Sequential optimization of a water-based polygeneration system in ethylene glycol production

T Prihatin*1, S Mahadzir2 and M I Abdul Mutalib2
1Processing Technique of Mineral and Coal Mining Products, Bukit Asam Mining Industry Community College, Tanjung Enim, Sumatera Selatan, Indonesia
2Chemical Engineering Department, Universiti Teknologi PETRONAS, Bandar Seri Iskandar, 31750, Tronoh, Perak, Malaysia
E-mail: triana.prihatin@gmail.com

Abstract. Polygeneration system as one of the promising strategies shows the great potential for natural resource sustainability, particularly water. In this paper, a model of water-based polygeneration system to minimize the freshwater consumption and the wastewater generation is presented. Water usage in a heat and power utility, cooling utility, and chemical production are modelled simultaneously. To demonstrate the applicability of the model, a case study on the synthesis of optimum water-based polygeneration system is developed for an ethylene glycol production. A two-steps optimization method is presented. The first step consists of the synthesis and design of the polygeneration unit in which a superstructure containing three subsystems, i.e. heat and power generations, re-circulating cooling water system, and chemical production is introduced. A mixed integer non linear program (MINLP) model is developed to allow the selection of choices. The second step includes the incorporating of wastewater treatment strategy and a non linear program model is developed. As results, comparing to the simultaneous method in the same case study, this approach gives a better result in a computation efficiency.

1. Introduction
Water will be one of the crucial issues of the 21st century. Water is probably the most widely used raw material in the process industries and it has been used in abundant quantities by chemical, petrochemical, petroleum refining, food and drink, pulp and paper, power generation and many other industries. Industrial development has led to the fast acceleration of freshwater withdrawals. Moreover, the freshwater consumption in industry creates more pressure on freshwater resources from the impacts of wastewater discharges [1]. According to the UN World Water Development Report [2], some 300-500 million tons of heavy metals, solvents, toxic sludge, and other wastes accumulate each year from industry, most of which gets into the freshwater supply. In some developing countries, 70% of industrial wastes are dumped into untreated waters where they pollute the drinking water [2]. However, instead of seen as a problem, wastewater should be recognized as a water resource.

Numerous approaches have been investigated to give a solution for that global water problem. Water polygeneration system is one of the promising strategies which shows the great potential to solve the
problem of limited freshwater resources. Polygeneration system is an integrated approach to an industrial process that generates multiple products from a single or more natural resources [3-4]. It is usually the integration of power generation and chemical process production. Different types of polygeneration system design have been presented, particularly on the energy-based focusing on the evaluation of existing plants and process technologies [5-7], the configuration and the performance of processes [8-10].

It is seen that there has been little discussion about polygeneration system based on water. Many of the recent studies on water minimization have only focused on the common points with a heat exchanger network problem and were mainly developed for targeting procedure but excluding 3R as a strategy to conserve water. In fact, a simultaneous demand for heat, power and cooling exists in certain chemical industries. The purpose of this study was to investigate a water-based polygeneration system co-producing heat and power, cooling water, and chemicals, incorporating 3R strategy (regenerate, reuse, and recycle). Efficient use of water in the industry can be achieved through 3R strategy within the processes, which can be designed using mathematical optimization approaches. Sequential optimization through two-steps procedure is presented. In the first step, an MINLP model is developed through a superstructure representation to select the appropriate configuration of water-polygeneration and in the second step 3R strategy is presented to the plant configuration in a NLP model to reduce freshwater consumption and wastewater generation. A water polygeneration system for ethylene glycol production is presented as a case study.

2. Optimization First Level (MINLP)

In this paper, a superstructure-based optimization approach is used for the design of water polygeneration. One common approach to systematically build superstructure is combining the detailed superstructures for each subsystem that perform multiple tasks or functions and interconnect them. All the feasible interactions would be taken into account. In this work, the water-based polygeneration system consists of three subsystems: heat and power generation, cooling water system and ethylene glycol (EG) synthesis process. The schematic of sequential optimization method is shown in Figure 1. General superstructure for the synthesis of an optimum water-based polygeneration system in an ethylene glycol production is shown in Figure 2.

![Figure 1. Schematic of sequential optimization method](image-url)
To illustrate the model superstructure, the set technology choices of three subsystems of the water-based polygeneration system have been determined. (1) Heat and power generation: conventional boiler (CB), cogeneration including heat recovery steam generator (HRSG), back-pressure steam turbine (BPST), condensing steam turbine (CST). (2) Recirculating Cooling Water System: forced mechanical draft and induced mechanical draft. There are two broad classes of cooling tower, i.e., natural draft and mechanical draft cooling towers. (3) Ethylene Glycol Production Process: EO production: O₂-based EO and air-direct oxidation, EG process reaction: EO hydrolysis and the Shell’s OMEGA process, reactor: continuous stirred tank reactor (CSTR) and plug flow reactor (PFR), separation: direct sequence, indirect sequence and dividing wall distillation column (DWC).

Ethylene glycol is used mainly as an antifreeze in automobile radiators, as an engine coolant and as a raw material for the manufacture of polyester fibers and plasticiser. There are two common methods of ethylene glycol production, i.e hydration of ethylene oxide [11] and mono-ethylene glycol (MEG) only technology [12]. Ethylene oxide (EO) is obtained by direct oxidation of ethylene with air or oxygen (O₂). Some literatures reported that a molar ratio of ethylene oxide to water may reach 1:5 to 1:30 [12-13]. The ethylene oxide is thermally hydrolyzed to ethylene glycol without a catalyst. Diethylene glycol, triethylene glycol and polyglycols are also produced, with respectively decreasing yield, and are separated from the ethylene glycol by series of distillation columns at reduced pressure. In the second method, ethylene glycol is manufactured by the reaction of ethylene oxide with carbon dioxide to form ethylene carbonat, an intermediate product, which can be hydrolyzed to ethylene glycol. In this work, the alternative process/technology in reactor and separation units of ethylene glycol plant is investigated in Aspen HYSYS simulation for superstructure configurations.

In this first-step optimization, the model of the superstructure presented is formulated as a MINLP problem in which binary variable 0 or 1 is assigned as a logical constraint, which acts as a yes-no decision for the potential existence of options. In this work, shortcut equations are developed for the MINLP model. Some equations and constraints in the simultaneous method [14] are eliminated, for instance, the energy balances around steam boiler and the enthalpy values. For heat and power generation subsystem, mass balance around steam boiler is expressed as:

\[ \sum_{j \in \mathcal{B}} F_{j,0} y_{b} - \sum_{k \in \mathcal{B}} F_{k,0} y_{b} = 0 \]  

(1)
where $F$ is flowrate of inlet streams $j$ and outlet streams $k$, respectively, and $y_b$ is a binary variable representing the selection of boiler, i.e conventional boiler or HRSG. While, mass balance of the steam turbines is given as follows:

$$
\sum_{j} \sum_{a} F_{j,a} y_a - \sum_{k} \sum_{a} F_{k,a} y_a = 0
$$

where $y_{st}$ is a binary variable for steam turbine options, i.e. back pressure and condensing steam turbines.

In equation (3), the power demand, $W_{st,i}$, and the steam enthalpy at steam pressure level, $\Delta H_{st,i}$, is correlated to the flowrate intake of the steam turbine, $FT_{st,i}$ [15].

$$
FT_{st,i} = W_{st,i} / \Delta H_{st,i}
$$

In this work, equation of power produced/demand in the boiler options is linearized as a function of water intake ($F_{dw}$) as follows.

$$
W = f (F_{dw})
$$

where $F_{dw}$ is flowrate of demin water.

In the cooling water subsystem, mass balance around the cooling tower is given by:

$$
\sum_{j} \sum_{a} F_{j,a} y_{cw} - \sum_{k} \sum_{a} F_{k,a} y_{cw} = 0
$$

where the binary variable $y_{cw}$ denotes the selection of cooling tower. The detailed mass balance at specific technology as below:

$$
F_{in} + F_{m} - F_{out} - F_{b} = 0
$$

where $F_{in}$ is the inlet water flowrate to cooling tower, $F_{out}$ is outlet water flowrate of cooling tower, $F_{m}$ is the cooling water makeup and $F_{b}$ is the cooling tower blowdown.

In the ethylene glycol production subsystem, most parameters in the model are estimated from the Aspen HYSYS simulation [16] using the UNIQUAC equation. This fluid package shows a good representation for liquid structure and can be applied to a wide range of mixtures containing water. The reactors are operated under isothermal conditions. The constraints for the supply of cooling, heating and power requirement in the chemical production are formulated as per process demands. Ethylene glycol reactor products will go through distillation column to produce mono-ethylene glycol which is purified from the by-products of water and other higher glycols derivatives. In this separation process, the product recovery and purification also consume water for the heating and cooling. Cooling water demand, $F_{CW}$, at ethylene glycol production, particularly in the separation process, is correlated mainly to condenser duty, $Q_{cd}$, as shown in equation (7).

$$
F_{CW} = \frac{Q_{cd}}{CP.\Delta T}
$$

where $CP = 4.18$ kJ/kg °C and $\Delta T = 20$ °C.

The objective function ($z$) includes the cost of freshwater intake, $C_{fw}$, as shown in equation (8). The cost is calculated on annual basis assuming AOT, annual operation time, of 8000 hrs/year. The cost of freshwater ($C_{fw}$) is 0.26 $/ton.
3. Second Level Optimization
Reduction of freshwater consumption and wastewater discharge has become one of the main targets of design and optimization of process design. Reducing wastewater affects both effluent treatment and freshwater costs. Therefore, the best configuration obtained from the first step is being optimized in this second step by implementing the 3R strategy for water recovery to obtain the optimum solution of water polygeneration system. 3R strategy includes reuse, regeneration reuse, and regeneration recycling. In reuse process, wastewater is reused directly in other operations subject to the level of contamination to operations within the process system. Regeneration is a term used to describe any treatment process that regenerates the quality of water such that it is acceptable for further use. Water recycling involves post-treatment process of the water quality to meet the next water user specification.

The mathematical model is formulated as a NLP problem. Water recovery unit will treat blowdown from the boiler and the cooling tower, and wastewater from ethylene glycol process production. During operation of cooling tower and boiler, there will be accumulation of solids that can impact to heat transfer or other unit operation. Based on Baliban et al [17], the concentrations of suspended solids (TSS) and dissolved solids (TDS) in the boiler blowdown stream are 10 ppm and 500 ppm, respectively. The solids concentration in the cooling tower blowdown will be 50 ppm for suspended solids and 2500 ppm for dissolved solids. These quantities are set in equation (12) as:

$$\text{(9)}$$

For ethylene glycol process production, the maximum allowable concentration of dissolved solids in the wastewater is 500 ppm as set as $C_{ww_{max}}$. $F_{ww_{out}}$ is flow rate of specified contaminants in wastewater and $F_{ww_{out}}$ is flow rate of wastewater leaving ethylene glycol plant, as following:

$$\text{(10)}$$

3.1. Objective Function
The objective function for the model is to optimize the flow rate of freshwater consumption (FW) at minimum total annualized cost as shown in equation (14). Minimization of freshwater consumption directly reduces freshwater intake by the ethylene glycol production. The objective function ($z$) includes the cost of freshwater intake, $C_{fw}$, and the cost of water recovery, $OP_{recovery}$.

$$\text{(11)}$$

The cost of water recovery takes into account the pumping cost as well as cost associated with water recycle, reuse, and regeneration (3R). The cost of water recovery is correlated to total power need for pumps operation ($P_i$) and cost of electricity ($C_{elect}$) by the following equations:

$$\text{(12)}$$
The cost of electricity \( C_{\text{elect}} \) is 0.064 $/kWh. The power consumed by the pump is a function of the water flowrate to the pump, \( F_p \), pump efficiency, \( \eta_p \), and density of water, \( \rho \), as given below [18]:

\[
P_p = 1283 \times \left( \frac{F_p}{\eta_p \rho} \right)^{0.476}
\]

(13)

4. Case Study Results

The proposed mathematical model is used to analyze the case study of a water-based polygeneration system in ethylene glycol production. This study concerns on water flow in the three subsystems that might detect the possibility of water recovery. The complexity of the work is the interactions between the chemical process and the subsystems of heating, power, and cooling. The overall model is developed in GAMS on Intel (R) Core of 2.20 GHz with a memory size of 4 GB. For first step, the model is solved to optimum using DICOPT++ as the MINLP solver. It is a non-convex model contains 15 binary variables, 109 continuous variables and 101 equations. It takes 885 iterations and 4 major iterations only to claim an optimum. These results are compared with the simultaneous method [14] in which water polygeneration system is formulated simultaneously with water recovery in MINLP model, as given in Table 1.

### Table 1. Results of water-based polygeneration system

| Method      | Freshwater consumption (t/hr) | Wastewater generation (t/hr) | Objective function ($/yr) | Computation efficiency       |
|------------|-------------------------------|-------------------------------|---------------------------|------------------------------|
| Simultaneous | 224.375                        | 5.154                         | 648,330                   | No equations: 309 No variables: 330 Binary: 20 Solver times: 0.47 (NLP) 1.01 (MIP) No iterations: 3683 |
| Sequential: |                               |                               |                           |                              |
| First step  | 448.886                        | 228.897                       | 933,682                   | No equations: 101 No variables: 109 Binary: 15 Solver times: 0.18 (NLP) 0.45 (MIP) No iterations: 885 |
| Second step | 225.144                        | 5.156                         | 650,248                   | No equations: 17 No variables: 17 No iterations: 3 |

In the second step, the NLP model with 3R strategy gives an optimal solution contains 17 continuous variables and 17 equations. The problem is solved using CONOPT. Freshwater is consumed as makeup for demineralized water in the heat and power subsystem, feed to the ethylene glycol reactor, and makeup water for the cooling tower. The blowdown from boiler and cooling tower units and wastewater generated in the ethylene glycol plant are mixed and recovered in the regeneration unit. This regenerated water then
recycled to the freshwater intake might reduce the cost of water consumption. As summarized in Table 1, through two-steps procedure the optimal solution can be obtained. The shortcut model can achieve a minimum freshwater consumption and cost of 225.14 t/hr and 650,248 $/yr respectively, close to the results through simultaneous method. This approach also show a significant reduction of the computational efficiency. To validate the work, the preliminary results obtained from the simulation model are compared with those from the simulation model of the utility plant by Bruno et al. [19]. The work of Bruno et al. is chosen as a validation of the simulation for this work due to the similarity of the model simulation for heat and power generation plant or utility plant.

5. Conclusion
A sequential optimization approach is applied to the synthesis and design of water-based polygeneration system as a strategy to address the problem of limited freshwater resources in the chemical process industry. In the first step (MINLP model), superstructure helps in assessing the potential of different design configurations to formulate a comprehensive model for the optimum water-based polygeneration system. The model features all possible configurations of heat and power generations, re-circulating cooling water system, as well as reaction and separation technologies for chemical production. In the next step, the 3R strategy for water conservation is implemented in the NLP model. The application of the model was demonstrated on an ethylene glycol production case study. The solutions derived with this approach are close to the simultaneous solutions because of the close approximation of the system by the shortcut model. The main benefit of sequential optimization is the efficiency in run time.

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