Process Systems Engineering PSE '85:

The Use of Computers in Chemical Engineering

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Preface

The second in a series entitled Process Systems Engineering, PSE '85 picks up the key themes of the previous PSE conference in Kyoto in 1982 and gives special emphasis to new topics and new directions.

The major sessions, each conducted by a Chairman-Rapporteur, concentrate on:-

Operability Considerations at the Design Stage
Design of Flowsheets
Aids for Plant Operations
Influence of new CAD Technology

In addition, a "poster session" is included where authors can present their latest ideas in the most effective way without the constraints imposed by a formal session. An exhibition of hardware/software runs concurrently with the conference and provides a shop window of the latest products relating to process engineering CAD.

PSE '85 is organised by the Institution of Chemical Engineers on behalf of the European Federation of Chemical Engineering in association with the Inter American Federation of Chemical Engineering and the Asian Pacific Confederation of Chemical Engineering. It continues a successful series of symposia held at the invitation of the EFCE Working Party on Computer Applications.

It has been a privilege and a pleasure to help organise the Conference. More than 120 papers of an excellent standard were submitted from all over the world — North, South, East and West! The task of reducing this number to a manageable volume was no easy one, and I am indebted to each and every one of my colleagues on the Technical Committee. The hours of lively debate being most stimulating! Finally, we are all of us especially indebted to Gillian Nelson and Julie Wearne for keeping us organised to our objectives and enduring our attempts to side-track their efforts.

This volume contains a compendium of some of the valuable work in progress in Process Systems Engineering today. Alas, there was no more room to publish the many worthy papers that we were forced to omit.

I sincerely hope you get as much pleasure from reading its contents as we did from compiling it.

F. A. PERRIS

Contents

iii

The Design of Flowsheets II
Session Chairman: Dr J.W. Ponton, University of Edinburgh

POSTER SESSION

Aids for Plant Operation

Influence of New CAD Technology

Design of Flowsheets

A. Simulation

B. Synthesis

* This paper is numbered out of page sequence

FORMAL SESSIONS

Operability Considerations at Design Stage II
Session Chairman: Prof O.A. Asbjornsen (University of Houston, USA)

REPRESENTING DISTRIBUTED UNITS IN AN EQUATION-ORIENTED SIMULATOR

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Summary

Equation-oriented flowsheeting programs work with algebraic equations, but some common process units are modelled by differential equations. Previous attempts to cope with the resultant difficulties are unsatisfactory. A new method is described, called the "black-box approach", in which subroutines are used which disguise the nature of the unit model, and instead pass to the main program a set of algebraic equations of the form it requires. The method is illustrated by successful application to a two-dimensional model of a packed bed catalytic reactor within a simple process flowsheet.

1. Introduction

In an equation-oriented simulator such as QUASILIN (1), the units are not represented by a modular simulator's familiar unit subroutines whose purpose is to calculate unit outputs given unit inputs. Instead the unit subroutine's job is to set equations. The algebraic equations describing the unit are divided into linear and non-linear. The former are retained unchanged and have appended to them the linear equations resulting from linearising, about some starting values, the non-linear equations. The equations thus set are combined with those of the other units in the flowsheet and solved. Iteration proceeds to converge.

If a unit is essentially distributed (i.e. is represented by ordinary differential equations (ODE's) or partial differential equations (PDE's), lacking a closed form solution) it does not fit naturally into this scheme, since it is not described by alegbraic equations. There are currently two ways of overcoming this difficulty.

Sarma and Reklaitis (2) used an equation-oriented model to represent a wall-cooled plug-flow tubular ethylene oxidation reactor. To convert their ODE's to algebraic form "approximate response surfaces had to be fit to the numerical solutions generated over a range of anticipated operating conditions. The response surfaces inherently have only a limited range of validity and hence may need to be refitted if their limits are exceeded during use of the model". Given the large number of variables which might be involved - feed composition and temperature, pressure, tube length and other reactor parameters (tube diameter, coolant temperature and so forth) - this approach has limited applicability. Kreule (3) developed a QUASILIN representation of a two-dimensional model (i.e. one with both axial and radial gradients) of a packed bed reactor using an intricate two-dimensional network of CSTR's. This has the disadvantages that:

(a) A new reactor model is required to replace an existing PDE model.

(b) As explained below, excessive storage requirements readily arise.

(c) There is no easy, cheap way of finding the derivative of an output
     variable w.r.t. reactor size, so limiting the usefulness of the
     model to simulation, as distinct from design or optimisation,
     calculations.

2. Direct Reduction to Algebraic Equations

An obvious alternative is to reduce the differential equation(s) to
algebraic equations using some approximate method e.g. orthogonal
collocation (OC). Unlike finite differencing, where the derivative at a
point is replaced by a linear combination of the variable values at that
point and neighbouring points only, OC replaces the derivative of a
variable C w.r.t. some co-ordinate x by a linear combination of the
variable values at a number of points x_i spaced over the whole domain of
x:

$$\frac{dc}{dx} \simeq \Sigma \ A_{ij} \ C_i \ ; \ \nabla^2 C \simeq \Sigma \ B_{ij} \ C_i, \ \text{where} \ C_i \equiv C(x_i) \qquad \{1\}$$

The points x_i, and weights A_{ij}, B_{ij} are calculated following Finlayson
(4): double precision calculation was found to be essential. The method
was applied to a two-point boundary value ODE model of an isothermal
tubular reactor with axial dispersion superimposed on plug flow, effect-
ing a 2nd order reaction ((4), pp 126-9). After substitution of {1}, the
differential equation of the model becomes a set of nonlinear algebraic
equations which QUASILIN handles successfully.

Consider next a second example from Finlayson: a two-dimensional plug-
flow model of a packed bed catalytic reactor which is host to an
exothermic isomerisation A → B at constant denisty. The model equations
are shown in Table 1 where they are written in terms of G_A, the molar
flowrate of A per unit cross-section area of reactor; T the temperature;
a, the dimensionless tube radius and z, the dimensional distance from the
reactor entrance. First, {1} is applied to the radial co-ordinate,
reducing the coupled PDE's to a set of coupled ODE's in the axial
co-ordinate

$$\frac{dG_{Aj}}{dz} = \alpha_m \sum_{i=1}^{N+1} B_{ij} \ G_{Ai} - \beta_m \ e^{-\gamma} \frac{G_{Aj}}{G} \qquad j = 1, 2 \ldots (N+1)$$

$$\frac{dT_j}{dz} = \alpha_h \sum_{i=1}^{N+1} B_{ij} \ T_j + \beta_h \ e^{-\gamma} \frac{G_{Aj}}{G^2} \qquad j = 1, 2 \ldots (N+1)$$

$$\sum_{i=1}^{N+1} A_{N+1, i} \ G_{Ai} = 0 \ ; \ \sum_{i=1}^{N+1} A_{N+1, i} \ T_i = Bi \ (T_w - T_{N+1})$$

$$z = 0 \ ; \ G_{Aj} = G_A \ \text{in}, \ T_j = T_{in} \qquad \{2\}$$

Here $G_{Aj} \equiv G_A(a_j)$ and $T_j \equiv T(a_j)$. Then OC is applied to {2}. Here a
difficulty arises: because of the shape of the axial profiles (Fig. 1)
with their severe changes of gradient, the power of OC is much reduced,

2

because e.g. the temperatures near the reactor inlet tell us nothing about the temperature gradients near the reactor outlet. This difficulty is avoided by applying OC within each of a sequence of 40 steps of z (4). Two interior collocation points were used within each step. This results in a total of 968 variables (and hence equations to be linearised) for QUASILIN: as shown below, probably 10 interior collocation points should have been used radially implying a total of 2904 variables. However, our operating system until recently imposed a limit of about 1000 variables for all the streams in a flowsheet. Thus direct reduction to algebraic equations may lead to exceeding permissible storage size. This same objection applies to attempts to model such a packed bed reactor by a network of CSTR's.

The escape from this restriction is obvious - a method must be found of modelling such reactors which requires QUASILIN to handle the variables in reactor inlet and outlet streams and not the variables relevant only to the interior of the reactor. Such an approach will hide from QUASILIN the detailed modelling of the reactor, so a reasonable name is:-

3. The "black box" Approach

For any process unit let the column vectors of input and output variables be $\underline{u}$ and $\underline{v}$ respectively. Then, from the point-of-view of QUASILIN, the unit may be treated as a "black-box", with its output related to its input by some functional relation

$$\underline{v} = \underline{h}\,(\underline{u}) \quad \text{or} \quad \underline{v} - \underline{h}\,(\underline{u}) = \underline{0} \qquad \{3,4\}$$

A Newton linearisation of {4} w.r.t. both $\underline{v}$ and $\underline{u}$ yields

$$\underline{\underline{I}}\,\underline{v}^{(m+1)} - \left[\frac{\partial \underline{h}}{\partial \underline{u}}\right]\underline{u}^{(m+1)} = \left[- (\underline{v} - \underline{h}\,(\underline{u})) + \underline{\underline{I}}\,\underline{v} - \frac{\partial \underline{h}}{\partial \underline{u}}\underline{u}\right]$$

which simplifies to

$$\underline{\underline{I}}\,\underline{v}^{(m+1)} - \left[\underline{\underline{J}}\right]\underline{u}^{(m+1)} = \left[\underline{h}\,(\underline{u}) - \underline{\underline{J}}\,\underline{u}\right] \qquad \{5\}$$

$$\text{where } \underline{\underline{J}} \text{ is the Jacobian } \partial \underline{h}/\partial \underline{u} \text{ i.e. } J_{ji} \equiv \partial h_j/\partial u_i \qquad \{6\}$$

Thus the set of linearised relations which QUASILIN requires, between the elements of $\underline{v}$ and $\underline{u}$, is given by {5}, whose use requires values for the elements of $\underline{\underline{J}}$, $\underline{h}$ and $\underline{u}$ within the square brackets. In contrast to a module in a modular simulator, the "black-box" must, for given $\underline{u}$, provide not only values of $\underline{h}$ but also of $\underline{\underline{J}}$. Two methods for finding these elements have been used, which will be illustrated with reference to plug-flow reactors.

(a) Perturbation

Here a subroutine is written to integrate the model ODE's with initial values available as $\underline{u}$ from the previous iteration. At the reactor exit, the values of $\underline{h}$ are obtained. By repeating the solution with one element of $\underline{u}$, u_i, perturbed to the value $u_i + \delta u_i$ where δu_i is typically ca. 1% of u_i, a new value of $\underline{h}$ is obtained, say $\underline{h}'$. Then $J_{ji} = \partial h_j/\partial u_i \simeq (h_j' - h_j)/\delta u_i$ is calculated for each element of $\underline{h}$. This perturbation-

3

plus-re-solution is repeated for each element of $\underline{u}$. The values of $\underline{u}$, $\underline{h}$ and $\underline{J}$ are used in the square brackets of {5}. The advantage of this method is that it allows use of a unit model originally devised for stand-alone or sequential-modular flowsheeting use. The disadvantage is that it requires (n+1) solutions of the model per flowsheet iteration, where n is the number of stream variables in the reactor input stream.

(b) Sensitivity coefficients

The set of ordinary differential equations describing a one-dimensional distributed model may be written generally as

$$\frac{dy}{dz} = \underline{f} \ (\underline{y}, \ \underline{p}, \ z) \qquad \{7\}$$

with initial values (IV's) $\underline{y}$ (o) = y_o.

Here z is the independent variable i.e. axial co ordinate, $\underline{y}$ is the column vector of dependent variables (temperature (T), molar flowrate of substance A (F_A) ....), $\underline{f}$ is the column vector of functional forms, and $\underline{p}$ is a vector of parameters. Differentiate the LHS of one eqn of the set w.r.t. one element of $\underline{y}_o$ and then reverse the order of differentiation on the LHS and apply the chain rule to the RHS.

$$\frac{\partial}{\partial y_{oj}} \frac{dy_i}{dz} = \frac{\partial f_i}{\partial y_{oj}} \quad \Rightarrow \quad \frac{d}{dz} \frac{\partial y_i}{\partial y_{oj}} = \sum_{\ell} \frac{\partial f_i}{\partial y_{\ell}} \frac{\partial y_{\ell}}{\partial y_{oj}}$$

Or, defining $\qquad \lambda_{ij} \equiv \dfrac{\partial y_i}{\partial y_{oj}}$, $S_{i\ell} \equiv \dfrac{\partial f_i}{\partial y_{\ell}}$ $\qquad \{8\}$

the result for all i,j summaries to

$$\frac{d}{dz} \underline{\lambda} = \underline{S} \ \underline{\lambda} \text{ with i.c. } \underline{\lambda}(o) = \underline{I} \qquad \{9\}$$

An alternative derivation is given in Rosenbrock and Storey (5), whose eqn (68) on p. 202 should be corrected.

Thus a set of coupled linear ODE's, called the auxiliary equations, describes the evolution of the sensitivity coefficients λ_{ij} along the reactor, where λ_{ij} summarises how y_i responds to some small change in the input variable y_{oj}. The matrix $\underline{\lambda}$ evaluated at the reactor outlet, z = L, therefore is identical to the matrix $\underline{J}$ of {5} and {6}. Thus, to obtain $\underline{h}$ and $\underline{J}$ for the square brackets in {5}, all that's required is the integration of the conservation equations {7} simultaneously with the auxiliary equations {9} to the reactor outlet. The IV's used for {7} are the values of $\underline{y}_o$ determined from the previous flowsheet iteration. For the isomerisation A $\rightarrow$ B at constant density, the particular form of {3} is

$$F_{A \ out} = h_1 \ (F_{A \ in}, \ F_{B \ in}, \ T_{in}) \ ; \ T_{out} = h_2 \ (F_{A \ in}, \ F_{B \ in}, \ T_{in}) \quad \{10\}$$

$$F_{B \ out} = F_{A \ in} + F_{B \ in} - F_{A \ out} \ ; \ F_{out} = F_{A \ out} + F_{B \ out} \qquad \{11\}$$

4

Hence from {10}

$$F_{A\ out} - \left(\lambda_{AA}^{out}\right) F_{A\ in} - \left(\lambda_{AB}^{out}\right) F_{B\ in} - \left(\lambda_{AT}^{out}\right) T_{in}$$

$$= \left(F_{A\ out} - \lambda_{AA}^{out} F_{A\ in} - \lambda_{AB}^{out} F_{B\ in} - \lambda_{AT}^{out} T_{in}\right)$$

and $T_{out} - \left(\lambda_{TA}^{out}\right) F_{A\ in} - \left(\lambda_{TB}^{out}\right) F_{B\ in} - \left(\lambda_{TT}^{out}\right) T_{in}$

$$= \left(T_{out} - \lambda_{TA}^{out} F_{A\ in} - \lambda_{TB}^{out} F_{B\ in} - \lambda_{TT}^{out} T_{in}\right)$$

{12}

where, for instance, $\lambda_{AA}^{out} \equiv \partial F_{A\ out} / \partial F_{A\ in} = \partial h_1 / \partial F_{A\ in}$, evaluated at the reactor outlet $z = L$. Equations {11} are retained unchanged since they are already linear, so {11} and {12} together correspond to {5}.

This technique has been applied successfully to the wall-cooled plug flow reactor (6) and to the isothermal plug flow reactor with multiple reactions (7). It is also possible to calculate other derivatives commonly required in design or optimisation. Thus $\partial y_i / \partial L$ is simply the value of the LHS of the ith member of {7} at the completion of integration at $z = L$. Further, the derivatives of the outlet values of $\underline{y}$ w.r.t. some of the parameter values might be required. Writing $\kappa \equiv \partial y_i / \partial p_j$ and $T_{ij} \equiv \partial f_i / \partial p_j$,

$$\frac{d}{dz} \underline{\kappa} = \underline{S} \underline{\kappa} + \underline{T} \text{ with i.c. } \underline{\kappa}(o) = \underline{0}$$

{13}

The required members of this additional set of auxiliary equations may also be integrated with the conservation equations {7}; the values of the elements of $\underline{\kappa}$ at the reactor outlet are the desired derivatives.

4. Application to a two-dimensional reactor model

There are two ways of applying sensitivity coefficients to PDE's. Either the PDE's are first discretised radially into ODE's - e.g. by OC using {1} - and then the method is applied to them; or the auxiliary PDE's are derived and then discretised radially for solution, which gives the same result. The auxiliary equations for the latter are shown in table 1, where, for simplicity, the parameters Bi, α_m, α_h are treated as being independent of F_A, F_B, T. The solution to the PDE's consists of quantities such as λ_{AA}^{out} at a number of radial positions a_i. However QUASILIN requires J_{ij} of {3-6}, which is clearly the derivative, with respect to inlet molar flowrate of A, of the radial (mixing cup) mean outlet molar flowrate of A, given by

$$F_{A\ out} = 2 \int_0^1 a F_A(L, a)\ da.$$

{14}

$$\lambda_{AA}^{out} = \frac{\partial}{\partial F_{A \; in}} 2 \int_0^1 a \, F_A \, (L, \, a) \, da =$$

$$2 \int_0^1 a \, \frac{\partial F_A (L, \, a)}{\partial F_{A \; in}} \, da = 2 \int_0^1 a \, \lambda_{AA} \, (L, \, a) \, da \qquad \{15\}$$

The integrals in {14} and {15} are calculated numerically from the integrand values at the a_j's.

In this way, all outlet variables and derivatives required in {11} and {12} are calculated. Calculations were performed using the parameter values listed in (8) which were derived from the dimensionless values of Finlayson. Axial integration was effected by three different methods (Runge-Kutta-Merson, Adams, Gear), and radial discretisation by two (OC, finite differencing). Figure 1 shows results obtained using 10 radial points. The (radial mean) temperature rises to a hot-spot, at which point the conversion of A is 98%. Thereafter the temperature sinks approximately exponentially towards the wall temperature and F_A falls towards zero. Hence at the outlet, all the sensitivity coefficients plotted are almost zero since only alteration of T_w could alter T_{out}, and F_A and F_B are scarcely alterable by any small change. At $z = 0.65$ m, just before the hot-spot, λ_{TT} goes through a maximum, because if T_{in} were raised a little, the hot-spot would occur nearer the entrance, and so T at 0.65 m would be much higher. Thereafter λ_{TT} falls to a negative value, since if T_{in} were raised, and hot-spot moved forward, near-exhaustion of reactants would mean that T at 0.80 m would be much lower. The other sensitivity coefficient plots can be explained similarly.

As a useful approximation, these reactor outlet results could be used in {12}, and integration of the auxiliary equations dispensed with altogether, if one could be sure that, at all flowsheet iterations, the reactor would be long enough, and T_{in} high enough, for profiles of this shape to develop. Figure 2 shows the results for the same case, but with only four radial points used. An unphysical kink appears in the F_A and T plots – an artefact of the poorer approximation to the PDE. The effect on the sensitivity coefficients at the bed exit is unimportant, but the effect at the kink is dramatic. If, during the flowsheet iterations, such misbehaviour should happen at the reactor outlet, then convergence would presumably be hampered. This implies, unusually, that for refined calculations towards the end of a simulation, it might be that a comparatively crude 4-point radial approximation to the PDE would suffice, whereas for preliminary calculations a more accurate 10-point radial approximation might be necessary. A cheaper solution would probably be to replace the two-dimensional model with a one-dimensional model in early iterations.

Comparison of numerical methods was not exhaustive, nor was any attempt made to optimise the subroutines used. However, it is noteworthy that OC was faster than finite differencing when only 4 radial points were used, as the literature (4) would lead one to expect. Unexpectedly, with 10 radial points finite differencing proved faster (9). With the same number of radial points, OC is the more accurate. Runge-Kutta-Merson was the fastest of the axial integration routines.

Satisfactory tests were made with the reactor module placed in a simple recycle loop with different values of L and F_{A-in} used (9). The perturbation method was used successfully to check several of the sensitivity coefficient calculations.

Discussion and Conclusions.

(i)   For one-dimensional plug flow reactor models, the black-box method works well. In a real flowsheet application, much of run-time will be spent in thermopacks i.e. on calculating thermodynamic properties (3). There will then be a heavy penalty for the multiple integra-tions of the perturbation method so the sensitivity coefficient method is preferred. This will require modification of existing modular flowsheeting routines by addition of the auxiliary equations. This is the recommended technique for any operation to be modelled by first-order ordinary differential equations with given initial values.

(ii) For a two-dimensional model, with its need for radial integration of output variables, there is no attractive alternative to a black-box module. Again, the sensitivity coefficient method is recommended.

(iii) As an alternative to the black-box approach, direct reduction to algebraic equations worked well for an isothermal boundary value problem. For more demanding non-isothermal boundary value problems, it seems likely that a black-box approach must be developed. Advances here may also prove useful for countercurrent cascades of stages (10).

(iv) It would be worth investigating the reduction in computing time realised by exploiting the linearity of the auxiliary equations (11).

(v)   Some methods of integrating the conservation ordinary differential equations generate the matrix $\underline{S}$ within the integration routine (12, 13). It should therefore be recovered and used in the auxiliary equations, which will thus be solved at almost no extra cost beyond that required for the conservation equations.

(vi) If resort is to be made to a response surface fit, it might best be derived by exploiting a black-box unit which provides both responses and their derivatives w.r.t. input variables and parameters.

(vii) Beginners find writing QUASILIN units difficult, partly because of the use of MORTRAN statements. Black-box units consist of FORTRAN coding, and might therefore be exploited as a route to making QUASILIN more user-friendly.

(viii) The sensitivity equation plots give some hint at the difficulties any flowsheeting method might face in representing a short reactor performing an irreversible exothermic reaction.

Acknowledgements

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