



# Experimental characterization of multi-component absorption in complex liquid: New method and apparatus



David Benizri, Nicolas Dietrich, Gilles Hébrard \*

Laboratoire d'Ingénierie des Systèmes Biologiques et des Procédés (LISBP), Université de Toulouse, CNRS, INRA, INSA, Toulouse, France

## HIGHLIGHTS

- The Henry's law constant determination for multi-component gases in presence of complex liquids.
- New experimental setup built to measure under pressure the Henry's law constants easily.
- Three types of water were tested and put in contact with a real biogas mix.
- Henry's law constants for carbon dioxide and methane were obtained with good accuracy.
- Henry's law constant can differ between tap water and recycled water; it is strongly reduced when recycled water is used.

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## ABSTRACT

The Henry's law constant is required to size absorption processes. However, its experimental determination for multi-component gases in presence of complex liquids remains problematic in the process industry and research field. This work presents a new experimental setup built to measure Henry's law constants easily. The apparatus brings gas and liquid phases into contact before and after equilibrium in medium pressure conditions (from 1 to 10 bars). Thanks to standard sensors, temperature, pressure and phase concentrations of the components are recorded continuously, permitting to calculate the Henry's law constant. Three types of water were tested and put in contact with a real biogas mix. Henry's law constants for carbon dioxide and methane were obtained with good accuracy and were consistent with the values reported in the literature for tap water. Results obtained with the apparatus show that values of Henry's law constant can differ between tap water and recycled water. Carbon dioxide absorption is strongly reduced when recycled water is used. We can conclude that the proposed apparatus is a convenient experimental set up for determining Henry's law constants with good accuracy and as it is movable it can be helpful for engineer in charge of biogas upgrading system.

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## 1. Introduction

The experimental characterization of fugacity equilibrium between a gas and a liquid phase in a complex gas/liquid system is a major issue in process sciences. It can be useful to validate thermodynamic modeling but it also brings answers for process design in cases where there is a lack of reliable models. The design of absorption and desorption processes in water requires a good knowledge of Henry's law constants (Roustan, 2003). Assuming that requirement and in order to design processes with water, chemical and process engineers use Henry's law constants that are commonly available (Sander, 2001; Reid and Prausnitz, 1986). Thus, engineers often choose to consider that non-polar

molecules, such as light hydrocarbons and Volatile Organic Compounds (VOC), do not dissolve in water. However, when the pressure is relatively high, this phenomenon should be considered. When the gas phase contains more than one gas dissolving in the liquid phase, the model of fugacity equilibrium between gas and liquid phases should be validated before designing the process. This experimental validation must be done in real conditions of operation (pressure, temperature and water quality). We can conclude that a simple method to determine Henry's law constants in real conditions could have a large field of application in the environmental industry and the development of renewable energies.

This work aims to set up and validate a new apparatus that is able to control and measure fugacity equilibrium between a gas and a liquid phase. For one compound (i), the fugacity equilibrium between gas and liquid is usually written in the following form:

\* Corresponding author.

E-mail address: [gilles.hebrard@insa-toulouse.fr](mailto:gilles.hebrard@insa-toulouse.fr) (G. Hébrard).

## Nomenclature

### Latin letters

$H$	Henry's law constant [Pa]
$M$	molar mass [ $\text{g}\cdot\text{mol}^{-1}$ ]
$n_{trans,i}$	amount of molecule transferred during absorption step [mol]
$P$	pressure [Pa]
$R$	perfect gas constant [ $\text{J}\cdot\text{mol}^{-1}\cdot\text{K}^{-1}$ ]
$T$	temperature [K]
$V_L$	liquid Volume [ $\text{m}^3$ ]
$x$	liquid molar rate [ $\text{mol}\cdot\text{mol}^{-1}$ ]
$y$	gaseous molar rate [ $\text{mol}\cdot\text{mol}^{-1}$ ]
$Z$	compressibility factor

### Greek letters

$\phi$	fugacity coefficient
$\gamma$	activity coefficient
$\rho$	density [ $\text{kg}\cdot\text{m}^{-3}$ ]

### Subscripts

$CH_4$	related to $CH_4$
$CO_2$	related to $CO_2$
$G$	related to the gas phase
$i$	related to the component $i$
$j$	related to the state $j$
$L$	related to the solvent phase

$$y_i \cdot P \cdot \phi_i = x_i \cdot H_i \cdot \gamma_i \quad (1)$$

where  $y_i$  and  $x_i$  are the molar fractions in the gas and liquid phases respectively,  $P$  is the pressure under test,  $\phi_i$  the fugacity coefficient,  $\gamma_i$  the activity coefficient and  $H_i$  the Henry's law constant.

A recent study (Mohebbi et al., 2012) focused on the determination of Henry's law constants for light hydrocarbon gases at low temperatures (274 K to 293 K) and under pressures up to 2 bars. An experimental process was built to measure *i*-butane dissolution and data in the literature was reviewed for other gases, such as methane. It is important to mention that the authors concluded that the results depended on temperature for *i*-butane and pressure for methane. As pressure was relatively low in their experiments, stronger variations can be expected with a process reaching higher pressures. The system the authors considered was made of a cooled chamber containing a determined volume of pure hydrocarbon gas (*i*-butane for example). The exact molar number in the gas phase was determined with the SRK equation of state because of its reliability (Soave, 1993). Water was then injected with a high pressure syringe and the mixture was stirred magnetically. The pressure was measured during water injection and stirring, to follow gas dissolution. Pressure was stabilized and a mass balance was obtained using the pressure variation as the gas phase was considered invariant in concentration (it was a pure gas). This protocol was suitable as there was no contact between water and gas during the compression of the gas (water was injected after the compression) so the molar number was perfectly known before absorption. Pressure was considered to be the only variable of the system because gas phase concentration did not evolve and this allowed the authors of the study to perform a molar balance on the gas phase only. However, this system is not suitable to characterize dissolution of a gas mix, because the gas phase concentration at equilibrium must be determined. With this system, such a measurement was impossible as the interface between water and gas was maintained after equilibrium. It led to desorption during the determination of the gas phase residue. On the other hand, balances won't be possible on gas phase only.

Another work (Knapp et al., 1987) reviewed experimental techniques to characterize phase equilibrium for the design of gas washing systems. One of these systems was intended to measure a solvent saturation in presence of a gas mix under high pressures. The set-up was particularly complicated as it achieved saturation of the solvent with a re-circulating gas. Gas samples could be taken during circulation and pressure was controlled. Both phases were analyzed by gas chromatography, resulting in very accurate analysis. Despite this, the authors did not explain how the gas phase was processed during absorption. It seems that the pressure was maintained but no description of how this was done was provided,

given that the experimental cell was closed. They could inject nitrogen during experiments, but they did not show how or why.

Recently, another work (Ouoba et al., 2015) proposed a new experimental method to determine the Henry's law constant of a volatile organic compound adsorbed in soil. They first presented a review of the different experimental methods proposed in the literature to determine the Henry's law constant (Robbins et al., 1993; Ji and Evans, 2007; Ashworth et al., 1988; Shimotori and Arnold, 2003; Chai et al., 2005; Dewulf et al., 1999; Roberts, 2005; Ip et al., 2009; Lee et al., 2013; Kish et al., 2013; Brockbank et al., 2013; McPhedran et al., 2013; Richon, 2011) before to present their new experimental method. Their method is based on a piston pump place above soil sample containing volatile organic compounds; as pressure applied on the sample can be varied by changing chamber volume controlled by a screw system's, different thermal and pressure equilibrium are reached successively, allowing by standard sensors to calculate Henry's law constant. However, this suitable method for soil contamination cannot be relevant for medium pressure condition encounter in biogas plants. From this short review of experimental techniques for Henry's law constant determination, it appears that a simple technique adapted to a very large range of operated conditions is not yet available. The aim of this work is to propose a simple and movable experimental setup, able to quantify gas mix absorption in solvents under pressure, in order to calculate the Henry's law constants.

Considering the former works, we can deduce that it is obviously important to control several factors. Agitation is a key point if the mass transfer is to reach equilibrium. The quantity of water (or solvent) must be known. To avoid non desired pressure modifications, the global volume (gas plus liquid) must not change. Pressure and temperature are the most important parameters. We must be able to measure concentrations in the gas phase and in the liquid phase before and after experiments. When determining the parameters cited, we must ensure that they exactly represent steady-state equilibrium.

Considering all those criteria, an experimental set-up was built. Called the "Saturo-meter", this is a batch contactor for gas and liquid, able to bring the phases into contact and separate them before and after equilibrium in medium pressure conditions (from 1 to 10 bar). Temperature, pressure and phase concentrations of the components are measured continuously or sequentially. This study presents the design background, the procedure, and the method for exploiting the results obtained with such an experimental setup. Some values are presented for the system comprising  $H_2O$ ,  $CO_2$ ,  $CH_4$  and compared with literature data. Three types of water were tested.

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