A Superstructure Optimization Approach for Water Network Synthesis with Membrane Separation-Based Regenerators

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Abstract

This work addresses the problem of water network synthesis. We propose a superstructure with fixed topology for a water network that consists of three layers, similar to a pooling problem: sources for reuse/recycle; regenerators for contaminants removal; and sinks for acceptance of water for reuse/recycle. The superstructure encompasses multiple freshwater sources, membrane separation-based partitioning regenerators of the industrially-favored ultrafiltration and reverse osmosis, and sinks for incineration and deep ocean discharge. A mixed-integer nonlinear program is formulated based on this superstructure to determine the optimal interconnections in terms of total flowrates and contaminant concentrations. The main decisions include determining the split fractions of the source flowrates, extents of regeneration, and mixing ratios of the sources and regenerated streams subject to compliance with the maximum allowable inlet contaminant concentration limits of the sinks and discharge regulations. We also develop linear models for the membrane regenerators that admit a more general expression for the retentate stream concentration based on liquid-phase

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recovery factors and removal ratios. Computational studies are performed using GAMS/BARON on an industrially-significant case study of a petroleum refinery water system. We incorporate linear logical constraints using 0–1 variables that enforce certain design and structural specifications to tighten the model formulation and enhance solution convergence. A globally optimal water network topology is attained that promotes a 27% savings equivalent to about $218,000/year reduction in freshwater use.

Keywords: Optimization; Water reuse; Superstructure; Mixed-integer nonlinear programming (MINLP); Membrane; Pooling problem

1. Introduction

High demand of water consumption in the process industry may result in process plants becoming vulnerable to the global water scarcity challenge. Coupled with the drive for achieving sustainable development, this work is undertaken to address industrial water network synthesis that minimizes costs as well as freshwater use and wastewater generation (Desai and Klanecky, 2011).

Within the realm of the process optimization philosophy for minimizing freshwater use and wastewater generation, *water reuse* and *water recycle* are concerned with channeling the effluent from a water-using operation to other operations, including the operations where it was generated. In further reducing freshwater and wastewater flowrates after exhausting recovery opportunities via direct reuse/recycle, *water regeneration* can be considered, which involves performing partial treatment on the effluent by using water treatment and purification units such as membranes and steam stripping prior to reuse/recycle.

In general, there are two major approaches for addressing the water network synthesis problem, namely insights-based techniques and mathematical optimization-based techniques. The former typically involves water pinch analysis algorithms, which offer low computational burden in generating solutions, yet often at the expense of requiring significant problem simplifications. On the other hand, optimization allows treatment of water network synthesis problems in their full complexity by considering representative cost functions, multiple contaminants, and various topological constraints, but it frequently suffers from the high computational expense required to achieve optimality especially for large-scale problems.

Recent work in this area increasingly have involved the development of optimization models, primarily solved using mathematical programming (Faria and Bagajewicz, 2010a; 2010b; 2009; Tan et al., 2009; Karuppiah and Grossmann, 2008; 2006; Tan et al., 2007b; Bringas et al., 2007; San Roman et al., 2007; Gunaratnam et al., 2005) as well as other approaches such as fuzzy programming (Tan and Cruz, 2004; Aviso et al., 2010a; Aviso et al., 2010b), Monte Carlo simulation (Tan et al., 2007a) and artificial intelligence-based metaheuristic algorithms (Jeżowski et al., 2007; Hul et al., 2007). The optimization-based approach mainly requires the construction of a superstructure network representation of design alternatives that leads to a mixed-integer linear/nonlinear program (MILP/MINLP). Optimization-based techniques for reuse/recycle and regeneration networks have also been developed by incorporating mass-integration strategies (Gabriel and El-Halwagi, 2005; El-Halwagi et al., 1996) and property-integration framework (Napoles-Rivera et al., 2010; Ng et al., 2010; Ponce-Ortega et al., 2010; Ponce-Ortega et al., 2009). They have also been coupled with physical insights derived from water pinch analysis (Alva-Argáez et al., 1998; Alva-Argáez et al., 2007b; 2007a; Alva-Argáez et al., 1999; Ng et al., 2009a; 2009b; Ng et al., 2010; Ng et al., 2009c).

In line with the aforementioned trend, this work is concerned with the superstructure optimization approach for water network synthesis in the process industry. The main contributions of our work are three-fold. First, we consider several extensions in the superstructure development, beginning with the incorporation of multiple freshwater sources. The regeneration subnetwork in the superstructure explicitly caters for partitioning regenerators, particularly membrane separation-based water treatment technologies such as ultrafiltration (UF) and reverse osmosis (RO). The major factor that motivates this emphasis in our work stems from the fact that UF and RO are gaining increasingly widespread practical applications in the process industry for more efficient separation between clean and contaminated water. RO is known to be the separation process with the lowest cost, whilst UF is touted as the technology of choice to assist plants in efficiently utilizing available water supplies (Desai and Klanecky, 2011). Besides the conventional permanent sinks of end-of-pipe treatment and environmental discharge, the superstructure also accounts for other permanent sink features namely a sink for incineration of untreated waste, which does not meet discharge regulations and a deep ocean discharge sink for brine disposal from the reject of an RO.

This source–regenerator–sink superstructure allows interconnections of its elements in many potential alternative configurations for implementing direct water reuse/recycle, regeneration–reuse, and regeneration–recycle. In particular, the regenerated water can be directed from one regenerator to another for multiple treatments using the same or different technology type in meeting the sink requirements for reuse/recycle or environmental limits. It is noteworthy that the resulting model formulation gives rise to a total water network synthesis problem, as defined by Faria and Bagajewicz (2010a), in which the sinks encompass end-of-pipe effluent treatment system (ETS) and discharge to the environment (see Figure 1).

Our second contribution is in the development of a linear model with fixed removal ratios for a membrane regenerator. This model treats the permeator and rejector at the outlet of a regenerator as tasks (instead of states), and we propose a more general expression for the rejector concentration in terms of liquid-phase recovery factor and removal ratio. The last contribution is to incorporate linear logical constraints using 0–1 variables in the model to tighten the formulation and enhance solution convergence to the global optimum by removing undesired solutions early enough during the enumeration procedure in the tool of our choice, GAMS/BARON.

The rest of the paper is organized as follows. Section 2 formally describes the problem addressed in this work. Section 3, which is the main part of this paper, presents the proposed superstructure and MINLP model formulation to handle the problem, with emphasis on the membrane separation-based regenerators. Section 4 details the development of logical constraints that are incorporated in the model as a strategy to speed up its solution convergence. Section 5 reports the computational experiments carried out for implementing the proposed approach on the water systems of an actual operating petroleum refinery. Finally, concluding remarks including future work to be undertaken are provided in Section 6. The Appendix lists the notations used in this work.

2. Problem Statement

Given is the following data for the elements of a water network:

- a set of fixed-flowrate water sources i, $i \in I$, with known (fixed) flowrates $F_{\mathrm{SO}}\left(i\right)$ and concentrations $C_{\mathrm{SO}}\left(i,q\right)$ of the contaminants $q \in Q$ to be removed, that are amenable to reuse/recycle;
- a set of fixed-flowrate water sinks j, $j \in J$, with known (fixed) flowrate requirements $F_{\mathrm{SI}}\left(j\right)$ and maximum allowable inlet concentration limit $C_{\max}\left(j,q\right)$ for the contaminants q;
- a set of water regenerators k, $k \in K$, with fixed removal ratios of targeted contaminants; and
- a set of freshwater sources, with variable flowrate (to be optimized) and known contaminant concentrations that can be purchased to supplement the availability of the water sources i, $i \in I$.

We aim to synthesize an optimal water network configuration in terms of its stream piping interconnections along with the corresponding water flowrates and contaminant concentrations. The optimal system configuration is one that minimizes the cost of freshwater use, the treatment cost of wastewater effluent discharges to the environment, and the capital and operating costs for both the regenerators and the interconnections. Regarding constraints, material balances on flows and concentrations around the sources, regenerators, and sinks are to be obeyed in addition to the sink limits and certain design and structural specifications.

A number of assumptions are made in our proposed model formulation to achieve the stated goal:

- the number of sources and sinks is fixed;
- the flowrates of all the water sources and sinks are fixed (i.e., they are fixed-flowrate type of operations) (Foo, 2009);
- the total flowrate of a source or sink is the same as that of pure water because the contaminant concentrations are at the level of parts per million (ppm) (Karuppiah and Grossmann, 2006; Bagajewicz et al., 2002).

Moreover, it is assumed that the water network operates under constant temperature conditions, therefore heat integration is not considered.

3. Optimization Model Formulation

Water-using operations can be broadly categorized into two model representations, which are mainly distinguished by the involvement of mass transfer (Foo, 2009). The first representation is the fixed contaminant mass load model, or simply *fixed-load model*, that caters for mass transfer-based water-using operations. This model is mainly concerned with the use of water as a lean stream that removes a certain amount of contaminant mass load from a rich stream. Since water is used as a mass separating agent, water losses and gains in such operations are always assumed to be negligible. Thus, a fixed-load unit has specified inlet and outlet flowrates that are equal. Typical examples include gas absorption, scrubbing, solvent extraction, and vessel cleaning.

The second representation is the *fixed-flowrate model* that caters also for non-mass transfer-based water-using operations. It is mainly concerned with the water flowrate requirement of an operation that generates and/or consumes a fixed amount of water. A fixed-flowrate unit has specified inlet and outlet flowrates that may or may not be equal, hence it is able to account for both water losses and gains. Typical examples include reactors with water as either a raw material or a product; cooling towers requiring periodic water makeup; and boilers requiring blowdown that releases water to remove solids and other impurities (Shenoy and Prakash, 2005).

In this work, we adopt a fixed-flowrate model mainly because it offers a more general representation that encompasses both mass transfer- and non-mass transfer-based water-using operations. Moreover, contaminant mass load data, namely the amount of contaminants picked up or transferred to the water streams, are seldom available in operating plants and thus saddled with significant uncertainty. A typical approach is to model fixed-flowrate water-using operations as water sources and/or sinks, which is a representation that we conveniently adopt in developing the superstructure and the resulting model formulation for our problem. It should however, be highlighted that a limitation of such a model is the absence of direct interaction between a sink and a source, particularly if they belong to the same water-using unit, in which case a fixed-load model can be used to investigate potential decrease in the total annualized cost.

3.1. Superstructure Representation of Water Network Systems

We consider a generic superstructure representation as shown in Figure 2 for an industrial water network based on El-Halwagi and Gabriel (2005) and Meyer and Floudas (2006). The superstructure admits a fixed network topology consisting of predetermined numbers of fixed-flowrate sources for water reuse/recycle, regeneration units for contaminants removal from water streams, and fixed-flowrate sinks for acceptance of water for reuse/recycle. It is noteworthy that since water network synthesis is an application of the classical pooling problem (Misener and Floudas, 2009), the three network elements are analogous to the latter as follows: feedstock, pooling tanks for intermediate storage, and final pooling products, respectively. The source–regenerator–sink superstructure allows interconnections of its three elements in all feasible ways to embed numerous potential alternative configurations for implementing direct water reuse/recycle, regeneration–reuse, and regeneration–recycle.

3.1.1. Source Subnetwork

Sources are water supply streams, which contain contaminants targeted for removal, that are amenable to direct reuse/recycle or to regeneration–reuse/recycle. These sources are complemented with multiple external sources of freshwater. The goal of the optimizer is to determine the optimal split fractions of the source flowrates at given contaminant concentrations for subsequent allocation to the regeneration and sink subnetworks.

3.1.2. Regeneration Subnetwork

The regeneration subnetwork considers a set of water treatment technologies to regenerate water streams for reuse/recycle in the sinks or for discharge to environment. In this work, we explicitly handle partitioning regenerators, in particular membrane separation-based water treatment technologies. These technologies include ultrafiltration (UF) and reverse osmosis (RO) that are gaining increasingly widespread practical applications in the process industry. Inlets to a regenerator consist of all sources and regenerated streams from every other regenerator. In this way, the regenerated water can be directed from one regenerator to another for multiple treatments using the same or different technology type in meeting the sink requirements for reuse/recycle or the environmental discharge limits. Within the regeneration network, a regenerator removes a fraction of selected contaminants from the incoming water streams, typically specified as a fixed removal ratio for each contaminant. As a result, a regenerated stream is altered in terms of flowrate and contaminant concentrations, while a stream that is not regenerated remains unchanged. The optimizer seeks to determine the extents of regeneration before the streams are directed to and mixed in the sinks for reuse/recycle. This goal is achieved by ensuring that all regulated contaminant concentrations in each sink do not exceed their maximum permissible levels (Hammer and Hammer, 2008), which in this work is designated as the maximum allowable inlet concentrations (MAIC) limit for a sink. For a discharge sink, the MAIC is equivalent to the effluent standard imposed by the environmental regulations of local authorities (or more precisely, the parameters listed on the individual site discharge permit and its municipality’s general sewer ordinance). If a stream that exits the regeneration network does not obey the MAIC for any part or all of the sinks, the superstructure allows the stream to undergo further treatment operations involving multiple units in series, either in another regenerator of the same or different technology type.

3.1.3. Sink Subnetwork

Sinks are water-using units, equipment, or operations that can accept water streams from sources or regenerators as long as they are compliant with the MAICs of those sinks. Optimization of the sink subnetwork aims to determine the optimal mixing ratios of the sources and regenerated streams for reuse/recycle in the sink operations. The superstructure include the two conventional permanent sinks of: (1) an offsite end-of-pipe effluent treatment system (ETS) plant for waste materials that are not reused/recycled in any of the sinks; and

(2) an outlet for discharge to surface water or groundwater of the environment, which is typically the main waterways of rivers. The superstructure also accounts for the option of bypassing the ETS by sending a stream directly to the discharge without treatment by the ETS plant first.

We consider extending the superstructure to account for other permanent sink features, notably a sink for incineration of contaminants whose fate do not end up being reused/recycled in any of the other sinks and are untreated wastes that do not meet discharge regulations (Metcalf and Eddy, 2004). The incineration sink is assigned with capital and operating costs that are relatively higher than all the other sinks in the objective function formulation in order for it to be selected only as a last resort. Another permanent sink incorporated in the proposed superstructure is ocean discharge. This sink is mainly intended for the deep sea discharge of brine disposal from the reject of an RO (Metcalf and Eddy, 2004).

A regeneration–recycle operation may involve regenerating a stream up to freshwater quality or even drinking water quality as dictated by operational requirements. To capture this aspect, we introduce a permanent sink structure called “city water tank” (Nystuen, 2011), which represents the storage for freshwater as purchased from a water retailer (or wholesaler) and from regeneration–recycle. The MAIC of the city water sink can be stipulated according to the international standard for drinking water limits as imposed by the World Health Organization (WHO, 2008) or the United States of America’s Environmental Protection Agency (EPA).

3.2. Regenerator Models

Conventional models for water treatment units typically entail a representation involving a single inlet flow stream and a single outlet flow stream (Karuppiah and Grossmann, 2006; 2008; Huang et al., 1999). We conveniently adopt such models for the non-membrane regenerators in this work. However such models must be adapted when applied to membrane regenerators, which typically consist of two main outlets: (1) a cleaner lean permeate stream called *permeator*, which is of lower concentration than (2) a dirtier concentrated retentate stream called *rejector*. Although the outlet flow of a regenerator in the conventional model could be easily separated into a permeator and a rejector stream by using a splitter, both streams would then share the same concentration (see Figure 3). Such a model is clearly not suitable for a membrane regenerator because the contaminant concentration in a rejector is relatively higher than a permeator.

A linear model for membrane regenerators has been previously reported in Tan et al. (2009), in which the feed stream is separated into a permeator and a rejector (see Figure 4). In an effort to address the concentration discrepancy issue, an important point of departure in our proposed model is the representation of the permeator and the rejector as tasks (units) instead of states (streams) in a superstructure (see Figure 5). Despite employing a representation that seems to treat the permeator and the rejector as two separate units, it will be evident in the complete network model presented in later sections that we have employed a suitable formulation approach which preserves the permeator and the rejector as two related subentities that make up the physical configuration of the outlets of a membrane regenerator.

The split ratio on the inlet flowrate of a regenerator is applied using the liquid-phase recovery factor α, $0 < \alpha < 1$, which represents a fixed fraction that exits in its permeator:

$$F_{\mathrm{MP}} = \alpha F_{\mathrm{in}}, \tag{1}$$

where F_{MP} and F_{in} are total flowrates of the permeator and the feed stream. The complement of the fraction is discharged as the rejector flowrate:

$$F_{\mathrm{MR}} = \left(1-\alpha\right) F_{\mathrm{in}}, \tag{2}$$

where F_{MR} is the total rejector flowrate of the membrane regenerator k. Another point of departure concerns the use of a more general expression for the rejector concentration, in terms of α and the removal ratio $R(q)$ for a contaminant q. The overall concentration balance of the membrane regenerator is given by:

$$F_{\mathrm{in}} C_{\mathrm{in}}\left(q\right) = F_{\mathrm{MP}} C_{\mathrm{MP}}\left(q\right) + F_{\mathrm{MR}} C_{\mathrm{MR}}\left(q\right), \quad \forall q \in Q \tag{3}$$

where $C_{\mathrm{in}}(q)$, $C_{\mathrm{MP}}(q)$, and $C_{\mathrm{MR}}(q)$ are the concentrations of a contaminant q of the feed stream, the permeator, and the rejector, respectively. Considering the removal ratio:

$$R\left(q\right) = \frac{F_{\mathrm{in}} C_{\mathrm{in}}\left(q\right) - F_{\mathrm{MP}} C_{\mathrm{MP}}\left(q\right)}{F_{\mathrm{in}} C_{\mathrm{in}}\left(q\right)}, \quad \forall k \in K_{\mathrm{M}}, \forall q \in Q, \tag{4}$$

expressions for the permeator and the rejector concentrations are obtained as:

$$C_{\mathrm{MP}}(q) = \frac{1 - R(q)}{\alpha} C_{\mathrm{in}}(q), \quad \forall q \in Q, \tag{5}$$

$$C_{\mathrm{MR}}(q) = \frac{R(q)}{1 - \alpha} C_{\mathrm{in}}(q), \quad \forall q \in Q. \tag{6}$$

Note that it is sufficient to formulate a model that includes the two relations defining α for the permeator and the rejector, since the total mass balance around a regenerator is redundant to them. Alternatively, α could also be defined in terms of the outlet streams since the total balance holds. Recall that both the permeator and the rejector of a regenerator possess the same removal ratio.

Most optimization models that are tailored for membrane separation-based water treatment technology in the literature are nonlinear (Saif et al., 2008b; 2008a; Bringas et al., 2007; San Roman et al., 2007; Galan and Grossmann, 1998). Hence at the outset, these models are relatively more computationally challenging to handle compared to our linear formulation. Nevertheless, the price that we pay in retaining linearity is to compromise in terms of capturing the detailed physics of the water regeneration processes occurring in each regenerator of a specific technology, which the detailed nonlinear models account for at the expense of limitations in handling a large number of regenerators of various technologies that are typical of industrial-scale problems. From this perspective, our formulation offers advantage in synthesizing site-wide or multisite water network models involving multiple water sources and sinks and their interactions with multiple regenerators of different technologies as illustrated in a practical case study in Section 5.

3.3. Optimization-Based Formulation of Water Network Synthesis Model

Based on the superstructure and the proposed regenerator models, an MINLP is formulated by using model structures previously reported in the literature as a basis (Meyer and Floudas, 2006; Gabriel and El-Halwagi, 2005). The aim of the model is to determine the piping interconnections (or pipelines) of an optimal water system configuration with its associated total stream flowrates and contaminant concentrations.

3.3.1. Material Balance Equations

3.3.1.1. Water Balances for Sources

Mass balances for a source:

$$F_{\mathrm{SO}}(i) = \sum_{k \in K_{\mathrm{NM}}} F_{\mathrm{SO,NM}}(i,k) + \sum_{k' \in K_{\mathrm{M}}} \left(F_{\mathrm{SO,MP}}(i,k') + F_{\mathrm{SO,MR}}(i,k') \right) + \sum_{j \in J} F_{\mathrm{SO,SI}}(i,j), \quad \forall i \in I. \tag{7}$$

3.3.1.2. Water Balances for Regenerators

Figure 6 shows a schematic of the regeneration subnetwork consisting of a set of non-membrane regenerators K_{NM} and a set of membrane regenerators K_{M}, in which the latter are each composed of a permeator MP and a rejector MR.

(a) Water Balances for Non-Membrane Regenerators

Mass balances for a non-membrane regenerator:

$$\begin{aligned}&\sum_{i\in I}F_{\mathrm{SO,NM}}(i,k)+\sum_{\substack{k'\in K_{\mathrm{NM}}\\k'\neq k}}F_{\mathrm{NM,NM}}(k',k)+\sum_{k''\in K_{\mathrm{M}}}\left(F_{\mathrm{MP,NM}}(k'',k)+F_{\mathrm{MR,NM}}(k'',k)\right)\\&=\sum_{j\in J}F_{\mathrm{NM,SI}}(k,j)+\sum_{\substack{k'\in K_{\mathrm{NM}}\\k'\neq k}}F_{\mathrm{NM,NM}}(k,k')+\sum_{k''\in K_{\mathrm{M}}}\left(F_{\mathrm{NM,MP}}(k,k'')+F_{\mathrm{NM,MR}}(k,k'')\right),\quad\forall k\in K_{\mathrm{NM}}.\end{aligned}\tag{8}$$

Concentration balances for a non-membrane regenerator:

$$\begin{aligned}&\left(1-R(k,q)\right)\left(\begin{aligned}&\sum_{i\in I}F_{\mathrm{SO,NM}}(i,k)C_{\mathrm{SO}}(i,q)+\sum_{\substack{k'\in K_{\mathrm{NM}}\\k'\neq k}}F_{\mathrm{NM,NM}}(k',k)C_{\mathrm{NM}}(k',q)\\&+\sum_{k''\in K_{\mathrm{M}}}\left(F_{\mathrm{MP,NM}}(k'',k)C_{\mathrm{MP}}(k'',q)+F_{\mathrm{MR,NM}}(k'',k)C_{\mathrm{MR}}(k'',q)\right)\end{aligned}\right)\\&=C_{\mathrm{NM}}(k,q)\left(\begin{aligned}&\sum_{j\in J}F_{\mathrm{NM,SI}}(k,j)+\sum_{\substack{k'\in K_{\mathrm{NM}}\\k'\neq k}}F_{\mathrm{NM,NM}}(k,k')\\&+\sum_{k''\in K_{\mathrm{M}}}\left(F_{\mathrm{NM,MP}}(k,k'')+F_{\mathrm{NM,MR}}(k,k'')\right)\end{aligned}\right),\quad\forall k\in K_{\mathrm{NM}},\forall q\in Q.\end{aligned}\tag{9}$$

(b) Water Balances for Membrane Regenerators

Mass balances for the permeator of a membrane regenerator:

$$\begin{aligned}&\sum_{i\in I}F_{\mathrm{SO,MP}}(i,k)+\sum_{k'\in K_{\mathrm{NM}}}F_{\mathrm{NM,MP}}(k',k)+\sum_{\substack{k''\in K_{\mathrm{M}}\\k''\neq k}}\left(F_{\mathrm{MP,MP}}(k'',k)+F_{\mathrm{MR,MP}}(k'',k)\right)\\&=\sum_{j\in J}F_{\mathrm{MP,SI}}(k,j)+\sum_{k'\in K_{\mathrm{NM}}}F_{\mathrm{MP,NM}}(k,k')+\sum_{\substack{k''\in K_{\mathrm{M}}\\k''\neq k}}\left(F_{\mathrm{MP,MP}}(k,k'')+F_{\mathrm{MP,MR}}(k,k'')\right),\quad\forall k\in K_{\mathrm{M}}.\end{aligned}\tag{10}$$

Mass balances for the rejector of a membrane regenerator:

$$\sum_{i\in I} F_{\text{SO,MR}}(i,k) + \sum_{k'\in K_{\text{NM}}} F_{\text{NM,MR}}(k',k) + \sum_{\substack{k''\in K_{\text{M}}\\ k''\neq k}} \left(F_{\text{MP,MR}}(k'',k) + F_{\text{MR,MR}}(k'',k)\right)$$
$$= \sum_{j\in J} F_{\text{MR,SI}}(k,j) + \sum_{k'\in K_{\text{NM}}} F_{\text{MR,NM}}(k,k') + \sum_{\substack{k''\in K_{\text{M}}\\ k''\neq k}} \left(F_{\text{MR,MP}}(k,k'') + F_{\text{MR,MR}}(k,k'')\right), \quad \forall k\in K_{\text{M}}. \tag{11}$$

Split ratio on flow based on the liquid-phase recovery for a permeator:

$$\alpha(k)\begin{pmatrix} \sum_{i\in I}\left(F_{\text{SO,MP}}(i,k) + F_{\text{SO,MR}}(i,k)\right) + \sum_{k'\in K_{\text{NM}}} \left(F_{\text{NM,MP}}(k',k) + F_{\text{NM,MR}}(k',k)\right) \\ + \sum_{\substack{k''\in K_{\text{M}}\\ k''\neq k}} \left(F_{\text{MP,MP}}(k'',k) + F_{\text{MP,MR}}(k'',k) + F_{\text{MR,MP}}(k'',k) + F_{\text{MR,MR}}(k'',k)\right) \end{pmatrix}$$
$$= \sum_{j\in J} F_{\text{MP,SI}}(k,j) + \sum_{k'\in K_{\text{NM}}} F_{\text{MP,NM}}(k,k') + \sum_{\substack{k''\in K_{\text{M}}\\ k''\neq k}} \left(F_{\text{MP,MP}}(k,k'') + F_{\text{MP,MR}}(k,k'')\right), \quad \forall k\in K_{\text{M}} \tag{12}$$

Similarly, split ratio on flow for a rejector:

$$\left(1-\alpha(k)\right)\begin{pmatrix} \sum_{i\in I}\left(F_{\text{SO,MP}}(i,k) + F_{\text{SO,MR}}(i,k)\right) + \sum_{k'\in K_{\text{NM}}} \left(F_{\text{NM,MP}}(k',k) + F_{\text{NM,MR}}(k',k)\right) \\ + \sum_{\substack{k''\in K_{\text{M}}\\ k''\neq k}} \left(F_{\text{MP,MP}}(k'',k) + F_{\text{MP,MR}}(k'',k) + F_{\text{MR,MP}}(k'',k) + F_{\text{MR,MR}}(k'',k)\right) \end{pmatrix}$$
$$= \sum_{j\in J} F_{\text{MR,SI}}(k,j) + \sum_{k'\in K_{\text{NM}}} F_{\text{MR,NM}}(k,k') + \sum_{\substack{k''\in K_{\text{M}}\\ k''\neq k}} \left(F_{\text{MR,MP}}(k,k'') + F_{\text{MR,MR}}(k,k'')\right), \quad \forall k\in K_{\text{M}}. \tag{13}$$

Concentration balances for the permeator of a membrane separation-based regenerator:

$$\left(1-R\left(k,q\right)\right)\left(\begin{array}{l}\sum_{i\in I}F_{\mathrm{SO,MP}}\left(i,k\right)C_{\mathrm{SO}}\left(i,q\right)+\sum_{k'\in K_{\mathrm{NM}}}F_{\mathrm{NM,MP}}\left(k',k\right)C_{\mathrm{NM}}\left(k',q\right)\\ +\sum_{\substack{k''\in K_{\mathrm{M}}\\ k''\neq k}}\left(F_{\mathrm{MP,MP}}\left(k'',k\right)C_{\mathrm{MP}}\left(k'',q\right)+F_{\mathrm{MR,MP}}\left(k'',k\right)C_{\mathrm{MR}}\left(k'',q\right)\right)\end{array}\right)$$
$$=C_{\mathrm{MP}}\left(k,q\right)\left(\begin{array}{l}\sum_{j\in J}F_{\mathrm{MP,SI}}\left(k,j\right)+\sum_{k'\in K_{\mathrm{NM}}}F_{\mathrm{MP,NM}}\left(k,k'\right)\\ +\sum_{\substack{k''\in K_{\mathrm{M}}\\ k''\neq k}}\left(F_{\mathrm{MP,MP}}\left(k,k''\right)+F_{\mathrm{MP,MR}}\left(k,k''\right)\right)\end{array}\right),\quad \forall k\in K_{\mathrm{M}},\forall q\in Q. \tag{14}$$

Concentration balances for the rejector of a membrane separation-based regenerator:

$$R\left(k,q\right)\left(\begin{array}{l}\sum_{i\in I}F_{\mathrm{SO,MR}}\left(i,k\right)C_{\mathrm{SO}}\left(i,q\right)+\sum_{k'\in K_{\mathrm{NM}}}F_{\mathrm{NM,MR}}\left(k',k\right)C_{\mathrm{NM}}\left(k',q\right)\\ +\sum_{\substack{k''\in K_{\mathrm{M}}\\ k''\neq k}}\left(F_{\mathrm{MP,MR}}\left(k'',k\right)C_{\mathrm{MP}}\left(k'',q\right)+F_{\mathrm{MR,MR}}\left(k'',k\right)C_{\mathrm{MR}}\left(k'',q\right)\right)\end{array}\right)$$
$$=C_{\mathrm{MR}}\left(k,q\right)\left(\begin{array}{l}\sum_{j\in J}F_{\mathrm{MR,SI}}\left(k,j\right)+\sum_{k'\in K_{\mathrm{NM}}}F_{\mathrm{MR,NM}}\left(k,k'\right)\\ +\sum_{\substack{k''\in K_{\mathrm{M}}\\ k''\neq k}}\left(F_{\mathrm{MR,MP}}\left(k,k''\right)+F_{\mathrm{MR,MR}}\left(k,k''\right)\right)\end{array}\right),\quad \forall k\in K_{\mathrm{M}},\forall q\in Q. \tag{15}$$

It is worthy to emphasize the point we indicated earlier that the model presented here, with particular respect to the use of set K_{M}, lends a natural formulation to the problem by maintaining the physical configuration of a membrane regenerator as being made up of the two outlets of a permeator and a rejector.

3.3.1.3. Water Balances for Sinks

Mass balances for a sink:

$$\sum_{i\in I} F_{\mathrm{SO,SI}}\left(i,j\right)+\sum_{k\in K_{\mathrm{NM}}} F_{\mathrm{NM,SI}}\left(k,j\right)+\sum_{k'\in K_{\mathrm{M}}}\left(F_{\mathrm{MP,SI}}\left(k',j\right)+F_{\mathrm{MR,SI}}\left(k',j\right)\right)=F_{\mathrm{SI}}\left(j\right),\quad \forall j\in J. \tag{16}$$

3.3.2. Constraints

Quality requirements for a sink as dictated by its MAIC:

$$\begin{aligned}&\sum_{i\in I} F_{\mathrm{SO,SI}}\left(i,j\right)C_{\mathrm{SO}}\left(i,q\right)+\sum_{k\in K_{\mathrm{NM}}} F_{\mathrm{NM,SI}}\left(k,j\right)C_{\mathrm{NM}}\left(k,q\right)\\&+\sum_{k'\in K_{\mathrm{M}}}\begin{pmatrix}F_{\mathrm{MP,SI}}\left(k',j\right)C_{\mathrm{MP}}\left(k',q\right)\\+F_{\mathrm{MR,SI}}\left(k',j\right)C_{\mathrm{MR}}\left(k',q\right)\end{pmatrix}\le F_{\mathrm{SI}}\left(j\right)\cdot C^{\max}\left(j,q\right),\quad \forall j\in J,\forall q\in Q.\end{aligned} \tag{17}$$

0–1 variables are introduced and big-*M* constraints are enforced to guarantee the existence of stream piping interconnections for non-zero flows and vice versa:

$$\begin{gathered}F^{\mathrm{L}}_{\mathrm{A,B}}\left(\beta,\gamma\right)y_{\mathrm{A,B}}\left(\beta,\gamma\right)\le F_{\mathrm{A,B}}\left(\beta,\gamma\right)\le F^{\mathrm{U}}_{\mathrm{A,B}}\left(\beta,\gamma\right)y_{\mathrm{A,B}}\left(\beta,\gamma\right),\\ \forall\left(\beta,\mathrm{A}\right)\in\left(I,\mathrm{SO}\right)\cup\left(K_{\mathrm{NM}},\mathrm{NM}\right)\cup\left(K_{\mathrm{M}},\mathrm{MP}\right)\cup\left(K_{\mathrm{M}},\mathrm{MR}\right),\\ \forall\left(\gamma,\mathrm{B}\right)\in\left(K_{\mathrm{NM}},\mathrm{NM}\right)\cup\left(K_{\mathrm{M}},\mathrm{MP}\right)\cup\left(K_{\mathrm{M}},\mathrm{MR}\right)\cup\left(J,\mathrm{SI}\right)\end{gathered} \tag{18}$$

where $F^{\mathrm{U}}_{\mathrm{A,B}}\left(\beta,\gamma\right)$ is suitably taken to be the maximum capacity of the piping interconnection between entities A and B (Biegler et al., 1997).

To decide whether a regenerator *k* is used or not used in the water network, a 0–1 variable is introduced and the following big-*M* constraints are enforced:

$$\left(\sum_{i \in I} F_{\text{SO,A}}\left(i,\alpha\right) + \sum_{\substack{k' \in K_{\text{NM}} \\ k' \neq k}} F_{\text{NM,A}}\left(k',\alpha\right) + \sum_{\substack{k'' \in K_{\text{M}} \\ k'' \neq k}} \left(F_{\text{MP,A}}\left(k'',\alpha\right) + F_{\text{MR,A}}\left(k'',\alpha\right) \right) \right) \leq F_{\text{A}}^{\max}\left(\alpha\right) y_{\text{A}}\left(\alpha\right), \quad \forall\left(\alpha,\text{A}\right) \in \left(K_{\text{NM}},\text{NM}\right) \cup \left(K_{\text{M}},\text{MP}\right) \cup \left(K_{\text{M}},\text{MR}\right) \tag{19}$$

where $F_{\text{A}}^{\max}\left(\alpha\right)$ is the capacity of the regenerator associated with α.

3.3.3. Objective Function

The objective function of the model involves minimizing the annualized total network cost as contributed by the economic and environmental sustainability drivers. The former consists of:

- the cost of piping interconnections, which is taken to be a linear function of 0–1 variables for the capital cost ($\text{CC}_{\text{piping}}$) and a linear function of flowrates for the operating cost ($\text{OC}_{\text{piping}}$);
- the capital cost associated with the regenerators ($\text{CC}_{\text{regenerator}}$), whose total equipment cost for each of the regenerators is approximated as a linear function of the 0–1 variables denoting its use (or non-use).

The latter consists of the operating cost of freshwater consumption ($\text{OC}_{\text{freshwater}}$) and the operating cost of wastewater treatment in the ETS (OC_{waste}), both of which are assumed to be linear functions of flowrates. The objective function is thus given by:

$$\min \text{CC}_{\text{piping}} + \text{OC}_{\text{piping}} + \text{CC}_{\text{regenerator}} + \text{OC}_{\text{freshwater}} + \text{OC}_{\text{waste}}, \tag{20}$$

with the annualized expressions for the four cost components defined as follows:

$$
\begin{aligned}
\mathrm{CC}_{\mathrm{piping}} = {} & \sum_{i\in I}\sum_{j\in J} d_{\mathrm{SO,SI}}(i,j)\, y_{\mathrm{SO,SI}}(i,j) + \sum_{i\in I}\sum_{k\in K_{\mathrm{NM}}} d_{\mathrm{SO,NM}}(i,k)\, y_{\mathrm{SO,NM}}(i,k) \\
& + \sum_{i\in I}\sum_{k\in K_{\mathrm{M}}} \left(d_{\mathrm{SO,MP}}(i,k)\, y_{\mathrm{SO,MP}}(i,k) + d_{\mathrm{SO,MR}}(i,k)\, y_{\mathrm{SO,MR}}(i,k) \right) \\
& + \sum_{k\in K_{\mathrm{NM}}}\sum_{\substack{k'\in K_{\mathrm{NM}}\\ k'\neq k}} d_{\mathrm{NM,NM}}(k,k')\, y_{\mathrm{NM,NM}}(k,k') \\
& + \sum_{k\in K_{\mathrm{NM}}}\sum_{k'\in K_{\mathrm{M}}} \left(d_{\mathrm{NM,MP}}(k,k')\, y_{\mathrm{NM,MP}}(k,k') + d_{\mathrm{NM,MR}}(k,k')\, y_{\mathrm{NM,MR}}(k,k') \right) \\
& + \sum_{k\in K_{\mathrm{M}}}\sum_{k'\in K_{\mathrm{NM}}} d_{\mathrm{MP,NM}}(k,k')\, y_{\mathrm{MP,NM}}(k,k') \\
& + \sum_{k\in K_{\mathrm{M}}}\sum_{\substack{k'\in K_{\mathrm{M}}\\ k'\neq k}} \left(d_{\mathrm{MP,MP}}(k,k')\, y_{\mathrm{MP,MP}}(k,k') + d_{\mathrm{MP,MR}}(k,k')\, y_{\mathrm{MP,MR}}(k,k') \right) \\
& + \sum_{k\in K_{\mathrm{M}}}\sum_{k'\in K_{\mathrm{NM}}} d_{\mathrm{MR,NM}}(k,k')\, y_{\mathrm{MR,NM}}(k,k') \\
& + \sum_{k\in K_{\mathrm{M}}}\sum_{\substack{k'\in K_{\mathrm{M}}\\ k'\neq k}} \left(d_{\mathrm{MR,MP}}(k,k')\, y_{\mathrm{MR,MP}}(k,k') + d_{\mathrm{MR,MR}}(k,k')\, y_{\mathrm{MR,MR}}(k,k') \right) \\
& + \sum_{k\in K_{\mathrm{NM}}}\sum_{j\in J} d_{\mathrm{NM,SI}}(k,j)\, y_{\mathrm{NM,SI}}(k,j) \\
& + \sum_{k\in K_{\mathrm{M}}}\sum_{j\in J} \left(d_{\mathrm{MP,SI}}(k,j)\, y_{\mathrm{MP,SI}}(k,j) + d_{\mathrm{MR,SI}}(k,j)\, y_{\mathrm{MR,SI}}(k,j) \right),
\end{aligned}
\tag{21}
$$

$$\begin{aligned}
\mathrm{OC}_{\mathrm{piping}} &= \sum_{i\in I}\sum_{j\in J} c_{\mathrm{SO,SI}}(i,j)\,F_{\mathrm{SO,SI}}(i,j) + \sum_{i\in I}\sum_{k\in K_{\mathrm{NM}}} c_{\mathrm{SO,NM}}(i,k)\,F_{\mathrm{SO,NM}}(i,k) \\
&+ \sum_{i\in I}\sum_{k\in K_{\mathrm{M}}} \left(c_{\mathrm{SO,MP}}(i,k)\,F_{\mathrm{SO,MP}}(i,k) + c_{\mathrm{SO,MR}}(i,k)\,F_{\mathrm{SO,MR}}(i,k)\right) \\
&+ \sum_{k\in K_{\mathrm{NM}}}\sum_{\substack{k'\in K_{\mathrm{NM}}\\ k'\neq k}} c_{\mathrm{NM,NM}}(k,k')\,F_{\mathrm{NM,NM}}(k,k') \\
&+ \sum_{k\in K_{\mathrm{NM}}}\sum_{k'\in K_{\mathrm{M}}} \left(c_{\mathrm{NM,MP}}(k,k')\,F_{\mathrm{NM,MP}}(k,k') + c_{\mathrm{NM,MR}}(k,k')\,F_{\mathrm{NM,MR}}(k,k')\right) \\
&+ \sum_{k\in K_{\mathrm{M}}}\sum_{k'\in K_{\mathrm{NM}}} c_{\mathrm{MP,NM}}(k,k')\,F_{\mathrm{MP,NM}}(k,k') \\
&+ \sum_{k\in K_{\mathrm{M}}}\sum_{\substack{k'\in K_{\mathrm{M}}\\ k'\neq k}} \left(c_{\mathrm{MP,MP}}(k,k')\,F_{\mathrm{MP,MP}}(k,k') + c_{\mathrm{MP,MR}}(k,k')\,F_{\mathrm{MP,MR}}(k,k')\right) \\
&+ \sum_{k\in K_{\mathrm{M}}}\sum_{k'\in K_{\mathrm{NM}}} c_{\mathrm{MR,NM}}(k,k')\,F_{\mathrm{MR,NM}}(k,k') \\
&+ \sum_{k\in K_{\mathrm{M}}}\sum_{\substack{k'\in K_{\mathrm{M}}\\ k'\neq k}} \left(c_{\mathrm{MR,MP}}(k,k')\,F_{\mathrm{MR,MP}}(k,k') + c_{\mathrm{MR,MR}}(k,k')\,F_{\mathrm{MR,MR}}(k,k')\right) \\
&+ \sum_{k\in K_{\mathrm{NM}}}\sum_{j\in J} c_{\mathrm{NM,SI}}(k,j)\,F_{\mathrm{NM,SI}}(k,j) \\
&+ \sum_{k\in K_{\mathrm{M}}}\sum_{j\in J} \left(c_{\mathrm{MP,SI}}(k,j)\,F_{\mathrm{MP,SI}}(k,j) + c_{\mathrm{MR,SI}}(k,j)\,F_{\mathrm{MR,SI}}(k,j)\right),
\end{aligned} \tag{22}$$

$$\mathrm{CC}_{\mathrm{regenerator}} = \sum_{k\in K_{\mathrm{NM}}} d_{\mathrm{NM}}(k)\,y_{\mathrm{NM}}(k) + \sum_{k'\in K_{\mathrm{M}}} \left(d_{\mathrm{MP}}(k')\,y_{\mathrm{MP}}(k') + d_{\mathrm{MR}}(k')\,y_{\mathrm{MR}}(k')\right), \tag{23}$$

$$\mathrm{OC}_{\mathrm{freshwater}} = c_{\mathrm{freshwater}} F_{\mathrm{SO}}(i)H, \quad \text{for } i \in \{\text{freshwater}\}, \tag{24}$$

$$\mathrm{OC}_{\mathrm{waste}} = c_{\mathrm{waste}} F_{\mathrm{SI}}(j)H, \quad \text{for } j \in \{\text{waste}\}. \tag{25}$$

4. Solution Strategy Using Valid Inequalities on Logical Constraints

Our proposed model is a nonconvex MINLP due to the presence of bilinear terms that arise in the concentration balances of the regenerators as a result of contaminant mixing in equations (9), (11), (14), and (16). Such bilinear terms can result in multiple local optimal solutions, thereby calling for the implementation of global optimization techniques to guarantee a reliable solution. Towards this end, we implement the model on GAMS 23.2.1 and solve it to global optimality using the general purpose global optimization solver BARON, which employs a branch-and-reduce algorithm (Sahinidis and Tawarmalani, 2005; Tawarmalani and Sahinidis, 2002). BARON performs convexification of bilinearity by constructing convex and concave envelopes as linear underestimators and overestimators, and implements a number of domain reduction heuristics as well as node partitioning or selection schemes to enhance the branch-and-bound search tree.

The recent work by Ahmetović and Grossmann (2011) demonstrates that with current computational infrastructure, BARON is now able to solve a well-posed water network synthesis problem with reasonable practical size to global optimality, particularly if good variable bounds are supplied. Nonetheless, as also advocated in the same paper and earlier in Karuppiah and Grossmann (2006; 2008), there are incentives in developing certain cuts that can help to speed up further the convergence of the spatial branch-and-bound procedure.

In this work, we consider adding extra cuts in the form of linear logical constraints to the MINLP model presented earlier to enhance the convergence speed. These constraints enforce certain design specifications and structural specifications on the interconnectivity relationships among the units and the streams. They also serve as a means to incorporate qualitative design knowledge based on engineering experience, physical insights, and heuristics for process synthesis problems. The logical constraints are otherwise implicitly enforced by material balances particularly the structural specifications. Thus, they are generally redundant to the original model because with sufficient computational time, the optimizer will eventually arrive at a solution that discards the forbidden options ruled out by these constraints (as driven by an appropriate objective function).

Application of logical constraints to speed up solution convergence can be found in work on MILP for oil refinery production scheduling by Shah and Ierapetritou (2011); MILP for refinery crude oil scheduling by Saharidis and Ierapetritou (2009); MINLP for paraxylene separation process using crystallization technology by Lima and Grossmann (2009); MINLP for reverse osmosis network synthesis for wastewater treatment by Saif et al. (2008a); nonlinear generalized disjunctive programming (GDP) for heat-integrated distillation sequences by Caballero and Grossmann (2006; 1999); and nonconvex GDP for process networks by Lee and Grossmann (2003). The pioneering work for process systems engineering applications can be traced back to the use of logic propositions by Raman and Grossmann (1992; 1993).

From a computational viewpoint, the logical constraints can be employed to cut off fractional solutions by imposing restrictions on the possible values of the 0–1 variables in a branch-and-bound scheme. In other words, they are able to reduce the number of nodes in a branch-and-bound search tree and hence, the computational load. They function as cutting planes in the form of logic cuts that increase convergence in solving nonconvex MINLP through tightening the bounds by recovering information lost due to the convex relaxations but without cutting off the optimal solution (Hooker et al., 1994).

The set of logical constraints incorporated into our water network synthesis model is as follows:

(1) Forbid freshwater from being sent directly to a waste treatment plant (i.e., the waste sink) and the main water ways for discharge to environment (i.e., the discharge sink) to prevent an environmentally unsustainable use of a valuable natural resource (see Figure 7):

$$y_{\mathrm{SO,SI}}(i, j) = 0, \quad \forall i \in \{\text{freshwater}\}, \forall j \in \{\text{waste, discharge}\} \tag{26}$$

It is noted that the use of freshwater has been reported in actual industrial practice for the purpose of diluting the effluent mixture in the waste sink to comply with its MAIC and to facilitate the wastewater treatment processes involved. Such a strategy is undertaken to comply with the maximum concentrations allowed by environmental regulations. Freshwater also has been reportedly used as an easy operational alternative (compared to blending with other polluted water streams) to provide a dilution effect in a discharge sink in meeting environmental limits for effluent removal (Eckenfelder, 2000). However, these practices are unsustainable ways of utilizing valuable freshwater resource. They are due to the perverse effect of issuing plant discharge permits based on concentrations and not on the actual amounts of contaminants discharged to the environment. Moreover, they only result in increased demand for freshwater in the face of global water scarcity challenge.

(2) Forbid regenerated water produced by non-membrane regenerators and permeators from directly entering the waste and discharge sinks (see Figure 8):

$$\left.\begin{array}{l} y_{\mathrm{NM,SI}}(k, j) = 0, \quad \forall k \in K_{\mathrm{NM}} \\ y_{\mathrm{MP,SI}}(k, j) = 0, \quad \forall k \in K_{\mathrm{M}} \end{array}\right\} j \in \{\text{waste, discharge}\} \tag{27}$$

It is clearly uneconomical to pay for the cost of water regeneration only to dispose it to the waste treatment plant. On the other hand, it is of course plausible for the low-quality concentrated rejectors to be sent to the waste sink directly.

(3) Forbid mixing of permeator and rejector of the same regenerator in a sink or in any other regenerator (see Figure 9):

$$y_{\mathrm{MP,SI}}\left(k,j\right)+y_{\mathrm{MR,SI}}\left(k,j\right)\leq 1,\quad \forall k\in K_{\mathrm{M}},\forall j\in J \tag{28}$$

$$y_{\mathrm{MP,NM}}\left(k,k'\right)+y_{\mathrm{MR,NM}}\left(k,k'\right)\leq 1,\quad \forall k\in K_{\mathrm{M}},\forall k'\in K_{\mathrm{NM}} \tag{29}$$

$$y_{\mathrm{MP,MP}}\left(k,k'\right)+y_{\mathrm{MR,MP}}\left(k,k'\right)\leq 1,\quad \forall k,k'\in K_{\mathrm{M}},k\neq k' \tag{30}$$

$$y_{\mathrm{MP,MR}}\left(k,k'\right)+y_{\mathrm{MR,MR}}\left(k,k'\right)\leq 1,\quad \forall k,k'\in K_{\mathrm{M}},k\neq k' \tag{31}$$

This logical constraint asserts the main purpose of a membrane regenerator of separating a feed stream into a high-quality permeator (which is of low contaminant concentrations) with a low-quality rejector (of high contaminant concentrations). Furthermore, it is thermodynamically undesirable to allow the re-mixing of these streams that have been separated beforehand.

(4) Forbid more than one piping interconnection between any two regenerators (see Figure 10):

$$y_{\mathrm{NM,NM}}\left(k,k'\right)+y_{\mathrm{NM,NM}}\left(k',k\right)\leq 1,\quad \forall k,k'\in K_{\mathrm{NM}},k\neq k' \tag{32}$$

$$y_{\mathrm{NM,MP}}\left(k,k'\right)+y_{\mathrm{MP,NM}}\left(k',k\right)\leq 1,\quad \forall k\in K_{\mathrm{NM}},k'\in K_{\mathrm{M}} \tag{33}$$

This is because pipes are single-choice interconnections that cannot handle flows in opposing directions.

(5) Enforce that the permeator and rejector of the same regenerator must exist or not exist together:

$$y_{\mathrm{MP}}\left(k\right)=y_{\mathrm{MR}}\left(k\right),\quad \forall k\in K_{\mathrm{M}} \tag{34}$$

This constraint is needed to maintain consistency with the physical configuration of a partitioning regenerator.

5. Case Study: Water Network of a Petroleum Refinery Complex

We implement the proposed MINLP on an industrial-scale case study of the water network of an operating petroleum refinery in South East Asia. A centralized ETS is utilized in the existing refinery water network under study. This system tends to mix all clean and contaminated water sources at various concetrations, which is a poor practice. There is potential for retrofit and for implementing a decentralized distributed treatment system approach to address this issue (Kuo and Smith, 1997). The distributed treatment approach allows local treatment for water regeneration to take place to avoid merging different contaminant levels of water from different operations, hence requiring simpler and cheaper wastewater treatment units. Moreover the capital and operating costs of treatment units is proportional to wastewater volume (Karuppiah and Grossmann, 2006). It is unnecessary to employ the highest quality of regeneration for cleaner wastewater except only for those streams that need advanced treatment.

A numerical example is presented here that involves 28 sources including a single freshwater source (see Table 1); 11 regenerators comprising 6 partitioning regenerators of 3 units of ultrafiltration and reverse osmosis each while the rest are non-membrane separation-based regenerators (see Table 2); and 8 sinks (see Table 3). Data on parameters for the objective function formulation are provided in Table 4. For this instance, our problem is focussed on investigating the organic carbon contaminant of chemical oxygen demand (COD), which is a measure of the quantity of oxygen required to fully oxidize matters present in water. The COD value for the discharge sink is designated according to environmental regulations to ensure discharge permit compliance of the refinery. The optimization is performed using the general purpose global optimization solver GAMS 23.2.1/BARON 7.8.1 with absolute optimality tolerance of 0 and relative optimality tolerance of 7%. The model will be made available publicly following its planned submission to the CMU–IBM Cyber-Infrastructure for MINLP collaborative website (http://minlp.org/).

Assigning relative magnitudes of capital and operating costs and MAIC for the sinks in the model is largely based on the following general structure. The discharge sink is assigned arbitrary costs with magnitudes that are higher than all other sinks because the overarching goal of our model is to promote the selection of options for reuse/recycle. The MAIC for this sink follows environmental regulations. For the waste sink, its arbitrary cost magnitudes are comparable to other sinks, but its MAIC is stipulated to be the largest among all the sinks. A similar structure is adopted for the incineration sink: it is designated the highest arbitrary cost magnitudes with an MAIC that is the second highest among the sinks.

The optimal water network configuration obtained is reported in Figure 11 and Table 5. It registers approximately 27% savings that is equivalent to a reduction of $218,400/year in freshwater use as compared to the existing base case refinery operations (see detailed calculations in Table 6 that summarizes the main computational results). The optimal configuration involves the use of freshwater as a diluent for the inlet streams to the multimedia filtration and RO regenerators. As indicated earlier, such a strategy agrees with industrial practice in facilitating the treatment processes to achieve higher regenerated water quality that comply with the MAIC of the sinks (see Figure 12). For sinks such as boilers that require operations with high water quality, the optimal design recommends the reuse of the permeator of the RO1 regenerator to supply the boiler feedwater.

Table 7 summarizes information on the model size and computational statistics. Due to the problem size particularly the large number of nonconvex bilinear terms, the model is solved using distributed parallel computing performed on the computational grid made available by the computer cluster maintained at the Department of Chemical Engineering at Imperial College London (http://wiki.ce.ic.ac.uk/tiki-index.php?page=The+Linux+Cluster). The cluster has 70 computing nodes with most nodes running on 12-core 3.47 GHz Intel® Xeon™ X5690 processors with 4 to 128 GB of RAM. The original formulation without the logical constraints could not guarantee global optimality in more than 11 days of CPU time (which is the imposed grid limitation) while the inclusion of logical constraints enables a global optimal solution to be achieved in approximately 11 days and 9 hours. To put into context the degree of the bilinearity of our problem and its computational burden, Table 8 compiles a number of recent representative work on pooling problems in terms of the largest number of bilinear terms handled in the numerical examples with the reported CPU time.

6. Concluding Remarks

This paper has addressed the optimal water network synthesis in the process industry by proposing a superstructure-based MINLP framework. Several important practically-driven extensions are considered in this work, mainly the incorporation of membrane separation-based partitioning regenerators, particularly ultrafiltration and reverse osmosis. The proposed approach is applied on an industrial case study with promising results of a 27% annual savings in freshwater use. We advocate the inclusion of physical insights-based linear logical constraints to the model in enhancing convergence to a global optimal solution that is practically reliable.

The huge computational expense advocates a need for future work that considers customized strategies for handling the presence of nonconvex bilinear terms in the model. In this respect, a recent work by Ruiz and Grossmann (in press) promotes a similar approach of adding redundant constraints derived from engineering knowledge and physical insights. These constraints serve the purpose of recovering the physical meaning of a problem, in particular the global concentration balances for each contaminant in water network synthesis, which is lost through relaxation of the nonconvexity in the associated model. This approach consists in a generalization of the reduction constraints method proposed in an earlier work by Liberti and Pantelides (2006). The latter provides a framework to formulate a well-posed nonconvex bilinear programs, in the spirit of the reformulation–linearization technique (RLT) introduced by Sherali and Alameddine (1992) to this class of problem. It is also noteworthy that advances in global optimization techniques to handle bilinearity that has gained a lot of attention recently is the use of MILP-based piecewise-affine relaxation schemes (also known as semilinear cuts) (Bergamini et al., 2008; Wicaksono and Karimi, 2008; Gounaris et al., 2009; Misener and Floudas, 2010a; 2010b; Misener et al., 2011). At this juncture, we hope that our approach may potentially be used in concert with these existing systematic methodologies for deriving bounds-strengthening cuts for water network synthesis problems.

Notations

Sets and Indices

I	set of sources i
J	set of sinks j
K	set of all types of regenerators k where $K = K_{\mathrm{M}} \cup K_{\mathrm{NM}}$
K_{NM}	set of non-membrane separation-based regenerators k
K_{M}	set of membrane separation-based regenerators k
Q	set of contaminants q

Parameters

$C^{\max}(q,j)$	maximum concentration of contaminant q at inlet to sink j
$F_{\mathrm{NM}}^{\max}(k)$	capacity of non-membrane regenerator k
$F_{\mathrm{MP}}^{\max}(k)$	capacity of permeator of membrane regenerator k
$F_{\mathrm{MR}}^{\max}(k)$	capacity of rejector of membrane regenerator k
H	annual operating time of the water systems plant
$R(k,q)$	removal ratio of contaminant q in membrane regenerator k
$\alpha(k)$	liquid-phase recovery factor of membrane regenerator k

Parameters on operating cost of stream piping interconnections:

$d_{\mathrm{SO,SI}}(i,j)$	from source i to sink j
$d_{\mathrm{SO,NM}}(i,k)$	from source i to non-membrane regenerator k
$d_{\mathrm{SO,MP}}(i,k)$	from source i to permeator of membrane regenerator k
$d_{\mathrm{SO,MR}}(i,k)$	from source i to rejector of membrane regenerator k
$d_{\mathrm{NM,NM}}(k,k')$	from non-membrane regenerator k to non-membrane regenerator k'
$d_{\mathrm{NM,MP}}(k,k')$	from non-membrane regenerator k to permeator of membrane regenerator k'
$d_{\mathrm{NM,MR}}(k,k')$	from non-membrane regenerator k to rejector of membrane regenerator k'
$d_{\mathrm{MP,NM}}(k,k')$	from permeator of membrane regenerator k to non-membrane regenerator k'

$d_{\text{MP,MP}}\left(k,k'\right)$	from permeator of membrane regenerator *k* to permeator of membrane regenerator *k′*
$d_{\text{MP,MR}}\left(k,k'\right)$	from permeator of membrane regenerator *k* to rejector of membrane regenerator *k′*
$d_{\text{MR,NM}}\left(k,k'\right)$	from rejector of membrane regenerator *k* to non-membrane regenerator *k′*
$d_{\text{MR,MP}}\left(k,k'\right)$	from rejector of membrane regenerator *k* to permeator of membrane regenerator *k′*
$d_{\text{MR,MR}}\left(k,k'\right)$	from rejector of membrane regenerator *k* to rejector of membrane regenerator *k′*
$d_{\text{NM,SI}}\left(k,j\right)$	from non-membrane regenerator *k* to sink *j*
$d_{\text{MP,SI}}\left(k,j\right)$	from permeator of membrane regenerator *k* to sink *j*
$d_{\text{MP,SI}}\left(k,j\right)$	from rejector of membrane regenerator *k* to sink *j*

Parameters on capital cost of stream piping interconnections:

$c_{\text{SO,SI}}\left(i,j\right)$	from source *i* to sink *j*
$c_{\text{SO,NM}}\left(i,k\right)$	from source *i* to non-membrane regenerator *k*
$c_{\text{SO,MP}}\left(i,k\right)$	from source *i* to permeator of membrane regenerator *k*
$c_{\text{SO,MR}}\left(i,k\right)$	from source *i* to rejector of membrane regenerator *k*
$c_{\text{NM,NM}}\left(k,k'\right)$	from non-membrane regenerator *k* to non-membrane regenerator *k′*
$c_{\text{NM,MP}}\left(k,k'\right)$	from non-membrane regenerator *k* to permeator of membrane regenerator *k′*
$c_{\text{NM,MR}}\left(k,k'\right)$	from non-membrane regenerator *k* to rejector of membrane regenerator *k′*
$c_{\text{MP,NM}}\left(k,k'\right)$	from permeator of membrane regenerator *k* to non-membrane regenerator *k′*
$c_{\text{MP,MP}}\left(k,k'\right)$	from permeator of membrane regenerator *k* to permeator of membrane regenerator *k′*
$c_{\text{MP,MR}}\left(k,k'\right)$	from permeator of membrane regenerator *k* to rejector of membrane regenerator *k′*
$c_{\text{MR,NM}}\left(k,k'\right)$	from rejector of membrane regenerator *k* to non-membrane regenerator *k′*
$c_{\text{MR,MP}}\left(k,k'\right)$	from rejector of membrane regenerator *k* to permeator of membrane regenerator *k′*
$c_{\text{MR,MR}}\left(k,k'\right)$	from rejector of membrane regenerator *k* to rejector of membrane regenerator *k′*
$c_{\text{NM,SI}}\left(k,j\right)$	from non-membrane regenerator *k* to sink *j*
$c_{\text{MP,SI}}\left(k,j\right)$	from permeator of membrane regenerator *k* to sink *j*
$c_{\text{MP,SI}}\left(k,j\right)$	from rejector of membrane regenerator *k* to sink *j*

Continuous variables

$F_{\text{SO}}\left(i\right)$	flow in outlet of source *i*
$F_{\text{SI}}\left(j\right)$	flow in inlet of sink *j*
$F_{\text{SO,SI}}\left(i,j\right)$	flow from source *i* to sink *j*
$F_{\text{SO,NM}}\left(i,k\right)$	flow from source *i* to non-membrane regenerator *k*

$F_{\mathrm{SO,MP}}(i,k)$	flow from source *i* to permeator of membrane regenerator *k*
$F_{\mathrm{SO,MR}}(i,k)$	flow from source *i* to rejector of membrane regenerator *k*
$F_{\mathrm{NM,NM}}(k,k')$	flow from non-membrane regenerator *k* to non-membrane regenerator *k′*
$F_{\mathrm{NM,MP}}(k,k')$	flow from non-membrane regenerator *k* to permeator of membrane regenerator *k′*
$F_{\mathrm{NM,MR}}(k,k')$	flow from non-membrane regenerator *k* to rejector of membrane regenerator *k′*
$F_{\mathrm{MP,NM}}(k,k')$	flow from permeator of membrane regenerator *k* to non-membrane regenerator *k′*
$F_{\mathrm{MP,MP}}(k,k')$	flow from permeator of membrane regenerator *k* to permeator of membrane regenerator *k′*
$F_{\mathrm{MP,MR}}(k,k')$	flow from permeator of membrane regenerator *k* to rejector of membrane regenerator *k′*
$F_{\mathrm{MR,NM}}(k,k')$	flow from rejector of membrane regenerator *k* to non-membrane regenerator *k′*
$F_{\mathrm{MR,MP}}(k,k')$	flow from rejector of membrane regenerator *k* to permeator of membrane regenerator *k′*
$F_{\mathrm{MR,MR}}(k,k')$	flow from rejector of membrane regenerator *k* to rejector of membrane regenerator *k′*
$F_{\mathrm{NM,SI}}(k,j)$	flow from non-membrane regenerator *k* to sink *j*
$F_{\mathrm{MP,SI}}(k,j)$	flow from permeator of membrane regenerator *k* to sink *j*
$F_{\mathrm{MP,SI}}(k,j)$	flow from rejector of membrane regenerator *k* to sink *j*
$C_{\mathrm{SO}}(i,q)$	concentration of contaminant *q* in outlet of source *i*
$C_{\mathrm{NM}}(k,q)$	concentration of contaminant *q* in outlet of non-membrane regenerator *k*
$C_{\mathrm{MP}}(k,q)$	concentration of contaminant *q* in outlet of permeator of membrane regenerator *k k*
$C_{\mathrm{MR}}(k,q)$	concentration of contaminant *q* in outlet of rejector of membrane regenerator *k k*

Binary variables

$y_{\mathrm{SO,SI}}(i,j)$	existence of interconnection from source *i* to sink *j*
$y_{\mathrm{SO,NM}}(i,k)$	existence of interconnection from source *i* to non-membrane regenerator *k*
$y_{\mathrm{SO,MP}}(i,k)$	existence of interconnection from source *i* to permeator of membrane regenerator *k*
$y_{\mathrm{SO,MR}}(i,k)$	existence of interconnection from source *i* to rejector of membrane regenerator *k*
$y_{\mathrm{NM,NM}}(k,k')$	existence of interconnection from non-membrane regenerator *k* to non-membrane regenerator *k′*
$y_{\mathrm{NM,MP}}(k,k')$	existence of interconnection from non-membrane regenerator *k* to permeator of membrane regenerator *k′*
$y_{\mathrm{NM,MR}}(k,k')$	existence of interconnection from non-membrane regenerator *k* to rejector of membrane regenerator *k′*
$y_{\mathrm{MP,NM}}(k,k')$	existence of interconnection from permeator of membrane regenerator *k* to non-membrane regenerator *k′*

$y_{\mathrm{MP,MP}}(k,k')$	existence of interconnection from permeator of membrane regenerator *k* to permeator of membrane regenerator *k′*
$y_{\mathrm{MP,MR}}(k,k')$	existence of interconnection from permeator of membrane regenerator *k* to rejector of membrane regenerator *k′*
$y_{\mathrm{MR,NM}}(k,k')$	existence of interconnection from rejector of membrane regenerator *k* to non-membrane regenerator *k′*
$y_{\mathrm{MR,MP}}(k,k')$	existence of interconnection from rejector of membrane regenerator *k* to permeator of membrane regenerator *k′*
$y_{\mathrm{MR,MR}}(k,k')$	existence of interconnection from rejector of membrane regenerator *k* to rejector of membrane regenerator *k′*
$y_{\mathrm{NM,SI}}(k,j)$	existence of interconnection from non-membrane regenerator *k* to sink *j*
$y_{\mathrm{MP,SI}}(k,j)$	existence of interconnection from permeator of membrane regenerator *k* to sink *j*
$y_{\mathrm{MP,SI}}(k,j)$	existence of interconnection from rejector of membrane regenerator *k* to sink *j*
$y_{\mathrm{NM}}(k)$	selection of non-membrane regenerator *k*
$y_{\mathrm{MP}}(k)$	selection of permeator of membrane regenerator *k*
$y_{\mathrm{MR}}(k)$	selection of rejector of membrane regenerator *k*

Subscripts

NM	index for non-membrane regenerators
MP	index for permeator of membrane regenerators
MR	index for rejector of membrane regenerators

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