Global Optimization in the 21st Century: Advances and Challenges

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Abstract
This paper presents an overview of the research progress in global optimization during the last five years (1998-2003), and a brief account of our recent research contributions. The review part covers the areas of (a) twice continuously differentiable nonlinear optimization, (b) mixed-integer nonlinear optimization, (c) optimization with differential-algebraic models, (d) optimization with grey box/black box/nonfactorable models, and (e) bilevel nonlinear optimization. Our research contributions part focuses on (i) improved convex underestimation approaches that include convex envelope results for multilinear functions, convex relaxation results for trigonometric functions, and a piecewise quadratic convex underestimator for twice continuously differentiable functions, and (ii) the recently proposed novel generalized αBB framework. Computational studies will illustrate the potential of these advances.

Keywords
Global Optimization; Nonlinear Optimization; Mixed-Integer Nonlinear Optimization; Differential-Algebraic Optimization; Optimization with Nonfactorable/Grey box models; Bilevel Nonlinear Optimization; Nonconvexities; Convex Envelopes; Convex Underestimators; Trilinear Monomials; Trigonometric Functions; Twice Continuously Differentiable Functions.

1 Introduction

It is now established that Global Optimization has ubiquitous applications not only in Chemical Engineering but also across all branches of engineering, applied sciences, and sciences (e.g., see the textbook by Floudas 2000). As a result, we have experienced significant interest in new theoretical advances, algorithmic and implementation related investigations, and their application to important scientific problems. A review paper discussed the advances in deterministic global optimization and their applications in the design and control of chemical process systems (Floudas (2000a)). A second review paper presented at the FOCAPD-1999 meeting outlined the Chemical Engineering research contributions in global optimization for the period 1994-1999, presented the advances, and identified research opportunities and challenges (Floudas and Pardalos, 1999). During the last five years, 1998-2003, several

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outstanding textbooks have been published addressing different facets of global optimization. These include the textbooks by Tuy (1998), Bard (1998), Sherali and Adams (1999), Floudas (2000), Horst *et al.* (2000), Tawarmalani and Sahinidis (2002), and Zabinsky (2003). A handbook of test problems in local and global optimization (Floudas *et al.* (1999)), as well as two edited volumes of the research contributions presented at the major conferences on Global Optimization held in 1999 and 2003 (Floudas and Pardalos (2000), Floudas and Pardalos (2003)), were published.

Global optimization addresses the computation and characterization of global optima (i.e., minima and maxima) of nonconvex functions constrained in a specified domain. Given an objective function f that is to be minimized and a set of equality and inequality constraints S, *Deterministic Global Optimization* focuses on the following important issues :

(a) Determine a global minimum of the objective function f (i.e., f has the lowest possible value in S) subject to the set of constraints S;

(b) Determine *lower* and *upper bounds* on the global minimum of the objective function f on S that are valid for the whole feasible region S;

(c) Determine an ensemble of qood quality local solutions in the vicinity of the global solution;

(d) Enclose all solutions of the set of equality and inequality constraints S.

In this review paper, we will discuss the deterministic global optimization advances during the last five years for the following classes of mathematical problems: (i) twice continuously differentiable nonlinear optimization, NLPs; (ii) mixed-integer nonlinear optimization, MINLPs; (iii) differential-algebraic systems, DAEs; (iv) grey-box and nonfactorable problems; and (v) bilevel nonlinear and mixed integer optimization. We will first present all the contributions in the aforementioned classes, and we will subsequently focus on a few advances from Princeton University on (a) convex envelope results for trilinear monomials, (b) convex relaxation results for trigonometric functions, (c) new convex underestimators based on piecewise convex quadratic representations, and (d) the generalized αBB global optimization approach.

2 Twice Continuously Differentiable NLPs

In the first part of this section, we will review the advances in convex envelopes and convexification techniques. We will subsequently focus on theoretical and algorithmic advances for (a) general C^2 NLPs; (b) concave, bilinear, fractional, and multiplicative problems; (c) phase equilibrium problems; and (d) parameter estimation problems.

Convexification Techniques and Convex Envelopes Adjiman *et al.* (1998a), Hertz *et al.* (1999) proposed several new rigorous methods for the calculation of the α parameters for (i) uniform diagonal shift of the hessian matrix and (ii) non-uniform diagonal shift of the hessian matrix, and they established their potential trade-offs. Adjiman *et al.* (1998b) presented the detailed implementation of the αBB approach and computational studies in process design problems such as heat exchanger networks, reactor-separator networks, and batch design under uncertainty.

Tawarmalani and Sahinidis (2001) developed the convex envelope and concave envelope for x/y over a unit hypercube, compared it to the convex relaxation proposed by Zamora and

Grosmmann (1998a),(1998b), (1999), proposed a semidefinite relaxation of x/y, and suggested convex envelopes for functions of the form $f(x)y^2$ and $f(x)/y$. Ryoo and Sahinidis (2001) studied the bounds for multilinear functions via arithmetic intervals, recursive arithmetic intervals, logarithmic transformation, and exponential transformation, and provided comparisons of the resulting convex relaxations. Tawarmalani *et al.* (2002a) showed that tigher linear programming relaxations are produced if the product of a continuous variable and the sum of several continuous variables is disaggregated, and applied it to the instance of rational programs that include a nuclear reactor reload pattern design, and a catalyst mixing in a packed bed reactor problem. Tawarmalani and Sahinidis (2002) introduced the convex extensions for lower semi-continuous functions, studied conditions under which they exist, proposed a technique for constructing convex envelopes for nonlinear functions, and studied the maximum separation distance for functions such as x/y. Tawarmalani *et al.* (2002b) studied $0-1$ hyperbolic programs, developed eight mixed-integer convex reformulations, proposed analytical results on the tightness of these reformulations, developed a global optimization algorithm and applied it to a p-choice facility location problem.

Liberti and Pantelides (2003) proposed a nonlinear continuous and differentiable convex envelope for monomials of odd degree, derived its linear relaxation, and compared to other relaxation. Björk *et al.* (2003) studied convexifications for signomial terms, introduced properties of power convex functions, compared the effect of the convexification schemes for heat exchanger network problems, and studied quasi-convex convexifications.

Meyer and Floudas (2003) studied trilinear monomials with positive or negative domains, derived explicit expressions for the facets of the convex and concave envelopes and showed that these outperform the previously proposed relaxations based on arithmetic intervals or recursive arithmetic intervals. Meyer and Floudas (2004a) presented explicit expressions for the facets of convex and concave envelopes of trilinear monomials with mixed-sign domains. Tardella (2003) studied the class of functions whose convex envelope on a polyhedron coincides with the convex envelope based on the polyhedron vertices, and proved important conditions for a vertex polyhedral convex envelope.

Caratzoulas and Floudas (2004) proposed novel convex underestimators for trigonometric functions which are trigonometric functions themselves. Akrotirianakis and Floudas (2004a) introduced a new class of convex underestimators for twice continuously differentiable NLPs, studied their theoretical properties, and proved that the resulting convex relaxation is improved compared to the αBB one. Meyer and Floudas (2004a) proposed two new classes of convex underestimators for general C^2 NLPs which combine the αBB underestimators within a piecewise quadratic perturbation, derived properties for the smoothness of the convex underestimators, and showed the improvements over the classical αBB convex underestimators for box-constrained optimization problems.

General C^2 NLPs Adjiman *et al.* (1998a), Adjiman *et al.* (1998b) introduced the αBB global optimization approach, which is applicable to general twice-continuously differentiable NLPs, and presented extensive computational studies in process design problems such as heat exchanger networks, reactor-separator networks, and batch design under uncertainty. Yamada and Hara (1998) proposed a global optimization approach based on the triangle covering for H-infinity control with constant diagonal scaling. Androulakis and Floudas (1998) studied the parallel computation issues that arise using the αBB global optimization approach.

Klepeis *et al.* (1998), and Klepeis and Floudas (1999a) proposed new global optimization approaches for the structure prediction of solvated peptides using area and volume accessible to the solvent models. A review of the global optimization activities in the areas of protein folding and peptide docking can be found in Floudas *et al.* (1999b). Klepeis and Floudas (1999b) proposed a novel deterministic global optimization approach for free energy calculations of peptides. Westerberg and Floudas (1999a) and Westerberg and Floudas (1999b) introduced a global optimization framework for the enclosure of all transition states of potential energy hypersurfaces, and studied the reaction pathways and dynamics helical formation with and without solvation. Klepeis *et al.* (1999) introduced a novel approach that combines deterministic global optimization and torsional angle dynamics for the prediction of peptide structures using a sparse set of NMR data.

Byrne and Bogle (1999) introduced a bound constrained linear relaxation, developed two classes of linear underestimators using the natural extension and mean value theorems of interval analysis, and showed that the interval LP is more efficient than other interval analysis approaches. Gau and Stadtherr (2002) studied the computational improvement of interval Newton/generalized bisection approaches, introduced a hybrid preconditioning strategy where a pivoting preconditioner is combined with the standard inverse midpoint method, and showed that this approach results in a large reduction of the needed subintervals and hence in significant computational improvements. Gau and Stadtherr (2002) studied synchronous work stealing, synchronous and asynchronous diffusive load balancing on a two-dimensional torus virtual network, developed a distributed computing interval Newton framework, and showed that superlinear speedups can be obtained for vapor-liquid equilibrium and parameter estimation problems. Lucia and Feng (2002) studied the least squares function landscape, introduced a differential geometry based framework for the determination of all physically meaningful solutions, singular points, and their connectivity, developed a global terrain algorithm,and illustrated the framework through one and two-dimensional examples from glass temperature calculations, equilibrium states in nanostructured materials, a simplied SAFT equation, and a CSTR equation. Klepeis *et al.* (2002) presented the advances in deterministic global optimization based on the αBB approach and its applications for structure prediction of oligopeptides, dynamics of helical formation, and protein-peptide interactions.

Zilinskas and Bogle (2003) studied the evaluation of ranges of functions through balanced random interval arithmetic, investigated the hypothesis on the normal distribution of the centers and radii of the evaluated balanced random intervals through several computational studies, and concluded that this hypothesis is incorrect. Klepeis and Floudas (2003b) introduced a deterministic global optimization approach, αBB, coupled with torsional angle dynamics for the protein structure prediction given restraints predicted from the identification of helices and β-sheets. Klepeis and Floudas (2003c) proposed the first principles framework, Astro-Fold, for the protein structure prediction, described the global optimization and mixed-integer optimization advances, and presented a variety of test systems including several blind protein predictions. Klepeis *et al.* (2003a) introduced a new class of hybrid global optimization methods denoted as integrated hybrids for the oligopeptide structure prediction. Klepeis *et al.* (2003b) proposed new alternating hybrid global optimization methods, studied and developed their distributed computing algorithms, and applied them to the structure prediction of met-enkaphalin and mellitin. These two classes of hybrid global optimization approaches combine the αBB for the generation of rigorous lower bounds with the modified genetic algorithm, CSA, for the upper bounding calculations. Lucia and Feng (2003) extended the terrain methodology to multivariable problems and integral curve bifurcations associated with valleys and ridges, showed that the terrain mathods are superior to arc homotopy continuation in the presence of parametric disconnectedness, and studied examples for the location of all azeotropes, retrograde flash calculations, and

CSTR problems.

Schafroth and Floudas (2004) studied the protein-peptide interactions via deterministic global optimization, atomistic-level modeling, and several solvation methods that include the area accessible to the solvent, the volume accessible to the solvent, and the Poisson-Boltzmann method, and reported excellent agreement on the binding motifs.

Akrotirianakis and Floudas (2004b) presented computational results of the new class of convex underestimators embedded in a branch-and-bound framework for box-constrained NLPs. They also proposed a hybrid global optimization method that includes the random-linkage stochastic approach with the aim at improving the computational performance.

Concave, Bilinear, Fractional and Multiplicative Models Zamora and Grosmmann (1998b) introduced a deterministic branch-and-bound approach for structured process systems that have univariate concave, bilinear and linear fractional terms. They proposed several properties of the contraction operation, embedded them in the global optimization algorithm and studied the contraction effects on several applications. Shectman and Sahinidis (1998) proposed a finite global optimization method for separable concave problems. Zamora and Grossmann (1999) proposed a branch-and-contract global optimization algorithm for univariate concave, bilinear, and linear fractional models. The emphasis was on reducing the number of nodes in the branch-and-bound tree through proper use of the contraction operator. Van Antwerp *et al.* (1999) studied the bilinear matrix inequality problem as a formulation of the globally optimal controller problem and applied a branch-and-bound global optimization approach to generate lower and upper bounds and prove optimality for a mass spring model and a reactive ion etching problem.

Adhya *et al.* (1999) studied bilinear models of the pooling problem, proposed a lagrangian relaxation approach for the generation of valid lower bounds, and showed that these bounds are tigher when compared to linear programming based relaxations. Ryoo and Sahinidis (2003) studied linear and generalized linear multiplicative models, applied the recursive arithmetic interval approach for the derivation of lower bounds, introduced greedy heuristics for a branch-and-reduce approach, and applied it to benchmark problems and randomly generated problems. Goyal and Ierapetritou (2003a) introduced an approach for the systematic evaluation of the infeasible domains using a simplicial outer approximation framework that is applicable to concave or quasiconvex constraints.

Phase Equilibriium Maier *et al.* (1998) applied an interval analysis based approach for the enclosure of homogeneous azeotropes. They employed the formulations proposed by Harding *et al.* (1997) and studied systems with activity coefficient and equation of state models. Meyer and Swartz (1998) proposed a new approach for testing convexity for phase equilibrium problems. McKinnon and Mongeau (1998) proposed a generic global optimization approach for the phase and chemical reaction equilibrium problem that is based on interval analysis and combines the stability criterion with the minimization of the Gibbs free energy. Hua *et al.* (1998a) applied an interval analysis method for the phase stability computations of binary and ternary mixtures using equation of state models. Hua *et al.* (1998b) introduced two enhancements on their interval analysis approach based on monotonicity and mole fraction weighted averages for improving the efficiency in the tangent plane stability analysis for cubic equations of state. Zhu and Xu (1999a) used simulated annealing for the tangent plane stability analysis criterion and they applied it to ternary systems. Zhu and Xu (1999b) studied the tangent plane stability analysis for the SRK cubic equation of state through a Lipschitz global optimization approach, and applied it to binary systems. Zhu and Xu (1999) used simulated annealing for the stability analysis of liquid-liquid equilibrium systems modeled via the NRTL and UNIQUAC equations for the activity coefficients and studied ternary systems with up to three liquid phases.

Harding and Floudas (2000a) introduced a novel global optimization approach for the phase stability of several cubic equations of state based on analytical findings and the principles of the αBB global optimization framework. Harding and Floudas (2000b) studied the enclosure of all heterogeneous and reactive azeotropes, developed a rigorous framework based on the αBB global optimization principles, and demonstrated its potential for a variety of case studies. Tessier *et al.* (2000) introduced monotonicity based and mole fraction weighted averages based enhancments for the application of interval Newton methods to the phase stability problem using the NRTL and UNIQUAC models. Zhu *et al.* (2000) proposed an enhanced simulated annealing algorithm for the tangent plane stability problem using the PR and SRK cubic equations of state.

Zhu and Inoue (2001) introduced a branch-and-bound approach based on a quadratic underestimating function and applied it to the tangent plane distance criterion using the NRTL equation. Xu *et al.* (2002) studied the phase stability criterion using the SAFT equation of state, introduced an interval Newton/generalized bisection approach, followed a volume-based formulation based on the Helmholtz energy, and applied to nonassociating, self-associating, and cross-associating systems. Cheung *et al.* (2002) studied the global minimum determination of clusters for the solvent-solute interactions in phase equilibrium. They introduced the OPLS force field, derived tight convex underestimators, derived bounds on the dependent variables, developed a branch-and-bound approach, and applied it to a butane molecule and a butane-ethylamine system.

Parameter Estimation Esposito and Floudas (1998) studied the error-in-variables approach and proposed the first global optimization method for the parameter estimation and data reconcilliation of nonlinear algebraic models using the principles of the αBB approach. Gau and Stadtherr (2000) introduced an interval analysis based approach for the error-in-variables method and studied vapor liquid equilibrium and reaction kinetics models. Gau *et al.* (2000) studied further the parameter estimation of vapor liquid equilibrium models via interval analysis, applied it using the Wilson equation for a variety of binary systems, and demonstrated that correct predictions of azeotropes are attained only based on the global optimum parameter solutions in direct contrast to the Dechema data collection. Gau and Stadtherr (2002) applied the interval-newton approach for the parameter estimation of a catalytic reactor model, a heat exchanger network model, and binary vapor-liquid equilibrium problems using the Wilson equation, and pointed out that problems of about two hundred variables can be addressed.

3 Mixed-Integer Nonlinear Optimization, MINLPs

Zamora and Grosmmann (1998a) derived thermodynamic-based convex underestimators, quadratic/linear fractional convex underestomators, and proposed a hybrid branch-and-bound and outer approximation method for the global optimization of heat exchanger networks with no stream splits. Westerlund *et al.* (1998) proposed an extended cutting plane approach for the global optimization of pseudoconvex MINLP problems, studied its convergence properties, and applied it to an example from the paper-converting industry. Vecchietti and Grossmann (1999) introduced a disjunctive programming approach for MINLPs, denoted as LOGMIP, discussed a hybrid modeling framework for process systems engineering which allows both binary variables and disjunctions as tools for discrete decisions, implemented a modified logic-based outer approximation approach, and presented computational studies on two process synthesis problems and an FTIR spectroscopy example. Sinha *et al.* (1999) studied the class of solvent design problems, modelled it as a nonconvex MINLP problem, identified the sources of nonconvexities in the properties and solubility parameter design constraints, proposed linear underestimators based on a multilevel representation approach for the functions, developed a reduced space branch-and-bound global optimization algorithm, and applied it to a single component blanket wash design problem. Noureldin and El-Halwagi (1999) studied mass integration problems for pollution prevention, proposed targets for the maximum achievable poluution, introduced an interval analysis framework for the determination of these targets, studied the pollution prevention via unit manipulation, recycle and interception, and employed the interval-based targets in a case study featuring the reduction of water usage and discharge in a tire-to-fuel plant. Pörn *et al.* (1999) proposed convexification schemes for classes of discrete and integer nonconvex models. They studied the exponential tranformation and potential-based tranformations and applied them to integer posynomial problems. Harjunkoski *et al.* (1999) studied the trim loss minimization problem for the paper converting industry, formulated it as a nonconvex MINLP, proposed tranformations for the bilinear terms that are based on linear representations and convex expressions, studied the reductions of the combinatorial space, investigated the role of different types of objective functions, developed and assessed several algorithmic alternatives, and showed that the global solution can be obtained with all strategies and certain convex formulations performed similarly to the linear models.

Adjiman *et al.* (2000) proposed two novel global optimization approaches for nonconvex mixed-integer nonlinear programming problems. The first approach, $SMIN-\alpha$BB is for separable continuous and integer domains and it is based on the principles of αBB type of convex underestimators and a branch-and-bound approach for the mixed set of continuous and binary variables. The second approach, $GMIN-\alpha$BB, is applicable to general mixed integer nonlinear problems which are not separable in the continuous and integer variables, and it is based on a branch-and-bound tree constructed only in the integer domain while the αBB principles are used to solve the nonconvex NLPs at each node so as to generate valid lower bounds. The first approach was applied to heat exchanger network problems, while the second one was applied to pump network configuration problems and trim loss minimization problems in addition to a variety of benchmark problems. Kesavan and Barton (2000) introduced a generalized branch-and-cut algorithm for nonconvex MINLPs, showed that decomposition-based approaches and branch-and-bound algorithms are special cases, and proposed a number of heuristics towards addressing the computational efficiency issues. Sahinidis and Tawarmalani (2000) presented two MINLP applications of global optimization for the design of just-in-time flowshops, and the design of an alterative to freon. In the first study, the model determines the stagewise number of machines needed that minimizes the total equipment costs, and they showed improvements compared to the heuristic approaches. In the second study, the model selects the constituent parts of a molecule so as to satisfy chemical and physical properties, economic, environmental constraints through a group contribution based approach, and provides a ranked order list of alternative compounds. Parthasarathy and El-Halwagi (2000) studied a systematic framework for the optimal design of condensation which an important technology for volatile organic compounds, formulated it as a nonconvex MINLP model, proposed an iterative global optimization approach which is based on physical insights and active constraint principles that allow for decomposition and efficient solution, and applied it to a case study for the manufacture of adhesive tapes.

Pörn and Westerlund (2000) introduced procedures for the successive linear approximation of the objective function and line search techniques, proposed a cutting plane method for addressing global MINLP problems that feature pseudo-convex objective function and constraints, studied its convergence properties and initialization schemes, and tested it on several benchmark problems arising in process synthesis and scheduling applications.

Lee and Grossmann (2001) studied nonconvex generalized disjunctive programming models, constructed the convex hull of each nonlinear disjunction, used convex underestimators for bilinear, linear fractional and concave separable functions, introduced a two level branch-and-bound algorithm where the lower bound requires a discrete search in the disjunctions space and the upper bound requires a spatial divide and conquer search in the nonconvex continuous space, and applied it to benchmark problems, a multicomponent separation problem, multistage design/synthesis of batch plants with parallel units, and heat exchanger network synthesis. Björk and Westerlund (2002) studied the global optimization of heat exchanger network synthesis through the simplified superstructure representation that allows only series and parallel schemes, applied convexification approaches for signomials via piecewise linear approximations, developed convex MINLP lower bounding models using the Patterson formula for the log mean temperature difference considering both isothermal and nonisothermal mixing, proposed a global optimization approach for alternative models, and presented extensive computational studies. Wang and Achenie (2002) studied solvent design problems which are formulated as nonconvex MINLPs, introduced a hybrid global optimization approach which combines outer approximation with simulated annealing, applied it to several benchmark problems, case studies for the extraction of acetic acid from water, and solvent design for reversible reactions, and showed that near optimal solutions can be located. Ostrovsky *et al.* (2002) studied nonconvex MINLP models in which most variables are in the conconvex terms and the number of linear constraints is much larger than the nonlinear constraints, introduced the idea of branching on a set of linear branching variables which depend linearly on the serach variables, proposed a tailored barnch and bound approach using linear understimators for tree functions based on a multilevel function representation, showed that there is a significant reduction in the branching variable space, and applied it to solvent design and recovery problems. Wang and Achenie (2002) studied the molecular design of solvents for extractive fermentation including solvent attributes such as biocompatibility, inertness and phase splitting, introduced a group contribution framework which results in a conconvex MINLP model, studied a local MINLP algorithm, OA/ER/AP, and applied it to case studies on ethanol extractive fermentation. Dua *et al.* (2002) proposed novel approaches for multiparametric mixed-integer quadratic models through the decomposition into a multiparametric quadratic MIQP model for the upper bound and a potentially nonconvex MINLP model for the lower bound, suggested ways of addressing the nonconvexity in the MINLP, and generated envelopes of parametric solutions and the enclosure of the multiparametric MIQP.

Sahinidis *et al.* (2003) revisited the design of alternative refrigerants problem, introduced an integer formulation for previously described structural constraints, proposed new structural constraints between one-bonded and higher-bonded groups in the absence of rings and new clique constraints for rings, applied a branch-and-reduce global optimization algorithm with a modification so as to generate all feasible integer solutions, and generated new compounds for refrigerants. Vaia and Sahinidis (2003) studied the simultaneous parameter estimation and model structure identification in infrared spectroscopy, proposed two methods out of which the second corresponds to a single nonconvex MINLP model, presented a branch-and-bound approach which is based on a relaxation of terms that are logarithmic, bilinear, and multilinear depending on the determinant of the covariance matrix, and presented comparative computational results. Ostrovsky *et al.* (2003) revisited their molecular design reduced dimension branch-and-bound algorithm by studying further the branching functions concept and the special tree function representation, proposed the sweep mathod for the construction of the linear underestimators, investigated the problem size dependency on the algorithmic performance, and showed that the computational effort increases almost linearly. Sinha *et al.* (2003) studied the systematic design of cleaning solvent blends for lithographic printing, modelled it as a nonconvex MINLP problem, introduced an interval analysis based global optimization approach with modifications on the upper bounding calculation and the local feasibility test which are solved via SQP, and an interval-based domain reduction algorithm, and presented computational results for the design of aqueous blanket wash blends. Zhu and Kuno (2003) proposed a hybrid global optimization method for nonconvex MINLPs which combines convex quadratic underestimation techniques with a revised form of the generalized benders decomposition, suggested its convergence properties, and illustrated it via a two variable problem. Goyal and Ierapetritou (2003b) studied MINLP models where the objective function is convex, and the constraints are convex, concave or quasi-concave, introduced the simplicial approximation of the convex hull of the feasible region, proposed algorithmic procedures and illustrated them via small benchmark problems. Kallrath (2003) studied a nonconvex product portfolio problem, modelled it as a nonconvex MINLP problem for the optimization of the number and size of batch process units, analyzed the sources of nonconvexity consisting of concave functions and trilinear products, investigated the piecewise linear approximation of the objective function, the use of a local MINLP solver, SBB, and a global optimization solver, Baron, and reported that for the large instances weak lower bounds are generated. Grossmann and Lee (2003) studied generalized disjunctive programming, GDP, problems which feature convex nonlinear inequalities in the disjunctions, proposed a convex nonlinear relaxation of the nonlinear GDP problem based on the convex hull representation of each of the disjunctions which was derived by variable disaggregation and reformulation, formulated the nonlinear GDP as a MINLP which was shown to produce improve bounds compared to big-M models, and presented comparative computational studies of the two formulations. Lee and Grossmann (2003) studied nonconvex GDP problems with bilinear equality constraints, derived convex underestimators and overestimators for the bilinear constraints using the reformulation/linearization approach, expressed the discrete choices as disjunctions which were subsequently relaxed by their convex hull representations, used their earlier two level global optimization approach (Lee and Grossmann (2001)), and presented computational studies for pooling problems, water usage problems, and wastewater network problems.

Lin *et al.* (2004) revisited the nonconvex product portfolio problem introduced by Kallrath (2003), presented an improved formulation consisting of a concave objective function with linear constraints in the continuous and binary variables, proposed several techniques for tightening the model and accelerating its solution, developed a customized branch-and-bound approach which addresses the problem to global optimality, applied it to small and large instances, and demonstrated that global solutions can be obtained very efficiently in contrast to commercial MINLP solvers. Kesavan *et al.* (2004) studied separable MINLP models with nonconvex functions, proposed two decomposition algorithms based on alternating sequences of relaxed master problems, two nonlinear programming problems, and outer approximation, showed that the first algorithm yields the global solution while the second provides a rigorous bound on the global solution, and presented computational results on several benchmark problems and heat excahnger network problems.

4 Differential-Algebraic Models, DAEs

Esposito and Floudas (2000a) studied the global optimization in parameter estimation of systems described by differential-algebraic models, proposed a rigorous global optimization approach based on a collocation framework and the αBB principles, proposed a global optimization approach based on an integration framework, and investigated a variety of benchmark problems and complex kinetic mechanisms. Esposito and Floudas (2000b) studied the deterministic global optimization of nonlinear optimal control problems, introduced the integration-based framework, investigated the properties of the input-output map of solutions, suggested three alternative ways of calculating the β values for the lower bounding problems, and demonstrated through several challenging case studies the algorithmic trade-offs of the different strategies, as well as the determination of the global solution. Barton *et al.* (2000) studied the optimization of hybrid discrete/continuous dynamic systems, presented a framework based on hybrid optimal control, investigated existence and sensitivity results, introduced a modified stochastic search approach, and presented computational results for a tank changeover problem. Esposito and Floudas (2001) pointed out the theoretical rigor and advantages of the proposed global optimization methods by Esposito and Floudas (2000a) and the differences between local search approaches and global optimization methods.

Esposito and Floudas (2002) studied the isothermal reactor network synthesis problem, formulated it as nonconvex NLP with differential-algebraic constraints, introduced a global optimization framework based on the integration approach and the αBB, investigated alternative types of reformulations, and reported extensive computational studies for complex reaction/reactor networks. Banga *et al.* (2002) studied the optimal experimental design for the parameter estimation of nonlinear dynamic systems, formulated it as an optimal control that optimizes the Fischer information matrix, introduced two stochastic global optimization approaches to address the nonsmoothness and the multiplicity of solutions, and applied it to the parameter estimation of a fed-batch bioreactor. Papamichail and Adjiman (2002) introduced a deterministic spatial branch-and-bound global optimization approach for nonconvex models with ordinary differential equations, proposed a convex relaxation based on the theory of differential inequalities which allowed them to generate rigorous bounds for the parametric ODEs and their sensitivities, and applied their framework to small optimal control problems and reaction kinetics parameter estimation models.

Adjiman and Papamichail (2003) developed further their branch-and-bound approach, proposed three convex relaxations for the parameter estimation of the initial value problem, and presented computational results on several parameter estimation problems in kinetics. Singer and Barton (2003), Singer and Barton (2004) studied the global optimization of integral objective functions subject to ordinary differential equations, derived convex relaxations for the integral based on a pointwise integrand scheme, developed a branch-and-bound global optimization approach on a Euclidean space which combines the integrand convex relaxations with differential inequalities, McCormick's composition approach, and outer approximation, and illustrated their approach with several small benchmark problems. Lee and Barton (2003) studied the global optimization of linear time varying hybrid systems which exhibit both discrete state and continuous state behavior, and extended their recently developed approach for the determination of the optimal mode sequence when the transition times are fixed (Barton and Lee (2003)), proposed a reformulation of the problem via binary variables while maintaining the linearity of the dymanical system, derived convex relaxations of Bolza-type functions using recent results for linear time varying continuous systems (Lee *et al.* (2004)), and applied it to benchmark problems and an isothermal plug flow reactor problem. Chachuat and Latifi (2003) introduced a spatial branch-and-bound global optimization approach for problems with ordinary differential equations in the constraints, presented results on the first and second order derivatives for the initial value problem and the two point boundary value problem, compared the sensitivity and the adjoint approaches, developed convex underestimators using the αBB principles, and presented computational studies and comparisons of the sensitivity versus the adjoint approach for several problems. Banga *et al.* (2003) studied integrated process design and operation, parameter estimation in bioprocess models, and focused on stochastic global optimization methods for dynamic systems, addressed handling of constraints in stochastic methods, presented hybrid approaches for dynamic optimization, and presented computational studies on the optimal control of bioreactors, the integrated design of a waste treatment plant (see also Moles *et al.* (2003)) where they provided comparisons for several algorithmic approaches, and discussed advances in the parameter estimation of bioprocesses. Banga *et al.* (2003) reviewed and introduced optimization as a key technology for food processing and discussed stochastic global optimization methods and their potential applicability in food process engineering.

5 Grey-Box and Nonfactorable Models

Byrne and Bogle (2000) studied the global optimization of modular flowsheeting systems, introduced an approach to modular based process simulation which is based on interval analysis and which can generate interval bounds, derivatives and their bounds for generic input-output modules, proposed a branch-and-bound global optimization algorithm, and applied it to an acyclic problem, and flowsheet with recycle.

Meyer *et al.* (2002) studied the global optimization of problems with nonfactorable constraints for which there does not exist an analytical form, proposed a sampling phase in which the nonfactorable functions and their gradients are sampled and a new blending function is constructed, presented a global optimization phase in which linear underestimators and overestimators are derived via interval anlysis and the interpolants are used as surrogates in a branch-and-cut global optimization algorithm, discussed a local optimization stage where the global optimum solution of the interpolation problem becomes the starting point for optimizing locally the original problem, and illustrated their approach through a small benchmark problem, an oilshale pyrolysis problem, and a nonlinear continuous stirred tank reactor model. Theoretical and algorithmic advances outside of Chemical Engineering in this area include the work by Jones *et al.* (1998), Jones (2001), Gutmann (2001), and the recent book by Zabinsky (2003).

6 Bilevel Nonlinear Optimization

Gumus and Floudas (2001) studied the global optimization of bilevel nonlinear programming problems which involve twice continuously differentiable functions, proposed a convex relaxation of the inner problem followed by its equivalent representation via necessary and sufficient optimality conditions, introduced the αBB global optimization principles, presented a branch-and-bound framework, and applied it to several benchmark problems and parameter estimation problems. Floudas *et al.* (2001) introduced the first rigorous global optimization approach for the calculation of the flexibility index and the feasibility test which are bilevel nonlinear optimization models, and demonstrated its applicability to a heat exchanger network problem, a pump and pipe run problem, a reactor-cooler system, and a prototype process flowsheet model.

Pistikopoulos *et al.* (2003) studied bilevel optimization models which are of linear-linear, linear-quadratic, quadratic-linear, or quadratic-quadratic type, and introduced approaches from parametric programming to transform the bilevel problem into a family of single level optimization problems which can be solved to global optimality, and presented computational results on several small benchmark problems. Gumus and Floudas (2004) studied the global optimization of bilevel mixed-integer optimization problems, proposed an approach that is applicable to mixed-integer nonlinear outer problem and twice continuously differentiable nonlinear inner problem, introduced another approach based on the convex hull representation of the inner problem, which is applicable when the inner level problem features functions which are mixed integer nonlinear in the outer variables and linear, polynomial, or multilinear in the inner integer variables, and linear in inner continuous variables; and applied it to several challenging benchmark problems.

In the remainder of this paper, we will present recent advances from Princeton University on (i) explicit facets for convex and concave envelopes for trilinear functions, (ii) convex underestimators for trigonometric functions, (iii) new convex underestimators based on a piecewise quadratic perturbation function, and (iv) the generalized αBB convex underestimators.

7 Explicit Facets of Convex and Concave Envelopes for Trilinear Monomials

Approximations of the convex envelope of nonconvex functions play a central role in deterministic global optimization algorithms and the efficiency of these algorithms is highly infuenced by the tightness of these approximations. Meyer and Floudas (2003), Meyer and Floudas (2004a) proposed explicit expressions defining the facets of the convex and concave envelopes for trilinear monomials, with mixed sign domains, as well as with positive or negative bounded domains for each variable. These advances are discussed in the sequel.

Facets of the Convex Envelope The description of the nonvertical facets depends on the signs of the bounds on $\mathbf{x}$. In this section, we present the set of facets for Case 1 (the complete set of cases can be found in the papers by Meyer and Floudas (2003), Meyer and Floudas (2004a)). The symbols x, y, and z are used to denote a permutation of x_1, x_2 and x_3. In addition to the signs of the bounds, in some cases there are auxiliary inequalities that must be satisfied for the facets to apply.

Case 1: $\underline{x} \geq 0,\ \underline{y} \geq 0,\ \underline{z} \geq 0$

Mapping $\{x_1, x_2, x_3\}$ onto $\{x, y, z\}$ in such a way that the following relations apply,

$$
\begin{array}{rcl}
\overline{x}\underline{y}\underline{z} + \underline{x}\overline{y}\overline{z} & \leq & \underline{x}\overline{y}\underline{z} + \overline{x}\underline{y}\overline{z}, \\
\overline{x}\underline{y}\underline{z} + \underline{x}\overline{y}\overline{z} & \leq & \overline{x}\overline{y}\underline{z} + \underline{x}\underline{y}\overline{z},
\end{array}
$$

the linear equalities defining the facets of $\underline{\mathcal{C}}_3(x)$ are:

$$
\begin{aligned}
w &= \underline{y}\,\underline{z}x + \underline{x}\,\underline{z}y + \underline{x}\,\underline{y}z - 2\underline{x}\,\underline{y}\,\underline{z} \\
w &= \overline{y}\,\overline{z}x + \overline{x}\,\overline{z}y + \overline{x}\,\overline{y}z - 2\overline{x}\,\overline{y}\,\overline{z} \\
w &= \underline{y}\,\overline{z}x + \underline{x}\,\overline{z}y + \overline{x}\,\underline{y}z - \underline{x}\,\underline{y}\,\overline{z} - \overline{x}\,\underline{y}\,\overline{z} \\
w &= \overline{y}\,\underline{z}x + \overline{x}\,\underline{z}y + \underline{x}\,\overline{y}z - \overline{x}\,\overline{y}\,\underline{z} - \underline{x}\,\overline{y}\,\underline{z} \\
w &= \frac{\theta}{\overline{x}-\underline{x}}x + \overline{x}\,\underline{z}y + \overline{x}\,\underline{y}z + \left(-\frac{\theta\underline{x}}{\overline{x}-\underline{x}} - \overline{x}\,\overline{y}\,\underline{z} - \overline{x}\,\underline{y}\,\overline{z} + \underline{x}\,\overline{y}\,\overline{z}\right), \\
& \quad \text{where} \quad \theta = \overline{x}\,\overline{y}\,\underline{z} - \underline{x}\,\overline{y}\,\overline{z} - \overline{x}\,\underline{y}\,\underline{z} + \overline{x}\,\underline{y}\,\overline{z} \\
w &= \frac{\theta}{\underline{x}-\overline{x}}x + \underline{x}\,\overline{z}y + \underline{x}\,\overline{y}z + \left(-\frac{\theta\overline{x}}{\underline{x}-\overline{x}} - \underline{x}\,\underline{y}\,\overline{z} - \underline{x}\,\overline{y}\,\underline{z} + \overline{x}\,\underline{y}\,\underline{z}\right), \\
& \quad \text{where} \quad \theta = \underline{x}\,\underline{y}\,\overline{z} - \overline{x}\,\underline{y}\,\underline{z} - \underline{x}\,\overline{y}\,\overline{z} + \underline{x}\,\overline{y}\,\underline{z}.
\end{aligned}
$$

Illustration To construct the lower bounding facets of $\underline{\mathcal{C}}_3(x)$ where $\mathbf{x} = [1,2] \times [1,2] \times [1,2]$ we first observe that all bounds are positive (i.e., Case 1). As the bounds on all the variables are the same it makes no difference how we map $\{x_1, x_2, x_3\}$ onto $\{x, y, z\}$. After substitution, the facets become:

$$
\begin{aligned}
w &= 1x_1 + 1x_2 + 1x_3 - 2, \\
w &= 4x_1 + 4x_2 + 4x_3 - 16, \\
w &= 2x_1 + 2x_2 + 2x_3 - 6, \\
w &= 2x_1 + 2x_2 + 2x_3 - 6, \\
w &= 2x_1 + 2x_2 + 2x_3 - 6, \\
w &= 2x_1 + 2x_2 + 2x_3 - 6.
\end{aligned}
$$

Note that the last four facets are identical and hence we only need three facets to define the convex envelope. Comparisons between the convex envelope and other approximation schemes are provided Figure 1. Explicit facets for the concave envelope and for the complete set of cases are presented in the papers by Meyer and Floudas (2003), Meyer and Floudas (2004a)).

Comparison with Other Bounding Schemes The recursive arithmetic interval (rAI) scheme for generating convex lower bounds for the multilinear monomial was compared with the arithmetic interval (AI) scheme and other bounding schemes studied by Ryoo and Sahinidis (2001).

The separation distances between the function xyz and the lower bounding functions $f_{\mathrm{AI}}(x,y,z)$ and $f_{\mathrm{rAI}}(x,y,z)$ are defined as $d_{\mathrm{AI}}(x,y,z) := xyz - f_{\mathrm{AI}}(x,y,z)$, and $d_{\mathrm{rAI}}(x,y,z) := xyz - f_{\mathrm{rAI}}(x,y,z)$. These separation distances are compared with $d_{\mathrm{C}}(x,y,z)$, the separation distance between xyz and the convex envelope. Two graphs are presented for each sign combination. In each graph y and z are constant, while the separation distances are plotted as a function of x.

In Figure 1, the AI and rAI systems are shown to generate poor bounds relative to the convex envelope.

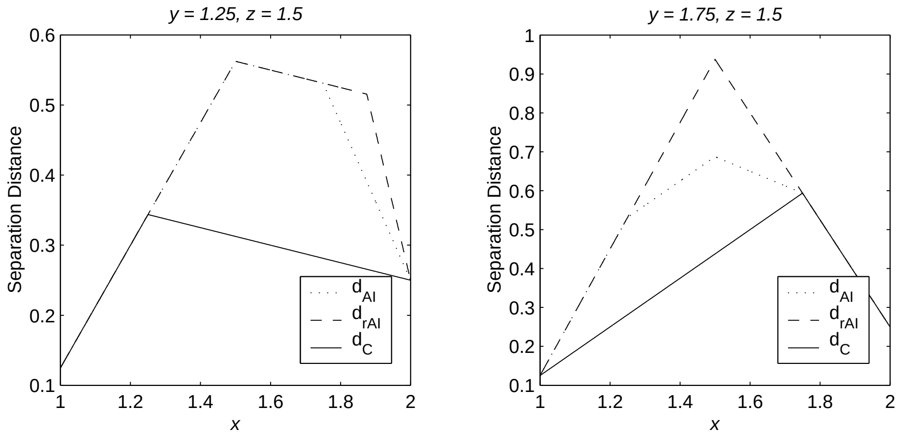

Figure 1: Comparison of Lower Bounding Separation Distances, $x \in [1,2]$, $y \in [1,2]$, $z \in [1,2]$.

8 Convex Underestimators for Trigonometric Functions

Caratzoulas and Floudas (2004) have recently proposed a C^∞ convex underestimator for the function

$$f(x) = \alpha \sin(x+s), \quad x \in [x_L, x_U], \quad \alpha > 0\,.$$

The underestimation method can be applied to one-dimensional as well as multi-dimensional problems involving trigonometric polynomials, since the product of trigonometric functions can always be decomposed into the sum of sin and cos functions with arguments that are linear combinations of the problem variables. The general case $\sin(k\,x)$, $x \in [x_L, x_U]$, reduces to the above equation form by appropriate scaling of the independent variable. A summary of the method is presented in the following for the translated function $g(x) = f(x + x_L)$, $x \in [0, D]$, where $D \equiv x_U - x_L$.

Caratzoulas and Floudas (2004) considered as underestimating function the following three-parameter (a, b, x_s) trigonometric function

$$\phi(x) = -a \sin[k(x - x_s)] + b, \quad x \in [0, D], \quad a \geq 0\,,$$

where $k = 2\zeta_1/L$, and L is the period of $\phi(x)$. For $\phi(x)$ to be convex, the conditions $x_s \leq 0$ and $L \geq 2\,(D - x_s)$ must be satisfied. They considered $L = 2\,(D - x_s) + M$, where $M \in \mathcal{R}^+$, an arbitrary positive, real number, that makes the period of $\phi(x)$ dependent on the phase shift x_s. They proved that any $M \geq 2\,D$ is sufficient, and by means of asymptotic analysis of the solution have further shown, rigorously, that the value of M does not affect how tight the underestimator will be.

Of the three equations necessary to uniquely determine the parameters a, b and x_s, two are obtained from the requirement that at the bounds of the domain $\phi(x)$ match $g(x)$, that is, $g(x_0) = \phi(x_0)$, $x_0 \in \{0, D\}$. If $g(x)$ is non-convex and the domain includes at least one minimum, the authors obtain a third equation by setting $g(q) = t(q)$, where $t(x) = g(x_0) + g'(x_t)(x - x_0)$, $x_0 \in \{0, D\}$, is tangent to $g(x)$ at the point x_t and passes through the point x_0; $q \in (0, D)$ denotes the minimum of $g(x)$ nearest to x_0. From these equations, Caratzoulas and Floudas (2004) obtained

$$a = \frac{\Delta + \delta_{\Delta,0}\,(f_L - T)}{\sin(kx_s) + \sin\{k[D - (D-q)\,\delta_{\Delta,0} - x_s]\}}$$

and

$$b = f_L - \frac{[\Delta + \delta_{\Delta,0}\,(f_L - T)]\sin(kx_s)}{\sin(kx_s) + \sin\{k[D - (D-q)\,\delta_{\Delta,0} - x_s]\}}\,,$$

where $f_L \equiv f(x_L)$, $f_U \equiv f(x_U)$, $\Delta \equiv f_L - f_U = g(0) - g(D)$; $T \equiv t(q)$; and $\delta_{\Delta,0} = 1$, if $\Delta = 0$, and zero otherwise. For the phase shift, x_s, they obtained

$$\tan(kx_s) = -\frac{\Delta\sin(kq) + (T - f_L)\sin(kD)}{\Delta(1-\cos(kq)) + (T - f_L)(1 - \cos(kD))}, \quad \Delta \neq 0$$
$$x_s = -M/2 > 0, \quad \Delta = 0\,.$$

This equation must be solved numerically (a few Newton iterations have proven sufficient) and it was shown that it always has a solution, that is, for given q and L (i.e., M) there always exists a unique $x_s < 0$ satisfying it.

For $x_0 = 0$, and $q = q_l$ the minimum of $g(x)$ nearest to $x = 0$, one obtains the tangent line $t_l(x)$. For $x_0 = D$, and $q = q_u$ the minimum of $g(x)$ nearest to the end point $x = D$, one obtains the tangent line $t_u(x)$. Thus, they obtain two sets of parameters, (a_l, b_l, x_{sl}) for $q = q_l$, and (a_u, b_u, x_{su}) for $q = q_u$, and the respective functions $\phi_l(x)$ and $\phi_u(x)$. If both $\phi_l(x)$ and $\phi_u(x)$ are underestimators, the tighter one is chosen — that is the one with the smaller amplitude parameter, a. Caratzoulas and Floudas (2004) proved that:

Property 1: For $M \geq 2D$, the function $F(x) = \phi_l(x) - \phi_u(x)$ cannot have a single root in the interval $[q_l, q_u]$.

Property 2: If $t_l(q_l) > t_u(q_l)$ and $t_l(q_u) < t_u(q_u)$, a sufficient condition for $\phi_l(x)$ ($\phi_u(x)$) to be an underestimator is that the function $(\phi_l - t_u)(x)$ ($(\phi_u - t_l)(x)$) has a root in $[q_l, q_u]$.

Theorem: At least one of the functions $\phi_l(x)$ and $\phi_u(x)$ constructed above is an underestimator.

If $g(x)$ is non-convex and the domain does *not* include a minimum, in the rather trivial case where $g(0) > g(D)$ and the tangent to $g(x)$ passing through $x = 0$ does not exist, namely $x_t \notin [0, D]$, the underestimator is a line through the end points; similarly if $g(0) < g(D)$ and the tangent to $g(x)$ passing through $x = D$ does not exist. That would also be the case if $g(0) = g(D)$. If, however, either one of the two tangents exists, an underestimator of the same form as before is sought. By enforcing the same end-points matching conditions as before, one obtains the equations for the parameters a and b. However, in the absence of a minimum point, the condition $\phi(q) = t(q)$ cannot be employed. Instead, they set $(\mathrm{d}\phi/\mathrm{d}x)_{x=0} = 0$ if $g(0) < g(D)$, or $(\mathrm{d}\phi/\mathrm{d}x)_{x=D} = 0$ if $g(0) > g(D)$, to obtain:

$$x_s = \begin{cases} -D - M/2, & \Delta < 0 \\ D - M/2, & \Delta > 0\,. \end{cases}$$

Caratzoulas and Floudas (2004) proved that the function $\phi(x)$ obtained in this manner is also an underestimator.

Maximum separation distance Caratzoulas and Floudas (2004) investigated the behaviour of the solutions with respect to the parameter $M \geq 2\,D$ as that becomes very large. In all cases, they showed that the curvature, $a\,k^2$, of $\phi(x)$ approaches a finite value. Based on their asymptotic analysis, they also have investigated the $\max_{x\in[0,D]}\{\min_{x\in[0,D]}[g(x)]-\phi(x)\}$ and its dependence on the domain size, D, as a measure of high tight an underestimator $\phi(x)$ is. Specifically, they showed that as $M \to \infty$

$$\max_{x\in[0,D]}\{\min_{x\in[0,D]}[g(x)]-\phi(x)\} \sim \begin{cases} \min_{x\in[0,D]}[g(x)] - f_L + \Delta/[4\,r\,D\,(1-r\,D)], & \Delta \neq 0 \\ \min_{x\in[0,D]}[g(x)] - f_L + (f_L - T)\,D^2/[4\,q\,(D-q)], & \Delta = 0 \end{cases}$$

where $r \equiv [\Delta\,q + (T-f_L)\,D]/[\Delta\,q^2 + (T-f_L)\,D^2]$, with $r \sim 1/D$ and $1-rD \sim \Delta\,q/[D\,(f_L - T)]$. As D increases, the quantity on the left-hand side grows linearly.

Illustration As an example, let us consider the following function: $f(x) = \sin x + \sin\frac{10x}{3} + \ln x - 0.84x,\ 1.5 \leq x \leq 12.484$. This function has a unique minimum with an objective function value of -8.7429 located at $x = 10.914$. Applying the proposed convex underestimation approach on this example to underestimate, individually, each of the first two terms in $f(x)$, the term $\ln x$, being concave, has been underestimated by a straight line connecting the end points of the domain. The first initial lower bound is -9.7818 at $x = 9.656$. Using αBB with the theoretical value $\alpha = 6.0007$, one obtains an initial lower bound of -185.2376 located at $x = 6.992$. Fig. 2 presents graphs of $f(x)$, of its trigonometric terms and their underestimators, and of the overall underestimator.

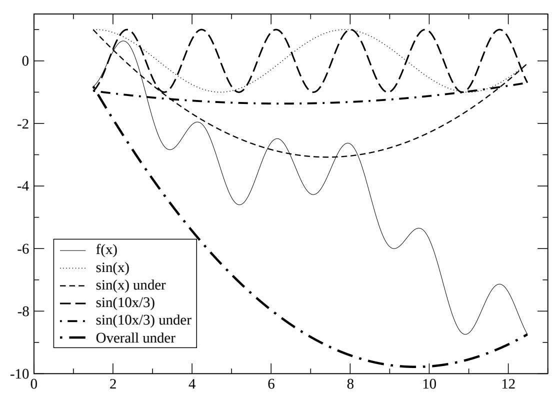

Figure 2: The function $f(x)$ in illustrative example and its underestimator. We also plot the trigonometric terms, $\sin x$ and $\sin(10x/3)$, of $f(x)$ and their individual underestimators, as computed by the method of Caratzoulas and Floudas (2004).

9 Convex Underestimators by Piecewise Quadratic Perturbation

Meyer and Floudas (2004b) introduced a refinement of the classical αBB convex underestimator, via a smooth, piecewise quadratic, perturbation function, q. In this section, we will briefly introduce the concepts behind the αBB type of underestimators, and we will subsequently focus on the new class of convex underestimators that are based upon a piecewise quadratic perturbation function.

The αBB algorithm is based on the idea of constructing a smooth convex underestimator of a nonconvex twice continuously differentiable function $f(x)$ using a convex quadratic perturbation function, $q(x)$ The convex underestimator $\phi(x)$ is defined as follows:

$$\phi(x) := f(x) - q(x).$$

The αBB convexification approach can be viewed as an approximate solution to a more general convexification problem, that of finding a convexifying perturbation function $q(x)$ which minimizes a measure, μ, of the separation between a nonconvex C^2 continuous function $f(x)$ and the convex underestimator $f(x) - q(x)$. The size of the domain $\mathbf{x}$ affects the result of every step in the α calculation and strongly influences the tightness of the resulting convex underestimator. In particular, reducing $\mathbf{x}$ reduces the mismatch between the assumed quadratic functional form and the ideal form; it reduces the overestimation in the interval extension of the Hessian matrix; and the maximum separation distance has been shown to be a quadratic function of interval length (Floudas (2000)). Constructing a convex underestimator using a number of different α vectors, each applying to a *subregion* of the full domain $\mathbf{x}$ can lead to improved convex underestimators and it is discussed in the sequel.

Let $f(x)$ be a C^2 continuous function. For each variable x_i, let the interval $[\underline{x}_i, \overline{x}_i]$ be partitioned into N_i subintervals. The endpoints of these subintervals are denoted $x_i^0, x_i^1, \cdots, x_i^{N_i}$ where $\underline{x}_i = x_i^0 < x_i^1 < \cdots < x_i^k < \cdots < x_i^{N_i} = \overline{x}_i$. In this notation the k^{th} interval is $[x_i^{k-1}, x_i^k]$. A smooth convex underestimator of $f(x)$ over $\mathbf{x}$ is defined by

$$\phi(x) := f(x) - q(x)$$

where

$$\begin{aligned} q(x) &:= \sum_{i=1}^{n} q_i^k(x_i) \quad \text{for } x_i \in [x_i^{k-1}, x_i^k], \\ q_i^k(x_i) &:= \alpha_i^k (x_i - x_i^{k-1})(x_i^k - x_i) + \beta_i^k x_i + \gamma_i^k. \end{aligned}$$

In each interval $[x_i^{k-1}, x_i^k]$, $\alpha_i^k \geq 0$ is chosen such that $\nabla^2 \phi(x)$, the Hessian matrix of $\phi(x)$, is positive semi-definite for all members of the set $\{x \in \mathbf{x} : x_i \in [x_i^{k-1}, x_i^k]\}$. $q_i^k(x_i)$ is the quadratic function associated with variable i in interval k. The function $q(x)$ is a piecewise quadratic function contructed from the functions $q_i^k(x_i)$.

The continuity and smoothness properties of $q(x)$ are produced in a spline-like manner. For $q(x)$ to be smooth the q_i^k functions and their gradients must match at the endpoints x_i^k. In addition, we require that $q(x) = 0$ at the vertices of the hyperrectangle $\mathbf{x}$. To satisfy these requirements, the following conditions are imposed for all $i = 1, \ldots, n$:

$$\begin{aligned} q_i^1(x_i^0) &= 0 \\ q_i^k(x_i^k) &= q_i^{k+1}(x_i^k) \quad \text{for all } k = 1, \ldots, N_i - 1 \\ q_i^{N_i}(x_i^{N_i}) &= 0 \\ \left.\frac{dq_i^k}{dx_i}\right|_{x_i^k} &= \left.\frac{dq_i^{k+1}}{dx_i}\right|_{x_i^k} \quad \text{for all } k = 1, \ldots, N_i - 1. \end{aligned}$$

Expanding and solving these equations, we obtain:

$$\begin{aligned}
\beta_i^1 &= \left(\sum_{k=1}^{N_i-1} s_i^k (x_i^k - x_i^{N_i})\right) / (x_i^{N_i} - x_i^0) \\
\beta_i^k &= \beta_i^1 + \sum_{j=1}^{k-1} s_i^j \ \text{ for all } k = 2, \ldots, N_i \\
\gamma_i^k &= -\beta_i^1 x_i^0 - \sum_{j=1}^{k-1} s_i^j x_i^j \ \text{ for all } k = 1, \ldots, N_i.
\end{aligned}$$

with $s_i^k = -\alpha_i^k (x_i^k - x_i^{k-1}) - \alpha_i^{k+1}(x_i^{k+1} - x_i^k)$.

This class of convex underestimators satisfies the following smoothness, underestimation, and convexity properties.

Property 1: $\phi(x) : \mathbf{x} \ni x \to \mathbb{R}$ is a continously differentiable function.

Property 2: If $\alpha_i^k \geq 0$ for all $k = 1, \ldots, N_i - 1$, and $i = 1, \ldots, n$, then $q(x)$ is concave over $\mathbf{x}$.

Property 3: $\phi(x)$ is an underestimator of $f(x)$, that is $\phi(x) \leq f(x)$ for all $x \in \mathbf{x}$.

Property 4: Let f be a function differentiable on an open set $\Omega \subset \mathbb{R}^n$, and let C be a convex subset of Ω. Then, f is convex on C if and only if its gradient ∇f is monotone on C.

Property 5: Let $f : \mathbb{R} \supset \mathbf{x} \to \mathbb{R}$ be a twice continuously differentiable function over $\mathbf{x}$. Let $\phi(x) := f(x) - q(x)$. If $\nabla^2 (f(x) - \sum_{i=1}^{n} q_i^k(x)) \geq 0$ for all $x \in I := [x_1^{k_1-1}, x_1^{k_1}] \times \cdots \times [x_n^{k_n-1}, x_n^{k_n}]$ where $x_i^{k_i} \in \{x_i^1, \ldots, x_i^{N_i-1}\},\ i = 1, \ldots, n$, then $\phi(x)$ is a convex function on $\mathbf{x}$.

Illustrative example Consider the Lennard-Jones potential energy function,

$$f(x) = \frac{1}{x^{12}} - \frac{2}{x^6}.$$

in the interval $[\underline{x}, \overline{x}] = [0.85, 2.00]$. The first term of this function is a convex function and dominates when x is small, while the second term is a concave function which dominates when x is large. The minimum eigenvalue of this function in an interval $[\underline{x}, \overline{x}]$ can be calculated explicitly as follows:

$$\min f'' = \begin{cases} \frac{156}{\overline{x}^{14}} - \frac{84}{\overline{x}^8} & \text{if} \quad \overline{x} \leq 1.21707 \\ -7.47810 & \text{if} \quad [\underline{x}, \overline{x}] \ni 1.21707 \\ \frac{156}{\underline{x}^{14}} - \frac{84}{\underline{x}^8} & \text{if} \quad \underline{x} \geq 1.21707. \end{cases}$$

The classical αBB underestimator for this function and interval is $f(x) - \frac{7.47810}{2}(\overline{x} - x)(x - \underline{x})$. The potential energy function, the classical αBB underestimator, and the $\phi(x)$ underestimators are shown in Figure 3. In this figure the α spline underestimator based on 2 subregions is denoted, $\phi^{(2)}$, while that based on 16 subregions is denoted, $\phi^{(16)}$.

10 The Generalized αBB Global Optimization Approach

In this section, the convex underestimators of the classical αBB global optimization approach are outlined first, the new class of convex underestimators is presented next along with their key theoretical properties and an illustrative example which compares the quality of the new convex understimators.

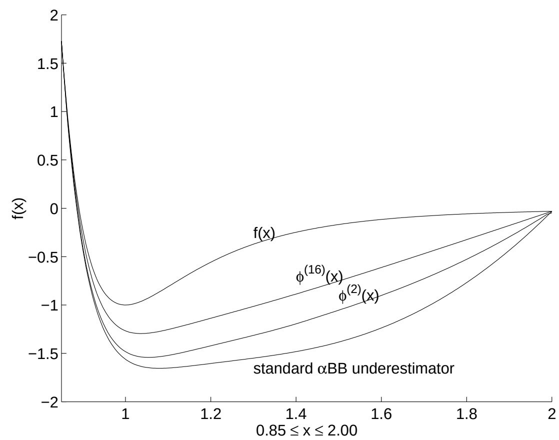

Figure 3: Lennard-Jones potential function and underestimators

Convex underestimators of the αBB method In αBB, a convex underestimator of a nonconvex function is constructed by decomposing it into a sum of nonconvex terms of special type (e.g., linear, bilinear, trilinear, fractional, fractional trilinear, convex, univariate concave) and nonconvex terms of arbitrary type. The first type is then replaced by very tight convex underestimators which are already known (Floudas (2000)). For the nonconvex terms of arbitrary type, whose convex envelops are not known, a convex underestimator is generated by adding to them the relaxation function, $\phi(x;\alpha)$:

$$\phi(x;\alpha) = -\sum_{i=1}^{n} \alpha_i (x_i - x_i^L)(x_i^U - x_i)$$

where $\alpha_i \geq 0, i = 1, 2, \ldots, n$. That is, if we assume that $f(x)$ is an arbitrarily nonconvex function, then

$$L_{\alpha BB}(x;\alpha) = f(x) + \phi(x;\alpha)$$

is an underestimator of $f(x)$. Note that since $\phi(x^L;\alpha) = \phi(x^U;\alpha) = 0$ the underestimator $L_{\alpha BB}(x;\alpha)$ coincides with $f(x)$ at the end-points of X. Also by noting that the relaxation function $\phi(x;\alpha)$ is separable we can derive the following relationship that exists among the Hessian matrices of $L_{\alpha BB}(x;\phi), f(x)$ and $\phi(x;\alpha)$

$$\nabla^2 L_{\alpha BB}(x;\alpha) = \nabla^2 f(x) + 2A$$

where $A = \nabla^2 \phi(x;\alpha) = diag\,\{\alpha_1, \alpha_2, \ldots, \alpha_n\}$. From the above equation it can be derived that $L_{\alpha BB}(x;\alpha)$ is convex if and only if $\nabla^2 L_{\alpha BB}(x;\alpha)$ is positive semi-definite matrix. It is shown in Adjiman *et al.* (1998a) that if the parameters $\alpha_i, i = 1, 2, \ldots, n$, have values greater than or equal to the negative one half of the minimum eigenvalue of the Hessian matrix $\nabla^2 f(x)$ in the whole domain $X = [x^L, x^U]$, then the underestimator $L_{\alpha BB}(x;\alpha)$ is convex function. The calculation of the smallest eigenvalue of the Hessian matrix of an arbitrarily nonconvex function is done by generating the interval hessian matrix and requiring that the interval hessian matrix is positive semi-definite.

Adjiman *et al.* (1998a), Floudas (2000) developed several methods that calculate appropriate values for all $\alpha_i, i = 1, 2, \ldots, n$ that ensure the positive semi-definiteness of the interval matrix $[\nabla^2 L_{\alpha BB}(x;\alpha)]$ and consequently the convexity of the underestimating function $L_{\alpha BB}(x;\alpha)$. These methods can be classified into two categories. The first category consists of methods that find a common value for every parameter α_i, whereas methods of the second category calculate different values for each α_i.

The most efficient of those methods is the scaled Gherschgorin. The value for each parameter α_i is determined by the equation

$$\alpha_i = \max\left\{0, -\frac{1}{2}(\underline{f}_{ii} - \sum_{j\neq i} \max\left\{|\underline{f}_{ij}|, |\overline{f}_{ij}|\right\}\frac{d_j}{d_i}\right\}$$

where $\underline{f}_{ij}$ and $\overline{f}_{ij}$ are the lower and upper bounds of $\partial^2 f/\partial x_i x_j$ as calculated by interval analysis, and $d_i, i = 1, 2, \ldots, n$ are positive parameters. A common choice for those parameters is $d_i = x_i^U - x_i^L$, which reflects the fact that variables with a wider range have a larger effect on the quality of the underestimator than variables with a smaller range.

The New Class of Convex Underestimators Akrotirianakis and Floudas (2004a) proposed the following new class of underestimating functions, $L_1(x;\gamma)$, of an arbitrary nonconvex function, $f(x)$:

$$L_1(x;\gamma) = f(x) + \Phi(x;\gamma)$$

where

$$\Phi(x;\gamma) = -\sum_{i=1}^{n}(1 - e^{\gamma_i(x_i - x_i^L)})(1 - e^{\gamma_i(x_i^U - x_i)})$$

and $\gamma = (\gamma_1, \gamma_2, \ldots, \gamma_n)^T$ is a vector of non-negative parameters. Akrotirianakis and Floudas (2004a) proved the following properties of the function $L_1(x;\gamma)$:

Property 1: $L_1(x;\gamma) \leq f(x)$, for all $x \in [x^L, x^U]$, because $\Phi(x;\gamma) \leq 0$ for all $x \in [x^L, x^U]$ and $\gamma \geq 0$.

Property 2: $L_1(x^C;\gamma) = f(x^C)$, for every corner point x^C of X, because $\Phi(x^C;\gamma) = 0$ for all $x^C \in X$.

Property 3: There exist certain values of the parameters γ_i so that $L_1(x;\gamma)$ is a convex function. This is due to the fact that the relaxation function $\Phi(x;\gamma)$ is convex for every $x \in X$ and $\gamma_i \geq 0, i = 1, 2 \ldots n$. Hence if the parameters γ_i have large enough values then all the non-convexities in the original function $f(x)$ can be eliminated, thereby producing a convex function $L_1(x;\gamma)$.

Property 4: The maximum separation distance of between the nonconvex function $f(x)$ and its underestimator $L_{G\alpha BB}(x;\gamma)$ is

$$\max_{x^L \leq x \leq x^U} \{f(x) - L_1(x;\gamma)\} = \sum_{i=1}^{n}(1 - e^{\frac{1}{2}\gamma_i(x_i^U - x_i^L)})^2$$

Property 5: The underestimators constructed over supersets of the current set are always less tight than the underestimator constructed over the current box constraints.

The values of the parameters $\gamma_i, i = 1, 2, \ldots, n$ are determined by an iterative procedure that not only guarantees the convexity of the underestimator $L_1(x;\gamma)$ but also ensures that $L_1(x;\gamma)$ is tighter than the αBB underestimator

$$L_{\alpha BB}(x;\alpha) = f(x) - \sum_{i=1}^{n} \alpha_i (x_i - x_i^L)(x_i^U - x_i)$$

The initial values of the γ_i parameters are selected by solving the system of non-linear equations

$$\ell_i + \gamma_i^2 + \gamma_i^2 e^{\gamma(x_i^U - x_i^L)} = 0, \; i = 1, 2 \ldots, n$$

where $\ell_i \leq 0, i = 1, 2, \ldots, n$. The parameters ℓ_i convey second order characteristics of the original nonconvex function into the construction process of the underestimator. Candidate values for these parameters can be provided by the scaled Gerschgorin method (Adjiman *et al.* (1998a)).

Akrotirianakis and Floudas (2004a) proved the following two important results regarding the relationship between the maximum separation distances between $f(x)$ and the two underestimators $L_1(x;\gamma)$ and $L_{\alpha BB}(x;\alpha)$.

Theorem 1: Let $\underline{\gamma} = (\underline{\gamma}_1, \underline{\gamma}_2, \ldots, \underline{\gamma}_n)^T$ be the solution of the above system. Then, the two underestimators $L_1(x;\underline{\gamma})$ and $L_{\alpha BB}(x;\underline{\alpha})$, where

$$\underline{\alpha} = \left(\frac{4(1 - e^{0.5\underline{\gamma}_1(x_1^U - x_1^L)})^2}{(x_1^U - x_1^L)^2}, \ldots, \frac{4(1 - e^{0.5\underline{\gamma}_n(x_n^U - x_n^L)})^2}{(x_n^U - x_n^L)^2}\right)^T$$

have the same maximum separation distance from $f(x)$.

Theorem 2: Let $\overline{\alpha} = (\overline{\alpha}_1, \overline{\alpha}_2, \ldots, \overline{\alpha}_n)^T$ be the values of the α parameters as computed by (10). Then, the two underestimators $L_1(x;\overline{\gamma})$ and $L_{\alpha BB}(x;\overline{\alpha})$, where

$$\overline{\gamma} = \left(\frac{2\log(1 + \sqrt{\overline{\alpha}_1}(x_1^U - x_1^L)/2)}{x_1^U - x_1^L}, \ldots, \frac{2\log(1 + \sqrt{\overline{\alpha}_n}(x_n^U - x_n^L)/2)}{x_n^U - x_n^L}\right)^T$$

have the same maximum separation distance from $f(x)$.

The above two theorems reveal that for any $\gamma \in [\underline{\gamma}, \overline{\gamma}]$ there exists an $\alpha \in [\underline{\alpha}, \overline{\alpha}]$, such that the underestimators $L_1(x;\gamma)$ and $L_{\alpha BB}(x;\alpha)$ have the same maximum separation distance from the nonconvex function $f(x)$. From all these pairs of underestimators, the only one that is known to be convex *a priori* is $L_{\alpha BB}(x;\overline{\alpha})$, since this is the one resulting from the classical αBB method. However, for most arbitrarily nonconvex functions the underestimators $L_{\alpha BB}(x;\alpha)$ and $L_1(x;\gamma)$ are convex within a large portion of the intervals $[\underline{\alpha}, \overline{\alpha}]$ and $[\underline{\gamma}, \overline{\gamma}]$ respectively. Based on the above observations, it is natural to search for a vector γ in the interval $[\underline{\gamma}, \overline{\gamma}]$ or for a vector α in the interval $[\underline{\alpha}, \overline{\alpha}]$, so that at least one of the underestimators $L_1(x;\gamma)$, $L_{\alpha BB}(x;\alpha)$ is convex.

Akrotirianakis and Floudas (2004a) proposed an approach that iteratively determines, using interval analysis, the minimum values of the γ or α parameters that result in an underestimator that is convex and tighter than the classical αBB method. They also developed the generalized αBB global optimization approach, denoted as GαBB, and perfomed extensive computational studies for box constrained global optimization problems (see Akrotirianakis and Floudas (2004b)).

Illustrative Example This example consists of the global minimization of a potential function describing the pseudoethane molecule (see Floudas (2000)) which takes the form:

$$\begin{aligned} f_1(x) &= \frac{588600}{(3r_0^2 - 4cos(\theta)r_0^2 - 2(sin^2(\theta)cos(x - \frac{2\zeta_1}{3}) - cos^2(\theta))r_0^2)^6} \\ &- \frac{1079.1}{(3r_0^2 - 4cos(\theta)r_0^2 - 2(sin^2(\theta)cos(x - \frac{2\zeta_1}{3}) - cos^2(\theta))r_0^2)^3} \\ &+ \frac{600800}{(3r_0^2 - 4cos(\theta)r_0^2 - 2(sin^2(\theta)cos(x) - cos^2(\theta))r_0^2)^6} \\ &- \frac{1071.5}{(3r_0^2 - 4cos(\theta)r_0^2 - 2(sin^2(\theta)cos(x) - cos^2(\theta))r_0^2)^3} \\ &+ \frac{481300}{(3r_0^2 - 4cos(\theta)r_0^2 - 2(sin^2(\theta + \frac{2\zeta_1}{3})cos(x) - cos^2(\theta))r_0^2)^6} \\ &- \frac{1064.6}{(3r_0^2 - 4cos(\theta)r_0^2 - 2(sin^2(\theta + \frac{2\zeta_1}{3})cos(x) - cos^2(\theta))r_0^2)^3} \end{aligned}$$

where r_0 is the covalent bond length ($r_0 = 1.54A$), θ is the covalent bond angle ($\theta = 109.5^o$) and x is the dihedral angle ($x \in X = [0, 2\zeta_1]$).

The value of the α parameter computed by the classical αBB method using the scales Gerschgorin approach is $\overline{\alpha} = 77.124$, and the corresponding value for the γ parameter, is $\overline{\gamma} = 1.0673$. Solving for γ we obtain $\underline{\gamma} = 0.8521$ and the corresponding value for the α parameter, is $\underline{\alpha} = 18.579$. The convexity verification algorithm of Akrotirianakis and Floudas (2004a) checks whether there exist values of $\gamma \in [\underline{\gamma}, \overline{\gamma}]$ and $\alpha \in [\underline{\alpha}, \overline{\alpha}]$ such that the underestimator $L_{\alpha BB}(x; \alpha)$ is convex. After 16 iterations it concludes that if $\alpha = \underline{\alpha}$, then $L_{\alpha BB}(x; \alpha)$ is a convex underestimator of $f_1(x)$. The minima of the two underestimators $L_{\alpha BB}(x; \overline{\alpha})$ and $L_{\alpha BB}(x; \alpha)$ are -762.2377, and -184.4244, respectively. Figure 4 compares the two underestimators $L_{\alpha BB}(x; \overline{\alpha})$ and $L_{\alpha BB}(x; \alpha)$ and shows the improvement.

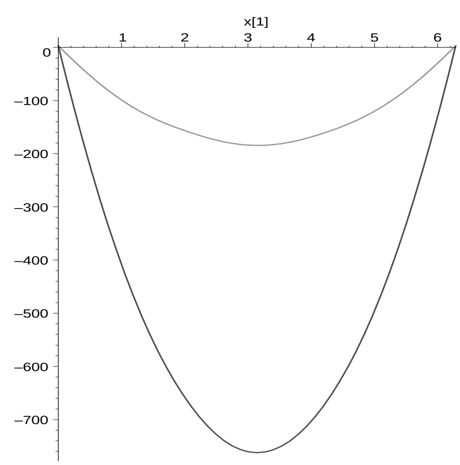

Figure 4: Comparison of the underestimators $L_{\alpha BB}(x; \overline{\alpha})$ and $L_{\alpha BB}(x; \alpha)$ of the nonconvex function $f_1(x)$

Summary

This paper reviewed the advances in global optimization during the period 1998-2003. The focal point was novel theoretical, algorithmic, and applications oriented advances on deterministic global optimization methods for (i) general twice differentiable NLPs, (ii) mixed integer nonlinear optimization problems MINLPs, (iii) models with differential-algebraic constraints, (iv) grey-box and nonfactorable models, and (iv) bilevel nonlinear and mixed-integer optimization. Recent advances from Princeton University were also presented on convex and concave envelopes for trilinear monomials, convex underestimators for trigonometric functions, a new class of convex smooth piecewise underestimators with a quadratic perturbation that use as a basis the classical αBB type of underestimators, and a new class of generalized and improved convex underestimators for twice continuously differentiable functions. Illustrative examples were presented to highlight the potential benefits of these recent advances.

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