Review of Optimization Models for Integrated Process Water Networks and their Application to Biofuel Processes

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Abstract. This paper provides an overview of recent development in the area of optimal synthesis of process water networks in which a major goal is to reduce the freshwater consumption by the reuse and recycle of process and treatment streams. The recent models can globally optimize these networks through mixed-integer nonlinear programming techniques. We discuss the application and impact of these techniques to biofuel plants, which are known to consume large amounts of water.

Introduction. Although water remains relatively cheap, it is an important natural resource that is increasingly becoming scarce as seen in Figure 1. In fact, it is projected that in 2025, two-thirds of the world population will experience water stress [1]. Since chemical, petroleum, and especially biofuel processes consume significant amounts of water, there is a clear incentive to reduce the net consumption of water in these processes. Furthermore, increasingly stringent environmental regulations on plant emissions as well as wastewater discharge have led to more effort devoted to reducing water consumption.

Reducing energy consumption through the use of systematic synthesis techniques [2] can often also lead to reduction of freshwater consumption. A simple example is heat-integrated distillation columns that leads to lowering the consumption of steam in the reboilers, which in turn decreases water use in the boiler loop. This coupling of the two process synthesis areas, energy and water, highlights the importance of performing simultaneous optimization, in which complex tradeoffs can be established among raw materials, investment cost, and energy and water consumption, which in turn leads to lower cost solutions. However, the traditional procedure for process designs uses a sequential approach, in which the process is first optimized without regard of energy and water integration. Next the heat exchanger network (HEN) is synthesized, followed by the synthesis of the water network (WN). In general, using such a sequential approach fixes a-priori important decision variables (e.g. temperature, flowrate), which may have major impact on the feasibility of optimal water and heat integration. Thus, at least conceptually, process flowsheets should be optimized simultaneously with heat and water integration.

A full simultaneous optimization approach, however, may be difficult to solve. First, optimal flowsheet synthesis problems usually take the form of nonconvex mixed-integer nonlinear programs (MINLP) where 0-1 variables are used to decide on the selection of the equipment, and nonlinear nonconvex terms arise in the objective function and process models. In addition, optimal synthesis models heat exchanger and water network synthesis are primarily characterized by MINLP problems and NLP problems, respectively [2,3]. Thus, the full integration of
these models for the simultaneous synthesis of flowsheets, heat exchange networks, and water networks may be very difficult to converge and computationally expensive to solve. To overcome this drawback, a targeting approach can be adopted, which determines the optimal levels of energy and water consumption without deciding on the final system configuration of the HEN and WN. Since the synthesis of HEN has been extensively covered in other reviews, and the concept of simultaneous optimization and heat integration has also been reported before, we focus on reviewing synthesis methods for water networks and a recent development for incorporating targets for water consumption in simultaneous synthesis strategies.

**Projected Global Water Scarcity, 2025**

![Projected Global Water Scarcity, 2025](image)

**Physical water scarcity:** More than 75% of river flows are allocated to agriculture, industries, or domestic purposes. This definition of scarcity — relating water availability to water demand — implies that dry areas are not necessarily water-scarce.

**Approaching physical water scarcity:** More than 60% of river flows are allocated. These basins will experience physical water scarcity in the near future.

**Economic water scarcity:** Water resources are abundant relative to water use, with less than 25% of water from rivers withdrawn for human purposes, but malnutrition exists.

**Little or no water scarcity:** Abundant water resources relative to use. Less than 25% of water from rivers is withdrawn for human purposes.

**Not estimated**

Source: International Water Management Institute.

**Figure. 1. Projected water scarcity in 2025 [39].**

**Review of Water Network Synthesis.** Conventional water use in the process industry does not reuse or recycle water streams of different quality. However, in principle a waste stream can be reused, recycled, or regenerated to lower the overall freshwater consumption. WN design has been a major topic in process synthesis for the past 20 years. The major elements in a WN formulation are water-using process units (PU) and wastewater treatment units (TU) which are interconnected through a set of mixers and splitters. A number of contaminant loads are given for each of the process units as well as fractional recoveries of them in the treatment units. Freshwater is supplied to the network, while all possible combinations of reuse, regeneration reuse, local recycling, and mixing of
streams are considered as illustrated in the superstructure in Fig. 2 for the case of 5 process units and 3 contaminants, where each requires a treatment unit. The objective is to minimize the freshwater flow into the network, or more generally the cost of the entire water process network.

Different models of WN have been proposed in the literature (e.g. water-using networks include water-using process units only, and wastewater treatment networks consider only wastewater treatment units) as discussed in the work by Bagajewicz and Faria [4]. The information required is stream flowrate and concentration of contaminants. Water-using process units can be classified either as mass-transfer process (e.g. extraction, absorption) or non-mass-transfer process (e.g. a reactor that consumes water in the reaction). A treatment unit design equation could be defined either for fixed outlet concentration or for a fixed contaminant removal ratio. The resulting formulation is a simplified model of the network. Both insight-based method and mathematical optimization-based technique have been investigated for water network synthesis.

Inspired by the pinch analysis for heat- and mass-exchange network synthesis, this insight-based approach has been applied to water network designs. A thorough review of insight-based methods is presented in Foo [5]. The first water pinch method was developed by Wang and Smith [6] where flowrate targeting as well as the detailed design of the network structure are determined in a sequential approach through the use of limiting composite diagram. Kuo and Smith [7] provided an extension to the previous work in order to address the design for treatment systems in both single and multiple contaminant cases. Hallale [8] presented a graphical approach called Water Surplus Diagram which provides a rigorous targeting technique for non-mass-transfer processes with a single contaminant. Then Tan et al. [9] developed a targeting approach to target non-mass-transfer water-using process units for two contaminants. The main difference between pinch analysis in HEN and WN is that whereas
only one quality—heat—is being transferred in HEN, WN involve multiple contaminants. Thus, even though the method is well-studied for WN with a single contaminant, a more general optimization model and optimization algorithm is required for a general multi-contaminant WN.

Mathematical optimization-based approach generally employs the superstructure that considers all feasible network structures like the one in Fig. 2. Extensive reviews of mathematical optimization techniques can be found in Bagajewicz [10] and Jezowski [11]. The major advantage of these techniques is that multiple-contaminants can be taken into consideration and that targeting and design can be optimized simultaneously. The first use of superstructure-based optimization approach for WN appears in the seminal work by Takama et al. [12]. The paper incorporates in the NLP model both water-using processes and treatment units for a multi-contaminant system in one framework. Alva-Argaez et al. [13] developed a mathematical optimization model that applies water pinch to wastewater minimization through the use of transshipment model. Savelski and Bagajewicz developed the necessary conditions of optimality for minimum freshwater requirement for the case of single contaminant [14] and then later for the case of multiple contaminants [15]. Huang, Chang, and Ling [16] presented an NLP model to determine the optimal freshwater consumption and wastewater treatment capacity.

Karuppiah and Grossmann [17] formulated an NLP problem to address both process and treatment units for a multi-contaminant system. Bilinear terms (flowrate multiplied by contaminant concentration) that are present in mixers give rise to nonconvexities in the model. As a result, a global optimization method is required. A non-redundant bound strengthening cut was developed to significantly improve the quality of the relaxation in the global optimization algorithm. In addition, this work proposes a generalized disjunctive programming (GDP) formulation to select among multiple treatment technologies for the removal of each type of contaminant. Ahmetović and Grossmann [3] expanded the model to include multiple freshwater sources and pretreatment options while considering all possible interactions among the units. The model is then formulated into an MINLP problem with binary variables to account for the cost of piping. The MINLP model for the example in Figure 2 involves 72 binary variables, 233 continuous variables, and 251 constraints. Using the global optimizer BARON with the special cut by Karuppiah and Grossmann [17], the problem can be solved to 1% optimality gap in about 3 minutes (197.5 CPUs). The optimal network design is shown in Figure 3. The network requires 40 ton/h of freshwater. In comparison, a conventional WN without recycling or reuse requires 300 ton/h of freshwater, almost an order of magnitude higher.
An extension to WN synthesis is simultaneous heat and water integration for a fixed flowsheet. Savulescu and Smith [18] is the first work to consider a heat-integrated single-contaminant WN with only water-using process units. The approach is an insight-based sequential procedure where direct heat transfer is maximized while maintaining the minimum freshwater consumption in the first stage, then the HEN is designed in the second stage. A superstructure-based mathematical optimization model is then presented by Bogataj and Bagajewicz [19] for non-isothermal multi-contaminant WN with only water-using process units. This MINLP model considers the typical WN model simultaneously with a modified HEN superstructure presented by Yee and Grossmann [2].

Similar to simultaneous process flowsheets and heat integration as demonstrated in Duran and Grossmann [20], water integration can also be incorporated. As discussed above, the challenge is solving the detailed HEN and WN, which are generally formulated as nonconvex MINLP or NLP. To simplify the model, Yang and Grossmann [21] proposed an LP targeting model for minimizing the freshwater consumption of a multi-contaminant system. This water targeting model, along with the available heat targeting model, is then incorporated into flowsheet optimization process to achieve minimal heating utility, cooling utility, and freshwater consumption. Examples reported by Yang and Grossmann [21] show that using both the heat and water targets, solutions can be obtained that not only yield lower costs, but also yield reduced energy and water consumption. While full simultaneous model as described here have not been applied to the design of biofuel processes, simultaneous flowsheet optimization and heat integration has been used, followed by optimal water network synthesis, yielding significant reductions in freshwater consumption as will be shown in the next section.
Applications for biofuels. There are two main water consumption stages in the production of biofuels: the water that is used in the production of the biomass, mainly due to irrigation [1], and that used along the process water that transforms the biomass into fuels.

Second generation of biofuels can reduce the water required by irrigation because of raw material integration. For instance, not only corn grain but also corn stover can be used for the production of ethanol. The second raw material can be processed either via hydrolysis and sugars (hexoses and pentoses) fermentation, or gasification. Thus the irrigation needed is cut by almost half since the yield from the plant to ethanol increases. Another lignocellulosic raw material, switchgrass, is a perennial warm-season grass which are deep-rooted and efficient in their use of nutrients and water, and therefore are relatively drought-tolerant. If switchgrass grows in its native habitat, yields from 4.5 to 8 dry tons per acre can be obtained without irrigation. However, if switchgrass is grown in non-native regions, irrigation is needed [22]. Considering algae, we can think of evaporation losses from the ponds [23]. According to the literature [24], it is estimated 3.15 to 3650 L of water per L of algae fuel are needed. However, the use of wastewater or saline water has also been reported in which case the consumption of freshwater in the production of oil form algae should be almost negligible. Another interesting example is the use of wastewater in the production of several biofuels from algae [25].

Process water, energy and water integration. The consumption of process water depends on the raw material used and the processing technology, namely low temperature and high temperature routes. The first ones include biochemical processes for bioethanol production from either corn or lignocellulosic raw materials as well as transesterification processes by using heterogeneous or homogeneous catalysts (alkali, acid or enzymes in both cases). These processes are characterized by complex energy integration, due to the moderate operating temperatures in the fermentation and transesterification reactions, dilute water - ethanol mixtures or the need of water for some washing steps. High temperature processes for bioethanol production include those involving biomass gasification or those using supercritical fluids for biodiesel production. Contrary to what might be expected, gasification does not result in larger water consumption, mainly due to the possibility of using air coolers as well as more efficient heat integration within the process.

Table 1.- Evolution of water consumption in corn based ethanol plants

| Corn based ethanol plants     | Water consumption (L/L) |
|-------------------------------|-------------------------|
| First plants [40]             | 11                      |
| Phillips 1998 [34]            | 5.8                     |
| Old plants in 2006 [26]       | 4.6                     |
| New plants 2008 [26]          | 3.4                     |
| Delta T (2011) [27]           | 1.5                     |
**Process water consumption.** Most of the available data from real plants is for corn-based ethanol. The lack of efficient designs and proper technology in these plants resulted in large water consumption. MTAP produced a report in 2008 [26] where the water consumed at old and new plants in the region was presented. The average consumption of water decreases from 4.6 L per L for old plants to 3.4 L per L for new plants, representing another 25% decrease in water consumption. In Table 1, we can see the evolution of the water consumption per L of ethanol produced from the beginning to the current plants. Recently, Delta T claimed values of 1.5 L per L [27].

To illustrate the impact of the process water network optimization models discussed in this article, we consider several examples of their application in the optimization of the production of biofuels. For the case of corn-based ethanol, a superstructure of alternatives was optimized first based on energy minimization. Karrupiah et al [28] identified that the most energy-consuming units are the distillation columns, and therefore the use of multieffect columns was proposed, which reduced the heat consumption by one half and the cooling load by more than two thirds. Based on the optimized flowsheet it was possible to identify the main water consumption units as well as treatment units to remove suspended solids, dissolved solids, and organics. A large amount of water was used in the fermentor, since ethanol concentration needed to remain low, below 15% by weight. Furthermore, water was used in washing the corn and in hydrolysis with steam. However, the process also generated wastewater containing solids from the washing step, and organics from the fermentation. Thus, it was possible to design an optimal network that efficiently treats and reuses the water by using the superstructure with multiple effects. By applying the model developed by Ahmetović and Grossmann [3], the water consumption was reduced to 1.5 L/L, a level similar to the one reported in the literature. Figure 4 shows the optimal network [29].

In terms of lignocellulosic raw materials, the water consumption ranges from 1 to 9.8 [30] depending on the particular raw material. These values must actually be divided between biochemical processing, for which values of 6 to 10 L/L are typically reported, and thermo-chemical or thermo-biochemical processes in which cases commonly 2 L/L are reported [22]. Coskata in 2008 reported a value of 1 L per L [31]. Using the same procedure as that presented for the corn-based ethanol, very encouraging results were obtained. A superstructure of options was formulated and optimized for simultaneous energy consumption, including the use of multieffect columns in case of sugars or syngas fermentation. Air cooling was used in place of cooling tower for further water use reduction since cooling tower incurs large evaporation losses. Before the design of the water network, we identify the units that use water or produce wastewater, which if properly treated can be reused. Table 2 shows the identification of units for ethanol production processes. Process units are those which generate contaminants in the water, demand units are the ones that need freshwater and sources are the ones that produce a secondary stream of contaminated water. Figure 5 shows some results of water consumption for different processes, from corn-based ethanol, to various processes to transform switchgrass into bioethanol, either via biochemical or thermo-chemical/thermобiochemical. We compare the results of the optimized processes design using energy and water saving technologies, Karrupiah et al [28], Martín & Grossmann [32], Martín & Grossmann [33], with those
by different studies from the NREL [34, 35]. We see that ethanol production is competitive with gasoline as long as water saving technologies, including air cooling are used [36].

Figure 4. Optimal water network for bioethanol production from corn. Flows in (t/h). [36]

Table 2.- Identification of units as a function of water use.

| Process units         | Demand         | Sources                   |
|-----------------------|----------------|---------------------------|
| Biomass Washing       | Fermentors     | Rectification column      |
| Cooling tower         | Cooling tower  | Beer column               |
| Boilers               | Boiler         | Scrubber                  |
|                       | Acid pretreatment | Condensation              |
Finally, the water consumption in biodiesel production processes is evaluated. Pate [37] reported values from 0.32 to 1 L per L. A simultaneous optimization and heat integration approach based on the model proposed by Duran & Grossmann [20] was used to optimize the production of biodiesel not only using methanol [23] but also ethanol [38]. After integration, a water network for ethanol was designed as in previous cases. Table 3 shows the energy and water consumption for the production of biodiesel, Fatty acid methyl ether (FAME) from methanol, and Fatty acid ethyl ether (FAEE) from ethanol. We see that values in the range predicted by Pate are achieved.

Table 3. Energy and water consumption for biodiesel production using different technologies

|             | Methanol | Ethanol |
|-------------|----------|---------|
|             | KOH      | H2SO4   | Superc | Het | Enzy | KOH | Superc | Het | Enzy |
| Thermal energy (MJ/L) |          |         |        |     |      |      |        |     |      |
| Algae       | 0.51     | 0.51    | 1.03   | 0.51 | 0.55 | 0.74 | 2.33   | 1.58 | 0.51 |
| Cooking     | 0.58     | 0.58    | 1.14   | 0.51 | 0.89 |      |        |      |      |
| Water (L/L) |          |         |        |     |      |      |        |     |      |
| Algae       | 0.60     | 0.87    | 0.87   | 0.59 | 0.72 | 0.47 | 1.98   | 1.08 | 0.35 |
| Cooking     | 0.32     | 0.67    | 0.61   | 0.33 | 0.59 |      |        |      |      |

Conclusion. We hope that this paper has shown both the importance of water as a natural resource, and of optimization models that have been developed for reducing freshwater consumption in process water networks. These models can be applied to any petroleum and chemical process, but here we have focused on biofuel
processes as these tend to consume large amounts of water. Although water continues to be cheap in many parts of the world, the goal of efficiently using water as has been outlined in this paper would seem to represent an important contribution towards the sustainable design of chemical processes.

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