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An Extended Superstructure Formulation for Non-Isobaric Flowsheet Synthesis

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ABSTRACT

Flowsheet synthesis is an integral step in process design, entailing the selection of a set of unit operations and their connectivity to convert raw materials to products. Superstructure optimisation represents a promising class of synthesis approaches, allowing for the systematic exploration of the flowsheet design space. Despite this, many superstructure formulations suffer from numerical instabilities, combinatorial explosion, and/or rely on restrictive assumptions on the types of flowsheet alternatives that can be considered. The modified state-operator network (MSON) formalism has recently been proposed to address some of these issues for isobaric flowsheets. The constant-pressure assumption restricts the applicability of the MSON to real process applications as pressure is a key process variable in many unit operations, such as distillation, reaction, and extrusion, and is necessary to elicit flow. In this work, we present the extended MSON (E-MSON) which inherits the numerical stability of MSON, whilst removing the isobaric assumption. This is achieved through the introduction of new constraints that further improve the numerical behaviour of the MSON. The E-MSON is then applied to a simple non-isobaric superstructure optimisation problem, the results of which demonstrate that the E-MSON can serve as a framework for non-isobaric flowsheet synthesis, enabling a broader range of flowsheet alternatives to be considered.

Keywords: Superstructure Optimisation, Process Synthesis, Optimisation, Process Design, gPROMS, MINLP

INTRODUCTION

Flowsheet synthesis is a key step in the broader problem of chemical process design, involving the selection of a feasible or even optimal configuration of unit operations to convert raw materials into valuable products. Traditional approaches to flowsheet synthesis and design are hierarchical and heuristic-based [1]. In these methods, the design procedure is decomposed into increasingly detailed stages that progress from high-level structural decisions to detailed unit design. These stages rely on ad hoc rules-of-thumb to eliminate potential configurations without performing detailed design calculations [2]. Although it is relatively straightforward to apply such an approach, decomposing the design problem makes accounting for the interdependence of decisions made at different stages largely intractable [1].

With the advancement of nonlinear and mixed-integer programming, superstructure optimisation has become an increasingly promising alternative to hierarchical process synthesis techniques [3]. Superstructure optimisation first involves the representation of all candidate flowsheet topologies in a superstructure. This superstructure is then translated into a mathematical programming problem which can then be solved to yield the optimal flowsheet configuration [4], allowing for a systematic exploration of the process design space.

In general, due to the binary decision variables representing unit selection or deselection in flowsheet alternatives, and nonlinearities in the models that describe process phenomena, superstructure optimisation problems can be represented as mixed-integer nonlinear programming (MINLP) problems. How to represent the superstructure is, however, non-trivial, having a significant

influence on the solvability of the subsequent problem, and on the quality of the result obtained [1]. For instance, one commonly encountered issue which depends on the formulation of choice is combinatorial complexity.

Among superstructure representations, there exist several graph-theoretic approaches, including the State Task Network [5], State Equipment Network [6], and the State Operator Network [7]. More recent approaches instead involve abstraction of the flowsheet into building blocks, such as the phenomena building block approach of Lutze et al. [8] or the building-block superstructure presented by Demirel et al. [9]. These abstract representations facilitate intensified or novel unit operations to be yielded as a solution of the optimisation problem, but this generality comes at the expense of the solvability of the subsequent mathematical programming problem [1].

In the State Operator Network (SON) [7], which is the subject of this work, nodes of the graph represent process units, this may include unit operations, e.g., a distillation column, discrete subsets of a unit operation, e.g., a stage of a distillation column, or an intensified unit operation, e.g., a vapour compression distillation which includes the distillation column and heat pump. The edges of the graph are streams, representing material flows from unit operation to the other. To allow for material to flow between units, conceptual mixers and splitters are introduced. Each inlet of each process unit is associated with a conceptual mixer that combines the properties of connected streams (edges) into the input to the units' model. Similarly, a splitter that splits material flows across streams is defined at each outlet.

To ensure that meaningful results are obtained upon the solution of superstructure optimisation problems, rigorous and high-fidelity models must be utilised [7]. In the SON, each process unit is described by a detailed process model that not only includes mass, energy, summation of composition, and heat balance equations, detailed thermodynamic models, and kinetic models when applicable, but also costing and sizing correlations. The model equations that represent each unit are therefore coupled, nonlinear, and high-dimensional, making the solution of superstructure optimisation problems challenging.

Superstructure optimisation problems may be solved in equation-oriented optimisation paradigms, e.g., Pyomo, or by using simulation-based optimisation. Simulation-based optimisation offers the advantages of in-built-process models and thermodynamic routines, as well as initialisation strategies which can be crucial to obtaining high-quality solutions. On the other hand, equation-oriented optimisation paradigms offer great flexibility in formulating the problem and a greater choice of numerical solvers. They also provide complete access to functional forms, allowing the construction of convex relations for global optimisation. Despite this, due to the sheer number of variables and constraints in these problems, global optimisation is typically intractable, meaning superstructure optimisation problems are generally only solved to local optimality.

Another complication associated with superstructure optimisation is numerical failure. When sections of the superstructure are deselected, whether that be building blocks or entire units, mass flows into these units must be set to zero. Thus, even the seemingly simple calculation of a mole fraction can lead to a numerical singularity and hence the failure of the optimisation solver. Several examples of numerically undefined behaviour or singularity arise when process units are deselected [10].

Several approximate methods to avoid numerical issues have been presented, including the relaxation of problematic equations to continuous and differentiable approximations [11], and the introduction of small non-zero flows into deselected units [12]. Exact, systematic formulations that are guaranteed to avoid numerical issues due to unit deselection have also been proposed. Yeomans and Grossmann [13] suggested to formulate the optimisation problem using generalised disjunctive programming (GDP), representing the binary decisions of unit selection and deselection as logical disjunctions. Using the logic-based outer approximation (LBOA) algorithm, the primal problem constructed at each iteration only includes the model equations pertaining to selected units with non-zero inlet flowrates. This avoids numerical failure entirely, however, the automatic generation of the primal problem in process simulation software can be challenging, as noted by Navarro-Amorós et al. [14]. Further issues pertaining to initialisation of the master problem of the LBOA algorithm may also occur [15].

Another promising solution to the problem of numerical failure arising from unit deselection is to modify the superstructure representation itself, as performed in the modified SON (MSON), presented by Gopinath and Adjiman [15, 16]. In the MSON, mixers are modified in the to have a fictitious stream. Fictitious stream flowrates are non-zero when unit operation is deselected and zero otherwise; splitters are similarly modified, ensuring that the solution (optimal point, objective function value) is unaffected by these fictitious flows.

The MSON formulation leads to improved numerical stability, eliminating the possibility of singularities in deselected units. Applied to the design of a counter-current carbon capture column, the optimisation of an MSON superstructure yielded a high-performance solution, indicating its applicability to complex synthesis problems. The MSON may be used in both equation-oriented and simulation-based optimization paradigms. Being based on the SON [7], the MSON also inherits an implicit reduction of combinatorial complexity, as compared to task-based representations such as the State Task Network.

In the definition of both the SON [7] and MSON [15], the pressure is assumed to be constant throughout the

flowsheet. Although an extension to treat pressure as an optimisation variable has been presented for the specific case of the synthesis of counter-current columns only [16], the MSON cannot directly be applied generally to non-isobaric flowsheet synthesis without modification.

Pressure is a key operating variable in many unit operations, due to its influence on chemical and phase equilibria [17], and being essential to elicit flow. Many unit operations, such as pressure-swing distillation, membrane-based processes, and high-pressure synthesis also cannot be modelled without allowing pressure to vary across the flowsheet [18-20]. Furthermore, compression and expansion can represent significant proportions of process costs, e.g., up to 75% of the capital expenditure for refrigeration systems [21]. Rigorous flowsheet modelling also requires accurately accounting for pressure drops.

Therefore, a superstructure formulation that inherits the mathematical tractability of the MSON, whilst relaxing its isobaric assumption, is highly desirable. It would broaden the scope of flowsheet alternatives and objectives that can be considered in a superstructure optimisation problem. Thus, in this work, we present the extended MSON (E-MSON), establishing a framework which facilitates the systematic and rigorous synthesis of non-isobaric flowsheets. We demonstrate the application of E-MSON on a test problem, successfully allowing the process units to operate at different pressures. In the next section, we present the superstructure representation of the E-MSON, highlighting the variables and sets of relevance, before giving its full derivation.

E-MSON: SUPERSTRUCTURE REPRESENTATION

The E-MSON formulation avoids numerical singularities through the introduction of strictly positive fictitious streams into deselected units [15]. Each stream has a mass flowrate, $f \in \mathbb{R}^+$, composition, $\boldsymbol{q} \in [0,1]^K$, where K is the number of components considered in the problem, and is at temperature, $T \in \mathbb{R}^+$, and pressure, $P \in \mathbb{R}^+$.

Let the set of all process units in the superstructure be represented by $\boldsymbol{\mathcal{L}}$, with each unit $u \in \boldsymbol{\mathcal{L}}$ being described by rigorous model equations (mass and energy balances, sizing, costing, etc.). We represent the set of conditional units, referring to units that can be deselected in a particular alternative, as $\boldsymbol{\mathcal{T}} \subseteq \boldsymbol{\mathcal{L}}$, such that the set of permanent units that must be included in all alternatives is given by $\boldsymbol{\mathcal{L}} \backslash \boldsymbol{\mathcal{T}}$. The set of units include all unit operations, including pressure-change equipment such as pumps, valves, and compressors, which we represent as $\boldsymbol{\mathcal{PC}} \subset \boldsymbol{\mathcal{L}}$. The set of all nodes of the graph can then be defined as $\boldsymbol{\mathcal{U}} = \boldsymbol{\mathcal{L}} \cup \boldsymbol{\mathcal{F}} \cup \boldsymbol{\mathcal{P}}$, where $\boldsymbol{\mathcal{F}}$ and $\boldsymbol{\mathcal{P}}$ refer to the sets of feeds and products containing the material sources and sinks to and from the superstructure respectively.

Each unit $u \in \boldsymbol{\mathcal{U}}$ is also associated with inlet and outlet indices corresponding to the sets $\boldsymbol{\mathcal{IN}_u}$ and $\boldsymbol{\mathcal{OUT}_u}$, the union of which give, for all $u \in \boldsymbol{\mathcal{U}}$, the sets of all mass inlet and outlet indices $\boldsymbol{\mathcal{I}}$ and $\boldsymbol{\mathcal{O}}$. At each inlet and outlet, we additionally specify an isenthalpic mixer and splitter, respectively, to facilitate connectivity between units. Permanent units $u \in \boldsymbol{\mathcal{L}} \backslash \boldsymbol{\mathcal{T}}$ are associated with standard mixers and splitters, denoted by m_i and s_o at inlet $i \in \boldsymbol{\mathcal{I}}$ and outlet $o \in \boldsymbol{\mathcal{O}}$ respectively. The inlet flowrate, composition, temperature, and pressure to a unit from a mixer at inlet i are labelled as $f_i^{in}, \boldsymbol{q_i^{in}}, T_i^{in}$, and P_i^{in}, and the outlet flowrate, composition, temperature and pressure from a unit to a splitter at outlet o as $f_o^{out}, \boldsymbol{q_o^{out}}, T_o^{out}, P_o^{out}$.

As in the MSON [15, 16], conditional units are connected to modified isenthalpic mixers and splitters, labelled as $\tilde{m}_i$ and $\tilde{s}_o$. At modified mixer $\tilde{m}_i$, the flowrate of the fictitious stream, f_i^M, takes the auxiliary value, f_i^A, when the unit is deselected. Additionally, when $f_i^M > 0$, the corresponding unit's inlet composition, temperature, and pressure are constrained to take the auxiliary values, $\boldsymbol{q_i^A}, T_i^A$, and P_i^A, defined *a priori* for the fictitious stream. Analogously, at the modified splitter, $\tilde{s}_o$, the variables $f_o^S, \boldsymbol{q_o^S}, T_o^S$, and P_o^S define the fictitious stream. An example of a completely labelled conditional unit following the E-MSON formalism can be seen in Figure 1.

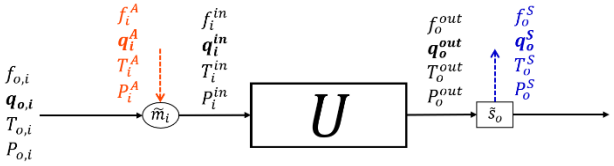

Figure 1. Example representation of a conditional unit $\underline{U}$ in the E-MSON formulation, including all variables of relevance. The fictitious streams of the modified mixer and splitter are coloured and marked by dashed lines.

The connectivity between mixers and splitters is captured in the sets $\boldsymbol{\mathcal{M}_i}$ and $\boldsymbol{\mathcal{S}_o}$, where $\boldsymbol{\mathcal{M}_i} \subseteq \boldsymbol{\mathcal{O}}$ represents the set of outlet indices connected to mixer m_i or $\tilde{m}_i$ and $\boldsymbol{\mathcal{S}_o} \subseteq \boldsymbol{\mathcal{I}}$ represents the set of inlet indices connected to splitter s_o or $\tilde{s}_o$. As with all other streams, the connections between mixers and splitters are parameterised in terms of flowrate, composition, temperature, and pressure, denoted as $f_{o,i}, \boldsymbol{q_{o,i}}, T_{o,i}$, and $P_{o,i}$, respectively.

To enable the selection and deselection of conditional units in the superstructure, each conditional unit $u \in \boldsymbol{\mathcal{T}}$ is also associated with a binary variable z_u that describes whether it is selected in $(z_u = 1)$ or deselected from $(z_u = 0)$ a particular flowsheet configuration. It is important to also define $\boldsymbol{\mathcal{C}_u}$, representing the set of "output" variables, that is, those variables corresponding to conditional unit $u \in \boldsymbol{\mathcal{T}}$ that are included in flowsheet-level constraints or the objective function.

In the case where continuous or discrete *design*

variables are present in the model of a conditional unit (such as a reboiler duty or the number of stages in a column), additional auxiliary variables must be defined to ensure model convergence. We represent these continuous and discrete design variables in the vectors $\boldsymbol{x}_{\mathcal{A}_u}$ and $\boldsymbol{y}_{\mathcal{A}_u}, \forall\, u \in \mathcal{T}$, respectively, and store the auxiliary values similarly in the vectors $\boldsymbol{x}^{\boldsymbol{A}}_{\mathcal{A}_u}$ and $\boldsymbol{y}^{\boldsymbol{A}}_{\mathcal{A}_u}, \forall\, u \in \mathcal{T}$. As with other auxiliary variables in the E-MSON approach, these should be assigned values in advance such that, in combination with $f_i^A, \boldsymbol{q_i^A}, T_i^A$, and P_i^A for all inlets $i \in \mathcal{IN}_u$, a solution is obtained to the model equations of process unit $u \in \mathcal{T}$.

We also define a binary variable, $z_{o,i}$ for all $o \in \mathcal{M}_i, i \in \mathcal{IN}_u, u \in \mathcal{L} \cup \mathcal{P}$. When outlet o and inlet i are connected, meaning $f_{o,i} > 0$, the binary variable $z_{o,i}$ takes a value of 1 and otherwise the binary variable is zero. Further, when $z_{o,i} = 1$, the pressure at the inlet i must be equal to the pressure $P_{o,i}$ of the stream that exits outlet o (cf. Figure 2b), this ensures the mixing of streams does not result physically in unintended flows, i.e.: from a high-pressure unit into a low-pressure one. When outlet o and inlet i are disconnected, when $z_{o,i} = 0$ and $f_{o,i} = 0$, the downstream pressure, P_i^{in}, is allowed to vary independently of $P_{o,i}$ (c.f Figure 2c).

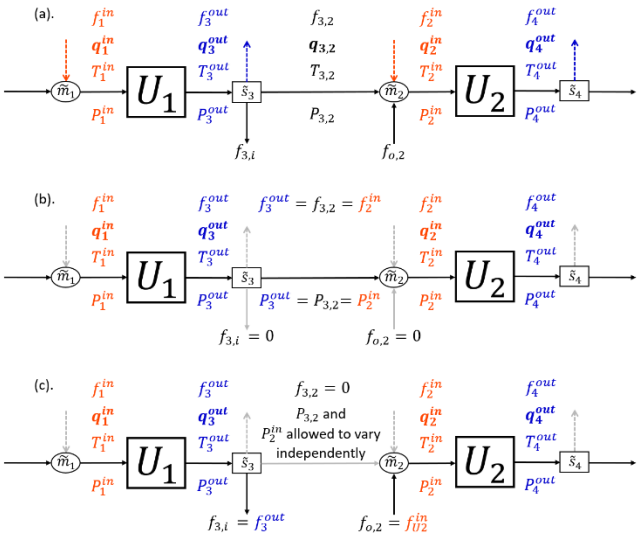

Figure 2. Representation of how pressures are connected within the E-MSON approach. (a) denotes the relevant variables of two conditional units, U_1, which has a single inlet and two outlets, and U_2, which has two inlets and a single outlet. (b) shows the pressure relationship when the two units are connected (when $z_{3,2} = 1$), and (c) illustrates the case when they are not connected (when $z_{3,2} = 0$).

Note that, due to Equations 9b to 9d and 10b to 10g, all streams leaving a splitter are at a composition, temperature, and pressure equal to the splitter's inlet.

Note that meaningful relations between the pressures of two units cannot be derived solely based on their selection in a flowsheet (as denoted by z_u). Inlet and outlet pressures should only be equal when two units are directly connected via flow, as indicated by $z_{o,i}$. In addition to enforcing correct pressure relationships, the introduction $z_{o,i}$ allows for the consideration of flowsheet alternatives where the same units are selected but different streams are active. For instance, the comparative performance of two flowsheet alternatives including the same set of conditional units, but with or without a recycle stream, can now directly be compared using the E-MSON formalism. Lastly, we mention the following assumptions made in the derivation of the MSON, which carry across to the E-MSON: all flows are assumed isenthalpic and frictionless, implying no pressure losses are incurred between units; all units are at steady state; no reaction occurs in mixers and splitters; and all mixers and splitters are isenthalpic and the work of mixing is zero.

E-MSON: FORMULATION

What follows is a complete definition of the E-MSON model equations; although many of these constraints are in common with the MSON approach [15, 16], they are included here for completeness. The MINLP reformulation of the E-MSON is given in Big-M form in this work, but alternative reformulations, such as those using convex hulls [22], could also be utilised. To constrain that the flowrate $f_{o,i}$ between outlet o and inlet i must be non-zero when $z_{o,i} = 1$, and zero when $z_{o,i} = 0$, the following constraints are defined, $\forall\, o \in \mathcal{M}_i, \forall\, i \in \mathcal{IN}_u, \forall\, u \in \mathcal{L} \cup \mathcal{P}$:

$$f_{o,i} \leq \bar{f} z_{o,i} \tag{1a}$$

$$\varepsilon_f z_{o,i} \leq f_{o,i} \tag{1b}$$

where $\bar{f}$ and ε_f are upper and lower bounds on flowrate respectively. Additionally, as in the MSON derivation, we constrain that all inter-unit flowrates directed to a deselected unit are zero, as follows:

$$f_{o,i} \leq \bar{f} z_u, \forall\, o \in \mathcal{M}_i, \forall\, i \in \mathcal{IN}_u, \forall\, u \in \mathcal{T} \tag{2}$$

and that all flowrates into process units are positive:

$$\varepsilon_f \leq f_i^{in}, \forall\, i \in \mathcal{IN}_u, \forall\, u \in \mathcal{L} \tag{3}$$

Further to this, we can impose the following linear relations between unit and stream binary variables, such that the number of feasible combinations of binary variables is reduced significantly. If a unit is deselected, the connected flows should accordingly be inactive:

$$z_{o,i} \leq z_u, \forall\, o \in \mathcal{M}_i, \forall\, i \in \mathcal{IN}_u, \forall\, u \in \mathcal{T} \tag{4a}$$

$$z_{o,i} \leq z_u, \forall\, i \in \mathcal{S}_o, \forall\, o \in \mathcal{OUT}_u, \forall\, u \in \mathcal{T} \tag{4b}$$

and, if a unit is selected, it should have at least one active inlet and one active outlet stream:

$$z_u \leq \sum_{i \in \mathcal{IN}_u} \sum_{o \in \mathcal{M}_i} z_{o,i}, \forall\, u \in \mathcal{T} \quad (5a)$$

$$z_u \leq \sum_{o \in \mathcal{OUT}_u} \sum_{i \in \mathcal{S}_o} z_{o,i}, \forall\, u \in \mathcal{T} \quad (5b)$$

The mass, component, and enthalpy balances around standard mixers are defined as follows, $\forall\, i \in \mathcal{IN}_u, u \in (\mathcal{L} \backslash \mathcal{T}) \cup \mathcal{P}$:

$$f_i^{in} = \sum_{o \in \mathcal{M}_i} f_{o,i} \quad (6a)$$

$$f_i^{in} q_{i,c}^{in} = \sum_{o \in \mathcal{M}_i} f_{o,i}\, q_{o,i,c}, \forall\, c \in \{1, \ldots, K\} \quad (6b)$$

$$f_i^{in} h(T_i^{in}, \boldsymbol{q_i^{in}}, P_i^{in}) = \sum_{o \in \mathcal{M}_i} f_{o,i}\, h(T_{o,i}, \boldsymbol{q_{o,i}}, P_{o,i}) \quad (6c)$$

where $h(T, \boldsymbol{q}, P)$ is the enthalpy per unit mass of a stream at temperature T, pressure P, and composition $\boldsymbol{q}$. Note the sum of all component fractions need not be constrained to one at mixers and splitters, given that it is satisfied by the input specifications at sources.

To ensure the desired behaviour of fictitious streams at modified mixers is enforced, at all inlets $i \in \mathcal{IN}_u, \forall\, u \in \mathcal{T}$, modified mixer constraints are defined. The mass, component, and enthalpy balances are given by:

$$f_i^{in} = \sum_{o \in \mathcal{M}_i} f_{o,i} + f_i^M \quad (7a)$$

$$f_i^{in} q_{i,c}^{in} = \sum_{o \in \mathcal{M}_i} f_{o,i}\, q_{o,i,c} + f_i^M q_{i,c}^A, \forall\, c \in \{1, \ldots, K\} \quad (7b)$$

$$f_i^{in} h(T_i^{in}, \boldsymbol{q_i^{in}}, P_i^{in}) = \sum_{o \in \mathcal{M}_i} f_{o,i}\, h(T_{o,i}, \boldsymbol{q_{o,i}}, P_{o,i}) + f_i^M h(T_i^A, \boldsymbol{q_i^A}, P_i^A) \quad (7c)$$

To ensure that the fictitious flowrate and inlet pressure are only equal to the auxiliary value when the unit is deselected, and that when the unit is selected, the fictitious flowrate is zero, the following constraints are also necessary, where M is a suitably large positive constant:

$$f_i^M = f_i^A (1 - z_u) \quad (7d)$$

$$-M z_u \leq P_i^A - P_i^{in} \leq M z_u \quad (7e)$$

Additionally, across *all* mixers, the inlet and outlet pressures should only be equal when the corresponding flowrate is non-zero (i.e.: $z_{o,i} = 1$), therefore, $\forall\, o \in \mathcal{M}_i, \forall\, i \in \mathcal{IN}_u, u \in \mathcal{L} \cup \mathcal{P}$:

$$-M(1 - z_{o,i}) \leq P_i^{in} - P_{o,i} \leq M(1 - z_{o,i}) \quad (8)$$

Equation 8 implies that if there are multiple non-zero flows from different outlets connected to an inlet, each of those outlets are at the same pressure. At each standard splitter, we require mass balance constraints and enforce the equality of compositions, temperatures, and pressures across the splitter, s_o. Hence, $\forall\, o \in \mathcal{OUT}_u, \forall\, u \in (\mathcal{L} \backslash \mathcal{T}) \cup \mathcal{F}$, we define:

$$f_o^{out} = \sum_{i \in \mathcal{S}_o} f_{o,i} \quad (9a)$$

$$q_{o,c}^{out} = q_{o,i,c}, \forall\, c \in \{1, \ldots, K\}, \forall\, i \in \mathcal{S_o} \quad (9b)$$

$$T_o^{out} = T_{o,i}, \forall\, i \in \mathcal{S_o} \quad (9c)$$

$$P_o^{out} = P_{o,i}, \forall\, i \in \mathcal{S_o} \quad (9d)$$

At modified splitters, we define the following, including the fictitious stream, $\forall\, o \in \mathcal{OUT}_u, \forall\, u \in \mathcal{T}$:

$$f_o^{out} = \sum_{i \in \mathcal{S}_o} f_{o,i} + f_o^S \quad (10a)$$

$$q_{o,c}^{out} = q_{o,i,c}, \forall\, c \in \{1, \ldots, K\}, \forall\, i \in \mathcal{S_o} \quad (10b)$$

$$T_o^{out} = T_{o,i}, \forall\, i \in \mathcal{S_o} \quad (10c)$$

$$P_o^{out} = P_{o,i}, \forall\, i \in \mathcal{S_o} \quad (10d)$$

$$q_{o,c}^{out} = q_{o,c}^S, \forall\, c \in \{1, \ldots, K\} \quad (10e)$$

$$T_o^{out} = T_o^S \quad (10f)$$

$$P_o^{out} = P_o^S \quad (10g)$$

As fictitious mass flow is added to the unit upon its deselection, to ensure that exact results are obtained from the remainder of the flowsheet, we must also enforce that when $z_u = 0, f_o^S = f_o^{out}$, by:

$$f_o^{out} - f_o^S \leq \bar{f} z_u \quad (11)$$

Furthermore, we must also ensure that no mass leaves in the fictitious stream when the unit is selected, restricting f_o^S to zero as follows:

$$0 \leq f_o^S \leq \bar{f}(1 - z_u) \quad (12)$$

To ensure the design variables of conditional units ($\boldsymbol{x_{\mathcal{A}_u}}$ and $\boldsymbol{y_{\mathcal{A}_u}}$) take the required auxiliary values upon deselection ($\boldsymbol{x_{\mathcal{A}_u}^A}$ and $\boldsymbol{y_{\mathcal{A}_U}^A}$), we introduce, $\forall\, u \in \mathcal{T}$:

$$-M z_u \leq x_{\mathcal{A}_u} - x_{\mathcal{A}_u}^A \leq M z_u \quad (13a)$$

$$-M z_u \leq y_{\mathcal{A}_u} - y_{\mathcal{A}_u}^A \leq M z_u \quad (13b)$$

As well as this, the "output" variables of a conditional unit, $x_i, i \in \mathcal{C_u}$, should only impact flowsheet-wide constraints if the unit is active. This is achieved by introducing an auxiliary vector of continuous variables, $\boldsymbol{x_i^S}$, and the following constraints, $\forall\, i \in \mathcal{C_u}, \forall\, u \in \mathcal{T}$:

$$-M(1 - z_u) \leq x_i - x_i^S \leq M(1 - z_u) \quad (14a)$$

$$-M z_u \leq x_i^S - c_{u,i} \leq M z_u \quad (14a)$$

where, $c_{u,i}$ is the relevant member of a vector of values $\boldsymbol{c_u}$, often of zeroes, which "output" variables should take upon unit deselection. Further, the flowsheet wide constraints and objective are expressed in term of auxiliary variables x_i^S instead of $x_i\, \forall\, i \in \mathcal{C_u}, \forall\, u \in \mathcal{T}$.

In addition to enforcing that physically meaningful values of pressure are obtained from the E-MSON, it is also of interest to include constraints related to the pressure-change units, $u \in \mathcal{PC}$. If $\mathcal{PC}$ is partitioned into a set $\mathcal{COM}$ of compressing units that lead to a positive pressure difference across them, and analogously a set $\mathcal{EXP}$

of expanding units that lower pressure, such that $\mathcal{PC} = \mathcal{EXP} \cup \mathcal{COM}$, it is possible to enforce that only the correct pressure-change occurs across them. Although the detailed models of such units should ensure that only the correct pressure difference can occur in compressors and expanders at feasible solutions (i.e. an increase across compressors, and a decrease across expanders), numerical difficulties and thus solver failure can still be encountered. For instance, if poor initialisation leads to a negative ΔP across a compressor or an NLP solver progresses in this direction, this can result in a negative value for power, which in many cost correlations would then be raised to a non-integer exponent [21] and thus cause a numerical singularity. To avoid this, we define the following for compressing units, $\forall\, i \in \mathcal{IN}_u, \forall\, o \in \mathcal{OUT}_u, \forall\, u \in \mathcal{COM}$:

$$\varepsilon_P \leq P_o^{out} - P_i^{in} \tag{15a}$$

where ε_P is a small positive constant to ensure a positive ΔP is obtained. Analogously for expanding units, $\forall\, i \in \mathcal{IN}_u, \forall\, o \in \mathcal{OUT}_u, \forall\, u \in \mathcal{EXP}$:

$$\varepsilon_P \leq P_i^{in} - P_o^{out} \tag{15b}$$

AN APPLICATION OF E-MSON

We now illustrate the use of the E-MSON formalism for variable pressure flowsheet synthesis on a simple test problem. The E-MSON representation of the problem is given in Figure 3.

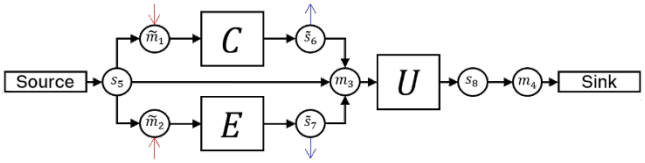

Figure 3. E-MSON representation of the test problem, where units C and E are a conditional compressor and expander, respectively, and U is an arbitrary permanent unit. Splitter s_8 and mixer m_4 are shown for completeness and may be eliminated in practice.

Three feasible flowsheet alternatives can be obtained from the superstructure, where exactly one of the compressor, C, the expander, E, or a bypass stream is active. Within the problem, the key decision variables are the two unit selection binary variables, z_C and z_E, the three flow activity binaries to indicate whether compression, expansion, or neither occurs ($z_{6,3}, z_{7,3}, z_{5,3}$), as well as the inlet pressure to the permanent unit, P_U^{in}.

For simplicity, streams in this problem are described only in terms of mass flowrate and pressure, implying any constraints involving composition and temperature from the full E-MSON derivation are not included. Simplified models are also used for each unit operation. The purpose of this problem is to provide initial evidence of the applicability of the E-MSON to non-isobaric superstructure optimisation problems. Indeed, as the pressure across the compressor and expander must vary from the source pressure, the MSON and SON formulations are inapplicable to even this rudimentary case.

The compressor and expander are assigned a fixed cost, and no mass loss is assumed across them. Notably, as the pressure difference across pressure-change units is constrained to be in the correct direction for each unit, neither the inlet pressure nor the outlet pressure is involved in the toy compressor and expander models. These models are given by, $\forall\, u \in \{C, E\} = \mathcal{PC} = \mathcal{T}$:

$$f_i^{in} = f_o^{out}, \forall\, i \in \mathcal{IN}_u, \forall o \in \mathcal{OUT}_u \tag{20a}$$

$$c_u = \mu_u \tag{20b}$$

where μ_u is a unit-specific constant.

The permanent unit is modelled as following, based on modification of the toy unit models in the case studies on which the MSON was first tested [15], $\forall\, u = U = \mathcal{L} \backslash \mathcal{T}$:

$$x_u f_i^{in} = f_o^{out}, \forall\, i \in \mathcal{IN}_u, \forall\, o \in \mathcal{OUT}_u \tag{21a}$$

$$x_u = \frac{P_i^{in}}{P_{UB}}, \forall\, i \in \mathcal{IN}_u \tag{21b}$$

$$c_u = \mu_u \left(f_i^{in} P_i^{in}\right)^{\upsilon_u}, \forall\, i \in \mathcal{IN}_u \tag{21c}$$

$$P_i^{in} = P_o^{out}, \forall\, i \in \mathcal{IN}_u, \forall\, o \in \mathcal{OUT}_u \tag{21d}$$

where υ_u is a unit-specific constant, x_u is a scaled pressure, and P_{UB} is an upper bound on pressure.

The objective is to maximise "profit", modelled as the revenue (the value of the stream being processed, given by the inlet flowrate into the sink and corresponding price c_f) minus the cost of each unit included in the selected configuration. It is given as:

$$\max\left(c_f f_{sink}^{in} - c_U - c_E^S - c_C^S\right) \tag{22}$$

where c_f is set to 1 without loss of generality, and c_E^S and c_C^S are auxiliary variables representing the cost of the expander and compressor, respectively, taking a value of zero when the unit is deselected. To complete the definition of the model, the unit-specific parameters are given in Table 1, and the additional E-MSON specific parameters are given in Table 2.

Table 1. Unit-specific parameters of the E-MSON test problem, where f_i^A, P_i^A, μ_u, and υ_u are the fictitious flowrate, fictitious pressure, and unit-specific parameters of the compressor, C, expander, E, and unit, U.

Unit	f_i^A	P_i^A	μ_u	υ_u
C	25.0	1.1	20.0	-
E	25.0	9.9	5.0	-
U	-	-	0.3	0.7

Table 2. Additional constants necessary to define the E-MSON test problem. Where f_{source} and P_{source} are the source flowrate and pressure, respectively, $\bar{f}$ and P_{UB} are the flowsheet-wide upper bounds on flow rate and pressure, P_{LB} is the lower bound, and ε_F and ε_P are small constants to ensure positive flowrates and pressure differences.

Parameter	Value
f_{source}	50.0
P_{source}	5.0
$\bar{f}$	50.0
P_{UB}	10.0
P_{LB}	1.0
ε_F	$1/\bar{f}$
ε_P	0.1

Table 3. Results from the E-MSON test problem, where P_U^{in} is the inlet pressure to the unit U in the superstructure.

Primal Iteration #	Selected Configuration	Objective Value	P_U^{in}
1	Bypass active	10.689	5
2	Expander active	5.390	1
3	Compressor active	6.751	10

The test problem was implemented in gPROMS Process 2025.2.0, utilising MINLPOA, a proprietary implementation of OAERAP [23], as the MINLP solver, NLPSQP as the primal NLP subproblem solver, and LPSOLVE as the master MILP problem solver. All the solvers utilised were run with standard settings, except for the convexity-based termination criterion of MINLPOA, which was disabled. The problem involved 72 variables, 8 of which were binary. The solver ran for 4 master problem iterations, obtaining solutions for each feasible combination of unit selection and flow activity binaries (3 combinations out of a possible 32), also indicating that the master problem at the final (fourth) iteration becomes infeasible. Convergence was achieved in less than 1 second. Further detail on the three primal problems that were solved is provided in Table 3, with the alternative in which no pressure-change unit was selected being found as optimal.

The results reported in Table 3 were obtained independently of which of the three configurations was selected as the initial guess to the problem. This was also found to be the case upon initialisation from an infeasible point, where all binary variables were set to zero.

CONCLUSION

In this work, we have presented E-MSON, a framework for the superstructure optimisation of non-isobaric flowsheets. It is a generalisation of the MSON approach in which modified isenthalpic splitters and mixers are introduced to avoid numerical difficulties, with the addition of pressure-change units and new constraints that ensure physically meaningful solutions. The extended MSON has then been tested on a toy superstructure containing an expander and a compressor, yielding promising results. Although this illustrative problem is small, it represents a non-isobaric flowsheet synthesis problem and is thus, to the authors' knowledge, beyond the scope of other exact and rigorous graph-theoretic superstructure formulations such as the SON, or MSON.

In future work the E-MSON will be applied to a broader range of problems, including more realistic process applications in which pressure represents a key operating variable. Beyond this, integrating such rigorous and exact superstructure formulations into molecular design problems is of interest, facilitating simultaneous molecular and process design. Additionally, constraints to further improve numerical convergence for units which require a particular inlet phase will be considered.

Furthermore, given the time-consuming nature of implementing such optimisation problems, automating the writing and execution of E-MSON superstructure optimisation problems is also of value. A Python library to do so within gPROMS Process is currently in development, permitting the user to visually represent the superstructure and generate the corresponding E-MSON problem formulation quickly.

DIGITAL SUPPLEMENTARY MATERIAL

Digital supplementary material, including the gPROMS Process 2025.2.0 file used in the solution of the test problem, and relevant case files, is available here: https://psecommunity.org/LAPSE:2026.0034

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