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A simple and timesaving method for the mass-transfer assessment of solvents used in physical absorption



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HIGHLIGHTS

- Heavy solvents (DEHA and PDMS) and water were assessed for the absorption of four VOC.
- A simple and timesaving experimental procedure was developed.
- Partition and mass-transfer coefficients were deduced from the experiments.
- The two organic solvents tested, especially DEHA, exhibited rather good affinity with all VOC.
- The liquid-film coefficient was 500% higher in PDMS and 30% higher in DEHA than in water.

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ABSTRACT

A simple dynamic absorption procedure to assess the mass-transfer performances of a solvent toward a selected gaseous solute is presented. Absorption was operated semi-continuously at transient state until the equilibrium was reached without solvent recirculation. Four volatile organic compounds (VOC) more or less hydrophobic (toluene, acetone, dichloromethane, isopropanol) were absorbed in water and two heavy organic solvents (Bis(2-ethylhexyl) adipate DEHA and polydimethylsiloxane PDMS). A numerical resolution procedure was developed to simulate the gas-liquid mass-transfer and to deduce the VOC partition coefficients, expressed as the Henry's law constants, as well as the overall liquid-phase mass-transfer coefficients. The overall liquid-phase mass-transfer coefficients were correlated to the diffusion coefficients using the Higbie penetration theory. The results confirm the high selectivity of water whereas the two organic solvents, especially DEHA, exhibit rather good affinity with all VOC even if the Henry's law constants of the most soluble and the less soluble compounds for those solvents differ by 1 or 2 orders of magnitude. The liquid-film mass-transfer coefficients in the two organic solvents, even being more viscous, are larger than in water which confirms their good potential for hydrophobic VOC treatment.

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1. Introduction

Combustion processes, road transport, waste and water treatments and many manufacturing industries generate gaseous emissions of volatile organic compounds (VOC) which are harmful for human health or environment [1]. Consequently, legislations in many developed countries become more and more drastic and impose to implement gas treatment processes to reduce VOC emissions.

Among the different gas treatment technologies, wet scrubbing is particularly attractive due to its simplicity and its high efficiency [2–5]. Gaseous pollutants are transferred in a liquid phase which

can be either aqueous or organic. Aqueous scrubbing liquids are suitable for hydrophilic, acidic and/or basic compounds or when a fast reaction can be implemented such as for the H₂S absorption in a chlorine solution [6–8]. However for hydrophobic compounds, due to a poor solubility in water, absorption in aqueous solution is ineffective and organic solvents should be considered as an alternative. In that case, besides a good affinity with VOC, several constraints must be respected such as a low vapor tension, a low viscosity, a rather low cost, good chemical and thermal stabilities, a good chemical compatibility and a low toxicity [9,10]. Since the last decade, a new generation of organic solvent, such as phthalates, polydimethylsiloxane (PDMS, a silicone oil), Bis(2-ethylhexyl) adipate (DEHA) or ionic liquids drew particular attention [11–16].

Besides the packing characteristics and some classic physico-chemical properties of the solvents such as its density, viscosity

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or surface tension, the mass-transfer rate in physical absorption mainly depends on two parameters related to the VOC/solvents interactions: (i) the gas–liquid partition coefficient (which accounts for the gas–liquid equilibrium) and (ii) the VOC diffusion coefficient (which accounts for the mass-transfer kinetics). The gas–liquid partition coefficients can be easily measured by static or dynamic methods [17–20]. Dynamic methods, whose the partition coefficient is deduced from the breakthrough curve, can be advantageously automated [17,19,20]. On the contrary, the diffusion coefficient determination requires sophisticated techniques and equipments to be measured, such as absorption in a laminar falling film contactor or thermogravimetric or manometric methods [14,21,22]. Moreover, no universal and accurate correlation exists in the literature to quantify the diffusion coefficients in viscous solvents such as DEHA or PDMS [21]. Consequently, up to now, the assessment of organic solvents for VOC absorption is often based only on the VOC/solvent affinity evaluation through the gas–liquid partition coefficient measurement [18]. To screen and assess many solvents in a first stage, the liquid film mass-transfer coefficient of a solute in an investigated solvent determination and its comparison with a well-know solvent, such as water, is a pertinent strategy to confirm that the solute diffusion is not a limiting parameter [23]. This strategy would be even more valuable if the partition coefficient could be deduced simultaneously. Thus, the diffusion coefficient would be determined in a second stage with a more sophisticated technique.

This study presents a simple, reproducible and timesaving dynamic absorption procedure to assess the mass-transfer performances of a solvent toward a selected solute. This absorption was operated semi-continuously at transient state until the equilibrium was reached without solvent recirculation to simplify as much as possible the experimental procedure. Four more or less hydrophobic VOC, which belong to different chemical families currently involved in industrial emissions (toluene, acetone, dichloromethane, isopropanol) were absorbed in water and two heavy organic solvents (DEHA and PDMS). The VOC/solvent partition coefficients and overall liquid-phase mass-transfer coefficients were deduced from the breakthrough curves using a numerical procedure developed in this work. Then, the gas and liquid film mass-transfer coefficients were deduced and correlated to the gas and liquid diffusion coefficients using the Higbie penetration theory. The gas and liquid diffusion coefficients were calculated owing to Fuller et al. correlation and Hayduk–Laudie (water) or Wilke–Chang (DEHA, PDMS) correlations respectively.

2. Material and methods

2.1. Selection of volatile organic compounds and solvents




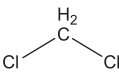
Tables 1 and 2 respectively present the four VOC and the three solvents selected. The diffusion coefficients in the gas phase D_G

(independent of the selected solvent) have been computed according to the Fuller et al. equation [8,24]. The diffusion coefficients in water (D_{water}) have been computed with the Hayduk and Laudi correlation which is recommended for aqueous solutions [24]. The correlations usually used for the calculation of diffusion coefficients in organic solvents (Wilke–Chang, Scheibel, Tyn–Calus, Siddiqui–Lucas, etc.) were not developed for the case of a small solute (such as VOC) diffusing in a solvent composed of large molecules (such as DEHA and PDMS). Therefore, they exhibit higher inaccuracy to predict diffusion coefficients in heavy solvents than in “traditional” organic or aqueous solvents [21,22]. The VOC diffusion coefficients in each solvent calculated with these correlations are summarized in the Appendix 1 and compared to the few experimental data found in the literature (Tables A.1–A.3). Except for the Wilke and Chang correlation, the molar volume of the solvents at their boiling point, which is unknown for DEHA and PDMS, were required. Therefore, for all these correlations, the DEHA and PDMS molar volumes were approximated at the ambient temperature instead of at the boiling temperature. The Tyn–Calus correlation undoubtedly overestimates the diffusion coefficients in DEHA and PDMS whereas the Hayduk–Minhas and Siddiqui–Lucas correlations seems to slightly underestimate them by comparison to the experimental values. The Scheibel and Wilke–Chang correlations appear more accurate providing values close to the experimental one. Nonetheless, the Scheibel correlation provides diffusion coefficients 40–75% higher for PDMS and 18–40% higher for DEHA than the Wilke–Chang correlation. For the case of phthalates, the Wilke and Chang correlation underestimates the experimental diffusion coefficient by 40–50% whereas the Scheibel equation overestimates it by 60–100% [21]. Therefore, the diffusion coefficients used in this paper were calculated with the Wilke–Chang correlation taking into account these observations and the fact that the unknown solvent molar volume at the boiling point is not required for this correlation. Nonetheless, the real diffusion coefficients might be slightly larger according to the Tables A.1 and A.2. Nevertheless, the mass-transfer rate is usually rather poorly sensitive to the diffusion coefficient (which is reflected on the large discrepancy of the experimental data), i.e. a rather large inaccuracy on the diffusion coefficient calculation can be tolerated.

2.2. Experimental set-up and procedure

The Flow and Process Diagram of the experimental set-up is presented in Fig. 1 (A picture of the experimental set-up is presented as Supplementary material). The liquid VOC was vaporized in a compressed air flow to generate a synthetic vicious effluent. The amount of VOC vaporized was carefully controlled using a syringe-driver (KDS100, KdScientific) and a gas-tight syringe (SGE) to target an inlet VOC concentration around 80–85 mg m⁻³ (20–25 ppmv). The VOC/air mixture was homogenized by a static mixer, placed after the syringe needle.

Table 1
Main physico-chemical properties of the selected VOC.

	Toluene	Acetone	Isopropanol	Dichloromethane
Formula				
Supplier	Acros organics	Acros organics	Acros organics	Acros organics
M (g mol ⁻¹)	92.32	58.08	60.095	84.93
Purity (%)	99	99	99	99
ρ at 293 K (kg m ⁻³)	866.9	790	785	1326.6
$10^5 D_G$ (m ² s ⁻¹)	0.74	1.04	1.01	1.02
$10^{10} D_{water}$ (m ² s ⁻¹)	7.96	10.2	9.88	11.3

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