

515f Gas Separation by a Novel Hybrid Membrane/Pressure Swing Adsorption Concept: Case Studies

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Abstract. A truly synergistically hybrid concept was recently developed for gas separation processes combining membrane permeation and pressure swing adsorption (PSA).

The possibility of integrating membrane operations in the gas separation industrial cycle is becoming a particular area of interest, because of several overall benefits. Although several published literature data concerning these hybrid systems are available^{4,5}, a truly synergistically concept was only found in X. Feng and Ghosh (1998) work. Either because the operational complexity of these type of integrations, either due to mathematical framework involved in these design procedures at cyclic steady state conditions, it seems that its still necessary a considerable amount of research concerning the study of feasible hybrid systems. Hence, the coupling of these two units into an integrated process has been studied for several binary gas mixtures (CO₂/N₂, CH₄/H₂, CH₄/CO₂) at different compositions and operating conditions. For the case studies formulated, the process characteristics were successfully predicted.

The configuration schemes developed use the PSA operative pressure range as driving force for permeation in the membrane module. The membrane produces a residue and a permeate streams, each one enriched in the component with more or less adsorption capacity towards the adsorbent bed. The obtained membrane streams are fed to the PSA at different steps of the cycle, in order to enhance the products purity and recovery in comparison with the performance of the conventional stand-alone units. Explicitly, the membrane assists the pressurization (PR) and high-pressure adsorption (HPA) steps of the PSA process. The integrated system occurs in a periodic state, where the permeation acquires a cyclic behavior when coupled with the conventional cycle step sequence of a PSA unit.

In order to explore the synergy of both units, the hybrid concept was developed to be capable of simulating under a wide range of operating and configurational conditions. Two main schemes were developed:

- MEMPSA – CASE A process, where the more permeable component is the least adsorbed one in the PSA bed,
- MEMPSA – CASE B process, where the more permeable component is the more adsorbed one in the PSA bed.

The feasibility of the integrated process was assessed through the effect of the various operating parameters on separation performance. Instead of the regular binary feed mixture, the PSA is fed with a mixture which is progressively enriched in the more adsorbed component along the PR and high HPA steps of a PSA cycle. Dynamic simulations were performed for the binary separations of CO₂/N₂, CH₄/H₂ and CH₄/CO₂ at different feed conditions. The dynamic computational results were obtained for both cases and compared with the modeled conventional units involved. We conclude that, the inclusion of a membrane module into a periodic PSA process, by a truly synergistically scheme, is indeed an innovative method to improve separation performance of several gas systems when compared with the more common isolated units. It is expected that this gain will make the integration of the two processes economically feasible.

Experimentally, adsorption equilibria measurements were performed using a gravimetric method with real-time acquisition of temperature, pressure and weight of the adsorbent sample. Experimental

permeation runs were also performed for CO₂, CH₄ and N₂ in a composite PES/PI (polyethersulfone/polyimide) membrane. Component permeances and ideal selectivities were determined in order to confirm those parameters values used in the modeling work. A laboratorial PSA unit was built and experiments were reported in order to validate the modeling work done. The real-time acquisition and control of the unit allowed the execution of breakthrough and dynamic pressure swing cyclic experiments. An experimental PSA cycle was developed for separation of CO₂/N₂ mixtures. The periodic cycle involves five steps: Pressurization (PR) with feed, High Pressure Adsorption (HPA), Co-current Blowdown (HBD), Countercurrent Blowdown (LBD) and Purge (LPG). The performance and dynamic behavior of the PSA process were successfully predicted, validating the modeling work. The temperature, component gas- and adsorbed-phases concentrations, and velocity profiles along the adsorbent bed were studied, as well as the histories of those variables during the PSA cycle and until the periodic steady-state was attained. In order to confirm the enhancements of the hybrid process predicted theoretically, a CO₂/N₂ separation under this concept was modeled exactly at the same conditions of the experimental runs and its simulations executed for the stand-alone PSA. The results obtained confirm the integrity of the hybrid concept and the improvement of the separation performance when compared with the conventional stand-alone units.

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