A Mathematical Programming Model for Discontinuous Water-Reuse System Design

Bao-Hong Li
Department of Chemical Engineering, Dalian Nationalities University, Dalian 116600, China

Chuei-Tin Chang*
Department of Chemical Engineering, National Cheng Kung University, Tainan, Taiwan 70101, Republic of China

A general mixed-integer nonlinear programming model (MINLP) is developed in this study for the design of discontinuous water-reuse systems. The proposed model formulation is believed to be more practical than the available models, in the sense that the unrealistic assumptions adopted in previous works have been removed and, also, the wastewater equalization options can be integrated into the system design. The resulting design specifications include the number and sizes of buffer tanks, the physical configuration of pipeline network, and the operating policies of water flows. The network structure of the water-reuse system can also be strategically manipulated by imposing suitable logic constraints. An illustrative example is presented at the end of this paper to demonstrate the effectiveness of our approach.

Introduction

All process plants are committed to improve the utilization efficiency of fresh water. Water reuse is not only a cost-cutting measure, but also an effective means of pollution prevention. In the literature, studies on water reuse have been mainly concerned with the continuous processes, whereas very little attention has been directed toward the development of water conservation strategies in batch processes. The latter issue is, in fact, more challenging, because the needs to consume fresh water and to discharge wastewater occur intermittently in each water user, according to a predetermined schedule. As a result, the unit model of continuous water-using operations is not applicable in the present case. Furthermore, additional auxiliary equipments, i.e., the buffer tanks, for temporarily storing the spent waters must be made available in the batch plants to facilitate reuse. Consequently, it is necessary to develop a rigorous approach to design discontinuous water-reuse systems.

Wang and Smith developed a modified version of the Pinch Method to minimize the amount of wastewater discharged from a batch process. Recently, Kim and Smith constructed a MINLP model to automate the design procedure of such discontinuous water-reuse systems. Although significant contributions have been made in these studies to address various water management issues in batch processes, it should be noted that their results may not be practically feasible, because of several unrealistic assumptions. Specifically, in each water user, the wastewater is required to be generated during the same period when the fresh water is being consumed. In addition, each storage tank is dedicated to a single reuse opportunity. Because none of the tanks are shared to serve multiple functions, the capital cost of the resulting water reuse system has a tendency to be higher than minimum.

On the other hand, Almato and co-workers and Puigjaner et al. developed a more-realistic nonlinear programming (NLP) model to optimize the water-reuse networks. Some of the previously mentioned simplification assumptions have been removed in their studies. However, their unit models are still too simple to adequately describe the water-using operations, and their superstructure is far from comprehensive. In the former case, only one pollutant index is considered in the mass-balance constraints, and the water quality and flow rate of the feed to every unit are not allowed to vary. In the latter case, the sizes of buffer tanks embedded in their superstructure are always fixed in advance. Also, all unusable wastewaters are collected in a single sink, whereas, in practice, some of them should be treated before being discharged into the environment.

McLaughlin et al. indicated that the capital cost of a wastewater treatment operation is usually proportional to its capacity. Thus, for economic reasons, flow equalization is needed to reduce the maximum flow rate of wastewater entering the treatment system. In addition, because the biological-treatment unit is included in most cases, the “shock loads” (mainly in concentration) must be avoided at all times, so that the embedded bacteria can be continuously maintained at the active state. In this situation, a buffer system may also be installed to equalize the wastewater flow rates and pollutant concentrations simultaneously. Although the design issues of a stand-alone equalization network has been fully addressed by Chang and Li, its interactions with the reuse system previously have never been analyzed.

Notice also that more than one water-using operation may be scheduled to be performed with the same equipment in different time intervals during a batch production cycle. It is thus conceivable that the actual configuration of the water-reuse network may be established and manipulated by incorporating extra structural constraints in the optimization procedure. Because this concept has never been fully exploited previously, the development of a systematic implementation procedure becomes necessary.

In the present paper, a general-purpose mathematical programming model is proposed for the design of integrated water-reuse and equalization systems in the batch processes. All of the limitations previously mentioned have been removed in this model. To illustrate the proposed approach, the rest of this paper...
The sets of all possible sources and sinks of a water-reuse system can be represented, respectively, as

\[
S = SA \cup SB
\]

(6)

\[
O = OA \cup OB \cup OC
\]

(7)

If a water-using operation both consumes and generates waters (e.g., equipment washing), it is referenced, in this paper, as a water user. Such operations are grouped into a set \( U \), i.e.,

\[
U = \{u|u \text{ is the label of a water user}\}
\]

(8)

This set can be divided into two subsets, according to the charging and discharging periods. In particular,

\[
UA = \{ua|ua \text{ is the label of a water user with distinct charging and discharging time intervals}\}
\]

(9)

\[
UB = \{ub|ub \text{ is the label of a water user with identical charging and discharging time intervals}\}
\]

(10)

All batch operations in the water-reuse system are assumed to be conducted in equipment that is defined below:

\[
E = \{e|e \text{ is the label of equipment that facilitates at least one operation defined in } U, SB, \text{ or } OC\}
\]

(11)

Furthermore, all operations implemented with a particular equipment \( e \in E \) can be represented as

\[
P_e = \{p|p \text{ is the label of an operation conducted in equipment } e \in E\}
\]

(12)

Notice that

\[
\bigcup_{e \in E} P_e = U \cup SB \cup OC
\]

(13)

As mentioned previously, a set of buffer tanks are needed to provide opportunities for water reuse and to equalize the flow rates and concentrations of wastewaters before entering the treatment systems. These tanks can be represented in another unit set that is defined as follows:

\[
B = \{b|b \text{ is the label of a buffer tank}\}
\]

(14)

Based on the aforementioned definitions, the design task in this study can be considered to be one of synthesizing a cost-effective water-reuse system and its operating policy to achieve the given water consumption and generation schedule. The physical structure of the water network can be assembled by connecting the given sources in set \( SA \), the given sinks in sets \( OA \) and \( OB \), the given equipment in set \( E \), and also a set of unspecified buffer tanks in set \( B \). It is also assumed that the following additional parameters are available: (i) the qualities and maximum supply rates of the fresh waters from the sources in set \( SA \), and the qualities and nominal flow rates of the secondary waters from the sources in set \( SB \); (ii) the upper and/or lower limits of the flow rate and pollutant indices in the wastewater stream discharged to each sink in set \( O \); (iii) the maximum allowable water consumption and generation rates of each water user in set \( U \), and the corresponding time intervals in which the charging and discharging operations occur; and
(iv) additional model parameters of each water user (i.e., the maximum pollutant indices allowed at the inlet and outlet, the accumulated mass loads of pollutants, and the total volume of water loss in every operation).

A proper design of the batch water-reuse system should at least include the following specifications: (i) the number of buffer tanks and their sizes, (ii) the network configuration, and (iii) the operating policy (i.e., the time profile of the flow rate in every pipeline of the reuse network).

Finally, note that the following two assumptions have been adopted in this work to simplify the network structure and also to enhance the operability of the water-reuse system:

(1) Any self-looping branch, either around a water-using operation or around a buffer tank, is forbidden; and

(2) All reuse water streams are taken from buffer tanks; i.e., a direct connection between any two water-using operations is not allowed.

The first assumption is adopted on the grounds that the self-looping design is seldom useful in practical applications. Note that the opportunities of direct water reuse arise only when multiple water-using operations are scheduled to be performed within the same time interval. Such opportunities are rare in realistic processes. In addition, notice that two parallel water-using operations are inevitably coupled if the wastewater generated by one is required to be directly consumed by the other. In other words, an external disturbance introduced to the former operation may propagate further to the latter. Because the operation flexibility of the water-reuse system can be greatly reduced by allowing such design options, it is our belief that the introduction of assumption 2 is also justifiable.

**Superstructure**

Similar to other optimization studies in process synthesis, a superstructure that is embedded with all possible flow configurations is built via the following steps:

(1) Place a mixing node at the inlet of each water user, buffer tank, and sink;

(2) Place a splitting node at outlet of each source, water user, and buffer tank;

(3) Connect the split branches from the splitting node after each source in \( S_A \) to all mixing nodes;

(4) Connect the split branches from the splitting node after every source in \( S_B \) and every water user in \( U \) to the mixing nodes before the buffer tanks in \( B \) and sinks in \( O_A \) and \( O_B \), but not to those before the water users in \( U \) and the sinks in \( O_C \); and

(5) Connect the split branches from the splitting node after each buffer tank in \( B \) to all mixing nodes except the one before itself.

The resulting superstructure is presented in Figure 2, in which \( S \) and \( M \) represent the splitting and mixing nodes, respectively. Notice that all other symbols have already been defined in the previous section.

To facilitate a concise model formulation, additional sets of the splitting and mixing nodes should be constructed accordingly, i.e.,

\[
SP = S \cup U \cup B \tag{15}
\]

\[
MX = U \cup B \cup O \tag{16}
\]

In these equations, \( SP \) is set of all splitting nodes and \( MX \) is set of all mixing nodes. Let us next label the sets of starting and ending nodes of the branches produced in steps 3, 4, and 5 as \( SP_{i} \) and \( MX_{i} (i = 1, 2, 3) \), respectively. They are assigned according to the following rules:

(a) In step 3, let \( SP_1 = S_A \) and \( MX_1 = MX \);

(b) In step 4, let \( SP_2 = S_B \cup U \) and \( MX_2 = B \cup O_A \cup O_B \);

(c) In step 5, let \( SP_3 = B \) and \( MX_3 = MX \).

Finally, notice that

\[
\bigcup_{i=1}^{3} SP_i = SP \tag{17}
\]

**Mathematical Programming Model**

For formulation convenience, the following set of all pollutant indices is now introduced:

\[
K = \{k | k \text{ is the label of a pollutant index}\} \tag{18}
\]

To account for the dynamic behaviors of batch systems in the mathematical programming model, it is necessary to divide the entire period of a production cycle (\( H \)) into a finite number of time intervals with constant duration (\( DT \)). To simplify the model notation, it is assumed that the starting and ending times of all water charging and discharging operations steps in the water-reuse system coincide with the interval boundaries.\(^{15}\) Thus, a set of discretized time intervals can be defined as

\[
T = \{t | t \text{ is the label of a discretized time interval}; \quad t = 1, 2, ..., N_T\} \tag{19}
\]

Here,

\[
N_T = \frac{H}{DT} \tag{20}
\]

and the time interval \( t \) is \( \{t(t - 1) \times DT, t \times DT\} \).

Based on the aforementioned definitions, the constraints of mathematical programming model can be formulated as follows.

**Water Sources**

There are two types of water sources in the proposed model. The external freshwater from a source in set \( S_A \) is assumed to be available at any time under the constraint of an upper flow limit. It is also assumed that the secondary water from a source in set \( S_B \) must be consumed completely in the water-reuse system. Thus, the flow constraint on a water source in set \( S_A \) can be expressed as
\[ f_{\text{source},s,t} \leq F_{ua} \quad (sa \in \text{SA}, \ t \in \text{T}) \] (21)

where \( f_{\text{source},s,t} \) represents the flow rate of freshwater from source \( sa \) during time interval \( t \); \( F_{ua} \) is the maximum allowable supply rate of fresh water from source \( sa \). On the other hand, the flow constraint on a water source in set \( \text{SB} \) can be written as

\[ f_{\text{source},s,b,t} = F_{sb,t} \quad (sb \in \text{SB}, \ t \in \text{T}) \] (22)

Here, \( F_{sb,t} \) is the nominal supply rate of secondary water from source \( sb \) during time interval \( t \). On the other hand, the water qualities of the fresh waters and secondary waters should be considered as known data, i.e.,

\[ c_{s,k,t} = C_{s,k,t} \quad (s \in \text{S}, \ k \in \text{K}, \ t \in \text{T}) \] (23)

where \( c_{s,k,t} \) denotes the pollution index \( k \) in the water stream from source \( s \) during time interval \( t \); \( C_{s,k,t} \) is a given parameter.

The material-balance constraint around the splitting node after each water source in set \( \text{SA} \) can be formulated as

\[ f_{\text{source},s,a,t} = \sum_{m \in \text{MX}} f_{s,a,m,t} \quad (sa \in \text{SA}, \ t \in \text{T}) \] (24)

in which \( f_{s,a,m,t} \) denotes the flow rate in the split branch from source \( sa \) to mixing node \( mx \). On the other hand, the mass-balance equations for the sources in set \( \text{SB} \) should be written in a slightly different form:

\[ f_{\text{source},s,b,t} = \sum_{b \in \text{B}} f_{sb,b,t} + \sum_{o \in \text{OA}} f_{sb,o,a,t} + \sum_{o \in \text{OA}} f_{sb,o,b,t} \quad (sb \in \text{SB}, \ t \in \text{T}) \] (25)

Finally, notice that the pollution indices in the water streams before and after a splitting node should be identical, i.e.,

\[ c_{s,k,t} = c_{s,k,t} \quad (s \in \text{S}, \ k \in \text{K}, \ t \in \text{T}) \] (26)

where \( c_{s,k,t} \) represents the pollution index \( k \) in all split branches from the splitting node after source \( s \) during time interval \( t \).

**Water Users**

First of all, the overall mass balances must be established for all water users. They can be written as

\[ DT \sum_{u \in \text{UA}_u} f_{u,a,t} = DT \sum_{u \in \text{UA}_u} f_{u,a,t} - DV_u \quad (u \in \text{U}) \] (27)

Here, \( DV_u \) represents the total water loss in the operation of water user \( u \); \( f_{u,a,t} \) and \( f_{u,a,b,t} \) respectively denote the input and output flow rates of water user \( u \) during interval \( t \); \( \text{TC}_u \) denotes the set of time intervals in which the charging operation of water user \( u \) occurs; \( \text{TD}_u \) is the set of time intervals in which the discharging operation of water user \( u \) occurs.

As mentioned previously, the water users can be classified into two sets, i.e., \( \text{UA} \) and \( \text{UB} \). The component balances for water users in set \( \text{UA} \) can be expressed as

\[ DT \sum_{u \in \text{UA}_u} f_{u,a,t}^{\text{in}} + c_{u,a,k,t} = DT \sum_{t \in \text{TD}_u} f_{u,a,b,t}^{\text{out}} + M_{u,a,k} = \] (28)

where \( c_{u,a,k,t} \) denotes the constant pollutant index \( k \) in the wastewater stream of water user \( ua \) during time interval \( t \); \( c_{u,a,k,t}^{\text{out}} \) denotes the constant pollutant index \( k \) in the wastewater stream of water user \( ua \) during time interval \( t' \); \( M_{u,a,k} \) is a given parameter that represents the accumulated mass load of pollutant index \( k \) in operation \( ua \).

For each water user in set \( \text{UB} \), the water-using operation is treated as a continuous one. Thus, the conventional unit model is used to express the mass balances in this case, i.e.,

\[ f_{u,b,t}^{\text{out}} + c_{u,b,k,t}^{\text{out}} = f_{u,b,t}^{\text{in}} + c_{u,b,k,t}^{\text{in}} + \mu_{u,b,k} \quad (ub \in \text{UB}, \ k \in \text{K}, \ t \in \text{T}_u) \] (29)

where \( \mu_{u,b,k} \) is the instantaneous mass load of pollutant index \( k \) in water user \( ub \). Notice that, because the charging and discharging periods are identical in this case, the set \( \text{T}_u \) in the aforementioned equation is

\[ \text{T}_u = \text{TC}_u = \text{TD}_u \] (30)

For operation simplicity, it is desirable to keep the charging and discharging rates of every water user constant. Therefore, the following additional constraints are also imposed in the proposed model:

\[ f_{u,a,t}^{\text{in}} = f_{u,a,t}^{\text{out}} = 0 \quad (u \in \text{U}, \ t \in \text{TC}_u, \ t' \in \text{TD}_u) \] (32)

For each water user in \( \text{UA} \), it is assumed that the pollutant indices in the generated wastewater are maintained at approximately constant levels, i.e.,

\[ c_{u,a,k,t}^{\text{out}} \approx c_{u,a,k} \quad (ua \in \text{UA}, \ k \in \text{K}, \ t \in \text{T}_u) \] (33)

In this equation, \( c_{u,a,k} \) represents pollutant index \( k \) in the wastewater stream generated from operation \( ua \) during time interval \( t \); \( c_{u,a,k,t}^{\text{out}} \) is the constant pollutant index \( k \) in the wastewater generated from operation \( ua \).

The mass balances around the mixing node before each water user can be written as

\[ f_{u,a,t}^{\text{in}} = \sum_{sa \in \text{SA}} f_{sa,a,u,t} + \sum_{b \in \text{B}} f_{sb,a,u,t} \quad (u \in \text{U}, \ t \in \text{TC}_u) \] (34)

\[ f_{u,a,t}^{\text{in}} = \sum_{sa \in \text{SA}} f_{sa,a,u,t} + \sum_{b \in \text{B}} f_{sb,a,u,t} + \sum_{s \in \text{SA}} f_{s,a,u,t} \quad (u \in \text{U}, \ t \in \text{TC}_u) \] (35)

where \( f_{sa,a,u,t} \) and \( f_{sb,a,u,t} \) respectively represent the flow rates of split branches from source \( sa \) and tank \( b \) to water user \( u \) during interval \( t \); \( c_{s,a,k,t} \) and \( c_{b,k,t} \) represent the values of the constant pollutant index \( k \) in the split branches from the splitting nodes after source \( sa \) and tank \( b \), respectively.
On the other hand, the material-balance constraints around the splitting node after every water user can be expressed as

\[ f_{u,t}^{\text{out}} = \sum_{b \in B} f_{u,b,t}^s + \sum_{\text{o w}} f_{u,o,t}^s + \sum_{\text{o b}} f_{u,o,b,t} (u \in U, k \in K, t \in T_{D}) \]  

\[ c_{u,k,t}^{\text{out}} = c_{u,k,t} (u \in U, k \in K, t \in T_{D}) \]  

The pollutant indices at the inlets and outlets of water users may be subject to additional inequality constraints to maintain operation efficiency, i.e.,

\[ c_{u,k,t}^{\text{in}} \leq \eta_{u,k}^{\text{in}} (t \in T, u \in U, k \in K) \]  

\[ c_{u,k,t}^{\text{out}} \leq \eta_{u,k}^{\text{out}} (t \in T, u \in U, k \in K) \]  

where the upper bounds \( \eta_{u,k}^{\text{in}} \) and \( \eta_{u,k}^{\text{out}} \) are given parameters.

**Buffer Tanks**

The buffer tanks are used for temporarily storing and mixing wastewaters. The mass-balance relations can be written as

\[ v_{b,t} = v_{b,t-1} + (f_{b,t}^{\text{in}} - f_{b,t}^{\text{out}}) DT (b \in B, t \in T) \]  

\[ v_{b,b,k,t} = v_{b,t-1} + (f_{b,t}^{\text{in}} c_{b,k,t}^{\text{in}} - f_{b,t}^{\text{out}} c_{b,k,t}^{\text{out}}) DT (b \in B, k \in K, t \in T) \]  

where, \( v_{b,t} \) represents the water volume in tank \( b \) at the end of time interval \( t \); \( c_{b,k,t}^{\text{in}} \) and \( c_{b,k,t}^{\text{out}} \) respectively denote the values of the pollutant index \( k \) in the inlet and outlet streams of tank \( b \) at the end of interval \( t \); \( f_{b,t}^{\text{in}} \) and \( f_{b,t}^{\text{out}} \) respectively represent the flow rates of the input and output streams of tank \( b \) during time interval \( t \).

Similar to the water users, every buffer tank in the super-structure is equipped with a mixing node in front and a splitting node at its back. The material-balance constraints around the mixing nodes are

\[ f_{b,t}^{\text{in}} = \sum_{\text{s p}} f_{b,s,p,t}^s (b \in B, t \in T) \]  

\[ f_{b,k,t}^{\text{in}} c_{b,k,t}^{\text{in}} = \sum_{\text{s p}} f_{b,s,p,k,t}^s (b \in B, k \in K, t \in T) \]  

For the splitting node after each buffer tank, the material-balance equations can be written as

\[ f_{b,t}^{\text{out}} = \sum_{\text{m x}} f_{b,m,x,t}^s (b \in B, t \in T) \]  

\[ c_{b,k,t}^{\text{out}} = c_{b,k,t} (b \in B, k \in K, t \in T) \]  

The total volume of water in a buffer tank at any instance should, of course, be larger than zero and, also, less than the storage capacity. These constraints can be expressed as

\[ v_{b}^{\text{max}} \geq v_{b,t} \geq 0 \quad (b \in B, t \in T) \]  

in which \( v_{b}^{\text{max}} \) is the needed size of tank \( b \).

Finally, because of the cyclic nature of batch production activities, it is assumed in this study that the operating conditions at the end of each cycle are the same as the initial conditions of the next cycle. In other words, the following constraints must also be imposed:

\[ v_{b,0} = v_{b,N} \quad \text{(b in B)} \]  

\[ e_{b,k,0} = e_{b,k,N} \quad \text{(b in B, k in K)} \]  

**Water Sinks**

All wastewaters that cannot be reused should be discharged into the water sinks. As mentioned previously, there are three types of water sinks in our model. The pollutant indices and flow rates of wastewaters sent to the sinks in set \( OA \) should comply with government regulations. On the other hand, the flow rates and pollutant indices of wastewaters are usually required to vary within a set of predetermined upper and/or lower limits to run the treatment systems in set \( OB \) smoothly. Finally, it is assumed in this study that the flow rate of the water stream supplied to every water-using operation in set \( OC \) must be controlled at the specified time-variant nominal value and, also, the corresponding pollution indices must be maintained below the given maximum allowable levels.

The material-balance constraints around the mixing nodes before the sinks in sets \( OA \) and \( OB \) can be written in a uniform format as

\[ f_{a,t}^{\text{sink}} = \sum_{\text{s p}} f_{a,s,p,t}^s (a \in OA, b \in OB, t \in T) \]  

\[ f_{a,t}^{\text{sink}} c_{a,k,t}^{\text{sink}} = \sum_{\text{s p}} f_{a,s,p,k,t}^s c_{a,s,p,k,t}^{\text{sink}} (a \in OA, b \in OB, k \in K, t \in T) \]  

Here, \( f_{a,t}^{\text{sink}} \) and \( c_{a,k,t}^{\text{sink}} \) respectively denote the flow rate and pollutant index \( k \) of wastewater discharged to sink \( a \) during time interval \( t \). The corresponding constraints for the sinks in \( OC \) should be

\[ f_{a,t}^{\text{sink}} = \sum_{\text{a c}} f_{a,s,a,c,t}^s + \sum_{b \in B} f_{b,o,c,t}^s (a \in OC, t \in T) \]  

\[ f_{a,t}^{\text{sink}} c_{a,k,t}^{\text{sink}} = \sum_{\text{a c}} f_{a,s,a,c,k,t}^s c_{a,s,a,c,k,t}^{\text{sink}} + \sum_{b \in B} f_{b,o,c,k,t}^s c_{b,o,c,k,t}^{\text{sink}} (a \in OC, k \in K, t \in T) \]  

The upper and lower bounds of flow rates and pollutant indices in the wastewaters discharged to the sinks in \( OA \) and \( OB \) can be expressed in the following general forms:

\[ \lambda_{a}^{\text{min}} \leq f_{a,t}^{\text{sink}} \leq \lambda_{a}^{\text{max}} \quad (a \in OA, b \in OB, t \in T) \]  

\[ \theta_{a,k}^{\text{min}} \leq c_{a,k,t}^{\text{sink}} \leq \theta_{a,k}^{\text{max}} \quad (a \in OA, b \in OB, k \in K, t \in T) \]  

where \( \lambda_{a}^{\text{min}} \) and \( \lambda_{a}^{\text{max}} \) respectively represent the minimum and maximum flows rate allowed by sink \( a \); \( \theta_{a,k}^{\text{min}} \) and \( \theta_{a,k}^{\text{max}} \) denote the corresponding minimum and maximum values of pollutant index \( k \). For the sinks in \( OA \), \( \lambda_{a}^{\text{min}} = 0 \) and \( \theta_{a,k}^{\text{min}} = 0 \). On the other hand, larger-than-zero upper and lower bounds are used in these inequality constraints for sinks in \( OB \).

Because the flow rates of wastewater streams delivered to the sinks in \( OC \) are required to be maintained at time-variant nominal levels, a set of equality constraints should be used accordingly, i.e.,

\[ f_{a,t}^{\text{sink}} = \lambda_{a,t} (a \in OC, t \in T) \]
where \( \lambda_{oc} \) denotes the nominal flow rate required by sink \( oc \) in interval \( t \). Finally, the constraints imposed upon the pollution indices in the wastewater streams discharged to the sinks in \( OC \) should be the same as those adopted for the sinks in \( OA \).

### Structural Constraints

From a practical standpoint, there is an obvious need to eliminate any branch that is used to transfer only a negligible amount of water during the entire production cycle. To prevent eliminating any branch that is used to transfer only a negligible amount of water during the entire production cycle, \( y_{sp,mx,il} \) must be limited to a level that is achievable with commercially available equipment. The corresponding inequality constraint can be written as

\[
\sum_{i \in T} f_{sp,mx,il} \geq y_{sp,mx,il} L^B (sp \in SP, mx \in MX, sp \neq mx)
\]

(54)

where \( i = 1, 2, 3; L^B \) denotes a user-specified lower bound of the total transported water volume during any branch during a production cycle; \( y_{sp,mx,il} \in \{0, 1\} \) signifies whether the corresponding branch is selected in the optimal network configuration. On the other hand, the maximum water flow rate in each branch must also be limited to a level that is achievable with commercially available equipment. The corresponding inequality constraint can be written as

\[
f_{sp,mx,il} \leq y_{sp,mx,il} U^F (sp \in SP, mx \in MX, sp \neq mx, t \in T)
\]

(55)

Here, \( i = 1, 2, 3; U^F \) is the upper bound of flow rate that must not be exceeded in each branch. If \( y_{sp,mx,il} = 0 \), eqs 54 and 55 will force the flow rate of \( f_{sp,mx,il} \) to be zero at any time interval \( t \). Notice that the aforementioned two constraints are imposed only on the existing branches in the superstructure, i.e., the non-self-looping branches from the splitting nodes in \( SP \) to the mixing nodes in \( MX \), and \( i = 1, 2, 3 \). The flow rates and the corresponding binary variables of the remaining connection branches between the nodes in \( SP \) and \( MX \) are all set to zero in the proposed model.

The number of buffer tanks embedded in the superstructure is usually larger than that which is actually needed in the optimal solution. To remove the unusually small tanks in system design, it is often necessary to impose a lower bound on the tank size, i.e.,

\[
x_b V^k \leq v^\text{max}_b (b \in B)
\]

(56)

where \( x_b \in \{0, 1\} \) is used to indicate if tank \( b \) exists and \( V^k \) is the lower limit of the volume of each tank. If buffer tank \( b \) does not exist, i.e., \( x_b = 0 \), the connecting branches in the superstructure should be eliminated completely. This logic operation can be realized with the following inequality constraints:

\[
x_p \geq y_{b,mx} (b \in B, mx \in MX, b \neq mx)
\]

(57)

\[
x_b \geq y_{sp,b} (b \in B, sp \in SP, b \neq sp)
\]

(58)

As mentioned previously, more than one water-using operation may be performed with the same equipment. In other words, the superstructure used in this study is, in fact, a fictitious process configuration. Therefore, additional structural constraints must be incorporated in the mathematical programming model to translate the optimal solution into the actual pipeline network of the resulting system design automatically.

Notice first that, if a branch in the superstructure is not used to facilitate the operation of a water user, it can be regarded as a physical pipeline. This feature can be characterized as

\[
z_{ij} = y_{ij} \quad (i \in SA \cup B, j \in B \cup OA \cup OB)
\]

(59)

where \( z_{ij} \in \{0, 1\} \) \((i \neq j)\) is used to signify the existence of a pipeline from \( i \) to \( j \). The binary values associated with the remaining connection branches can be interpreted according to the logic operators suggested by Raman and Grossmann, i.e.,

\[
z_{e}^e \leq \sum_{p \in P_e} y_{e,p} \quad (e' \in SA \cup B, e \in E)
\]

(60)

\[
z_{e}^e \geq y_{e,p} \quad (e' \in SA \cup B, e \in E, p \in P_e)
\]

(61)

\[
z_{e}^e \leq \sum_{p \in P_e} y_{e,p} \quad (e \in E, e' \in B \cup OA \cup OB)
\]

(62)

\[
z_{e}^e \geq y_{e,p} \quad (e \in E, e' \in B \cup OA \cup OB, p \in P_e)
\]

(63)

To simplify the network structure, it is also desirable to limit the numbers of pipelines attached to the mixing and splitting points in the actual pipeline network. These inequality constraints can be written in the following forms:

\[
\sum_{i \in IA} z_{i,a,e} + \sum_{b \in B} z_{b,e} \leq NM_e \quad (e \in E)
\]

(64)

\[
\sum_{b \in B} z_{b,e} + \sum_{o \in OA} z_{e,o,a} + \sum_{b \in B} z_{e,ob} \leq NS_e \quad (e \in E)
\]

(65)

\[
\sum_{i \in IA} z_{i,a,b} + \sum_{b \in B} z_{b,b'} + \sum_{o \in OA} z_{a,b,o} + \sum_{b \in B} z_{b,ob} \leq NM_b \quad (b \in B)
\]

(66)

\[
\sum_{e \in E} z_{b,e} + \sum_{b \in B} z_{b,b'} + \sum_{o \in OA} z_{a,b,o} + \sum_{b \in B} z_{b,ob} \leq NS_b \quad (b \in B)
\]

(67)

\[
\sum_{i \in IA} z_{i,a,e} + \sum_{b \in B} z_{b,b'} + \sum_{o \in OA} z_{a,b,o} + \sum_{b \in B} z_{b,ob} \leq NS_{ab}
\]

(68)

\[
\sum_{i \in IA} z_{i,a,e} + \sum_{b \in B} z_{b,b'} + \sum_{o \in OA} z_{a,b,o} + \sum_{b \in B} z_{b,ob} \leq NS_{oa}
\]

(69)

where \( NM_e \) and \( NS_e \) denote the upper bounds of the pipeline numbers connected to the mixing point and splitting point of equipment \( e \), respectively. Notice that the other upper bounds have a similar meaning.

### Objective Function

The objective function \( \text{obj} \) of our optimization problem is the sum of annual water cost, annualized installation cost, and annual treatment cost, i.e.,

\[
\text{obj} = \Gamma_{SA} \Phi_{SA} + \Gamma_{B} \Phi_{B} + \Gamma_{OB} \Phi_{OB}
\]

(70)

where \( \Gamma_{SA}, \Gamma_{B}, \) and \( \Gamma_{OB} \) assume only binary values. They are used in this formulation to account for various different combinations of objective functions used in practical applications. \( \Phi_{SA} \) represents the annual freshwater cost from the
sources in SA; \( \Phi _{B} \) and \( \Phi _{OB} \) denote the annualized installation cost of buffer tanks and annual treatment cost, respectively.

The annual water and treatment costs are calculated with the following formulas:

\[
\Phi _{SA} = N_{cycle} DT \sum _{sa} \omega _{sa} \left( \sum _{t \in T} u_{sa,t} \right) \\
\Phi _{OB} = N_{cycle} DT \sum _{ob} \omega _{ob} \left( \sum _{t \in T} u_{ob,t} \right)
\]

where \( N_{cycle} \) is the number of production cycles performed per year; \( \omega _{sa} \) is the raw-material cost per unit volume of consumed fresh water from source \( sa \); \( \omega _{ob} \) is the treatment cost per unit volume of wastewater discharged to sink \( ob \). The annualized installation cost of buffer tanks is determined according to

\[
\Phi _{B} = \sum _{b} \left[ \alpha _{b} x_{b} + \beta _{b} \left( u_{b}^{max} \right)^{0.6} \right]
\]

where \( \alpha _{b} \) is the fixed charge and \( \beta _{b} \) is the cost coefficient of tank \( b \).

Finally, note that the bilinear terms in constraints and exponential terms in the objective function of the aforementioned mixed-integer nonlinear programming (MINLP) formulation inevitably cause nonconvexities and local optima. The development of an efficient search algorithm for the true optimum is not the focus of present study; therefore, the optimal solution of a given design problem was obtained by solving the proposed model more than once with randomly selected initial guesses.

### An Illustrative Example

The optimal solutions of the example problem in Figure 1 are presented here to illustrate the implementation procedure of the proposed approach. The process data of all water-using operations in this system can be found in Table 1. The labels of every operation and the corresponding equipment are listed in columns 1 and 2, respectively. The limiting water flow rates at its inlet and/or outlet are provided in column 3; a negative flow rate implies that the corresponding water stream is consumed in the operation, and a positive value represents the wastewater generation rate. For a water user, i.e., \( u_{1} \), \( u_{2} \), \( u_{3} \), or \( u_{4} \), they represent the water flow rates required to maintain the maximum allowable inlet and outlet concentrations. Note that, although these four operations must be executed according to the given schedules, their input and output flow rates in the final design are not required to be the same as the limiting rates. The optimal flow rates to and from a water user are allowed to vary, as long as the given amount of accumulated mass load can be removed during operation. On the other hand, because operations \( oc_{1} \) and \( sb_{1} \) are treated in this example as the water sink and source, respectively, it is meaningless to specify their mass loads. In such cases, the water consumption rate of \( oc_{1} \) and the water generation rate of \( sb_{1} \) should be kept identical to the limiting flow rates listed in Table 1. Notice that \( u_{4} \) can be classified as a water-using operation in set UA, whereas the other water users can be considered as members in set UB. It can also be observed that there are two pollutants \( (k_{1} \) and \( k_{2} \)) in the water streams. The maximum pollutant concentrations in the inlet and/or outlet streams of every water-using operation are specified in columns 6 and 7 of Table 1, and the total water loss and accumulated mass loads are provided in the last three columns in the table.

It is assumed, in this example, that a complete production cycle lasts 20 h. The durations of the charging and/or discharging steps in a cycle are specified in the fourth and fifth columns of Table 1, where \( T_{e} \) and \( T_{c} \) respectively denote the starting and ending times of an operation step. Thus, the length of each fixed time interval used in the mathematical model (DT) must be equal to 0.5/\( n \) hours (where \( n \) is a positive integer). It is obvious that, if a larger value of \( n \) is selected, the solution will be more accurate but the computation that is needed will be more expensive. In the present study, the appropriate interval size is determined on a trial-and-error basis by solving the MINLP model repeatedly, according to decreasing DT values. The final network design should not vary if the selected interval number is increased further.

In this example, only one external water source is considered (that is, \( sa_{1} \)) and its fresh water is assumed to be free from any pollutant. It is also assumed that the maximum water supply rate from source \( sa_{1} \) is 15 m\(^3\)/h and its cost is 1 U.S. dollar/m\(^3\). On the other hand, we assumed that only one wastewater treatment system (\( ob_{1} \)) is available and the corresponding treatment cost is 2 US dollars/m\(^3\). Furthermore, the flow rate of wastewater stream discharged to this sink is required to be maintained within a range of 1–4 m\(^3\)/h, and the corresponding concentrations of \( k_{1} \) and \( k_{2} \) must be kept within the concentration intervals of 10–20 mg/L and 10–25 mg/L, respectively. An additional environment sink \( oa_{1} \) is also assumed to be available in this example. The upper bounds of its inlet pollutant concentrations are both set to be 2 mg/L. Other assumptions adopted in this example are listed in the following:

1. In the cost model of buffer tank, the fixed charge is 48000 U.S. dollars and the cost coefficient is 280 000 U.S. dollars/m\(^3\);
2. The annual depreciation rate is 10%;
3. The plant operates 7200 h per year;

<table>
<thead>
<tr>
<th>operation</th>
<th>equipment</th>
<th>limiting flow rate (m(^3)/h)</th>
<th>Duration (h)</th>
<th>Maximum Concentration (mg/L)</th>
<th>water loss (m(^3))</th>
<th>Accumulated Mass Load (kg)</th>
</tr>
</thead>
<tbody>
<tr>
<td>( u_{1} )</td>
<td>( e_{1} )</td>
<td>10</td>
<td>5.0</td>
<td>0.0</td>
<td>0.0</td>
<td>0.100</td>
</tr>
<tr>
<td>( u_{1} )</td>
<td>( e_{1} )</td>
<td>10</td>
<td>2.5</td>
<td>0.0</td>
<td>0.0</td>
<td>0.160</td>
</tr>
<tr>
<td>( u_{2} )</td>
<td>( e_{2} )</td>
<td>10</td>
<td>5.0</td>
<td>0.0</td>
<td>0.0</td>
<td>0.160</td>
</tr>
<tr>
<td>( u_{2} )</td>
<td>( e_{2} )</td>
<td>10</td>
<td>2.5</td>
<td>0.0</td>
<td>0.0</td>
<td>0.160</td>
</tr>
<tr>
<td>( u_{3} )</td>
<td>( e_{3} )</td>
<td>10</td>
<td>5.0</td>
<td>0.0</td>
<td>0.0</td>
<td>0.160</td>
</tr>
<tr>
<td>( u_{3} )</td>
<td>( e_{3} )</td>
<td>10</td>
<td>2.5</td>
<td>0.0</td>
<td>0.0</td>
<td>0.160</td>
</tr>
<tr>
<td>( u_{4} )</td>
<td>( e_{3} )</td>
<td>10</td>
<td>5.0</td>
<td>0.0</td>
<td>0.0</td>
<td>0.160</td>
</tr>
<tr>
<td>( u_{4} )</td>
<td>( e_{3} )</td>
<td>10</td>
<td>2.5</td>
<td>0.0</td>
<td>0.0</td>
<td>0.160</td>
</tr>
<tr>
<td>( oc_{1} )</td>
<td>( e_{1} )</td>
<td>10</td>
<td>5.0</td>
<td>0.0</td>
<td>0.0</td>
<td>0.160</td>
</tr>
<tr>
<td>( sb_{1} )</td>
<td>( e_{2} )</td>
<td>10</td>
<td>5.0</td>
<td>0.0</td>
<td>0.0</td>
<td>0.160</td>
</tr>
</tbody>
</table>

Table 1. Process Data of Example 1
Table 2. Comparison of the Results Obtained in Three Case Studies

<table>
<thead>
<tr>
<th></th>
<th>case 1</th>
<th>case 2</th>
<th>case 3</th>
</tr>
</thead>
<tbody>
<tr>
<td>objective function ($\times 10^3$ U.S. dollars/yr)</td>
<td>245.05</td>
<td>264.38</td>
<td>267.91</td>
</tr>
<tr>
<td>freshwater cost ($\times 10^3$ U.S. dollars/yr)</td>
<td>17.33</td>
<td>20.89</td>
<td>22.02</td>
</tr>
<tr>
<td>treatment cost ($\times 10^3$ U.S. dollars/yr)</td>
<td>26.02</td>
<td>33.15</td>
<td>35.41</td>
</tr>
<tr>
<td>installation cost ($\times 10^3$ U.S. dollars/yr)</td>
<td>201.70</td>
<td>210.34</td>
<td>210.48</td>
</tr>
<tr>
<td>number of selected tanks</td>
<td>2</td>
<td>2</td>
<td>2</td>
</tr>
<tr>
<td>number of branches</td>
<td>18</td>
<td>16</td>
<td>12</td>
</tr>
<tr>
<td>number of pipelines</td>
<td>15</td>
<td>11</td>
<td>8</td>
</tr>
<tr>
<td>total amount of consumed $sa_1$ (m$^3$)</td>
<td>48.13</td>
<td>58.04</td>
<td>61.18</td>
</tr>
<tr>
<td>volume of tank $b_1$ (m$^3$)</td>
<td>18.50</td>
<td>14.00</td>
<td>19.19</td>
</tr>
<tr>
<td>volume of tank $b_2$ (m$^3$)</td>
<td>1.18</td>
<td>4.00</td>
<td>1.53</td>
</tr>
<tr>
<td>CPU time (s)</td>
<td>12.00</td>
<td>15.48</td>
<td>12.44</td>
</tr>
</tbody>
</table>

Table 3. Design Conditions of the Water-Using Operations in Case 1

<table>
<thead>
<tr>
<th>operation</th>
<th>flow rate (m$^3$/h)</th>
<th>Concentration (mg/L)</th>
<th>from/to</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>$k_1$</td>
<td>$k_2$</td>
</tr>
<tr>
<td>$u_1$</td>
<td>$-10.0$</td>
<td>0.0</td>
<td>0.0</td>
</tr>
<tr>
<td>$u_1$</td>
<td>10.0</td>
<td>5.0</td>
<td>8.0</td>
</tr>
<tr>
<td>$u_2$</td>
<td>$-8.2$</td>
<td>$\leq 5.0$</td>
<td>$\leq 8.0$</td>
</tr>
<tr>
<td>$u_2$</td>
<td>8.2</td>
<td>14.0</td>
<td>10.4</td>
</tr>
<tr>
<td>$u_3$</td>
<td>$-6.1$</td>
<td>$\leq 13.5$</td>
<td>$\leq 11.5$</td>
</tr>
<tr>
<td>$u_3$</td>
<td>6.1</td>
<td>20.0</td>
<td>26.7</td>
</tr>
<tr>
<td>$w_4$</td>
<td>$-8.0$</td>
<td>$\leq 5.0$</td>
<td>$\leq 6.2$</td>
</tr>
<tr>
<td>$w_4$</td>
<td>4.0</td>
<td>$\leq 25.0$</td>
<td>$\leq 26.4$</td>
</tr>
<tr>
<td>$oc_1$</td>
<td>$-10.0$</td>
<td>$\leq 6.4$</td>
<td>$\leq 4.8$</td>
</tr>
<tr>
<td>$sb_1$</td>
<td>4.0</td>
<td>10.0</td>
<td>13.0</td>
</tr>
</tbody>
</table>

Table 4. Design Conditions of the Water-Using Operations in Case 2

<table>
<thead>
<tr>
<th>operation</th>
<th>flow rate (m$^3$/h)</th>
<th>Concentration (mg/L)</th>
<th>from/to</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>$k_1$</td>
<td>$k_2$</td>
</tr>
<tr>
<td>$u_1$</td>
<td>$-10.0$</td>
<td>0.0</td>
<td>0.0</td>
</tr>
<tr>
<td>$u_1$</td>
<td>10.0</td>
<td>5.0</td>
<td>8.0</td>
</tr>
<tr>
<td>$u_2$</td>
<td>$-8.1$</td>
<td>$\leq 5.0$</td>
<td>$\leq 8.0$</td>
</tr>
<tr>
<td>$u_2$</td>
<td>6.0</td>
<td>14.6</td>
<td>22.3</td>
</tr>
<tr>
<td>$u_3$</td>
<td>8.1</td>
<td>14.0</td>
<td>10.3</td>
</tr>
<tr>
<td>$u_3$</td>
<td>$-6.0$</td>
<td>$\leq 7.0$</td>
<td>$\leq 6.3$</td>
</tr>
<tr>
<td>$u_4$</td>
<td>$-8.0$</td>
<td>5.0</td>
<td>6.4</td>
</tr>
<tr>
<td>$u_4$</td>
<td>4.0</td>
<td>25.0</td>
<td>26.9</td>
</tr>
<tr>
<td>$oc_1$</td>
<td>$-10.0$</td>
<td>0.0</td>
<td>0.0</td>
</tr>
<tr>
<td>$sb_1$</td>
<td>4.0</td>
<td>10.0</td>
<td>13.0</td>
</tr>
</tbody>
</table>

Table 5. Design Conditions of the Water-Using Operations in Case 3

<table>
<thead>
<tr>
<th>operation</th>
<th>flow rate (m$^3$/h)</th>
<th>Concentration (mg/L)</th>
<th>from/to</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>$k_1$</td>
<td>$k_2$</td>
</tr>
<tr>
<td>$u_1$</td>
<td>$-10.3$</td>
<td>0.0</td>
<td>0.0</td>
</tr>
<tr>
<td>$u_1$</td>
<td>10.3</td>
<td>4.9</td>
<td>7.8</td>
</tr>
<tr>
<td>$u_2$</td>
<td>$-8.8$</td>
<td>14.0</td>
<td>7.8</td>
</tr>
<tr>
<td>$u_2$</td>
<td>8.8</td>
<td>14.0</td>
<td>11.2</td>
</tr>
<tr>
<td>$u_3$</td>
<td>$-3.3$</td>
<td>0.0</td>
<td>0.0</td>
</tr>
<tr>
<td>$u_3$</td>
<td>3.3</td>
<td>15.0</td>
<td>30.0</td>
</tr>
<tr>
<td>$u_4$</td>
<td>$-7.0$</td>
<td>0.0</td>
<td>0.0</td>
</tr>
<tr>
<td>$u_4$</td>
<td>7.0</td>
<td>20.0</td>
<td>18.7</td>
</tr>
<tr>
<td>$oc_1$</td>
<td>$-10.0$</td>
<td>0.0</td>
<td>0.0</td>
</tr>
<tr>
<td>$sb_1$</td>
<td>4.0</td>
<td>10.0</td>
<td>13.0</td>
</tr>
</tbody>
</table>

(4) For every split branch in the superstructure, the lower bound of total transported water volume is 6 m$^3$ and the upper bound of instantaneous flow rate is 20 m$^3$/h; and

(5) The lower bound of tank volume is 1 m$^3$. The mathematical programming model was solved with the module DICOPT within the GAMS environment. A personal computer with Pentium 4 and a computer processing unit (CPU) frequency of 2.80 GHz was used to facilitate the computation. The basic principle for setting the initial guesses in the present study is essentially the same as that adopted in the continuous water system design. Specifically, the initial value of flow rate in each branch of the superstructure at every time interval is first estimated with heuristics to satisfy the water balances. The bilinear component balance equations can be converted to a set of linear equations by substituting the estimated water flow rates. The initial guesses of pollutant concentrations can then be calculated according to these transformed equations. For the MINLP model presented in this paper, its relaxed version (RMINLP) should be solved in advance. The resulting optimal solution is subsequently used as the initial guess for solving the strict MINLP model.

The results of three case studies are presented here. In the first case, no limits were imposed on the number of pipelines connected to the mixing or splitting points in the water-reuse system. These upper bounds were all set to a value of 3 in the second case study. In the third case, the maximum number of pipelines entering the mixing point and leaving the splitting point of every equipment in set $E$ were both set to 1, whereas the pipeline numbers on the remaining mixing and splitting points were not constrained. In all cases, the number of buffer tanks embedded in the superstructure was three, but only two of them were eventually selected in the optimal solutions. The basic features of the network designs obtained in these three cases are presented in Table 2.

More-detailed information about the input and/or output streams of each water-using operation in these design cases 1, 2, and 3 can be found in Tables 3, 4, and 5, respectively. Notice that the water flows to and from the water-using operations are allowed to occur only in the time intervals specified in the fourth and fifth columns of Table 1. This constraint should also be applicable to the flow rates listed in the three tables just mentioned. In the proposed model, the pollutant concentrations at the outlets of water users in set $UA$, i.e., $u_1$, $u_2$, and $u_3$, are assumed to be constant, but the corresponding inlet concentrations may or may not vary with time. On the other hand, the pollutant concentrations in the input and output streams of the water-using operations in set $UB$, i.e., $u_4$, can be either maintained at steady values or time-variant. The same characterization can also be given to the inlet concentrations of $oc_1$, which is an operation in set $OC$. Therefore, every “less than or equal to” symbol (\(\leq\)) in Table 3 or 4 is used to denote the fact that the corresponding concentration is a function of time and only the maximum value is given. For case 2, additional results are plotted to provide further insight into the system dynamics.

In particular, the time profiles of liquid volumes in tanks $b_1$ and $b_2$ are presented in Figures 3 and 4, respectively. In addition, the flow-rate variation of wastewater discharged to sink $ob_1$ can be observed in Figure 5. In Figures 3 and 4, one can see that the liquid volume in either tank varies dramatically and, in certain instances, the tank is empty. The latter observations indicate that the tank sizes obtained in the system design are, indeed, minimum. Figure 5 shows that the inlet flow rate of sink $ob_1$ can be successfully equalized to within the specified range.
Finally, the physical network configurations of the system designs obtained in cases 2 and 3 are individually shown in Figures 6 and 7, respectively. From these figures, it can be observed that the network configuration in the latter case is, indeed, simpler. Furthermore, the results in Table 2 indicate that one can conclude that the inequality constraints given in eqs 64–69 do have an obvious impact on the network design. Specifically, the total number of pipelines is reduced from 15 in case 1 to 11 in case 2, at a cost of an 8% increase in the objective value, and from 11 to 8 in case 3, at an additional cost of only a 1% increase.

Conclusions

A general mathematical programming model has been developed in this study for the optimal design of discontinuous water-reuse systems. The number and sizes of buffer tanks, the physical configuration of pipeline network, and the operating policies of water flows can be determined based on this model. The proposed formulation is believed to be more comprehensive than the previous formulations, in terms of not only the unit models of water users and buffer tanks but also their connection scheme. To address the practical needs in wastewater treatment, the flow and concentration equalization options are also incorporated into the superstructure. The correspondences between the flow branches in superstructure and the actual pipelines in water-reuse system are established with additional logic constraints. The feasibility and effectiveness of this mathematical program is clearly demonstrated in the illustrative example.

Acknowledgment

This work is supported by the National Science Council of the ROC government, under Grant No. NSC93-2816-E006-00036.

Literature Cited


Received for review April 6, 2005
Revised manuscript received April 9, 2006
Accepted May 10, 2006