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110th Anniversary: A Generalized Nonsmooth Operator for Process Integration

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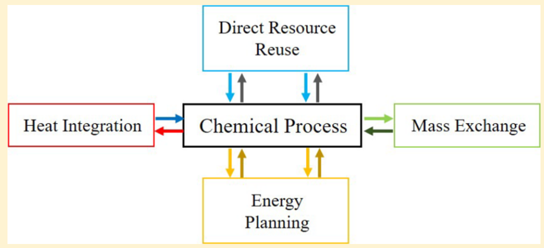
ABSTRACT: This paper presents a novel, generalized method for solving resource integration problems: the nonsmooth integration operator. Compared to current approaches, such as cascade analysis or the pinch location method, the nonsmooth integration operator is generalizable to any resource, including multiple resources simultaneously. Additionally, it is uniquely able to both solve for process variables and scale moderately with the number of sources and sinks in the system, and thus is well-equipped to handle large and complex multiplant systems, easily embedded in process optimization problems, and readily extendable to new applications. The nonsmooth integration operator is a system of two nonsmooth equations per resource that describe optimal conditions for pinch-constrained resource transfer limited to a single contaminant with preclassified sources and sinks. The operators for multiple resources can be combined with process models, and the resulting equation system is solved by using new advances in nonsmooth equation-solving. The operator can also be extended to automatically identify threshold problems. This paper details the formulation and use of the nonsmooth integration operator and includes several example problems to demonstrate its strengths and flexibility. These problems show that the nonsmooth integration operator can solve for unknown process variables, include process models, and simultaneously integrate multiple resources. The problems also cover a wide range of integration types, including mass integration, water and hydrogen networks, and carbon-constrained energy planning, to show the utility of a generalizable approach to the integration problem.



■ INTRODUCTION

In the face of increasing resource scarcity and costs, and stricter regulations on waste discharge, there are significant incentives to reduce resource use in chemical processes. Process integration addresses this problem of resource waste for both proposed and existing processes by identifying opportunities for optimally reusing resources throughout the system. As a result, methods for performing process integration have been widely proposed and utilized for a variety of resources, originating with heat in heat integration problems, then extending to materials in mass exchange networks (MENs) and water in water allocation problems. Seminal works in these fields include Hohman and Linnhoff et al. for heat integration,[1,2] El-Halwagi and Manousiouthakis for mass integration,[3] and Wang and Smith and Dhole et al. for water allocation.[4,5]

More recently, integration methods have also been applied beyond these traditional areas to new resources such as hydrogen,[6] oxygen,[7] carbon dioxide,[8] electrical power,[9] and even time in inventory and scheduling problems.[10] Additionally, to further decrease resource use, integration is being considered for increasingly large systems, including resource sharing between plants colocated in eco-industrial parks.[11] However, to have a significant impact on sustainability, the increasing application and scope of process integration requires integration methods that are generalizable, scalable, and efficient.

Process integration consists of two stages, first, determining the minimum attainable fresh and waste resource flows for the process, and second, designing a conservation network for resource reuse that can approach these targets. Integration approaches can solve these two stages either sequentially or simultaneously. Simultaneous approaches, first introduced by Grossmann and Sargent for heat integration,[12] use superstructures to optimize over different network configurations. While these approaches are generalizable to a wide array of integration problems and can in theory obtain globally optimal solutions, to guarantee optimality, the superstructures must embed all possible configurations and require solving large mixed-integer nonlinear programs (MINLP) that are often nonconvex.[13−15]

Therefore, in this work, we consider the former resource-targeting problem without the network design step. This approach avoids complex superstructure techniques and is able to consider resource usage and constraints more efficiently when screening and optimizing process designs.

The simplest class of targeting approaches are graphical pinch analysis and trans-shipment (also referred to as cascade) formulations, first proposed by Linnhoff et al. and Papoulias and Grossmann.[2,16] These approaches are physically intuitive and easy to use, but require process variables to be known a priori in order to construct quality intervals between which resources can be transferred. As a result, they cannot be used

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to simulate systems with known resource targets or for simultaneous process integration and optimization.

To address these limitations for heat integration, Duran and Grossmann developed the pinch location method,[17] which avoids the explicit construction of temperature intervals by proposing an optimization formulation with nonsmooth inequalities that automatically selects which streams should be included above potential pinch temperatures. This nonsmooth optimization problem can be solved using smoothing approximations or with disjunctions that either express the stream positions relative to the pinch temperature[18] or deal directly with the nonsmooth terms.[19] However, these smoothing approximations require a user-specified parameter that must be tuned to avoid inaccuracies and ill-conditioning, and the disjunctive formulations both solve MINLPs that scale quadratically in both the number of constraints and binary variables as the numbers of hot and cold streams in the problem increase. As a more intuitive alternative to the pinch location method, Navarro-Amoros et al. have also proposed a cascade-type model that uses disjunctions to determine temperature intervals,[20] but this model again requires solving a MINLP and exhibits cubic scaling.

To avoid approximations and improve the scaling of problem size with the number of streams, Watson et al.[21] reformulated the inequalities in the pinch location method to create a system of two nonsmooth equations that simulates multistream heat exchangers and retains the same number of equations regardless of the problem size. By describing the optimality conditions using explicitly nonsmooth equations, Watson et al. were also able to take advantage of new methods in nonsmooth equation solving, which use lexicographic directional (LD) derivatives to solve the equation system with Q-quadratic local convergence.[22]

Although the formulation of Watson et al. is simple and compact, it is only applied to multistream heat exchangers. Therefore, in this work, we extend their approach to the general integration problem to consider the simultaneous integration of any pinch-constrained resources. We generalize the variables in their formulation and add consideration for external utilities to create the generalized nonsmooth integration operator. Our method scales well with problem size, requires only equation-solving approaches, and can solve for process variables while simultaneously considering the integration of multiple resources. We also present a simple modification that identifies and solves threshold problems automatically. To our knowledge, this is the only approach to the targeting problem that has all of these properties, and the only solution presented explicitly for the general problem. Additionally, when applied to a heat integration problem without external utilities, our operator reduces to the equations of Watson et al.[21] However, we note that our approach currently applies only to the process integration problem, not combined integration and optimization, and thus cannot be expanded to additional degrees of freedom or objective functions other than the minimization of resource use.

In the sections below, we first present background on nonsmooth equation-solving methods and the generalized integration problem. We then detail the formulation of our nonsmooth integration operator, including variable selection for different resource types and modifications for the threshold problem. We follow with a series of examples with the operator being applied to a wide range of integration problems: carbon-constrained energy planning, a water threshold problem, a hydrogen conservation network with unknown process variables, and a combined mass and water integration problem with a process model. We conclude with a discussion of the features and limitations of our approach and our plans to address these in future work.

■ NONSMOOTH EQUATION SOLVING BACKGROUND

Nonsmooth analysis is a well-developed field, and many algorithms for nonsmooth equation-solving have been defined with desirable theoretical properties that are even competitive with their smooth counterparts. Such algorithms include Newton-type methods such as semismooth or linear-programming (LP) Newton.

The semismooth Newton method is analogous to the smooth Newton method; however, the evaluation of a generalized derivative element, $\mathbf{F}(\boldsymbol{x}_k)$, is required at each iteration instead of the Jacobian matrix. Therefore, the kth iteration is given by solving the linear equation

$$\mathbf{F}(\boldsymbol{x}_k)(\boldsymbol{x}_{k+1} - \boldsymbol{x}_k) = -\mathbf{f}(\boldsymbol{x}_k)$$

where $\mathbf{f}$ is semismooth and $\mathbf{F}(\boldsymbol{x}_k)$ is square and nonsingular.

The LP-Newton method relaxes the singularity requirement by iteratively solving the LP

$$\min_{\gamma \in \mathbb{R}, \boldsymbol{x} \in X} \gamma \quad \text{s. t. } \|\mathbf{f}(\boldsymbol{x}_k) + \mathbf{F}(\boldsymbol{x}_k)(\boldsymbol{x} - \boldsymbol{x}_k)\|_\infty \le \gamma \,\|\mathbf{f}(\boldsymbol{x}_k)\|_\infty^2,$$
$$\|\boldsymbol{x} - \boldsymbol{x}_k\|_\infty \le \gamma \,\|\mathbf{f}(\boldsymbol{x}_k)\|_\infty$$

where X is a polyhedral set.[23] For global convergence, a simple backtracking line search is performed after each LP iteration.[24] If $\mathbf{f}$ is piecewise differentiable,[25] the generalized derivatives are elements of the Bouligand (B-) subdifferential (the set of limiting Jacobians), and certain regularity conditions are met, both of these methods exhibit local Q-quadratic convergence.

Despite both the desirable performance of these algorithms and the ability of nonsmooth equations to describe many physical systems naturally and compactly, nonsmooth equation solving has generally been avoided due to the difficulty in calculating generalized derivative elements. Elements of the B-subdifferential cannot be found directly using automatic differentiation (AD) since they do not obey sharp calculus rules, nor can they be found from directional derivatives in the coordinate directions or component-wise limiting gradients.[26]

However, Khan and Barton have recently defined the LD-derivative, which follows a sharp chain rule, and can therefore be calculated using the nested computations of AD.[22] Additionally, the LD-derivative can be used to compute elements of the B-subdifferential for piecewise differentiable functions. Combined, these properties allow for the automatic calculation of useful generalized derivative elements for nonsmooth equation solving in complex settings and make explicitly nonsmooth approaches to process simulation viable.

In this work, we solve nonsmooth equation systems using either the semismooth or LP Newton method as specified. Both methods are supplied with generalized derivative elements calculated with vector forward AD for LD-derivatives as detailed by Khan and Barton and Barton et al. and implemented using operator overloading.[22,26,27] All algorithms are implemented in MATLAB 2017A.

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■ THE GENERAL INTEGRATION PROBLEM

This paper introduces a new approach to process integration using the nonsmooth methods detailed above. In particular, our nonsmooth method is designed to solve what we introduce as the "general integration problem," which describes the simultaneous minimization of the fresh supplies of an arbitrary number of resources. The following sections precisely define this general integration problem and thus the scenarios in which our approach can be applied.

Problem Structure and Assumptions. The general integration problem considers a set of resources, T, for integration. For each resource, there is a set of sources and a set of sinks, where each source or sink has a quality that changes with resource transfer and a constant state that determines the rate at which this quality changes. The source and sink qualities determine whether resource transfer is feasible between them based on enforced quality limits or driving force limitations. For each resource, the integration problem also incorporates a fresh utility that can supply any sink and a waste utility that can take in resource from any source. This system of resource sources and sinks is connected by a process model, which is dependent on the resource utilities and process variables. The objective of the general resource-targeting problem is then to determine the system specifications, either resource targets or process variables, at which minimal feasible resource use and waste production occur. Note that the general resource-targeting problem considers process integration but not optimization. For instance, this problem type does not include applications that minimize costs by selecting between multiple utilities with different costs and qualities. However, multiple external utilities can be incorporated in the general integration problem by including them in the set of sources.

Mathematically, the general integration problem can be represented by a system of equations describing a process model and a set of embedded optimization problems in parallel that minimize the fresh loads of each resource and are parametric in the process variables. For each resource type $n \in T$, we denote a vector of utilities, $\boldsymbol{y}_n = (R_{SR,n}, r_{SK,n})$, where $R_{SR,n}$ is the fresh resource supply and $r_{SK,n}$ is the waste resource flow. Then, for a process model, $\boldsymbol{h}$, and a set of process variables, $\boldsymbol{x}$, an outline of the structure of the general integration problem is

$$0 = \boldsymbol{h}(\boldsymbol{x}, \boldsymbol{y}_1, ..., \boldsymbol{y}_{|T|}),$$
$$\left.\begin{array}{ll} \{\boldsymbol{y}_n\} = \underset{\boldsymbol{y}_n}{\arg\min}\ R_{SR,n}(\boldsymbol{x}) & \\ \text{s. t.} & \text{resource balance holds} \\ & \text{resource transfer is feasible} \end{array}\right\} \forall\ n \in T$$

Note that the minimum for each embedded optimization problem is guaranteed to be unique because $R_{SR,n}$ is the objective function value and $r_{SK,n}$ is given explicitly in terms of $R_{SR,n}$ by the resource balance. The resource balance also guarantees that minimizing $R_{SR,n}$ is equivalent to minimizing $r_{SK,n}$.

Within this problem structure, we also make assumptions about the nature of the transfer of each resource. For each resource transferred from a set of sources SR_n to a set of sinks SK_n, using the notation presented by Foo,[28] we assume the sources $i \in SR_n$ have constant states $S_{i,n}$ that change in quality from $Q_{i,n}^{\text{in}}$ to $Q_{i,n}^{\text{out}}$ for a resource output $R_{i,n}$ according to $R_{i,n} = S_{i,n}(Q_{i,n}^{\text{in}} - Q_{i,n}^{\text{out}})$. Correspondingly, the sinks $j \in SK_n$ have constant states $s_{j,n}$ that change in quality from $q_{j,n}^{\text{in}}$ to $q_{j,n}^{\text{out}}$ for a resource input $r_{j,n}$ according to $r_{j,n} = s_{j,n}(q_{j,n}^{\text{out}} - q_{j,n}^{\text{in}})$. For resource transfer to be feasible between a source i and sink j, the source qualities must be higher than those of the sink by a minimum feasible quality difference $\Delta Q_{\min}$, that is, $Q_{i,n}^{\text{out}} \geq q_{j,n}^{\text{in}} + \Delta Q_{\min}$ and $Q_{i,n}^{\text{in}} \geq q_{j,n}^{\text{out}} + \Delta Q_{\min}$.

To describe this constrained resource transfer, there are a number of mathematical formulations that have been presented in the literature. Here we use the trans-shipment formulation of Papoulias and Grossmann[16] as an example of how to fully express the general integration problem using existing methods. In addition to being one of the first numerical approaches to resource-targeting, the trans-shipment formulation is still commonly used, particularly to describe cascade tables. This approach first defines a set of K_n quality intervals for each resource, partitioned by the sorted inlet and outlet qualities of the sources and sinks so that interval k spans higher quality values than $k + 1$. Thus, each interval with width $\Delta Q_{k,n}$, is predefined and independent of the current source or sink being considered. Given these quality intervals, we identify the sets $SR_{k,n} \subset SR_n$ and $SK_{k,n} \subset SK_n$, which are the sources and sinks, respectively, that have changes in quality that span interval k. The transfer constraints for each resource calculate the hypothetical resource flows $F_{k,n}$ that are available for transfer from interval k to the lower quality interval $k + 1$ using resource balances. (Note these flows are distinct from the individual source outputs $R_{i,n}$ because they are net quantities that consider all of the sources and sinks in k.) Feasibility is enforced by constraining the flows $F_{k,n}$ to be non-negative. Thus, the general integration problem, which we wish to solve for a selection of unknowns from $\boldsymbol{x}$ and $\boldsymbol{y}_n$, can be written as

$$0 = \boldsymbol{h}(\boldsymbol{x}, \boldsymbol{y}_1, ..., \boldsymbol{y}_{|T|}),$$
$$\left.\begin{array}{l} \{\boldsymbol{y}_n\} = \underset{\boldsymbol{y}_n, F_{1,n}, ..., F_{K_n-1,n}}{\arg\min}\ R_{SR,n}(\boldsymbol{x}), \\ \text{s. t.} \\ 0 = R_{SR,n} - F_{1,n} + \Delta Q_{1,n}\left(\sum_{i\in SR_{1,n}} S_{i,n} - \sum_{j\in SK_{1,n}} s_{j,n}\right), \\ 0 = F_{k-1,n} - F_{k,n} + \Delta Q_{k,n}\left(\sum_{i\in SR_{k,n}} S_{i,n} - \sum_{j\in SK_{k,n}} s_{j,n}\right), \\ \quad \forall\ k \in \{2, ..., K_n - 1\}, \\ 0 = F_{K_n-1,n} - r_{SK,n} + \Delta Q_{K_n,n}\left(\sum_{i\in SR_{K_n,n}} S_{i,n} - \sum_{j\in SK_{K_n,n}} s_{j,n}\right), \\ 0 \leq F_{k,n}, \quad \forall\ k \in \{1, ..., K_n - 1\}, \end{array}\right\} \forall\ n \in T$$

Although the above formulation is a complete representation of the general integration problem as embedded linear programs, when qualities are unknown, it is a nontrivial process to determine the quality intervals, k, and the mapping of the sources and sinks to these intervals to find $SR_{k,n}$ and $SK_{k,n}$ as functions of the qualities. Therefore, the qualities in the problem cannot be unknowns in $\boldsymbol{x}$ and must be known a priori. As discussed in the introduction, these limitations have been addressed using the pinch location method and disjunctive formulations;[17−20] however, these approaches require solving potentially nonconvex NLPs with smooth approximations or large MINLPs. In this work, we present an alternative approach to handling unknown qualities that uses

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Table 1. Resources, Qualities, and States for a Sample of Integration Formulations (Adapted from Foo[28])

integration types	resource quantity	quality	state
heat[2]	heat transfer rate	temperature	heat capacity flow rate
mass[3]	contaminant mass load flow rate	concentration in reference stream	scaled solvent mass flow rate
fixed-load water[4]			
RCN[30]	mass flow rate	cumulative difference in property loads (e.g., contaminant mass flow rate)	reciprocal of a linear mixing property, sorted by increasing property value (e.g., contaminant concentration)
fixed-flow water[31]			
hydrogen[6]			
oxygen[7]			
carbon[9]			
carbon-constrained energy[8]	electrical power	cumulative difference in emission masses	reciprocal emission factor, sorted by increasing emissions
inventory[10]	time	cumulative volume	usage or production rate

nonsmooth equations to avoid solving challenging NLPs and to improve scaling with the number of sources and sinks integrated.

Resource Types. As mentioned above, there is a significant body of work proposing different integration types, each of which can be described by the general integration problem. The resources, states, and qualities for a representative sample of integration types are summarized in Table 1. Note that the same terminology is often used to refer to integration formulations with different selections of resources, and formulations that are fundamentally the same are referred to in different ways. For example, water integration approaches can be of either the "fixed-load" or "fixed-flow rate" type. The former problem considers process units with constant water flow rates and is treated the same as a mass integration problem with the contaminant mass as the integrated resource; whereas, the latter directly integrates the water flows.[29] Similarly, carbon integration can either integrate power production with different emission quantities, or flows between CO_2 sources and sinks.[8,9] In addition, both of the latter water and carbon formulations are types of resource conservation networks (RCNs), in which the resource is a material flow that can be reused directly without considering heat or mass transfer.[28]

■ GENERALIZED NONSMOOTH OPERATOR FORMULATION

In this section, we introduce a new approach to solving the general resource-targeting problem that uses explicitly nonsmooth equations to improve scaling compared to current approaches while retaining the ability to solve for unknown process variables including qualities. This method also has the flexibility to automatically identify threshold problems. As an alternative to the nonsmooth LPs of the trans-shipment formulation or nonconvex MINLPs of the pinch location method, our nonsmooth formulation uses systems of equations to express the solutions of the embedded minimization problems. Each of these nonsmooth systems, or nonsmooth integration operators, consists of two equations per integrated resource: an overall resource balance, which ensures that resource transfer is feasible based on availability, and a resource balance below potential resource transfer pinch points, which enforces optimal transfer given quality limits or driving force limitations. The nonsmooth integration operators for each resource are coupled with the process model, and the resulting system can be solved efficiently using the nonsmooth equation-solving methods described in the Background.

In each nonsmooth integration operator, the pinch point balance uses a simple nonsmooth expression to enforce both the feasibility of resource transfer due to quality constraints and minimal resource use. For resource transfer to be feasible, at each pinch candidate quality, the total resource quantity produced by the lower quality sources must be less than or equal to the capacity of the sinks (including the waste resource flow) that can accommodate resource at these qualities, which in the trans-shipment formulation is expressed by the directionally constrained resource flows from high to low quality. In addition, according to pinch analysis theory, for resource transfer to be optimal, there must exist at least one quality pinch point for the resource, below which the source production and sink capacities are equal. Otherwise, fresh resource must be used to fulfill the sink demands below each quality level and is cascaded down to the waste resource sink. Therefore, both the feasibility and optimality criteria can be easily expressed using an explicitly nonsmooth equation by setting the minimum resource balance over the problem qualities to zero.

Because the general integration operator assumes constant state sources and sinks, this property can be equivalently enforced with a pinch point balance that considers a finite set of potential pinch points. Again drawing on pinch analysis, for constant state sources and sinks, the potential pinch point candidates are the inlet source and sink qualities that define distinct quality intervals for the problem. Additionally, enforcing feasibility at each of these points ensures feasibility at all qualities between them. Therefore, the pinch point balance in our nonsmooth integration operator considers potential pinch points in this set.

Then, neglecting the index n for clarity, the resulting nonsmooth integration operator for a single given resource is

$$0 = \sum_{i \in SR} S_i(Q_i^{\text{in}} - Q_i^{\text{out}}) - \sum_{j \in SK} s_j(q_j^{\text{out}} - q_j^{\text{in}}) + R_{SR} - r_{SK} \tag{1}$$

$$0 = \min_{p \in P}\{RBP_{SK}^p - RBP_{SR}^p\} + r_{SK} \tag{2}$$

where P is the finite index set of pinch point candidates and

$$RBP_{SR}^p := \sum_{i \in SR} S_i[\max\{0, Q^p - Q_i^{\text{out}}\} - \max\{0, Q^p - Q_i^{\text{in}}\} - \max\{0, Q^{\min} - Q^p\} + \max\{0, Q^p - Q^{\max}\}], \quad \forall\, p \in P$$

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$$RBP_{SK}^{p} := \sum_{j \in SK} s_j[\max\{0, (Q^p - \Delta Q_{\min}) - q_j^{\text{in}}\} - \max\{0, (Q^p - \Delta Q_{\min}) - q_j^{\text{out}}\} + \max\{0, (Q^p - \Delta Q_{\min}) - q^{\max}\} - \max\{0, q^{\min} - (Q^p - \Delta Q_{\min})\}], \quad \forall p \in P$$

where $\Delta Q_{\min}$ is the minimum feasible quality difference between a source and sink at which resource transfer can occur and the source qualities at the potential pinch points are

$$Q^p = \begin{cases} Q_i^{\text{in}}, & \forall p = i \in SR \\ q_j^{\text{in}} + \Delta Q_{\min}, & \forall p = j \in SK \end{cases}$$

The expressions RBP_{SR}^p and RBP_{SK}^p are the cumulative source and sink resource quantities that can be exchanged at qualities lower than Q^p. The nonsmooth max terms capture the position of the inlet and outlet qualities of each source or sink in relation to the potential pinch point quality, and therefore whether the source or sink should be, either partially or wholly, included in the resource balance. Thus, these expressions allow us to avoid the explicit ordering and construction of quality intervals as required in the trans-shipment formulation. $Q^{\min,\max}$ and $q^{\min,\max}$ are the minimum and maximum qualities across the sources or the sinks, respectively, and the max terms containing these variables create nonphysical extensions to the cumulative resource quantities, which avoid additional singular regions or infinite solutions by ensuring the difference between the source and sink resource balances is always defined. Note that, so the pinch point balance correctly selects resources transferred below the pinch quality, we define the inlet and outlet qualities such that $Q_{\text{in}} \geq Q_{\text{out}}$ and $q_{\text{out}} \geq q_{\text{in}}$.

For the case of heat integration, Watson et al. provide a detailed justification for why this system of equations enforces the existence of a pinch point.[21] This analysis can be applied analogously to the case of a general resource whose transfer obeys the linear relations $R_{i,n} = S_{i,n}(Q_{i,n}^{\text{in}} - Q_{i,n}^{\text{out}})$ and $r_{j,n} = s_{j,n}(q_{j,n}^{\text{out}} - q_{j,n}^{\text{in}})$ where the qualities have values that increase with increasing purity as described in the section below. Therefore, for any pinch-constrained resource transfer with this property, eqs 1 and 2 will enforce a pinch point and thus describe optimal resource transfer.

Graphically, the source and sink balances define quality versus quantity composite curves, and eqs 1 and 2 ensure that the source composite curve is always at higher qualities than the sink composite curve and that the composite curves touch at a pinch point. Figure 1 illustrates this graphical conceptualization of the resource balances and nonsmooth integration operator.

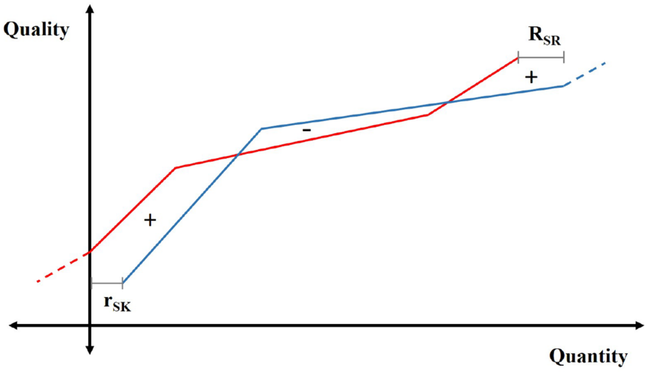

Figure 1. Graphical illustration of the nonsmooth integration operator. The red and blue plots are the source and sink composite curves, the qualities of which are generated by RBP_{SR}^p and RBP_{SK}^p, respectively, at each potential pinch point, and the dashed lines show the curve extensions. The sign of $RBP_{SK}^p - RBP_{SR}^p + r_{SK}$ is indicated for each region. These particular curves do not satisfy eqs 1 and 2 because $RBP_{SK}^p - RBP_{SR}^p + r_{SK}$ is not constrained to be nonnegative.

■ DETERMINING INTEGRATION VARIABLES

Before the nonsmooth operator can be used to solve a general integration problem, qualities and states must be defined for each resource, and often the provided data must be preprocessed in order to calculate these state and quality values. The sections below present some special considerations when defining and calculating integration variables, particularly for RCNs.

Quality Sign Selection. The pinch point balance as defined above calculates the net quantity of resource transferred at qualities below the pinch point quality, including the waste resource quantity. Therefore, our formulation requires the waste resource flow to have the lowest quality value of all the sources and sinks and the fresh resource to have the highest. Thus, while the equations as written are applicable to any general integration problem, the qualities used here may need to be transformed from those presented in other works. For example, in many types of integration problems, including common RCNs, the qualities are cumulative values such as property loads. In the typical RCN formulation, summarized by Foo,[28] the cumulative property loads, P_i and p_j, are determined by summing up changes in load from a selected fresh resource load $P^{\min} = p^{\min}$ according to

$$P_1^{\text{out}} = P^{\min} \tag{3}$$

$$P_i^{\text{in}} = P_i^{\text{out}} + \Delta P_i, \quad \forall i \in SR \tag{4}$$

$$P_i^{\text{out}} = P_{i-1}^{\text{in}}, \quad \forall i \in SR, \quad i \neq 1 \tag{5}$$

for the source loads and

$$p_1^{\text{in}} = p^{\min} \tag{6}$$

$$p_j^{\text{out}} = p_j^{\text{in}} + \Delta p_j, \quad \forall j \in SK \tag{7}$$

$$p_j^{\text{in}} = p_{j-1}^{\text{in}}, \quad \forall j \in SK, \quad j \neq 1 \tag{8}$$

for the sinks. $P^{\min}$ is usually chosen to be zero; however, the actual value is arbitrary since only the changes in property load are relevant. Therefore, in this formulation, the source exists at the lowest property load and the sink at the highest, so our nonsmooth operator cannot use this definition of the property load as the quality in the integration problem.

Instead, for RCNs, we define the qualities by subtracting the changes in property load from a selected fresh resource quality $Q^{\max} = q^{\max}$. For the source qualities,

$$Q_1^{\text{in}} = Q^{\max} \tag{9}$$

$$Q_i^{\text{out}} = Q_i^{\text{in}} - \Delta Q_i, \quad i = 1, ..., |SR| \tag{10}$$

$$Q_i^{\text{in}} = Q_{i-1}^{\text{out}}, \quad i = 2, ..., |SR| \tag{11}$$

and for sink qualities,

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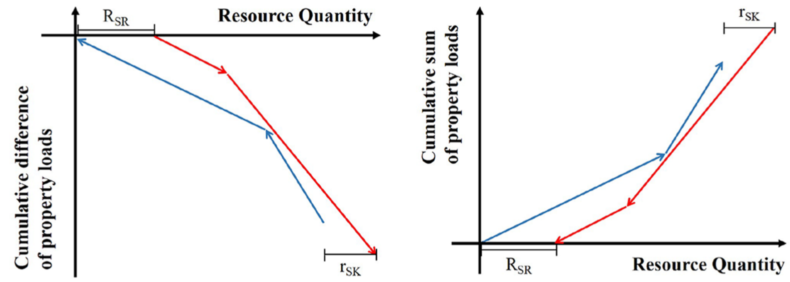

(a) Defining qualities through a cumulative difference so the source is at the highest quality value and the sink at the lowest.

(b) Reformulating the integration operator to include the fresh resource below the pinch point.

Figure 2. Illustration of approaches to determining problem qualities that can be used with our method. The source and sink composite curves are red and blue, respectively.

$$q_1^{\text{out}} = q^{\max} \tag{12}$$

$$q_j^{\text{in}} = q_j^{\text{out}} - \Delta q_j, \quad j = 1, ..., |SK| \tag{13}$$

$$q_j^{\text{out}} = q_{j-1}^{\text{in}}, \quad j = 2, ..., |SK| \tag{14}$$

where $\Delta Q_i = \Delta P_i$ and $\Delta q_j = \Delta p_j$. Again, the value of $Q^{\max}$ is arbitrary and is usually set to zero. Now, the source is at the highest quality value, the sink is at the lowest, and we maintain $Q_i^{\text{in}} \geq Q_i^{\text{out}}$ and $q_j^{\text{out}} \geq q_j^{\text{in}}$. Thus, we can apply the nonsmooth integration operator to these qualities.

To easily transfer the nonsmooth integration operator between problems, in this work, for cases where the high purity streams have low property loads, we always use this approach of defining the qualities through a cumulative difference so that they are consistent with the operator formulation. If desired, an alternative approach is to interchange the source and sink variables in the operator. With this change, the pinch point balance now includes the fresh resource source instead of the waste resource sink in the resource exchanged below the pinch point. Figure 2 visually demonstrates the difference between these two approaches. Figure 2a shows the transformation of the cumulative loads so that the waste sink is correctly included below the potential pinch point qualities, and Figure 2b shows swapping the sources and sinks so that the qualities of the source composite curve are lower than those of the sinks.

Sorting Property Values. In the integration problems such as RCNs discussed above, in which the qualities are cumulative values, the qualities, and thus the calculated resource targets, are highly dependent on the ordering of the sources and sinks. For RCNs where resource reuse is limited by composition, El-Halwagi et al. prove that resource targets are minimized if the sources and sinks are each sorted by increasing concentration,[31] and Kazantzi and El-Halwagi extend this principle to general property integration where the sources and sinks are also sorted by their property values according to decreasing purity.[30] To perform this sorting, most approaches to RCNs require all source and sink properties, which are the states S_i or s_j in the general integration problem and are usually the reciprocal of the property value, to be known a priori. The few formulations that can solve for the properties are typically superstructures that must be solved with MINLPs that scale poorly with problem size. However, sorting algorithms are inherently continuous but nonsmooth with respect to the sorted elements because the sorted order only changes at the finite set of points at which the two elements are equal. Thus, they can be incorporated directly into the nonsmooth integration operator so that we can solve for properties and states in RCNs using only equation-solving methods.

To incorporate sorting when it is required, we treat the sorting algorithm as a nonsmooth function that maps the unsorted input to a sorted output. Then, the overall nonsmooth system that simulates the integrated process is the composition of the integration operator and the sorting operation. In practice, we preprocess the problem data to create lists of property and property load pairs for the sources and sinks and sort each list in order of nonincreasing purity (e.g., nondecreasing contaminant concentration).

By selecting a sorting algorithm for which we can calculate LD-derivatives, because they obey a sharp chain rule, we can find the generalized derivative elements for the composite function by supplying the derivatives for the sorted pairs with respect to the unknown variables to the integration operators. Then these generalized derivative elements can be used to solve the equation system using the nonsmooth equation-solving methods detailed above. For all of the examples in this work, we have used a simple bubble sort algorithm as shown in Figure 3. Because the only operations required are taking the max or min of two functions and these operations are performed the same number of times for any input of a given size, we can easily incorporate this sorting algorithm into code for the nonsmooth integration operator and apply AD methods to calculate the LD-derivatives for both the sorting process and the composite integration equations. Therefore, with the use of this method, the nonsmooth integration operator can solve for property values in RCNs, a feature unique to our approach.

```
Input: An unsorted list, A, with entries A[1], ..., A[m]
Output: The list A, with the m entries sorted in order of increasing values
for i ← 1 to m do
    for j ← 1 to m − 1 do
        a ← min(A[j], A[j + 1])
        b ← max(A[j], A[j + 1])
        A[j] ← a
        A[j + 1] ← b
return A
```

Figure 3. A simple bubble sort algorithm.

Extension to the Threshold Problem. Another significant advantage of modeling integrated systems using nonsmooth equations is the ability to easily incorporate additional

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scenarios such as threshold problems. A threshold scenario can occur for a resource when the resource utilities are unknowns, and all other variables that the resources flows depend on, the process variables, are fixed, that is, the traditional targeting problem. For this problem type, it may be infeasible for a pinch point to occur, in which case, resource usage will be optimal when the fresh or waste resource flow is zero and the pinch point balance in eq 2 will be positive instead of zero. To capture this behavior, a simple min function wrapper can be added to eq 2 so it becomes

$$0 = \min\{\min_{p \in P}\{RBP_{SK}^{p} - RBP_{SR}^{p}\} + r_{SK}, R_{SR}, r_{SK}\} \quad (15)$$

This expression captures all possible cases for the basic resource-targeting problem: either a pinch point exists, so the pinch point balance is zero and the utilities are nonnegative, or a pinch point does not exist, so the pinch point balance is positive, and one of the utilities is zero and the other is nonnegative. Therefore, when the process variables related to a resource are known, eq 15 should be used in place of eq 2 for that resource. Then, unlike other approaches to process integration, the nonsmooth operator will identify threshold cases even if they are not known a priori.

It is important to note that eq 15 should not be applied when process variables are unknown. In this case, the free process variables ensure that a pinch point is obtainable. Thus, if a pinch point is not enforced at the solution, both the fresh and waste resource flows could feasibly be reduced, and the solution does not describe a system under optimal resource reuse. Additionally, if one of the external resource utilities is specified to be zero, the integration operator will be underdetermined because eq 15 will automatically be satisfied. Therefore, we use eq 2 in these scenarios to ensure the existence of a pinch point.

Nonsmooth Operator Implementation. To solve the general integration problem using the nonsmooth integration operator, one operator is constructed for each integrated resource using the specifications detailed above. The states and qualities for each resource are identified along with any operations, including sorting, required to calculate them from the problem data and unknowns. Then, the appropriate pinch point balance is selected from eqs 2 and 15 depending on whether the resource states and qualities are dependent on any unknown process variables. Once the integration operators are constructed, they are combined with a process model to form a system of nonsmooth equations.

The resulting system can be solved for different selections of unknowns, for which the degrees of freedom are determined by the size of the equation system, using the nonsmooth equation-solving methods described in the background section. Regardless of the number of sources and sinks in the problem, the integration operator for each resource contributes only two equations to the system. This feature results in a solution process that scales compactly with problem size compared to optimization approaches such as the pinch location method that at best scale quadratically with both the number of constraints and binary variables. Additionally, this strategy is applicable to any generalized integration problem, and we show its implementation for a variety of specific problems in the examples below.

■ EXAMPLE PROBLEMS

In this section, we present a series of examples that demonstrate the use and potential of the nonsmooth integration operator. These examples begin with a classic resource-targeting problem and increase in complexity to include unknown process variables, process models, and the simultaneous integration of multiple resources. They also cover a wide range of resource types to show the utility of a truly generalizable approach to the integration problem.

Example 1: Carbon-Constrained Energy Planning. In this example, we demonstrate the ability of the nonsmooth integration operator to solve for fresh and waste targets for any general integration problem in which the resource transfer is limited by a pinch point. We consider the carbon-constrained energy-planning problem presented by Tan and Foo,[8] which examines how best to utilize energy sources under carbon limits during the transition to clean energy. In this scenario, there is a set of geographical regions, which each have an expected energy consumption and a CO_2 emission limit for the planning horizon, and a set of energy resources with different emission factors (tonnes CO_2 emitted per TJ of energy produced) and availabilities. The provided data for this example is given in Table 2. We need to determine the

Table 2. Problem Data for Example 1[8]

energy resource	emission factor (t CO_2/TJ)	available resource (10^6 TJ)	demand region	expected demand (10^6 TJ)	emission limit (10^6 t CO_2)
coal	105	0.6	region I	1.0	20
oil	75	0.8	region II	0.4	20
natural gas	55	0.2	region III	0.6	60

quantity of emission-free renewable energy sources required to meet the emission targets for each region and the quantity of high-emission sources that will go unused.

The first step in solving this problem with the generalized nonsmooth operator is determining the problem states and qualities. Here, the energy is the resource transferred from the production sources to the different regions where it is consumed, and Tan and Foo use cumulative emission versus energy pinch plots to solve the integration problem for optimal energy transfer. As for standard RCNs, the cumulative emission loads are determined by sorting the source and sinks by increasing emission factor. This approach suggests that the cumulative emission loads and reciprocal emission factors can be considered as the qualities and states for this problem, respectively, which is consistent with our definitions since the energy transfer is constrained by the net carbon loads released from the energy sources and gained by the different geographic regions and the change in carbon load for each source or sink is proportional to its limiting emission factor. However, Tan and Foo's approach to calculating the cumulative emission loads, as in eqs 3−8, results in the zero-emission energy source having the lowest cumulative load and the excess power sink the highest. Therefore, for this problem, to be consistent with the nonsmooth integration operator, we chose to use eqs 9−14 to calculate the qualities as the cumulative difference of the changes in emission load.

We then transformed the problem data to these states and qualities to be used with the integration operator. The resulting values are shown in Table 3. For the energy resources, the

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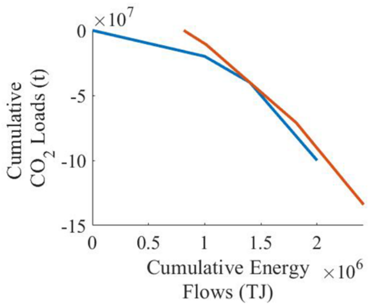
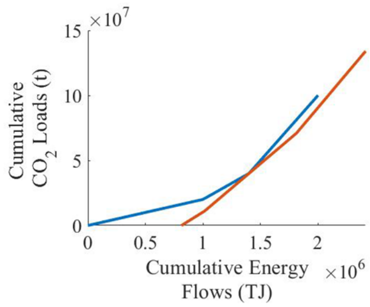
Table 3. Calculated States and Qualities Used in the Nonsmooth Integration Operator for Example 1

energy resource	S_i (10^{-3} TJ/t CO_2)	Q_i^{in} (10^6 t CO_2)	Q_i^{out} (10^6 t CO_2)	demand region	s_j (10^{-3} TJ/t CO_2)	q_j^{in} (10^6 t CO_2)	q_j^{out} (10^6 t CO_2)
natural gas	18.2	0	−11	Region I	50	−20	0
oil	13.3	−11	−71	Region II	20	−40	−20
coal	9.5	−71	−134	Region III	10	−100	−40



(a) Quality versus quantity plot showing the negative qualities used in the integration operator.



(b) Standard pinch representation.

Figure 4. Pinch plots for Example 1. The source and sink composite curves are red and blue, respectively.

states can be calculated directly as the reciprocals of the emission factors. Then, the changes in emission load for each source can be found from the known energy flows, R_i, according to $\Delta Q_i = R_i/S_i$. Using these values, we sorted the $(S_i, \Delta Q_i)$ pairs by nonincreasing state (nondecreasing emission load) to be used in eqs 9−11 to find Q_i^{out} and Q_i^{in}. Here, the sources can be sorted using any algorithm since the emission factors can be calculated explicitly from the problem data. Note that the sorting procedure changes the order of the energy resources as shown in Table 3. For the demand regions, we are given r_j and Δq_j, so we determined the states from $s_j = r_j/\Delta q_j$ and found q_j^{in} and q_j^{out} from the sorted $(s_j, \Delta q_j)$ pairs and eqs 12−14. We also set $\Delta Q_{min} = 0$ since no driving force is required for power transfer between the energy resources and demand regions.

Once the states and qualities are calculated, the appropriate operator equations can be applied to these state and quality values, and the system can be solved using one of the nonsmooth equation-solving methods described above. For this problem, since all process variables are known, we used eq 15 in addition to eq 1 to ensure potential threshold problems are identified. We solved this system for R_{SR} and r_{SK} using the semismooth Newton method to give $R_{SR} = 0.81 \times 10^6$ TJ and $r_{SK} = 0.81 \times 10^6$ TJ as expected. Figure 4 shows the pinch plot at the solution both in terms of the qualities used in the integration operator and using the standard representation for this problem type.

Semismooth Newton is a good equation-solving method for any application in which all unknowns are resource targets because the resource targets are not present in any nonsmooth terms, and therefore, the algorithm cannot encounter any singular generalized derivative elements along its solution path. Accordingly, in this example, semismooth Newton converged to the solution for all initial guesses tested in only 1 or 2 iterations.

Example 2: Water Threshold Problem. The next example shows the use of our integration operator to automatically identify and solve threshold problems. Here, we consider two cases presented by Foo for fixed-flow water integration with the flow rates and concentrations given in Tables 4 and 5.[32] The first data set describes a zero-discharge network in which all of the source water can be used by the system, and the second a water network that requires no fresh water feed.

Table 4. Zero Discharge Problem Data for Example 2[32]

water source	flow rate (g/min)	concentration (ppm)	water sink	flow rate (g/min)	concentration (ppm)
1	20	20	1	50	20
2	50	100	2	20	50
3	40	250	3	100	400

Table 5. Zero Fresh Resource Problem Data for Example 2[32]

water source	flow rate (t/h)	concentration (ppm)	water sink	flow rate (t/h)	concentration (ppm)
1	500	100	1	1200	120
2	2000	110	2	800	105
3	400	110	3	500	80
4	300	60			

Analogous to the carbon integration problem in Example 1, since Foo approaches these problems using cascade tables with water and cumulative contaminant flow rates sorted by increasing contaminant concentration, we selected the cumulative difference of the contaminant flow rates and the reciprocal source and sink concentrations as the qualities and states, respectively. We then transformed the problem data to these states and qualities and applied eqs 1 and 15 with $\Delta Q_{min} = 0$ since no driving force is required for resource transfer. For both problems, we solved for the fresh and wastewater flow rates using the semismooth Newton method, and we converged to the solutions in 1 to 2 iterations across a wide range of initial guesses. As desired, for the zero-discharge problem, the integration operator determined the correct zero wastewater flow rate and a fresh water flow rate of 60 g/min, and for the other, it found a zero fresh water flow rate and a wastewater flow rate of 700 t/h.

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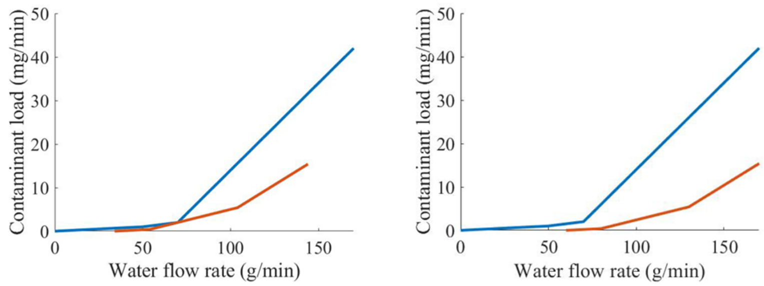

(a) Equation 2 enforces a pinch point.

(b) Equation 15 correctly identifies the solution in the threshold case.

Figure 5. Comparison of approaching the zero-discharge threshold problem using eqs 2 and 15. The source and sink composite curves are red and blue, respectively.

Table 6. Problem Data for Example 3.[28]

sink	hydrogen consumption (mol/s)	concentration (mol %)	source	hydrogen production (mol/s)	concentration limit (mol %)
HCU	2495.0	19.39	HCU	1801.9	25.0
NHT	180.2	21.15	NHT	138.6	25.0
CNHT	720.7	24.86	CNHT	457.4	30.0
DHT	554.4	22.43	DHT	346.5	27.0
			SRU	623.8	-
			CRU	415.8	20.0

In comparison, using eq 2 for these targeting problems does not correctly identify threshold cases, giving infeasible waste and fresh water flow rates of −26 g/min and −9.1 t/h for the zero-discharge and zero fresh flow cases, respectively. Pinch plots comparing the results from using eqs 2 and 15 for the zero-discharge problem are given in Figure 5. Although not appropriate for identifying threshold problems in resource-targeting cases, when solving for process variables, using eq 2 enforces pinch points in the system to ensure resources are used as efficiently as possible as demonstrated in the examples below.

Example 3: Hydrogen Conservation Network. The next example highlights the ability of our formulation to solve for process variables in integration problems, particularly properties that must be sorted to determine optimal resource transfer. In this example, we examine a refinery hydrogen recovery network similar to that from Alves and Towler.[6,28] The network contains four hydrogen-consuming processes, a hydrocraking unit (HCU), a naphtha hydrotreater (NHT), a cracked naphtha hydrotreater (CNHT), and a diesel hydrotreater (DHT), and two in-plant hydrogen-producing facilities, a catalytic reforming unit (CRU) and a steam-reforming unit (SRU). There is also an external feed of hydrogen that can be purchased with an impurity content of 6.5%. The limiting data for the hydrogen network is given in Table 6. We assume there is potential to upgrade the SRU to produce higher-purity hydrogen, and we wish to determine the required flow rate of external hydrogen and purity of the hydrogen produced by the SRU to achieve a waste flow rate of 100 mol/s.

To solve this problem using the integration operator, we note that the system is a RCN of the same form as the water networks in Example 2. Therefore, we again selected qualities that are the cumulative differences of the impurity loads and states that are the reciprocal concentrations. In this case, because the external hydrogen source has a fixed impurity concentration and is not the highest possible purity, we treated it as an additional source stream with an unknown flow rate and set R_{SR} = 0. Because process variables are unknown, we applied eqs 1 and 2, and to solve for the SRU concentration, we used the bubble-sort algorithm presented in Figure 3 to determine the generalized derivatives of the functions composed with the sorting process. Using these generalized derivatives in the semismooth Newton algorithm gave a SRU hydrogen purity of 7.00 mol % and an external hydrogen feed of 268.8 mol/s in 1 to 2 iterations. Figure 6 gives the pinch plot for this result, which shows the node corresponding to the SRU correctly sorted among the other hydrogen sources to achieve a pinch point in the optimally integrated system.

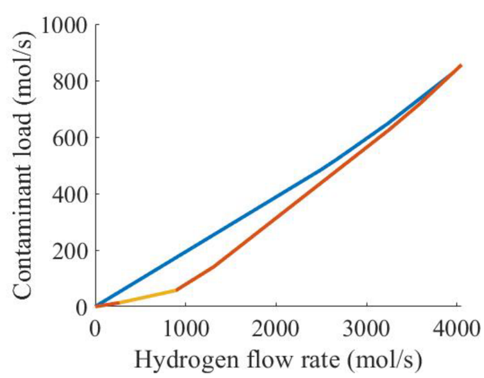

Figure 6. Pinch plot for Example 3. The source and sink composite curves are red and blue, respectively. The source node corresponding to the SRU is highlighted in yellow.

In comparison to our approach, most other integration methods are unable easily to solve for state variables in RCNs due to the complexity of the sorting operations. Historically, determining these variables has required repeatedly solving the integration problem at different quality values or using large mixed-integer program.[6,33] Instead, our approach allows for the efficient identification of network properties by solving only a single system of equations. However, when using the nonsmooth integration operators to solve for process variables, to ensure the system is well-defined, it is important to be

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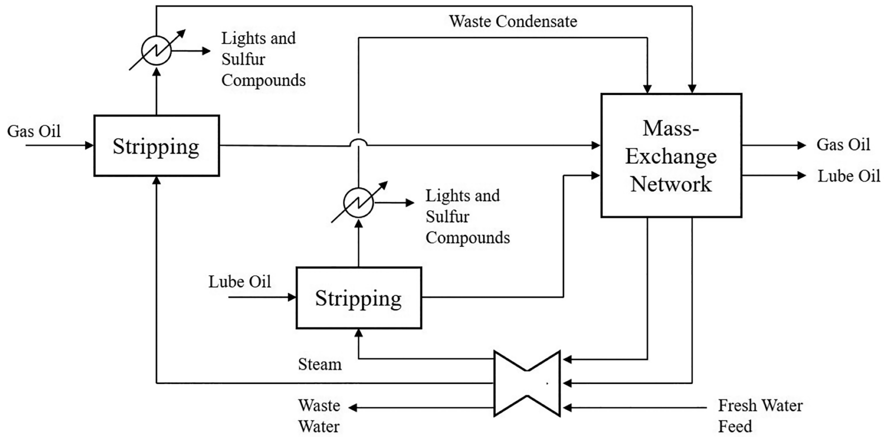

Figure 7. Simplified process diagram for the proposed process in Example 4.

mindful of where the process variables are present in the operator equations when they are transformed to the appropriate qualities and quantities. For example, when using flow rate and property data for a RCN, the overall resource balance in eq 1 is only a function of the resource flow rates, so the integration operator is only able to solve for one property value. As demonstrated in the next example, this limitation can be mitigated by including a process model or integration operators for other resources to fix unknown properties.

Example 4: Dephenolization and Recycling of Aqueous Wastes. This final example extends the use of the integration operator to a problem that includes both a process model and the joint integration of multiple resources, the flows of which are dependent on the process variables. We have adapted a problem presented by El-Halwagi,[34] which involves an oil recycling facility that uses steam strippers to remove sulfur and other light compounds from the oil streams. The main contaminant of concern in the stripper condensates is phenol, which can be removed through transfer to the oil streams in a MEN. Here, we analyze a proposed retrofit of a facility that processes lube oil, which considers both adding capacity for recycling gas oil and the possibility of reuse of the stripper condensate after dephenolization to reduce both fresh water consumption and wastewater production. The proposed plant therefore includes two steam strippers, a mass integration network for reducing the condensate phenol concentration, and a water network to reuse the water output in the strippers. Figure 7 shows a simplified process diagram for the plant.

We wish to determine the minimum attainable fresh and wastewater flow rates as well as the phenol concentrations and water flow rates throughout the system, particularly in the new gas oil stripper. We require that no external utilities are used in the MEN and that the concentrations of the two stripper condensates are the same when they exit the MEN. We also assume that there are phenol concentration limits in the boilers that limit the inlet concentrations to the strippers and that the mass of phenol transferred in each stripper is constant. (This assumption can be replaced by more complex stripper models if desired.) The parameters for this system are given in Table 7.

To solve this problem, we applied two integration operators, one for the allocation of water and one for the mass exchange of phenol. As in Example 2, the water network qualities and states were taken as the cumulative differences in contaminant (phenol) load and the reciprocal contaminant concentration, respectively. Again, we obtained the sorted mass fractions

Table 7. System Parameters for Example 4[a]

stream	flow rate (kg/h)	inlet mass fraction	outlet mass fraction
lube oil	5	0.005	0.015
gas oil	3	0.010	0.030
stripper 1 steam	2.5	0.005	z_2
stripper 2 steam	z_1	0.002	z_4
stripper 1 wastewater	2.50	z_2	z_3
stripper 2 wastewater	z_1	z_4	z_5
fresh water	z_6		
wastewater	z_7		

stripper 1 mass load: 0.11 kg/h
stripper 2 mass load: 0.03 kg/h
equilibrium relation for lube oil: $y = 2.00x_1$
equilibrium relation for gas oil: $y = 1.53x_2$
minimum MEN concentration difference in oil streams: $\epsilon = 0.001$

[a]Lube oil is processed in stripper 1, and gas oil in stripper 2.

required to determine these states and qualities using the bubble sort algorithm. For mass integration, the qualities and states are analogous to those defined in the common heat integration problem; as shown in Table 1, the qualities are the stream contaminant concentrations, and the states are the stream flow rates. However, to ensure that concentrations can be compared in a meaningful way, the equilibrium expressions must be used to transform the stream concentrations to their equivalent values in a selected reference stream, which we chose as the stripper condensate. The stream flow rates in the system must also be transformed to ensure the overall resource balance holds. Additionally, in this problem, the minimum MEN concentration difference is given in reference to the oil streams, so we included this concentration difference in our transformations, that is, $y = m(x + \epsilon)$, and set $\Delta Q_{\min} = 0$.

In addition to the two integration operators, we also included process equations describing the constant mass transfer in the strippers and equating the MEN water outlet concentrations:

$$m_1 = V_1(z_2 - y_{0,1})$$

$$m_2 = z_1(z_4 - y_{0,2})$$

$$z_3 = z_5$$

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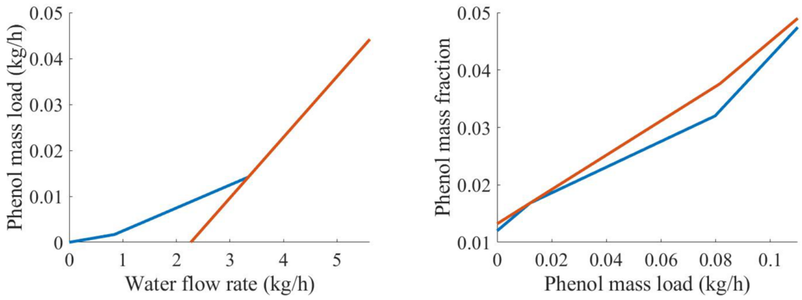

(a) Water allocation network.

(b) Mass integration network.

Figure 8. Composite curves for the optimized dephenolization system. The source and sink curves are red and blue, respectively.

where $m_{1,2}$ are the mass loads of phenol transferred in each stripper, V_1 is the steam flow rate through Stripper 1, and $y_{0,1}$ and $y_{0,2}$ are the inlet steam concentrations to the strippers.

With these process equations, we obtained a nonsmooth system of seven equations which we solved for seven unknowns using a semismooth Newton method to give $z = (0.84, 0.049, 0.013, \ 0.038, 0.013, 2.27, 2.27)$. For a range of initial guesses, the semismooth Newton method quickly converged to the solution in 3 to 9 iterations. The mass and water composite curves for this solution are given in Figure 8 and demonstrate that our solution method produces the optimal pinch behavior. These results highlight the unique ability of our approach to simultaneously integrate multiple resources that are coupled through process variables.

■ CONCLUSIONS

In this work, we present a new, generalizable, and efficient approach for solving resource-targeting problems using compact, nonsmooth operators. These operators are nonsmooth systems of only two equations per integrated resource, regardless of the number of sources and sinks in the system. New methods in AD for LD-derivatives make it easy to solve these operators, in combination with process models, for resource targets or any process variable, including states that require sorting. We include a series of examples that, together, demonstrate the ability of our approach to automatically identify threshold problems, solve for sorted qualities, include process models, and be applied to any pinch-constrained resource, including multiple resources simultaneously.

The current formulation is only applicable to problems with preclassified sources and sinks and can only consider a single contaminant for each resource. However, because our approach is nonsmooth, there is the potential to easily incorporate other work that uses explicit max and min expressions to address unclassified streams and multicontaminant problems while retaining the desirable features of our methods.[33,35] Additionally, our current method is only applicable to process integration, not optimization, and has degrees of freedom limited by the size of the equation system. Nevertheless, with advances in nonsmooth optimization methods, nonsmooth integration operators can be embedded in mathematical programming problems to perform simultaneous process integration and optimization and increase the degrees of freedom. This approach promises significant improvements in scaling and efficiency compared to existing methods. Nonsmooth operators would introduce two equality constraints per resource regardless of the size of the process without requiring embedded optimization problems or large numbers of constraints and binary variables that increase rapidly with the number of sources and sinks.

Our nonsmooth formulation is the only approach to the resource-targeting problem that can solve for any unknown quantity while scaling compactly, only requires equation-solving methods, and is explicitly generalizable to multiple resources. Therefore, the nonsmooth integration approach is a good candidate for performing integration for large, even interplant, systems and can be easily extended as pinch analysis is applied to new problems. Thus, we have formulated a readily adaptable approach that significantly reduces problem complexity and can provide computationally practical solutions to a wide variety of new integration problems to improve resource use and sustainability in chemical processes.

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The authors declare no competing financial interest.

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