Synthesis of Reverse-Osmosis Networks for Waste Reduction

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The purpose of this work is to introduce the novel notion of synthesizing reverse-osmosis networks (RONs) for waste-reduction applications. The RON design task aims at synthesizing a network of reverse-osmosis units, booster pumps and energy-recovery devices that can separate a set of waste streams into lean (product) streams and rich (retenate) streams at minimum total annualized cost. A systematic and generally applicable procedure for tackling RON synthesis problems is developed. First, a structural representation is devised to embed all potential network configurations. Then, the problem is formulated as an optimization program whose objective is to minimize the total annualized cost of the network while satisfying all environmental and technical requirements. The solution to this program provides the optimal arrangement, types and sizes of the reverse-osmosis units, the booster pumps, and the energy-recovery turbines to be employed in the network. Furthermore, the solution also provides the optimum stream distribution, operating conditions, and separation levels. The applicability of the proposed synthesis technique is demonstrated by tackling case studies and comparing the optimal solutions with current industrial practices.

Introduction

Hazardous-waste minimization means the reduction, to the extent feasible, of hazardous waste that is generated prior to the treatment, storage, or disposal of the waste. It represents the most efficient method for preventing the release of hazardous substances to the environment. An effective way of tackling waste-minimization problems is through the use of on-site recycle/reuse separation networks. In addition to its favorable environmental impact, the adoption of this waste-recovery approach can provide an economically attractive waste-management alternative. It can lead to significant energy and raw-materials savings and possibly provide income from the salable wastes. With more than 250 million tons of hazardous wastes disposed of at the generating sites throughout the United States annually (Warren, 1990), the proposition of waste recycle/reuse is becoming an essential industrial practice.

The growing awareness of the consequences of discharging effluents into natural resources have lead to corrective measures both voluntary and legislated as demonstrated by the consistent trend to tighten environmental requirements on the concentration of hazardous species in the industrial waste streams. This trend calls for achieving unprecedented and progressively higher degrees of separation. In this context, reverse-osmosis "RO" systems are finding growing applications in the field of hazardous-waste minimization. The availability of numerous types of strongly selective membranes makes it possible to accomplish very high separation levels for virtually all hazardous species (for example, Clark, 1990; Palmer et al., 1988; Borup and Middlebrooks, 1987; and Cushnie, 1985). Since RO does not involve phase changes, its energy consumption is normally moderate. By virtue of their size compactness, RO systems can be conveniently added to existing plant structures to accommodate any prospective environmental regulations. Furthermore, since an RO system is typically installed as a network of modules, its modular configuration can be exploited to provide favorable flexibility characteristics for design and operation. Sections of the network can be turned on or off in response to quantitative and qualitative variations in the influx of the waste streams.

While the behavior of individual RO modules has been extensively studied in the past three decades, very little attention has been directed towards the task of designing systems of multiple RO modules. Since an RO process typically involves the use of several (in some cases hundreds of) units, the complex problem of optimally designing these systems should be thoroughly investigated. In fact, the success in the challenge of effectively employing RO systems in the novel applications of hazardous-waste minimization appears to hinge upon the development of systematic procedures for the optimal design for large-scale RO processes. The establishment of such an optimal-design methodology represents a knowledge breakthrough which can significantly alter the current industrial practices of waste management. Since the present work aims at generating cost-effective and environmentally-sound recycle/reuse networks, its success can considerably enhance the competitiveness membrane-separation processes in waste-reduction applications.

The development of a systematic methodology for the optimal design of RO systems is not a straightforward optimization task. The problem complexity stems from the following considerations:

- Since the RO process may involve a large number of units, the task of optimally arranging these modules (in a series, a parallel or a hybrid scheme) is highly combinatorial. For each unit, optimum type, capacity and operating conditions should be identified.
- In addition to RO units, the use of booster pumps and energy-recovery devices should be thoroughly examined. The optimization task involves the determination of their locations and sizes.
- Besides the arrangement of RO units, pumps and turbines, an optimal distribution of the streams should be sought with the purpose of identifying the necessary recycle, bypass and/or mixing of the streams.
- Multiple waste streams may be fed to the recycle/reuse network. Each of these streams may contain several hazardous species to be removed.

The above complications present us with a challenging task of practical importance. The high dimensionality of the problem renders any exhaustive-enumeration technique for screening all the design alternatives impractical or even impossible. Instead, a systematic and practical procedure ought to be developed for tackling this important design problem. The application of process-synthesis techniques has been instrumental to the efficient design of other separation systems. This has lead to well-established procedures for the optimal design of distillation networks [see Liu (1987) and Westerberg (1985) for very good reviews] as well as mass-exchange networks (El-Halwagi and Srinivas, 1992; El-Halwagi and Manousiouthakis, 1990a,b, 1989). The development of equivalent procedures for RO systems can lead to profound impact on the economic as well as the technical viability of these systems.

The present work aims at developing a systematic and generally applicable methodology for tackling the problem of optimally designing reverse-osmosis networks (RONs). The problem will be formulated as an optimal network-synthesis task. This entails the development of a finite structural representation of a RON which is rich enough to embed all potential process alternatives of interest. Such a general configuration will account for all the system components (RO units, pumps and energy-recovery devices) as well as stream distribution (bypass, recycle, mixing, and so on). The problem is then formulated as an optimization program which seeks to meet all the design objectives (economic, environmental, technical, and so on) no matter how incompatible they may be. The solution of this optimization problem provides the optimal arrangement of the units, size(s) and type(s) of the RO modules to be employed (hollow fiber, tubular, and/or spiral wound), distribution of the streams, energy consumption and recovery, and separation levels. Illustrative examples will be presented to demonstrate the effectiveness of the proposed procedure.

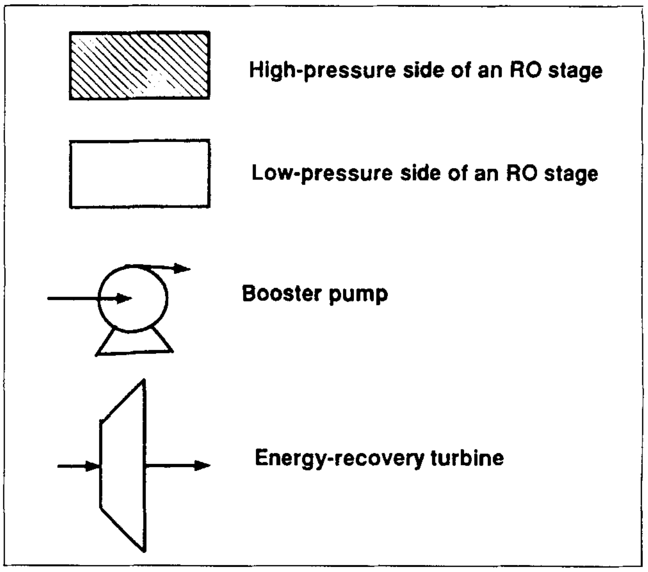

Figure 1. Symbols used in representing an RO system.

Literature Survey

While vigorous research efforts have been directed towards the study of individual RO modules, much less work has been conducted in analyzing systems of multiple RO modules. The earliest attempt to optimize an RO system with several modules dates back to the work of Fan et al. (1968, 1969). In their work, Fan et al. analyzed a multistage RO configuration with intermediate pumps. Each stage may be composed of several RO units connected in parallel. The reject stream leaving each stage was split into two portions; the first was fed to the next stage while the latter was recycled to the stage inlet. An energy-recovery device was proposed to exploit the pressure energy of the final reject stream leaving the system. Figure 1 illustrates the symbols used in representing the RO systems, whereas Figure 2 shows a schematic representation of the sequential arrangement proposed by Fan et al. By employing the discrete version of the maximum principle, Fan et al. minimized the

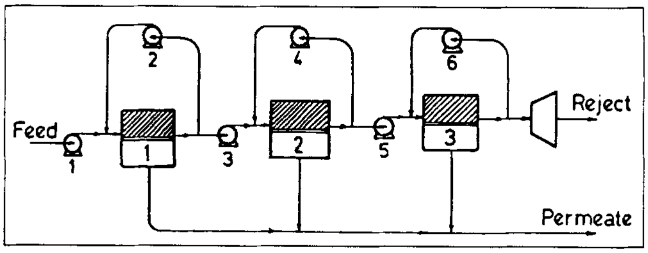

Figure 2. RO process configuration proposed by Fan et al. (1968).

Courtesy of Elsevier Science Publishers and Fan et al.

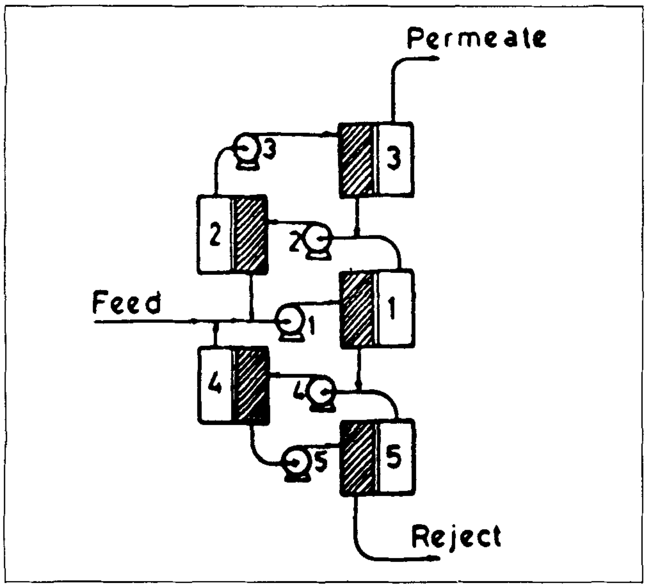

Figure 3. Multistage RO cascade of Kimura et al. (1969).
Courtesy of the American Chemical Society.

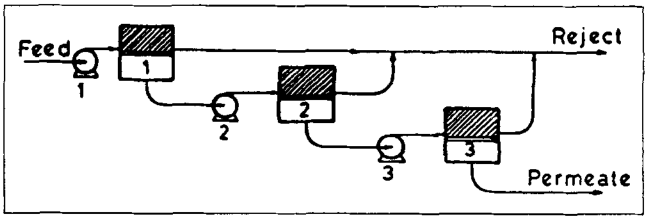

Figure 5. Permeate-processing RO system.

total cost per unit quantity of water produced for a three-stage series system. Later, Hatfield and Graves (1969) employed nonlinear programming techniques to maximize the productivity of a sequential structure with respect to the membrane fabrication temperatures. They used an RO serial structure similar to that of Fan et al., yet they did not allow for intermediate reject pumping or recycle.

By capitalizing on the conceptual representation of a multistage fractionation column, Kimura et al. (1969) proposed an RO scheme composed of two sections; one above the feed for the purification of the permeate and one below the feed for the concentration of the reject (Figure 3). They employed stream recycle for each stage to emulate the role of reflux in distillation. Following the formalism of the multistage distillation process along with several simplifying assumptions on the unit modeling, Kimura et al. derived design equations for the RO systems. Later, Perona and Dillon (1972) investigated the optimal membrane permeability for the RO configuration of Kimura et al. They have deduced that the minimum number of RO stages does not often correspond to the minimum total cost.

Bansal and Wiley (1973) investigated the optimal membrane area and operating conditions for a three-stage RO system with sequential reject concentration. They proposed the use of bypass around the second stage and recycle for the third stage (Figure 4). In contrast to the reject staging employed by Fan et al. (1968, 1969), one can use permeate staging to enhance the product purity. Such configurations (Figure 5) have been proposed in literature (McCutchan and Goel, 1974; Evangelista, 1989).

Influenced by the abovementioned research efforts, industrial practice in installing large-scale RO systems appears to favor two modular structures: the straight-through (or squared-off) and the tapered (or Christmas-tree) flow arrangements (for example, Rautenbach and Albrecht, 1989; Kimura et al., 1985; van Dijk et al., 1984; and Hwang and Kammermeyer, 1975). Both of these schemes consist of a number of stages in series. Within each stage, multiple RO modules are connected in parallel. In the straight-through flow scheme, the number and size of the parallel modules in any stage are identical to those in all other stages. Typically, the reject stream leaving each stage represents the feed to the next stage (Figure 6). As the permeate is removed, the flow rate of the feed entering each stage falls steadily. This reduction in the feed velocity worsens the mass-transfer coefficients and increases concentration polarization. These undesirable effects can be counteracted by employing successively lower numbers or smaller sizes of RO units in each stage. This is the essence of the tapered-flow arrangements (Figure 7). Based on a number of heuristic rules, several short-cut methods were proposed for the design of straight-through and tapered RO systems (Harris et al., 1976; Sirkar and Rao, 1981; Sirkar et al., 1982; Sirkar and Rao, 1983; Evangelista, 1985, 1986a,b, and 1989). In all these methods, neither intermediate pumps, nor energy-recovery devices were employed. Furthermore, the use of stream recycle and bypass was not allowed.

Despite the usefulness of the abovementioned computational procedures for designing RO systems, each of these methods suffers from at least one of the following serious limitations:

Inadequate Problem Representation. Invariably, none of

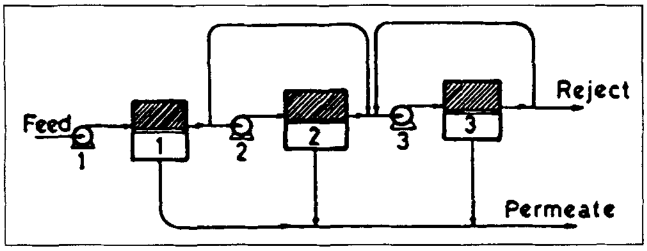

Figure 4. Three-stage configuration of Bansal and Wiley (1973).
Courtesy of *Tappi*.

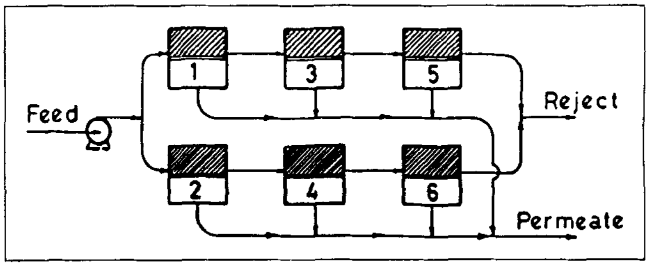

Figure 6. Straight-through reject-processing flow arrangement of an RO system.

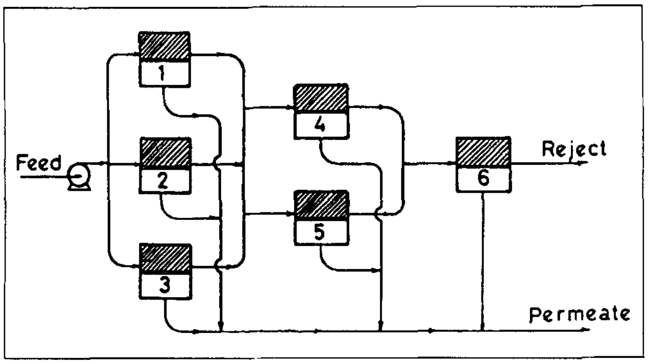

Figure 7. Tapered reject-processing flow arrangement of an RO system.

the foregoing works has presented a problem representation that is rich enough to embed all potential process configurations of interest. This observation can be readily inferred by noting that none of the schemes demonstrated by Figures 2–8 represents a superset that contains all the other configurations shown in these Figures. Indeed, each of these arrangements lacks more than one of the following topographies.

Stream Bypass and/or Recycle. As has been previously discussed, only the work of Bansal and Wiley accounted for bypass in the problem representation; albeit it was around one stage. Similarly, recycle was included only in the works of Fan et al. (1968, 1969); Kimura et al. (1969); and Perona and Dillon (1972). Nonetheless, only the recycle of reject to the proceeding stage was allowed. Based upon mass-transfer arguments, one can show that it may be advantageous to have partial recycle of the permeate. Similarly, it may be desirable to allow for stream bypass and/or recycle that is not necessarily implemented around the same stage. Furthermore, mixing of streams with different compositions (although not thermodynamically desirable) may be economically favorable.

Intermediate Pumps and/or Energy-Recovery Devices. With the exception of the work of Fan et al. (1968, 1969), the use of an energy-recovery device was not exploited despite their economic potential.

Multiple Types of RO Units. In all the foregoing research efforts, single types of RO units were exclusively employed. Since the selection of each type of RO modules (hollow fiber, tubular and spiral wound) is influenced by the operating conditions which may change significantly throughout the RON, it may be advantageous to simultaneously employ more than one type of RO units.

Unrealistic Assumptions and/or Models. In developing short-cut procedures, the majority of the abovementioned works have invoked a number of unrealistic assumptions and/or unreliable models (for example, no solute transport through the membrane, no pressure drop across the unit, constant permeate recovery ratio for all units, oversimplified mixing patterns in the high-pressure side, and so on). Such assumptions may prevent the design procedure from generating a meaningful solution.

Single Feed Streams. All the previously reviewed literature works were developed to tackle RO problems with a single feed stream. This is not the case for waste-minimization problems which may involve multiple waste streams. Due to the potential interaction of these streams, an optimal-design methodology must consider waste recovery from all these streams simultaneously.

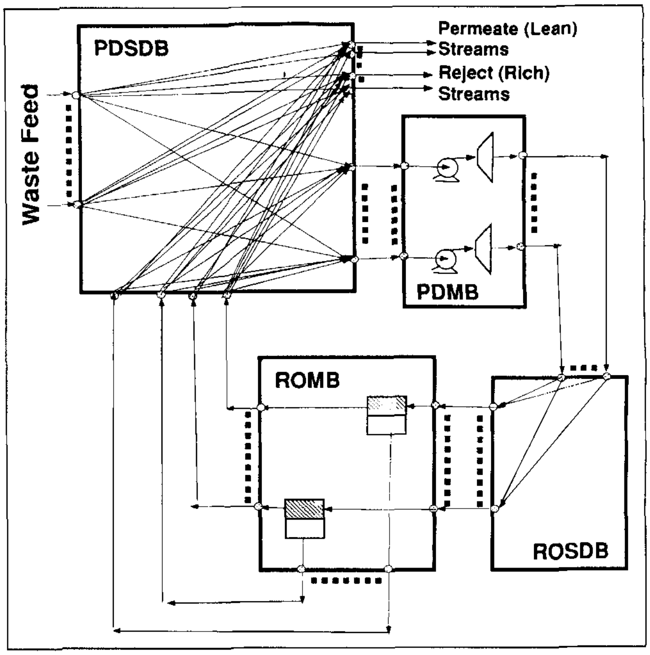

Figure 8a. State-space representation of a RON.

One-Component Separations. Since the majority of the previous large-scale RO research efforts were developed to address water-desalination problems, all of these works were invariably confined to separation tasks with a single (key) component. Nonetheless, waste-reduction problems are typically characterized by the need to simultaneously recover several hazardous species. Indeed, environmental constraints on

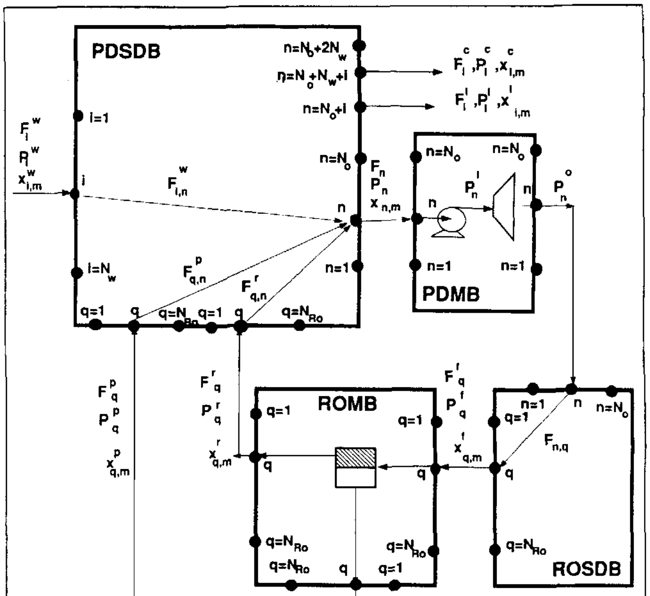

Figure 8b. Nomenclature employed in RON representation.

all the undesirable components must be met, no matter how incompatible they may be.

Problem Description

Problem statement

Given a set $W=\{i|i=1, N_w\}$ of waste streams each containing a set $S=\{m|m=1, N_s\}$ of hazardous species, it is desired to synthesize a cost-effective network of RO units, pumps and energy-recovery devices that can separate each of the waste streams into an environmentally-acceptable lean (permeate or product) stream and a rich (reject or retenate) stream in which the hazardous compounds are concentrated.

Given also are:

- The flow rate of each waste stream, F_i^w, its pressure, P_i^w and a composition set $X_i^w=\{x_{i,m}^w | m=1, N_s\}$
- A lower bound on the flow rate of each lean stream (a minimum required flow rate of product), $F_i^{l,min}$, that is

$$F_i^l \geq F_i^{l,\min} \quad i \in W \tag{1}$$

- The concentration of each compound in a lean stream is bounded by a given environmentally-based composition, $x_{i,m}^e$, that is

$$x_{i,m}^l \leq x_{i,m}^e \quad \begin{matrix} i \in W \\ m \in S \end{matrix} \text{ and} \tag{2}$$

Network representation

The first step in synthesizing the RON is to develop a structural representation of the network that is rich enough to embed all potential configurations of interest. Such a general configuration will account for all the system components (RO units, pumps, and energy-recovery devices) as well as stream distribution (bypass, recycle, mixing, and so on). The proposed representation is inspired by the "state-space approach" that has been recently introduced (El-Halwagi and Manousiouthakis, 1990; Manousiouthakis et al., 1990; Manousiouthakis and Bagajewicz, 1990) for the synthesis of mass, heat, and distillation networks. The RON is conceptualized as being composed of four boxes: a pressurization/depressurization stream-distribution box (PDSDB), a pressurization/depressurization matching box (PDMB), a reverse-osmosis stream-distribution box (ROSDB) and a reverse-osmosis matching box (ROMB). The objective of the PDSDB and the ROSDB is to span the necessary space for the distribution of all the streams. On the other hand, the role of the PDMB and the ROMB is to embed all potential matching states for the distributed streams. Figure 8a demonstrates a schematic representation of the proposed state-space conceptualization. Figure 8b presents the state-space representation for a generic stream along with the nomenclature employed.

Each stream entering the PDSDB passes through an inlet junction. Thus, the PDSDB has a set $J1=\{i|i=1, N_w\}$ of inlet junctions for the waste feed streams, a set $J2=\{q|q=1, N_{RO}\}$ of inlet junctions for the permeate streams entering the PDSDB and a set $J3=\{q|q=1, N_{RO}\}$ of inlet junctions for the reject streams entering the PDSDB. Furthermore, the PDSDB features a set $J4=\{n|n=1, N_o+2N_w\}$ of outlet junctions. Each stream entering the PDSDB is split into N_o+2N_w "substreams" each of which connects an inlet and outlet junctions. Hence, $F_{i,n}^w$, $F_{q,n}^P$, and $F_{q,n}^r$ denote the flow rates of the distributed substreams connecting an outlet junction n with the inlet junctions i (of a waste feed stream), q (of a permeate stream), and q (of a reject stream), respectively. Each of the first N_o outlet junctions is assigned to an outlet stream whose flow rate, pressure and composition are denoted by F_n, P_n and $x_{n,m}$, respectively. These streams proceed to the PDMB. On the other hand, the streams leaving the next N_w outlet junctions of the PDSDB represent the lean (product) streams leaving the network. Their flow rates, pressures, and compositions are represented by F_i^l, P_i^l, and $x_{i,m}^l$, respectively, where $i=1, 2, \ldots, N_w$. Similarly, the concentrated rich (retenate) streams leave the next N_w outlet junctions of the PDSDB. These streams have flow rate, pressures, and compositions denoted by F_i^c, P_i^c, and $x_{i,m}^c$, respectively, where $i=1, 2, \ldots, N_w$.

The outlet streams leaving the PDSDB are next fed to the PDMB. Each stream enters the PDMB via an inlet junction, then passes through a pump followed by a turbine and eventually leaves the PDMB through an outlet junction. Intuitively, at most one of the sequential pump and turbine exists in the solution. While the flow rate and composition of any stream remain unchanged within the PDMB, its pressure may change to become P_n^i after the pump and P_n^o after the energy-recovery device.

Each stream leaving the PDMB passes to an inlet junction $n \in JI=\{n|n=1, N_o\}$ of the ROSDB and, then, splits into N_{RO} "substreams" emanating from that junction to the N_{RO} outlet junctions. Subsequently, one stream leaves each outlet junction $q \in JO=\{q|q=1, N_{RO}\}$ to form a feed stream to the ROSDB. The flow rate, pressure, and composition of a stream leaving the qth outlet junction of the ROSDB are designated as F_q^f, P_q^f, and $x_{q,m}^f$, respectively.

Each feed stream entering the ROMB via an inlet junction is assigned to one of the N_{RO} reverse-osmosis modules in the ROMB. Each stage, q, consists of a number, N_q^S, of parallel and identical RO modules. Module type and size in each stage may differ from those in other stages. Depending upon the optimal solution, each of these RO stages may exist or vanish. In addition, N_q^S is an unknown whose value can be determined via optimization. Subsequently, each feed stream entering the qth RO unit is separated into a permeate stream (whose flow rate, pressure and composition are denoted by F_q^P, P_q^P, and $x_{q,m}^P$, respectively) and a reject stream (whose flow rate, pressure, and composition are designated by F_q^r, P_q^r, and $x_{q,m}^r$, respectively). The permeate as well as the reject streams leaving the ROMB are fed to the PDSDB.

The proposed state-space representation provides a powerful tool for embedding all potential configurations of the RON. Indeed, it can be shown that all the previous works, demonstrated by Figures 2-7, can be readily represented via the proposed formulation (see Figures 9-14). Furthermore, other network configurations which have not been previously considered are also embedded in this representation. This fact becomes particularly important when one recalls the serious limitations associated with the previous research efforts, as discussed in the literature survey. Since the suggested formulation truly accounts for all network configurations, it provides the necessary degrees of freedom for optimally designing the RON.

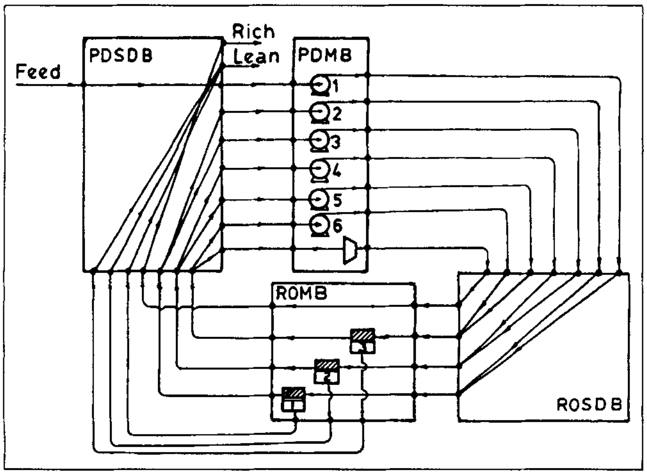

Figure 9. State-space representation of the multistage RO system of Fan et al. (1968).

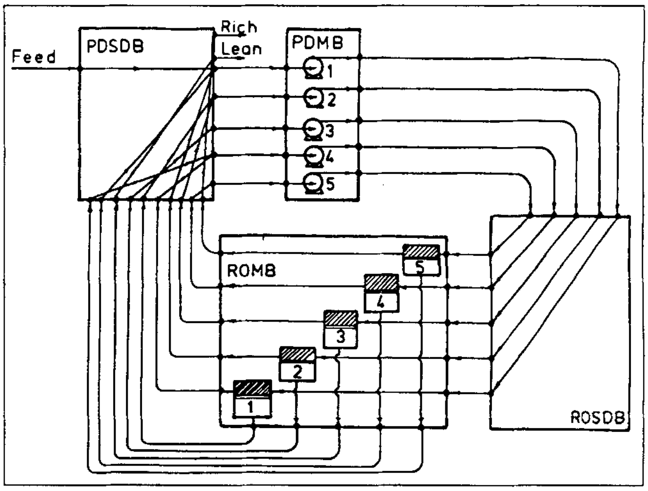

Figure 10. Implementation of Kimura et al.'s RO cascade via a state-space representation.

Mathematical description

In this section, the task of synthesizing a RON will be formulated as an optimization problem. The objective function aims at minimizing the total annualized cost of the RON. The total annualized cost of the RON is the sum of the annualized fixed cost (for all the RO units, pumps, and energy-recovery devices) and the operating cost (cost of power necessary for pumping less the value of power generated by turbines, cost of membrance replacement, and so on). Depending upon the types of RO units, pumps and turbines, one can derive an explicit expression for the objective function in terms of the various flow rates, pressures, and concentrations throughout the RON as well as a number of binary integer variables that indicate the existence or absence of RO modules, pumps, and turbines.

The constraints account for the various environmental and technical aspects of the problem as follows:

Material balances for the inlet junctions of the PDSDB:

$$F_i^w = \sum_{n=1}^{N_o+2N_w} F_{i,n}^w \quad i = 1, 2, \ldots, N_w$$

$$F_q^P = \sum_{n=1}^{N_o+2N_w} F_{q,n}^P \quad q = 1, 2, \ldots, N_{RO}$$

$$F_q^r = \sum_{n=1}^{N_o+2N_w} F_{q,n}^r \quad q = 1, 2, \ldots, N_{RO}$$

Material balances for the outlet junctions of the PDSBD:

$$F_n = \sum_{i=1}^{N_w} F_{i,n}^w + \sum_{q=1}^{N_{RO}} F_{q,n}^P + \sum_{q=1}^{N_{RO}} F_{q,n}^r \qquad n = 1, 2, \ldots, N_o + 2N_w$$

$$F_n x_{n,m} = \sum_{i=1}^{N_w} F_{i,n}^w x_{i,m}^w + \sum_{q=1}^{N_{RO}} F_{q,n}^P x_{q,m}^P + \sum_{q=1}^{N_{RO}} F_{q,n}^r x_{q,m}^r \quad n = 1, 2, \ldots, N_o + 2N_w$$

$$m = 1, 2, \ldots, N_s$$

where, according to the nomenclature used in the network representation

$$F_i^l = F_{N+i} \qquad i = 1, 2, \ldots N_w$$

$$x_{i,m}^l = x_{N_o+i,m} \qquad i = 1, 2, \ldots, N_w \qquad m = 1, 2, \ldots, N_s$$

$$x_{i,m} = x_{N_o+N_w+i,m} \qquad i = 1, 2, \ldots, N_w \qquad m = 1, 2, \ldots, N_s$$

$$F_i^c = F_{N_o+N_w+i} \qquad i = 1, 2, \ldots, N_w$$

Product-demand constraints:

$$F_i^l \ge F_i^{l,\min} \quad i = 1, 2, \ldots, N_w$$

Environmental constraints:

$$x_{i,m}^l \le x_{i,m}^e \quad i = 1, 2, \ldots, N_w \qquad m = 1, 2, \ldots, N_s$$

Since only streams with equal pressures can be mixed together, the following constraints are necessary:

$$(P_n - P_i) F_{i,n}^w = 0 \quad i = 1, 2, \ldots, N_w \qquad n = 1, 2, \ldots, N_o + 2N_w$$

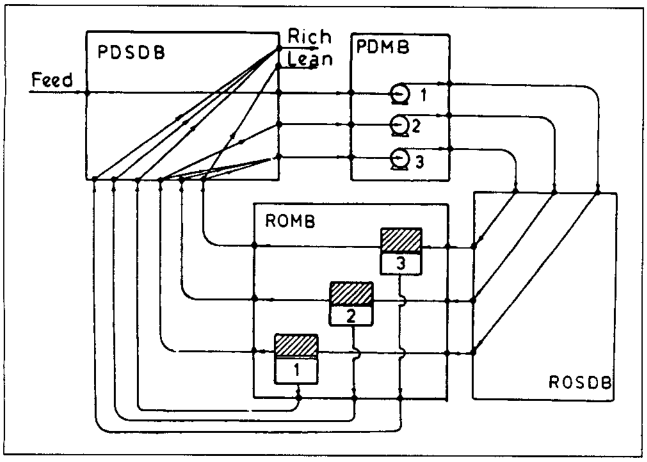

Figure 11. Realization of the three-stage RO system of Bansal and Wiley via the state-space representation.

$$(P_n - P_q^P)F_{q,n}^P = 0 \quad q = 1, 2, \ldots, N_{RO}$$
$$n = 1, 2, \ldots, N_o + 2N_w$$

and

$$(P_n - P_q^r)F_{q,n} = 0 \quad q = 1, 2, \ldots, N_{RO}$$
$$n = 1, 2, \ldots, N_o + 2N_w$$

Constraints for the PDMB

Since a pump will only raise the pressure of a stream while a turbine can only reduce the pressure of a stream, the following constraints are necessary:

$$P_n^i - P_n \geq 0 \quad n = 1, 2, \ldots, N_o$$

and

$$P_n^i - P_n^o \geq 0 \quad n = 1, 2, \ldots, N_o$$

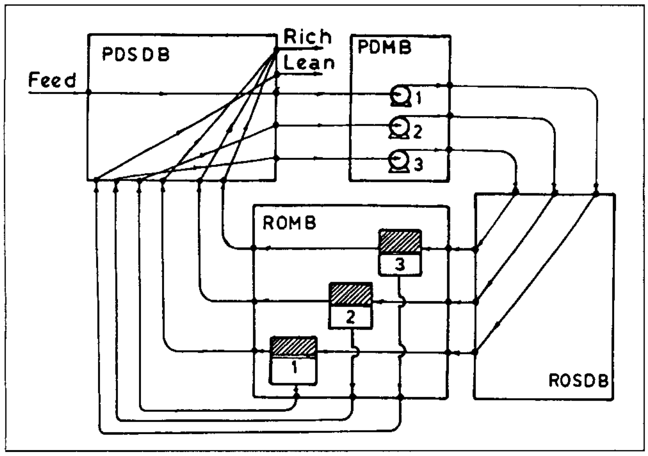

Figure 12. State-space representation of the permeate-staging system.

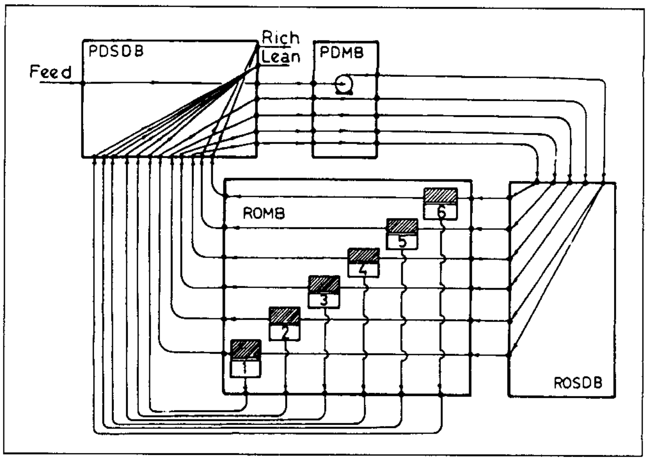

Figure 13. State-space representation of a straight-through flow arrangement.

Now, let us employ the binary integer variables B_n and T_n to account for the existence of the nth booster pump and turbine, respectively. When the nth pump (turbine) exists, $B_n(T_n)$ assumes the value of one, whereas if it is absent, the binary variable becomes zero. Therefore, the following constraints identifying the values of the binary variables are necessary:

$$U * B_n \geq P_n^i - P_n \geq L * B_n \quad n = 1, 2, \ldots, N_o$$

where U and L are arbitrary large and small numbers, respectively. The foregoing constraint forces B_n to become one if P_n^i is larger than P_n (that is, the stream entering the nth junction of the PDMB is pressurized), otherwise it becomes zero. A similar constraint can be written for the turbines:

$$U * T_n \geq P_n^i - P_n^o \geq L * T_n \quad n = 1, 2, \ldots, N_o$$

Since, it is illogical to pressurize a stream and immediately depressurize it, the following constraint is needed:

$$B_n + T_n \leq 1 \quad n = 1, 2, \ldots, N_o$$

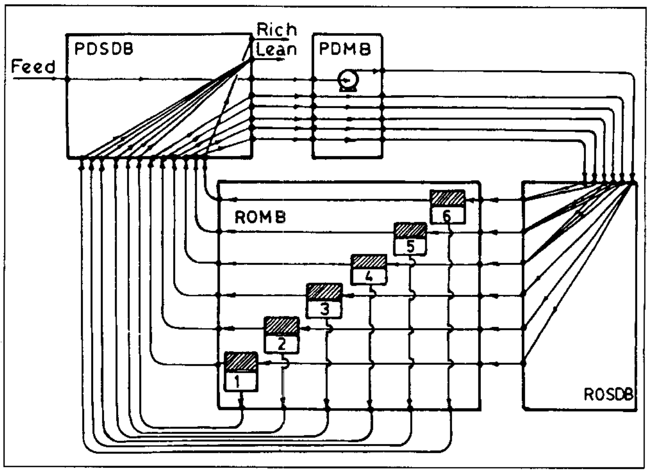

Figure 14. State-space realization of a tapered flow scheme.

Table 1. Geometrical Data of the RO Modules Employed in Example Problems (Evangelista, 1985)*

Module / Property	B-10 (Desalination Example)	B-9 (Dephenolization Example)
Fiber length, l, m	0.750	0.750
Fiber seal length, l_s, m	0.075	0.075
Outer radius of fiber, r_o, m	50×10^{-6}	42×10^{-6}
Inner radius of fiber, r_i, m	21×10^{-6}	21×10^{-6}
Membrane area, S_m, m^2	152.00	180.00

*Courtesy of the American Chemical Society.

Constraints for the ROSDB

Material balances for the inlet junctions of the ROSDB:

$$F_n = \sum_{1=1}^{N_{RO}} F_{n,q} \quad n=1, 2, \ldots, N_o$$

Material balances for the outlet junctions of the ROSDB:

$$F_q^f = \sum_{n=1}^{N_o} F_{n,q} \quad q=1, 2, \ldots, N_{RO}$$

$$F_q^f x_{q,m}^f = \sum_{n=1}^{N_o} F_{n,q} x_{n,m} \quad q=1, 2, \ldots, N_{RO} \quad m=1, 2, \ldots, N_s$$

Isobaric mixing of streams:

$$(P_q^f - P_n^o)F_{n,q} = 0 \quad q=1, 2, \ldots, N_{RO} \quad n=1, 2, \ldots, N_o$$

Constraints for the ROMB

Stage-design equations:

It is essential to have a set of equations relating the flow rates and composition of the reject and the permeate leaving a stage to the flow rate, pressure and concentration of the stream entering that stage as well as the pressures of the permeate and the reject leaving that stage. In general, these equations can be expressed as:

$$F_q^P = \phi_1(F_q^f, x_{q,m}^f, P_q^f, P_q^r, P_q^P, N_q^S) \quad q=1, 2, \ldots, N_{RO}$$

$$F_q^r = \phi_2(F_q^f, x_{q,m}^f, P_q^f, P_q^r, P_q^P, N_q^S) \quad q=1, 2, \ldots, N_{RO}$$

$$x_{q,m}^P = \phi_3(F_q^f, x_{q,m}^f, P_q^f, P_q^r, P_q^P, N_q^S) \quad q=1, 2, \ldots, N_{RO}$$

and

$$x_{q,m}^r = \phi_4(F_q^f, x_{q,m}^f, P_q^f, P_q^r, P_q^P, N_q^S) \quad q=1, 2, \ldots, N_{RO}$$

where ϕ_1, ϕ_2, ϕ_3, and ϕ_4 are the functional relations constituting the model equations for a single RO stage.

Operational constraints

Typically, manufacturers of RO modules recommend operating ranges for pressure, flow rate and concentration. These can be expressed as:

$$F^{\min} \le F_q^f / N_q^S \le F^{\max} \quad q=1, 2, \ldots, N_{RO}$$

$$P_q^f \le P^{\max} \quad q=1, 2, \ldots, N_{RO}$$

$$x_{q,m}^r \le x_m^S \quad m=1, 2, \ldots, N_s$$

(for example, physical constraints to prevent solute precipitation) $\quad q=1, 2, \ldots, N_{RO}$

The following constraint will be employed to identify the existence or absence of a certain RO unit:

$$U * R_q \ge F_q^P \ge L * R_q \quad q=1, 2, \ldots, N_{RO}$$

where R_q is a binary integer variable that assumes the value of one when the qth RO stage exists and vanishes when this stage is absent.

The above mathematical formulation is a mixed-integer nonlinear optimization program (MINLP). Its solution requires the identification of the optimal values of continuous as well as discrete variables. The solution of this MINLP identifies the network configuration, stream distribution and operating conditions.

Having outlined the mathematical description of the problem, it is now appropriate to demonstrate the usefulness of the proposed synthesis procedure by applying it to two example problems. As has been previously mentioned, detailed design procedures of RO systems in the literature are confined to single feed, single component systems. By far, the best studied system is that of water desalination. Although the primary objective of this article is to address waste-reduction situations, the applicability of the proposed procedure extends beyond waste minimization. It is, therefore, useful to compare the approach developed in this work with current practices for the well-known water-desalination system. Next, a second example is tackled with the objective of separating two chlorophenolic compounds from two waste streams. In the sequel, both examples problems are presented.

Illustrative Example

Seawater desalination

This example (Evangelista, 1985) deals with the desalination of seawater using DuPont B-10 hollow-fiber reverse-osmosis modules. Geometrical properties of these modules are given in Table 1. The input data of this case study are summarized in Table 2. In tackling this problem, Evangelista (1985) employed the following module modeling equations:

$$F_q^P / N_q^S = A S_m (\Delta P_q - \pi_q)\gamma \quad (3)$$

where

Table 2. Input Data for the Seawater Desalination Example (Evangelista, 1985)*

Feed flow rate, F_1^w, kg/s	19.290
Feed composition, $x^w_{1,1}$	0.03480
Minimum acceptable product (permeate) flow rate, $F_1^{p,min}$, kg/s	5.787
Maximum allowable product (permeate) composition, $x^e_{1,1}$	0.00057
Maximum flow rate per module, kg/s	0.270
Minimum flow rate per module, kg/s	0.210
Maximum feed pressure, N/m²	68.88×10^5
Pressure drop per module, N/m²	0.22×10^5
Pure water permeability, A, kg/s·N	1.20×10^{-10}
Solute transport parameter, K_1, kg/m²·s	4.00×10^{-6}

*Courtesy of the American Chemical Society

$$\gamma=\frac{\eta}{1+16A\mu r_o l l_s \eta / r_i^4} \tag{4}$$

and

$$\eta=\frac{\tanh[(16A\mu r_o/r_i^2)^{1/2}(l/r_i)]}{[(16A\mu r_o/r_i^2)^{1/2}(l/r_i)]} \tag{5}$$

Also

$$x^P_{q,m}=\frac{K_m x_{q,m}}{A(\Delta P_q-\pi_q)\gamma} \tag{6}$$

By applying an explicit design methodology to the abovementioned seawater desalination problem, Evangelista (1985) developed a tapered RO system. This configuration involves one pump and two RO stages (131 modules) with reject processing. The basic features of this design are shown in Figure 15. The data shown on each stream represent mass flow rate, mass fraction of salt and pressure, respectively. Employing the economic data outlined in the Appendix, one can compute the total annualized cost of Evangelista's solution as $280,503 per year.

It is now appropriate to apply the RON synthesis procedure to the desalination case study. To unify the basis of comparison, Eqs. 3-6 are employed in the ROMB constraints. It is worth pointing out that more realistic models for describing the performance of hollow-fiber RO modules have been developed recently (for example, El-Halwagi et al., 1991). The previously described MINLP is formulated and solved using the software GINO (Liebman et al., 1985; Nabar and Schrage, 1990). This package is based upon the "generalized reduced gradient method" and does not necessarily guarantee the identification of the global solution. The optimization program was executed on a Sun Sparcstation II in 117 CPU seconds. The solution of this program is shown in Figure 16. It features one pump, two RO stages (106 modules) and two energy-recovery turbines. The total annualized cost of this network is $237,990 per year which is 15% less expensive than Evangelista's solution. This result is further appreciated when recalling that the design techniques of RO desalination plants

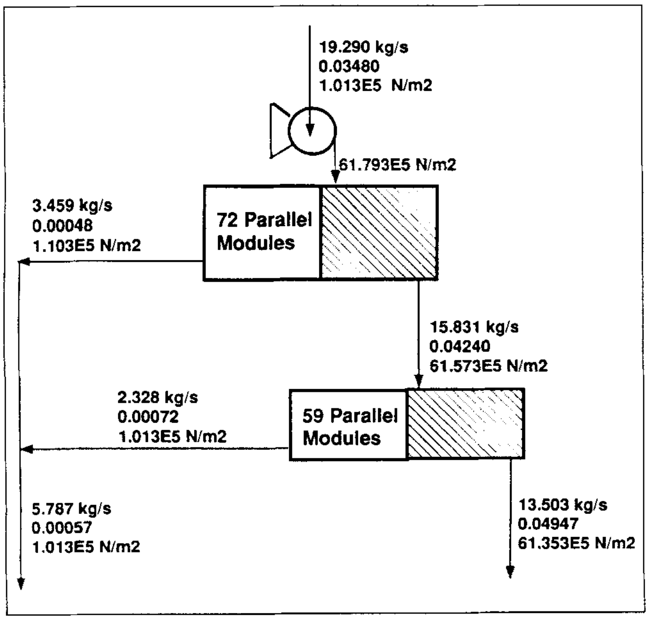

Figure 15. Tapered design for the seawater desalination example (Evangelista, 1985).

Courtesy of the American Chemical Society.

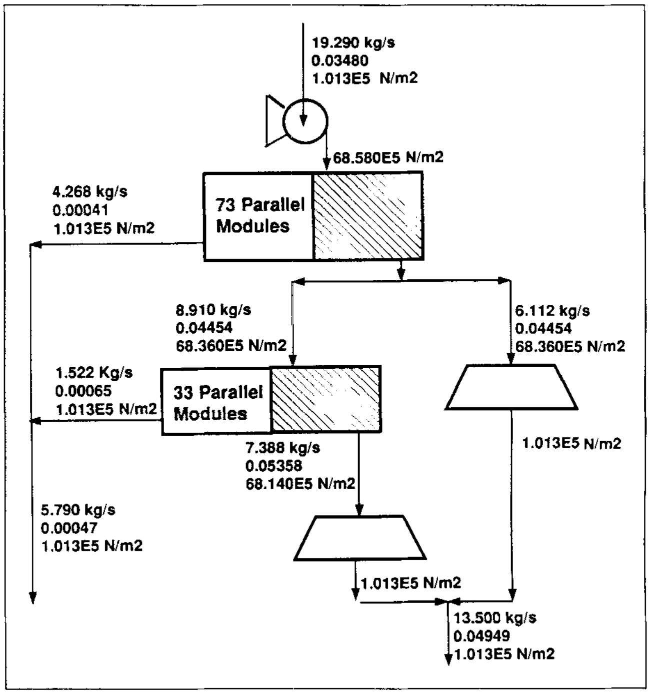

Figure 16. Optimal RON for the seawater desalination example.

are reported to be the most effective among all other membrane-separation applications.

Having addressed a traditional RO system, it is now appropriate to proceed to a novel application in the area of waste reduction; namely the dephenolization of pulp-and-paper aqueous wastes. Basic data of the following example are obtained from US EPA (1990), Austin (1984), and Smook (1982).

Dephenolization of Aqueous Wastes of a Pulp-Bleaching Plant

The production of pulp is a major step in the paper industry. About 80% of the wood pulp in the United States is produced via the Kraft, or sulfate, chemical pulping process. In this process as shown in Figure 17 wood chips are mixed with white liquor (NaOH, Na_2S, NA_2CO_3 and water) in digestor whereby the wood is cooked to release cellulose and hemicellulose from lignin. The solution leaving the digester, called weak black liquor, is concentrated in multiple-effect evaporators to yield strong black liquor. This strong liquor is burned in a recovery furnace to produce molten chemicals than can be dissolved in water given a "green liquor" (NaOH, Na_2S, NA_2CO_3 and water). The green liquor is reacted with lime to regenerate the white liquor which is recycled back to the digestor.

The brightness of the pulp leaving the digestor can be improved through bleaching. This is commonly accomplished in a stepwise sequence utilizing different chemicals in each stage with intermediate washing carried out between stages. One way of carrying out bleaching is by first chlorinating the pulp. Chlorine reacts with lignin which is primarily responsible for the color of the pulp. The chlorinated pulp is then washed with water and filtered. The wastewater leaving this stage contains mono-chloro phenol (MCP) and tri-chloro phenol (TCP) as the two principal hazardous species. To avoid the serious health and environmental consequences of these two compounds, the wash water leaving the chlorination stage should be treated for the removal of MCP and TCP prior to environmental discharge. The chlorinated pulp is then fed to a caustic-extraction stage to remove colored and degraded products and prepare the fibers to more effective treatment in subsequent oxidative stage. Following extraction, the pulp is washed and filtered. Again the wash water leaving the extraction stage should be treated for the removal of hazardous chlorophenols (primarily MCP and TCP).

The waste-reduction task is to synthesize a RON which can

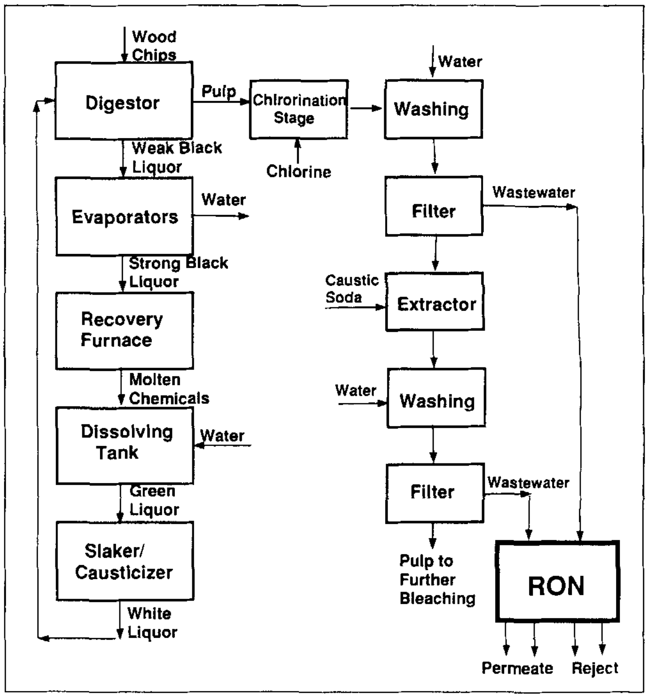

Figure 17. Simplified flowsheet of the kraft pulping process.

Table 3. Input Data for the Dephenolization Example

Feed flow rate of first waste stream, F_1^w, kg/s	6.00
Feed flow rate of second waste stream, F_2^w, kg/s	25.00
Feed composition of MCP in first waste stream, $x^w_{1,1}$	26.00×10^{-6}
Feed composition of TCP in first waste stream, $x^w_{1,2}$	3.00×10^{-6}
Feed composition of MCP in second waste stream, $x^w_{2,1}$	12.00×10^{-6}
Feed composition of TCP in second waste stream, $x^w_{2,2}$	4.00×10^{-6}
Minimum acceptable flow rate of product (permeate) for the first waste stream, $F_1^{l,min}$, kg/s	4.500
Minimum acceptable flow rate of product (permeate) for the second waste stream, $F_2^{l,min}$, kg/s	9.000
Maximum allowable composition of MCP in the product (permeate) of the first waste stream, $x^e_{1,1}$	8.8×10^{-6}
Maximum allowable composition of TCP in the product (permeate) of the first waste stream, $x^e_{2,1}$	1.4×10^{-6}
Maximum allowable composition of MCP in the product (permeate) of the second waste stream, $x^e_{2,1}$	8.8×10^{-6}
Maximum allowable composition of TPC in the product (permeate) of the second waste stream, $x^e_{2,2}$	1.4×10^{-6}
Maximum flow rate per module, kg/s	0.460
Minimum flow rate per module, kg/s	0.210
Maximum feed pressure, N/m^2	25.58×10^5
Pressure drop per module, N/m^2	0.405×10^5
Pure-water permeability, A, kg/s·N	1.20×10^{-10}
MCP solute transport parameter, K_1, $kg/m^2\cdot S$	2.43×10^{-4}
TCP solute transport parameter, K_2, $kg/m^2\cdot s$	2.78×10^{-4}

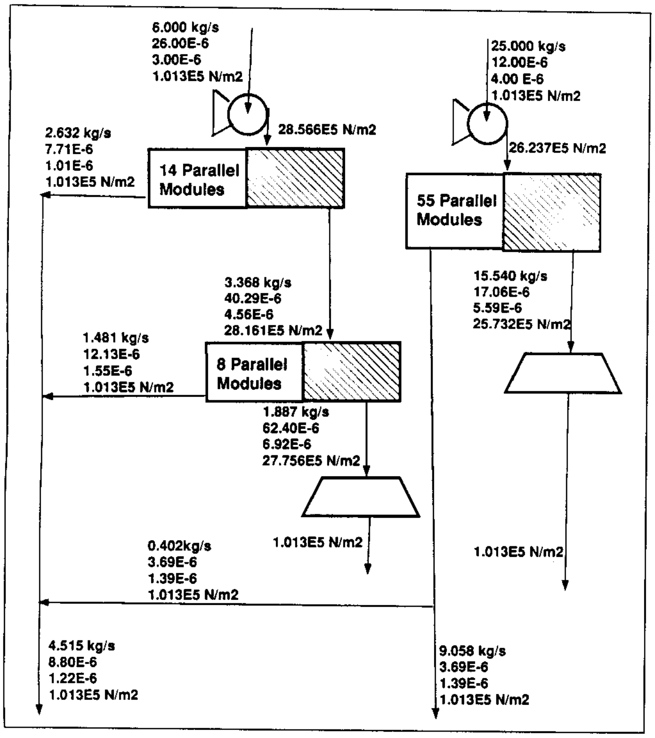

Figure 18. Optimal RON for the dephenolization problem.

separate each of the two waste streams into a lean (permeate) stream which meets environmental regulations set for the control of MCP and TCP discharge and a concentrated rich (reject) stream at minimum total annualized cost. The use of Du-Pont B-9 RO modules is considered (see Table 1 for geometrical properties of modules). The necessary input data for this case study are summarized in Table 3. The basic economic data employed in the objective function of the MINLP are given in the Appendix and the module modeling Eqs. 3-6.

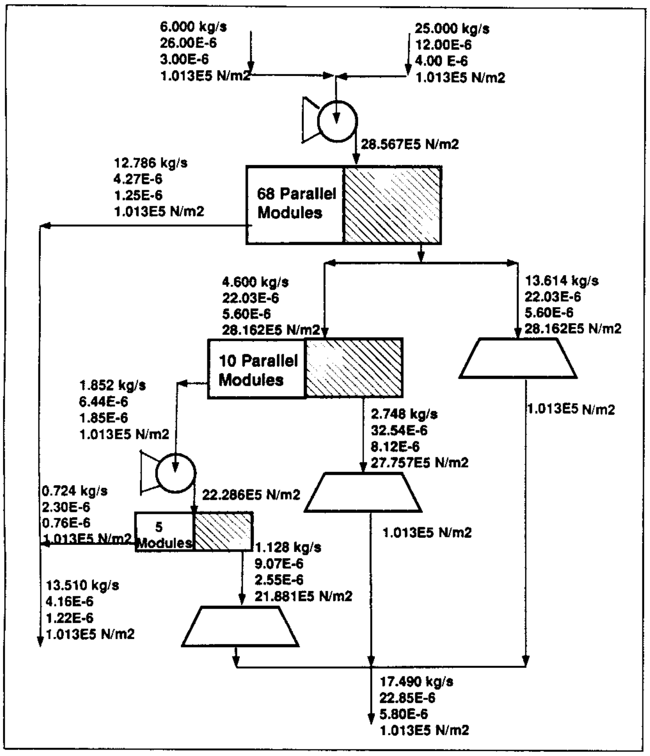

Figure 19. Optimal RON for the dephenolization problem with mixed feed of waste streams.

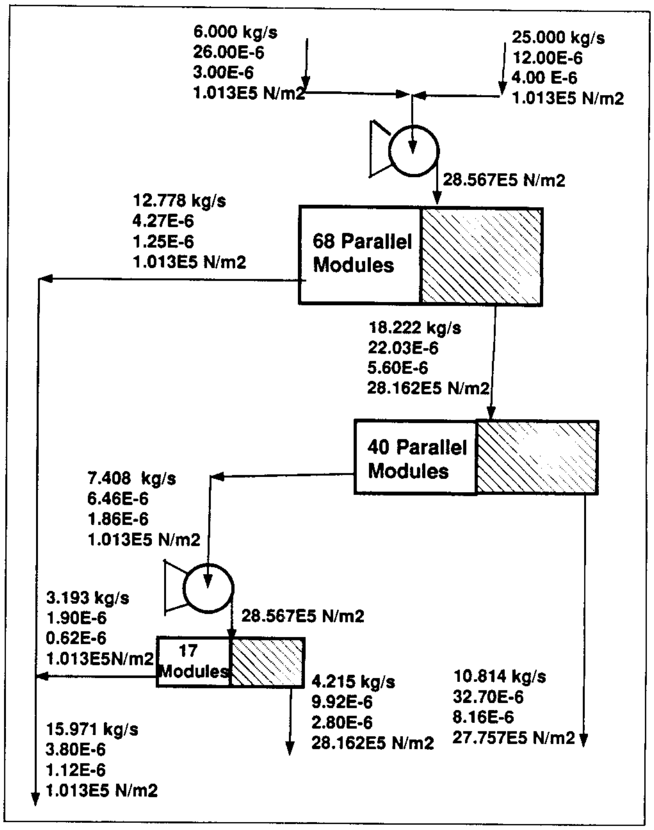

Figure 20. Tapered design for the dephenolization problem with mixed feed of waste streams (without energy-recovery).

Using the RON synthesis procedure, the problem is formulated as an MINLP. The solution, which was obtained by using the software GINO, is shown in Figure 18. Computing time is 237 CPU seconds on a Sun Sparcstation II. The data shown on each stream represent mass flow rate, mass fraction of MCP, mass fraction of TCP and pressure, respectively. It requires three RO stages (77 modules), two pumps and two turbines. The total annualized cost of this scheme is \$193,420 per year.

It is interesting to compare the obtained solution with alternate configurations. If one decides to follow the common industrial practice of mixing waste streams, then a single-feed RON problem can be formulated. The resulting MINLP is solved (computing time is 218 CPU seconds using GINO on a Sun Sparcstation II) to yield the solution demonstrated in Figure 19. This entails the use of three RO stages (83 modules), two pumps, and three turbines. The corresponding total annualized cost is \$210,154 per year. This result suggests that the common industrial approach of mixing waste streams prior to treatment is not always proper. Next, following the principles of the design procedure of Evangelista (1989), a tapered RO system with reject and permeate staging is designed to tackle

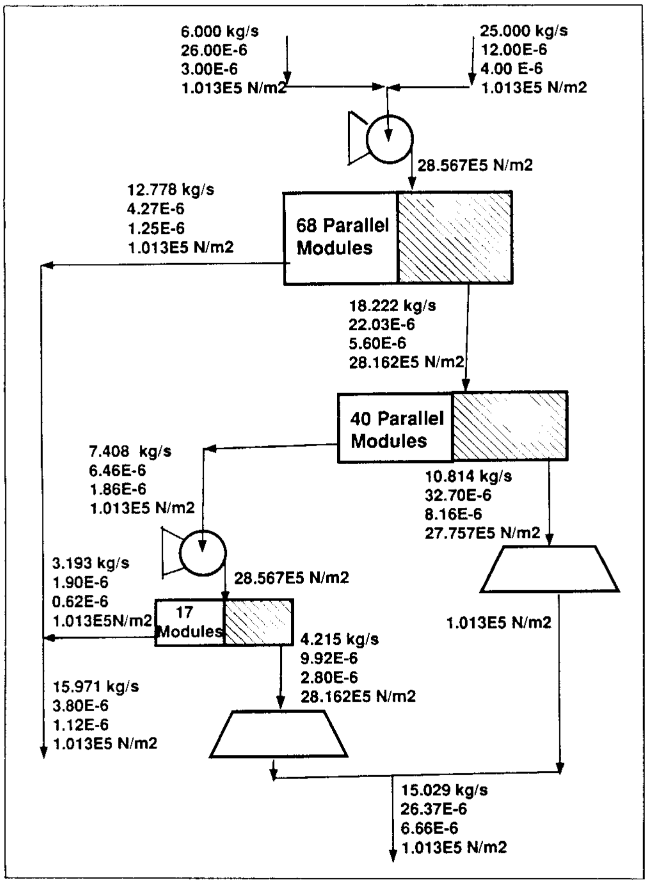

Figure 21. Tapered design for the dephenolization problem with mixed feed of waste streams (with energy-recovery).

the mixed-feed dephenolization problem. The result is shown in Figure 20. It features three RO stages (125 stages) and a booster pump. The corresponding total annualized cost is $304,377 per year which is 57% more expensive than the cost of the optimal RON. If one decides to supplement the foregoing tapered design with energy-recovery turbines (Figure 21), the total annualized cost of the system becomes $292,800 per year which is 51% more expensive than the cost of the optimal RON.

Conclusions

The new problem of synthesizing RON's has been introduced. A structural representation which embeds all potential configurations of interest has been presented. Then, the synthesis task was formulated as a MINLP which seeks to minimize the total annualized cost of the network while complying with all environmental and technical considerations. Comparisons with current industrial practices for two case studies indicate that significant cost savings can be accomplished by adopting the devised RON synthesis procedure.

Notation

A = pure-water permeability, kg/s·N
F_i^c = flow rate of concentrated rich (retenate) stream i leaving the RON, kg/s
F_i^l = flow rate of lean (product) stream i leaving the RON, kg/s
$F_i^{l,min}$ = minimum allowable flow rate of lean (product) stream i leaving the RON, kg/s
F^{max} = maximum flow rate per module (as recommended by manufacturer), kg/s
F^{min} = minimum flow rate per module (as recommended by manufacturer), kg/s
F_i^w = flow rate of waste feed stream i entering the RON, kg/s
$F_{i,n}^w$ = flow rate of substream connecting inlet junction i and outlet junction j in the PDSDB, kg/s
F_n = flow rate of stream leaving the PDSDB via outlet junction n, kg/s
$F_{n,q}$ = flow rate of substream connecting inlet junction n with outlet junction q in the ROSDB, kg/s
F_q^f = flow rate of stream leaving the ROSDB through outlet junction q and entering the qth RO stage in the ROMB, kg/s
F_q^P = flow rate of the permeate leaving the qth RO stage in the ROMB, kg/s
$F_{q,n}^P$ = flow rate of substream connecting the qth inlet junction of a permeate stream with the nth outlet junction in the PDSDB, kg/s
F_q^r = flow rate of the reject leaving the qth RO stage in the ROMB, kg/s
$F_{q,n}^r$ = flow rate of substream connecting the qth inlet junction of a reject stream with the nth outlet junction in the PDSDB, kg/s
i = index for the waste feed streams or their corresponding inlet junctions to be PDSDB
JI = set of inlet junctions to the ROSDB
JO = set of outlet junctions from the ROSDB
$J1$ = set of inlet junctions of the waste feed streams to the PDSDB
$J2$ = set of inlet junctions of the permeate streams to the PDSDB
$J3$ = set of inlet junctions of the reject streams to the PDSDB
$J4$ = set of outlet junctions from the PDSDB
K_m = transport parameter of solute m, m/s
l = fiber length, m
l_s = fiber seal length, m
L = arbitrary small number
n = index for the pumps/turbines in the PDMB or their corresponding outlet junctions from the PDSDB and inlet junctions to the ROSDB
N_o = number of pumps/turbines in the PDMB or their corresponding outlet junctions from the PDSDB and the inlet junctions to the ROSDB
N_q^S = number of parallel RO modules in the qth stage of the ROMB
N_{RO} = number of RO units in the ROMB or their corresponding outlet ROSDB junctions, inlet ROMB junctions, outlet ROMB permeate junctions, outlet ROMB reject junctions, inlet PDSDB permeate junctions or inlet PDSDB reject junctions
N_s = number of undesirable compounds to be separated from the waste streams
N_w = number of waste feed streams or their corresponding inlet PDSDB junctions, outlet PDSDB lean (product) junctions or outlet PDSDB rich (retenate) junctions
ΔP_q = pressure difference across the membrane of any module in the qth stage in the ROMB
P_i^c = pressure of concentrated rich (retenate) stream i leaving the RON, N/m^2
P_i^w = pressure of waste feed stream i entering the RON, N/m^2
P_n = pressure of stream leaving the PDSDB via outlet junction n, N/m^2
P_q^f = pressure of stream leaving the ROSDB through outlet junction q and entering the qth RO unit in the ROMB, N/m^2
P^{max} = maximum allowable pressure for RO units (as recommended by module manufacturer), N/m^2
P_q^P = pressure of the permeate leaving the qth RO unit in the ROMB, N/m^2
P_q^r = pressure of the reject leaving the qth RO stage in the ROMB, N/m^2
q = index for the RO stages in the ROMB or their corresponding outlet ROSDB junctions, inlet ROMB junctions, outlet ROMB permeate junctions, outlet ROMB reject junctions, inlet PDSDB permeate junctions or inlet PDSDB reject junctions

r_i = inner radius of the hollow fiber, m
r_o = outer radius of the hollow fiber, m
R_q = binary integer variable that corresponds the qth RO stage in the ROMB
S = set of hazardous species
S_m = membrane area per module, m^2
T_n = binary integer variable to indicate the existence or absence of the nth turbine in the PDMB
U = arbitrary large number
W = set of waste feed streams
$x^c_{i,m}$ = mass fraction of component m in concentrated rich (retenate) stream i leaving the RON
$x^e_{i,m}$ = maximum allowable mass fraction of species m in lean (product) stream i leaving the RON, as imposed by environmental-protection regulations
$x^l_{i,m}$ = mass fraction of component m in lean (product) stream i leaving the RON
$x^w_{i,m}$ = mass fraction of undesirable species m in waste feed stream entering the RON
X^w_i = set of inlet composition of waste feed stream i
$x_{n,m}$ = mass fraction of compound m in the stream that is leaving the PDSDB via outlet junction n
$x^f_{q,m}$ = mass fraction of component m in the stream that is leaving the ROSDB through outlet junction q and entering the qth RO stage in the ROMB
$x^P_{q,m}$ = mass fraction of component m in the permeate leaving the qth RO stage in the ROMB
$x_{q,m}$ = average mass fraction of species m in the high-pressure side of any module in the qth stage in the ROMB
$x^r_{q,m}$ = mass fraction of species m in the reject leaving the qth RO stage in the ROMB
x^s_m = maximum allowable operating composition of component in any RO unit

Greek letters

γ = defined by Eq. 4
η = defined by Eq. 5
μ = viscosity, kg/m·s
π_q = average osmotic pressure on the high-pressure side of the qth RO stage in the ROMB, N/m^2

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Appendix

Summary of Cost Data

Throughout this paper the following economic data are employed:

Annualized fixed cost of pumps (\$/yr) = 0.0157 [flow rate through pump (kg/s)* pressure difference across pump $(N/m^2)]^{0.79}$ (A1)

Annualized fixed cost of turbines (\$/yr) = 0.4182 [flow rate through turbine (kg/s)* pressure difference across turbine $(N/m^2)]^{0.47}$ (A2)

Cost (value) of electric power = 0.06 \$/kWR (A3)

Mechanical efficiency of pumps (or turbines) = 65% (A4)

Annualized cost of RO modules for the seawater desalination example (including annualized installed annualized installed cost, membrane replacement, labor and maintenance) $= 1{,}450 \dfrac{\$}{\text{module}\cdot\text{yr}}$ (A5)

Annualized cost of RO modules for the dephenolization example (including annualized installed cost, membrane replacements, labor and maintenance) $= 1{,}140 \dfrac{\$}{\text{module}\cdot\text{yr}}$ (A6)

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