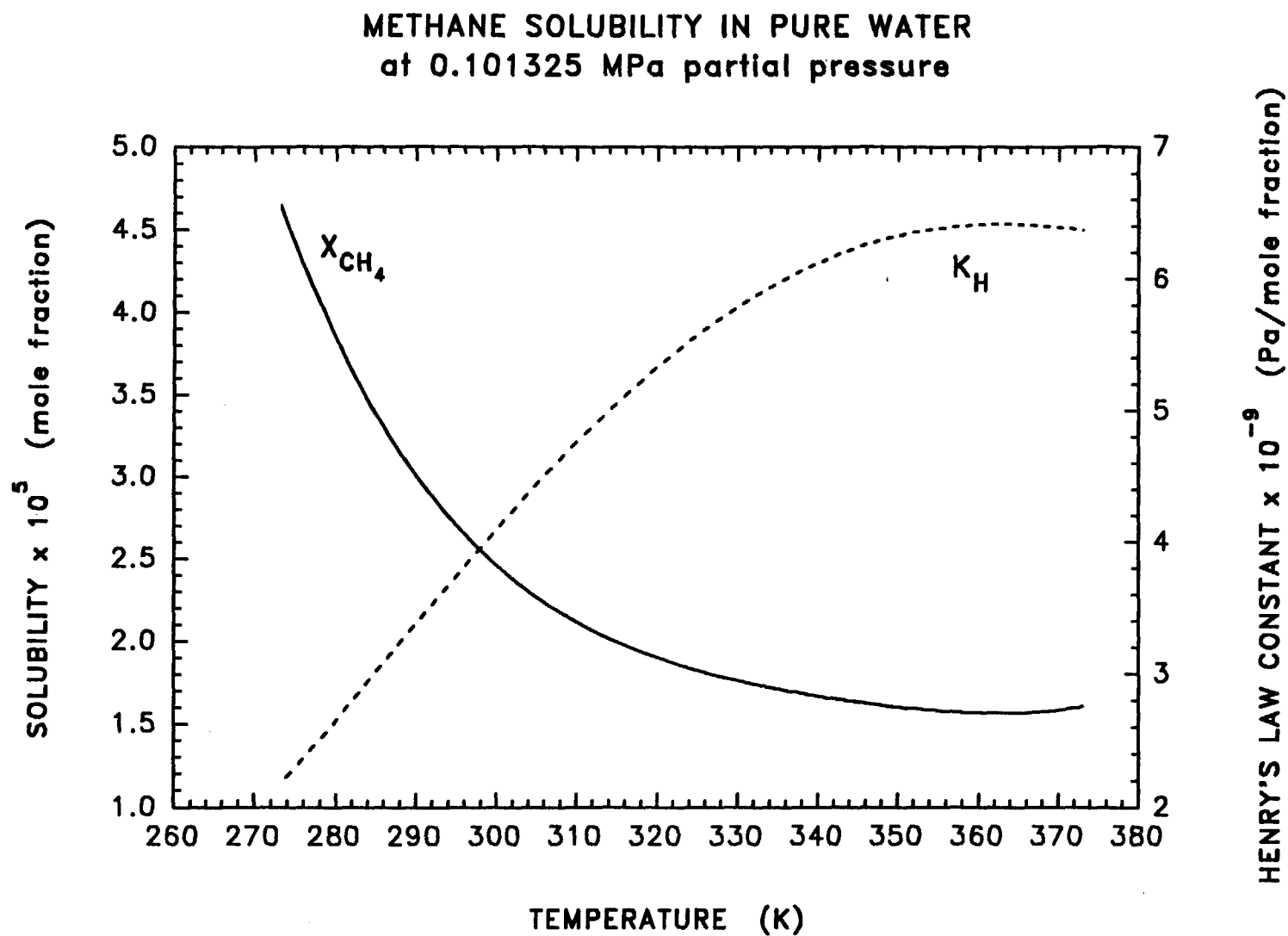


Figure 4

## 3.5 Carbon Dioxide

The solubility of carbon dioxide in pure water at 0.101325 MPa is summarized in Figure 5. The smooth curve represents the best fit of the available data based upon the compilation of Dodds et al. (1956) and the measurements of Ellis (1959), Stewart and Munjal (1970), and Munjal and Stewart (1971). A large decrease in solubility with temperature is displayed for the given temperature range. Figure 5 also displays the Henry Law's constants based upon a carbon dioxide partial pressure of 0.101325 MPa. Figure 1 presents the carbon dioxide solubility data relative to that of the other gases. At 273.15 K carbon dioxide exhibits a mole fraction solubility that is more than thirty times greater than that of the other three gases. The chemical reaction of the water solvent with carbon dioxide to create carbonic acid results in this drastic increase in solubility relative to the more inert gases.

# CARBON DIOXIDE SOLUBILITY IN PURE WATER
## at 0.101325 MPa partial pressure
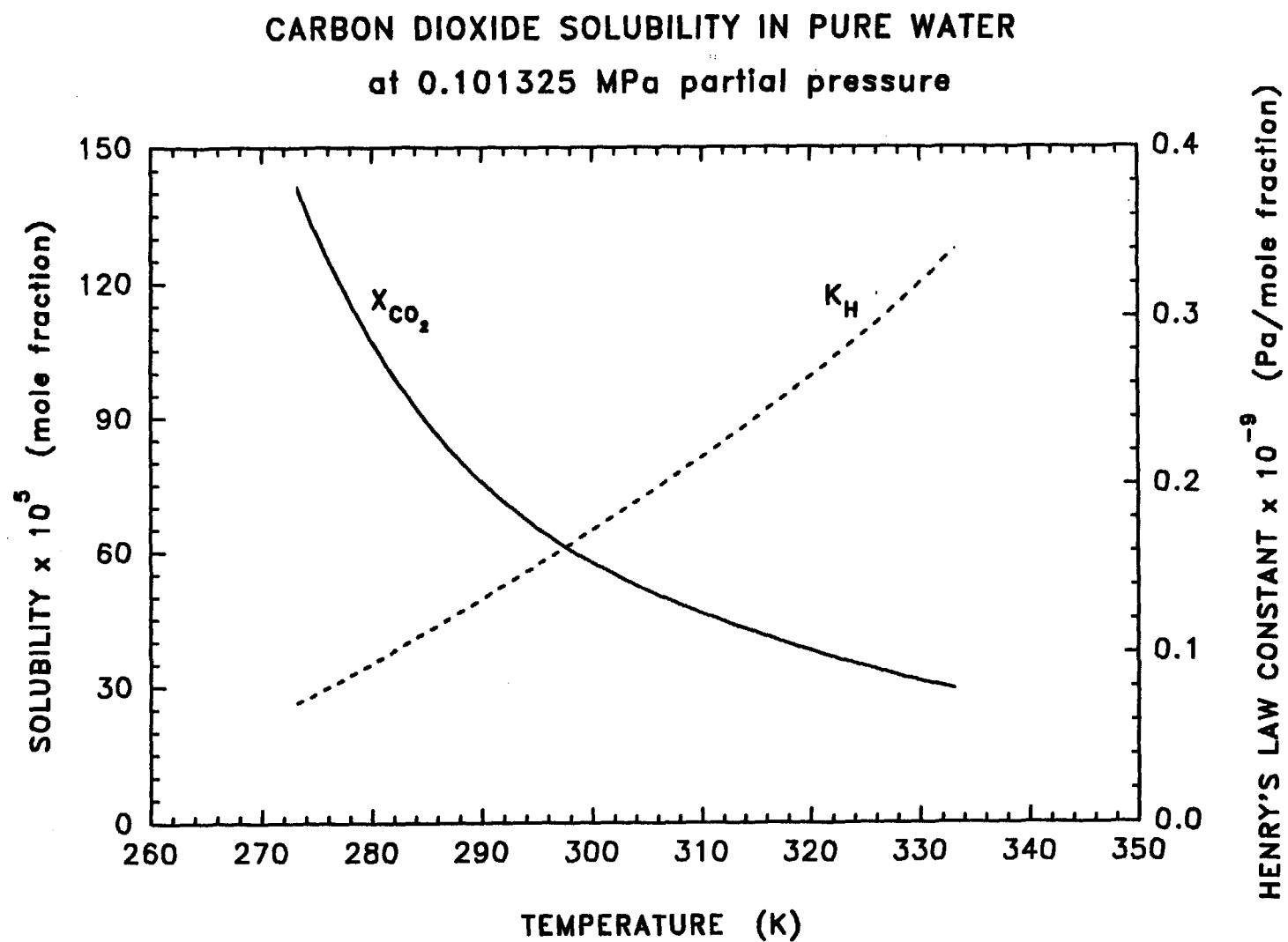


Figure 5

# 4  GAS SOLUBILITY IN PURE WATER AT ELEVATED PRESSURES

## 4.1  Hydrogen

The data of Wiebe and Gaddy (1934) are recommended as the best
available measurements of the solubility of hydrogen in pure water at
elevated pressures (see Young, 1981).  More recent solubility data exist
for hydrogen, however, these data are not as precise nor internally
consistent as this older study, nor do they cover an extensive pressure
range.  Figure 6 presents smoothed isobars for the mole fraction
solubility of hydrogen as a function of temperature.  The low pressure
solubilities exhibit very little dependence, if any, upon temperature.
The isobars for the higher pressures (up to 101.325 MPa) exhibit a
minimum in the solubility curves.  Figure 7 provides the same hydrogen
solubility data in the form of isotherms for the given pressures.

## 4.2  Nitrogen

Battino (1982) recommends the use of the data of Wiebe et al.
(1933) and O'Sullivan and Smith (1970) for applications requiring the
solubility of nitrogen in water at elevated pressures.  These two
studies provide solubilities for pressures up to 50 MPa which are in
fairly good agreement with each other.  The isobars of the solubility
for each study are presented in Figure 8 as a function of temperature.
The temperature dependence of the nitrogen solubility is similar to that
exhibited by hydrogen.  Figure 9 provides a plot of the data of the two

# HYDROGEN SOLUBILITY IN PURE WATER
## at elevated pressures



Figure 6

# HYDROGEN SOLUBILITY IN PURE WATER
## at elevated pressures
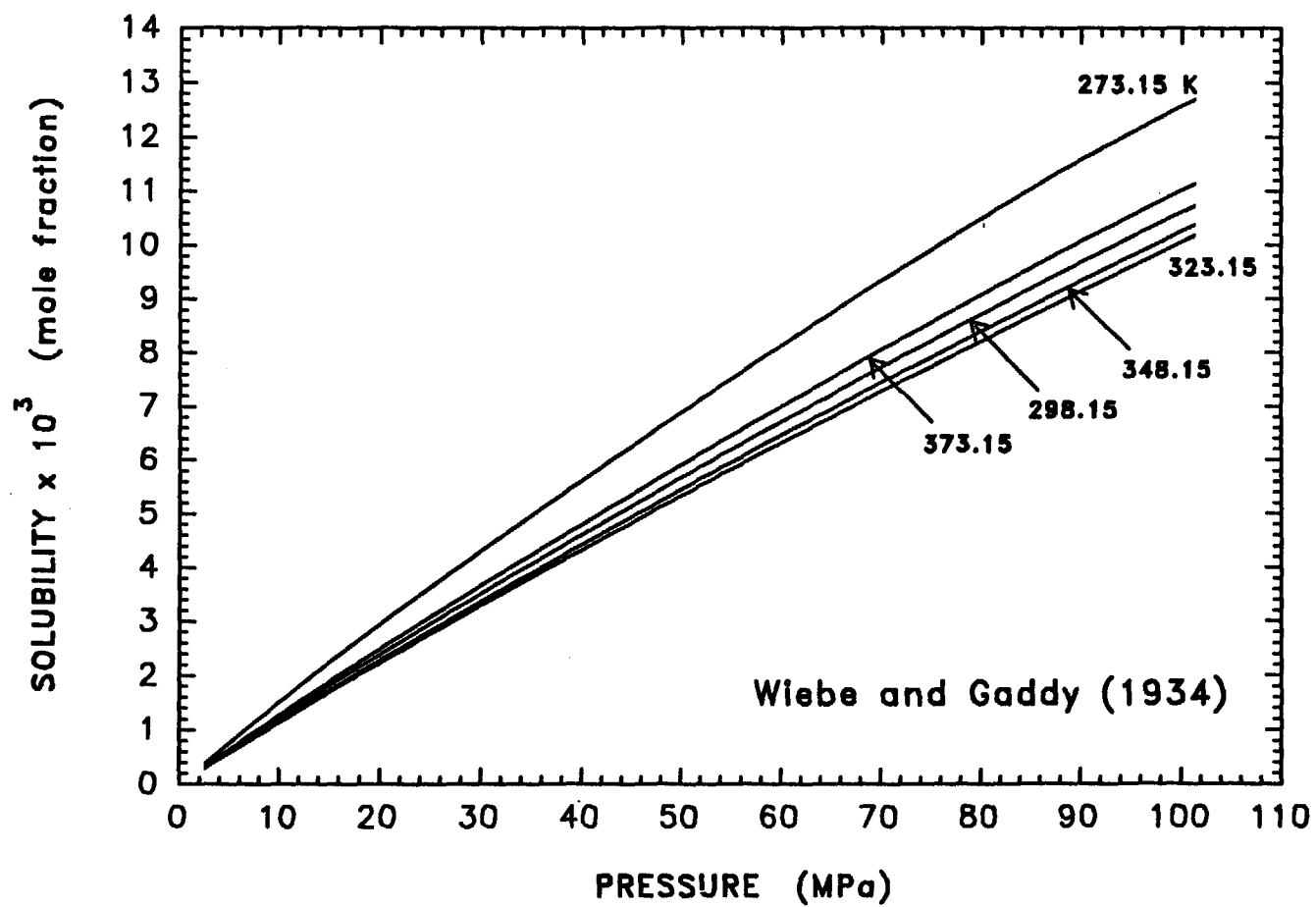


Figure 7

NITROGEN SOLUBILITY IN PURE WATER
at elevated pressures

Figure 8

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# NITROGEN SOLUBILITY IN PURE WATER
## at elevated pressures
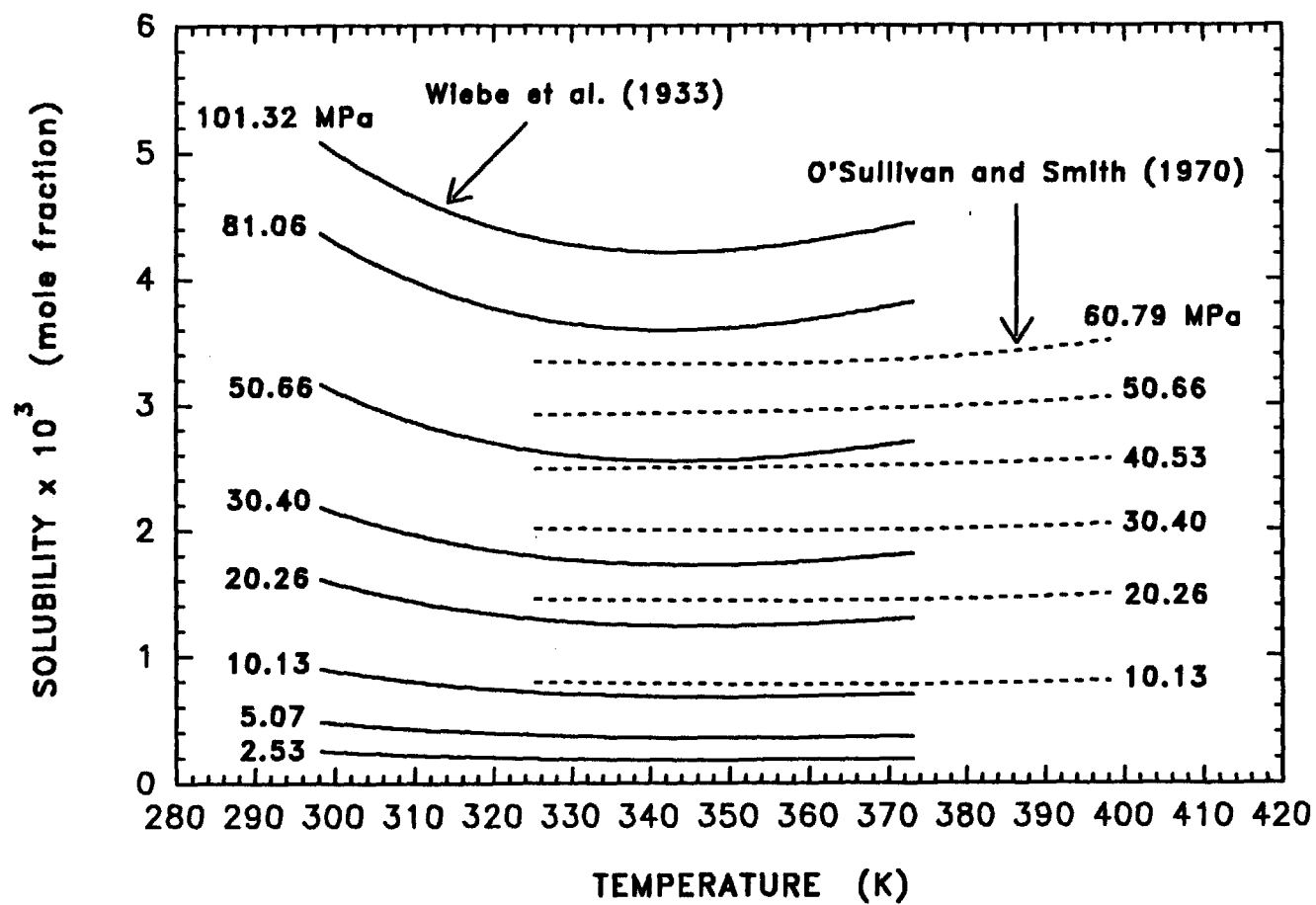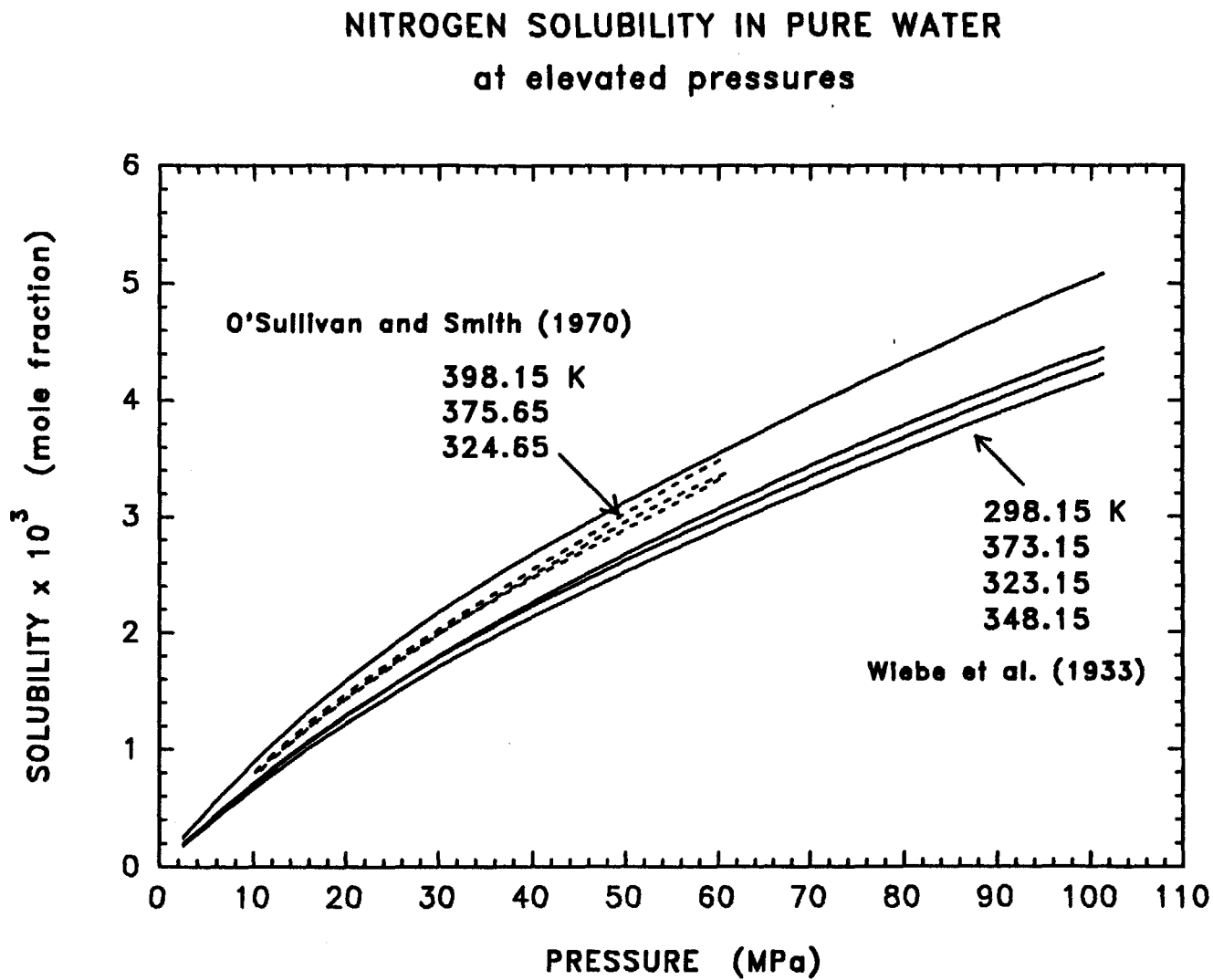


Figure 9

recommended studies expressed as isotherms over the experimental

pressure range.


## 4.3 Methane


Measurements of methane and other associated hydrocarbon solubility

in water at elevated pressures are much more prevalent in the literature

because of the significance of these data in evaluating petroleum

reserves in natural fluids.  Clever and Young (1987) provides an

assessment of the available studies from the literature and supplies a

smooth curve (based on equation (15)) fit to over 240 data points for

the temperature and pressure dependence of the solubility.  The data

sources include Duffy et al. (1961), O'Sullivan and Smith (1970), Price

(1979), Stoessell and Byrne (1982) and Cramer (1984).  Other studies of

note but not included in this evaluation are Kobayashi and Katz (1953),

Price (1981), Crovetto et al. (1982), Ashmyan et al. (1984).  A

graphical summary for the smoothed data is provided in Figures 10 and 11

where the respective isobars and isotherms are plotted.  The isobars

exhibit minima which are enhanced by increasing pressure.  The isotherms

display a roughly linear dependence upon pressure.  The solubility data

isobars of McGee et al. (1981) are presented in Figure 12.  Section 2.6

discusses the uniqueness of this particular study and the results are

presented here separately.  The McGee et al. (1981) solubility values

are slightly higher than those presented in Figure 10, but the shapes of

the curves are quite similar.  Note that Figure 12 displays a decrease

in solubility for the low pressure curves as the temperature increases

beyond 500 K.

# METHANE SOLUBILITY IN PURE WATER
## at elevated pressures
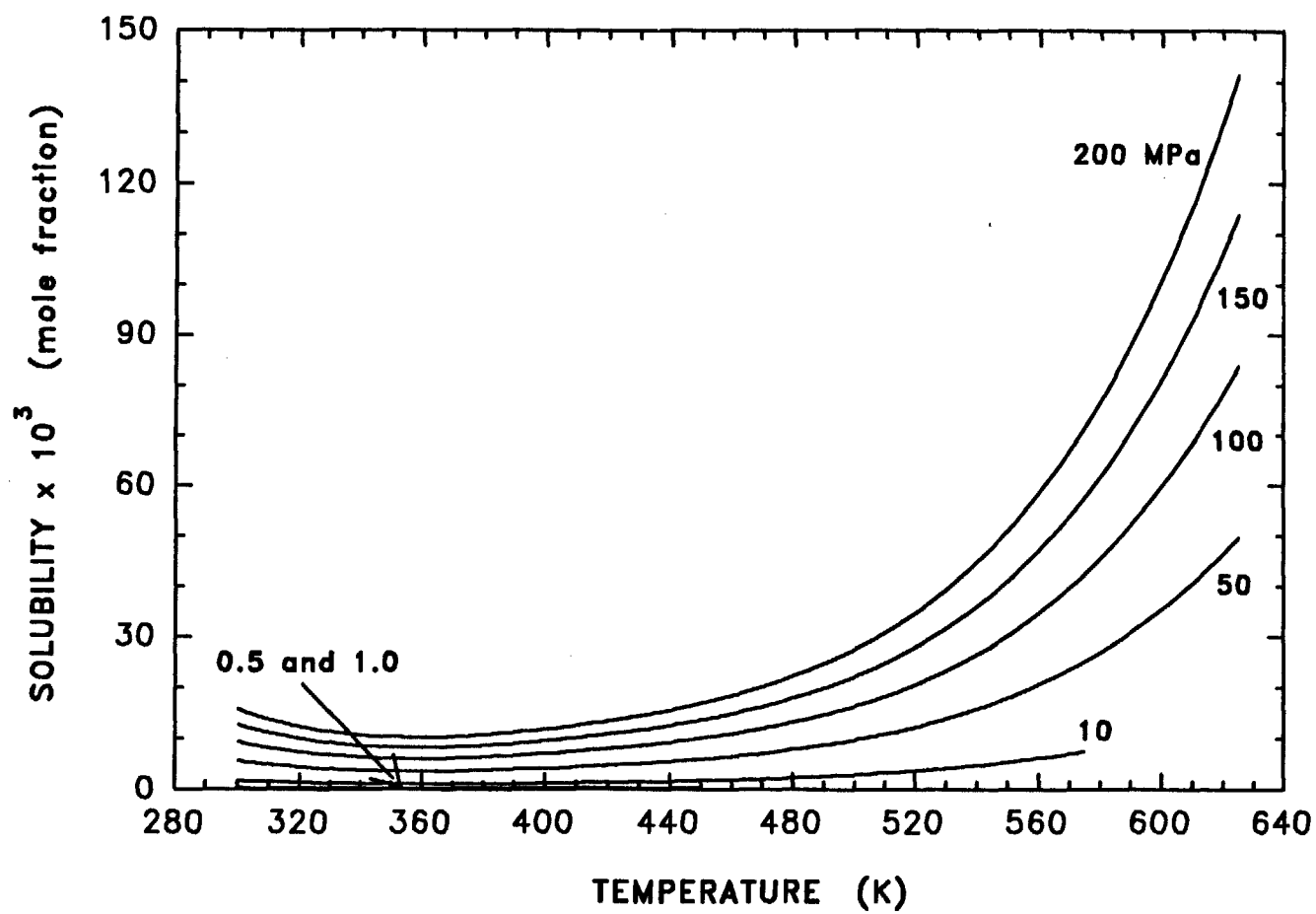


Figure 10

METHANE SOLUBILITY IN PURE WATER
at elevated pressures



Figure 11

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**METHANE SOLUBILITY IN PURE WATER**
**at elevated pressures**

McGee et al. (1981)
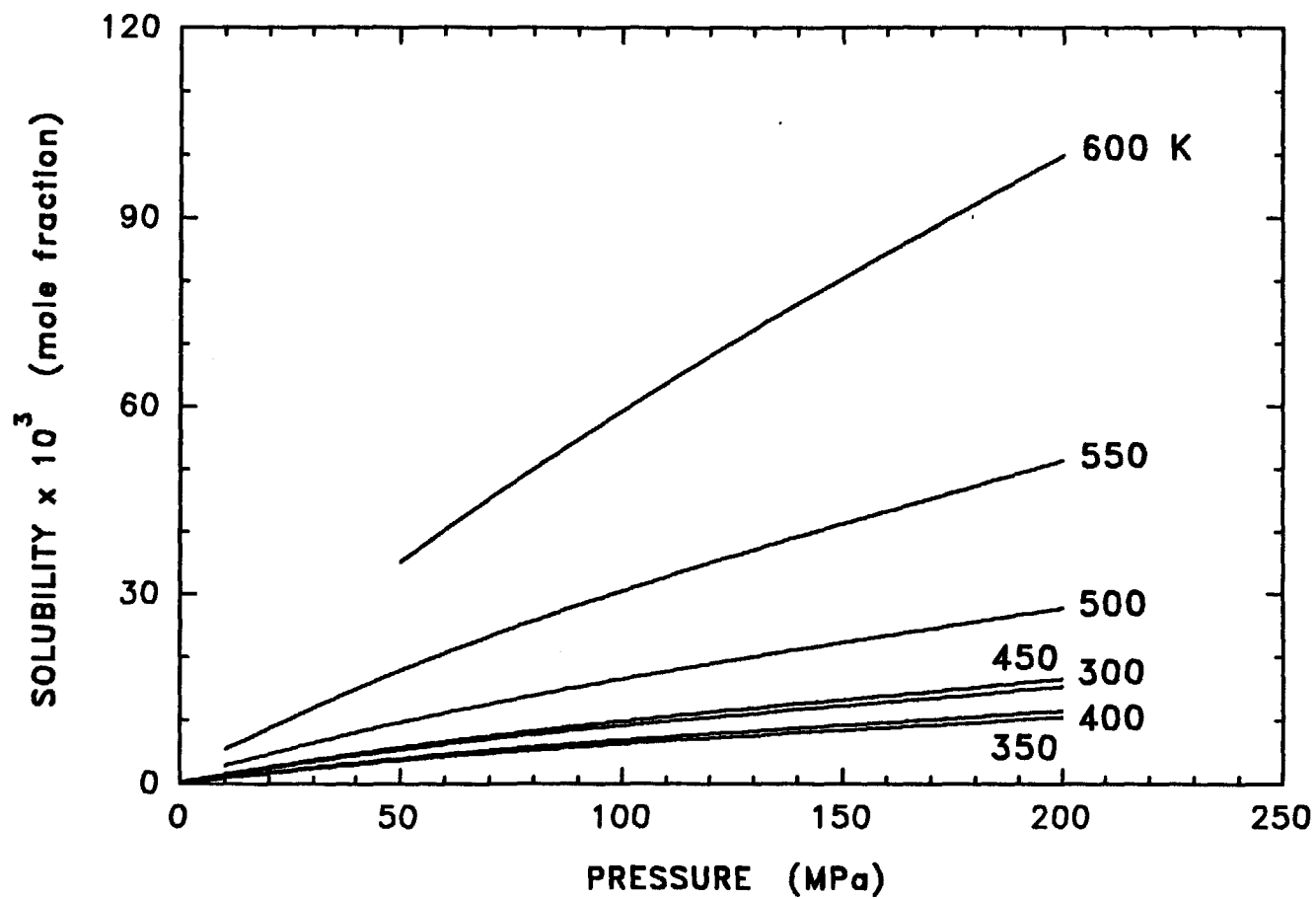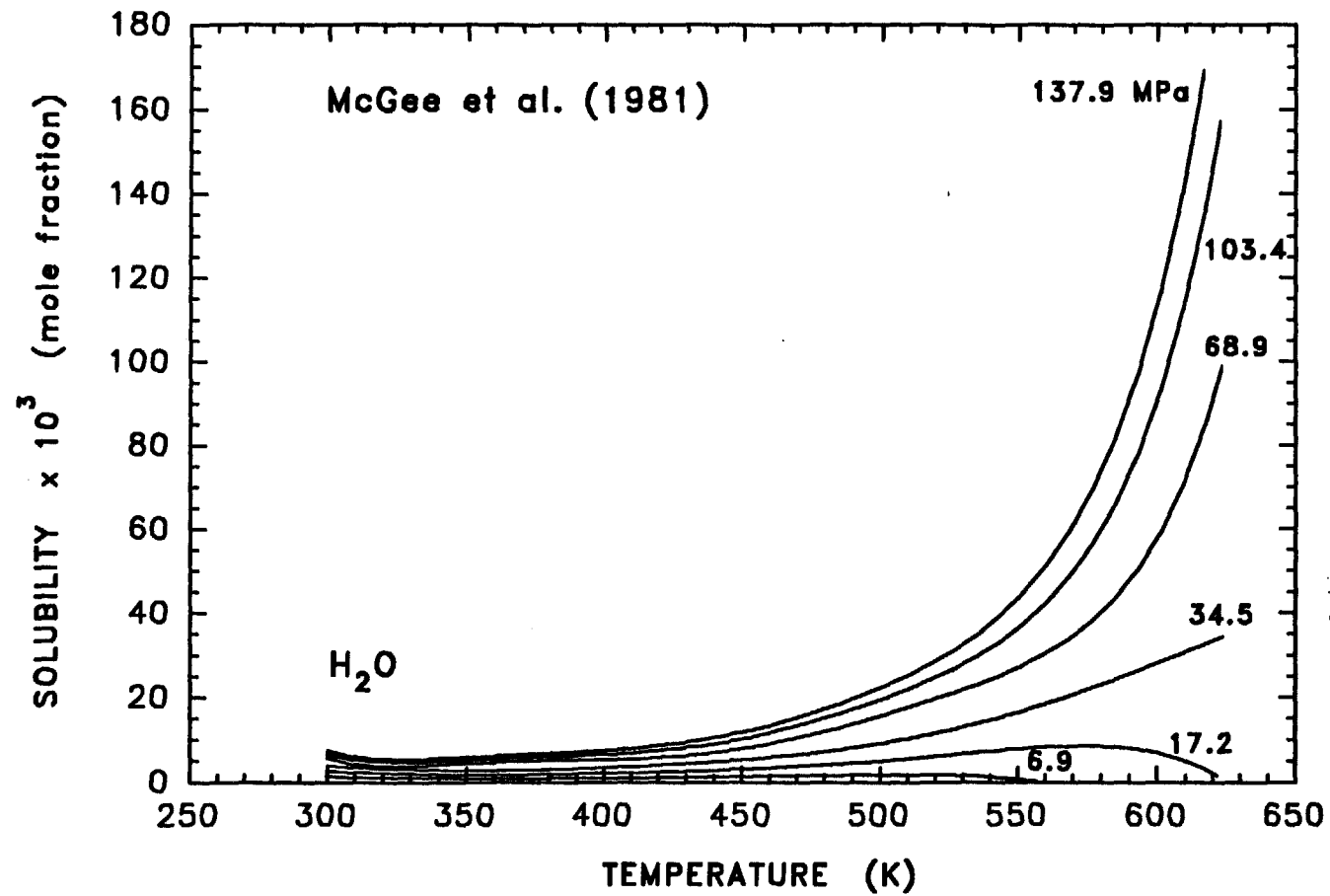
Figure 12

## 4.4 Carbon Dioxide

A summary of the solubility data for carbon dioxide in pure water at elevated pressures is provided in Figure 13. Isobars were fit to the compiled data of Dodds et al. (1956) which includes the measurements of Wiebe and Gaddy (1939, 1940, 1949), Wiebe (1941), and Prutton et al. (1945). The solubility values for carbon dioxide for these pressures are greater than those observed for the other three gases. The isobars display minima for only the highest pressure data. At the lower pressures the solubility first decreases with increasing temperature then levels off at the higher temperatures. Figure 14 provides the recent solubility data of Nighswander et al. (1989) as a function of pressure for several high temperatures. The solubility data of Takenouchi and Kennedy (1964) are presented in Figures 15 and 16. Figure 16 displays the response of the solubility isotherms as the critical point of the system is approached at high temperature.
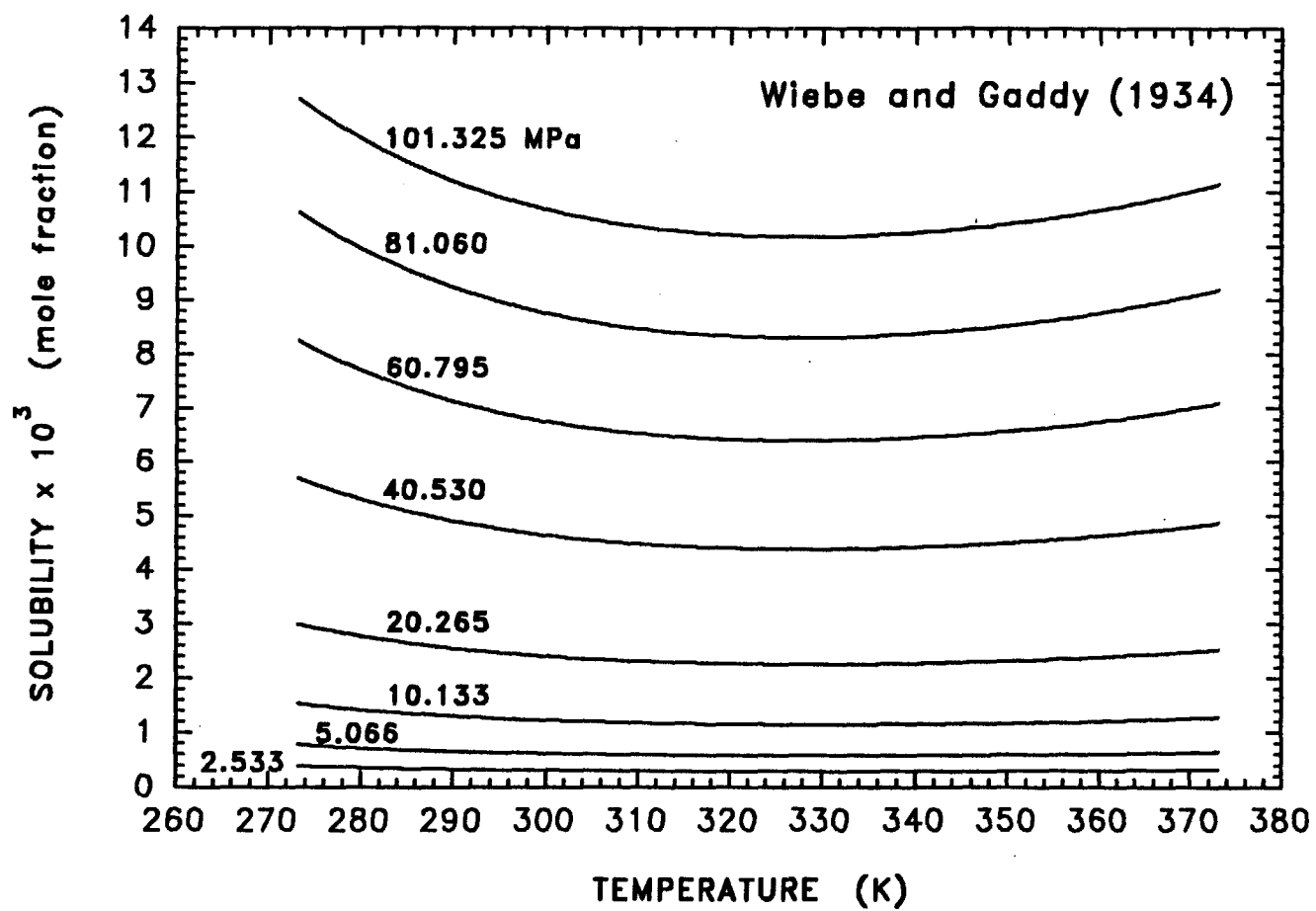
CARBON DIOXIDE SOLUBILITY IN PURE WATER
at elevated pressures

Figure 13

# CARBON DIOXIDE SOLUBILITY IN PURE WATER
## at elevated pressures
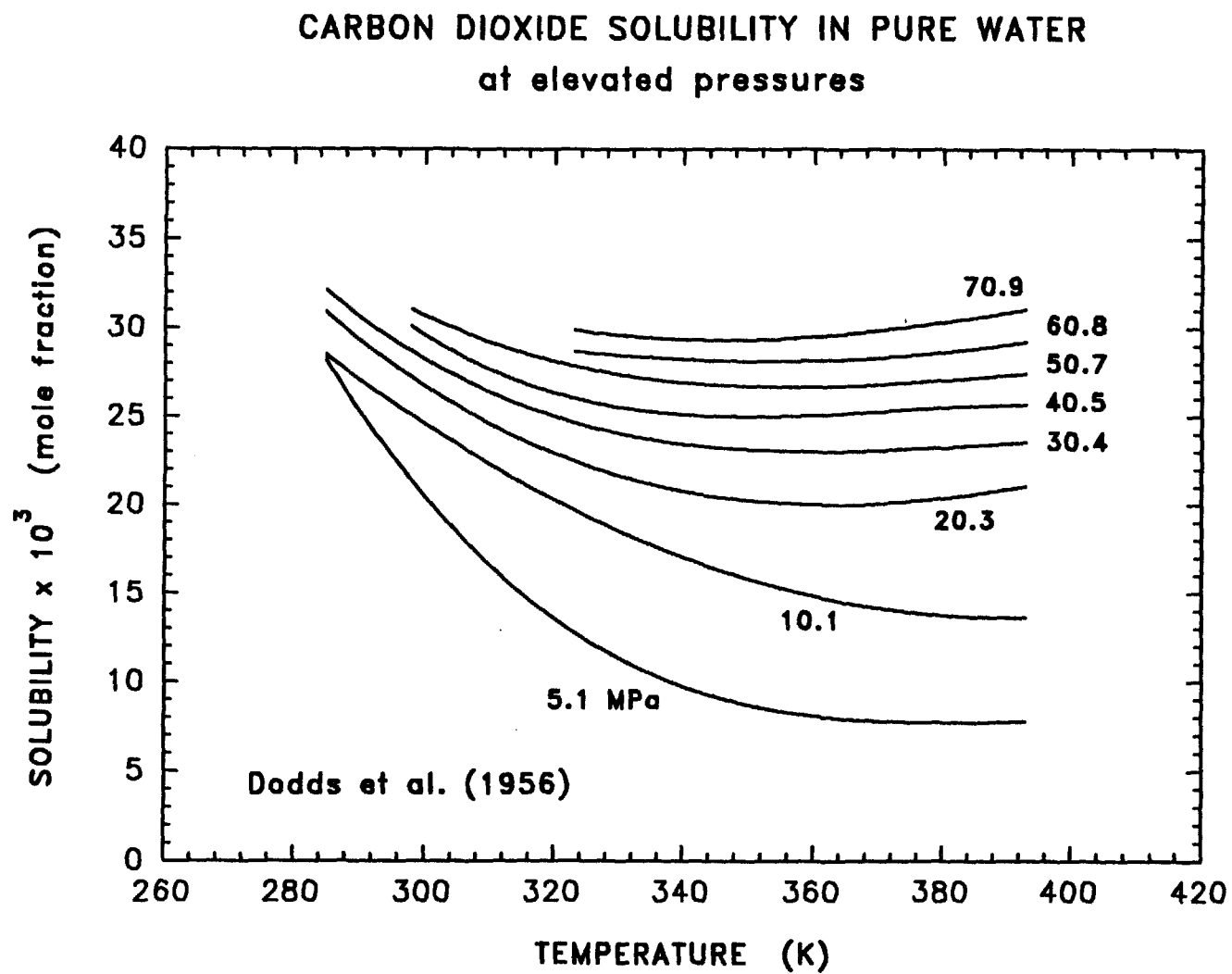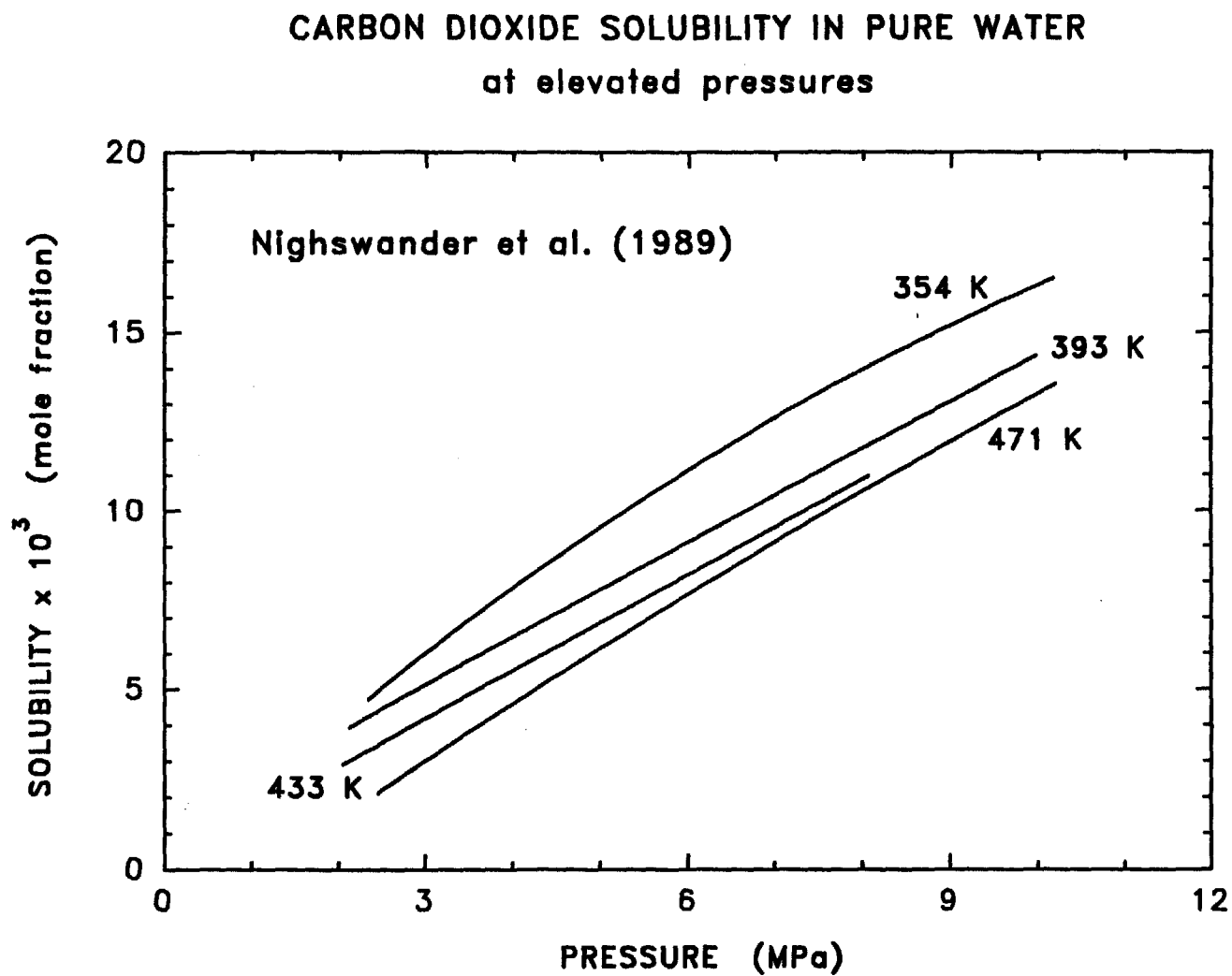


Figure 14

# CARBON DIOXIDE SOLUBILITY IN PURE WATER
## at elevated pressures



Takenouchi and Kennedy (1964)
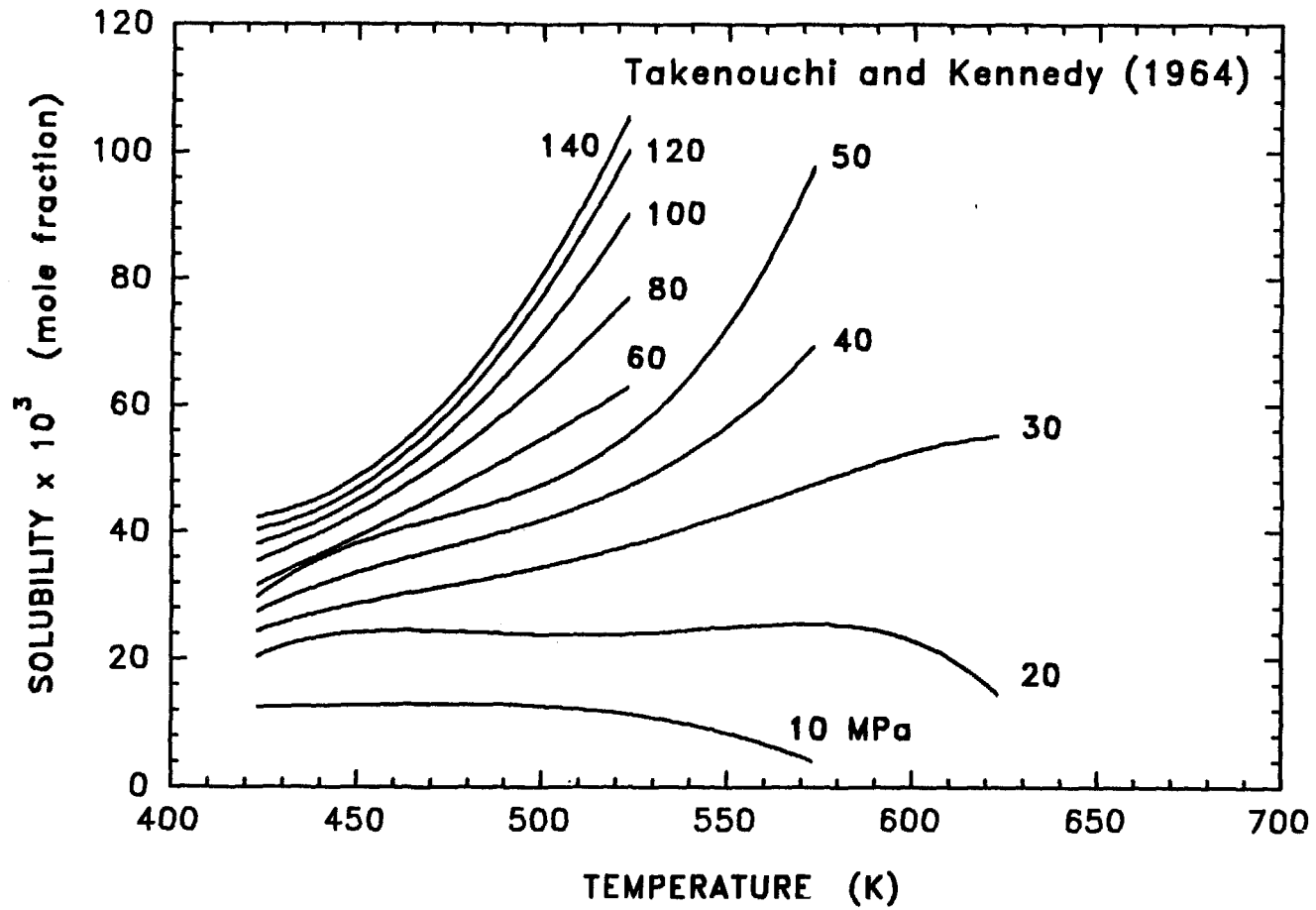
Figure 15

# CARBON DIOXIDE SOLUBILITY IN PURE WATER
## at elevated pressures
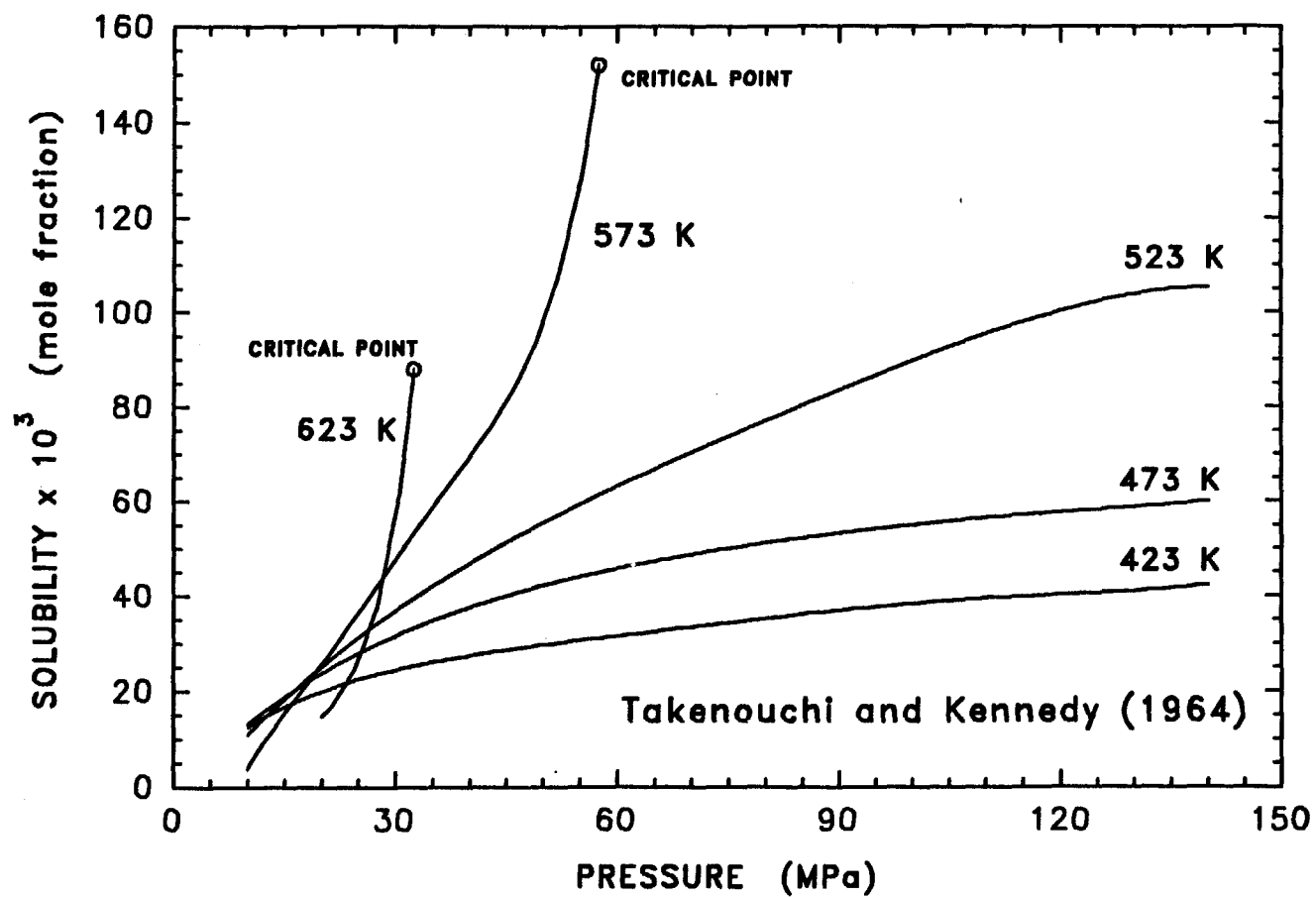


Figure 16

# 5  GAS SOLUBILITY IN BRINE

## 5.1  Hydrogen

A decrease in the gas solubility generally occurs by introducing sodium chloride into the water solvent and creating an electrolyte solution.  This physiochemical process is referred to as salting-out and is quantified by the Sechenov coefficient (see Section 2.3.2).  The temperature dependence of the Sechenov coefficient for hydrogen gas solubility in brine (NaCl solution) at 0.101325 MPa is presented in Figure 17 as recommended by Young (1981).  The Sechenov curve is derived from the best fit of data from Morrison and Billet (1952) and Crozier and Yamamoto (1974) based upon solubility values derived from Bunsen coefficients (Sechenov coefficients are presented as $k_{scx}$ rather than $k_{sc\alpha}$).  Only data for 1 N NaCl solutions are available.  The limited data suggest a linear decrease of the Sechenov coefficient with temperature.

The Sechenov coefficients are easily implemented for calculating the mole fraction solubility of the electrolyte solution given that the solubility of the gas in pure water solvent is known.  In addition, they can be used to extrapolate gas solubilities measurements to other concentrations of the electrolyte assuming that the Sechenov coefficient is not a function of salt concentration (see Malinin, 1978).  Equation (13) can be rearranged to provide the following useful version of the Sechenov relation:

## HYDROGEN SOLUBILITY IN SODIUM CHLORIDE SOLUTION
### derived from 1 N NaCl at 0.101325 MPa partial pressure
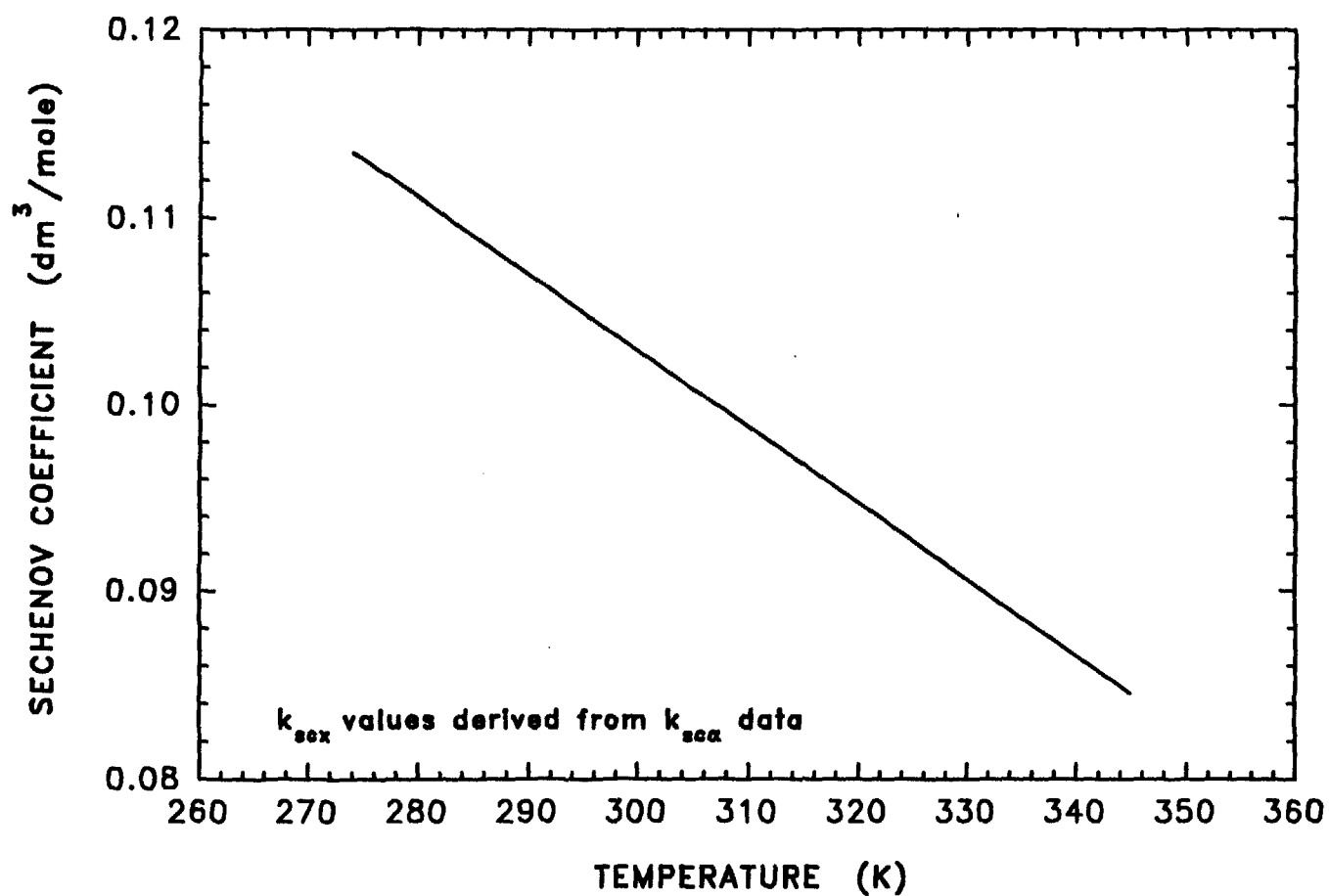


Figure 17

$$S_i = S_i^o \, 10^{-k_s C_i} \qquad (17)$$

Figure 18 provides the solubility curves for hydrogen in sodium chloride solution at 0.101325 MPa based upon this extrapolation of the 1 N NaCl data. The solubilities for the brines are less than those for the pure water as would be expected for positive Sechenov coefficient values (salting out process). The 5 N NaCl solution exhibits an extrapolated solubility curve that is approximately at third the value for hydrogen solubility in pure water. Minima in the solubility curves for the brines are similar to those observed for the pure water but are displaced toward lower temperatures.

## 5.2 Nitrogen

Sechenov coefficients for nitrogen solubility in brine at 0.101325 MPa are presented in Figure 19 based upon the recommended values from Battino (1982). The smooth curve represents the best fit of the $k_{scx}$ data for the temperature dependence of Sechenov values from Morrison and Billett (1952), Smith et al. (1962), O'Sullivan et al. (1966), and O'Sullivan and Smith (1970). No compositional dependency of the Sechenov values is observed for brine concentrations up to 5.5 N NaCl. The derived solubility curves as a function of temperature are reported in Figure 20 for several brine concentrations. The 5 N NaCl solution displays a solubility value at 298 K that is approximately five times less than the value that is observed for pure water. Minima in the solubility curves are reduced with increasing brine concentration. The high concentration solutions exhibit very little temperature dependence.

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HYDROGEN SOLUBILITY IN SODIUM CHLORIDE SOLUTION
derived from 1 N NaCl at 0.101325 MPa partial pressure

Figure 18

# NITROGEN SOLUBILITY IN SODIUM CHLORIDE SOLUTION
## derived from 1 N NaCl at 0.101325 MPa partial pressure



$k_{scx}$ values derived from $k_{scc}$ data

SECHENOV COEFFICIENT $(dm^3/mole)$

TEMPERATURE (K)
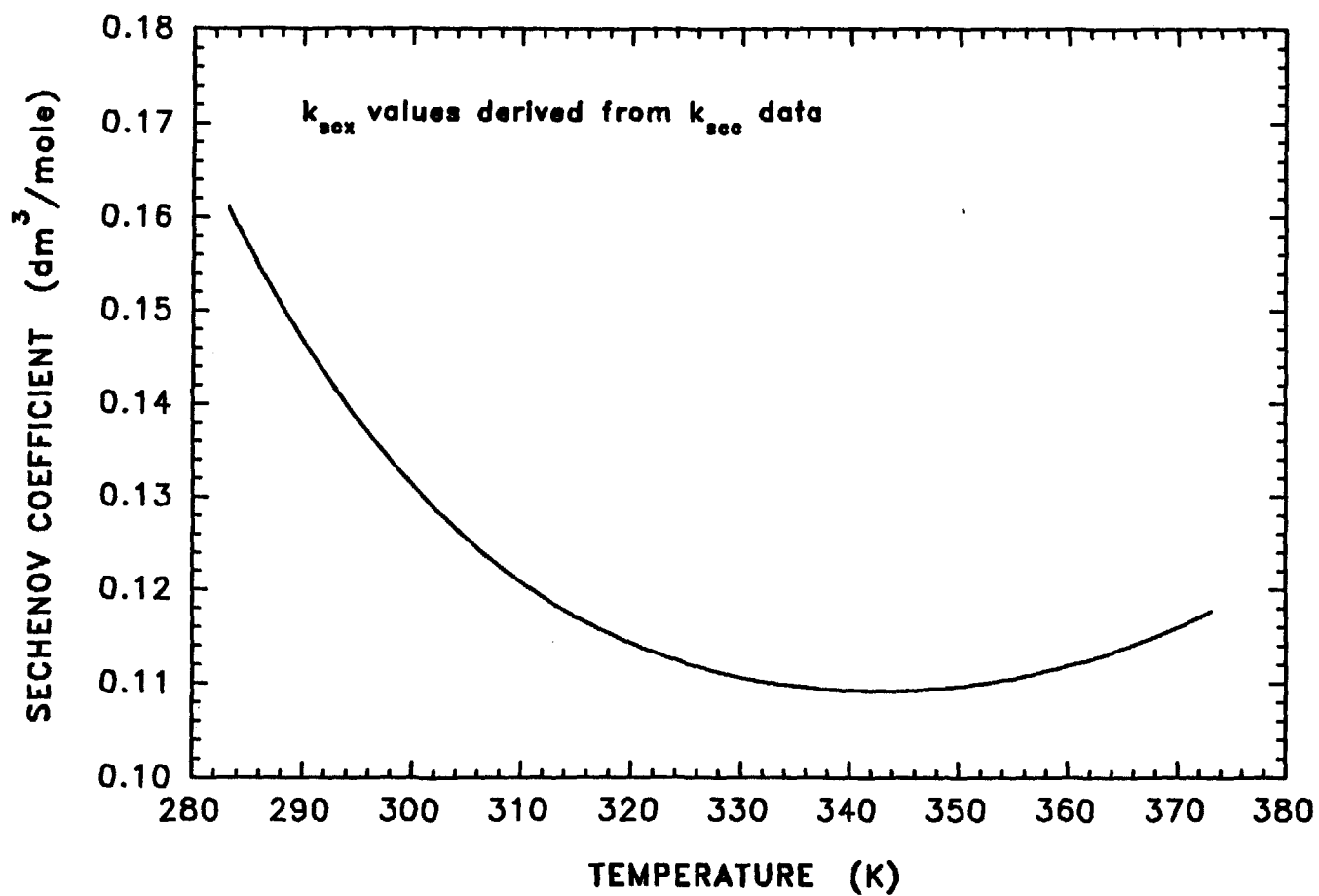
Figure 19

## NITROGEN SOLUBILITY IN SODIUM CHLORIDE SOLUTION
### derived from 1 N NaCl at 0.101325 MPa partial pressure
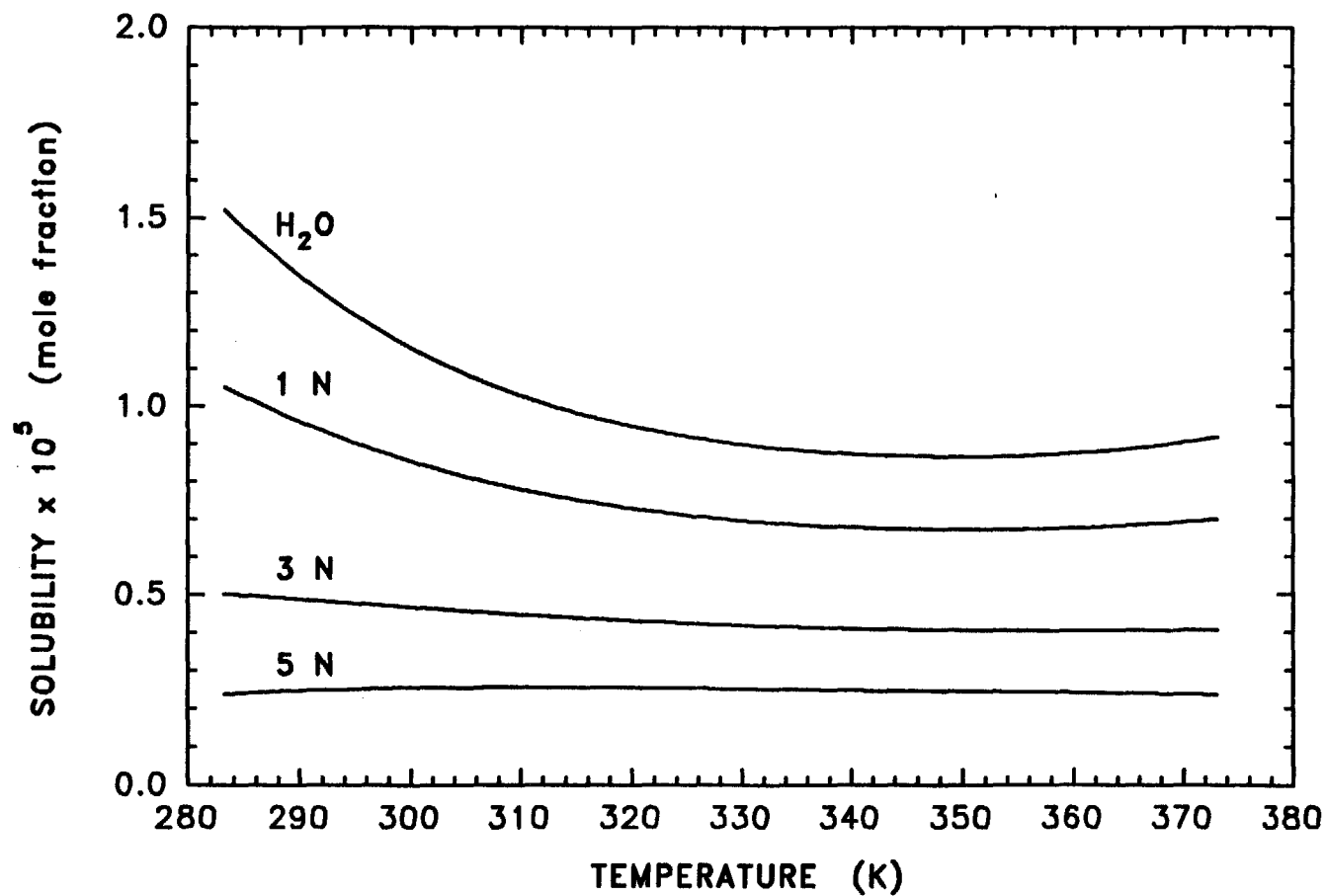


Figure 20

## 5.3 Methane

Clever and Young (1987) provide a tabulated summary of the Sechenov coefficients for methane in sodium chloride solutions at 0.101325 MPa from twelve experimental studies including Michels et al. (1936), Duffy et al. (1966), O'Sullivan and Smith (1970), and Ben-Naim and Yaacobi (1974). A linear dependence of the coefficients, converted to $k_{scx}$ values, with temperature is observed and is presented in Figure 21. The scatter of the data symbols relative to the fitted line provides an estimate of the variability of the data from the separate studies. Clever and Young (1987) determined that there appears to be no dependency of the reported Sechenov coefficients with sodium chloride concentration or with pressure. Figure 22 presents the solubility curves derived from the Sechenov data for brine solutions up to 5 N NaCl as a function of temperature. Methane solubility in 5 N NaCl solution is decreased by more than a factor of five at 298 K relative to the solubility in pure water.

## 5.4 Carbon Dioxide

The Sechenov coefficients for the solubility of carbon dioxide in NaCl brine at 0.101325 MPa from Markham and Kobe (1941), Ellis and Golding (1963), Onda et al. (1970), and Malinin (1978) are represented in the fitted curves of Figure 23. Curves are presented for 1 N and 5 N NaCl solutions. The $k_{scx}$ values for carbon dioxide behave similarly to those of nitrogen by exhibiting a minimum in their temperature

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## METHANE SOLUBILITY IN SODIUM CHLORIDE SOLUTION
## derived from 1 N NaCl at 0.101325 MPa partial pressure



Figure 21

METHANE SOLUBILITY IN SODIUM CHLORIDE SOLUTION
derived from 1 N NaCl at 0.101325 MPa partial pressure
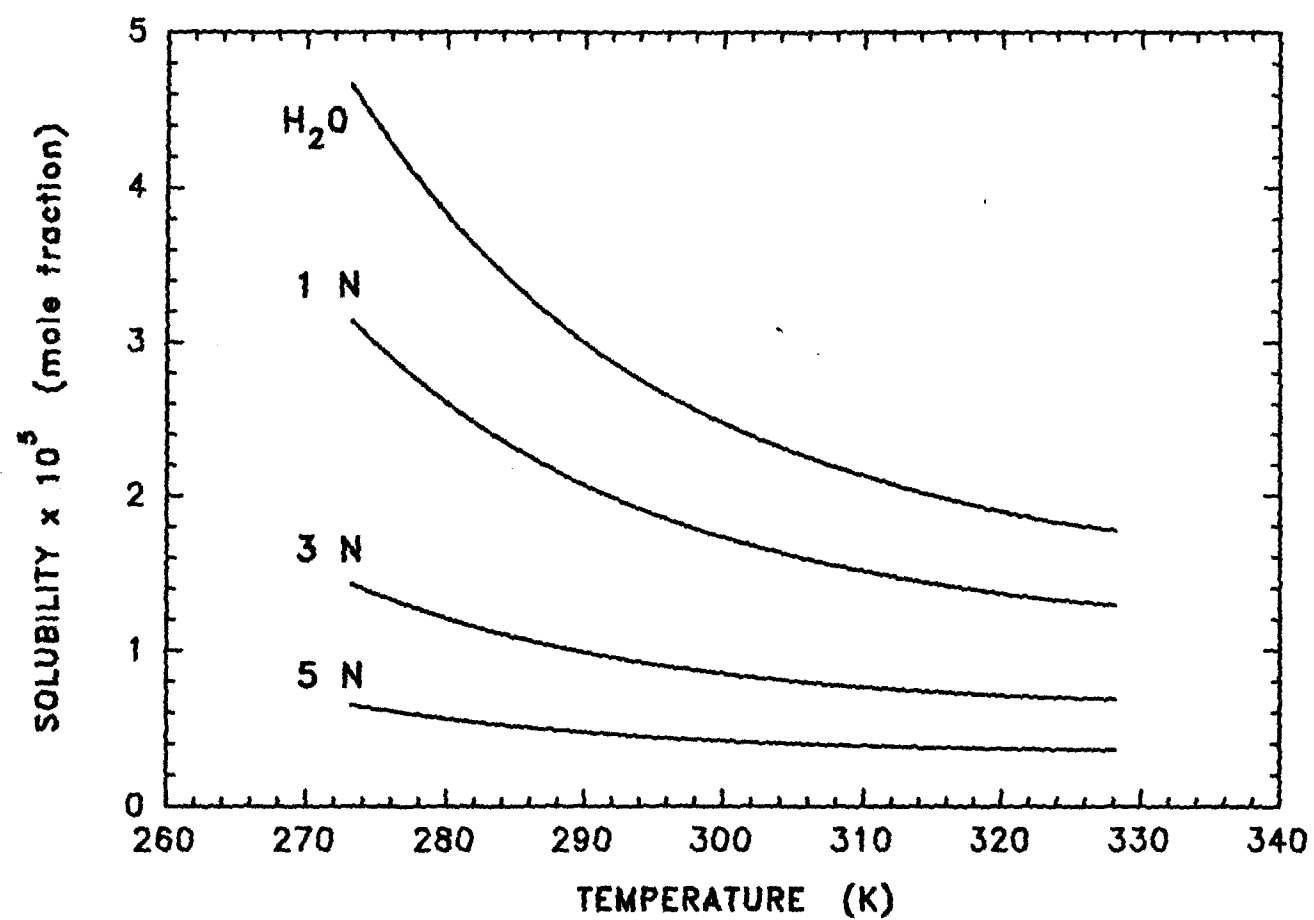


Figure 22

# CARBON DIOXIDE SOLUBILITY IN SODIUM CHLORIDE SOLUTION
## at 0.101325 MPa partial pressure
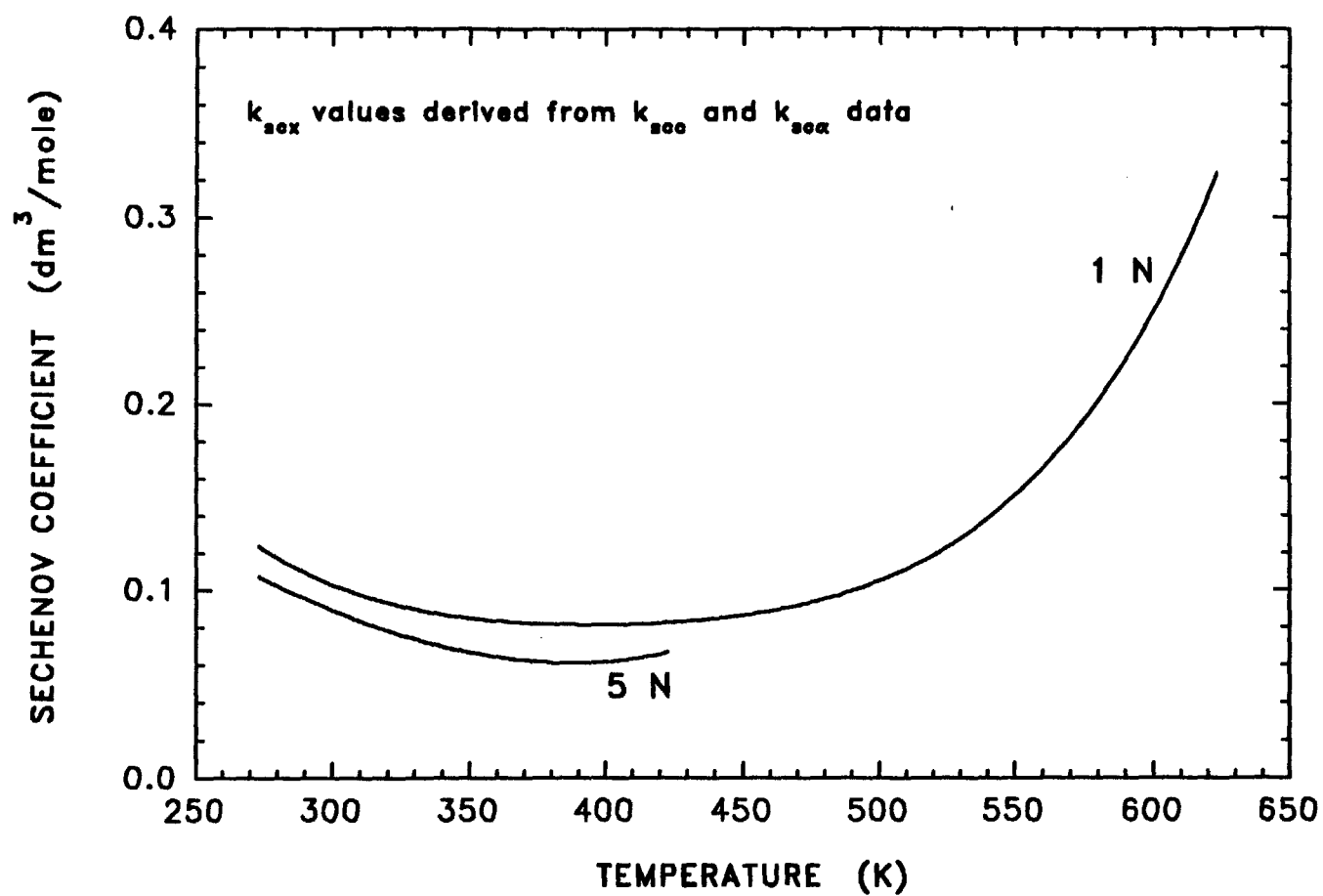
Figure 23

dependence.  Approximately 10 to 20 percent variability of the Sechenov coefficient is observed for the values between the two concentration curves up to a temperature of 420 K.  Figure 24 presents the solubility curves for carbon dioxide in NaCl brines as extrapolated using the 5 N NaCl Sechenov values and equation (17).  A similar calculation using the 1 N NaCl Sechenov coefficients provided extrapolated solubilities that are within 10 percent agreement.  Carbon dioxide solubility in 5 N NaCl solution at 298 K is decreased by a factor of four relative to that for pure water.
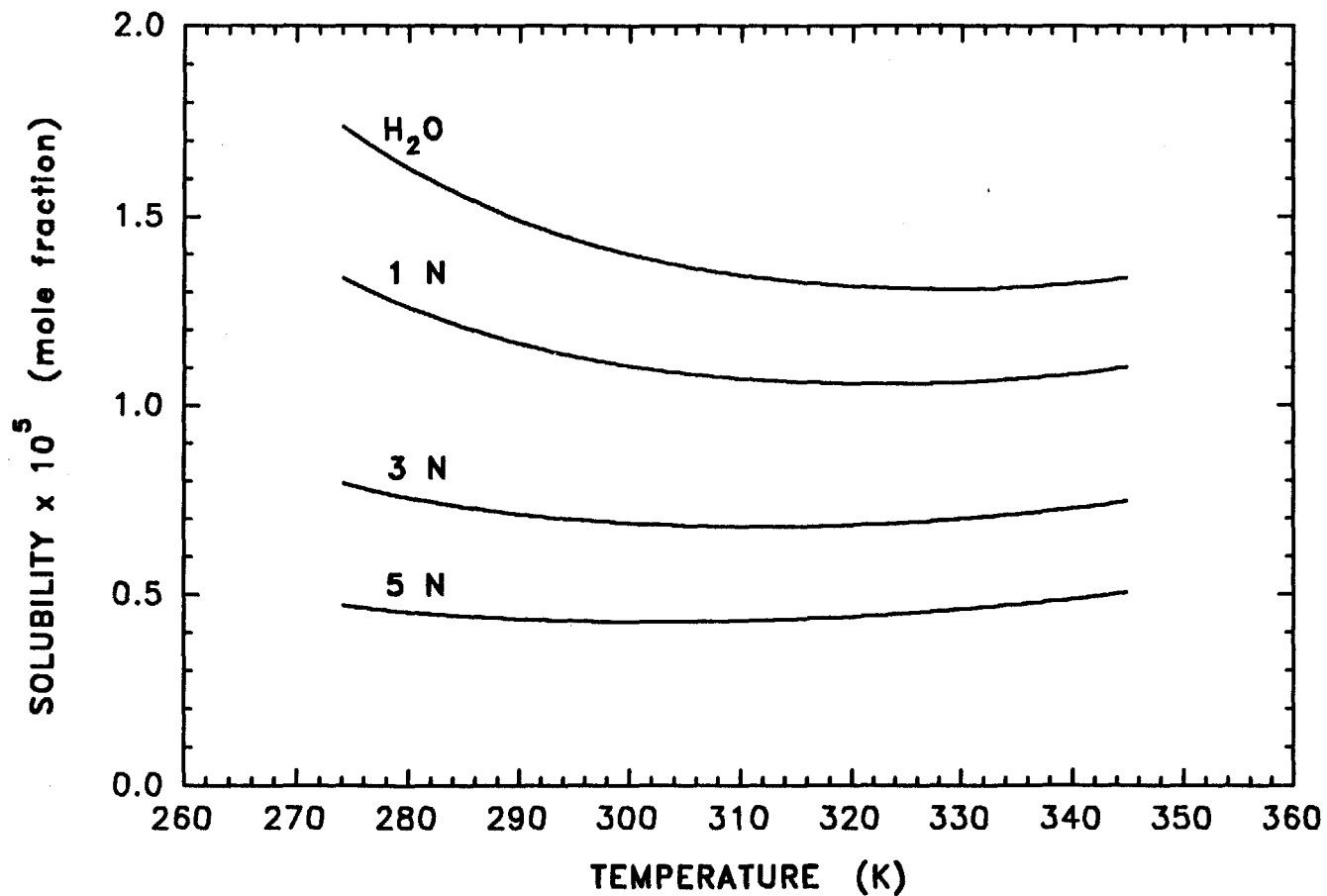
CARBON DIOXIDE SOLUBILITY IN SODIUM CHLORIDE SOLUTION
derived from 5 N NaCl at 0.101325 MPa partial pressure
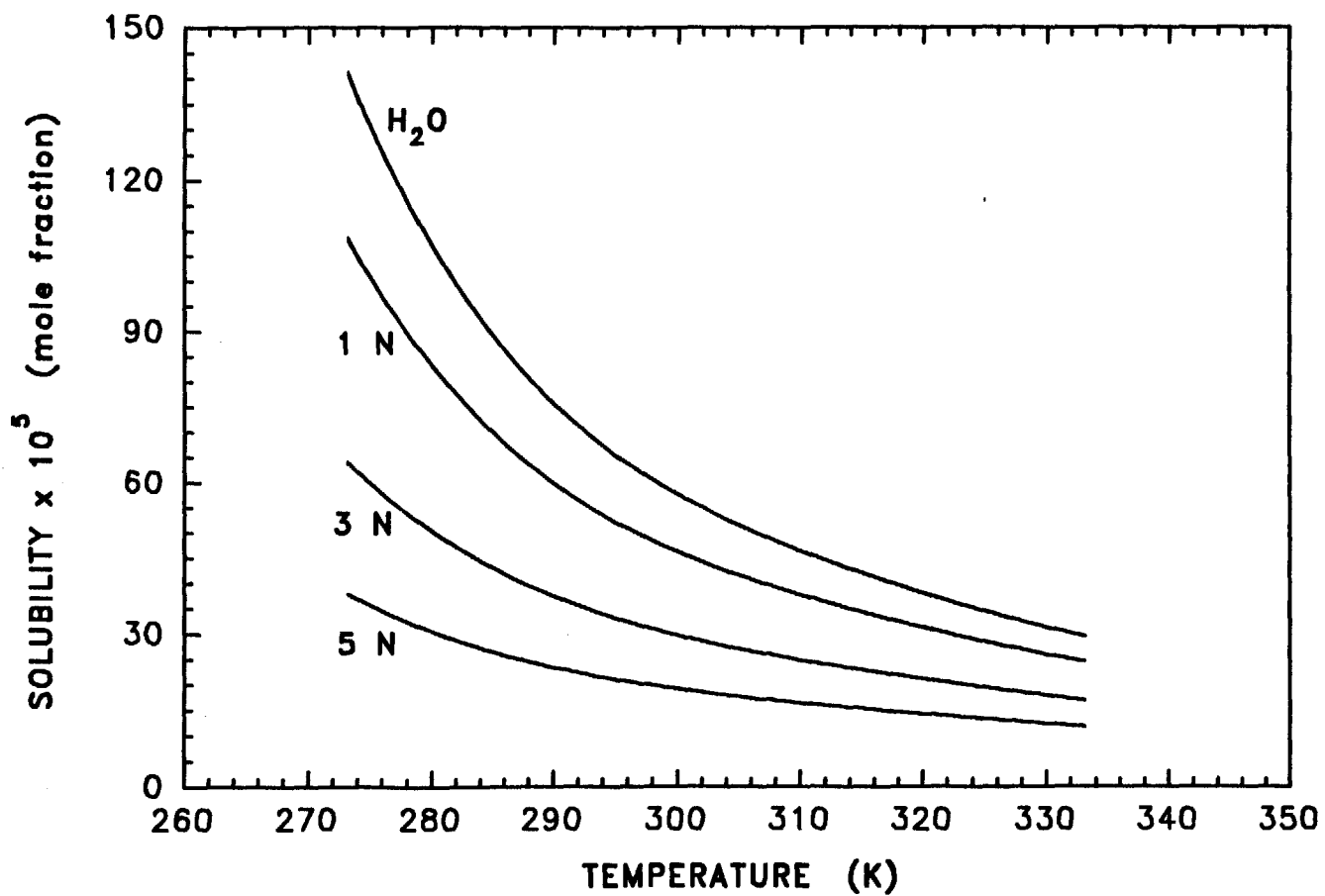


Figure 24

# 6  GAS SOLUBILITY IN BRINE AT ELEVATED PRESSURES

## 6.1  Hydrogen

No data exist for the solubility of hydrogen in brine at elevated pressures. A discussion is presented in Section 7 for estimating the solubility of hydrogen under these conditions based upon an extrapolation of the low pressure solubility data.

## 6.2  Nitrogen

Nitrogen solubility in NaCl brine at elevated pressures is best represented by the measurements of Wiebe et al. (1933), Smith et al. (1962), O'Sullivan et al. (1966), and O'Sullivan and Smith (1970). These studies provide solubility values for nitrogen that are within 2 percent agreement of each other. Several other studies are reported in the literature but have been critically rejected for inconsistencies in data, contamination, or some other rejection criterion (see Battino, 1982). Figure 25 provides the nitrogen solubility results of Smith et al. (1962) for pure water and brine solutions up to 5.47 N NaCl at 303.15 K as a function of pressure up to 7 MPa. Figure 26 presents a similar solubility plot for the data of O'Sullivan and Smith (1970) but at several higher temperatures as a function of pressure up to 60 MPa. Very little effect of temperature upon the nitrogen solubility is apparent. The mole fraction solubility data follow similar trends of nearly linearly increasing values with pressure for both pressure

NITROGEN SOLUBILITY IN SODIUM CHLORIDE SOLUTION
at low pressures

Figure 25

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NITROGEN SOLUBILITY IN SODIUM CHLORIDE SOLUTION
at elevated pressures
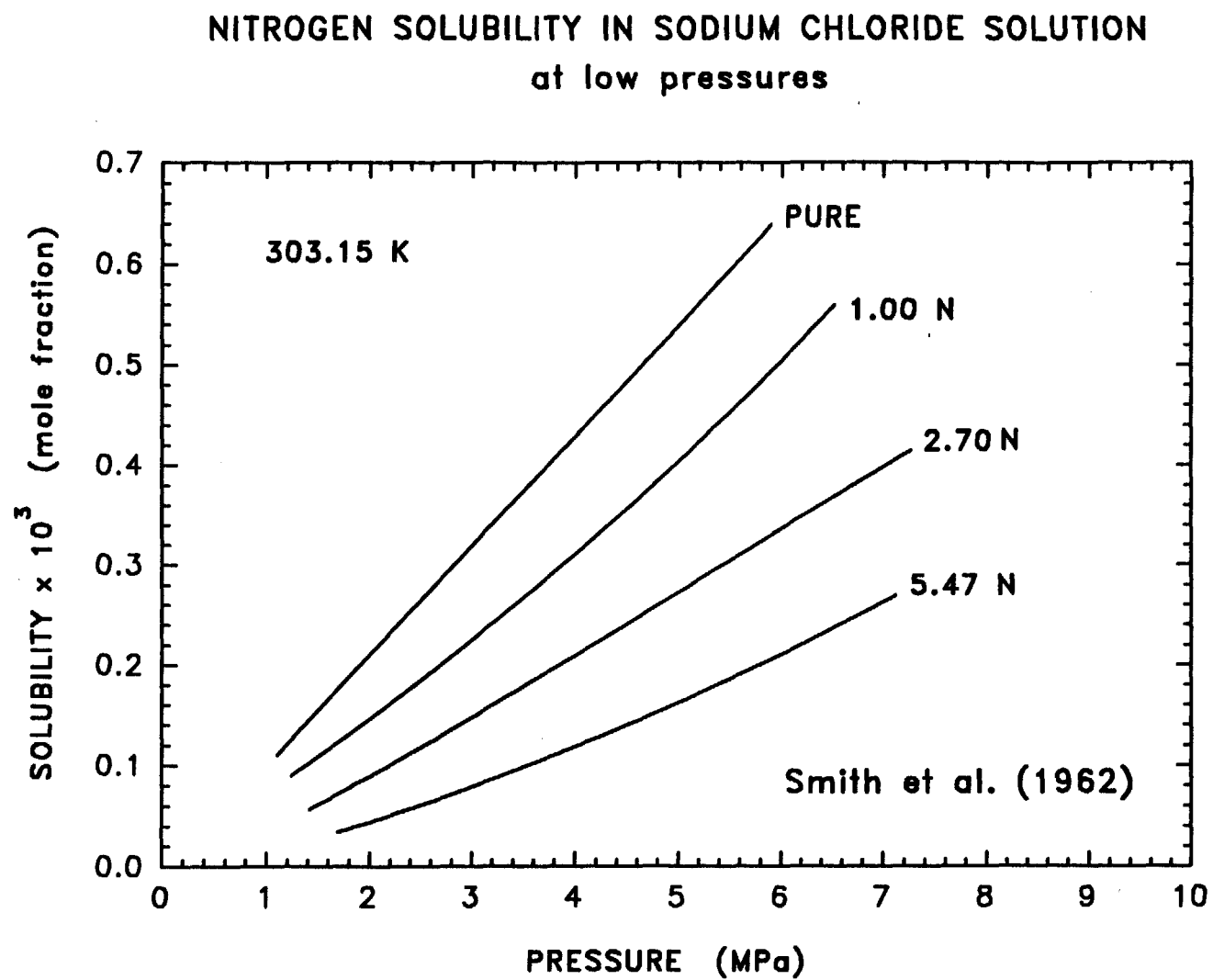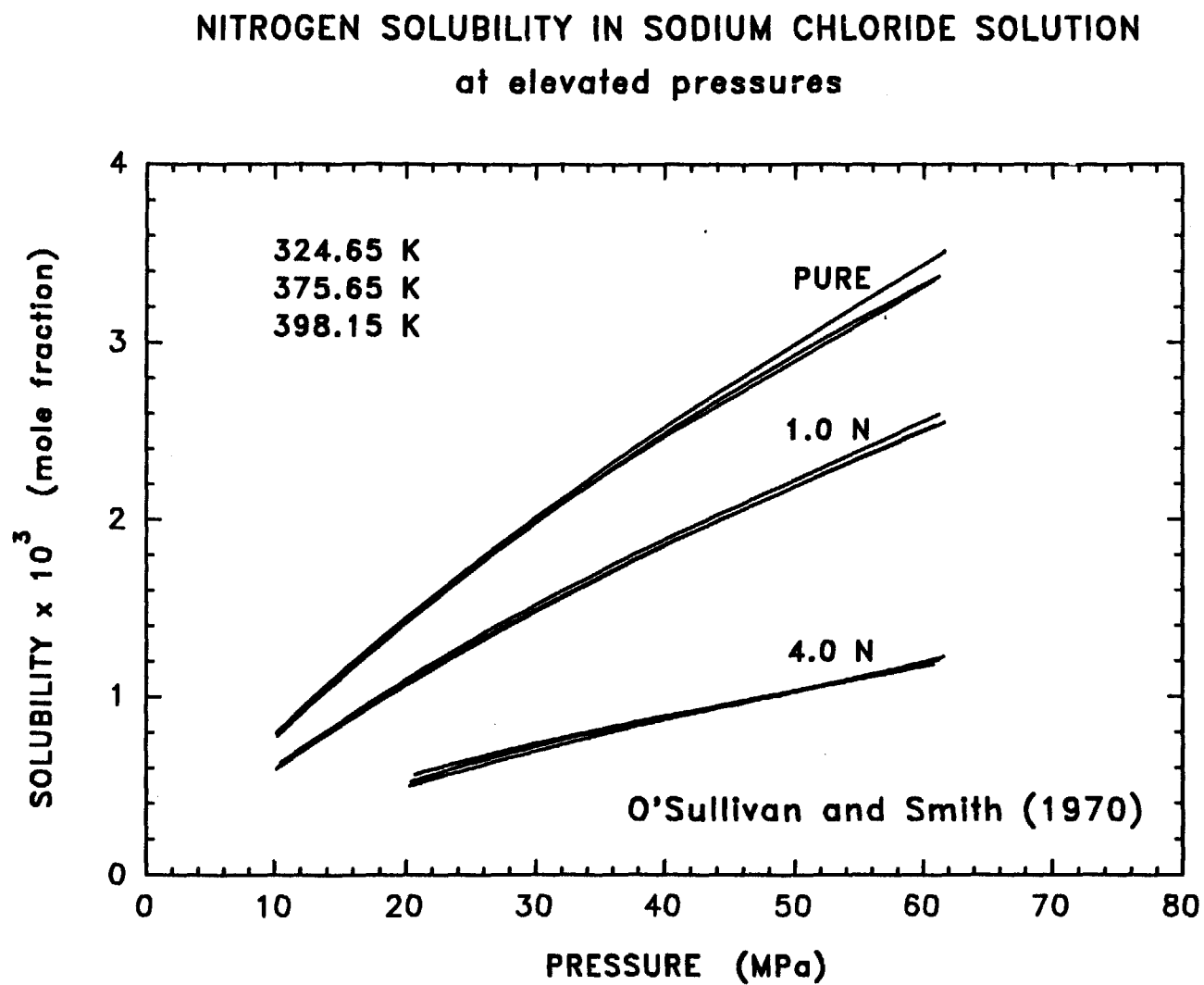


Figure 26

ranges. Increasing the brine concentration decreases the solubility as expected. The pressure derivatives of the solubility decrease considerably with increasing brine concentration.

## 6.3 Methane

Numerous studies reported in the literature have examined the solubility of methane in brine solutions at elevated temperatures and pressures. The studies of Michels et al. (1936), Culberson et al. (1950), Culberson and McKetta (1951), Duffy et al. (1961), O'Sullivan and Smith (1970), Blanco and Smith (1978), Haas (1978), Blount et al. (1980), Hanor (1980, 1981), Johnson (1980), McGee et al. (1981), Byrne and Stoessell (1982), Stoessell and Byrne (1982), Cramer (1984), and Johnson and Chao (1988) have examined the variability of methane solubility as a function of temperature and pressure for a wide variety of electrolyte compositions. Clever and Young (1987) have critically evaluated most of these studies. The recommended methane solubilities that provide internally consistent data sets and which are in general agreement with each other are presented in the following figures.

Figure 27 provides the results of Duffy et al. (1961) for the variation of solubility for water and brine concentrations up to 5.4 N NaCl at 303.15 K as a function of pressure up to 7 GPa. Figure 28 presents a similar solubility plot for the data of O'Sullivan and Smith (1970) but at several higher temperatures as a function of pressure up to 60 MPa. As with the previous nitrogen solubility data, little effect of temperature upon the methane solubility is displayed. The mole fraction solubility data follow similar trends of nearly linearly

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METHANE SOLUBILITY IN SODIUM CHLORIDE SOLUTION
at low pressures

Figure 27

METHANE SOLUBILITY IN SODIUM CHLORIDE SOLUTION
at elevated pressures
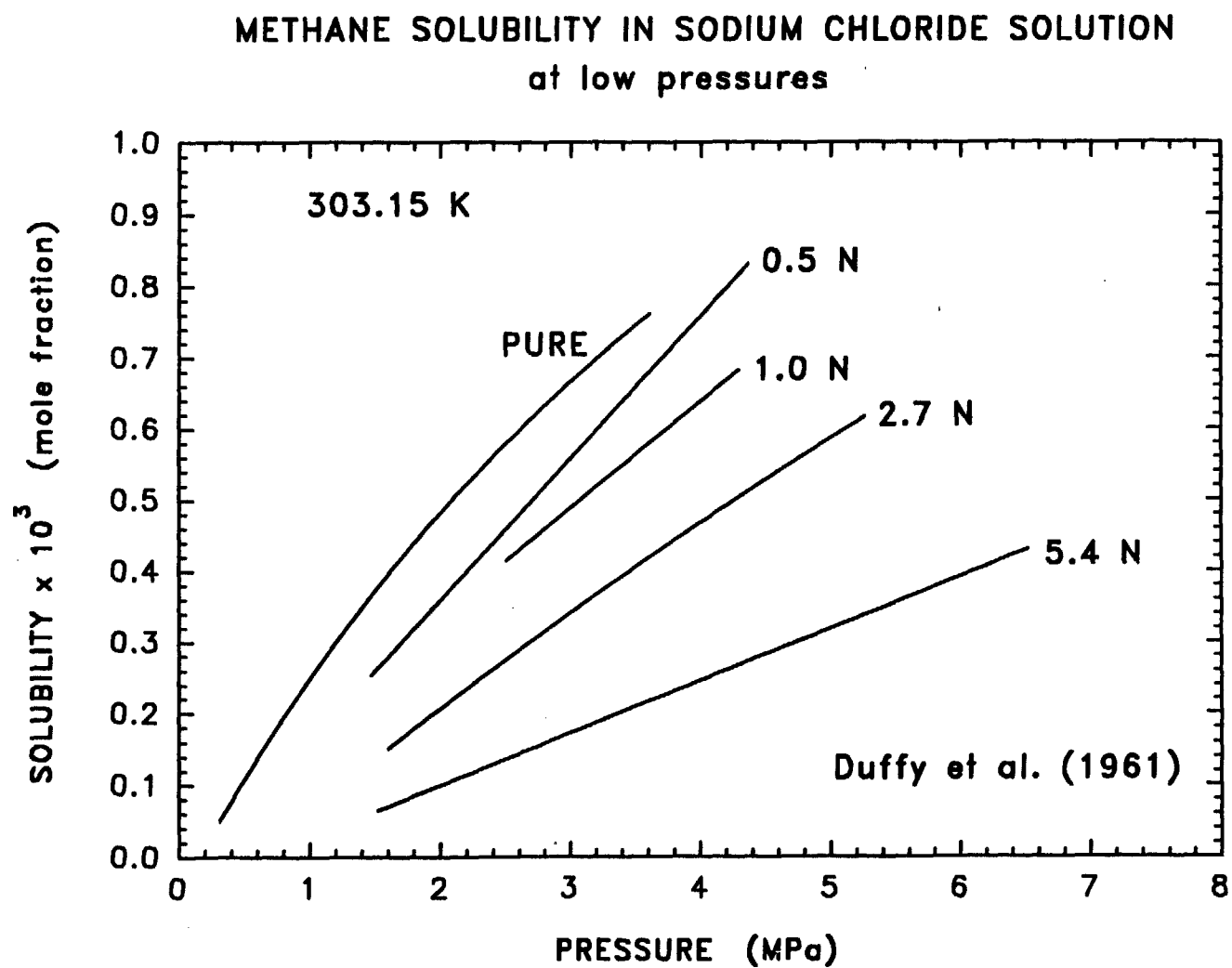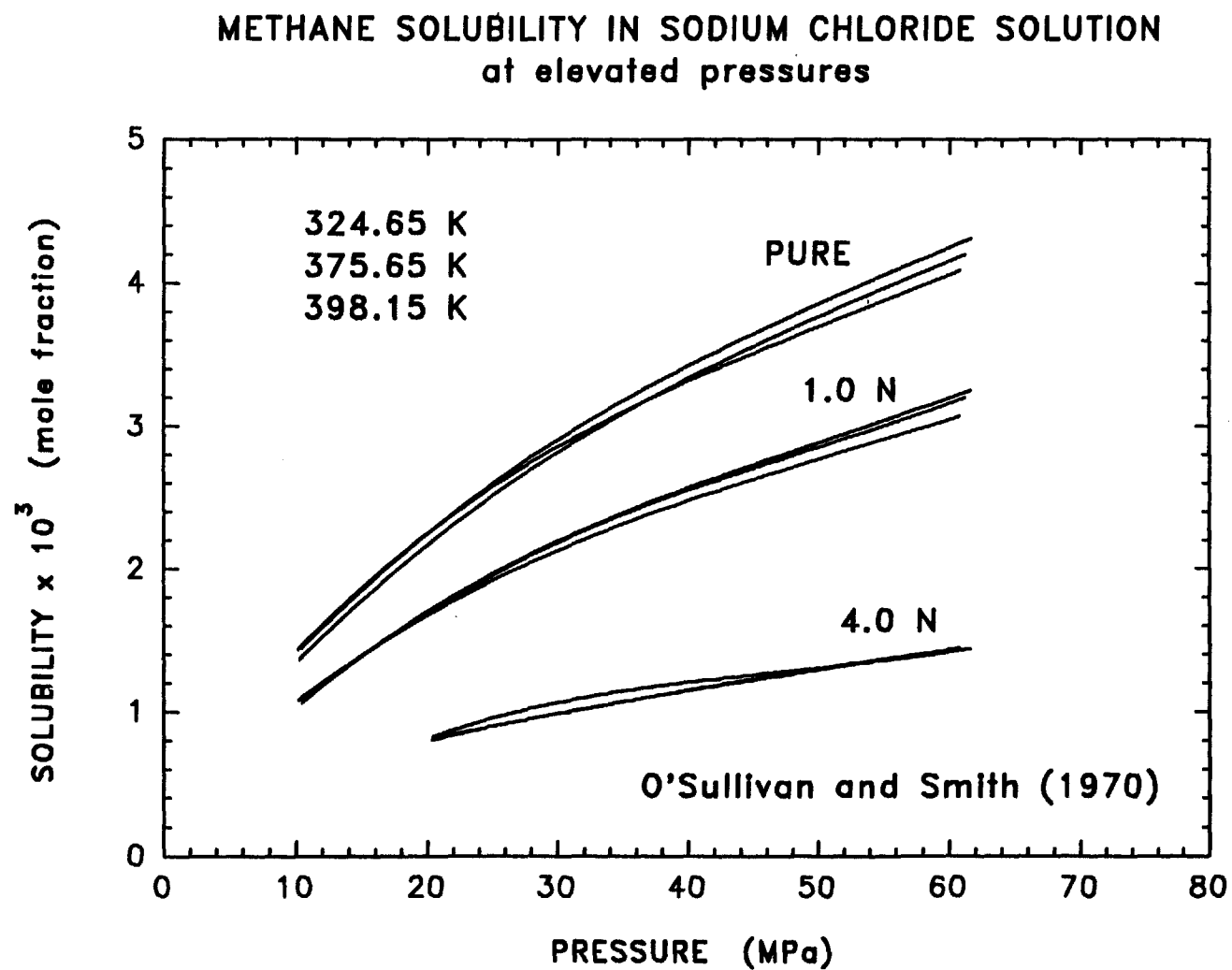
Figure 28

increasing values with pressure for both pressure ranges. Increasing

the brine concentration decreases the solubility as expected. The

pressure derivatives of the methane solubility decrease with increasing

brine concentration. Figure 29 provides the solubility data of Michels

et al. (1936) for water and several sodium chloride solutions at 298.15

K as a function of pressure up to 47 GPa. Slightly lower solubility

values than those of O'Sullivan and Smith (1970) are exhibited.


Figures 30 through 33 present the methane solubility data of McGee

et al. (1981) for various sodium chloride brines as a function of fairly

elevated temperatures and pressures. The solubility data for the pure

water solvent from this study are given in Figure 12. Section 2.6

discusses the details of the experimental approach used by McGee et al.

(1981). These results exhibit consistent trends of the methane

solubility to decrease with increasing sodium chloride content. The

high pressure isobars display slight minima in the solubility and a

significant increase in solubility at the high temperature (above 550

K).


6.4 Carbon Dioxide


Data for the solubility of carbon dioxide in aqueous sodium

chloride solutions at elevated pressures is provided in the studies of

Ellis and Golding (1963), Takenouchi and Kennedy (1965), Malinin and

Savelyeva (1972), Malinin and Kurovskaya (1975), and Nighwsander et al.

(1989). The study of Takenouchi and Kennedy (1965) provides a complete

data set of solubilities for a broad range of conditions. Figures 34

METHANE SOLUBILITY IN SODIUM CHLORIDE SOLUTION
at elevated pressures

298.15 K

PURE

1.0 N

1.7

2.5

2.7

3.2

5.4

4.2

Michels et al. (1936)

SOLUBILITY x $10^3$ (mole fraction)

PRESSURE (MPa)

Figure 29

METHANE SOLUBILITY IN SODIUM CHLORIDE SOLUTION
at elevated pressures
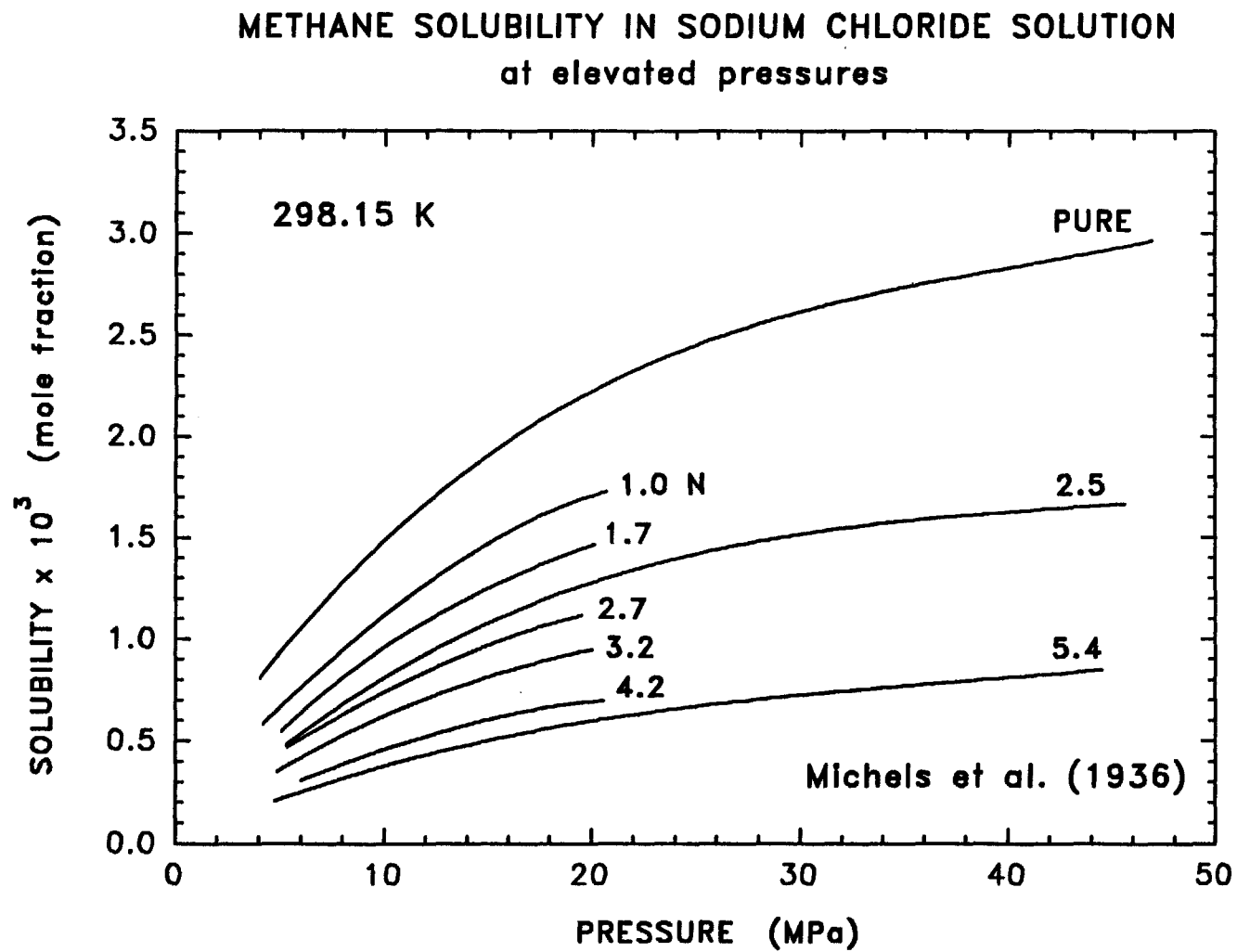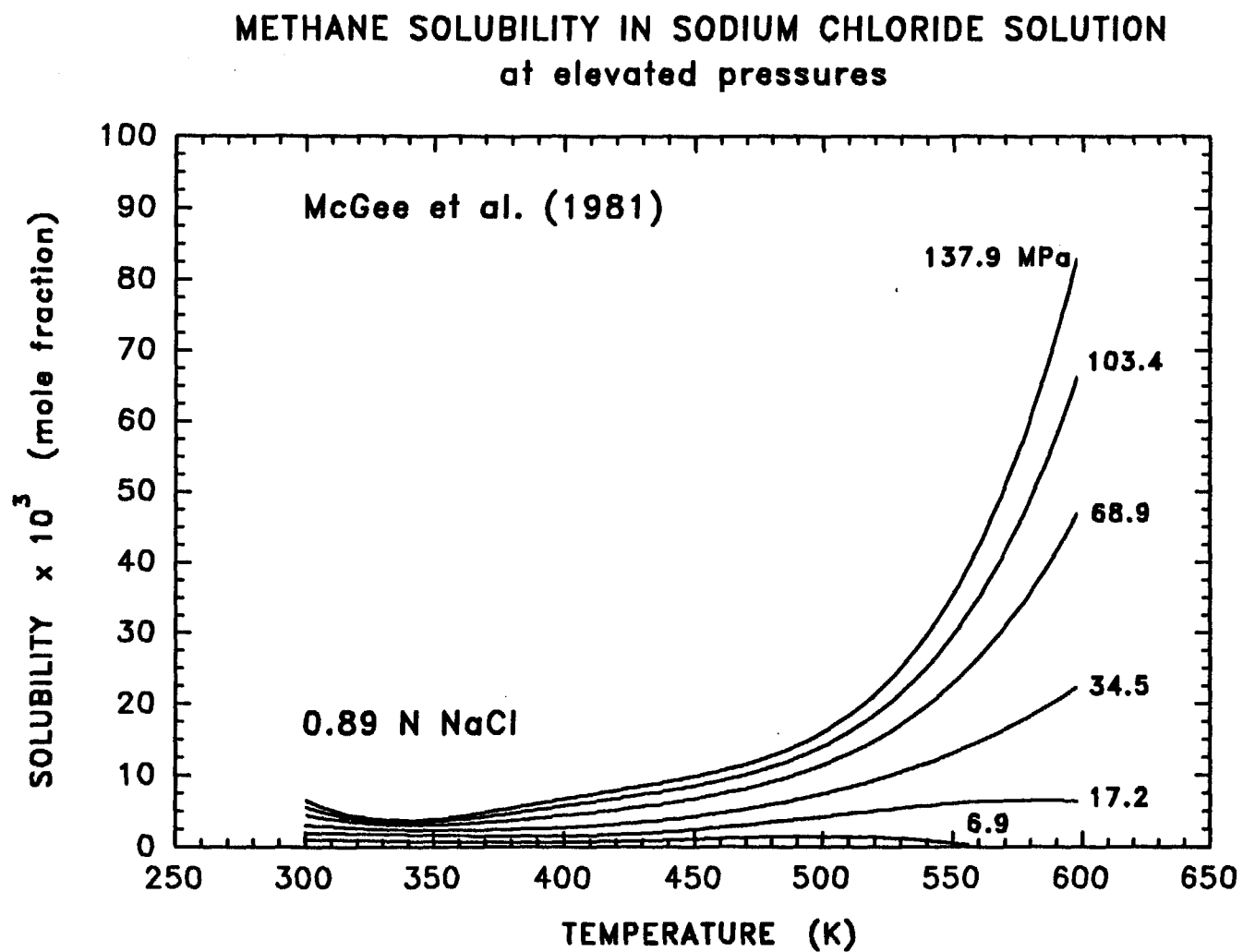
Figure 30

METHANE SOLUBILITY IN SODIUM CHLORIDE SOLUTION
at elevated pressures
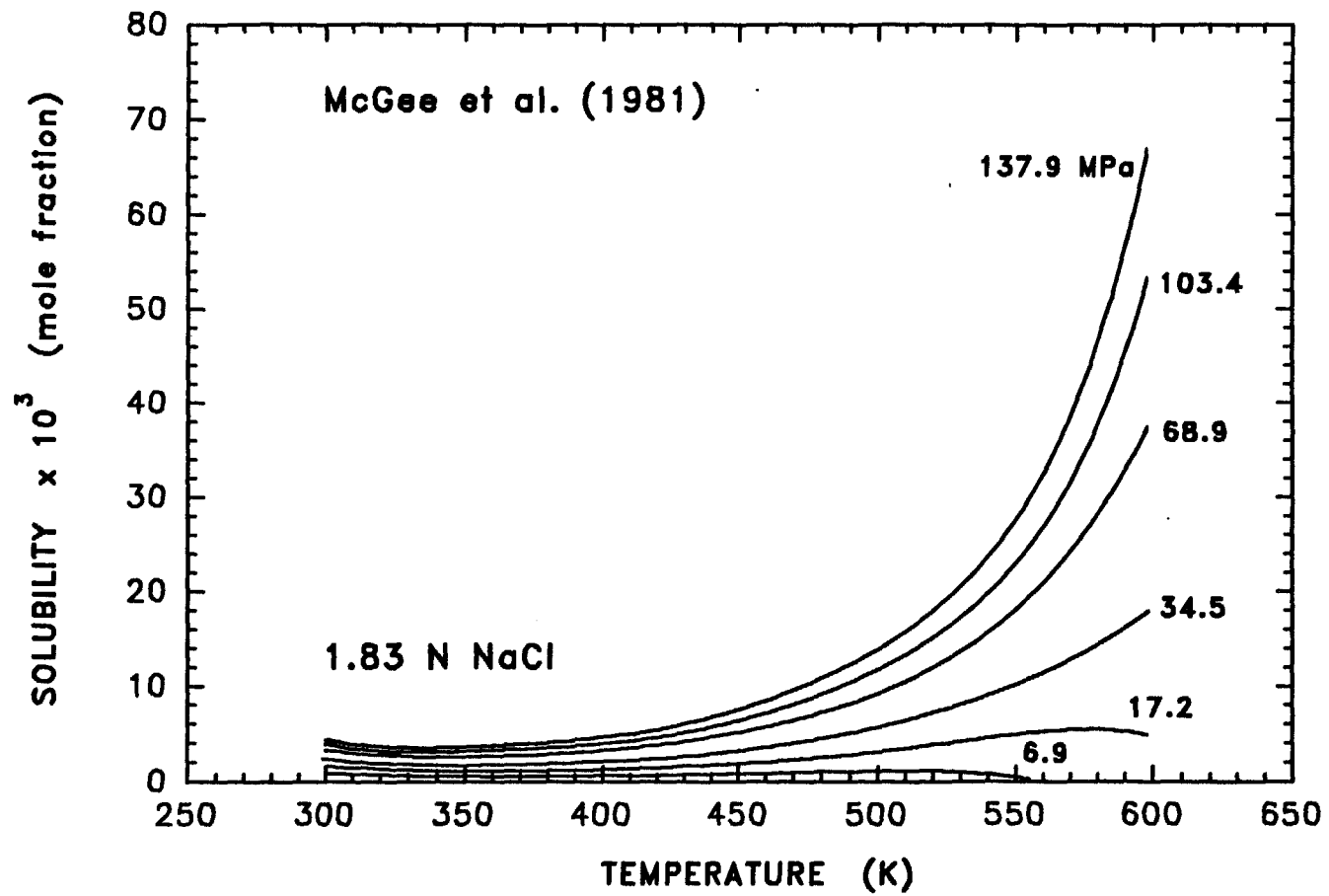
Figure 31

# METHANE SOLUBILITY IN SODIUM CHLORIDE SOLUTION
## at elevated pressures
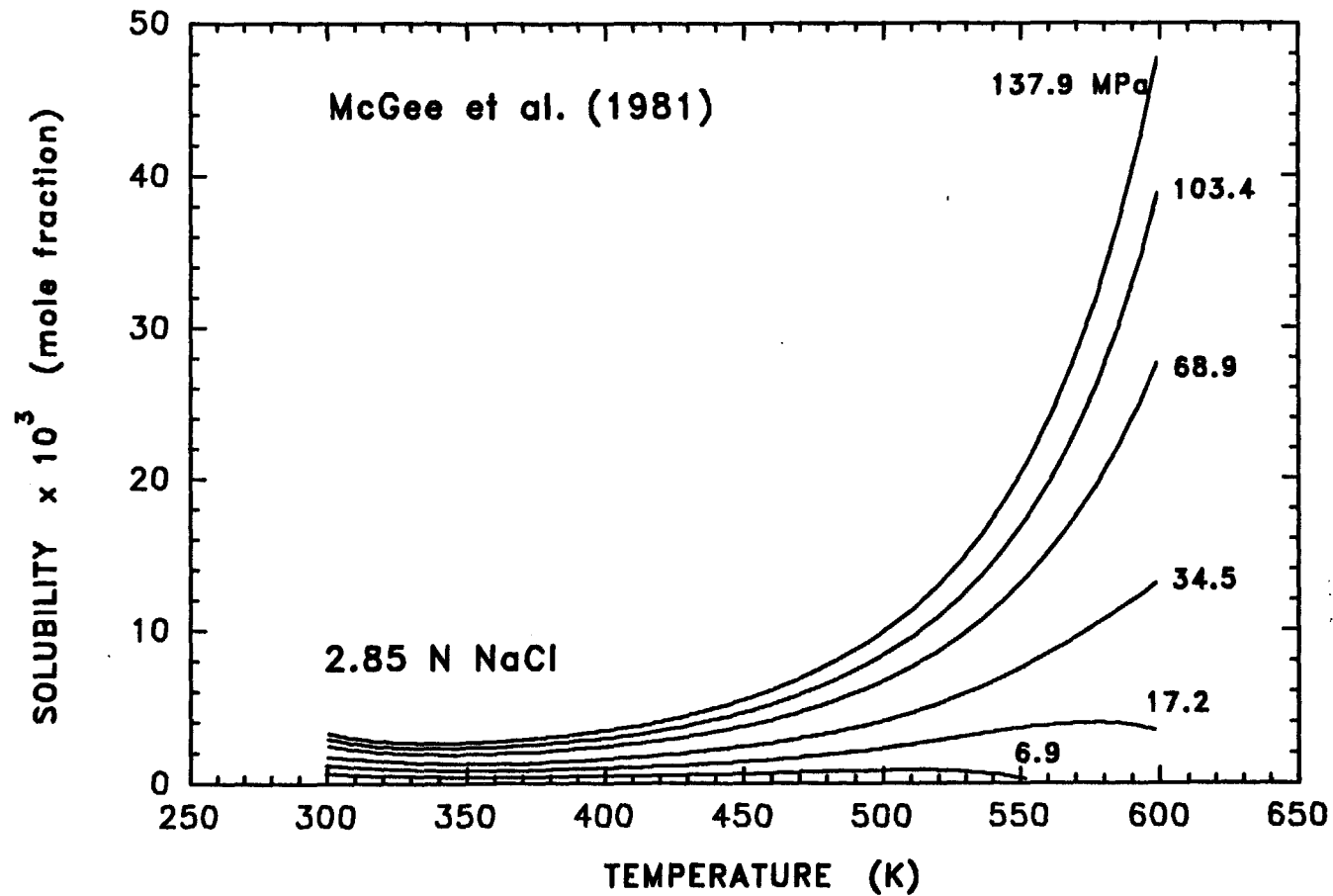


Figure 32

METHANE SOLUBILITY IN SODIUM CHLORIDE SOLUTION
at elevated pressure
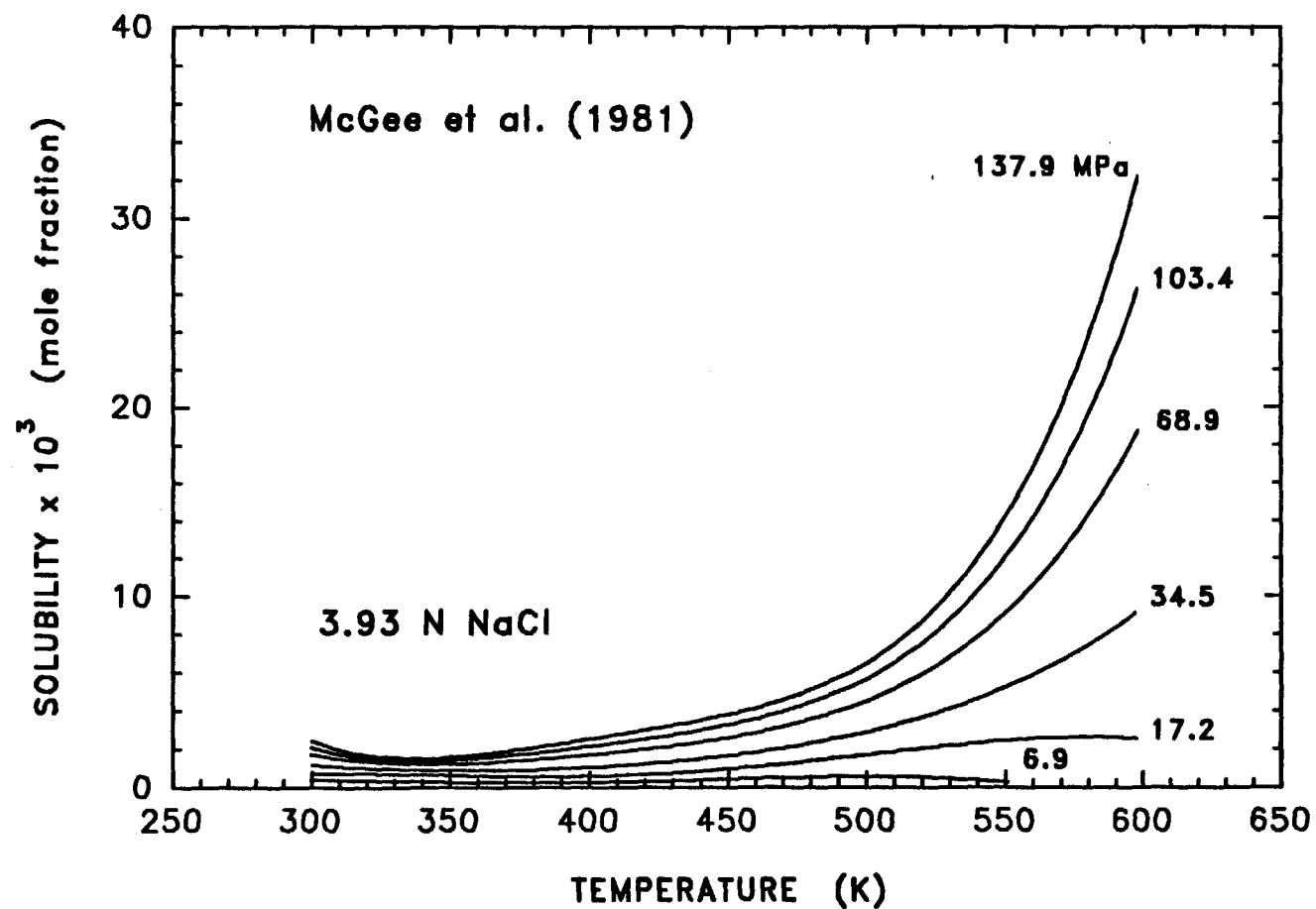
Figure 33

and 35 provide these results in plots of carbon dioxide solubility isotherms for, respectively, 1.07 N NaCl and 3.93 N NaCl solutions as a function of pressure up to 140 MPa. The solubility data for pure water solvent under these pressure-temperature conditions is given in Figure 16. As observed for nitrogen and methane, the solubility of the gas decreases with increasing sodium chloride content. The high temperature isotherms exhibit the greatest pressure dependence due to the proximity to the critical point of the system. The critical point is displaced to higher temperatures and pressures as the solution becomes more concentrated in sodium chloride (compare with Figure 16). Figure 36 presents the data of Malinin and Savelyeva (1972) in terms of the mole fraction solubility at 4.90 MPa as a function of solution concentration for three relatively low temperatures. The solubility curves display less of a concentration dependence with increasing temperature.

CARBON DIOXIDE SOLUBILITY IN SODIUM CHLORIDE SOLUTION
at elevated pressures

Takenouchi and Kennedy (1965)

1.07 N NaCl
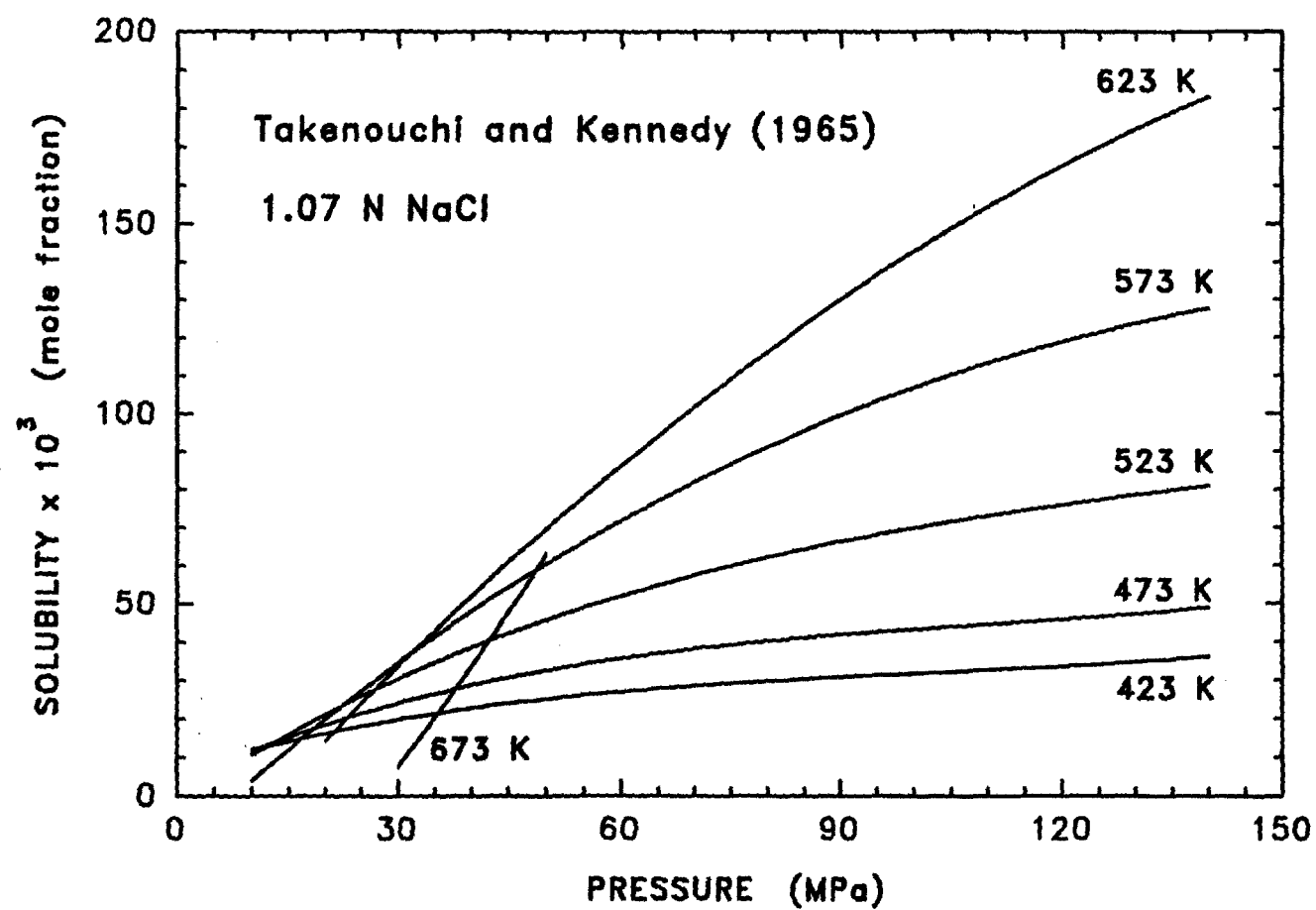
Figure 34

**CARBON DIOXIDE SOLUBILITY IN SODIUM CHLORIDE SOLUTION**
**at elevated pressures**
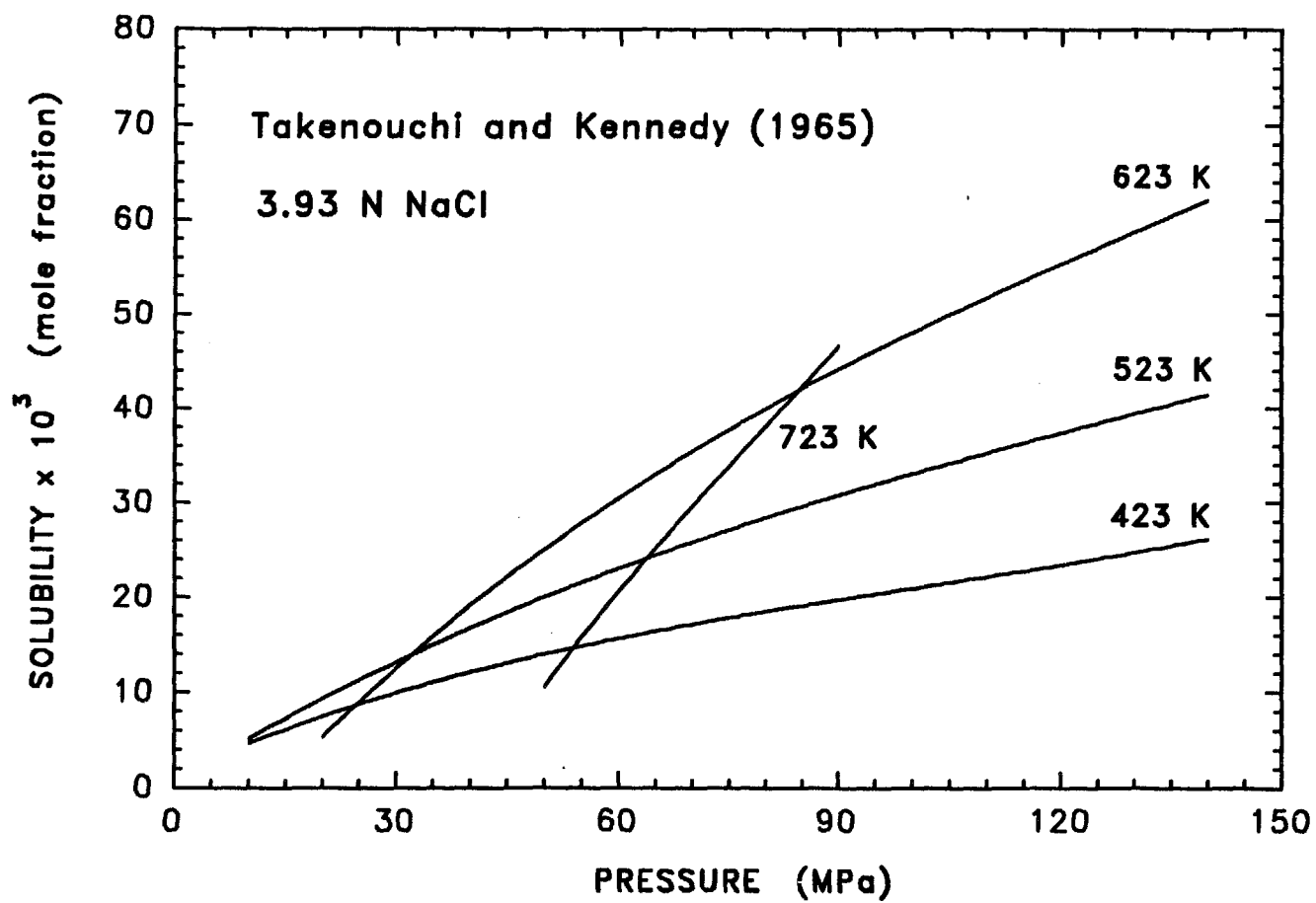


Figure 35

CARBON DIOXIDE SOLUBILITY IN SODIUM CHLORIDE SOLUTION
at 4.90 MPa partial pressure
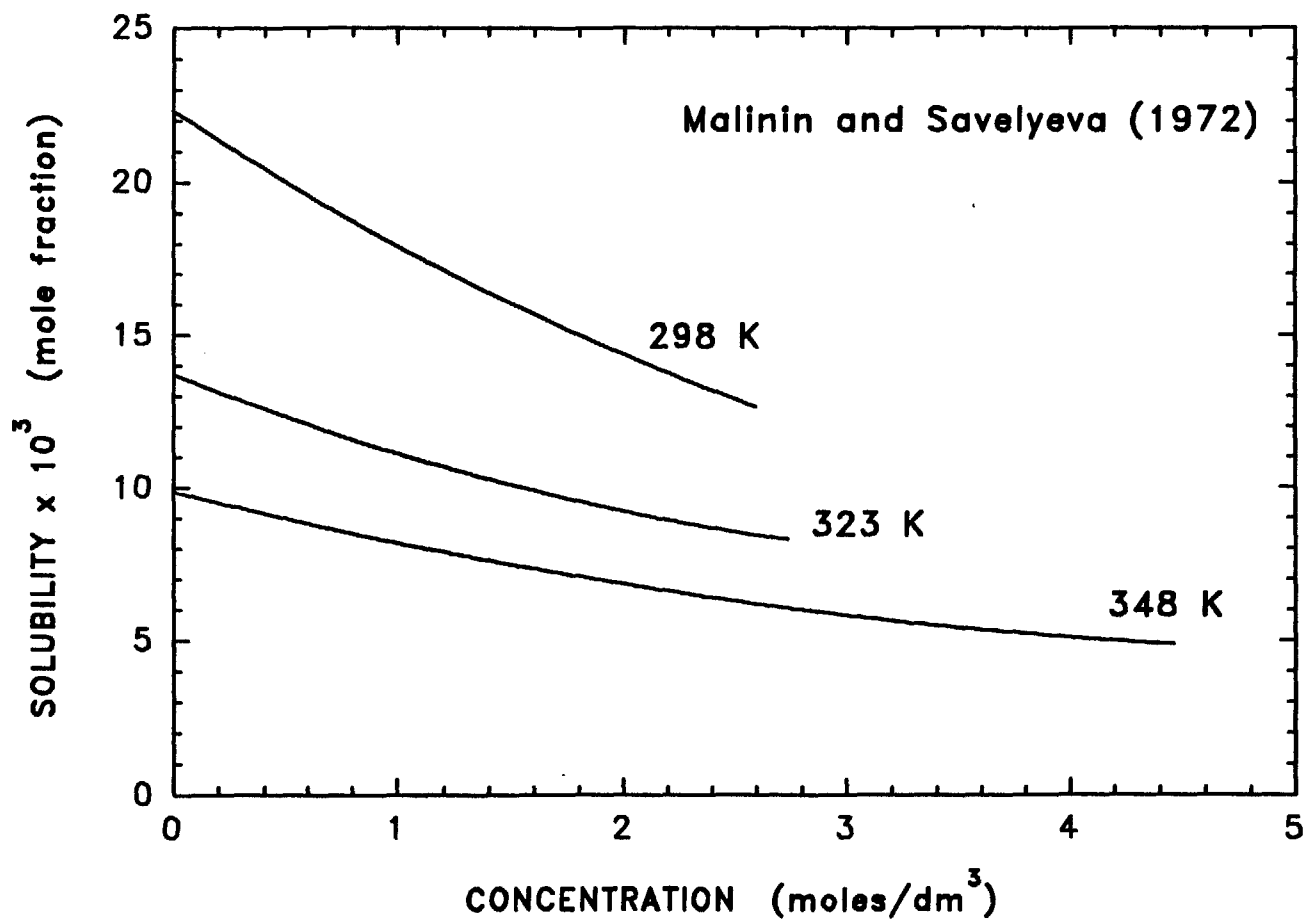


Figure 36

# 7  ESTIMATION OF GAS SOLUBILITY


As noted in Section 6.1 there are no data for the solubility of hydrogen in brines, or similar electrolyte solutions, at elevated pressure.  This paucity of hydrogen solubility data reflects the limited occurrence of hydrogen gas at elevated pressures in nature and, probably, in engineering needs.  In addition, safety concerns over the handling of combustible hydrogen gas at these pressures would limit any experimental study.  On the other hand, suitable data for nitrogen, methane, and carbon dioxide solubility in NaCl brine at pressures up to 140 MPa are available (see Sections 6.2 - 6.4).  It would be beneficial to examine the existing solubility pressure derivatives in order to estimate the behavior of hydrogen solubility in brine at these elevated pressures.


The mole fraction solubility of hydrogen gas in pure water as a function of pressure is presented in Figure 7 based upon the data of Wiebe and Gaddy (1934).  More recent solubility data exist for hydrogen at elevated pressure (see Young 1981), however, these data are not as precise nor internally consistent as this older study, nor do they cover an extensive pressure range.  There is some general agreement of the solubility values for these recent studies with those of Wiebe and Gaddy (1934).


To estimate the effect of pressure upon the solubility of hydrogen in NaCl brine, it is first necessary to evaluate the solubility of

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hydrogen in brine at 0.101325 MPa. This is accomplished using the

Sechenov coefficients of Figure 17 derived from the 1 N NaCl data of

Morrison and Billet (1952) and Crozier and Yamamoto (1974), and the

solubility data for hydrogen in pure water (Figure 7). Calculated

results for a 5 N NaCl solution as a function of temperature are

presented in Figure 18. Only the solubility data derived for room

temperature 298.15 K will be incorporated in the estimation scheme.


The effect of pressure upon the hydrogen solubility for the 5 N

NaCl solution is quite difficult to evaluate without experimental data.

Nonetheless, an extrapolation of 0.101325 MPa hydrogen solubility can be

performed by making use of the available nitrogen data. The pressure

dependence of hydrogen solubility in the electrolyte solution is assumed

to be similar to that of nitrogen solubility, for which high pressure

data for brines are available (see Figure 26). Additionally, both

hydrogen and nitrogen have similar physiochemical properties and exhibit

approximate ideal behavior at low pressures. The pressure dependence of

hydrogen and nitrogen solubilities in pure water exhibit minimal effects

of changing temperature (see Figures 6-9). This is also true for the

solubility of nitrogen in solutions of various concentrations of

electrolytes (see Figure 26).


An analytical derivative was obtained for the pressure dependence

of the nitrogen solubility in NaCl solutions from the third order

polynomial regression presented in Figure 26. The 4.0 N NaCl data for

nitrogen of O'Sullivan and Smith (1970) was chosen for this purpose in

order to approximate the desired 5 N NaCl brine concentration for

hydrogen. A maximum estimate of hydrogen solubility will therefore be obtained from these approximations because solubility decreases with increasing NaCl concentration. Note also that the pressure dependence of gas solubility normally decreases with increasing NaCl concentration as exhibited in Figure 26 for the nitrogen data. It is then possible to generate the hydrogen solubility in 5 N NaCl brine as a function of pressure from the previously calculated value of hydrogen solubility at 298.15 K and 0.101325 MPa. This involves the use of a mid-pressure numerical extrapolation using the pressure derivative from the nitrogen data.

The results of these calculations are presented in Figure 37 for 298.15 K and pressures ranging from 0 MPa (0.101325 MPa) to 50 MPa. The graph provides the hydrogen solubilities (filled circle symbols) obtained from the third order polynomial regression of the experimental data of Wiebe and Gaddy (1934) for pure water and those estimated for the 5 N NaCl solution. The solid lines represent the regressions of these data to the two term pressure dependence equation for gas solubility (modified form of equation (16)):

$$\ln x_{H_2} = D_0 + D_1 \ln P \qquad (18)$$

$x_{H2}$ is the hydrogen solubility in mole fraction and P is the pressure (partial pressure) in MPa. The best fit values of $D_0$ and $D_1$ are, respectively, -8.8980 and 0.9538 for pure water and -10.0789 and 0.8205 for the 5 N NaCl brine.

CALCULATED HYDROGEN SOLUBILITY IN SODIUM CHLORIDE
SOLUTION AT ELEVATED PRESSURES
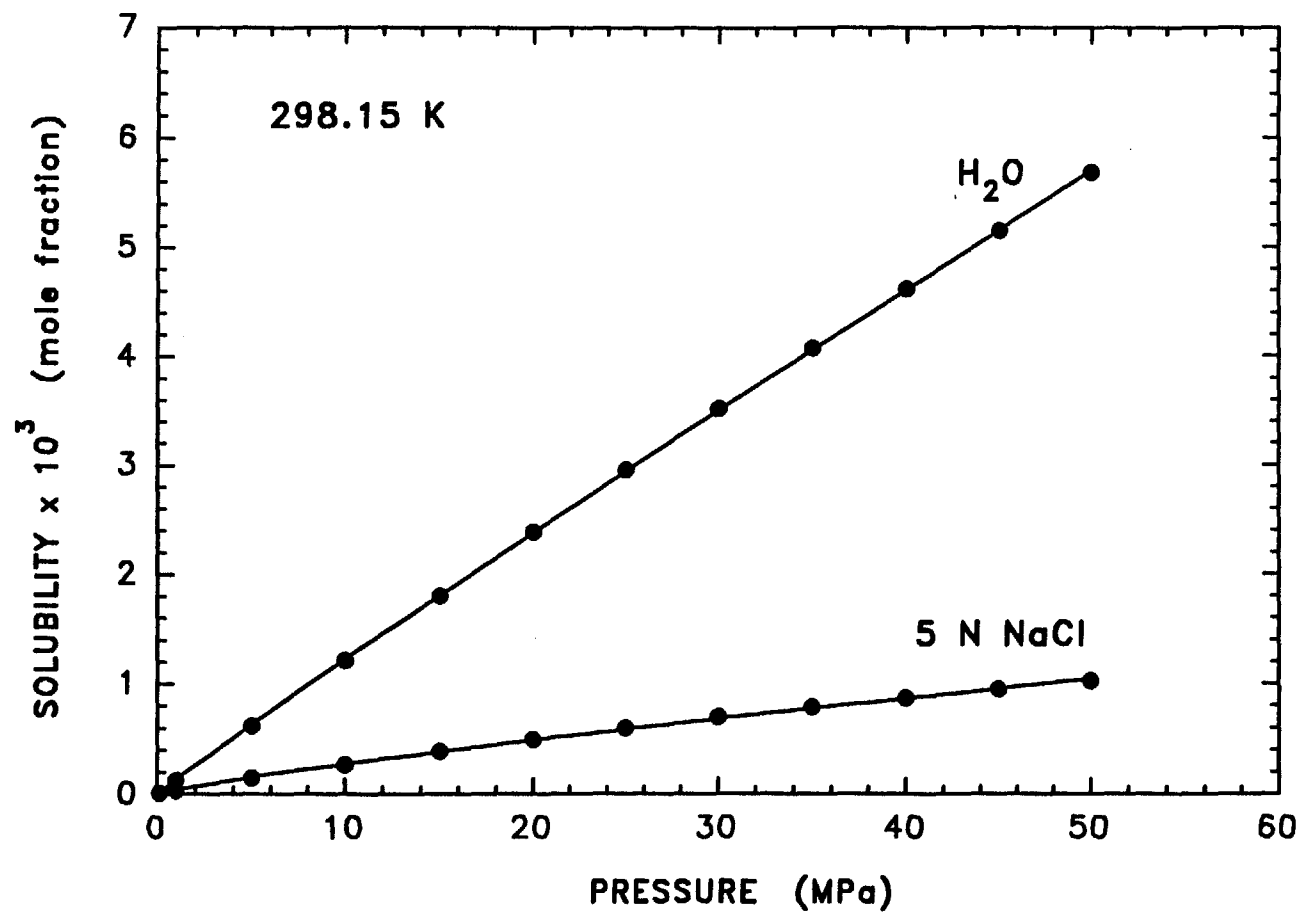
Figure 37

It should be emphasized that these calculations represent an educated estimate of the solubility behavior of hydrogen in NaCl brine at elevated pressure. These estimates should be treated as such and should in no way be construed as accurately representing the real hydrogen gas solubility. Unfortunately, due to the assumptions and methods used in the above calculations it is impossible to provide limits or confidence levels on the accuracy of the estimated hydrogen solubilities. Furthermore, there is no substitute for precise and accurate gas solubility experiments performed at the desired temperature and pressure conditions.

# 8  SUMMARY AND CONCLUSIONS

The difference in solubilities observed for gases in liquid is
dictated by the fundamentals of thermodynamics in assessing the free
energy difference between gas in a vapor phase and the gas dissolved in
a liquid.  Sechenov coefficients quantify the role of solution
electrolytes in modifying the solubility of a gas and, in addition,
provide a means of evaluating the activity coefficient of the gas
species.  A critical evaluation of numerous gas solubility studies from
the literature has provided a graphical summary of the data for the
solubility of hydrogen, nitrogen, methane, and carbon dioxide in pure
water and NaCl brine.

In general, the mole fraction solubility of a gas in water or brine
decreases with increasing temperature at a fixed pressure.  All of the
gases show a slight increase in solubility or a trend towards increasing
solubility at temperatures above 320 K.  This effect is enhanced at
elevated pressures.  Because of its reactivity with the water solvent,
carbon dioxide exhibits an approximate fifty-fold higher mole fraction
solubility relative to the other gases for a given temperature.  In pure
water, the solubility of the gas increases with increasing partial
pressure at a fixed temperature.  Sechenov coefficients for NaCl
solutions tend to decrease with increasing temperature, although the
nitrogen and carbon dioxide values exhibit an increase at temperatures
above 350 K.  Increasing the electrolyte concentration of a solution
will decrease the gas solubility.  The data for nitrogen, methane, and

carbon dioxide suggest that their solubilities at approximately 50 MPa will be decreased by a factor of 3 to 5 as the NaCl brine concentration increases from zero to near saturation. No solubility data for hydrogen in NaCl brine at elevated pressures are available, although calculations suggest that hydrogen will have a similar decrease in solubility.

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