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A hybrid optimization approach to interaction parameter identification in thermodynamic model problems

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Abstract

The interaction parameter identification problem in thermodynamic models is an important requirement and a common task in many areas of chemical engineering because these models form the basis for synthesis, design, optimization and control of process. For bad starting values the use gradient based result in local optimal solutions. To overcome this drawback, a global optimization approach, Simulated Annealing (SA) and genetic algorithm(GA), has been coupled with a Nelder-Mead Simplex(NMS) method. To improve the accuracy of the interaction parameter estimate. The experimental ternary LLE data for extraction of 1-propanol from water with n-hexane were considered in the NRTL and UNIQUAC activity coefficient model. In conclusion, the different obtained results of the prediction of liquid–liquid equilibrium are compared. These results were obtained to justify that the process of optimization recommended is very practical to estimate the interaction parameters of this ternary system.

Keywords: hybrid optimization approach, genetic algorithm, simulated annealing, parameter estimation;

1. Introduction

The accurate prediction of physical properties and equilibrium conditions of chemical systems can be considered one of the most important applications of thermodynamics in chemical and process engineering[1].

The most common way to face this task is to fit the experimental data to a thermodynamic model and use the obtained model with fitted parameters for predicting properties at other conditions. One of the drawbacks of this approach is that the parameter estimation rarely comprises an optimization problem with unique result (solution), i.e., the optimization problem is frequently a non-convex response surface with several local minima. Liquid–liquid phase equilibrium is a much more severe problem because the excess Gibbs free energy expressions generally used in the thermodynamic modeling are strongly dependent on composition and temperature. Due to this fact, the development of robust strategies for phase equilibrium calculations and parameter optimization that can afford reliable solutions with relatively low computational effort is welcomed.

Modeling liquid phases in equilibrium with activity coefficient models, such as NRTL (Non-Random, Two-Liquids) and UNIQUAC, frequently provides several local minima thus making the parameter estimation step a nontrivial issue [1]. In order to reduce the complexity involved a hybrid algorithm may be required for this purpose.

This paper compares hybrid Genetic Algorithms (HGA) with hybrid Simulated annealing (HSA) that not only is efficient in its search strategy, but also is statistically guaranteed to find the function optima. We first review Genetic Algorithms as evolutionary optimization methods, and illustrate their operation. We next introduce the SA algorithm and we then compare HGA with HSA on a prediction of LLE data for extraction of 1-propanol with n-hexane, the experimental protocol has been previously discussed [2]. Our preliminary results show that the objective function values obtained using hybrid genetic are less than other proposed methods.

2. Parameter estimation algorithm

In the previous work, the optimal values of the interaction parameters are obtained by using deterministic and stochastic methods, the best of these parameters are obtained with genetic algorithm (GA) method. To overcome the drawbacks of GA such as: a long time to acquire the global optimum, prematurity and easily trapping in local optimum of the standard genetic algorithm. A hybrid approach has been used, which couples the SA or GA by Nelder-Mead simplex method. A main advantage of creating a hybrid [3-6] of global optimization with traditional methods is to improve the global search and convergence speeds. The SA or GA procedure is used in the first stage to find a solution within the attraction domain of the global optimum. This solution is then used as starting values for the Nelder-Mead simplex method

2.1. Genetic algorithm (GA)

Genetic algorithm (GA) is a stochastic technique that simulates natural evolution on the solution space of the optimization problems. It operates on a population of potential solutions (i.e., individuals) in each iteration (i.e., generation). By combining some individuals of the current population according to predefined operations, a new population that contains better individuals is produced as the next generation. The first step of GA is to create randomly an initial population of Npop solutions in the feasible region. GA works on this population and combines (crossover) and modifies (mutation) some chromosomes according to specified genetic operations, to generate a new population with better characteristics. Individuals for reproduction are selected based on their objective function values and the Darwinian principle of the survival of the fittest [7]. Genetic operators are used to create new individuals for the next population from those selected individuals of the current population, and they serve as searching mechanisms in GA. In particular, crossover forms two new individuals by first choosing two individuals from the mating pool (containing the selected individuals) and then swapping different parts of genetic information between them. This combining (crossover) operation takes place with a user-defined crossover probability (Pcros) so that some parents remain unchanged even if they are chosen for reproduction. Mutation is a unary operator that creates a new solution by a random change in an individual. It provides a guarantee that the probability of searching any given string will never be zero and acting as a safety net to recover good genetic material which may be lost through the action of selection and crossover. The mutation procedure proceeds with a probability Pmut. Selection, crossover and mutation procedures are recursively used to improve the population and to identify promising areas for optimization. This algorithm terminates when the user-specified criterion is satisfied.
Usually, GA stops after evolving for the specified number of generations (Gmax). The GA subroutine used in this study is from the OptimToolbox of MATLAB®.

2.2. Simulated Annealing

The simulated annealing (SA) algorithm is motivated by an analogy to the statistical mechanics of annealing of solids [8]. The system is said to be in thermal equilibrium at a temperature T if the probability of being in state i with energy Ei is governed by a Boltzman distribution. The annealing process leads to this probability law for energy states. A particular configuration for the annealing of physical systems is analogous to the vector of variables, and the energy is analogous to its objective functional value for minimization problem. SA for continuous variables was originally proposed by Vanderbilt and Louie [9]. Karoonsoontawong and Waller [10] developed the SA based on the work by C. Lee [11] to solve the UODTA-based NDP, and also calibrated the SA parameters for the NDP. In this paper, we employ optimtool for MATLAB as a tool for simulation of this method.

2.3. Hybrid approach

Most successful implementations existing today are actually hybrids of more than one technique. Hybridization basically aims to combine and extend strengths of individual techniques and alleviate their weaknesses. For example, GAs are effective at sampling large areas of the search space, whereas local search heuristics are effective at fine-tuning small areas of the search space, and hence the effectiveness of GAs can often be enhanced by hybridizing with some local search techniques. In fact, most hybrid implementations of stochastic methods involve adding some iterative search techniques such as local search. Local search can also be viewed as a means of integrating problem specific knowledge in the stochastic methods. Today, these hybrids are very common, known under different names: hybrid genetic algorithms, hybrid simulated annealing, hybrid particle swarm, hybrid harmony search, hybrid ant colony...
To ensure an effective and efficient implementation of hybrid Genetic Algorithms (HGA) for example, one should balance several factors. They include the timing of the local search within the algorithm (e.g. after each operator or once after the end of the generation), the frequency of embedding local search (e.g. at each generation or once every few generations), the extent of local search (e.g. until a local optimum is found or for a few iterations only), the number of individuals subjected to local search (e.g. to the entire population or to the best individual only). In addressing these issues, one has to evaluate the benefits of hybridization against any additional expenses incurred, since costs of individual techniques in a hybrid contribute to the total computational cost of the resulting algorithm.

2.4. Genetic hybrid approach

A hybrid algorithm genetic based approach has been used in this study; the genetic approach is an innovative method for solving optimization problems applied to constrained and unconstrained problems [12,13]. The genetic algorithm is based on repeatedly change of a population from individual solutions. At each step, the genetic algorithm selects individuals at random from the current population of species parents and uses them to

produce new generations for the next generation. Over successive generations, the population evolves toward an optimal solution. We can apply the genetic algorithm to solve a variety of complex optimization problems that are not well suited for standard optimization algorithms, including problems in which the objective function is discontinuous, non differentiable, stochastic, or highly non- linear. In each step, genetic algorithm uses three main types of rules (operators) to create the next generation of the population: Selection, Crossover, and Mutation.

The selection is carried out by choosing pairs of individuals surviving from one generation to another and those involved in the reproduction process of the future population. This selection is based on the adaptation of individuals. A certain percentage of the population size is maintained from one generation to another, another percentage, called survival, remains constant over generations. The method of selecting the most traditional, developed by [Goldberg], is based on a random lottery wheel bias. The Goldberg scroll wheel enables the random selecting of individuals having the highest fitness function, without banning the selection of less well-adapted individuals. Indeed, it is important to allow some degree of weak selection of individuals to ensure a degree of genetic diversity within the future generation, to make certain that a good exploration of the solution space should be obtained.

The crossover allows the enrichment of the population by acting on the structure of chromosomes. The crossing is applicable to two individuals drawn randomly from a population above the current population. These two individuals are mated to give birth to two other individuals. Despite the randomness, this exchange of information gives genetic algorithms power in their work: Sometimes 'good' genes from one parent will replace the "bad" genes and create another son better adapted to the environment.

The mutation operator for all these individuals generated in the new population. The mutation provides genetic algorithms property of flexibility in space. This property indicates that the genetic algorithm will be able to reach all points of the space. In the case of binary encoding, the traditional method, after determining the coordinates to mutate, is to reverse a bit in a chromosome.

The hybrid algorithm in this work is a combination between a genetic method and a classical method such as Levenberg-Marquardt, the basic steps of the algorithm can be summarised as follows:

1- Initialization of population of individuals
2- Evaluation of the fitness of each individual in that population
3- Continue repeatedly on this generation until convergence achieved
4- Choose the best-fit individuals for reproduction
5- Mate individuals using crossover and mutation operations to give birth to new individuals
6- Evaluation the fitness function of new individuals
7- Save least-fit population with new individuals as a vector of solution
8- Use the obtained vector by GA as initial vector to be used by Levenberg-Marquardt
9- Calculating new vector by Levenberg-Marquardt method
10- Checking the objective function
11- Repeat calculation until convergence

The purpose of the hybrid method is the benefit of each method, for example the genetic algorithm can be used to get a local minimum and from this value one can use a second method (Nelder-Mead) to ensure that a global minimum is reached. It's to notice that the hybrid function start at the best vector parameter returned by the genetic function. A hybrid function is an optimisation method that start running when genetic function stop in order to improve the fitness function. Several methods displayed in literature can be used as hybrid function; one can cite the simplex search method, pattern search algorithm, large scale-optimisation (trust region approach), and trust-region reflective optimisation. Each hybrid function previously cited can have limitation (facing stabilities problem near local minimum or stopping at this latter) when used alone to calculate the parameters.

2.5. Simulated annealing approach

Simulated annealing is considerably simpler than genetic approaches. The convergence can be guaranteed with suitable values for the initial annealing temperature, the cooling rate and the number of cycles performed.

However, SA has some serious drawbacks when it comes to computational efficiency. As a global solution can only be guaranteed for a small cooling rate the number of function evaluations is very large. Even if the algorithm has reached a near optimal solution, still a large number of iterations are necessary to achieve the optimal solution.

In this study, SA-Hybrid algorithm we choose the same hybrid function as with GA-Hybrid method. The major steps to construct and implement SIMPSA method are:

1) Setting annealing control parameters
2) Constructing initial simplex
3) Running the SA algorithm
4) Checking the convergence
5) Simplex iteration steps
6) Stopping criteria.

3. Results and discussion

The NRTL and UNIQUAC activity coefficient model was used to correlate the experimental results. The NRTL activity coefficient model for component i are expressed as follows [14]:

$$\ln\gamma_i = \frac{\sum_j \tau_{ji} G_{ji} x_j}{\sum_K G_{Ki} x_K} + \sum_j \frac{x_j G_{ij}}{\sum_K G_{Ki} x_K}\left[\tau_{ij} - \frac{\sum_r x_r \tau_{rj} G_{rj}}{\sum_K G_{Kj} x_K}\right] \quad (1)$$

$$G_{ji} = \exp\left(-\alpha_{ji}.\tau_{ji}\right) \quad (2)$$

$$\tau_{ij} = \left(\frac{g_{ji} - g_{ii}}{R.T}\right) = \left(\frac{A_{ij}}{R.T}\right) \quad (3)$$

The NRTL model for a solution with n components is in the following form [15]:

$$\ln\gamma_i = \ln\frac{\phi_i}{x_i} + \frac{z}{2} q_i \ln\frac{\theta_i}{\phi_i} + l_i - \frac{\phi_i}{x_i}\sum_{j=1}^{n} x_j l_j + q_i\left[1 - \ln\left(\sum_{j=1}^{n}\theta_j\tau_{ji}\right) - \sum_{j=1}^{n}\frac{\theta_j\tau_{ij}}{\sum_{k=1}^{n}\theta_k\tau_{kj}}\right] \quad (4)$$

$$\theta_i = \frac{q_i x_i}{q_T} \quad (5)$$

$$q_T = \sum_{k=1}^{n} q_k x_k \quad (6)$$

$$r_T = \sum_{k=1}^{n} r_k x_k \quad (7)$$

$$\phi_i = \frac{r_i x_i}{r_T} \quad (8)$$

$$l_i = \frac{z}{2}\left(r_k - q_k\right) + 1 - r_k \quad (9)$$

$$\tau_{ij} = \exp\left(-\frac{A_{ij}}{T}\right) \quad (10)$$

The values of r and q used for these ternary systems are presented in the following table.

Table 1. The UNIQUAC structural parameters *r* and *q* [available within the HYSYS® data base]

Component	*r*	*q*
Water	0.9200	1.3997
1-propanol	2.7799	2.5120
n-hexane	4.998	3.856

Ternary LLE experimental data were used to determine the optimum NRTL and UNIQUAC binary interactions parameters between water, 1-propanol and n-hexane. The thermodynamics models were fitted to experimental data using an iterative computer program, based on combination of the Newton Raphson method and the hybrid simulated annealing or hybrid genetic algorithm.

Figure 1 shows the value of the best-quality solution in the population as a function of the generation of the GA and HGA algorithms, HGA found the best solutions (the optimal interaction parameters) at least as good as GA.

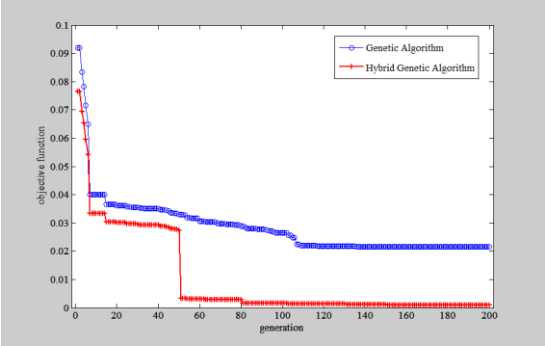

Figure 1. Objective function .vs. of generations.

This figure illustrates how much faster HGA is to converge compared to GA. Moreover, HGA not only found better-quality solutions, but did so in less CPU time.

Figure 2 shows the values of the objective function is almost always less by HSA than the standard SA and the number of function evaluation in standard SA is greater than HAS, also HSA not only found better-quality solutions, but did so in less CPU time.

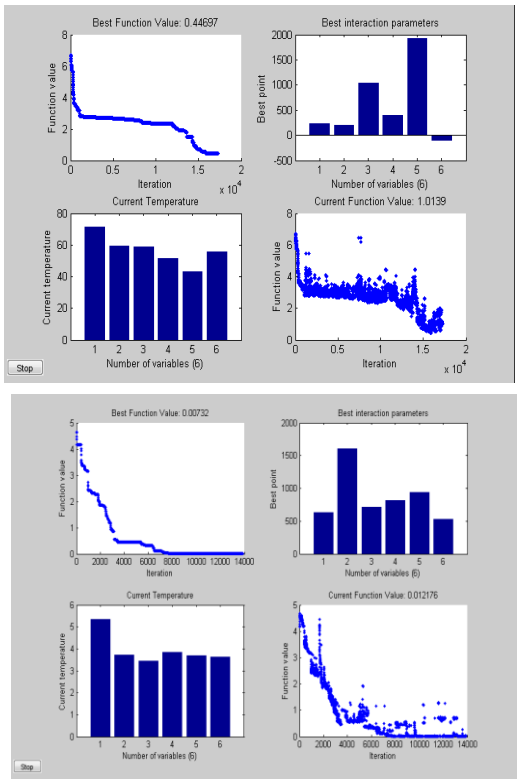

Figure 2. The convergences and the best interaction parameter (for NRTL model) using FS function with T0=100; (a): HAS, (b): SA

The root-mean-square deviation is a measure of the agreement between the experimental data and the calculated values. The RMSD value is defined as follows [16]

$$RMSD = \left[\sum_i \sum_j \sum_k \left(x_{ijk}^{\exp} - x_{ijk}^{cal} \right)^2 / 6.M \right]^{1/2} \quad (11)$$

Where, M is the total number of tie lines, xexp and xcal are the experimental and calculated mass fractions, and the subscripts i, j and k designate the component, phase and tie line respectively.

The table 1, present the RMSD's values for parameters estimated using: GA, SA, NELDER–MEAD ALGORITHM, HGA and HSA. It is seen that the RMSD values obtained using HGA and HSA are less than other methods (making it more reliable for process design and simulation), the optimal values of binary interaction parameters involved in this benchmarking system are shown in Table 2.

Table 1. Comparison of RMSD values with NRTL and UNIQUAC models for water–1-propanol+n-hexane system at 25°C.

Methods	NRTL	UNIQUAC
GA	0.0200	0.0165
SA	0.051	0.068
NM-Simplex	0.1067	0.0919
HGA	0.0081	0.0101
HSA	0.0104	0.0121

Table 2. The NRTL and UNIQUAC binary interaction parameters and RMSD values for water +1-propanol+n-hexane system at 25°C. ternary system obtained with HGA.

model	Interaction parameters (cal/mol)			
	i-j	Aij	Aji	RMSD
NRTL	1-2	-284.5	-450.7	0.0081
	1-3	1185.3	1124.7	
	2-3	68.45	-607.4	
UNIQUAC	1-2	408.1	-185.5	0.0101
	1-3	1253.5	780.2	
	2-3	670.8	-154.9	

4. Comparisons of both algorithms

4.1. Comparison based on the Performance measures

The various RMSD values of GA, SA, NMS, GA-NMS and SA-NMS are given in figures 3-4. These figures present the performance of each algorithm used by giving the iteration number for NRTL and UNIQUAC models in support of water+1-propanol+n-hexane ternary system. It was also found that the RMSD values obtained using HGA and HSA are less than other methods (see figures 3 and 4).

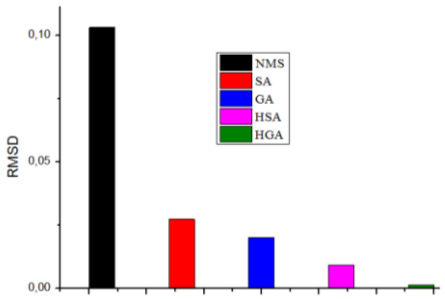

Figure 3. Comparison of RMSD values for several algorithms using NRTL model for water +1-propanol+n-hexane system at 25°C.

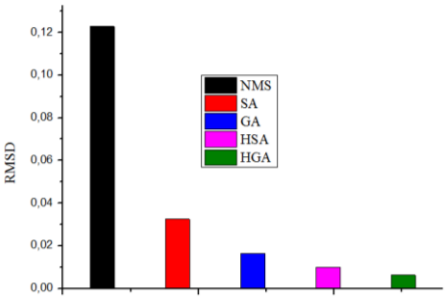

Figure 4. Comparison of RMSD values for several algorithms using UNIQUAC model for water +1-propanol+n-hexane system at 25°C.

4.2. Running time:

Comparing execution times between the GA, SA, NMS, HGA and HSA algorithms, it is evident to see that the NMS, SA and HSA algorithms requires the way less time to found the optimum (see figures 5-6). These methods (NMS, SA and HSA) can be considered the most effective algorithm in the term of running time.

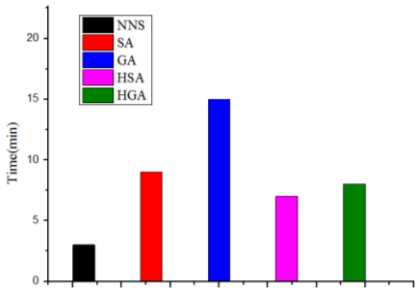

Figure 5. Comparison of optimization methods in terms of time taken (running time) using NRTL model for water +1-propanol+n-hexane system at 25°C.

In summary, all the algorithms are feasible for phase equilibria calculation and interaction parameters estimation problems. On the other hand, it can be concluded that the HGA and HSA algorithms can outperform the other algorithms in terms of performance measures (RMSD values). However, in terms of the running time to achieve the optimum the NMS, HGA and HSA was better than other algorithms.

5. Conclusion

A hybrid optimization approach has been used for estimating interaction parameters of activity coefficient model such as NRTL and UNIQUAC in liquid-liquid equilibrium problem. A stochastic global optimization approach, genetic algorithm and Simulated Annealing, has been used to find interaction parameter values in the attraction domain of the global optimum. The global approach has been coupled with NELDER–MEAD – Simplex method to decrease the objective function values toward the end of the optimization procedure. All the algorithms used in this work are suitable for LLE calculations. The maximum calculated RMSD between the experimental and calculated mass fractions was 0.0121.

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