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Optimal design of dual-reflux pressure swing adsorption units via equilibrium theory

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HIGHLIGHTS

- A design procedure for DR-PH-A units, a configuration of DR-PSA, is proposed.
- Three parameters are identified as key to the effective design of the separation.
- A region of complete separation with triangular shape is found in a suitable space.
- The effect of various process parameters on the design parameters is discussed.

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ABSTRACT

An optimal design strategy for dual-reflux pressure swing adsorption units targeted at achieving the separation of binary gas mixtures into two pure components (complete separation) is presented. Based on the equilibrium theory, detailed study of a specific configuration (feed to high pressure bed and pressure swing using heavy gas) reveals that feed location along the adsorption column, capacity ratio of the *purge* step (ratio between amount of feed to be processed and amount of adsorbent) and light recycle ratio (ratio of pure light reflux to feed rate) are the three most relevant design parameters. With respect to previous literature, two major achievements can be mentioned: (i) the development of an expression to evaluate optimum feed location and (ii) the identification of an operating region inside which complete separation at cyclic steady-state conditions can be established. In the plane defined by the two quantities feed position-capacity ratio of the *purge* step, such region has triangular shape and optimal conditions correspond to a vertex.

1. Introduction

Pressure swing adsorption (PSA) is a well-established technology for the separation and/or purification of gaseous mixtures. It is employed in a wide array of industrial applications, such as hydrocarbon separation, hydrogen purification, air drying and air separation among the most popular. A vast majority of PSA systems are either stripping type, based on Skarstrom (1959) cycle, or rectifying type, developed by Diagne et al. (1994) and Ebner and Ritter (2002). Stripping PSA systems are capable of producing only the light product (weakly adsorbed; referred to as species B) at high purity from a binary feed gas mixture, since the purity of the heavy product (strongly adsorbed; referred to as species A) is confined by thermodynamic constraints (Subramanian and Ritter, 1997). On the other hand, also rectifying PSA systems, known as enriching reflux PSA (Yoshida et al., 2003), have thermodynamic constraints on the purity of the light product, thus resulting in the capability of producing only the heavy product at high purities.

To counteract such thermodynamic limits, *duplex* PSA (Leavitt, 1992) also known as dual-reflux pressure swing adsorption (DR-PSA) processes were proposed, which disclosed the possibility of producing both pure light and heavy products from a binary feed gas mixture. Typically, DR-PSA systems employ feed injection along the adsorbent bed axis in a combined two-bed system. The feed injection position, Z_F, divides each bed into *stripping* and *rectifying* sections with two reflux streams from each of these sections. Depending on the column operating pressure (high pressure, P_H, or low pressure, P_L) to which the binary feed gas mixture is supplied and the type of gas (A or B) with which the pressure swing is carried out, four different cycle configurations can be identified (Kearns and Webley, 2006a). Coming to the actual experimental implementation of this separation process, Diagne et al. (1994, 1995a, 1995b) explored the application of DR-PSA for CO_2 removal from air through experimental studies and

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later performed a numerical analysis for the same (Diagne et al., 1996). McIntyre et al. (2002, 2010) reported the results of their experimental studies conducted using DR-PSA for the recovery and enrichment of dilute ethane from nitrogen. These experimental studies proved that DR-PSA is certainly capable of producing both light and heavy product streams at high purities, thus overcoming the PSA thermodynamic limitations.

Different types of models have been applied to design and optimize PSA units, ranging from very simple to complex (cf. Ruthven et al., 1994; Spoorthi et al., 2011; Thakur et al., 2011; Sivakumar and Rao, 2011a, 2011b, 2012; Turncok and Kadlec, 1971) were among the earliest to put forth a detailed PSA model considering nonlinear adsorption equilibrium along with pressure drop and temperature effects, and this approach has been recently used also for DR-PSA (e.g., Thakur et al., 2011). At the opposite extreme, a much simpler modeling approach was presented by Shendalman and Mitchell (1972), which assumed instantaneous linear equilibrium throughout the columns, ignored all transport phenomena and exclusively considered mass conservation. Such oversimplified approach is usually named Equilibrium Model (or equilibrium theory) and it is often analytically solvable by the method of characteristics (Rhee et al., 1986). Even though approximate, this type of solution offers a basic insight into the system behavior and frequently represents a very effective design tool to elucidate process behavior (Ruthven et al., 1994). The equilibrium theory solution for conventional PSA systems and linear isotherms was fully explained by Knaebel and Hill (1985) more than two decades ago.

This same modeling approach was applied by Ebner and Ritter (2004) to DR-PSA systems. Focusing on a single configuration (feed to P_L and pressure swing using A), the authors analyzed the effect of changing the feed injection position (Z_F) along the axis of the adsorption column between a minimum ($Z_{F,\,Min}$) and a maximum value ($Z_{F,\,Max}$). Kearns and Webley (2006a) found that this design approach did not ensure the complete utilization of the adsorbent. Although the model assumed the production of two pure product streams, it did not account for the composition profiles inside the adsorption columns which converge during pressurization and purge steps resulting in shock waves. Hence, the actual achievement of cyclic steady-state conditions (*CSS*) was verified using external flows only, without any detailed reconstruction of the concentration profiles inside the unit. Nonetheless, this equilibrium model was the first one clearly proving that the complete separation of binary feed gas mixture is in fact possible using DR-PSA. Hence, it paved the way for better understanding of the process and opened a new realm of possibilities for future process development.

Later, Kearns and Webley (2006a) applied the equilibrium theory model proposed by Ebner and Ritter (2004) to four DR-PSA cycle configurations. A single, specific feed position was considered; given this feed position, a sole reflux rate was required to achieve *CSS* in each of the four configurations. This methodology ensured the most efficient use of the adsorbent, i.e. its minimum quantity for a specific separation. However, the relationship between this specific feed position and the range proposed by Ebner and Ritter (2004) for the same variable was not clearly established.

In this work, our aim is to dwell further on the equilibrium theory to provide an optimal design procedure for DR-PSA systems. It can be regarded as an attempt to enhance understanding of the separation behavior by careful tracking of the trajectories of the characteristic curves, as well as of the corresponding shock transitions, during constant and non-constant pressure steps of the cyclic process. Based on the productivity and energy consumption criterion discussed by Kearns and Webley (2006b) for DR-PSA when separating dilute binary feed gas mixtures (i.e. when mole fraction of A is smaller than that of B), in this work we focus exclusively on the best configuration (DR-PH-A: feed to P_H and pressure swing using A). An expression for the optimal feed injection position $Z_{F,\,opt}$ (which can ensure maximum utilization of the adsorbent) has been developed. Moreover, a region of complete separation at *CSS* conditions is identified in terms of feed position and a parameter called capacity ratio of the *purge* step ($\mathbb{C}$, ratio between amount of feed to be processed and amount of adsorbent): any operating point inside this region ensures complete separation, it is operated at constant value of the light recycle ratio ($\mathbb{G}$, stated as the ratio of pure light reflux to feed rate) and exhibits different robustness with respect to changes of the process parameters.

2. DR-PH-A cycle description

The schematic diagram of a typical twin-bed DR-PH-A system (Kearns and Webley, 2006a) under consideration in this work is depicted in Fig. 1. In this particular configuration, each of the two identical adsorption beds (*Bed − I* and *Bed − II*) undergo a four step cyclic process: two steps are simultaneously executed at constant pressure and remaining two at non-constant pressure in both the beds. Note that in Fig. 1, only half-cycle is depicted since the same steps occur with the column numbers transposed. The feed injection position along the bed, defined in terms of normalized axial coordinate (Z_F), divides each column into *stripping* and *rectifying* sections; it is a dimensionless value, Z being the axial coordinate normalized with respect to the column length, $Z = z/L_{bed}$. The section to the left of the feed injection position ($Z < Z_F$) is termed as '*Stripping Section*' or *SS* (light material is either injected-in or pushed-out of the end of this section, $Z=0$) and the section to the right of the feed injection position ($Z > Z_F$) is termed as '*Rectifying Section*' or *RS* (heavy material is either injected-in or pushed-out of the end of this section, $Z=1$), as depicted in Fig. 1.

Binary feed gas mixture with flowrate $\dot{N}_F$ and composition y_F (mole fraction of A in feed gas mixture) is supplied to *Bed − I* which is maintained at constant P_H during the feed step (*FE*);

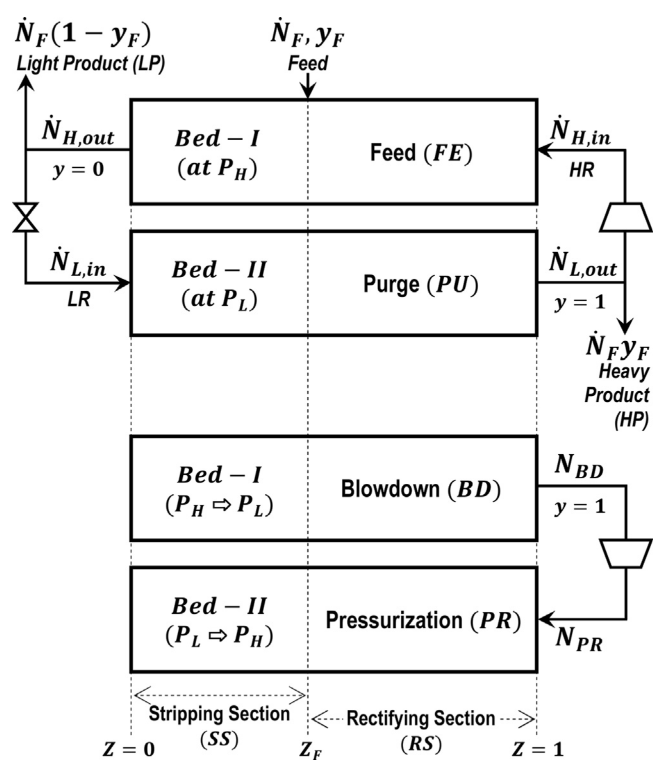

Fig. 1. DR-PH-A cycle steps and flows.