

# Designing the inner surface corrugations of hollow fibers to enhance CO<sub>2</sub> absorption efficiency

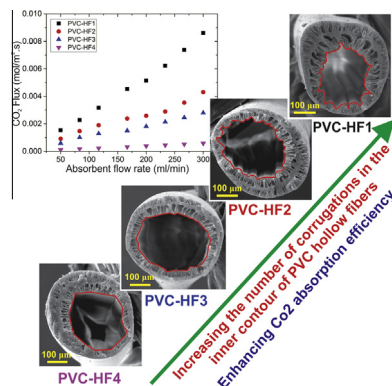


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## GRAPHICAL ABSTRACT



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## ABSTRACT

For the first time, a low cost strategy is introduced to enhance the efficiency of CO<sub>2</sub> absorption using gas-liquid membrane contactors. This is implemented by designing the corrugations in the inner layer of poly (vinyl chloride) hollow fibers (PVC HFs) through changing the bore fluid composition. In fact, the number of corrugations in the HF inner layer is engineered via changing the phase separation time within the inner layer. Such that expedited phase separation leads to highly corrugated inner layer. In contrast, decelerated phase separation is responsible for reduced number of inner layer corrugations. Phase separation causes the initial polymer solution with low viscoelastic moduli to be transferred into polymer-rich domains with high viscoelastic moduli. These domains resist against stretching-induced radial forces toward the center of HF; therefore, the inner layer of HF buckles. Delayed phase separation defers formation of polymer-rich domains and hence, HF with less corrugated inner surface is expected. The phase separation within the HF inner layer is controlled through changing the rate of solvent/nonsolvent exchange. This is conducted by variation the solvent content in the bore fluid; as higher as solvent content, as slower as solvent/nonsolvent exchange.

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## 1. Introduction

Nowadays, global warming caused by greenhouse gases particularly carbon dioxide (CO<sub>2</sub>) is considered as one of the most important predicaments in our world. This dilemma poses a serious

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challenge to natural resources and all animals including melting sea ice and glaciers, shifting precipitation patterns and setting animals on the move [1,2]. In these regards, enormous efforts have been devoted to prevent the sustained increase in emission level of greenhouse gases. For example, the recent united nations climate change conference (30 November to 12 December 2015) was held to raise the scientific understanding and global awareness regarding the whole world's warming up and climate changes. Furthermore, many attempts have been made to develop modern and efficient techniques to capture the greenhouse gases before reaching to the atmosphere. These separation methods include chemical absorption, physical absorption, solid adsorption, column absorption, cryogenic distillation and gas adsorption using molecular sieves [2,3]. Besides, the presence of CO<sub>2</sub> in the natural gas is another controversial issue causing problems such as reduction in the heating value, pipeline corrosion and occupying the volume in the pipeline. Currently, bubble column, packed bed and spray towers are widely used in different industries to deal with this problem [3,4].

Considering the above discussion, it can be concluded that although conventional CO<sub>2</sub> capture processes have attained remarkable successes and benefit from advantages such as maturity and large-scale implementation [5], however, they cannot provide high throughput to meet the requirements of the advanced processes. Therefore, looking for modern CO<sub>2</sub> capture methods with high efficiency and reliability is a hot topic. In this respect, membrane-based technologies have been able to attract great attention as a promising alternative to currently available CO<sub>2</sub> absorption methods [5].

Gas-liquid membrane contactor (GLMC) firstly developed by Qi and Cussler [6], allows absorbent liquid and gas to flow independently in different sides of a membrane inside a module. Actually, in this configuration, the membrane provides an interface through which the gas molecules can be transferred. Among different geometries of membrane, *i.e.* flat sheet and hollow fiber membranes, the latter has gained too much attention to make GLMCs. This package also offers some advantages over the conventional CO<sub>2</sub> capture devices. Specifically, the independent flow of gas and liquid makes it possible to alter the liquid and gas flow rates, independently. Additionally, providing high mass transfer interfacial area per unit volume, being cost effective, operationally flexible and modular, having small size, easy to scale-down & -up, providing high driving force for mass transfer along with very high selectivity, all make GLMCs as promising candidates for CO<sub>2</sub> absorption with high efficiency [3,4,7–9].

The efficiency of GLMCs is predominantly determined by morphology of hollow fibers. Hollow fibers employed in GLMCs have to fulfill some essential conditions including extremely good chemical resistance against different liquid absorbent, high hydrophobicity to provide long service time, low mass transfer resistance and microporosity to afford high permeability. A rich literature exists focusing on controlling the morphology of hollow fibers to achieve GLMCs with the highest level of efficiency for CO<sub>2</sub> absorption and the longest service time. Below are different technical approaches considered to control the hollow fiber morphology to achieve the highly effective CO<sub>2</sub> absorption:

- Using different polymers including poly(tetrafluoro ethylene) (PTFE) [10,11], polypropylene (PP) [11,12], polyetherimide (PEI) [13], polyethersulfone (PES) [14], polysulfone (PSF) [15,16] and poly(vinylidene fluoride) (PVDF) [3,9,17] to manufacture hollow fibers
- Improving surface hydrophobicity of PVDF hollow fibers through solution blending with surface modifying macromolecules (SMM) [3,18] or post-spinning process employed on the activated surface [19]

- Changing the polymer concentration as well as the rheological properties of the spinning solution [17,20]
- Engineering the inner skin morphology of hollow fiber [16,21]
- Superimposing rheological images on ternary phase diagram to design hollow fibers with sub-layer large cavities and macrovoids [9]
- Using different additives in the spinning dopes [3,22–25]

According to the above discussion, it can be said a great deal of research has been conducted to assess how various operational and spinning parameters contribute to morphology evolution of hollow fibers and their capacities to capture CO<sub>2</sub> using GLMCs. However, to our knowledge, no attention has been paid towards the influence of inner contour shape of hollow fibers on CO<sub>2</sub> absorption efficiency. Exploring this effect is the cornerstone of the present research work. This is conducted through producing poly(vinyl chloride) hollow fibers with different bore fluids composed of various compositions of solvent (NMP) and nonsolvent (water). Implications regarding the interplay among liquid-liquid phase separation, viscoelastic properties of polymer-rich phases and buckling instability to develop inner contour of hollow fibers are discussed.

## 2. Experimental

### 2.1. Materials

Poly (vinyl chloride) (PVC), and the solvent, N-methyl pyrrolidone (NMP), manufactured by Sigma Aldrich, Inc. were obtained. Distilled water in addition as non-solvent for PVC/NMP solutions was also used for absorbent liquid for CO<sub>2</sub> absorption, measurement of pure water permeability and dynamic water contact angle of the produced hollow fibers. The materials were utilized as received with no additional purification.

### 2.2. Characterization of water/NMP/PVC phase behavior

Localization of the binodal and spinodal boundaries in ternary phase diagram was employed for investigation of phase behavior of water/NMP/PVC ternary system at 25 °C. This was achieved both experimentally and theoretically. Theoretical calculations were performed using the first and second derivatives of Gibbs free energy of mixing ( $\Delta G_{mix}$ ) with respect to the number of moles of component  $i$  ( $n_i$ ) ( $\Delta\mu_i = [\partial\Delta G_{mix}/\partial n_i]_{T,P,n_{j\neq i}}$ ,  $i = 1$  (nonsolvent), 2 (solvent) and 3 (polymer)) and volume fraction of component  $j$  ( $\varphi_j$ ) ( $G_{22}G_{33} = (G_{23})^2$ ,  $G_{ij} = v_1(\partial^2(\Delta G_M)/\partial\varphi_i\partial\varphi_j)_{T,P,n_k \neq ij}$ ), respectively. The former was considered to calculate binodal curve through equalizing the chemical potentials ( $\Delta\mu_i$ ) of three components involved in the membrane formation in two corresponding coexisting phases connected by tie-lines. The latter was evaluated to obtain spinodal curve. Conventional Flory-Huggins model expanded for ternary systems, was considered to calculate  $\Delta G_{mix}$  [26]. More details about the theoretical and numerical procedures can be found in our previously published works [9,27,28]. The binary interaction parameters required for the calculations have been listed in Table 1. Water/NMP interaction parameter ( $g_{12}(u_2)$ ,  $u_2 = \varphi_2/(\varphi_1 + \varphi_2)$ ) was obtained from Ref. [29]. NMP/PVC interac-

**Table 1**  
Binary interaction parameters (BIP) used to construct ternary phase diagram of water/NMP/PVC system at 25 °C.

BIP	$g_{12}(u_2)$	$\chi_{23}$	$\chi_{13}$
Value	$\alpha(= 0.316) + \frac{\beta(=0.468)}{1-\gamma(=0.499)u_2}$	0.47	2.1

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