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Estimation of heat transfer parameters in a trickle-bed reactor using differential evolution and orthogonal collocation

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Abstract

A new non-sequential technique is proposed for the estimation of *effective* heat transfer parameters using radial temperature profile measurements in a gas–liquid co-current downflow through packed bed reactors (often referred to as trickle bed reactors). Orthogonal collocation method combined with a new optimization technique, differential evolution (DE) is employed for estimation. DE is an exceptionally simple, fast and robust, population based search algorithm that is able to locate near-optimal solutions to difficult problems. The results obtained from this new technique are compared with that of radial temperature profile (RTP) method. Results indicate that orthogonal collocation augmented with DE offer a powerful alternative to other methods reported in the literature. The proposed technique takes less computational time to converge when compared to the existing techniques without compromising with the accuracy of the parameter estimates. This new technique takes on an average 10 s on a 90 MHz Pentium processor as compared to 30 s by the RTP method. This new technique also assures of convergence from any starting point and requires less number of function evaluations.

Keywords: Trickle-bed reactor; Heat transfer parameters; Differential evolution; Orthogonal collocation; Radial temperature profile method; Two-dimensional pseudo-homogeneous model

1. Introduction

Trickle-bed reactors are widely used in petroleum and petro-chemical industries, and to a lesser extent in chemical and pharmaceutical industries. They also have a potential application in waste water treatment and in bio-chemical reactions. Various flow regimes such as trickle flow (at low liquid and low gas rates), pulse flow (intermediate gas and liquid rates), dispersed bubble flow (at high liquid and low gas rates), and spray flow (at low liquid and high gas rates) are encountered in a trickle-bed reactor depending upon the flowrates and physical properties of flowing phases and the packing geometry. The most common mathematical model for the non-adiabatic trickle-bed catalytic reactor is the two-dimensional pseudo-homogeneous model (Tsang et al., 1976; Babu, 1993), which consists of coupled partial differential equations of the parabolic type. The inherent characteristic of the homogeneous model is that the system can be considered as a continuum; no distinction is made between the solid phase and the fluid phase. The assumption implies that the reactant and the product concentrations in the bulk fluid phase are same as that on the surface of the catalyst pellet. A similar implication holds for the bed temperature. The homogeneous model that describes the physical and chemical processes is as follows:

Mass transfer

$$u\frac{\partial C_A}{\partial z}+\rho_B r_A=\varepsilon D_{er}\left(\frac{\partial^2 C_A}{\partial r^2}+\frac{1}{r}\frac{\partial C_A}{\partial r}\right). \tag{1}$$

Heat transfer

$$u\rho_f C_p\frac{\partial T}{\partial z}+\rho_B(-\Delta H)r_A=k_{er}\left(\frac{\partial^2 T}{\partial r^2}+\frac{1}{r}\frac{\partial T}{\partial r}\right). \tag{2}$$

with boundary conditions

$$z=0,\quad 0\leqslant r\leqslant R\quad C_A=C_o,\ T=T_o$$

$$r=0,\quad 0\leqslant z\leqslant Z,\quad \frac{\partial C_A}{\partial r}=\frac{\partial T}{\partial r}=0,$$

$$r=R,\quad 0\leqslant z\leqslant Z,\quad \frac{\partial C_A}{\partial r}=0,\ -k_{er}\frac{\partial T}{\partial r}=h_w(T-T_w)$$

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In the typical operation of a trickle-bed reactor the heat transfer parameters, the effective radial thermal conductivity of the bed, k_{er}, and the effective wall-to-bed heat transfer coefficient, h_w, are unknown and need to be estimated. These parameters are extremely important in design and in process analysis. Once these parameters are estimated, the temperature profile can be generated numerically. The temperature profile in the reactor is important because it affects the selectivity, the yield and the stability of the reactor.

Froment (1967) has demonstrated the sensitivity of the homogeneous model to the effective parameters. He concluded that changing the effective Peclet number of mass transfer had a negligible effect on the temperature and conversion, whereas a 10% increase either in k_{er} or h_w greatly changed the temperature and conversion profiles in the reactor. Smith (1973) has analyzed the relative importance of the heat and mass transfer effects in the fixed-bed reactor and concluded that the radial temperature gradient is the most important heat transfer characteristic. Only a few studies have been reported on the heat transfer characteristics (Weekman and Myers, 1965; Hashimoto et al., 1976; Muroyama et al., 1978; Matsuura et al., 1979a; b; Specchia and Baldi, 1979; Crine, 1982; Lamine et al., 1996; Babu and Rao, 1997), which are essential for the proper design of a trickle-bed reactor, using only air–water and air–glycerol systems; although a lot of information is available on hydrodynamics and mass transfer for the same. Deviations of 30–40% were reported for the prediction of h_w with their own empirical correlations by different authors even for their own data. Moreover, the effect of gas rate on k_{er} and h_w is not well understood with respect to the flow regimes encountered in two-phase flow, especially in the pulse flow with packing geometry. Further, the pulse properties especially liquid holdup and pulse frequency could play an important role on the heat transfer parameters in pulse flow, and none of the earlier studies except Babu (1993) detailed their effects on heat transfer. These factors emphasize the importance of heat transfer and suggest the further need for study of the heat transfer phenomena.

The study of heat transfer characteristics in a trickle bed can be simplified by testing the system with no reaction occurring. In this case only the heat transfer balance equation without the reaction term is needed, i.e.,

$$(LC_{pL} + GC^*_{pG})\frac{\partial T}{\partial z} = k_{er}\left(\frac{\partial^2 T}{\partial r^2} + \frac{1}{r}\frac{\partial T}{\partial r}\right) \tag{3}$$

with boundary conditions

$$z = 0, \quad T = T_o, \tag{4}$$

$$r = 0, \quad \frac{\partial T}{\partial r} = 0, \tag{5}$$

$$r = R, \quad -k_{er}\frac{\partial T}{\partial r} = h_w(T - T_w). \tag{6}$$

Various methods can be used to integrate Eq. (3) subject to the boundary conditions. Among the most popular are analytical solution, finite difference techniques and the method of weighted residuals, the last two being numerical methods. Virtually all of the previous work on estimating the *effective* parameters has been based on the analytical solution, which is

$$\frac{T(r,z) - T_w}{T_o - T_w} = 2\sum_{i=1}^{\infty}\frac{BiJ_o(b_i^r{}_R)\exp(-k_{er}b_i^2 z/(LC_{pL} + GC^*_{pG})R^2)}{(Bi^2 + b_i^2)J_o(b_i)}, \tag{7}$$

where

$$Bi = \frac{h_w R}{k_{er}} = b_i\frac{J_1(b_i)}{J_o(b_i)}. \tag{8}$$

Since the eigenvalues b_i increase as i increases, there are certain ranges of parameters within which only the first few terms of the infinite series in Eqs. (7) and (8) is significant (Coberly and Marshall, 1951; Tsang et al., 1976; Specchia and Baldi, 1979). Most of the previous studies, except Specchia and Baldi (1979) used graphical methods for estimating k_{er} and h_w considering only the first term of the infinite series in the analytical solution (Eq. (7)) of two-dimensional energy equation for simplicity leading to uncertainty in the estimated values. Babu (1993) concluded that the first seven terms of the infinite series would be sufficient for ensuring good convergence. Differing numbers and locations of temperature measurements made on the packed bed system have led to several types of parameter estimation methods using the analytical solution. The relative advantages and disadvantages have been accounted by Tsang et al. (1976).

Tsang et al. (1976) proposed a technique using orthogonal collocation in an inverse problem. They used both gradient and gradient-free optimization schemes for parameter estimation. They showed that the results were accurate enough and the computational time required was less. They used Graeffe's method along with Newton's method for finding out the roots of the Jacobi polynomial. But the Newton's method is highly dependent on the initial guess, is less accurate and takes more computational time (Acton, 1970). Graeffe's method involves in squaring the polynomial and then taking the square root of the solution, which reduces the accuracy as the degree of the polynomial increases (Acton, 1970). It has been shown that the objective function is very flat near the minimum or the contours are long and narrow (Tsang et al., 1976). For such kinds of problems the gradient-based optimization techniques fail and computational time taken is also large (Acton, 1970). Though gradient-free optimization solves some of the problems of the gradient methods, the global optimum is not assured and the computational time taken is still large. Thus, to

develop a new technique for the estimation of the heat transfer parameters in a trickle bed, which is not only accurate but also guarantees faster convergence is the main objective of our study.

The results obtained from DE in the present study are compared with those obtained from the radial temperature profile method employing Powell's method for optimization. This new technique is highly robust and is very fast in terms of computation time when compared to the radial temperature profile method. The results show that the Orthogonal Collocation augmented with the DE's offer a powerful alternative to the conventional estimation techniques while demonstrating the potential of Orthogonal Collation for solving boundary value problems.

2. Orthogonal collocation

The trickle-bed reactor model is not amenable to an analytical solution when the chemical reaction term is non-zero. In this case, a numerical integration method such as a finite difference technique must be used. However, the orthogonal collocation method (Villadsen and Stewart, 1967; Villadsen and Michelsen, 1978; Finlayson, 1972, 1980), a technique categorized as a method of weighted residuals, has been shown by Ferguson and Finlayson (1970), and Finlayson (1972, 1980) to be superior in some respects to the finite difference approaches. Furthermore, the orthogonal collocation method, besides obtaining the solution, gives the possibility of exploring the local stability within the system (Perlmutter, 1972; Bosch and Padmanabhan, 1974; Sorenson et al., 1973). Young and Finlayson (1973) have used the orthogonal collocation technique to approximate the boundary condition at the entrance of a reactor and solved the coupled non-linear partial differential equations which take both the axial and radial dispersions into account. Finlayson (1971) has also shown when the two-dimensional reactor model must be used instead of one-dimensional model. Karanth and Hughes (1974) used collocation to simulate the adiabatic packed-bed reactor. Carey and Finlayson (1975) combined the orthogonal collocation method with finite element method to solve the catalyst pellet problem with large Thiele modulus.

The orthogonal collocation method has been used in the above studies for the simulation, i.e., as a numerical method to solve the boundary value problems. Tsang et al., (1976) used the orthogonal collocation method in an inverse problem for the estimation of the heat transfer parameters in a packed-bed reactor. Bosch and Hellinckx (1974) used Lobatto quadrature combined with the collocation method to estimate the parameters in the differential equations. This yielded a non-linear programming problem. Polis et al. (1973) have used the Galerkin technique to estimate the parameters in distributed systems. The Galerkin method is also classified as one of the methods of weighted residuals which can be used to reduce the partial differential equation to a set of ordinary differential equations. The set of ordinary differential equations can then be used to simulate the system response iteratively.

However unlike all other weighted residual methods which determine the undetermined coefficients of the trial function, the orthogonal collocation method gives the solution of the dependent variables at the collocation points directly. One can select the measurement locations to coincide with the collocation points. The various problems in estimating the parameters in a distributed system from the point of view of accurate parameter estimates have been discussed by Goodson and Polis (1974). The details of the use of the orthogonal collocation to estimate k_{er} and h_w are given below.

By rewriting Eqs. (3)–(6) in dimensionless form, the parameter estimation problem becomes
Minimizing

$$\mathscr{F} = \varepsilon^{\mathrm{T}} Q \varepsilon \tag{9}$$

subject to

$$\frac{\partial \tilde{T}}{\partial \tilde{z}} = P'_{er} \frac{1}{\tilde{r}} \frac{\partial}{\partial \tilde{r}} \left(\tilde{r} \frac{\partial \tilde{T}}{\partial \tilde{r}} \right), \tag{10}$$

$$\tilde{z} = 0, \quad \tilde{T} = \tilde{T}_d, \tag{11}$$

$$\tilde{\mathrm{r}} = 0, \quad \frac{\partial \tilde{T}}{\partial \tilde{r}} = 0, \tag{12}$$

$$\tilde{\mathrm{r}} = 1, \quad -\frac{\partial \tilde{T}}{\partial \tilde{r}} = Bi\tilde{T}, \tag{13}$$

where $\tilde{T} = T - T_w$, the modified Peclet number $P'_{er} = k_{er}Z/(GC^*_{pG} + LC_{pL})R^2$, $\tilde{r} = r/R$, $\tilde{z} = z/Z$ and ε is defined as an $N \times 1$ column vector:

$$\varepsilon = \frac{(\tilde{T}_{\mathrm{exp}}(r_i, Z) - \tilde{T}_{\mathrm{cal}}(r_i, Z))}{\tilde{T}_d},$$

N is the number of collocation or measurement points. Q is a $N \times N$ positive-definite weighting matrix, the identity matrix being used in the present study. The estimation criterion used, when Q is taken to be an identity matrix reduces from weighted least squares to a simple least-squares estimation. A detailed derivation of the solution of Eqs. (10)–(13) using orthogonal collocation is given in Appendix A. Using orthogonal collocation method, the estimation problem reduces to

Minimizing Eq. (9) subject to

$$\frac{\mathrm{d}\tilde{\mathbf{T}}}{\mathrm{d}\tilde{z}} = P'_{er} \left(\mathbf{B} - \frac{\mathscr{B}(\mathscr{A})^{\mathrm{T}}}{Bi + A_{N+1,N+1}} \right) \tilde{\mathbf{T}}, \tag{14}$$

$$\tilde{\mathbf{T}}(0) = \tilde{\mathbf{T}}_d. \tag{15}$$

where **B**, $\mathscr{B}$ and $\mathscr{A}$ are the computational collocation matrix and vectors given by Villadsen and Stewart (1967). However, instead of using Graeffe's method and Newton's method, in the present study, the *Laguerre's* method (Ralston and Rabinowitz, 1978) for finding the roots of the Jacobi polynomial has been used. Laguerre's method is guaranteed to converge to all types of roots: real, complex, single or multiple from any starting point (Acton, 1970; Press et al., 1996). We used *LU decomposition* and *LU back-substitution* for finding out the inverse of a matrix required in calculating the collocation matrices (Villadsen and Stewart, 1967). The integration of the ordinary differential equation, Eq. (14), was computed using the fifth-order Runge–Kutta method with adaptive step size control (Cash and Karp, 1990; Press et al., 1996) for greater accuracy.

3. Differential evolution

Since their inception three decades ago, Genetic Algorithms (GA) have evolved like the species they try to mimic (Goldberg, 1989). Just as competition drives each species to adapt to a particular environmental niche, so too, has the pressure to find efficient solutions across the spectrum of real-world problems forced genetic algorithms to diversify and specialize (Davis, 1991; Moros et al., 1996; Wolf and Moros, 1997; Chakraborti and Sastry, 1997, 1998). Differential Evolution (Price and Storn, 1997) is a search procedure similar to a GA applied on real variables which is significantly fast at numerical optimization and is also more likely to find a function's true global optimum. DE is in similar to a real coded GA combined with an adaptive random search (ARS) (Boender and Romeijn, 1995; Maria, 1998) with a normal random generator. Among DE's advantages are its simple structure, ease of use, speed and robustness. DE has been used to design several complex digital filters (Price and Storn, 1997) and to design fuzzy logic controllers (Sastry et al., 1998). DE has been used in the previous works for design and control application. In this paper, DE is used for estimation purpose.

Upreti and Deb (1996) used GAs to optimize the length of ammonia reactor by solving coupled differential equations. They used binary strings to code the parameters; however this choice limits the resolution with which an optimum can be located to the precision set by the number of bits in the integer (Davis, 1991; Price and Storn, 1997; Wolf and Moros, 1997). Wolf and Moros (1997) encoded a floating point codes into mantissa, exponent and sign of exponent; however the string encoding can be completely circumvented by using the floating point codes di

codi
these-551.3(1997mutns.)--3(In)-285.31ofisis5-532(416(-3910.7(numbr-657.30be1s)-3n2T [(the)-2ough7.8(use)-383.55can)mutns.

Recombination (crossover) provides an alternative and complementary means of creating viable vectors from the components of existing vectors. Previous studies on GA for solving PDEs (Upreti and Deb, 1996; Wolf and Moros, 1997) used a uniform crossover, however, a DE uses a nonuniform crossover that can take child vector parameters from one parent more often than it does from the other. In order to increase the diversity of the parameter vectors, the vector $\mathbf{u}_{i,\Gamma}$ with

$$(\mathbf{u}_{i,\Gamma})_j = \begin{cases} v_j & \text{for } j = \langle n \rangle_D, \langle n+1 \rangle_D, \ldots, \langle n+M-1 \rangle_D, \\ (x_{i,\Gamma})_j & \text{otherwise} \end{cases} \tag{19}$$

is formed where the acute brackets $\langle\ \rangle_D$ denote the modulo function with modulus number equal to dimension D. A certain sequence of the vector elements of $\mathbf{u}$ is identical to the elements of $\mathbf{v}$, the other elements of $\mathbf{u}$ acquire the original values of $\mathbf{x}_{i,\Gamma}$. Choosing a subgroup of parameters for mutation is similar to a process known as *crossover* in evolution theory. The integer M is drawn from the interval $[0,\ D-1]$ with the probability $Pr(M=\gamma) = (CR)^{\gamma}$, $CR \in [0, 1]$ is the crossover probability and constitutes a control variable for the above-mentioned scheme. The random decisions for both n and M are made anew for each trial vector $\mathbf{v}$.

Unlike many GAs, DE does not use proportional selection, ranking or even an annealing criterion that would allow occasional uphill moves. Instead the cost of each trial vector is compared to that of its parent target vector. The vector with the lower cost is rewarded by being selected to the next generation. This selection of the individuals to the next generation resembles *tournament selection* except that each child that is pitted against one of its parents, not against a randomly chosen competitor. If the resulting child vector yields a lower objective function value than a predetermined population member, the child vector replaces the parent vector with which it was compared. The comparison vector can, but need not, be a part of the generation process mentioned above. In addition, the best parameter vector $\mathbf{x}_{\text{best},\Gamma}$ is evaluated for every generation Γ in order to keep track of the progress that is made during the minimization process.

4. Experimental setup and procedure

Experiments were carried out to obtain the data on radial temperature profile in a trickle-bed reactor (gas–liquid co-current downflow through packed beds). The schematic diagram of the setup is shown in Fig. 1. The experimental setup mainly consists of a packed-bed column of 50 mm diameter, comprising of air–liquid

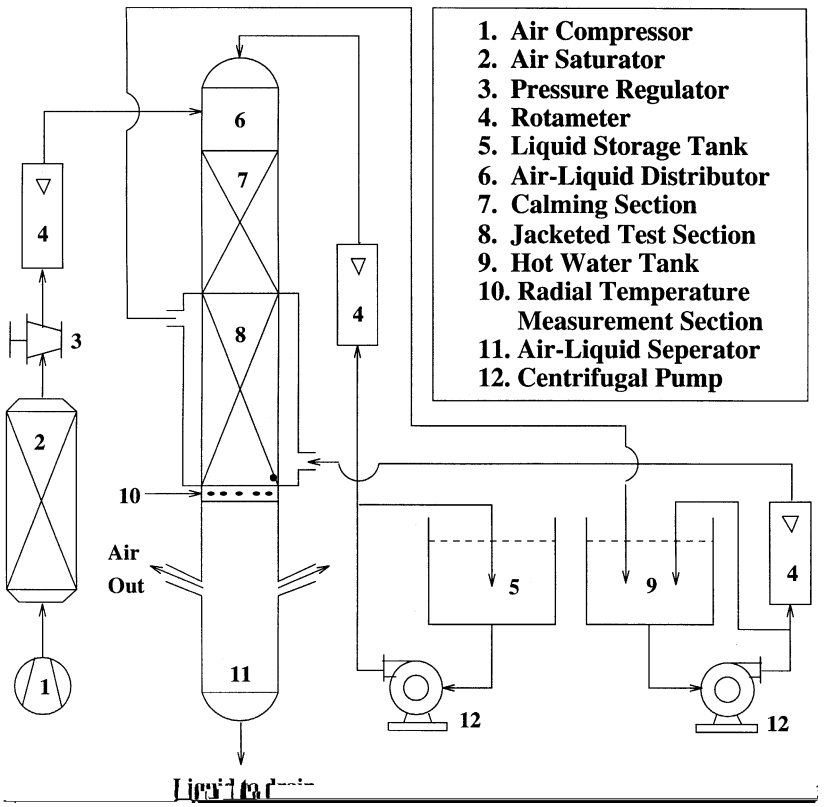

Fig. 1. Schematic diagram of the experimental setup.

distributor, calming section, jacketed test section and air–liquid separator with other auxiliary parts. Air was drawn from 3.7 kW double piston–double action compressor, of maximum volumetric capacity of 14.98 m^3/s at STP and a working pressure of 12 atm. The air drawn from the compressor was saturated with water in a saturater. The saturated air was introduced at the top of the column through a set of pre-calibrated rotameters to the air–liquid distributor at a constant pressure of 4 atm, monitored by a pneumatic pressure regulator. The air flowrates in the rotameter were controlled by needle valves. The air was passed through a filter before entering the distributor to remove traces of oil and dust, if any. Water was pumped through a 4.5 kW pump and was metered through a set of pre-calibrated rotameters to the air–liquid distributor at the top of the column. Water flowrates to the column were controlled by means of globe valves. Air and water were uniformly distributed through an air–liquid distributor at the top of the calming section. The air–liquid distributor essentially consists of two sets of openings, 21 copper tubes for distributing liquid and 16 nozzles for distributing air. The tubes and the nozzles were alternately arranged on a triangular pitch over the column cross-section. The number of liquid distribution tubes per unit area was approximately equal over the entire cross-section to ensure equal distribution of liquid. The calming section consisted of a long tube, which ensured a fully developed equilibrium gas–liquid flow before it entered the test section.

The jacketed test section was designed for heat transfer studies. It consists of a jacket in order to circulate hot water at 60°C. Hot water was pumped with a 4.5 kW and metered through a set of pre-calibrated rotameters. Below the heat transfer section radial temperature profile measurement section was provided. An air–liquid separator was provided at the bottom of the column to separate the air and the liquid phases coming out of the test section. The radial temperature profile was obtained by measuring the temperatures at the bottom of the test section at three radial positions at $\tilde{r} = 0.0$, 0.4, and 0.8, and at three symmetric angular positions (120° apart) for each radial position. Wall temperature was measured by installing a thermocouple at the inside wall 3 mm above from bottom of the jacketed test section. Thermocouples were also installed at various locations to measure the inlet temperature of test liquid, and inlet and outlet temperatures of hot water. An INSREF constant temperature bath having an accuracy of 0.01°C and a MINCO platinum resistance thermometer bridge (MINCO RTB8078, Model No. S7929 Pail120C) with an accuracy of 0.025°C as a standard thermometer were used for calibrating all the chormel–alumel thermocouples used in the present study. All these thermocouples were connected to an APTEK multi-channel digital temperature scanner for recording the temperatures. The detailed description of the experimental set-up, and the data collection and reduction procedures are reported elsewhere (Babu, 1993; Babu and Rao, 1994, 1997).

Air and water were fed to the column from the top at the desired flowrates by means of pre-calibrated rotameters. Hot water was circulated through the jacket around the test section at sufficiently high flowrates (25–30 l per minute) in order to maintain nearly constant wall temperature, and the minimum and maximum temperature difference between the inlet and the outlet hot water streams were 0.3°C at low flowrates to 2°C at high flowrates respectively of the flowing fluids. In general, it took 20–40 min for attaining the steady state. After steady state was attained, which was confirmed from the constant values of flowrates and temperatures, the flowrates of air and water and the temperatures were recorded. The average of the three angular positions was taken as the temperature at each radial position. This procedure was repeated for a wide range of air (0.01–0.898 $kg/m^2 s$) and water flowrates (3.16–71.05 $kg/m^2 s$), covering trickle, pulse and dispersed bubble flow regimes. The length of the heat transfer test section used for heat transfer experiments was 0.715 m. The packing employed were 2.59 mm ceramic spheres, 4.05 and 6.75 mm glass spheres and 4.0 and 6.75 mm ceramic raschig rings.

5. Results and discussions

The psuedocode of the DE algorithm used in the present study is shown below:

- Initialize the values of D, NP, CR, F, λ and maximum number of generations MaxGen.
- Initialize all the vectors of the population randomly between a given lower bound LB, and
  upperbound UB
  for i = 1 to NP
    for j = 1 to D
      $(\mathbf{x}_{i,0})_j$ = LB + RandomNumber × (UB - LB)
- Evaluate the cost of each vector. Cost here is the value of the objective function to be minimized.
  for i = 1 to NP
    $\mathscr{F}_i = \varepsilon^2|_{\mathbf{x}_{i,0}}$
- Find out the vector with lowest cost i.e., the best vector so far
  $\mathscr{F}_{min} = \mathscr{F}_1$ and best = 1
    for i = 2 to NP
    if ($\mathscr{F}_i < \mathscr{F}_{min}$)
  then $\mathscr{F}_{min} = \mathscr{F}_i$ and best = i
- While the current generation is less than the maximum number of generations perform recombination, mutation, reproduction and evaluation of the objective function. while ($\Gamma <$ MaxGen) do {

for i = 1 to NP {
- Select two distinct vectors randomly from the population other than the vector $\mathbf{x}_{i,\Gamma}$
  do r_1 = RandomNumber × NP while(r_1 = i)
  do r_2 = RandomNumber × NP while((r_2 = i) OR ($r_2 = r_1$))
- Perform D binomial trails, change at least one parameter of the trial vector $\mathbf{u}_{i,\Gamma}$ and perform mutation.
  j = RandomNumber × D
  for $n = 1$ to D {
    if ((RandomNumber < CR) OR (n = (D-1)))
      then $(\mathbf{u}_{i,\Gamma})_j = (\mathbf{x}_{i,\Gamma})_j + \lambda * ((\mathbf{x}_{\text{best},\Gamma})_j - (\mathbf{x}_{i,\Gamma})_j) + F * ((\mathbf{x}_{r_1,\Gamma})_j - (\mathbf{x}_{r_2,\Gamma})_j)$
    else $(\mathbf{u}_{i,\Gamma})_j = (\mathbf{x}_{i,\Gamma})_j$
    $j = \langle n+1 \rangle_D$ }
- Evaluate the cost of the trial vector.
  $\mathscr{F}_{trial} = \varepsilon^2|_{\mathbf{u}_{i,\Gamma}}$
- If the cost of the trial vector is less than the parent vector then select the trial vector to the next generation.
  if ($\mathscr{F}_{trial} \leqslant \mathscr{F}_i$) {
    $\mathscr{F}_i = \mathscr{F}_{trial}$
    if ($\mathscr{F}_{trial} < \mathscr{F}_{min}$)
      $\mathscr{F}_{min} = \mathscr{F}_{trial}$ and best = i } } /* for $i = 1$ to NP ends */
- Copy the new vectors $\mathbf{u}_{i,\Gamma}$ to $\mathbf{x}_{i,\Gamma}$ and increment Gamma
  $\Gamma = \Gamma + 1$
- Check for convergence and break if converged. } /* while $\Gamma \cdots$ ends. */

• Print the results.

The collocation points or the measurement points were chosen to be that of the radial temperature profile measurements, i.e., $r/R = 0, 0.4, 0.8$. The values of NP, CR, λ and F are fixed empirically following certain heuristics (Price and Storn, 1997; Sastry et al., 1998): (1) F and λ are usually equal and are between 0.5 and 1.0, (2) CR usually should be 0.3, 0.7, 0.9 or 1.0 to start with, (3) NP should be of the order of $10 \times D$ and should be increased in case of misconvergence, and (4) if NP is increased then usually F has to be decreased. In the present study, the values of D, NP, F, λ and CR were taken as 2, 20, 0.7, 0.7 and 0.9, respectively. The maximum number of iterations was kept as 100; however, in all the runs the algorithm converged within 15 generations. The initial values of Bi and P'_{er} were generated using Knuth's uniform random variate (Press et al., 1996). The matrix inversions involved in computing the collocation matrices were achieved using LU decomposition and LU back-substitution. Runge–Kutta method with adaptive step size control was used for integrating the differential equation (Eq. (14)).

Altogether 232 experimental data points were obtained covering a wide range of liquid and gas flowrates using five packings of different size and shape. The DE algorithm in conjunction with orthogonal collocation method was employed using the experimental temperature profile obtained for all the 232 data points. The typical radial temperature profile given by DE and RTP methods is shown in Table 1which shows that the temperature profiles generated by both the methods are almost similar to the experimental profile. Similar trends were observed for all data points. The minimum and maximum sum square error for DE being 1.905052×10^{-6} and 7.57026×10^{-4}, respectively, and that for RTP being 1.905064×10^{-6} and 7.757032×10^{-4}, respectively with the present experimental data. The close agreement with the analytical solution and the experimental value shows that non-optimal selection of collocation points, which are taken to be the measurement points and not the roots of the Jacobi polynomial, does not cause significant errors. The estimation errors (sum square error) given in Table 2 indicate that the temperature profile by DE is slightly better than that with RTP (DE's sum square error is 0.0001–0.001% less than RTP). Computational time taken by DE and RTP algorithm for randomly selected sample data points is compared in Fig. 2. Although the

Table 1
Radial temperature profile generation: DE vs RTP

$G = 0.0107$, $G^* = 8952.44$, $\frac{LC_{pL}}{GC^*_{pG}} = 137.8$, $L = 3.16$, $D_e = 8.1$, TF			$G = 0.2041$, $G^*_{pG} = 6809.25$, $\frac{LC_{pL}}{GC^*_{pG}} = 165.01$, $L = 54.89$, $D_e = 4.89$, DBF			$G = 0.5$, $G^*_{pG} = 9447.63$, $\frac{LC_{pL}}{GC^*_{pG}} = 2.79$, $L = 11.11$, $D_e = 2.59$, PF		
EXP(°C)	DE(°C)	RTP(°C)	EXP(°C)	DE(°C)	RTP(°C)	EXP(°C)	DE(°C)	RTP(°C)
56.48	56.605865	56.605843	38.00	38.075127	38.074937	57.30	57.276149	57.276099
56.95	56.776015	56.775936	38.98	38.876623	38.876634	57.46	57.493691	57.493661
57.20	57.248915	57.248689	41.09	41.118606	41.119144	58.08	58.069785	58.069799
58.91	58.91	58.91	49.85	49.85	49.85	59.12	59.12	59.12

The unit of D_e is mm.

Table 2
Estimation characteristics: DE vs RTP

Sum square error		h_w		k_{er}		Parameters			
DE	RTP	DE	RTP	DE	RTP	L	G	D_e	Flow
$4.637816e^{-5}$	$4.637820e^{-5}$	894.626343	894.511169	17.688799	17.693483	3.16	0.017	8.1	TF
$2.333386e^{-4}$	$2.333388e^{-4}$	2508.752686	2507.566406	27.927656	27.941372	9.57	0.2401	8.1	PF
$1.764492e^{-4}$	$1.764498e^{-4}$	3195.485596	3195.852539	53.098129	53.086544	19.83	0.5000	8.1	PF
$2.432793e^{-4}$	$2.432798e^{-4}$	3999.604736	3999.616943	63.374241	63.372269	18.38	0.8980	8.1	PF
$1.568811e^{-4}$	$1.568813e^{-4}$	1645.458130	1645.295044	26.286270	26.290203	6.52	0.0459	8.1	TF
$3.369437e^{-5}$	$3.369470e^{-5}$	4505.336914	4505.336914	99.590958	99.575050	54.89	0.2401	4.89	DBF
$1.905052e^{-6}$	$1.905064e^{-6}$	1761.488281	1761.581177	16.218536	16.217493	3.16	0.5000	4.89	TF
$1.340671e^{-4}$	$1.340675e^{-4}$	4035.566406	4035.837646	65.492363	65.486969	36.87	0.0459	4.89	DBF
$6.927241e^{-4}$	$6.927244e^{-4}$	9719.313477	9718.518555	41.572712	41.573727	45.31	0.1020	2.59	DBF
$7.757026e^{-4}$	$7.757032e^{-4}$	11872.214844	11868.007812	58.668274	58.678162	60.18	0.0459	2.59	DBF

The unit of D_e is mm.

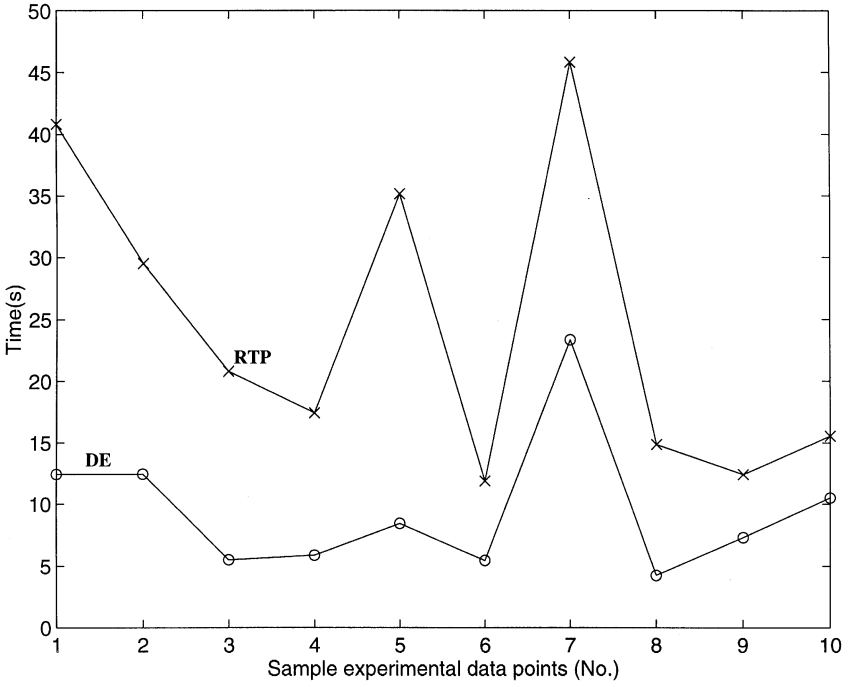

Fig. 2. Computational time: DE vs RTP.

qualitative trends by both the methods is more or less the same, the average time taken, based on all data points, for an estimation by DE is 10 s as compared to 30 s by RTP on a 90 MHz Pentium processor. The function evaluations computed for sample data points by RTP and DE is shown in Fig. 3. DE also takes less number of function evaluations as compared to RTP (DE takes average of 800 function calls as compared to 2000 evaluations by RTP). The estimation of values of h_w and k_{er} using both DE algorithm and RTP methods, are compared in Figs. 4 and 5, respectively, which shows that the estimation by DE is as accurate as the well-proven RTP algorithm. The estimations are also tabulated in Table 2 which show that the estimation error of DE is lower when compared to that of RTP algorithm. It also indicates that the estimation accuracy depends on the accuracy of the measured temperature profile which, in the present study is accurate only to two decimal places. Since a wide range of air and water flowrates was used covering the trickle, pulse and dispersed bubble flow for generalization of the estimation algorithm, the estimated parameters cover a wide range (894–11872 W/m^3 K for h_w and 16-99 W/mK for k_{er}).

The convergence criterion used for RTP is very stringent: the objective function, its relative change, the parameter values and their relative changes and the gradient of the objective function were all checked before the optimization scheme was terminated. The RTP

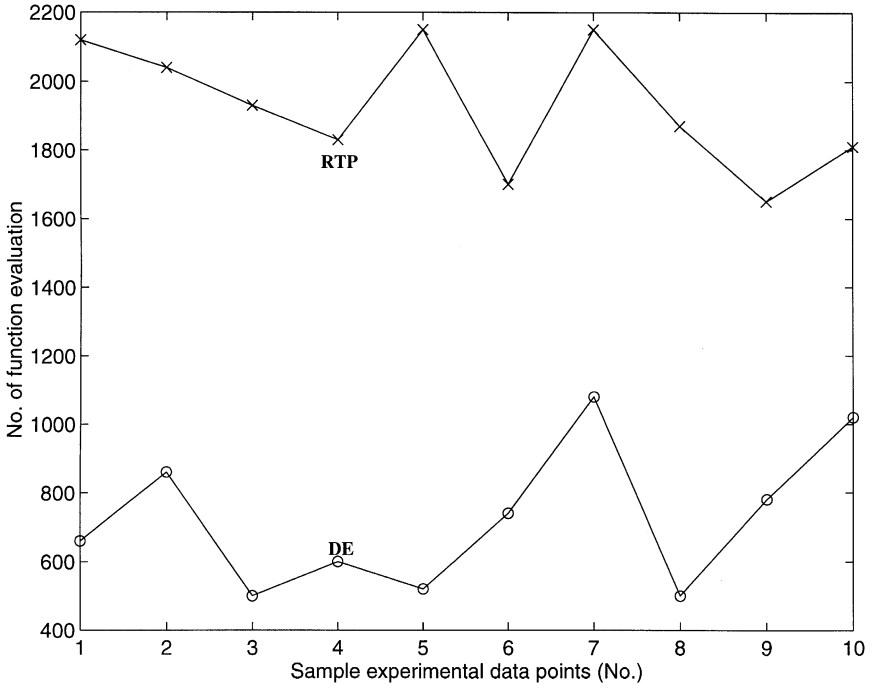

Fig. 3. Number of function evaluations: DE vs RTP.

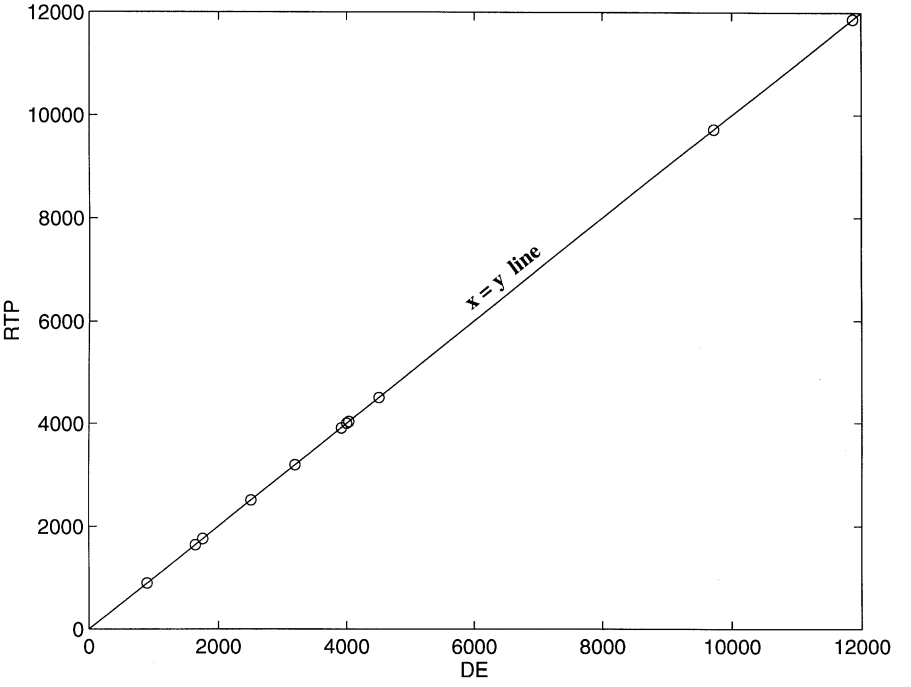

Fig. 4. Estimated value of h_w(W/m^{2}K): RTPvsDE.

algorithm is said to have converged if it satisfies all the conditions given below (Eq. (20)–(25)):

$$\mathscr{F}_\Gamma < \delta_1 \tag{20}$$

$$|\mathscr{F}_\Gamma - \mathscr{F}_{\Gamma-1}| < \delta_2, \tag{21}$$

$$k_{er,min} \leqslant k_{er,\Gamma} \leqslant k_{er,max}, \tag{22}$$

$$h_{w,min} \leqslant h_{w,\Gamma} \leqslant h_{w,max}, \tag{23}$$

$$|k_{er,\Gamma} - k_{er,\Gamma-1}| < \delta_3, \tag{24}$$

$$|h_{w,\Gamma} - h_{w,\Gamma-1}| < \delta_4, \tag{25}$$

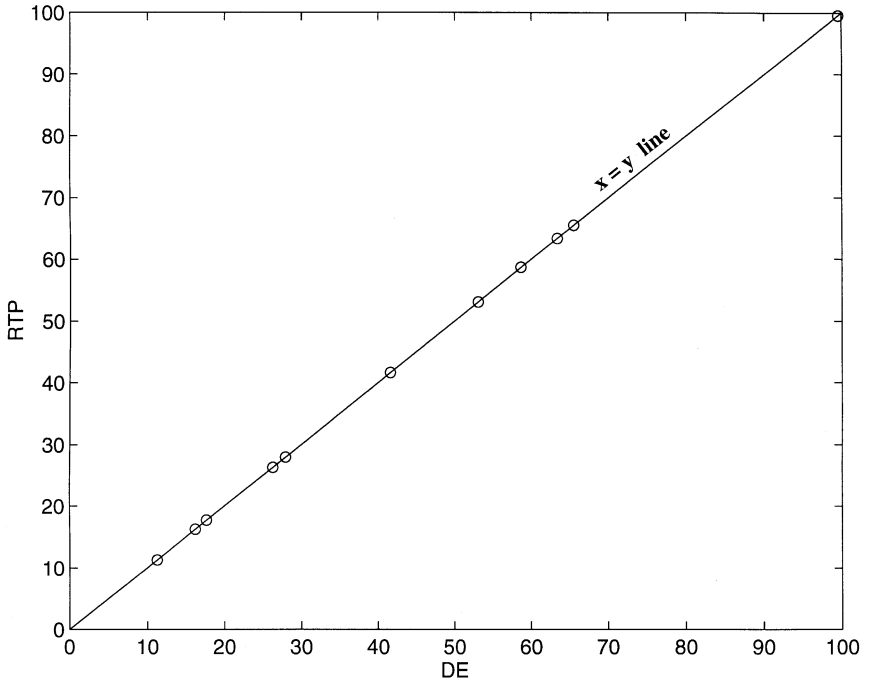

Fig. 5. Estimated value of k_{er} (W/m K): RTP vs DE.

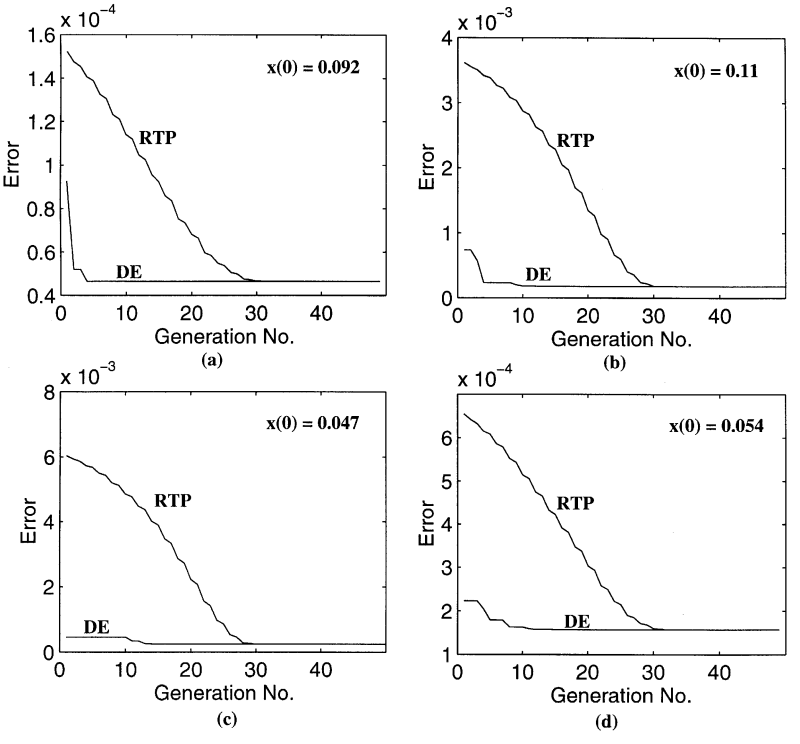

Fig. 6. Convergence of DE and RTP (x(0) is RMS error of the best initial guess of DE).

where δ_1, δ_2, δ_3 and δ_4 are constants. On the contrary, in case of DE the termination criterion is when 90–95% of the population have the same cost. The convergence criterion used for DE is:

$$|\sigma_\Gamma - \sigma_{\Gamma-1}| < \delta, \tag{26}$$

where δ is a constant (in the present study $\delta = 1.0 \times 10^{-4}$) and σ_Γ is the cost variance given by

$$\sigma_\Gamma = \frac{\sum_{i=1}^{NP} (\mathscr{F}_{i,\Gamma} - \bar{\mathscr{F}}_\Gamma)^2}{NP - 1}. \tag{27}$$

RTP either failed or took a long time to converge when the initial guess of h_w and k_{er} was bad. Besides, DE initial guesses generated randomly were spread throughout the search space. Still the convergence of DE is faster than that of RTP. The convergence of DE and RTP is compared for sample data points in Fig. 6a–d, which clearly shows that DE converges much faster than RTP irrespective of the initial guesses. As depicted in Fig. 6a even though the initial estimation error of the best vector (the vector with least cost) of DE algorithm is 0.092, and in RTP method the initial estimation error is only 1.52×10^{-4}, DE converged in 10 generations whereas RTP took 32 iterations. Similarly, as shown in Fig. 6b–d, the initial estimation errors of the best vector of DE algorithm are 0.11, 0.047 and 0.54, respectively, and it took 10, 15 and 12 generations respectively to converge. On the other hand, even though the initial estimation errors are 3.6×10^{-3}, 6.0×10^{-3} and 6.5×10^{-4}, RTP method took 30, 30 and 31 iterations, respectively, to converge. The results clearly illustrate that DE is a design tool of great utility that is immediately accessible for practical applications.

The previous studies (Weekman and Myers, 1965; Hashimoto et al., 1976; Muroyama et al., 1978; Matsuura et al., 1979a, b; Specchia aand Baldi, 1979; Crine, 1982; Lamine et al., 1996; Babu and Rao, 1997) on heat transfer effects in trickle-bed reactors have been conducted on non-reacting systems (air–water or air–glycerol). In the present study, the DE algorithm was applied on an air–water system as, to the best of our knowledge, no radial temperature profile data is available for trickle-bed reactors with reacting systems. As stated earlier, with the reaction term the pseudo-homogeneous model equations cannot be solved analytically. Since the proposed DE algorithm uses orthogonal collocation for solving the model equations which can also be applied for systems with reaction. However, from the results shown above, it can be predicted that the DE algorithm will be equally effective in estimating heat transfer parameters in the presence of reaction effects.

6. Conclusions

The present study clearly shows the potential for using DE in estimating the heat transfer parameters in trickle-bed reactors. In most of the studies carried out earlier in the estimation of heat transfer parameters in packed bed reactors, researchers have focused on using the first few terms of the analytical solution of the model equation and have used either gradient based or non-gradient based traditional optimization techniques for the estimation of h_w and k_{er}. Since the parameters are floating point and also due to its simple structure, ease of use, speed and robustness, it has been shown that a DE is the more appropriate choice for optimization. The results were compared with that of a RTP using analytical solution with Powell's method for estimation. In previous studies, DE has been used for design and control purposes, but in the present study DE is used as an estimator.

DE algorithm is much faster, has less computational burden when compared to the RTP algorithm and the estimation is much more accurate. It is also observed that DE algorithm converges to the global optimum irrespective of its initial population, whereas the RTP-Powell algorithm needed an initial guess nearer to the global optimum for convergence. The results presented in this study depict the scope of Differential Evolution in estimating the heat transfer parameters of a trickle-bed reactor. DE is more effective in terms of faster convergence, greater accuracy, lesser number of function evaluation and robustness. Based on these results, it is concluded that DE can be a very valuable resource for accurate and faster estimation of the heat transfer parameters, in multi-phase reactors such as trickle-bed reactors.

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Appendix A. Orthogonal collocation method

The equations of the boundary value problem to be solved (Eq. (10)–(13)) are

$$\frac{\partial\tilde{T}}{\partial\tilde{z}} = P'_{er}\,\frac{1}{\tilde{r}}\,\frac{\partial}{\partial\tilde{r}}\left(\tilde{r}\,\frac{\partial\tilde{T}}{\partial\hat{r}}\right),$$

$$\tilde{z}=0,\quad \tilde{T}=\tilde{T}_d$$

$$\tilde{r}=0,\quad \frac{\partial\tilde{T}}{\partial\tilde{r}}=0,$$

$$\tilde{r}=1,\quad -\frac{\partial\tilde{T}}{\partial\tilde{r}} = Bi\tilde{T}$$

In orthogonal collocation method, the unknown solution is expanded in terms of known expansion functions with arbitrary coefficients. One choice is

$$\tilde{T}(\tilde{r},\tilde{z}) = \tilde{T}(1,\tilde{z}) + (1-\tilde{r}^2)\sum_{i=0}^{N} a_i P_{i-1}(\tilde{r}^2) \tag{A.1}$$

in which the $P_i(\tilde{r}^2)$ are polynomials of degree i in $\tilde{r}^2$ and are defined to be orthogonal with the condition

$$\int_0^1 W(\tilde{r}^2)P_k(\tilde{r}^2)P_m(\tilde{r}^2)\tilde{r}\,\mathrm{d}\tilde{r} = 0,\quad k\leqslant m-1 \tag{A.2}$$

The polynomials defined by Eq. (A.2) are Jacobi polynomials and are given explicitly for cylindrical coordinates by

$$P_i(\tilde{r}^2) = f(-i, i+2, 1, \tilde{r}^2) \tag{A.3}$$

$$= 1 + \frac{(-i)(i+2)}{(1!)^2}\tilde{r}^2 + \cdots + \frac{(-i)(-i+1)\cdots(-1)(i+2)\cdots(i+2+i)}{(i!)^2}\tilde{r}^{2i} \tag{A.4}$$

The gradient and the Laplacian operators for the function $\tilde{T}(\tilde{r}, \tilde{z})$ of Eq. (A.1) are given by

$$\left(\frac{\partial \tilde{T}}{\partial \tilde{r}}\bigg|_{r=r_i}\right) = \sum_{j=1}^{N+1} A_{ij}\tilde{T}(\tilde{r}_j, \tilde{z}), \tag{A.5}$$

$$\nabla^2 \tilde{T}|_r = r_i = \left(\frac{1}{\tilde{r}}\frac{\partial}{\partial \tilde{r}}\left(\tilde{r}\frac{\partial \tilde{T}}{\partial \tilde{r}}\right)\bigg|_{r=r_i}\right) = \sum_{j=1}^{N+1} B_{ij}\tilde{T}(\tilde{r}_j, \tilde{z}). \tag{A.6}$$

Using Eqs. (A.5) and (A.6), the boundary value problem for a symmetric profile (Eqs. (10)–(13)) reduces to an initial value problem as follows:

$$\frac{\mathrm{d}\tilde{T}_i}{\mathrm{d}\tilde{z}} = P'_{er}\sum_{j=1}^{N+1} B_{ij}\tilde{T}_j, \quad j = 1, 2, \ldots, N, \tag{A.7}$$

$$\mathrm{I}C \qquad \tilde{T}_i(0) = \tilde{T}_d, \tag{A.8}$$

$$\mathrm{B}C \qquad -\sum_{j=1}^{N+1} A_{N+1,j}\tilde{T}_j = Bi\tilde{T}_{N+1}. \tag{A.9}$$

The summation limit $N+1$ arises from N collocation points plus one boundary condition.

Nomenclature

$\mathscr{A}$	collocation vector
A_{ij}	element of the collocation matrix **A**
b_i	coefficients of the analytical solutions
B	collocation matrix
$\mathscr{B}$	collocation vector
Bi	biot number
B_{ij}	element of the collocation matrix **B**
C_A	concentration of reactant **A**, kmol/m^3
C_o	inlet concentration, kmol/m^3
C_p	specific heat, J/kg K
C^*_{pG}	rate of change of enthalpy with respect to the rise in temperature of the gas phase, J/kg K
C_{pL}	specific heat of liquid, J/kg K
CR $(0 \leqslant CR \leqslant 1)$	crossover constant
ΔH	heat of reaction, J/kg
D	dimension
D_e	effective diameter of the packing, m
D_{er}	effective diffusivity, m^2/s
$\mathscr{F}$	cost or the value of the objective function
$\bar{\mathscr{F}}$	average cost
F $(0 < F \leqslant 1.2)$	scaling factor
G	gas flowrate, kg/m^2 s
h_w	Wall heat transfer coefficient, W/m^2 K
k_{er}	effective thermal conductivity of the bed, W/m K
L	liquid flowrate, kg/m^2 s
N	measurement points or number of collocation points
NP	population size
M $(0 < M < D-1)$	random integer
P_i	Jacobi polynomial of order i
P'_{er}	modified Peclet number, $(= k_{er}Z/(LC_{pL} + GC^*_{pg})R^2)$
Pr	binomial probability function
Q	positive-definite weighting matrix
r	radial position in trickle bed, m
$\tilde{r}$	dimensionless radius, r/R
r_A	reaction rate
R	trickle-bed radius, m
T	temperature, K
$\tilde{T}$	temperature vector, $(= T(\tilde{r}, \tilde{z}) - T_w)$ K
$\tilde{T}_d$	Temperature vector, $(= T_o - T_w)$, K
T_o	inlet fluid temperature, K
T_w	wall temperature, K
u	fluid velocity, m/s
u, **v**	trial vectors
$\mathbf{x}_{best,\Gamma}$	vector with minimum cost in generation Γ
$\mathbf{x}_{i,\Gamma}$	ith vector in generation Γ
$\mathbf{x}_{\Gamma 1,\Gamma}$, $\mathbf{x}_{r2,\Gamma}$	randomly selected vector
Z	length of the trickle bed, m
z	axial direction in the trickle bed, m
$\tilde{z}$	dimensionless axial position

Greek letters

$\delta, \delta_1, \delta_2, \delta_3, \delta_4$	constants
ϵ	dimensionless temperature vector, $\tilde{T}(\tilde{r}, 1)/\tilde{T}_d)$
ε	void fraction of the bed
γ	integer
Γ	generation number
$\lambda (0 < \lambda < 1.2)$	greediness scaling factor
ρ_B	catalyst bulk density, kg/m^3
ρ_f	fluid density, kg/m^3
σ_Λ	cost variance in generation Λ

Abbreviations

ARS	adaptive random search
DBF	dispersed bubble flow
DE	differential evolution
GA	Genetic Algorithms
PDE	Partial differential equation
PF	pulse flow
RTP	radial temperature profile method
TF	trickle flow

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