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On the development and implementation of knowledge-driven optimisation schemes: an application in non-isothermal reactor network synthesis

Victoria M. Ashley, Patrick Linke*
Centre for Process and Information Systems Engineering
School of Engineering, University of Surrey
Guildford GU2 7XH United Kingdom

Abstract
Knowledge driven optimisation has been developed in an attempt to overcome difficulties in applying existing reactor network synthesis methods to complex systems. Knowledge derived from kinetic relationships is applied to superstructure optimisation in the form of a customised rule-based Tabu Search where rules are used to guide optimisation decisions. Nonisothermal behaviour is represented using temperature profiles along the length of a reactor. Results show the method outperforms a standard Tabu Search both in solution quality and speed of convergence.

Keywords: Knowledge driven optimisation, data mining

1. Introduction

Existing methods for reactor network synthesis include graphical based Attainable Region (Horn, 1964; Glasser et al., 1987), superstructure optimisation using both deterministic (Achenie & Biegler, 1990; Kokossis & Floudas, 1990; Schweiger & Floudas, 1999) and stochastic optimisation methods (Marcoulaki & Kokossis, 1999; Linke & Kokossis, 2003), and targeting and combined approaches (Balakrishna & Biegler, 1992; Lakshmanan & Biegler, 1996). Dimensionality limitations, initialisation and convergence problems, and long computational times are some of the challenges that face existing methods. Knowledge driven reactor network synthesis (Ashley & Linke, 2004a,b) has been developed in an attempt to overcome problems suffered by existing techniques when applied to complex reaction systems.

Ashley and Linke (2004a,b) have presented an approach for isothermal reactor network synthesis that uses knowledge of the process system to guide the search. Data mining techniques analyse the kinetic model to uncover relationships between process variables and high performance regions. This knowledge is translated into rules that guide the optimisation search towards promising spaces by suggesting moves based on rule violations.

* Author to whom correspondence should be addressed: p.linke@surrey.ac.uk

As many industrial applications are nonisothermal, the work presented in this paper extends the novel approach to enable application with reaction schemes whose reaction rates are highly temperature dependent. A similar approach to the basic knowledge-driven rule-based methodology outlined in Ashley and Linke (2004a,b) is adopted and temperatures are incorporated within the reactor network representation by adopting temperature profiles along the length of the reactor, as proposed by Mehta and Kokossis (2000).

2. Method

Knowledge driven reactor network synthesis derives information from a kinetic model to devise rules describing high performance regions and guide the optimisation search (Ashley & Linke, 2004a,b). This paper briefly describes extensions required for application to nonisothermal systems. Contrary to the isothermal approach where temperature is assumed constant, nonisothermal applications take into account the temperature dependencies of reaction rates, which are often defined according to the Arrhenius equation:

$$k_{ir} = A_{ir} e^{-\frac{E_{ir}}{RT}} \tag{1}$$

where k_{ir} = reaction rate constant for reaction ir, A_{ir} = Arrhenius constant for reaction ir, E_{ir} = activation energy for reaction ir, T = reactor temperature, R = universal gas constant. The procedure for nonisothermal systems follows the three main steps of knowledge driven optimisation for isothermal reactor networks: 1) Reaction pathway analysis, 2) Data mining and rule formulation, and 3) Optimisation and synthesis.

2.1 Reaction Pathway Analysis

Reaction pathways are generally considered independent of temperatures for a given system and the procedure for isothermal systems applies directly to nonisothermal applications. Reaction pathway analysis involves defining the reaction network through construction of a reaction network diagram. This allows the identification of serial and parallel reaction pathways, which are highly significant when determining the optimal structure. Rate indicators are defined as ratios of reaction rates for serial and parallel connections and allow the comparative progress of the reactions to be monitored. Concentration and temperature profile plots are observed to gain familiarity with the process system.

2.2 Data Mining and Rule Formulation

Knowledge discovery and data mining are conducted to extract relationships and trends from multitudes of data that may not be immediately apparent due to the sheer amount of data or the complex interactions between different data fields. A database is generated based on kinetic information, and consists of combinations of temperatures and component concentrations, from which reaction rate constants, reaction rates and

rate indicators are calculated. Data mining is conducted using classification trees to reveal more information about the relationships between the rate indicators and concentrations, temperatures and rate constants. Trends leading to good performance are identified and these are translated into rules that can be applied during optimisation searches. The rules define upper and lower bounds on concentrations, reaction rates, rate indicators or temperatures that lead to high quality solutions.

2.3 Optimisation and Synthesis

Optimisation and synthesis is conducted using superstructure optimisation based on Tabu Search (Glover 1989) and enriched by the uncovered rules. The rules are implemented as upper and lower bounds on process variables, against which an existing structure is assessed. Rule violations suggest a move direction, from which multiple moves may be selected. At each iteration, Tabu Search generates a set of new solutions in the neighbourhood of the current structure and the best performing solution is accepted provided it is not on the Tabu List.

The superstructure representation consists of all possible combinations of ideal reactor units including continuous stirred tank reactors (CSTR), plug flow reactors (PFR) and distributed side stream reactors (DSSR). Plug flow behaviour is approximated as a cascade of equal volume sub-CSTRs (Kokossis & Floudas, 1990). Variations in temperature are achieved by adopting temperature profiles along the length of each non-CSTR reactor as first proposed by Mehta and Kokossis (2000). Temperature profiles are implemented by varying the temperature between sub-CSTRs. This is equivalent to an isothermal reactor with heat transfer to maintain constant temperature, but with the flexibility of selecting appropriate temperatures depending on the progress of the reactions. The approach allows the incorporation of nonisothermal systems without the complication of solving energy balances.

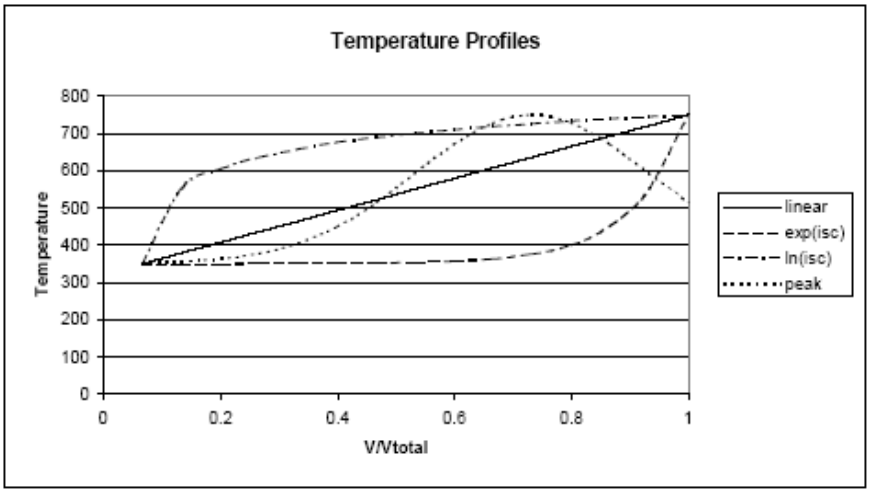

Figure 1. Nonisothermal temperature profiles, T_{in} = 350 K, T_{out} = 750 K

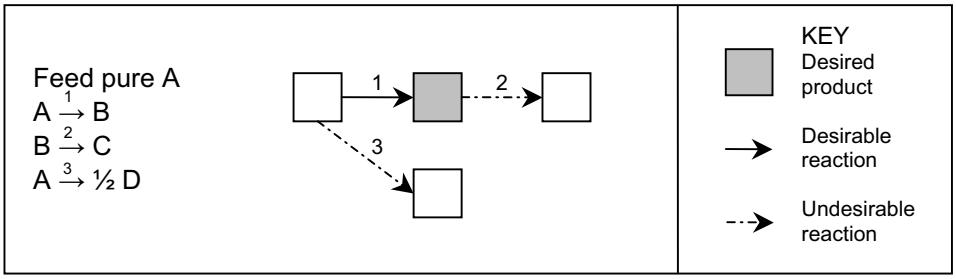

Figure 2. Reaction network diagram for the nonisothermal Van de Vusse example

Temperature profiles include 1) flat, 2) linear, 3) exponential, 4) logarithmic and 5) peaked profiles, as shown in Figure 1. A set of moves is included to allow for changes in temperature and temperature profiles.

3. Illustrative Example

A nonisothermal version of the Van de Vusse reaction (Mehta & Kokossis, 2000) is considered as an example (Figure 2). The first two reactions are of first order while the third is of second order. The feed consists of 10 mol/s of pure A at a concentration of 1 mol/L. The objective is to maximise the concentration of component B. The reaction rate constant is dependent upon the Arrhenius temperature relation (1) above. Reaction rate data for the nonisothermal Van de Vusse example is contained in Table 3. The allowed temperature range is between 177 – 537 degrees Celsius (450 – 800 Kelvin). The volume of each reactor is allowed to vary between 3.6 – 600L. Figure 2 shows the reaction network diagram. Rules developed from data mining results are shown in Table 4. Move categories are explained in Ashley and Linke (2004a).

Table 1. Reaction rate data for nonisothermal Van de Vusse example

Rxn (*ir*)	A_{ir}	E_{ir} *(cal/mol)*
1	5.4×10^{9} (1/h)	15840
2	3.6×10^{5} (1/h)	7920
3	1.6×10^{12} (1/h·mol)	23760

Table 2. Nonisothermal Van de Vusse rule summary

No.	Location (reactor)[1]	Condition	Limit (X)	Move Category
1	1	$r_1 / r_3 < X$	6.70	5
2	n	$C_B = X$, $C_C = X$	0.00	1
3	n	$C_B = X$, $C_C > X$	0.00	2
4	n	$C_C > X * C_B$	1.00	8
5	n	$C_B < X$, $C_C < X$	0.50	1
6	n	$C_B < X$, $C_C > X$	0.50	2
7	n	$r_1 / r_2 < X$	0.15	8
8	n	$r_1 / r_2 > X$	5.40	5

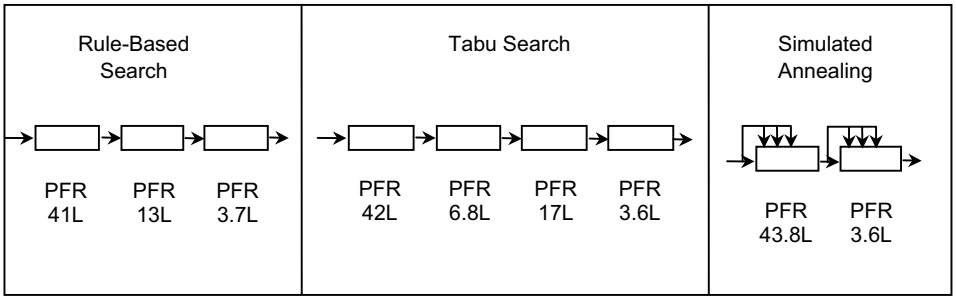

Figure 3. Optimal structures for the nonisothermal Van de Vusse example

Optimisation studies were conducted with a neighbourhood size of 12 and a Tabu List size of 1. The superstructure included up to 4 reactors and PFRs were represented by a cascade of 15 serial equal volume CSTRs. Termination criteria were set at 500 total iterations or 50 unimproving iterations. 10 statistical runs with different initial structures were conducted on 2.4 GHz processors. To monitor the performance of the rule-based algorithm, standard Tabu Searches with random move selection were also conducted.

Comparative results for the different stochastic methods are presented in Figure 3 and Table 5. Figure 3 shows the simple PFR structures of Tabu Search and the rule-based method differ slightly to the Simulated Annealing solution which features side feed streams, and the total volumes differ also. Despite this, Table 5 shows the rule-based search gives solutions of the same high quality as those reported by Mehta and Kokossis (2000), while Tabu Search does not perform as well. The best and average best objectives for Tabu Search are poorer while the function evaluations are higher than the rule based method. Comparing the two temperature profiles illustrated in Figure 4, the rule-based search profile matches more closely the trend reported by Mehta and Kokossis (2000). This indicates that the solution quality is much more dependent on the temperature profiles than reactor size or even mixing strategy.

Table 3. Nonisothermal Van de Vusse results

Nonisothermal Van de Vusse	Statistics[1]	Best Objective	Function Evaluations	Total Volume
Rule-based	average	7.95	1884	51.82L
	maximum	8.13	2580	105.81L
	minimum	7.63	1055	5.42L
Tabu Search	average	7.90	1934	51.88L
	maximum	8.09	3617	84.91L
	minimum	7.60	874	5.34L
Simulated Annealing (Mehta & Kokossis, 2000)[1]	average	7.95	2100	49.9L
	maximum	8.13	-	64.1L
	minimum	-	-	24.6L

[1]Summary statistics based on 10 runs, except Simulated Annealing where literature reported 6 runs

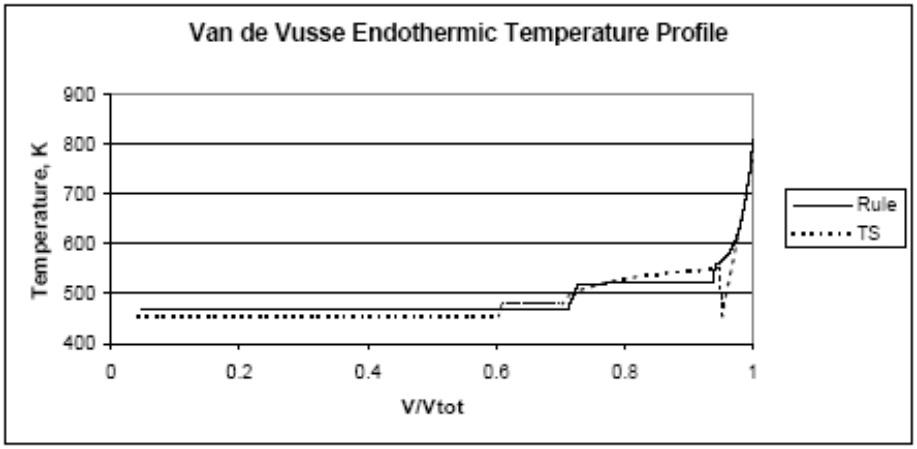

Figure 4. Nonisothermal Van de Vusse temperature profiles for best solution

4. Conclusions

This paper has presented extensions to the knowledge driven reactor network synthesis method to cater for nonisothermal problems and involved applying temperature profiles along the reactors and developing new moves for modifying temperatures. This approach proved successful in finding the optimal results for the nonisothermal Van de Vusse example reported in the literature. The results were superior to Tabu Search in terms of best objective as well as function evaluations, and faster than Simulated Annealing.

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