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Strategies for the simulation of multi-component hollow fibre multi-stage membrane gas separation systems

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ABSTRACT

Gas separation membranes allow the preferential removal of certain gases from a mixture of gases. If the separation objective is to obtain high product purity and either high removal efficiency or high product recovery it is often necessary to implement a multi-stage network of membranes. However, in the literature most modelling approaches consider the simulation of single-stage membranes. Hence, the aim of this study is to identify stable and computationally efficient strategies for simulating complex multi-stage membrane systems. For this purpose a multi-stage membrane modelling framework is developed and six different stable solution strategies are evaluated and compared in terms of the computational effort required to solve the resulting sets of equations. These solution strategies vary according to the sequence in which the individual membrane models are solved (sequential and simultaneous approaches) and the manner in which those membrane models are initialised. In all these strategies a Newton–Raphson method is employed to solve the mass balance equations in both single-stage and multi-stage membrane systems. Comparisons are made using example simulations of 10 different configurations of membranes containing 1–4 membranes with different numbers of connections and recycle streams present.

1. Introduction

A single-stage membrane used for gas separation will typically have a single stream of feed gas and two exit streams called the permeate and retentate as shown in Fig. 1. The permeate stream contains gases which have passed through the membrane and the retentate contains any remaining gases from the feed.

For hollow fibre membranes there is a choice to be made as to whether to insert the feed into the bore side (inside the hollow fibre) or the shell side (outside the hollow fibre). In either case the hollow fibres will generally be encased in a module containing multiple fibres so that the pressures of both sides of the membrane are contained and pressurised appropriately. Additionally there are three basic configurations including co-current, counter-current and cross-flow which vary depending on the direction of flow, location of the exit streams and the influence of permeate-side mixing.

There have been a number of articles which describe methodology for the simulation of single-stage membranes. The equations which are used to describe these systems normally form a boundary value problem, i.e. sets of differential equations with boundary conditions at the inlet and outlets. One of the most commonly used and cited method for the simulation of multi-component membrane separations is the work of Pan [1] and they implement an iterative method (a shooting-type method [2]) to solve this boundary value problem where they repeatedly integrate along the membrane until reaching convergence. However, a number of authors have made simplifying assumptions and modifications which allow the conversion of this problem into an initial value problem [3,4] which is simpler to solve.

Alternatively, authors such as Coker et al. [5] and Katoh et al. [6] have tackled the boundary value problem directly by dividing the membrane into a number of sections or tanks which is equivalent to a finite-difference approach to solve the problem.

The methods for obtaining numerical solutions describing these membrane separations can then be divided into those which require initial values for flow rates, compositions and pressures inside the membrane such as those of Pan [1] and Coker et al. [5] and other methods which state that no initial values are required [4,7] (methods which require no initial values should converge starting from a poor/crude initial guess rather than requiring a good initial values). The method of Kaldis et al. [7] uses collocation

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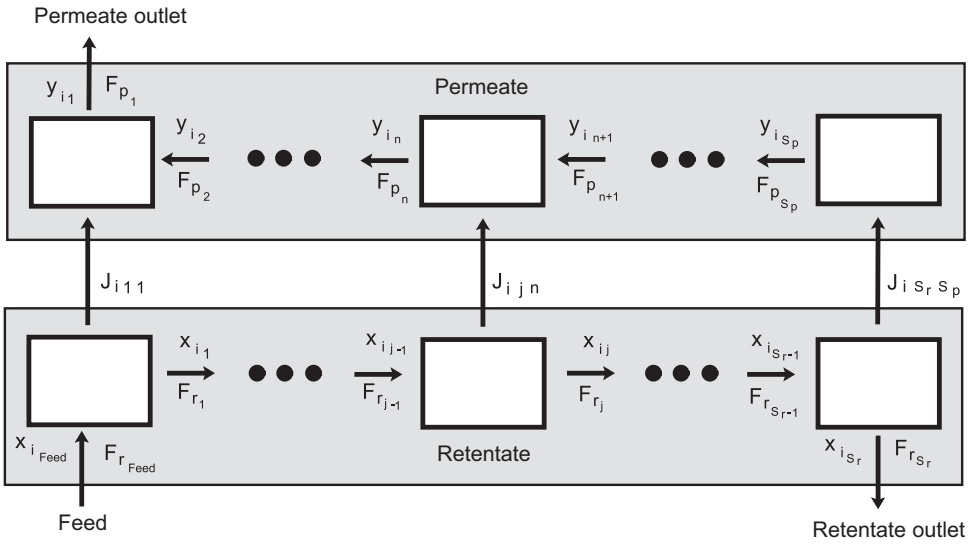

Fig. 1. Tanks-in-series model representation of a hollow-fibre membrane operated in the counter-current configuration.

together with the Brown method to solve the equations for a single-stage membrane while the method of Kundu et al. [4] converts the problem in to an initial value problem which is solved through dynamic integration using Gear's method incorporating a variant of Newton's method. In both cases the authors state that their methods are both stable and computationally efficient.

However, the accuracy and computational effort required by these approaches and by the finite difference methods depend on the size of the time steps for integration methods, the number of collocation points for collocation methods and the number of sections/tanks for finite-difference methods. These numbers are typically chosen such that the models are sufficiently accurate to reproduce experimental results. For example Kaldis et al. [7] state that only 6 collocation points are required to reproduce the experimental results of Pan [1] within an accuracy of 5% while Coker et al. [5] suggest that 100 finite difference tanks are sufficient for most modelling purposes.

Additionally, a major limitation of most membrane simulation methodologies is that they consider only the solution of single-stage membranes. This is a significant point because it is known that multi-stage membrane systems are normally required to obtain high purity and high removal efficiency. For example Baker [8] suggests that due to pressure ratio and selectivity limits in commercial membranes a single-stage system may be unable to provide the required separation. Additionally Low et al. [9] show that for CO_2 capture a single-stage membrane is unable to simultaneously give high CO_2 purity and recovery.

Hence, there are many articles in the literature such as that of Ahmad et al. [10] which consider the simulation and optimisation of multi-stage membrane systems. Although it is possible to use single-stage methods to model multi-stage systems through the sequential simulation of the different connected membranes this can be a computationally inefficient approach. In particular for cases where one or more of the outlet streams are recycled (e.g. to enhance product recovery) this sequential solution strategy may require a large number of iterations before it converges to a steady-state solution for the whole system. This is shown by Makaruk and Harasek [11] who implement a methodology which sequentially and repeatedly simulates single-stage membranes in order to obtain solutions for multi-stage membrane systems. In their results they show that for cases involving 2 membrane stages with a recycle thousands of iterations are required to obtain a solution. Hence, single-stage methods can be used to model multi-stage systems but depending on the membrane configuration and the algorithm used this can be a computationally inefficient approach.

Ahmad et al. [10] have developed methodology for the simulation of single-stage membranes which they use repeatedly within a process simulator environment to simulate multi-stage membrane systems. However, other authors including Khalipour et al. [12] and Kundu et al. [4] have presented methodology for simulation of single-stage membranes and state that their methodologies can be applied to multi-stage membrane systems but they do not specify how their methodology should be extended to these multi-stage membrane systems. Hence, it is presumed that they use a similar approach as Ahmad et al. [10] repeatedly using their single-stage membrane methodology.

This sequential approach has also been used inside optimisation with the Levenberg–Marquardt method for the upgrading of both biogas from natural gas [13] and for the extraction of hydrogen from biomass gasification [14]. However, in both cases it is assumed that a large computational effort was required due to the large numbers of iterations required for each simulation.

Additionally a number of authors have suggested methods for the simultaneous solution of multi-stage membrane systems. For example authors such as Qi and Henson [15] and Scholz et al. [16] have included the equations for multi-stage membrane systems as non-linear constraints inside their MINLP optimisation methods. As these authors do not mention any convergence issues (e.g. due to numerical stiffness) which might occur at high or low stage cut it is assumed that implicit methods together with damping are used which should be able to handle most cases without difficulty. Also, in both cases these authors have used either simplified or shortcut models and they do not state the computational requirements of their multi-stage simulations or of the optimisations which are carried out with these models. Katoh et al. [6] have also considered the simultaneous solution of multi-stage membrane systems, in their case using a dynamic relaxation method which should be a stable method for solving such systems but may require large numbers of small time steps to reach a steady-state solution. Although the number of steps can often be greatly reduced using variable step-length methods the number of steps required can still be very large in some cases (depending on the stiffness of the problem, the algorithm used and tolerances specified). Hence, dynamic methods may be computationally slow (generally much slower than steady-state methods), but they are essential if the dynamic behaviour of the system is being studied.

So in summary for the solution of multi-stage membrane