

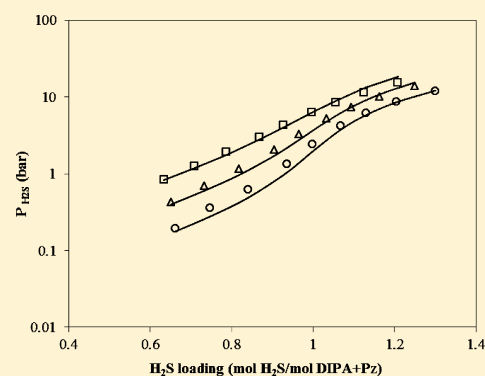
# Solubility of H<sub>2</sub>S in Aqueous Diisopropanolamine + Piperazine Solutions: New Experimental Data and Modeling with the Electrolyte Cubic Square-Well Equation of State

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**ABSTRACT:** The solubilities of H<sub>2</sub>S in aqueous solutions of diisopropanolamine (DIPA) and in aqueous mixtures of DIPA and piperazine (Pz) have been measured. The molality of DIPA was fixed at 2.96 *m*, and the concentration of Pz was (1.20 or 1.80) *m*. Experiments were carried out at (40, 60, and 80) °C over the pressure range (19 to 1554) kPa. For modeling of the DIPA + Pz + H<sub>2</sub>S + H<sub>2</sub>O system, the electrolyte cubic square well (eCSW) equation of state (EOS) (Haghtalab, A.; Mazloumi, S. H. *Fluid Phase Equilib.* 2009, 285, 96–104) was applied to predict the total and partial pressures of H<sub>2</sub>S over aqueous solutions of DIPA, Pz, and DIPA + Pz using only the interaction parameters,  $k_{ij}$ , of the H<sub>2</sub>S + H<sub>2</sub>O system. The reasonable agreement of the eCSW EOS and experimental results demonstrates the good accuracy of the eCSW EOS for thermodynamic modeling of the solubilities of acid gases in aqueous alkanolamine solutions.



## INTRODUCTION

The experimental determination and modeling of the solubilities of acid gases in aqueous alkanolamine solutions, such as aqueous solutions of monoethanolamine (MEA), methyldiethanolamine (MDEA), diethanolamine (DEA), diisopropanolamine (DIPA), and diglycolamine (DGA), is one of the major fields in chemical engineering thermodynamics, because of their applications in the design of various chemical plants, such as natural gas, refinery, and tail gas treatments. DIPA is one of the major H<sub>2</sub>S-selective alkanolamines, and it is low-corrosive and has high potential for removal of the other sulfur compounds, such as COS and CS<sub>2</sub>. This amine is utilized in the sulfinol process, which is usually used for those streams that have a H<sub>2</sub>S/CO<sub>2</sub> ratio greater than 1, where it is not necessary to remove CO<sub>2</sub> at the same levels as required for H<sub>2</sub>S removal. Also, it is used in the Shell Claus off-gas treating (SCOT) process when selective elimination of H<sub>2</sub>S over CO<sub>2</sub> is needed.<sup>1</sup> One of the reasons for the applicability of this alkanolamine is that its reactivity with H<sub>2</sub>S is fast, as for the other amines, so the protonation reaction can progress instantaneously without kinetic control. On the other hand, the reaction of CO<sub>2</sub> with DIPA, and alkanolamines generally, is kinetically controlled and slow. To improve the potential of DIPA for better removal of CO<sub>2</sub>, an alkanolamine with high reactivity with CO<sub>2</sub> should be added to the solvent as an activator. One widely used activator that is already known to be a very good additive to accomplish this purpose is piperazine (Pz).<sup>2</sup>

In spite of the wide application of DIPA in acid gas removal units, experimental data on the solubilities of acid gases in this alkanolamine by itself or in its blends are rare,<sup>3</sup> and thus, the

first and main aim of this study was to obtain experimental data on the solubility of H<sub>2</sub>S in aqueous solutions of DIPA and activated DIPA (i.e., DIPA + Pz). The other objective of the present work was to model the experimental data using the equation of state (EOS) approach, which is a rather new application of EOSs for such systems.<sup>4–9</sup> Thus, the electrolyte cubic square well (eCSW) EOS<sup>10,11</sup> was applied to correlate and predict the solubilities of H<sub>2</sub>S in aqueous solutions of DIPA, Pz, and DIPA + Pz.

## EXPERIMENTAL SECTION

**Materials.** H<sub>2</sub>S (mass fraction purity > 0.999) was purchased from Linda Company, DIPA (mass fraction purity 0.98) from Merck, and Pz (mass fraction purity 0.99) from Acros, and they were used without further purification. Distilled, deionized, and degassed water were employed to prepare aqueous alkanolamine solutions.

**Apparatus.** In this work, the solubility measurements were carried out using a synthetic–static apparatus in which a predefined amount of the solution is fed into a cell, the equilibrium temperature and pressure are monitored, and then the amount of the dissolved gas is calculated.<sup>5</sup> Figure 1 presents the apparatus used in this study. The main part of the system is the equilibrium cell, which is made of 316 stainless steel, has one side glass window, has a volume is 117 ± 0.7 cm<sup>3</sup>, and is equipped with a heat jacket. Agitation of the liquid phase is

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