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Use of a Distributed Computer System for Tubular Reactor Profile and Catalyst Activity Identification

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INTRODUCTION

In the last decade or so, a relatively complete theory of sequential distributed parameter filtering theory has appeared in the literature (Tzafestas, 1978). All of the distributed parameter filters described to date are dynamic in nature. The dynamic feature of these filters adds considerable complexity, making them very difficult to implement on-line. In fact, applications of these fully dynamic filters are scarce and are limited to rather simple systems (Ray, 1978). Fortunately, the full complexity involving dynamic modeling and filtering of all process states and parameters is not required for many processes that are characterized by certain fast dynamic modes and also long-term, slowly varying effects.

A quasi-steady state approach is suitable for these industrial processes. This approach allows one to decompose the full dynamic distributed filter into two simpler ones: a steady state distributed parameter filter that can be used for the fast dynamic states and a simpler dynamic filter that can track the slower modes or parameters. The on-line implementation of two such filters, which are developed to estimate the states and to identify the catalyst activity of a packed-bed styrene monomer reactor, is presented here.

PROCESS DESCRIPTION, MATHEMATICAL MODEL, AND FILTER EQUATIONS

The process considered here is the thermal dehydrogenation of ethylbenzene to form styrene. Ethylbenzene is passed through a tubular reactor packed with an iron oxide catalyst and dehydrogenation takes place. There are ten distinct chemical species associated with this process. Since the main dehydrogenation reaction is strongly endothermic, a 50°C temperature drop down the reactor is typical. A two-dimensional, heterogeneous mathematical model is derived and presented elsewhere (Kuruoglu, 1977). A general model is simplified via order-of-magnitude scaling arguments. The final mathematical model is summarized in Table 1. This model is further simplified to allow for convenient on-line implementation. By assuming the gas phase temperature to be that of the catalyst phase, the two energy balance equations may be combined. Then the method of orthogonal collocation (Kuruoglu, 1982) is used to eliminate radial partial derivatives. Jacobi polynomials for a single interior collocation point are used to approximate the radial profiles. After these simplifications the mathematical model has the form, when expressed at a collocation point

$$\frac{\partial x}{\partial t} = F\left(x, \frac{\partial x}{\partial z}\right) \tag{1}$$

with $x(0,z)$ and $x(t,0)$ given. Here, x is the process state vector of temperature and nine compositions; F is a set of nonlinear differential expressions corresponding to the deterministic model of the process; t is time; and z is the axial location in the reactor from the entrance.

For identification purposes, the states of the process are augmented by a catalyst activity parameter related to the main dehydrogenation rate by

$$r = \alpha k e^{-E/RT} \tag{2}$$

where r is the reaction rate, α is the catalytic activity parameter, k is a frequency factor, E the activation energy, R the gas law constant, and T temperature. The state equation for the augmented parameter is postulated to be

$$\frac{d\alpha}{dt} = -\frac{\alpha}{\tau} \tag{3}$$

where τ is a first-order time constant.

Since the typical dynamic range of the state variables is on the order of 1 sec (or 1 min for temperature), while the activity decay time constant is of the order of hours or even days, a quasi-steady state assumption is used and the process dynamics are assumed to be instantaneous. Hence, the steady state form of Eq. 1 is used to describe the process state profiles. This is a set of ordinary differential equations, and white noise with a priori statistics is added to the right-hand side of Eqs. 1 and 2 in order to introduce model

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TABLE 1. SUMMARY OF THE MATHEMATICAL MODEL

Gas Phase Energy Balance

$$\rho\overline{C}_p \frac{\partial T_g}{\partial t} = -\rho\overline{C}_p v \frac{\partial T_g}{\partial z} + \lambda_r \left(\frac{1}{r}\right) \frac{\partial \left(r \frac{\partial T_g}{\partial r}\right)}{\partial r} - \frac{ha}{\epsilon}(T_g - T_c) \tag{a}$$

$$\text{with } T_g = T_{g0} \text{ at } z = 0; \quad \frac{\partial T_g}{\partial r} = 0 \text{ at } r = 0; \quad -\lambda_r \frac{\partial T_g}{\partial r} = h_w(T_g - T_w) \text{ at } r = R;$$

$$\text{and } T_g(r,z,o) = T_{g_{in}} \text{ at } t = 0$$

Catalyst Phase Energy Balance

$$\rho_c C_{p_c} \frac{\partial T_c}{\partial t} = k_c \left(\frac{1}{r}\right) \frac{\partial \left(r \frac{\partial T_c}{\partial r}\right)}{\partial r} + \frac{ha}{(1-\epsilon)}(T_g - T_c) + \frac{\epsilon}{(1-\epsilon)} \sum_{i=1}^{M} (-\Delta H_i) r_i \tag{b}$$

$$\text{with } T_g = T_c \text{ and } \frac{\partial T_c}{\partial r} = 0 \text{ at } r = 0; \quad T_c = T_w \text{ at } r = R; \quad T_c(r,z,o) = T_{c_{in}} \text{ at } t = 0$$

Component Balance for Species *i*

$$\frac{\partial C_i}{\partial t} = -\frac{\partial (vC_i)}{\partial z} + \mathscr{D}_r \left(\frac{1}{r}\right) \frac{\partial \left(r \frac{\partial C_i}{\partial r}\right)}{\partial r} - r_i, \; i = 1,2,\ldots,N \tag{c}$$

$$\text{with } C_i = C_{i_o} \text{ at } z = 0; \quad \frac{\partial C_i}{\partial r} = 0 \text{ at } r = 0, \text{ and at } r = R; \quad C_i(r,z,o) = C_{i_{in}} \text{ at } t = 0$$

and parameter uncertainties. Measurements of process states are available only at discrete spatial locations along the reactor in the form of temperatures. Temperature and concentration data are available at the reactor exit. All measurements are assumed to be corrupted with white noise of known statistics.

With the process and mathematical model summarized above, complete system identification requires a steady state estimator for the spatially distributed states, x, and a time-varying lumped parameter estimator for the catalyst activity. A steady state, sequential, distributed parameter filter has been developed to estimate state temperatures and composition profiles. The details of the theoretical development of this filter are presented elsewhere (Kuruoglu, 1982; Kuruoglu et al., 1982). A summary of the steady state filter is presented in Table 2. A second lumped parameter dynamic filter was also derived (Kuruoglu et al., 1982) to estimate the slowly varying catalyst activity. Table 3 summarized this dynamic Kalman filter.

ON-LINE CONTROL, ESTIMATION, AND IDENTIFICATION

A schematic diagram of the styrene pilot plant used in this work is shown in Figure 1. The stainless steel reactor is approximately 2.5 cm inside diameter and 20 cm long. Further details of the apparatus have been published elsewhere (Clough and Ramirez, 1976; Kuruoglu, 1982). The pilot plant is monitored and controlled by a distributed computer system. A Data General MP-100 microcomputer is interfaced directly to the experimental apparatus and handles the local control and data acquisition tasks. It communicates via a fiber optic link to a larger Data General Eclipse minicomputer, which serves to store experimental data and carry out the estimation and identification tasks. All programming is done in a multitasking version of FORTRAN.

Applications software written for the MP-100 microcomputer consists of three major tasks and a master task for scheduling. These three tasks are process input/output, console input/output, and intercomputer communication. A second program is written for the Eclipse minicomputer in a multiuser environment—the Eclipse also serves the needs of several other MP-100 micros in other research labs. Software in the Eclipse solves the mathematical model of the styrene reactor, computes the steady state distributed filter and lumped Kalman filter, exchanges data with the MP-100, and stores data to disk memory.

As mentioned earlier, there are ten distinct chemical components and temperatures described by the full mathematical model. This model was simplified for implementation of the filters to include only those components that exist in significant quantity. Only temperature and two major components, ethylbenzene and styrene, were included for direct on-line estimation. This reduced the computational problem from one of 66 coupled partial differential equations to six. See Table 2 for a summary of the distributed parameter filter equations. The compositions that are not directly

TABLE 2. STEADY STATE DISTRIBUTED FILTER EQUATIONS

Equations for the Initial Estimates

$$\frac{\partial \hat{\boldsymbol{x}}^-}{\partial t} = \mathbf{O} = \boldsymbol{F}\left(x^-, \frac{\partial \hat{\boldsymbol{x}}^-}{\partial z}\right) \tag{a}$$

$$\frac{\partial \boldsymbol{P}^-}{\partial t} = \mathbf{O} = \left(\frac{\partial \boldsymbol{F}}{\partial \hat{\boldsymbol{x}}^-}\right)_z \boldsymbol{P}^-(z,z') + \boldsymbol{P}^-(z,z') \left(\frac{\partial \boldsymbol{F}}{\partial \hat{\boldsymbol{x}}^-}\right)_{z'}^T + \left(\frac{\partial \boldsymbol{F}}{\partial \hat{\boldsymbol{x}}_z^-}\right)\left(\frac{\partial \boldsymbol{P}^-(z,z')}{\partial z}\right) + \left(\frac{\partial \boldsymbol{P}^-(z,z')}{\partial z'}\right)\left(\frac{\partial \boldsymbol{F}}{\partial \hat{\boldsymbol{x}}^-{}_{z'}}\right) + \boldsymbol{Q}(z,z') \tag{b}$$

Update Equations

$$\hat{\boldsymbol{x}}^+(z) = \hat{\boldsymbol{x}}^-(z) + \frac{1}{M} \sum_{i=1}^{M} \boldsymbol{K}(z,z_i)[y(z_i) - \boldsymbol{H}_i \hat{\boldsymbol{x}}^-(z_i)] \tag{c}$$

$$\boldsymbol{P}^+(z,z') = \boldsymbol{P}^-(z,z') - \frac{1}{M} \sum_{i=1}^{M} \boldsymbol{K}(z,z_i) \boldsymbol{H}_i \boldsymbol{P}(z_i,z') \tag{d}$$

$$\boldsymbol{K}(z,z_i) = \boldsymbol{P}^-(z,z_i) \boldsymbol{H}_i^T [\boldsymbol{H}_i \boldsymbol{P}^-(z_i,z_i) \boldsymbol{H}_i^T + M \boldsymbol{R}(z_i,z_i)]^{-1} \tag{e}$$

TABLE 3. DYNAMIC FILTER EQUATIONS

Equations for the Initial Estimates

$$\bar{\alpha}^n = \Phi\hat{\alpha}^{n-1} \qquad \text{(a)}$$

(discretized form of Eq. 3)

$$M^n = \Phi P^{n-1}\Phi + Q^{n-1} \qquad \text{(b)}$$

where $\Phi = e^{-\Delta T/\tau}$

Update Equations

$$\hat{\alpha}^n = \bar{\alpha}^n + P^n \sum_{i=1}^{M} \left(\frac{\partial h_i}{\partial \alpha}\right)^T_{\bar{\alpha}^n} R_i^{-1}(y_i^n - h_i^n(\bar{\alpha}^n)) \qquad \text{(c)}$$

$$P^n = \left(\tilde{M}^n + \sum_{i=1}^{M} \left(\frac{\partial h_i}{\partial \alpha}\right)^T_{\bar{\alpha}^n} R_i^{-1} \left(\frac{\partial h_i}{\partial \alpha}\right)_{\bar{\alpha}^n}\right)^{-1} \qquad \text{(d)}$$

where $\tilde{M}^n = (M^n)^{-1}$

$$\frac{\partial h_i^n}{\partial \alpha} = \begin{pmatrix} \dfrac{\partial T_i}{\partial \alpha} \\ \dfrac{\partial C}{\partial \alpha} \end{pmatrix}^n \text{ for } i = 1, \ldots 4$$

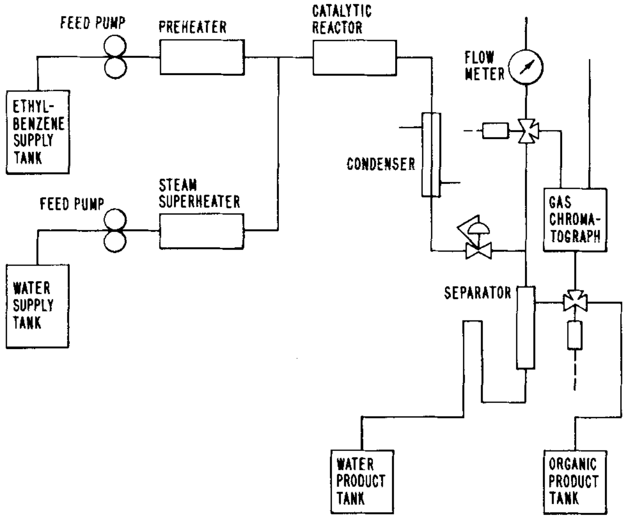

Figure 1. Styrene pilot plant.

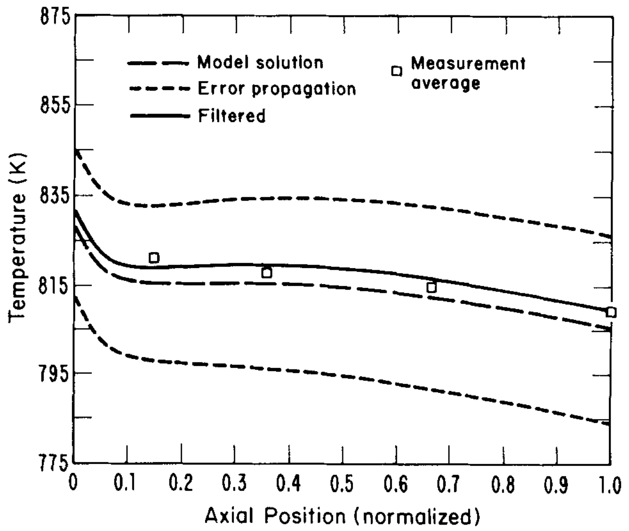

Figure 2. Temperature profile estimate with α = 1.

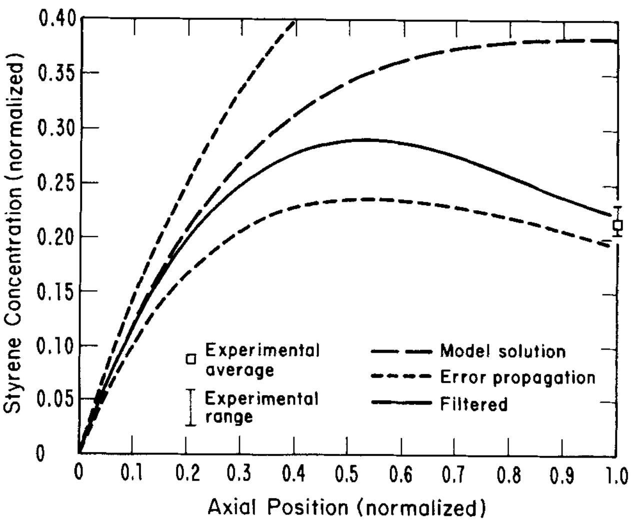

Figure 3. Styrene concentration profile estimate with α = 1.

estimated by the filter equations are determined via the on-line solution of the mathematical model. This model was also simplified by neglecting the water-gas shift reactions, which reduced it from eleven to six coupled differential equations. This latter simplification was justified by comparison of the full model solution to that of the simplified one. Nominal errors were less than 1%, which is commensurate with the model uncertainty used in filtering.

The model and measurement uncertainty values and the distribution functions used for on-line distributed parameter state estimation are given in Table 4. For dynamic filtering, a decay time constant of 180 hr and 3% model and initial uncertainty values were used for the catalyst activity parameter. The differential equations were solved using simple, explicit, finite-difference techniques (Kuruoglu, 1982). The computational time involved in solving the model, steady state, and dynamic filters on the Eclipse was nearly 20 min. Since measurements are available only every 35 min (constrained by the gas chromatograph's elution time for liquid plus gas samples), the on-line implementation of the filter is feasible using this distributed control system.

EXPERIMENTAL RESULTS FOR ON-LINE FILTERING

The steady state distributed filter and the lumped dynamic filter were tested on-line using the styrene pilot plant and the networked computers described above. Both filters processed the "live" noisy measurements and the "uncertain" model. The on-line experiments were run continuously over a period of ten days.

With an initial catalyst activity estimate of unity, ten sets of measurements were processed for reactor profile estimation using the distributed filter. Each set of measurements consists of reactor exit concentrations of ethylbenzene and styrene and four temperatures (measured at 3, 7, 13 and 20 cm) and is available for transmission by the MP-100 to the Eclipse at 35-min intervals. The distributed filtering results at the end of the tenth set are summarized in Figures 2 and 3. Figure 2 shows the model and filter estimates of reactor temperature profiles, while Figure 3 shows the styrene composition profile with the initial activity coefficient estimate of unity. From Figure 2 it is observed that the filtered estimate represents the reactor temperature profile better than the model estimate when compared to the actual measurement values. Figure 3 indicates that the initial activity estimate is much higher than the actual catalyst activity, and the actual conversion at 560°C with steam-to-oil ratio, SOR equal to 5 (steam-to-ethylbenzene in the feed, an operating parameter), to be 21% rather than the model value of 38%. Since the conversion estimate is mainly influenced by the concentration measurements, which are available only at the reactor exit, the estimated styrene concentration profile goes through a maximum, which is not realistic. This behavior is attributed to the significant error in the initial estimate of catalyst activity.

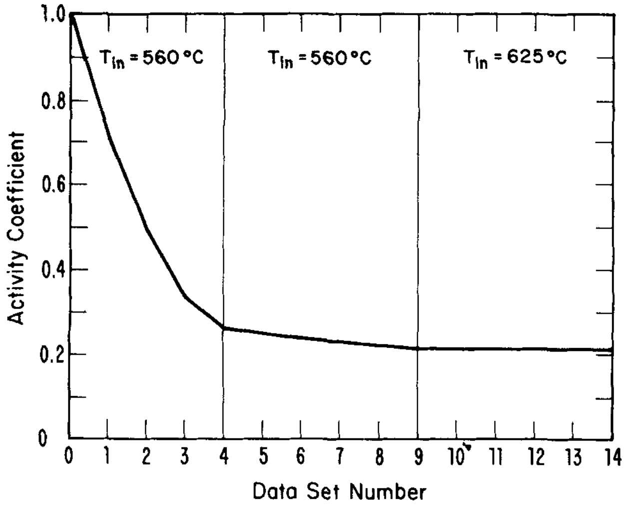

Figure 4. Summary of activity identificiation for SOR = 5.

TABLE 4. ERROR COVARIANCE FUNCTIONS USED FOR THE STEADY-STATE FILTERING

For Temperature

$Q_{11}(z,z') = 0.0001 \exp(-|z - z'|)$ (a)

$R_{11}(z_i,z_i) = 0.0004,\ i = 1, \ldots 4$ (b)

$P_{11}(0,z') = 0.0004 \exp(-z')$ (c)

For Concentration

$Q_{nn}(z,z') = 0.0225 \exp(-|z - z')$ (d)

$R_{nn}(z_i,z_i) = 0.0025,\ i = 4$ (e)

$P_{nn}(0,z') = 0.0025 \exp(-z')$ (f)

where $n = 2,3$

Figure 4 shows the convergence of the activity coefficient to the value of 0.215 after several iteractions of the lumped dynamic filter. With this converged value, the model estimates and the distributed filter estimates of temperature and concentration profiles agree extremely well. This agreement is illustrated in Figures 5 and 6.

After the convergence to an acceptable value of the catalyst activity, the reactor inlet temperature was raised to 625°C. This was accomplished by raising the temperature of the superheated steam to 650°C, while the ethylbenzene preheater exit temperature was maintained at 520°C. While keeping the SOR at 5.0, ten sets of measurements were processed for steady state distributed parameter state estimation. Catalyst activity was also identified after each two sets of measurements, yielding five more activity estimates of 0.215, 0.214, 0.213, 0.213, and 0.213. These activity values demonstrate that a 65°C increase in the reactor inlet temperature did not affect the catalyst activity. These estimates are also shown in Figure 4. The increase in inlet temperature did cause, however, an increase in ethylbenzene conversion from 21 to 37%, which is expected.

This experimental program demonstrated the effectiveness of on-line state estimation and parameter identification. This feasibility of using a distributed minicomputer/microcomputer network is also apparent. Estimated state variable profiles and catalyst activity values were viable and demonstrate the appropriate blend of model prediction and measurement which optimal filters should yield.

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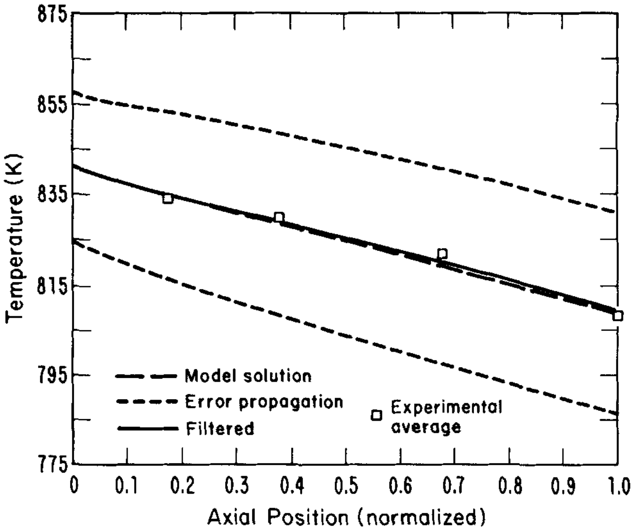

Figure 5. Temperature profile estimate with α = 0.22.

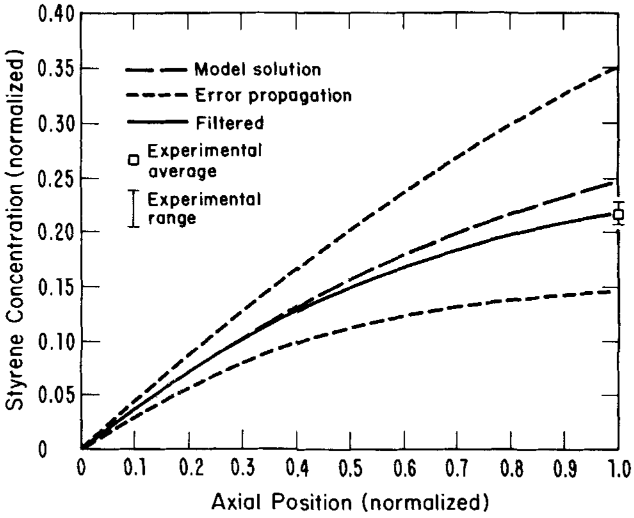

Figure 6. Styrene concentration profile estimate with α = 0.22.

NOTATION

a = catalyst surface area per unit volume of reactor
C_i = concentration of species i
$\overline{C}_p$ = weighted average heat capacity
$\mathcal{D}$ = mass dispersion coefficient
$\mathbf{H}$ = measurement matrix
h = local gas phase/catalyst bed heat transfer coefficient
$\mathbf{h}$ = measurement vector relating the activity coefficient to measurements
h_w = fluid/wall heat transfer coefficient
ΔH_j = heat of reaction for jth reaction
$\mathbf{K}$ = filter gain matrix
k = rate constant
k_c = conductive heat transfer coefficient of catalyst bed
M = number of measurement locations (M is also used for the total number of reactions for the mathematical model)
M^n = error covariance of the a priori estimate at nth time interval
N = number of interior collocation points (N is also used for the total number of chemical components)
$\mathbf{P}$ = error covariance of the estimate
$\mathbf{Q}$ = error covariance of the model uncertainty
r = radial position
R = reactor radius

r_i = rate of disappearance for species i
t = time
T = temperature
ΔT = sampling time interval
v = average gas velocity
x = state vector
$\hat{x}$ = state vector estimate
y = measurement vector
z,z' = axial position

Greek Letters

α = catalyst activity coefficient
α_w = overall heat transfer coefficient at the wall
ϵ = porosity of packed bed
λ = heat dispersion coefficient
ρ = mass density
τ = catalyst activity decay time constant
Φ = state transition function

Subscripts

c = catalyst bed
i = discrete spatial location
j = discrete time state
g = gas phase
r = radial component
w = wall
z = axial component

Superscripts

$+$ = *a posteriori* estimate
$-$ = *a priori* estimate

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Modification of Petersen's Model for Porous Solids

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In his pioneering work in the field of kinetics of porous solids, Petersen (1957) introduced the notion that surface area and therefore reaction rate will vary as solid is consumed. The developing surface was modeled as a sum of cylindrical pore contributions reduced by the sum of the effects of the random intersections. The result is a quadratic function of pore radius, which was recast by Szekely et al. (1976) in the form

$$S = 2\pi r L - K r^2 \qquad (1)$$

Equation 1 predicts

$$S = 0 \quad \text{at} \quad r = 2\pi L/K \qquad (2)$$

Since pore volume is the integral of surface with respect to radius

$$\epsilon = \pi L r^2 - (K/3) r^3 \qquad (3)$$

which has a maximum value of

$$\epsilon = \frac{4}{3}\frac{\pi^3 L^3}{K^2} \quad \text{at} \quad r = 2\pi L/K \qquad (4)$$

By using the initial condition that $\epsilon = \epsilon_0$ at $r = r_0$, Eq. 3 yields

$$\epsilon_0 = \pi L r_0^2 - \left(\frac{K}{3}\right) r_0^3 \qquad (5)$$

Dividing Eq. 3 by 5 gives

$$\frac{\epsilon}{\epsilon_0} = \left(\frac{r}{r_0}\right)^2 \left[\frac{G - (r/r_0)}{G - 1}\right] \qquad (6)$$

where

$$G = 3\pi L / K r_0 \qquad (7)$$

In order to develop an algorithm for computing G from available initial porosity data, it is useful to evaluate Eq. 6 at $r = (2\pi L/K)$, where

$$\frac{\epsilon}{\epsilon_0} = \frac{4}{27}\frac{G^3}{G - 1} \qquad (8)$$

By assuming that $\epsilon = 1$ for this maximum position, Petersen (1957) rearranged Eq. 8 to obtain the cubic relation

$$\frac{4}{27}\epsilon_0 G^3 - G + 1 = 0 \qquad (9)$$

But this assumption is in fact contradicted by Eq. 4. If, instead, the correct value