Pergamon

PII: S0098-1354(96)00306-7

Computers chem. Engng Vol. 21, No. 7, pp. 733–738, 1997

0098-1354/97 $17.00+0.00

Synthesis under uncertainty with simulators

Prosenjit D. Chaudhuri and Urmila M. Diwekar*

Department of Civil & Environmental Engineering and Engineering & Public Policy, Carnegie Mellon University, Pittsburgh, PA 15213, U.S.A.

(Received 6 January 1995; revised 20 May 1996)

Abstract

Synthesis capabilities based on conventional mathematical programming techniques such as MINLP algorithms can pose certain difficulties, especially with black-box, sequential modular simulators (SMS). Further, since commercial simulators are based on a deterministic framework, synthesis of processes under uncertainty is not possible. This paper presents a generalized synthesis capability built around the public version of the ASPEN Process Simulator based on stochastic annealing — a new algorithm for synthesis under uncertainty. It is complementary to MINLP synthesis capabilities in ASPEN, and provides an automated approach to stochastic synthesis. Further, it achieves the trade-off between accuracy and computational efficiency, by selecting the optimal number of samples. The implementation of this new synthesis capability in ASPEN is illustrated by synthesizing a bench-mark process in chemical synthesis — the HDA process. The results show that stochastic annealing, which overcomes many of the problems associated with a MINLP approach in a simulation environment, can be a potential tool for stochastic synthesis with deterministic, black-box simulators. © 1997 Elsevier Science Ltd. All Rights reserved

1. Introduction

Over the past decade, sequential modular simulators, such as FLOWTRAN, PRO-II or ASPEN, have been used widely in chemical engineering design. Although these simulators have been used to simulate new processes and analyze existing ones, they do not possess any synthesis capability. However, since these simulators are equipped with rigorous process, sizing and cost models, and are used more widely commercially than equation-oriented simulators, synthesis capabilities built around them are more desirable. A MINLP synthesis capability built around the public version of ASPEN represents a first step towards synthesis with sequential modular simulators (Diwekar *et al.*, 1992).

However, a MINLP approach to synthesis may pose certain problems, especially with sequential modular simulators:

- The more efficient Outer Approximation (OA) (Duran and Grossmann, 1986) and Generalized Benders Decomposition (GBD) (Benders, 1962; Geoffrion, 1972) algorithms are based on a two-level optimization procedure. The black-box nature of the models in sequential modular simulators prevents the upper level MILP master problem from receiving linearization information from the lower level NLP sub-problem. This leads to the *Implicit Constraint Problem*, which can be circumvented through the addition of pseudo-decision variables (Diwekar and Rubin, 1993).
- A flowsheet decomposition strategy is necessary to sub-optimize non-existing units in the superstructure through a Lagrangian decomposition scheme (Kocis and Grossmann, 1989).

Further, typical algorithms such as OA rely on a feasible NLP sub-problem (Duran and Grossmann, 1986). Functional discontinuities in a process synthesis application (Painton and Diwekar, 1995, 1994), and the problem of non-convexities (Floudas *et al.*, 1989), may introduce additional strategies to be incorporated for a MINLP algorithm to be applicable. An alternative approach to synthesis is based on simulated annealing, which is complementary to MINLP synthesis (Floquet *et al.*, 1993). Although such an approach circumvents the problems addressed previously associated with MINLP synthesizers around SMS, and can be easily incorporated as a synthesis capability in a sequential modular environment, it is computationally intensive compared to MINLP synthesis techniques.

Synthesis under uncertainty, on the other hand, poses a difficult problem to be handled by conventional MINLP techniques or the simulated annealing algorithm due to the computational intensity associated with it. The

* To whom correspondence should be addressed.

idea of a new algorithm for synthesis under uncertainty, referred to as the stochastic annealing algorithm, has been presented in the context of a design of a multi-stage compression/expansion system in the presence of uncertainties but involved discrete decisions only (Painton and Diwekar, 1995). The general process synthesis problem involves discrete and continuous decisions, and can involve uncertainties in process and economic parameters as well. The stochastic annealing algorithm, although a variant of the simulated annealing algorithm, has been extended to incorporate both types of decisions (i.e. discrete and continuous) and different probabilistic objective functions (e.g. mean, variance) encountered in a general flowsheet synthesis problem. It exhibits a systematic, automated framework developed for synthesis under uncertainty within a sequential modular environment. It can overcome many of the problems associated with the MINLP synthesis capability around SMS and is more efficient than the simulated annealing approach for synthesis under uncertainty. In the next section, this new algorithm is presented, followed by a discussion on the implementation of the new synthesis capability around the public version of the ASPEN Process Simulator. Finally, the new synthesis capability is demonstrated via a bench-mark chemical process in synthesis — the HDA process.

2. The stochastic annealing algorithm

The stochastic annealing algorithm is a variant of simulated annealing — a probabilistic-based, combinatorial approach to optimization (vanLaarhoven and Aarts, 1987). Simulated annealing requires definition of a *configuration* and a *temperature* analogous to the physical annealing process (Kirkpatrick *et al.*, 1983). The configuration of the system is defined by the decision variables, and for each configuration the objective function is evaluated. For a minimization problem, any decrease in the objective function owing to a *move* (a random change from one configuration to another) is always accepted. On the contrary, if the move results in an increase (Δ) in the objective function, the move is accepted according to the Metropolis criteria [i.e. with a probability $\exp(-\Delta/T)$]; where Δ is the corresponding change in the objective function and T is the corresponding annealing temperature. This offers the alternative of a random uphill move, so the system can jump out of a local minima towards (at least theoretically) a global minima. A detailed overview of simulated annealing is not presented here and the interested reader is referred to Rutenbar (1989).

The stochastic annealing algorithm is designed to automate the synthesis of processes under uncertainty by selection of the optimal number of samples. The generalized approach to synthesis under uncertainty usually involves the solution of a stochastic optimization problem. This can be computationally intensive, since the process model has to be run with each set of the sampled values of the uncertain parameters recursively for a given sample size, to obtain cumulative distribution of the process objective and constraints. The optimizer analyzes this cumulative probability distribution of the objective function and constraints, and then predicts new decision variables. In order to reduce the computational burden, the new stochastic annealing algorithm is designed to predict the minimal sample size automatically, the sample size being another decision variable (Chaudhuri and Diwekar, 1996). The estimate of the objective function (in this case a probability function, e.g. mean, fractile or variance) depends on the sample size. Consequently, the goal of the stochastic annealing algorithm is to select the sample size judiciously, since the sample size required for the desired sampling accuracy depends on the current process configuration. This automatic sample size selection is achieved by the addition to the objective function of a penalty term, which incorporates the error band-width of any probabilistic function such as the mean, fractile or variance (Chaudhuri and Diwekar, 1996). The penalty term is weighted by a factor $b(t) = b_0/k^t$; where b_0 and k are constants and t is the current annealing temperature level. The constants b_0 and k are usually evaluated by running the process models several times, such that the penalty term is usually less than 5% of the objective function. This ensures that the augmented objective function is unchanged from the real objective. The penalty term, which is a function of the error band-width (and hence the sample size), therefore discourages a large sample size in the initial stage of the annealing procedure, when the algorithm is exploring the configuration space to identify regions of local optima. Once the process configuration is close to the optimum, the weighted factor $b(t)$ maintains the sample size constant. This enables an automatic, adaptive manipulation of the sample size such that the computational intensity resulting from running the model with a fixed, large sample size is overcome easily. In the next section, this new synthesis capability as implemented around the ASPEN Process Simulator is discussed.

3. Implementation of the stochastic annealing synthesis capability in ASPEN (public version)

The input structure of the stochastic annealing process synthesizer in ASPEN, follows ASPEN's keyword input structure (ASPEN User Manual, 1982). The ASPEN input file, which defines the various process alternatives (including stream and block connectivities, calculation sequences, loop convergence parameters, etc.), is also keyword driven and contains detailed information essential for the synthesis of the flowsheet. Such information incorporates the discrete variables which define subsystems of flowsheet, continuous variables (e.g. equipment sizes, pressures and temperatures associated with a chosen subsystem of the flowsheet, etc.) and any other input data for the process flowsheet. It contains also the parameters essential for the stochastic annealing algorithm, such as the initial starting temperature, cooling schedule, number of moves at a given temperature,

number of temperature levels, upper and lower bounds of the variables, and the initial configuration.

A schematic diagram of this synthesis capability under uncertainty in ASPEN is illustrated in Fig. 1. The first step is essentially the formulation of a superstructure which has embedded in it all the alternative flowsheet structures. The presence/absence of units in the superstructure are determined by the fraction of flows through these units. The process synthesis environment in ASPEN based on the stochastic optimization framework consists of two loops: an inner and an outer loop. The inner one is the sampling loop and consists of a stochastic block (STOCHA), which assigns probability distributions to the uncertain parameters and generates a sample set based on a sampling technique. The outer one consists of the synthesis or optimization loop and involves a user-defined optimization or synthesis block (STANOP), similar to an unit-operation block in ASPEN (public version). This block predicts the decision variables (both discrete and continuous) and the sample size, which is used by STOCHA to generate the samples. The decision variables and the sample set for the uncertain parameters are transferred to the ASPEN model of the superstructure using the FORTRAN access block (SIMACCESS).

In addition to the optimizer, the stochastic block, and the superstructure, there are two FORTRAN blocks for controlling the recycle of the sampling and the optimization loops. After transferring the set of the decision variables and the sample set of the uncertain parameters to the flowsheet, the flowsheet is evaluated repeatedly for each sample set, until all the samples have gone through the cycle. This completes the cycle of the sampling loop, the probability objective function (mean, variance, or the fractile) and the constraints are then computed along with the penalty function, and transferred to the optimizer. The optimizer based on the stochastic annealing algorithm decides new decision variables (including the sample size), and the cycle repeats. The termination criteria for the outer or synthesis/optimization loop is governed by (i) tolerance imposed on the change in the objective function at the end of each temperature level; or (ii) the number of temperature levels in the annealing schedule.

4. Synthesis of the HDA process

The process chosen to illustrate the features of the stochastic annealing algorithm is the hydrodealkylation (HDA) of toluene (Douglas, 1988). The problem presented and solved here is the selection of the flowsheet configuration and some of the operating conditions that maximize profit. It has been assumed that uncertainties exist in some of the process and economic parameters, and are represented by probability distributions (Table 1).

4.1. HDA process description

The pertinent reactions of the HDA process are:

$$C_6H_5CH_3+H_2\rightarrow C_6H_6+CH_4.$$

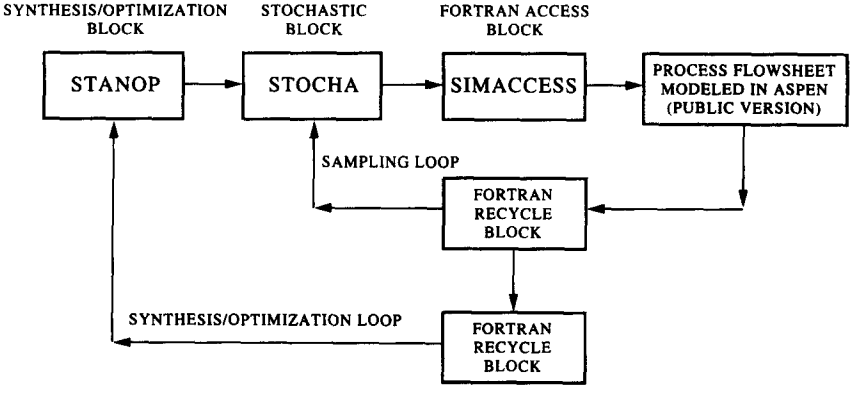

Fig. 1. Schematic diagram of the synthesis capability in ASPEN using stochastic annealing.

Table 1. Description of uncertainties in process and economic parameters*

Uncertainty	Distribution type	Lower bound	Upper bound
Fixed charge cost of compressor	Normal	0.8	1.2
Fixed charge cost of the stab. column	Uniform	0.6	1.4
Cost of hydrogen feed	Normal	0.6	1.4
Cost of toluene feed	Triangular	0.5	1.5 (1.0)
Cost of benzene product	Normal	0.7	1.3
Cost of diphenyl product	Lognormal	0.6	1.1
Purity of the benzene product	Normal	0.95	1.04

*For normal and lognormal distributions the lower and the upper bounds represent 0.001 and 0.999 quantiles, respectively. However, for the triangular distribution, the value within parentheses represents the middle apex of the triangle representing the distribution.

In addition to this desired reaction, an undesired reaction:

$$2C_6H_6 \rightleftharpoons C_6H_5 + H_2$$

also occurs. These homogeneous gas-phase reactions occur in the range of 894–974 K. The first reaction is exothermic and can be carried out in either an adiabatic reactor ($y1=1$, $y2=0$) or an isothermal reactor ($y2=1$, $y1=0$). A molar ratio of at least 5:1 hydrogen to aromatics is maintained to prevent coking and the reactor effluents must be quenched to 894 K to prevent coking in the heat-exchanger following the reactor. The superstructure for the HDA process synthesis problem is presented in Fig. 2.

The raw hydrogen stream has a purity of 95% (the rest is methane) and is mixed with fresh inlet stream of toluene, recycle hydrogen and toluene streams. The feed is heated in a furnace before being fed to the reactor. The reactor effluent containing unreacted hydrogen and toluene, desired product benzene and undesired diphenyl and methane is quenched and subsequently cooled in a flash separator to condense the aromatics from non-condensable hydrogen and methane. The vapor stream from the flash unit contains hydrogen, which needs to be recycled. The liquid stream containing traces of hydrogen and methane needs to be separated from the aromatics in a secondary separation unit which consists of either a stabilizing column ($y3=1$, $y4=0$) or a second flash unit ($y4=1$, $y3=0$). Having separated the hydrogen and undesired methane, the liquid stream from the secondary unit consisting of benzene, diphenyl and toluene are separated in two distillation columns. The first column separates the product benzene from diphenyl/toluene, while the second separates the diphenyl from toluene which is recycled back into the reactor.

4.2. *Results and discussion*

The mixed-discrete optimization problem involves four binary (0–1) variables ($y1$–$y4$) and six continuous variables, namely furnace temperature, temperature of the isothermal reactor, conversion in the two reactors, and the molar flow-rates of the hydrogen and toluene streams. The objective function (annualized profit) is subjected to the following constraints:

1. The benzene production rate must be maintained at 120 kmol/h.
2. The isothermal and the adiabatic reactors must have a volume less than 500 m^3.
3. The maximum number of trays allowable in the stabilizing column is 100.
4. The hydrogen feed must have a purity of 95%.
5. The purity of the benzene feed is approximately 95%. The allowable uncertainty in the purity measurement of the benzene product varies between 0.95 and 1.04 of the nominal value of 95% product purity.
6. The net change in the enthalpy after reaction in the isothermal reactor must be maintained constant at 1.1 kcal/mol by circulating cooling water.

The deterministic case (i.e. when no uncertainties are present in the process/economic parameters) has been formulated as a MINLP problem and solved using an alternating sequence of NLP subproblems and MILP master problems using a decomposition strategy (Diwekar *et al.*, 1992). Since simulated annealing presents a deterministic framework and is the basis for the new

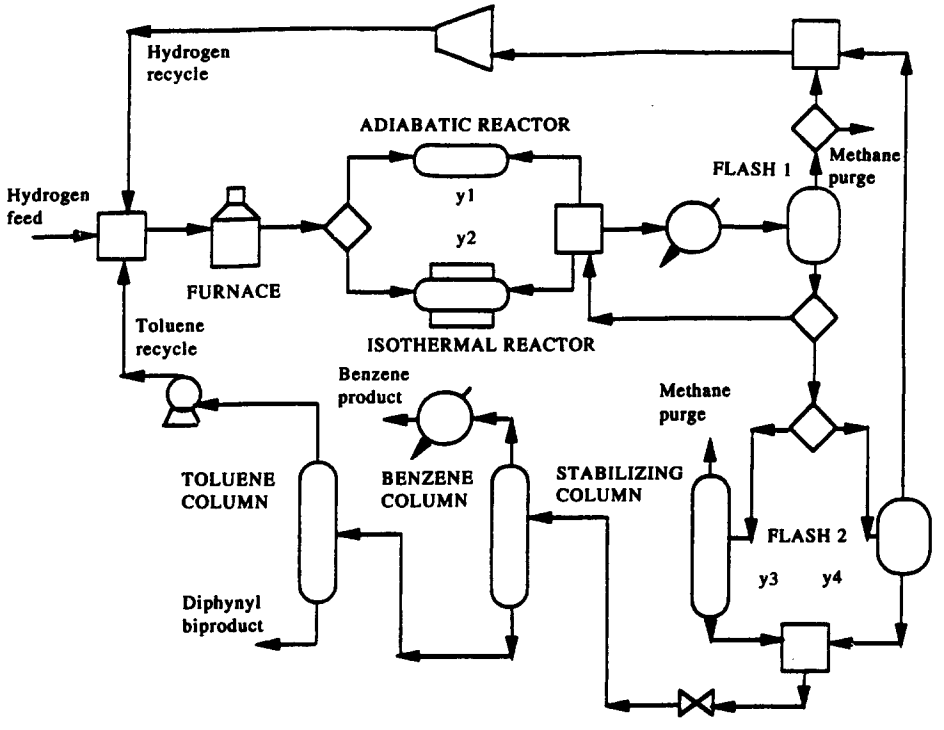

Fig. 2. Superstructure of the HDA process synthesis problem.

Table 2. Summary of results for the HDA problem (Deterministic Case)

Decision variables	Algorithm	
	MINLP	Sim. annealing
y1	1	1
y2	0	0
y3	0	0
y4	1	1
Conversion	0.632	0.612
Reactor temp.	866.3 K	865.7 K
Furnace temp.	894.1 K	893.5 K
Molar flow-rate (hydrogen feed)	247.78 kmol/h	250.137 kmol/h
Molar flow-rate (toluene feed)	128.35 kmol/h	127.21 kmol/h
Maximized profit, $/yr	1041.5	1035.6

Table 3. Summary of results for the HDA problem (Probabilistic Case)

Decision variables	Algorithm	
	Sim. annealing (sample size fixed)	Stoch. annealing
y1	1	1
y2	0	0
y3	0	0
y4	1	1
Conversion	0.603	0.609
Reactor temp.	868.5 K	866.8 K
Furnace temp.	891.8 K	893.0 K
Molar flowrate (hydrogen feed)	252.44 kmol/h	252.62 kmol/h
Molar flowrate (toluene feed)	126.84 kmol/h	127.31 kmol/h
CPU time	74000 s	14516 s
Maximized profit, $/yr	621.1	649.7

stochastic annealing algorithm, it was deemed necessary to validate the simulated annealing algorithm with the MINLP technique for the deterministic case. It was observed that simulated annealing predicted the same solution, but was computationally intensive compared to conventional MINLP methods, particularly because of the absence of the definition of a suitable step-size for continuous variables in simulated annealing. The optimal structural topology for the deterministic model requires the presence of the adiabatic reactor and the second flash unit (Table 2).

Uncertainties in the process and economic parameters result in a probabilistic model. The objective function in this case is an expected value of the annualized profit. The flowsheet was synthesized using stochastic annealing and compared with simulated annealing modified for a probabilistic framework with a fixed sample size (50). Stochastic annealing determined the optimal flowsheet configuration using fewer samples at each temperature level than the simulated annealing approach with fixed sample size, resulting in 80% savings in CPU time. The presence of uncertainties, however, resulted in a decrement of the annualized expected value of the maximum profit attainable (Table 3). This is attributed to the fact that all the uncertainties except for the purity of benzene were assumed to be in the economic parameters. The decision variables are more dependent on process parameters and are practically unaffected by uncertainties in cost. On the contrary, the objective function (expected value of the annualized profit), being an economic index, is affected more by uncertainties in economic parameters.

5. Summary

An automated approach for synthesis under uncertainty based on stochastic annealing, a new variant of simulated annealing, has been implemented as a stochastic synthesis capability in a sequential modular simulation environment — the ASPEN Process Simulator (public version). The proposed algorithm is complementary to MINLP methods for synthesis and overcomes almost all the problems associated with the MINLP synthesis capability around an SMS. It achieves the trade-off between computational efficiency and accuracy by selecting the optimal number of samples. The stochastic annealing algorithm determined the optimal flowsheet configuration with considerable savings in CPU time (80%), and is more efficient than a simulated annealing approach with a fixed sample size for synthesis under uncertainty. Special techniques currently are being investigated to speed up the annealing procedure with large number of continuous variables and to characterize accurately a suitable penalty term for more efficient sampling techniques based on fractal dimensions.

References

ASPEN User's Manual. Massachusetts Institute Of Technology, Cambridge (1982).

Benders, J. F. (1962) Partitioning procedures for solving mixed-variables programming problems. *Numer. Math.* **4**, 238

Chaudhuri, P. D. and Diwekar, U. M. (1996) Process synthesis under uncertainty: a penalty function approach. *AIChE J.* **42**, 3 742

Diwekar, U. M., Grossmann, I. E. and Rubin, E. S. (1992) An MINLP Process Synthesizer for a sequential modular simulator. *Ind. Engng Chem. Res.* **31**, 31

Diwekar, U. M. and Rubin, E. S. (1993) Efficient handling of the implicit constraints problem for the ASPEN MINLP synthesizer. *Ind. Engng Chem. Res.* **32**, 2006

Douglas, J. M. (1988) *Conceptual Design of Chemical Processes*. McGraw–Hill, New York.

Duran, M. A. and Grossmann, I. E. (1986) An outer-approximation algorithm for a class of mixed-integer nonlinear program. *Math. Programming* **36**, 307

Floquet, P., Pibloulleau, L. and Domenech, S. (1993) Recent trends in process optimization. *Desalination* **92**, 1

Floudas, C., Aggrawal, S. and Ciric, A. (1989) Global optimum search for nonconvex NLP and MINLP problems. *Computers Chem. Engng* **13**, 1117

Geoffrion, A. M. (1972) Generalized benders decomposition. *J. Optim. Theory Appl.* **10**, 4 237

Kirkpatrick, S., Gelatt, C. and Vecchi, M. (1983) Optimization by simulated annealing. *Science* **220**, 670

Kocis, G. R. and Grossmann, I. E. (1989) A modeling and decomposition strategy for MINLP optimization of process flowsheets. *Computers Chem. Engng* **13**, 797

Painton, L. A. and Diwekar, U. M. (1995) Stochastic annealing for synthesis under uncertainty. *Eur. J. Oper. Res.* **83**, 489

Painton, L. A. and Diwekar, U. M. (1994) Synthesizing optimal design configurations for a Brayton cycle power plant. *Computers Chem. Engng* **18**, 369

Rutenbar, R. A. (1989) Simulated annealing algorithms: an overview. *IEEE Circ. Dev. Mag.* **100**, 19

vanLaarhoven, P. J. M. and Aarts, E. H. L. (1987) *Simulated annealing: Theory and Applications*. Reidel, Holland.