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Global bounds on optimal solutions in chemical process design

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In this paper a new approach for computing global bounds on optimal solutions of mixed-integer nonlinear programs is presented. These type of problems frequently arise in optimal design of chemical processes. The approach is based on a hierarchy of polyhedral relaxations leading to mixed-integer linear programs, which can be solved rigorously. Application is demonstrated for the optimal design of combined reaction distillation processes and for feasibility studies of simulated moving bed chromatographic processes.

1. Introduction

The optimal design of chemical processes using mathematical optimization often leads to mixed-integer nonlinear programs (MINLP). Due to nonconvexity MINLP problems are usually difficult to solve. Typically either gradient based local optimization methods are used for this purpose or stochastic optimization methods like simulated annealing or genetic algorithms (see e.g. [8]). However, in both cases no guarantee can be given that the solution found by the algorithm is the global optimum.

To overcome this problem, a new global approach is proposed in this paper. It is based on techniques to derive polyhedral approximations of the underlying nonlinear equations in such a way that a mixed-integer linear relaxation of the original problem is obtained, which can be solved rigorously. Since the feasible set of the original nonlinear problem lies within the feasible set of the relaxed problem, the latter provides global lower bounds for the optimal solution of the original problem. The lower bound approaches the true global optimum as the number of grid points of the relaxation is increased.

The applicability of the approach is demonstrated for two different challenging fields. The first application is concerned with the optimal design of reaction distillation processes

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involving reactor separator recycle systems, reactive distillation columns and side reactor concepts. The second application is concerned with the feasibility of simulated moving bed chromatographic processes. With the new method one can prove whether it is possible to meet given product specifications with a given process configuration and given column efficiency.

2. Methods

Optimal design of chemical processes often leads to mixed-integer nonlinear programs, which can be written in the following generic form

$$\begin{array}{lrcl} \min & f(x,y) & & \\ \text{s. t.} & h(x,y) & = & 0, \\ & g(x,y) & \leq & 0, \\ & x & \in & X \subseteq \mathbf{R}^n, \\ & y & \in & Y \subseteq \mathbf{Z}^d, \end{array} \qquad \text{(MINLP)}$$

where $f : \mathbf{R}^{n\times d} \to \mathbf{R}$ is a real function, $h : \mathbf{R}^{n\times d} \to \mathbf{R}^q$ and $g : \mathbf{R}^{n\times d} \to \mathbf{R}^p$ are vectors of real functions, $X := \{x \in \mathbf{R}^n \mid Ax \leq a\} \subseteq \mathbf{R}^n$ is a compact polyhedron, and $Y := \{y \in \mathbf{Z}^d \mid By \leq b\} \subseteq \mathbf{Z}^n$ is the set of integer point lying in a compact polytope.

Several approaches for deriving bounds on the model (MINLP) have been suggested in the literature that we briefly survey below. Of major importance are the Generalized Bender Decomposition approach and the outer-approximation technique (see e.g. [3,4] and references therein). The two types of algorithms work in two phases: In the outer phase only the integer variables are manipulated. In each inner iteration a nonlinear subproblem is solved for the continuous variables for fixed values of the integer variables. The inner optimal solution is used to construct a relaxation in form of a second order problem to determine better values for the integer variables. Both approaches differ in the way the second order problem is constructed. Whereas Generalized Bender Decomposition methods use dual information, the outer-approximation approaches construct a mixed-integer linear relaxation for the primal problem. It is worth noting, that only in special cases these approaches yield global results.

An important tool for treating general nonlinear functions occurring in model (MINLP) is to resort to convex underestimators for the corresponding functions. This ensures that relaxations of MINLP can be defined that only involve convex functions. Neglecting the integrality requirements yields a convex relaxation for model (MINLP) that is well tractable. Following this approach, interesting global bounds on the optimal value of synthesis problem can be given if one is able to derive good convex underestimators. For special functions convex underestimators have been given (see e.g. [2] and references therein).

In this paper, we propose an alternative approach, where integrality requirements for the discrete variables y are explicitly taken into account. Instead of using convex underestimators a hierarchy of polyhedral relaxations is constructed. The construction of these relaxations is based on combinatorial substructures arising from the nonlinearities of the specific application. Each member of this hierarchy is a mixed-integer linear program in an extended space of variables and defines a global bound on the optimal value of the overall problem, because it contains all feasible solutions of the original nonlinear model. In the past decades several algorithmic techniques have been proposed to solve such MILPs in practice. Most notably, clever enumeration strategies in combination with advanced preprocessing and cutting plane techniques make instances tractable today that were out of any reach even ten years ago [1].

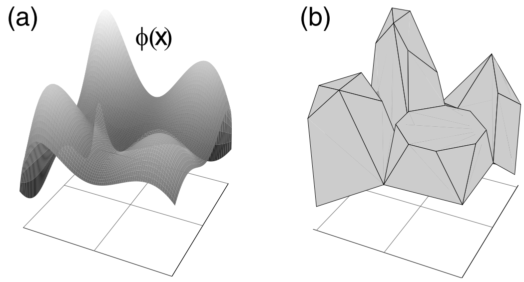

Figure 1. For the nonlinear function $\Phi(x)$ on a box $[l, u]$ in (a), a polyhedral relaxation for the graph of $\Phi(x)$ is given in (b): The domain is subdivided into four subregions. For each subregion a polyhedron is defined enclosing the graph of $\Phi(x)$.

In order to apply this idea, it is crucial to decompose the function f and each component function of h and g, i. e., to express the function as the sum of functions $\phi_1 \ldots, \phi_r$. Next, one introduces a new variable for the value of ϕ_i. More precisely, the following relaxation procedure is applied as illustrated in Fig. 1: To this end, let $(x, y)^\top \in \mathbf{R}^n \times \mathbf{Z}^d$ be a vector of n continuous variables $x_i \in [l_i, u_i]$, $i = 1, \ldots, n$, and d integer variables $y_j \in \{L_j, \ldots, U_j\}$, $j = 1, \ldots, d$, and consider a nonlinear function $\phi : \mathbf{R}^n \times \mathbf{Z}^d \to \mathbf{R}$ restricted to $\mathcal{D} := \prod_{i=1}^n [l_i, u_i] \times \prod_{j=1}^d \{L_j, \ldots, U_j\} \subseteq \mathbf{R}^n \times \mathbf{Z}^d$. The main steps of our relaxation procedure includes

- a subroutine ANALYZE in which the nonlinear function $\phi(x, y)$ is investigated for local and global properties, e. g., local and global extrema, discontinuities, etc., using elementary differential geometry.
- a subroutine BINMOD that returns a hierarchy of subdivisions of the domain $\mathcal{D}$. Each subdivision consists of a set of support vectors. A subdivision that is higher in the hierarchy contains all the support vectors of the lower-level ones.
- a subroutine POLYPROX that defines an *enclosing* (not interpolating) polyhedron for the graph of $\phi(x, y)$ on every subregion $\mathcal{D}^\nu$ based on the results of the subroutine ANALYZE.
- a subroutine COMBINE that, based on *combinatorial substructures* given by linear (and nonlinear) relations between the variables occurring in $\phi(x, y)$, determines *valid inequalities* involving the corresponding binary variables.

The hierarchy of subdivisions provided by subroutine BINMOD gives rise to a hierarchy of nonlinear mixed-integer optimization problems. Each such instance is an extended formulation of (MINLP) by introducing additional variables. It attains the following form

$$
\begin{array}{lrcll}
\min & \sum_{k=1}^{r} f_k \phi_k(x,y) & & & \\
\text{s. t.} & \sum_{k=1}^{r} h_{j,k}\phi_k(x,y) & = & 0, & \text{for all } j=1,\dots,q, \\
& \sum_{k=1}^{r} g_{j,k}\phi_k(x,y) & \leq & 0, & \text{for all } j=1,\dots,p \\
& \sum_{\iota=1}^{\nu_i} s_\iota^i \beta_\iota^i \quad \leq x_i & \leq & \sum_{\iota=1}^{\nu_i} s_{\iota+1}^i \beta_\iota^i, & \text{for all } i=1,\dots,n, \\
& \sum_{\iota=1}^{\nu_i} \beta_\iota^i & = & 1, & \text{for all } i=1,\dots,n, \\
& x & \in & [l,u] \subseteq \mathbf{R}^n, & \\
& y & \in & [L,U] \cap \mathbf{Z}^d, & \\
& \beta = (\beta^1,\dots,\beta^n)^\top & \in & \prod_{i=1}^{n} \{0,1\}^{\nu_i} &
\end{array}
\qquad \text{(MDEF)}
$$

Therein, the additional binary variables β_ι^i indicate subintervals $[s_\iota^i, s_{\iota+1}^i] \subseteq [l_i, u_i]$ for the original domain of x_i. Such a division into subintervals can be represented as a list of supporting points, $S^i := \{s_1^i, \dots, s_{\nu_i+1}^i\} \subseteq [l_i, u_i]$, with $s_1^i = l_i$, $s_{\nu_i+1}^i = u_i$, and $s_\iota^i < s_{\iota+1}^i$, for all $\iota \in \{1,\dots,\nu_i\}$. In this manner, the original domain $[l,u]$ is divided into $\prod_{i=1}^{n} \nu_i$ subboxes. Analytic and geometric properties of the underlying nonlinear functions ϕ_k for specific examples give rise to specific lists of supporting points $S^i, i=1,\dots,n$.

In the integer programming community problems of the form (MDEF) involving linear functions only are known as fixed-charge mixed-integer optimization problems that include a large variety of applications (see [7]). In the remainder, focus will be on applications arising in the design of chemical processes. In order to tackle these problems we make use of a variety of new combinatorial relaxations. These arise from nonlinear flow conservations, mixed-integer Knapsack constraints and stable sets in special conflict graphs.

3. Applications

3.1. Optimal design of combined reaction separation processes

The first application is concerned with the optimal design of combined reaction distillation processes, which play an important role in chemical industry. Depending on the physical properties of the system, the given constraints on production rate, product specifications, and the given costs different process candidates can be attractive including reactive distillation columns, or nonreactive distillation columns with side- and/or pre-reactors as illustrated in Fig. 2.

In principle, the best process configuration can be found by optimizing some suitable superstructures, which include the relevant process alternatives. The objective are minimal total costs comprising investment and operating costs. This will lead to a complex problem of type (MINLP), which could be solved rigorously with a sequence of linear relaxations of type (MDEF) with successive refinement until the global optimum is reached. This however, can be computationally very expensive. Therefore a combined strategy is proposed, where a quick preliminary ranking of relevant process candidates is obtained with available local MINLP optimization methods. Afterwards the results can be checked

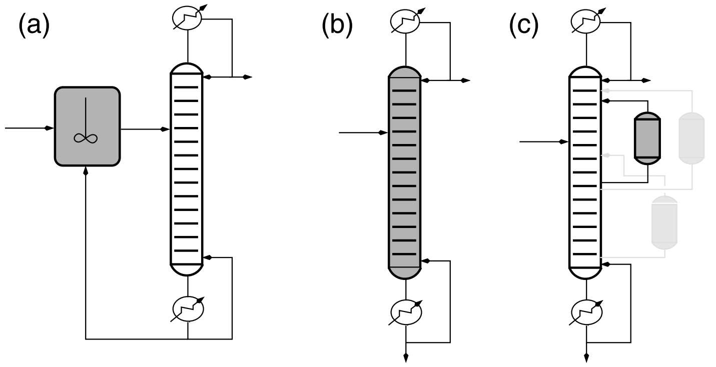

Figure 2. Characteristic process alternatives for combined reaction distillation processes: (a) reactor with nonreactive distillation column and recycle, (b) reactive distillation column, (c) nonreactive distillation column with side reactors.

with global lower bounds obtained for each subproblem through polyhedral relaxation. The ranking of any two process candidates is either proven if the lower bound on the cost function of the second best candidate is greater than the known local solution of the best candidate. If that can not be achieved with successive refinement of the polyhedral relaxation, we may use the polyhedral relaxation to generate new starting values for the local optimization to find better global minima. Application of the procedure has been demonstrated for production of 2,3-dimethylbutene-1 by isomerization of 2,3-dimethylbutene-2. A detailed description of the mathematical formulation is given elsewhere [5].

3.2. Feasibility of simulated moving bed chromatographic processes

The second application is concerned with the feasibility of simulated moving bed (SMB) chromatographic processes. SMB is an advanced technology to separate isomers and in particular enantiomers. For high purity separations with highly efficient columns triangle theory developed by Storti et al. [9] is used for process design. Triangle theory is based on a true moving bed model and assumes thermodynamic equilibrium between the solid and the fluid phase corresponding to an infinite column efficiency. The theory allows to determine suitable values for the flow rate ratios $m^I, \ldots m^{IV}$ in the four zones of the process, which allow for complete separation of a given mixture with components A and B as illustrated in Fig. 3.

Maximum productivity, i.e. maximum feed rate is obtained for values of m^{II} and m^{III} at the vertex of the triangle (point W in Fig. 3. The triangle represents the feasible region for complete separation. It should be noted that for infinitely efficient columns total separation is always possible for mixtures with different adsorptivities, which can be described with linear or Langmuir isotherms. Or, in other words, in this case the feasible region is always non empty, and, in particular a maximum value for the feed rate different from zero can be found.

However, in practice often lower purities are acceptable and cheaper columns with reduced efficiency can be applied. In this case, the question arises whether a given product

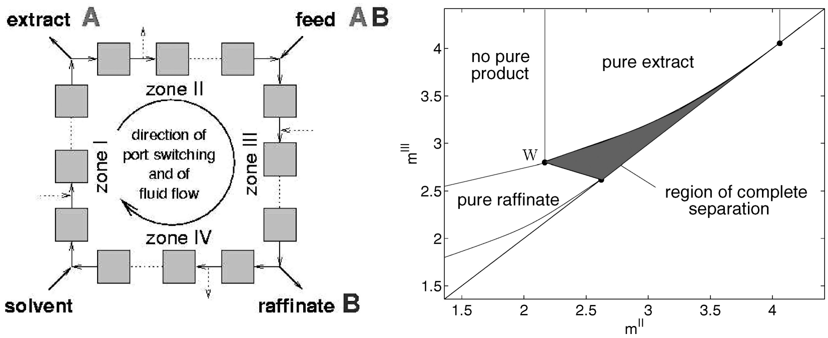

Figure 3. Characteristic process alternatives for combined reaction distillation processes: (a) reactor with nonreactive distillation column and recycle, (b) reactive distillation column, (c) nonreactive distillation column with side reactors.

purity can be achieved with a given column efficiency corresponding to a given number of theoretical stages in our model formulation. To answer this questions polyhedral relaxations are applied to an optimization problem, where we maximize the feed rate for given purity constraints. By means of polyhedral relaxations on upper bound for the optimal feed rate is obtained. If this upper bound is zero, than it is proven that it is not possible to achieve the required purity for any values of $m^I, \dots m^{IV}$.

Application has been demonstrated for a process separating a mixture of fructose-dextran T9 and fructose-raffinose. A detailed description of the mathematical formulation is given elsewhere [6].

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