Gas-Separation Membrane Cascades Utilizing Limited Numbers of Compressors

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A systematic procedure is presented to drawn n-compressor membrane cascades for gas-separation applications with $n \geq 1$. In this method, a parent cascade structure is created that contains the n-compressor cascades as substructures. These substructures are then revealed by systematically eliminating unwanted membrane stages and recycle compressors from the parent cascade. The procedure is illustrated by generating arrays of one- and two-compressor cascade schemes. It is found that these arrays contain the cascade schemes known from the literature and also lead to some new schemes. This procedure increases the likelihood of finding an economically viable membrane cascade for a given separation application.

Introduction

Membrane-based gas separation processes have been the subject of intense study and development during the past two decades and several processes have been fully commercialized (Ho and Sirkar, 1992; Koros and Fleming, 1993). A significant number of these commercial applications use membrane cascades involving recycle streams. Cascades are required when the available membrane selectivities are limited and the desired separation cannot be economically achieved from a simple one-stage membrane. Available membrane selectivities are limited for many gas-separation applications (Robeson, 1991). This has led to the development of several membrane cascade schemes for gas separation.

A classic application of a membrane cascade was for the recovery of ^{235}U isotope from natural uranium by gaseous diffusion of UF_6 through a microporous barrier. Because of the very low separation factor for the separation of uranium isotope ^{235}U from ^{238}U, thousands of gaseous-diffusion stages were required to produce 90% ^{235}U from a feed containing 0.7% ^{235}U (King, 1980). Unlike the uranium isotope enrichment case, the use of membranes in most other gas-separation applications has to compete with alternative, non-membrane-based separation processes. This often requires that the capital portion of the processing cost of a membrane process be decreased. Cascades with only a few membrane stages are used in order to decrease the number and cost of the compressors. Decreasing the number of recycle streams used in a cascade process generally increases energy consumption, but the capital-energy trade-off generally favors cascades with only a few compressors. Most commercial gas-separation cascade processes use only one or two compressors (Spillman, 1989; Prasad and Thompson, 1992).

In the last two decades, a number of papers (Ohno et al., 1978a,b; Laguntsov et al., 1992; Xu and Agrawal, 1996b; Agrawal and Xu, 1996) and patents (Iwata and Tamura, 1987; Prasad, 1992; Prasad, 1993, Gottzmann et al., 1994; Prasad, 1994; Thompson, 1994; Xu, 1994a,b) have appeared in the literature suggesting different gas-separation membrane cascades using one or two recycle compressors. While a number of studies have analyzed the usefulness of these cascades (Matson et al., 1983; McCandless, 1985; Matson et al., 1986; Bhide and Stern, 1991; Pettersen and Lien, 1994; Bozhenko and Bozhenko, 1995), a systematic procedure to draw these cascades has been lacking. To date, each of these cascade schemes seems to have been drawn as a result of the inventive activity of a researcher trying to find an economic solution for a particular application. This raises the question as to how one ensures that for a given application the choice of a membrane cascade with a given number of recycle compressors is the optimum one; what other cascade schemes with the same number of recycle compressors could be used for this given application? Answers to these questions require *a priori* identification of all possible membrane cascade schemes for the application.

This article develops a stepwise systematic procedure for generating gas-separation membrane cascades using limited numbers of recycle compressors. For a given number of com-

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pressors, this leads to all the known cascade schemes and also provides an array of new cascade schemes hitherto unknown in the literature; therefore, a nearly exhaustive list of cascade schemes is generated. For a given application, the problem is thus reduced to that of finding the most optimum cascade from an array of available cascade schemes.

Hwang and Kammermeyer (1965) introduced the concept of an ideal membrane cascade for a binary separation as one where the compositions of two streams, which are combined before feeding to a membrane stage, are exactly the same. This avoids exergy losses due to mixing, and in principle should lead to the most efficient system. Although cascade schemes with inherent mixing losses have been suggested and studied in the literature (Ohno et al., 1978a,b; Bhide and Stern, 1991; Petterson and Lien, 1994), we believe that such nonideal cascade schemes are of limited utility (Bhide and Stern, 1991; Xu and Agrawal, 1996b). Therefore, no attempt has been made to generalize the proposed method to generate nonideal cascade schemes.

It is not the objective of this article to discuss the situations under which the proposed cascade schemes would be economically viable. Some discussion on the applicability and usefulness of one and two compressor membrane cascades can be found elsewhere (Xu and Agrawal, 1996b; and Agrawal and Xu, 1996).

Procedure to Draw Cascades with Limited Numbers of Compressors

The theory of membrane cascades using reasonably large numbers of recycle compressors and membrane stages is very well developed (Pratt, 1967; Brigoli, 1979; Benedict et al., 1981). In its simplest form, a cascade arrangement involves multiple membrane stages where the permeate from a stage after compression is sent as feed to the succeeding stage, while the nonpermeate stream from each stage is sent to the previous stage (Figure 1a). A cascade is generally characterized in terms of parameters p and q, where p is the number of succeeding membrane stages that the compressed permeate stream is fed to and q is the number of previous membrane stages that the nonpermeate stream is sent to. When the values of cascade parameters p and q are unequal, the cascade is referred to as unsymmetrical cascade (Nikolaev et al., 1980). In Figure 1a, the value of both the parameters p and q is one and it is a symmetrical cascade. Figure 1b and 1c show examples of unsymmetrical cascades. In Figure 1b, the permeate stream from a membrane stage, after compression is not sent to the next membrane stage but to the one after it. Therefore, the value of parameter p is two. In Figure 1c, while permeate stream is sent to the next membrane stage, the nonpermeate stream skips the immediate previous stage and is fed to a membrane stage before it. Therefore, while the value of p is one, the value of q is two. It is worth noting that cascades for any suitable values of p and q can be drawn; however, when the numerator and denominator of the fraction $q/(p+q)$ have a common factor other than one, the cascade reduces to independent parallel cascades with lower values of p and q (Pratt, 1967). For example, a cascade with $p=2$, $q=2$ consists of two independent parallel cascades with $p=1$ and $q=1$.

The procedure to draw cascades with limited numbers of compressors starts with the modification of known cascade schemes such as the ones shown in Figure 1. (By limited numbers of compressors it is implied that only a few recycle compressors are used in a cascade; it means either one or more than one but not a large number of recycle compressors are associated with the cascade.) The modification to the initial starting cascade is slightly dependent on whether the feed is at a high pressure or a low pressure. A feed is said to be at a high pressure if it can be directly fed to the high-pressure

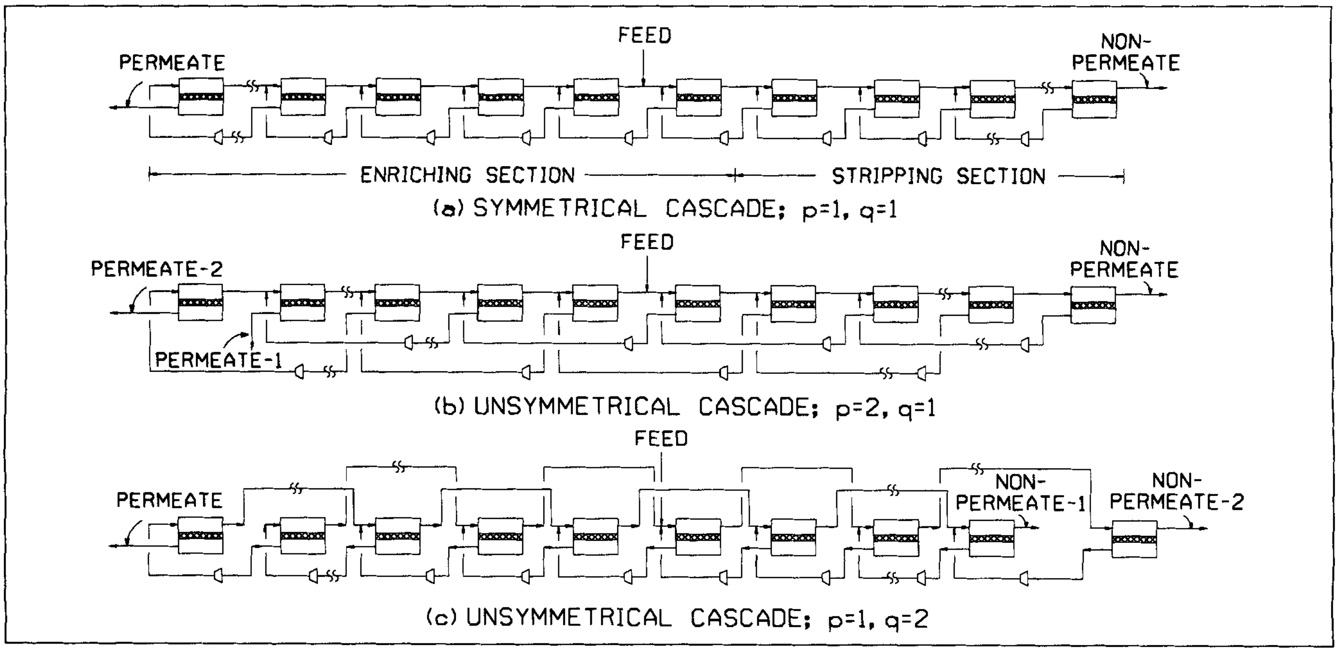

Figure 1. Some known membrane cascade schemes using large numbers of recycle compressors.
(a) Symmetrical cascade; $p=1$, $q=1$. (b) Unsymmetrical cascade; $p=2$, $q=1$. (c) Unsymmetrical cascade; $p=1$, $q=2$.

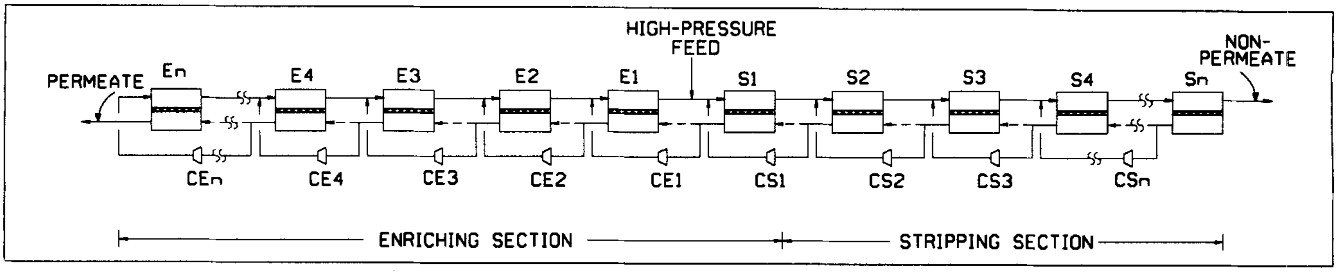

Figure 2. A modified symmetrical cascade with high-pressure feed to yield parent cascade; $p=1$, $q=1$.

side of a membrane unit without further compression. Similarly, a low-pressure feed would need some pressure boost before it could be fed to the high-pressure side of a membrane unit. A separate description of the procedure for each high-pressure feed and low-pressure feed case is now presented.

High-pressure feed case

In this section, the procedure will be illustrated by drawing cascades with one and two recycle compressors when the feed can be directly fed to the high-pressure side of a membrane unit.

The first step is to choose a scheme from the known cascade schemes, such as the ones shown in Figure 1, with certain values of cascade parameters p and q. Let us start with the simplest of the cascades shown in Figure 1a. In the next step this cascade is modified as shown in Figure 2 to yield a parent cascade scheme; the dotted permeate streams show that the permeate from a membrane stage can either be compressed and fed to the high-pressure side of the next membrane stage or directly fed as sweep stream to the low-pressure side of the next membrane stage. Now n-compressor cascades, which are substructures of this parent cascade, can easily be derived. These feasible substructures are derived by systematically eliminating the unwanted membrane stages and recycle compressors. Before starting the process of eliminating unwanted membrane stages and recycle compressors, it is important to note that the membrane stage S1, to which feed is directly fed, cannot be eliminated. Even when all other membrane stages and recycle compressors are eliminated, stage S1 is needed to yield a simple one-stage membrane process. In order to generate distinct cascade schemes, if a membrane stage receives the permeate from the previous membrane stage as sweep, then its permeate discharge stream should not be sent as sweep to the low-pressure side of the next membrane stage. If this step is not followed, then membrane cascades will be created where a membrane stage would be shown as simply two or more membrane units connected in series. Generally the membrane area available in a membrane unit is limited and in a given application a membrane stage within a cascade can consist of two or more such units connected either in series or in parallel. Therefore, a cascade with a membrane stage consisting of multiple membrane units connected in series is not considered a distinct cascade in this article.

Consider the generation of *one-compressor cascades*, which are substructures of the parent cascade scheme of Figure 2. In order to derive the first one-compressor cascade, let us start with a recycle compressor in the stripping section (the right of the feed location in Figure 3). Let the first choice be recycle compressor CS2. Since recycle compressor CS1 is eliminated, the permeate from membrane stage S2 should be directly sent as sweep to the low-pressure side of membrane stage S1. Similarly, any low-pressure feed to stage S2 from the permeate stream of stage S3 is eliminated due to use of

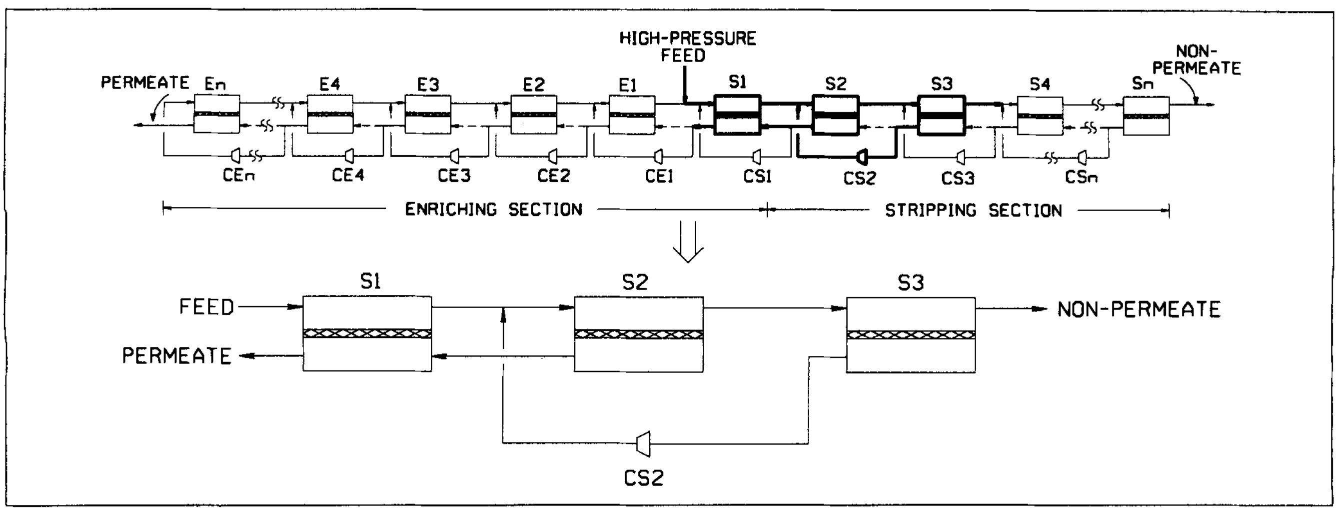

Figure 3. Derivation of cascade scheme HP1-11-3a from the parent symmetrical cascade with $p=1$ and $q=1$.

recycle compressor CS2. All other membrane stages and recycle compressors are eliminated to yield cascade HP1-11-3a shown in Figure 3. The low-pressure permeate discharge stream from stage S1 cannot be sent to any membrane stage and is the permeate product stream. It is worth noting that if instead of recycle compressor CS2, the choice was made to retain recycle compressor CS3, the cascade scheme obtained would still be HP1-11-3a. This is because in this case the permeate from stage S3 will fed to the low-pressure side of stage S2 and stages S2 and S1 would be effectively one membrane stage.

Before proceeding further with the generation of other cascade schemes, we are going to describe an alphanumeric system adopted by us to identify various cascade schemes with limited numbers of compressors. A derived cascade is designated either HPα-pq-$\beta\gamma$ or LPα-pq-$\beta\gamma$. The first two letters indicate whether the feed is at a high-pressure (HP) or a low-pressure (LP), the next number, α, indicates the total number of compressors and vacuum pumps used, the following two numbers, p and q, are the values of the cascade parameters characterizing the parent cascade from which the cascade under consideration is derived, the value of β stands for the number of membrane stages in the cascade, and the last letter, γ, is the assigned alphabetic serial of the cascade scheme. Thus HP1-11-3a for the cascade in Figure 3 indicates that the given feed is at a high pressure (needs no further compression). This cascade requires one compressor, it is derived from a symmetrical cascade with parent cascade parameters $p = 1$ and $q = 1$, the cascade uses three membrane stages, and its assigned serial is a.

Returning to the derivation of one-compressor cascades from the parent cascade of Figure 2, it is worth noting that for one recycle compressor in the stripping section, cascade HP1-11-3a provides a cascade scheme with the maximum number of membrane stages. A cascade with fewer stages would be derived by retaining recycle compressor CS1 and the associated membrane stage S2 from the parent cascade of Figure 2. The resulting two-membrane stage cascade scheme HP1-11-2a is shown in Figure 4a. This cascade was originally introduced as "series type two unit membrane cell" (Ohno, 1978a).

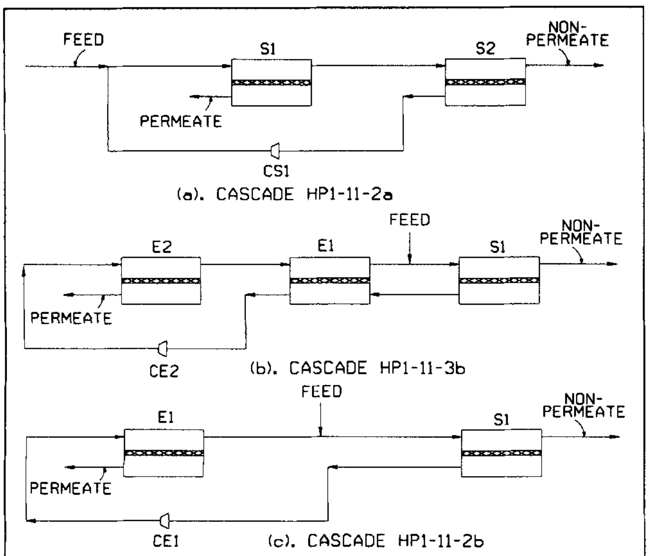

Figure 4. One-compressor high-pressure feed cascades derived from the parent cascade with $p = q = 1$.

(a) Cascade HP1-11-2a. (b) Cascade HP1-11-3b. (c) Cascade HP1-11-2b.

Once the possibilities with a recycle compressor in the stripping section have been exhausted, then cascades with a recycle compressor in the enriching section may be derived. As stated earlier, during the process of eliminating membrane stages, it is necessary to retain membrane stage S1. A cascade with the maximum number of membrane stages is derived by retaining from the set of usable recycle-feed compressors the one that is farthest away from the feed location. Thus, from the parent cascade scheme of Figure 2, recycle compressor CE2 and its associated membrane stages E1 and E2 are retained while other membrane stages (besides S1) and recycle compressors are eliminated. Once again, the elimination of recycle compressor CE1 implies that the low-pressure permeate stream from stage S1 be sent as sweep stream to the low-pressure side of stage E1. The resulting HP1-11-3b cascade scheme is shown in Figure 4b (Laguntsov et al., 1992). The next possibility is to retain recycle compressor CE1 and its associated membrane stage E1 and eliminate all other unwanted membrane stages and recycle compressors from the parent cascade; the resulting two-membrane stage cascade scheme HP1-11-2b is shown in Figure 4c (Gottschlich et al., 1989; Spillman, 1989). This completes the possibilities of one-compressor high-pressure feed cascades that can be derived from the parent cascade with $p = q = 1$.

We can now follow the same procedure to construct *two-compressor cascades* from the parent cascade of Figure 2. Let us start again by using both the recycle compressors in the stripping section of the parent cascade (Figure 5). The maximum number of membrane stages results when recycle compressor CS4, which is farthest away from the feed, is utilized, and recycle compressor CS2 is retained. The rest of the recycle compressors and unwanted membrane stages are eliminated. The resulting scheme HP2-11-5a is shown in Figure 5. Other possible distinct cascades derived with both recycle compressors in the stripping section are shown in Figure 6a–6c; recycle compressor CS4 has been eliminated from all these schemes. Use of recycle compressor CS3 with CS1 results in cascade HP2-11-4a (Figure 6a); and the use of CS3 with CS2 results in HP2-11-4b in Figure 6b. Elimination of all other recycle compressors except CS2 and CS1 leads to the three-membrane stage cascade HP2-11-3a, shown in Figure 6c.

Similar to the use of two recycle compressors in the stripping section, cascades with both the recycle compressors in the enriching section can be derived from the parent cascade scheme of Figure 2. These cascades are shown in Figure 6d–6g. The cascade with maximum number of membrane stages, HP2-11-5b, is derived by retaining recycle compressors CE4 and CE2. Next recycle compressor CE4 is also eliminated and possible combinations of recycle compressor CE3 with another recycle compressor are considered to provide cascade schemes HP2-11-4c and HP2-11-4d (Figures 6e and 6f). Finally, recycle compressor CE3 is eliminated and recycle compressors CE1 and CE2 are retained, resulting in cascade HP2-11-3b of Figure 6g.

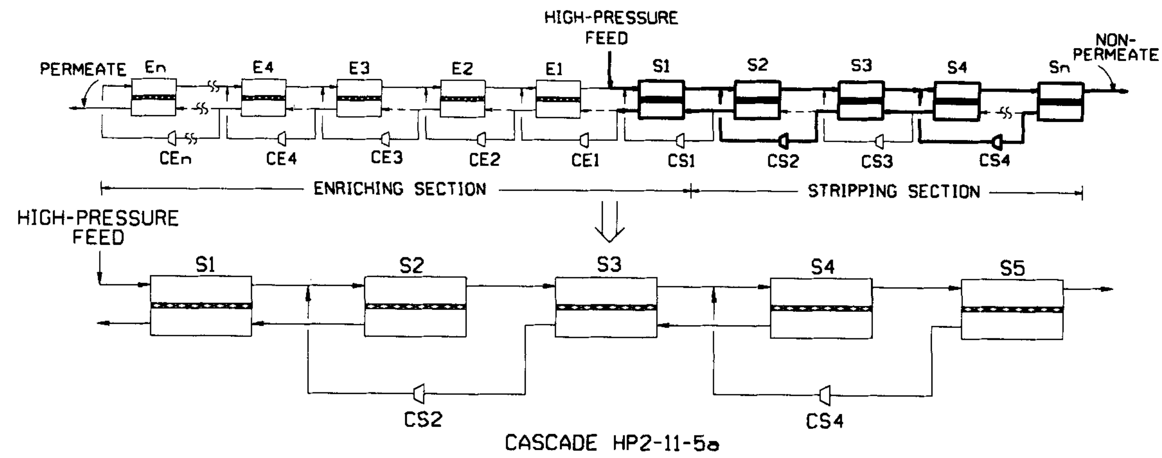

Figure 5. Derivation of cascade scheme HP2-11-5a from the parent symmetrical cascade with $p=q=1$.

To exhaust all possible two-compressor cascades that can be derived from the parent cascade scheme of Figure 2, it is necessary to consider the scheme whereby one recycle compressor is used in the stripping section and the other in the enriching section. Four such cascades are shown in Figure 6h–6k. These are easily generated by following the steps described for the previous two-compressor cascades.

The procedure can now easily be applied to generate all

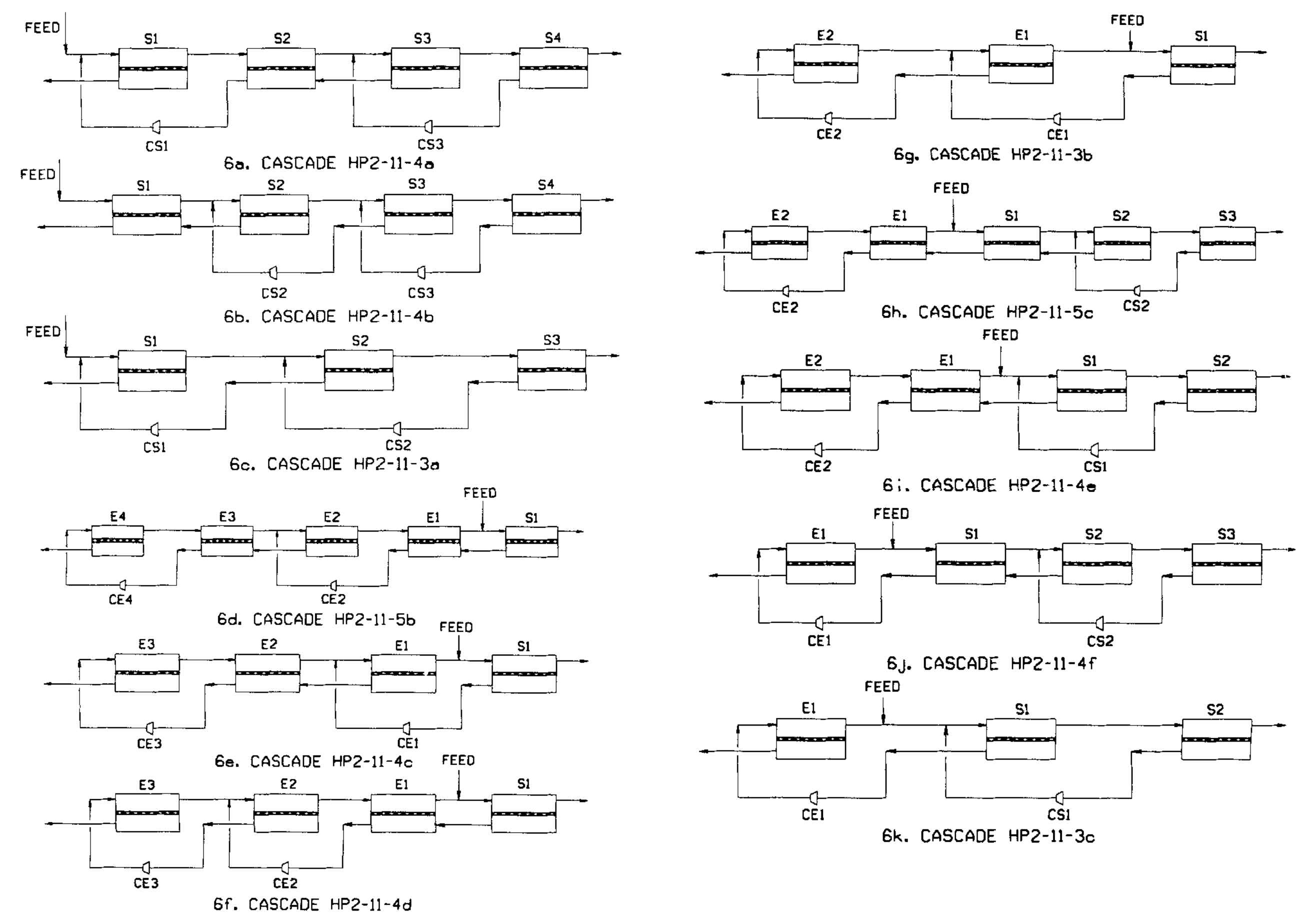

Figure 6. Two-compressor cascades derived from the parent symmetrical cascade of Figure 2.

(a) Cascade HP2-11-4a. (b) Cascade HP2-11-4b. (c) Cascade HP2-11-3a. (d) Cascade HP2-11-5b. (e) Cascade HP2-11-4c. (f) Cascade HP2-11-4d. (g) Cascade HP2-11-3b. (h) Cascade HP2-11-5c. (i) Cascade HP2-11-4e. (j) HP2-11-4f. (k) Cascade HP2-11-3c.

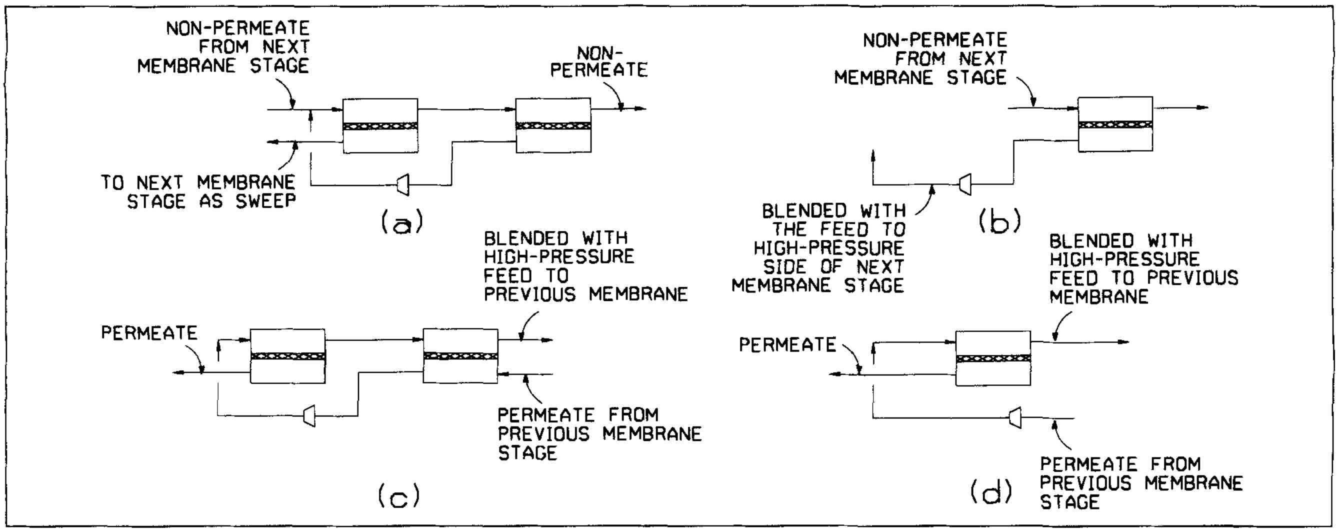

Figure 7. Repeating blocks for the cascades derived from the parent symmetrical cascade scheme of Figure 2.
(a) and (b) For the stripping section. (c) and (d) For the enriching section.

cascades with three or more compressors that are substructures of the symmetrical parent cascade with $p = q = 1$ of Figure 2. From Figures 3, 4b, 5, 6d, and 6h, it is observed that in such n-compressor cascades the maximum number of membrane stages is $2n+1$. This observation makes the task of generating cascades a little easier.

Another interesting observation can be made by comparing the one-compressor cascade schemes of Figures 3 and 4 with the two-compressor cascades of Figures 5 and 6. It is apparent that the two repeating blocks for each stripping and enriching section can be added to the one-compressor cascade schemes to generate all the two-compressor cascades (Figure 7). Therefore, the repeating block shown in Figure 7a can be added in the stripping sections of Figures 3 (HP1-11-3a) and 4 to generate two-compressor cascades of Figures 5 (HP2-11-5a), 6a, 6h, and 6j. Similarly, addition of the repeating block in Figure 7b to these one-compressor cascades leads to the two-compressor cascades of Figure 6b, 6c, 6i, and 6k. The addition of the enriching section block of Figure 7c to the enriching sections generates the cascades shown in Figure 6h, 6i, 6d, and 6e. A similar exercise with the enriching block of Figure 7d leads to the cascade in Figure 6j, 6k, 6f and 6g. Clearly, all of the two-compressor cascades may be derived through this exercise. A similar exercise of adding the repeating blocks to two-compressor cascades will yield all the three-compressor cascades. This provides an alternative procedure for generating n-compressor substructures of the symmetrical parent cascade with $p = 1$ and $q = 1$.

Not all of the known one- and two-compressor cascades are substructures of the symmetrical parent cascade of Figure 2. These other cascade schemes may, however, be derived by applying the same modifications and steps to other cascades with different values of cascade parameters p and q; specifically to two unsymmetrical cascades with $p = 2$, $q = 1$ and $p = 1$, $q = 2$.

First consider the unsymmetrical cascade with $p = 2$, $q = 1$ (Figure 1b). The parent unsymmetrical cascade is created by allowing for the possibility of the permeate stream to be introduced as sweep stream to the low-pressure side of the next membrane stage; this is shown by dotted lines in Figure 8a. All the n-compressor cascades can now be created by systematically generating schemes containing all combinations of n-compressors. For example, one-compressor cascades can be created by retaining either one of the compressors from CS1, CS2, CS3, CE1, or CE2. Figure 8b and 8c show the substructure containing compressor CS1 leading to cascade HP1-21-3a (Xu, 1994a). Note that although the one-compressor cascades shown in Figures 3 and 4 are different from the one-compressor cascade HP1-21-3a, these are also embedded as substructures in the parent unsymmetrical cascade of Figure 8a. For example, elimination of all compressors but CS3 leads to HP1-11-3a, retention of CS2 leads to HP1-11-2a, use of only CE2 gives HP1-11-3b, and the substructure containing only CE1 is HP1-11-2b. It can therefore be concluded that the parent unsymmetrical structure with $p = 2$, $q = 1$ contains all the feasible substructures of the parent symmetrical structure with $p = 1$, $q = 1$, and is more general. The procedure for generating cascades with more than one compressor is the same as that described for symmetrical cascades with $p = 1$, $q = 1$, and will not be repeated here; some examples of such two-compressor cascades can be found elsewhere (Agrawal and Xu, 1996).

Now consider the unsymmetrical cascade with $p = 1$, $q = 2$ (Figure 1c). Besides the modification on the low-pressure side, this cascade also required that a similar modification be made on the high-pressure side (Figure 9a). Thus a high-pressure stream discharging from a membrane stage can either be fed to the immediately preceding membrane stage (dotted lines in the figure) or to the second previous membrane stage. After the creation of the parent cascade scheme of Figure 9a, all desired n-compressor cascade schemes can be generated by retaining n-compressor substructures. An example of a two-compressor cascade HP2-12-3a is illustrated in Figure 9. Note that any n-compressor substructure of the parent symmetrical cascade with $p = q = 1$ is also embedded in the parent unsymmetrical cascade with $p = 1$, $q = 2$. It seems that with the suggested procedure for drawing n-compressor cascades with $n > 1$, an unsymmetrical cascade provides a more general structure than the corresponding symmetrical cascade.

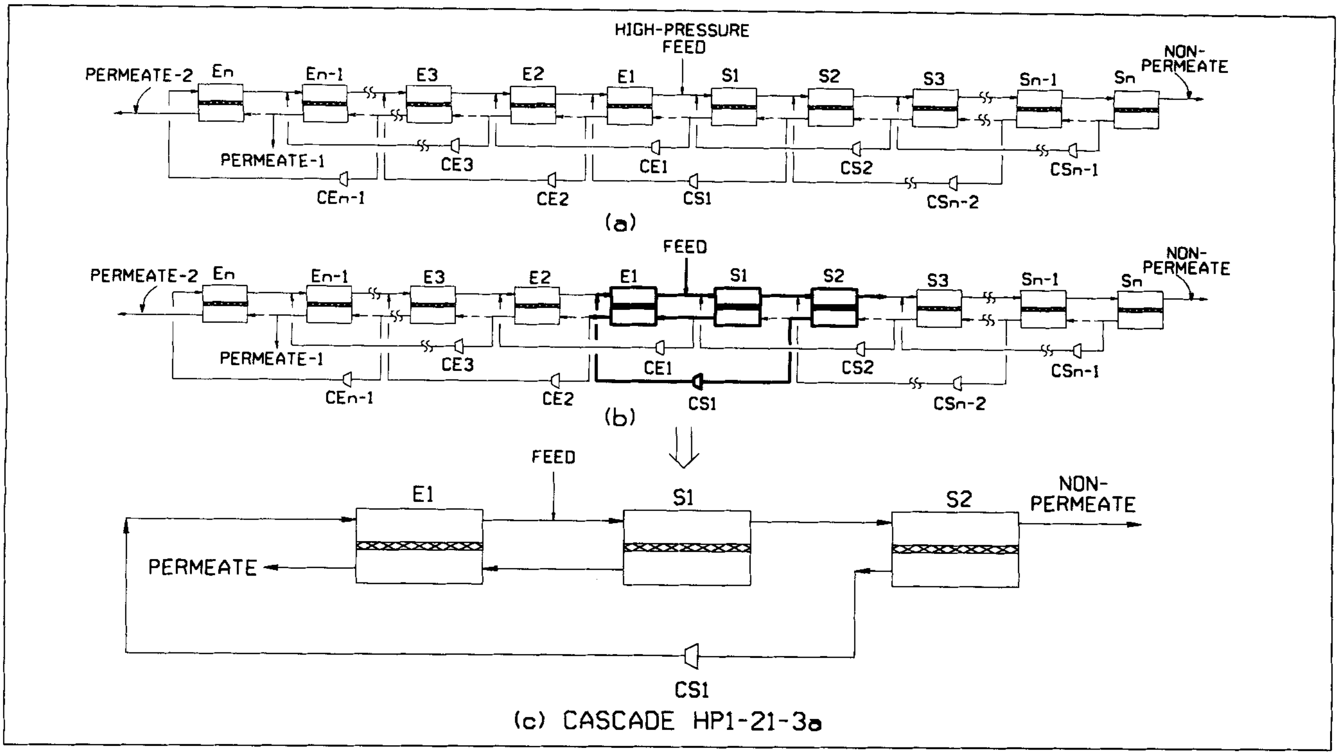

Figure 8. Derivation of cascade HP1-21-3a from the parent unsymmetrical cascade with $p = 2$, $q = 1$.

(a) parent cascade; (b) parent cascade with substructure HP1-21-3a highlighted; and (c) Cascade HP1-21-3a.

The procedure for a high-pressure feed is now complete and can be applied to a cascade with a given set of parameters p and q to generate n-compressor cascades with $n \geq 1$. The parameters p and q can have any suitable set of values.

Low-pressure feed case

A feed gas mixture is often available at a low pressure and needs to be compressed before a separation can be performed. Even though all the high-pressure feed cascades can

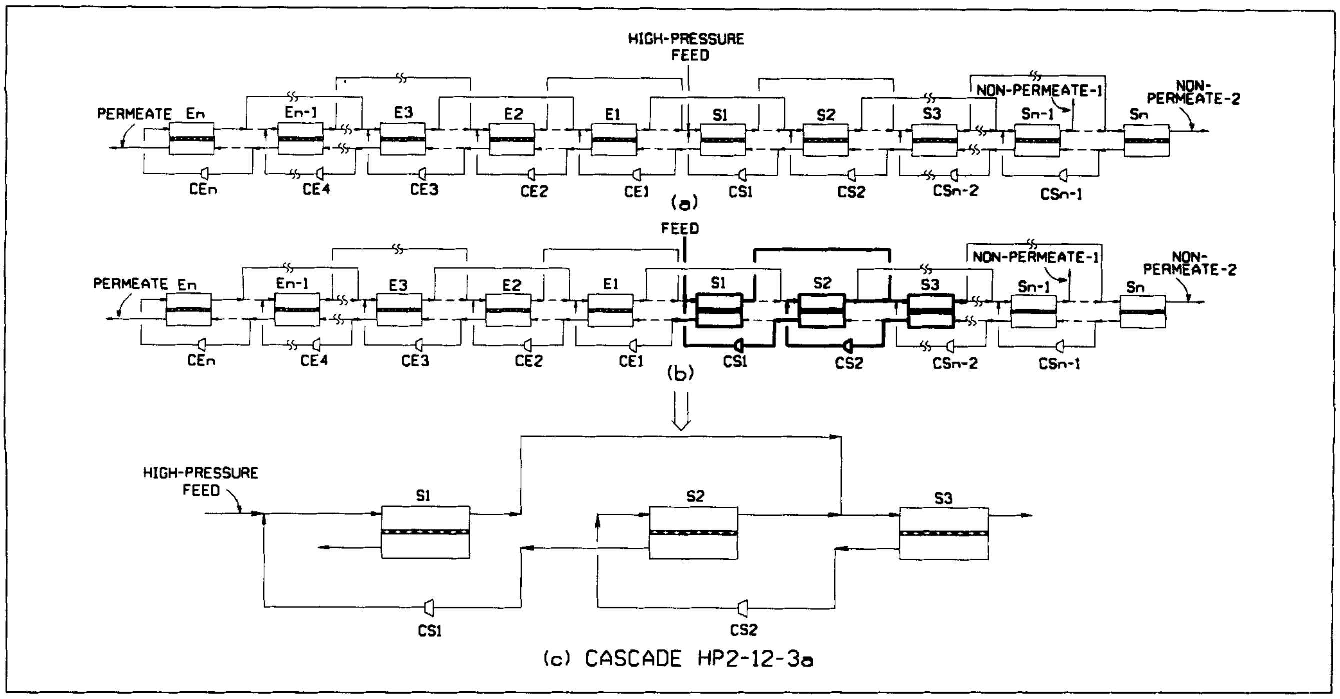

Figure 9. Derivation of cascade HP2-12-3a from the parent unsymmetrical cascade with $p = 1$, $q = 2$.

(a) parent cascade; (b) parent cascade with substructure HP2-12-3a highlighted; and (c) Cascade HP2-12-3a.

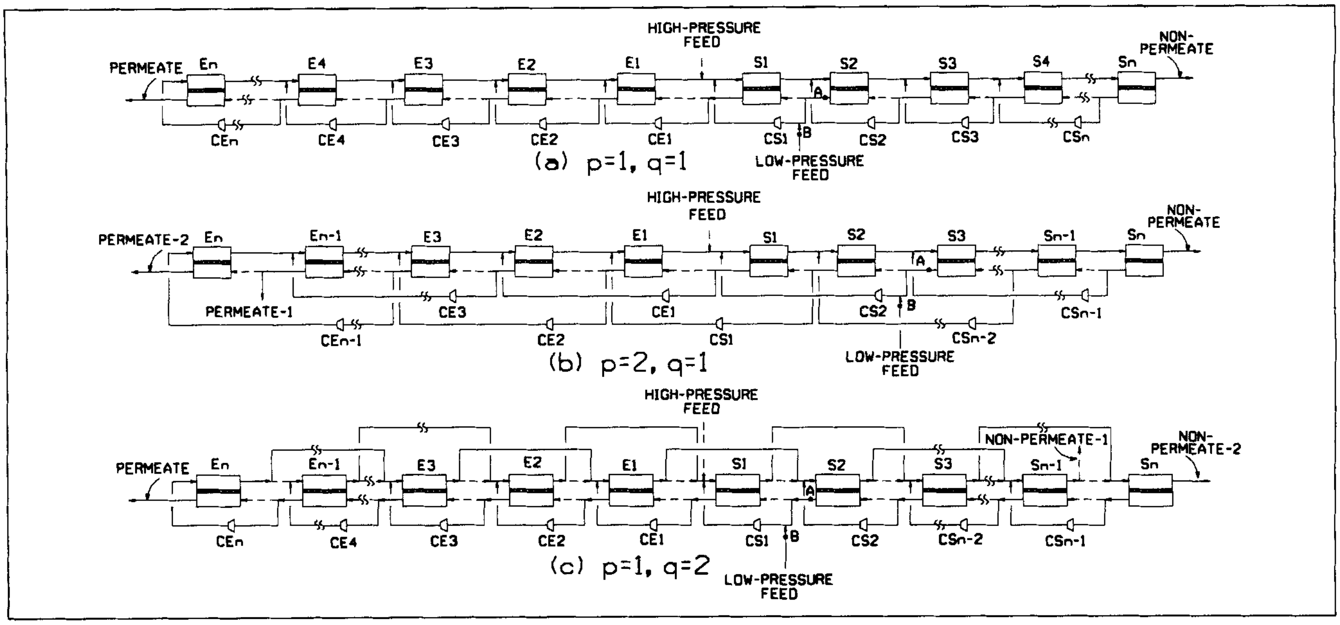

Figure 10. Parent cascades for low-pressure feed case.

(a) $p = 1$, $q = 1$, (b) $p = 2$, $q = 1$. (c) $p = 1$, $q = 2$.

be applied to the low-pressure feed case by adding feed compressors, additional alternative cascade schemes may also be drawn. Once again the procedure for generating these additional n-compressor cascades ($n \geq 1$) starts with the modification of known cascades with a set of values for parameters and p and q (Figure 1). The process of eliminating the unwanted recycle compressors and membrane stages to obtain the desired substructure from a parent cascade is similar to that for the high-pressure feed case. The entire procedure, including the generation of parent cascades for a low-pressure feed, will now be discussed.

The first step is to modify high-pressure feed cascades, such as the ones shown in Figure 1, for the low-pressure feed condition (Figure 10). The modification is achieved by feeding low-pressure feed to one of the recycle compressors. In Figure 10, three modified parent cascades are shown. In these figures the possibility of having a portion of the feed at high pressure is retained. However, without any loss of generality, only cases where all the feed is available at low pressure will be discussed in this section.

Before starting the process of elimination to create cascades that are substructures of a parent cascade, it is worthwhile to note that there are a total of nine distinct possibilities for introducing a low-pressure feed (Figure 11). These nine options differ in the manner in which the permeate stream from the previous membrane stage (flow at point A) is mixed with the low-pressure feed (flow at point B):

Case 1. All of the permeate flow at point A is combined with the low-pressure feed at point B, compressed, and fed to the high-pressure side of a suitable membrane stage (Figure 11a).

Case 2. Permeate flow at point A is not mixed with the low-pressure feed; instead, it is fed as a low-pressure sweep stream to the next membrane stage, and the low-pressure feed is compressed alone and sent to the high-pressure side of a suitable membrane stage (Figure 11b).

Case 3. Permeate flow at point A is not mixed with the low-pressure feed; instead, it is compressed alone and sent to the high-pressure side, and all of the low-pressure feed is fed to the low-pressure side (Figure 11c).

Case 4. All the low-pressure feed is combined with the permeate stream and fed as sweep to the low-pressure side of the next membrane stage (Figure 11d).

Case 5. Only a portion of the low-pressure feed is mixed with the permeate stream and directly fed to the low-pressure side of the next membrane stage, while the other portion of the low pressure feed is compressed and sent to the high-pressure side (Figure 11e).

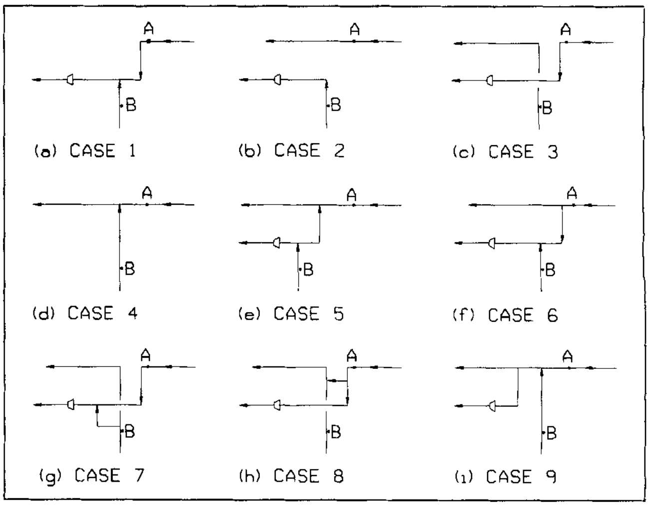

Figure 11. Different cases showing alternate ways to feed a low-pressure feed to a cascade.

Point A denotes permeate stream from the previous membrane stage and point B the low-pressure feed. (a) Case 1. (b) Case 2. (c) Case 3. (d) Case 4. (e) Case 5. (f) Case 6. (g) Case 7. (h) Case 8. (i) Case 9.

Case 6. Only a portion of the permeate stream is combined with all the low-pressure feed; the mixed stream is compressed and sent to the high-pressure side; whereas the other portion of the permeate stream is directly fed to the low-pressure side of the next membrane stage (Figure 11f).

Case 7. A portion of the low-pressure feed is mixed with all of the permeate stream, compressed, and sent to the high-pressure side, whereas the other portion of the low-pressure feed is fed to the low-pressure side of the next membrane stage (Figure 11g).

Case 8. Only a portion of the permeate stream is combined with all of the low-pressure feed, the mixed stream is sent to the low-pressure side of the next membrane stage; the other portion of the permeate stream is compressed and sent to the high-pressure side of a membrane stage (Figure 11h).

Case 9. The low-pressure feed and the permeate streams are mixed and then divided into two portions. One portion is sent to the low-pressure side of the next membrane stage, while the other portion is compressed and sent to the high-pressure side of a suitable membrane stage (Figure 11i).

Basically the preceding nine cases result from all possible combinations of handling the low-pressure feed and the permeate streams. Either the two streams are not mixed at all (Cases 2 and 3) or completely mixed (Cases 1 and 4) or one of the steams can be partially mixed with another stream (Cases 5–8). Furthermore, one has an option to send either of the resulting streams through the compressor or directly as the sweep stream to the permeate side of the next membrane stage. This results in eight options. The last option results when the completely mixed stream is split into two and each resulting stream is treated separately (Case 9).

All of the high-pressure feed cascades can be used as low-pressure feed cascades with a feed compressor. Therefore, when n-compressor substructures are created from the parent cascades, such as the ones shown in Figure 10 for a low-pressure feed, a large number of them are duplicates of the n-compressor high-pressure feed cascades.

In order to create practical n-compressor cascades for a low-pressure feed, it is required that at least one compressor that is downstream of the low-pressure feed point must be retained during the process of elimination of the unwanted compressors and membrane stages from a parent cascade. This ensures that at least one compressor is used in the enriching section and a high-pressure feed is available to a membrane stage in this section. Therefore, in Figure 10a, at least one compressor from compressors CS1, or from series CE must be used. Similarly in Figure 10b, a compressor from CS2, CS1 or from series CE must be used.

The process of elimination and generation of n-compressor cascades with $n \geq 1$ from a parent cascade for low-pressure feed is the same as for the high-pressure feed case. Now it will be briefly discussed for each of the nine cases. Unless specifically mentioned, the illustrative examples of n-compressor cascades will be for those derived from the parent cascade with $p = q = 1$ (Figure 10a). Examples of some of the n-compressor cascades derived from the two other parent cascades with $p = 2$, $q = 1$ and $p = 1$, $q = 2$ can be found elsewhere (Agrawal and Xu, 1996).

Substructures generated according to case 1 are identical to those already available for the high-pressure feed case. For example, cascades HP1-11-2a (Figure 4a), HP2-11-4a, HP2-11-3a, HP2-11-4e, and HP2-11-3c (Figures 6a, 6c, 6i, and 6k) result for this case from the parent cascade with $p = 1$, $q = 1$ (Figure 10a); the trivial modification needed is that the low-pressure feed and the permeate streams are first combined and then sent to the compressor. Similarly, a known low-pressure feed version of cascade HP2-12-3a (Figure 9c) results according to case 1 from the parent cascade with $p = 1$, $q = 2$ of Figure 10 (Prasad, 1994).

For case 2, where one of the recycle compressors is used to compress the low-pressure feed and the permeate stream from the corresponding membrane stage is sent to the low-pressure side of the next membrane stage, all the resulting n-compressor substructures are duplicates of some of the high-pressure feed cascades. An $(n+1)$-compressor cascade with a compressor on the low-pressure feed is equivalent to an n-compressor high-pressure feed cascade. Thus a two-compressor cascade with a compressor in the stripping section derived from the parent cascade of Figure 10a is equivalent to HP1-11-3a (Figure 3). Two three-compressor cascades obtained by retaining two compressors in the stripping section are equivalent to cascades HP2-11-5a (Figure 5) and HP2-11-4b (Figure 6b). When only the compressors in the enriching section are used, the two membrane stages S1 and S2 combine together to effectively provide one membrane stage. In this case, two-compressor cascades that can be derived are equivalent to HP1-11-3b and HP-11-2b (Figure 4b and 4c); and three-compressor cascades are analogous to HP2-11-5b, HP2-11-4c, HP2-11-4d, and HP2-11-3b (Figure 6d–6g). When only one compressor is retained in each of the stripping and enriching sections, the resulting cascades are equivalent to HP2-11-5c and HP2-11-4f (Figure 6h and 6j). When no additional compressors are used in either of the sections, a trivial case results with one membrane stage unit and a feed compressor. It is worth noting that cases 1 and 2 together provide all the equivalent low-pressure feed cascades of the high-pressure feed cascades in Figures 3 through 6 derived earlier for $p = q = 1$.

The modified low-pressure feed parent cascade according to case 3 for $p = q = 1$ is shown in Figure 12a. The one- and two-compressor cascade schemes derived from this structure are shown in Figure 12b–12f. One recycle compressor downstream of the low-pressure feed is always retained. Whenever a membrane stage in the stripping section is used, compressors CS1 must not be eliminated; the only exception is when the permeate from the membrane stage S2 can be treated as a product stream and need not be recycled. For this case, case 3, the maximum number of membrane stages used in an n-compressor cascade is $2n$.

The parent cascade derived according to case 4 for $p = q = 1$ is shown in Figure 13a. Notice that during the process of generating substructures, membrane stages S1 and S2 cannot be eliminated. The resulting substructures containing one and two compressors are shown in Figure 13b–13f. The maximum number of membrane stages used in this case by an n-compressor cascade is $2n+1$.

The modified parent cascade for case 5 with $p = q = 1$ is shown in Figure 14a. In order to generate cascades that are distinct from the other cases, membrane stage S1 and compressor CS1 must always be retained; and a portion of the low-pressure feed should also be sent to the low-pressure side of membrane stage S1. One has an option to eliminate mem-

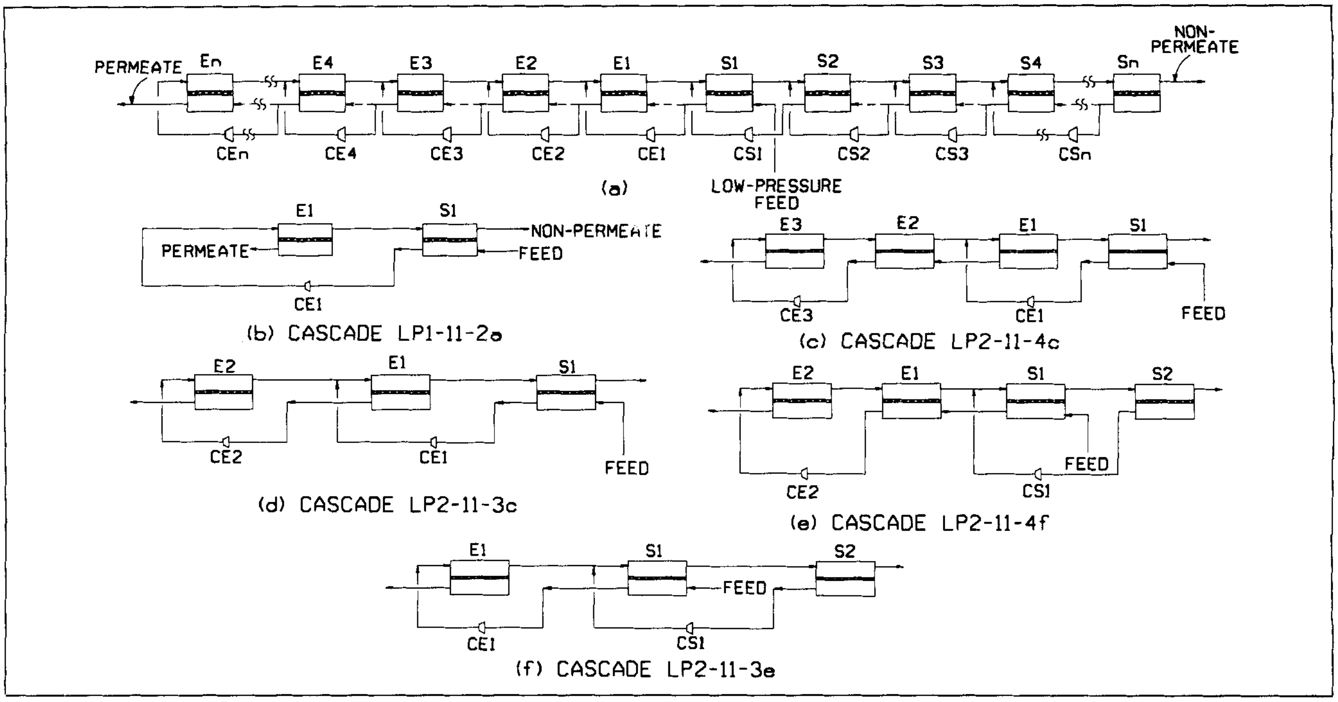

Figure 12. (a) Parent cascade for case 3 with $p=q=1$; (b)–(f) substructures of one- and two-compressor cascades.
(b) Cascade LP1-11-2a. (c) Cascade LP1-11-4c. (d) Cascade LP2-11-3c. (e) Cascade LP2-11-4f. (f) Cascade LP2-11-3e.

brane stage S2 and its associated permeate stream for the substructures when only the membrane stages downstream of S1 are retained, that is, it is possible to eliminate membrane stage S2 when all other membrane stages in the stripping section are eliminated. The resulting one-compressor cascades are shown in Figure 14b and 14c. There are six options for two-compressor cascades that are not shown in Figure 14. The maximum number of membrane stages used in an n-compressor cascade is $2n$.

The parent cascade for case 6 with $p=q=1$ is shown in

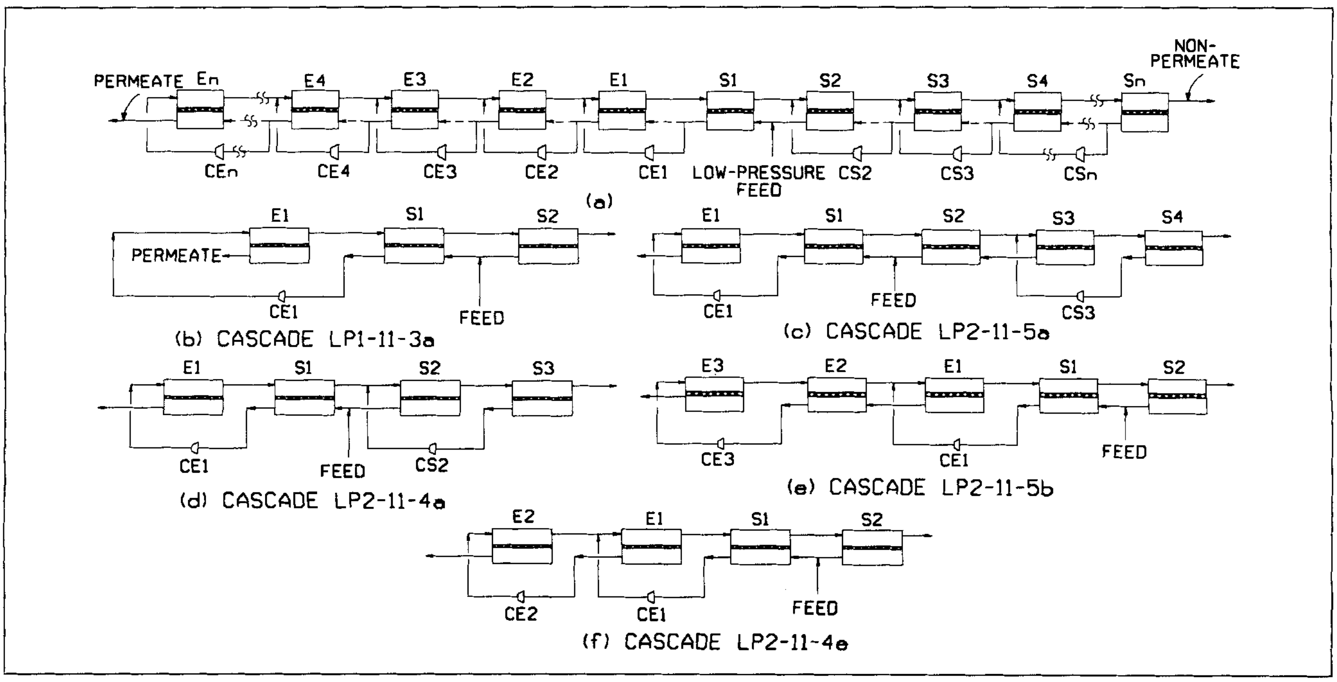

Figure 13. (a) Parent cascade according to case 4 for $p=q=1$; (b)–(f) substructures of one- and two-compressor cascades.
(b) Cascade LP1-11-3a. (c) Cascade LP2-11-5a. (d) Cascade LP2-11-4a. (e) Cascade LP2-11-5b. (f) Cascade LP2-11-4e.

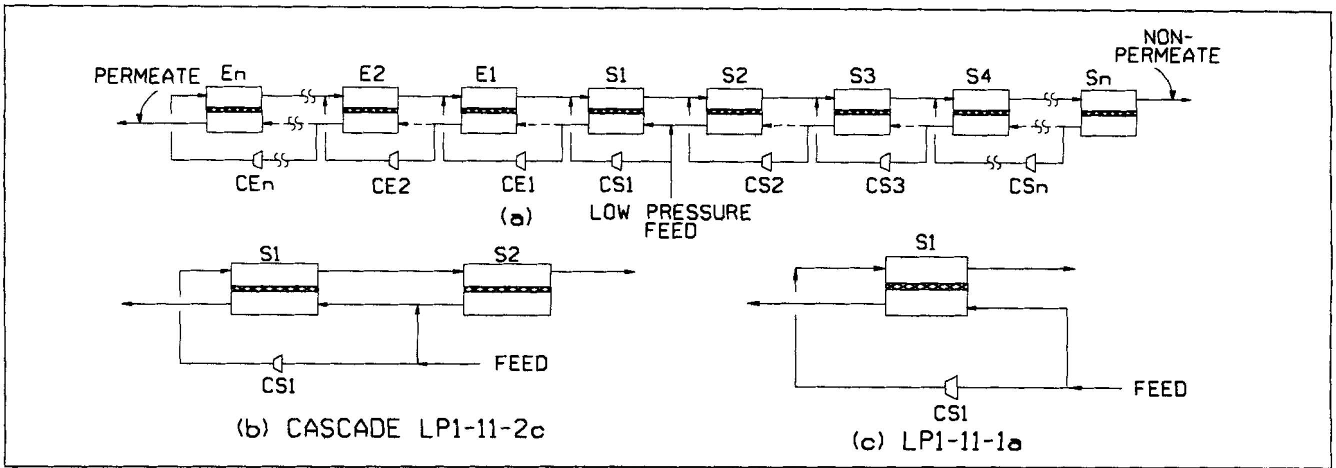

Figure 14. (a) Parent cascade according to case 5 for $p=q=1$; (b) and (c) substructures of one-compressor cascades.

(b) Cascade LP1-11-2c. (c) LP1-11-1a.

Figure 15a. For the substructures of this case to be distinct from other cases, it is essential that both membrane stages S1 and S2 and compressor CS1 be retained in all the derived substructures. A portion of the permeate from membrane stage S2 is sent to membrane stage S1 and the other portion is mixed with the low-pressure feed. Therefore, there is only one substructure with one compressor. When substructures from case 6 are compared with substructures from case 5, it becomes evident that all the substructures from case 5 that contain both membrane stages S1 and S2 can be trivially modified to yield the substructures for case 6. The trivial modification is that rather than mixing a portion of the feed

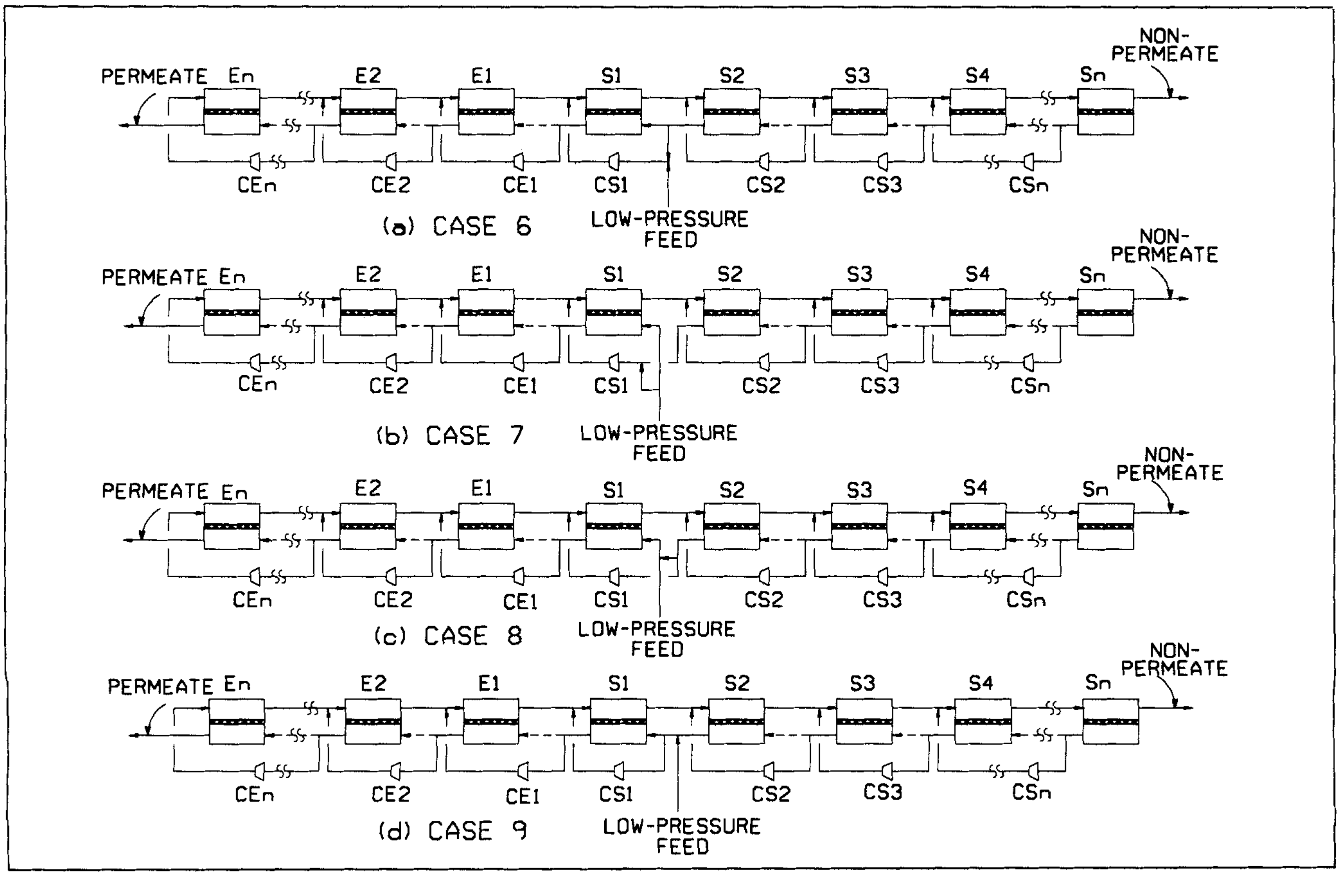

Figure 15. Parent cascades modified according to cases, 6, 7, 8, and 9 for $p=q=1$.

(a) Case 6. (b) Case 7. (c) Case 8. (d) Case 9.

with permeate from S2, now a portion of the permeate is mixed with all of the feed stream and the mixed stream is sent to compressor CS1. For a given feed flow rate, case 6 will have higher flow through compressor CS1 than case 5. There are four cascades that are two-compressor substructures of the parent cascade in Figure 15a, and the maximum number of membrane stages used in these cascades is four.

Figure 15b shows the modified parent cascade according to case 7 ($p = q = 1$). Once again, for all the substructures to be distinct from all other cases, both membrane stages S1 and S2 and compressor CS1 must be retained in all the substructures. Clearly the substructures for this case can also be easily obtained by trivial modification of corresponding cascade schemes that contain both membrane stages S1 and S2 in case 5. The trivial modification is that the mixed stream of the permeate and the feed is to be sent to compressor CS1 and the nonmixed portion of the feed is fed to the low-pressure side of membrane stage S1.

The parent cascade for case 8 with $p = q = 1$ is shown in Figure 15c. Distinct cascades result as substructures when both membrane stages S2 and S1 and compressor CS1 are retained during the process of elimination. Once again substructures for this can be obtained by trivial modification of substructures derived according to case 5 for $p = q = 1$ and that contain both membrane stages S2 and S1.

The modified parent cascade according to case 9 for $p = q = 1$ is shown in Figure 15d. To derive distinct cascades, both membrane stages S1, S2, and compressor CS1 must be retained in all the substructures. The relation between the substructures derived for this case and the cascades derived for case 5 is similar to the relation between cascades from either cases 6, 7, or 8 and cascades from case 5. It is worth noting that when the composition of the feed and the permeate stream to be mixed is same, then cases 6, 7, and 8 become subsets of case 9; also substructures containing both membrane stages S1 and S2 in case 5 become subsets of this case.

The preceding procedure described for generating low-pressure feed cascade containing n-compressors ($n \geq 1$) from a parent cascade with $p = q = 1$ can be applied to any parent cascade with a different value of the cascade parameters p and q.

Additional comments

Now some comments will be offered to incorporate other known means of operating membrane stages within a membrane cascade.

Membrane Flow Mode. In this article all the membrane stages are shown as operating in countercurrent mode. However, the procedure described is independent of the mode of operation and is equally valid when a membrane stage is operating in a cocurrent, crossflow, or a mixed-flow mode.

Membrane Pressure Ratio. Within a cascade it is not necessary that each membrane stage operate at the same pressure ratio. The overall performance of a cascade could be improved by adjusting the pressure ratio for each membrane stage to bring it closer to its optimum value. For example, in cascade HP1-11-3a (Figure 3), the pressure ratio in membrane stage S3 could be different from the ones used in membrane stages S2 and S1. Furthermore, it has been suggested that in certain cases it can be beneficial to use intermediate permeate compressions. In such cases, a membrane stage is broken into multiple substages, with permeate from one substage compressed and sent to the next substage as sweep on the permeate side (Xu, 1993). This decreases the pressure ratio along the direction of the permeate flow in a membrane stage. An analysis method based on the local thermodynamic efficiency of permeation is presented by Xu and Agrawal (1996a) to identify situations where this concept can be applied. In such cases, any suitable membrane stage of an n-compressor cascade can be easily modified to incorporate intermediate permeate compressions.

Membrane Elements. It is worth mentioning that in an n-compressor cascade, all the membrane stages may not contain the same membrane elements. Depending on the availability, different membrane stages could use membrane elements with different preselective properties to achieve a better overall solution.

Feeds at Multiple Pressures. In the derivation of n-compressor cascades with $n \geq 1$, it has been assumed that all the feed is available as one feed stream either at high pressure or at low pressure. If a feed is available at two pressure levels, then the corresponding substructures can be easily derived by having both the low- and high-pressure feed streams in the parent cascade, as shown in Figure 10. If feed streams are available at more than two pressure levels, then if a recycle compressor is a multistage compressor, one may be able to feed these streams in different stages of the recycle compressor. The same concept may be applied to cases where a permeate stream is to be mixed with the low-pressure feed and then compressed for feeding to the next membrane stage (e.g., cases 1, 6, and 7 and Figure 11), but their pressures do not match; in this case the permeate and the low-pressure feed streams may be fed to different stages of a compressor.

Feeds of Different Compositions. When multiple feed streams of different compositions are available, then a parent cascade can be easily modified by feeding each feed stream to an appropriate membrane stage. For binary feeds, the composition of the feed stream should generally match that of the stream with which it is being mixed. Thus a parent cascade can be modified to incorporate multiple feed streams of varying compositions and pressures to yield the desired n-compressor substructures.

Multiple Product Streams. In all of the n-compressor cascades derived in this article, there is only one permeate and one nonpermeate product stream. However, if needed, the ability to produce more than one permeate or nonpermeate product streams can be incorporated. This can be done in several ways. In the first method, no modification to the parent cascade is made and a derived n-compressor cascade producing one permeate and one nonpermeate product stream is taken as the starting point. This n-compressor cascade is then modified to produce an additional product stream that is either a portion or all of a suitable recycle stream from an appropriate membrane stage to yield a cascade with multiple permeate product streams. For example, cascade HP2-12-3a (Figure 9c) can be modified to produce two additional permeate product streams by taking a portion of the permeate stream from membrane stage S3 or by taking a portion or all of the permeate stream from stage S2 as product streams. The case where all of the permeate stream from membrane S2 is taken as a product stream, along with the permeate product stream from stage S1, leads to a membrane cascade that was originally studied by Spillman (1989) for treating natural gas. Similarly, from cascade HP2-12-3a, additional nonpermeate product streams can be produced by taking a portion of the nonpermeate stream from stage S1 or a portion or all of the nonpermeate stream from stage S2 as product streams. It is worthwhile mentioning that there is always the possibility of producing additional product streams from an intermediate location of any membrane stage. Thus, from an intermediate location of the membrane stages S1, S2, or S3, a permeate or a nonpermeate product stream can be produced by dividing the membrane stage into two substages.

Probably the easiest way to draw cascades with multiple product streams is to incorporate this possibility in the parent cascade structure. Unsymmetric parent cascades, such as the one described in Figures 8a, 9a, 10b and 10c, have some of this possibility already built in. In Figure 8a, a portion or all of the permeate stream from membrane stage E_{n-1} can be recovered as an additional product stream; the same is true for the recovery of a nonpermeate stream from stage S_{n-1} in Figure 9a. In order to create more possibilities, some additional modification to the structure of a parent cascade may be needed. One modification would be to add the possibility that a portion or all of the permeate stream from a membrane stage can be collected as additional product stream. This possibility would be in addition to the one that the permeate stream from this membrane stage can either be compressed and recycled to the high-pressure side of the succeeding pth membrane stage or fed as low-pressure sweep stream to the next membrane stage. Similarly, the possibility of producing a portion of the nonpermeate product stream from a membrane stage can be added to the feeding of this nonpermeate stream to the previous qth membrane stage. In some cases, the possibility exists of producing from certain membrane stages all the nonpermeate stream rather than a portion of this stream as an additional product stream.

Splitting of Permeate Flows. Note that a possibility for deriving n-compressor cascades that has not been discussed in detail in this article is to not recycle all the permeate from a membrane stage to the high-pressure side of the succeeding pth membrane stage, but to do so with only a portion of the permeate stream, and to send the rest of the permeate stream to the low-pressure side of the next membrane stage as sweep. If needed, this possibility can be easily included in any parent cascade or in an n-compressor cascade already derived according to the procedure outlined in this article. For example, in cascade HP1-11-3a (Figure 3) a portion of the permeate stream from membrane stage S3 could be sent as sweep to the low-pressure side of membrane stage S2.

Conclusion

A systematic procedure has been introduced to draw n-compressor cascades with $n \geq 1$ for gas separation. The procedure starts with the selection of a known membrane cascade with certain values of the cascade parameters p and q. This cascade is then modified by adding the possibility of sending the permeate stream from one membrane stage to the next membrane stage. Similarly, for cases where $q > 1$, an option is added of sending the nonpermeate stream from one membrane stage to the immediately preceding membrane stage. For a high-pressure feed that can be fed directly to the high-pressure side of a membrane stage, no further modification is needed and one obtains a parent cascade. However, for a low-pressure feed case, where the feed stream must be compressed prior to feeding it to the high-pressure side of a membrane stage, it is added to the low-pressure side of a membrane stage to yield the corresponding parent cascade structure.

The structure of a parent cascade contains various n-compressor cascades as substructures. These substructures are derived by eliminating unwanted membrane stages and recycle compressors. For a given number of compressors, generally more than one substructure exists. This array of n-compressor cascades can then be analyzed for a given application. The process of eliminating the unwanted membrane stages and recycle compressors while retaining the required ones to yield all possible substructures is illustrated for one- and two-compressor cases. This is done for both high-pressure and low-pressure feed cases.

Low-pressure feed situations present many more optional configurations than do high-pressure feed cases. This results primarily from the manner in which the low-pressure feed stream and the permeate stream from a membrane stage can be mixed; nine different cases are identified for this purpose. Since all the high-pressure feed cascades can be treated as low-pressure feed cascades with a feed compressor, the high-pressure feed cascades can be derived from parent cascades for low-pressure feeds.

The proposed method yields an array of one- and two-compressor cascades for both high-pressure and low-pressure feed cases. It not only gives all the known cascades in the literature but provides a large number of new cascade schemes. These new cascades present many new options for any particular separation task. This increases the likelihood of finding an economically viable membrane cascade for gas separation.

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