unhindered information flow reversals and equal component difficulty plays an important role as an introduction to the more complete problem.

In subsequent reports we will discuss the ordering of recycle calculations, the effects of direction dependent component computational difficulties, and the implications of partial acyclicity. These topics, along with the methods presented in this first report, may lead to a more orderly approach to complex systems calculations.

Obviously, the bipartite graph is only useful as a method of developing the algorithms, and in a practical problem the ordering of the equations and the selection of design variables would be performed without the use of the graph, applying the concept of list processing to a list of design equations in which the occurrence of each variable has been noted.

ACKNOWLEDGMENT

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NOTATION

c_i = composition of stream i
F = degrees of freedom
f_i = i^{th} function
k = number of tears
k^* = minimum number of tears
M = total number of variables
N = total number of equations
q_i = mass flow rate of stream i
$R(c, T, V)$ = reaction rate relationship
S = separation ratio
T = reaction temperature
V = reactor volume
v_j = j^{th} variable
$\rho(A)$ = local degree of node A

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Rate Data and Derivatives

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In most rate process problems, the original experimental data must be differentiated to obtain the derivatives, which are the information actually sought. For first derivatives all the commonly used methods give about the same result. However, the common methods of calculating derivatives can give very different second derivatives for the same set of starting data. Thus, in problems in which the desired result is a second derivative, the answer obtained is a strong function of the method used to calculate second derivatives. This is illustrated by several examples.

Churchill (*1*) has pointed out one important similarity between heat, mass, and momentum transfer and reaction kinetics. In all of these fields, the engineer is interested in determining rates (heat, mass, momentum transfer, and chemical reaction rates). These rates are generally obtained by differentiating integral data. For example, the momentum transfer rate to a particle (force) is proportional to the acceleration. The most common way (*14*) to obtain such accelerations is to differentiate position–time (integral) data twice to obtain d^2x/dt^2. Similarly, chemical reaction rates are determined by differentiating composition–time (integral) data and heat transfer rates by differentiating temperature–time or temperature–distance data.

In some cases these rates can be obtained from steady state experiments, for example, steady state heat transfer, steady state mass transfer, chemical reactions at constant composition (continuous stirred-tank reactors), or from force measurements on supported bodies. Even these data are often differentiated to obtain more interesting information; for example, steady state chemical reaction data are differentiated with respect to space velocity to elucidate kinetics (*16*).

Thus, in all these fields the desired derivatives are obtained from integral data. Given a table of x-y data, there are several widely known ways to find dy/dx and d^2y/dx^2. As will be shown here, all give practically the same values of dy/dx, but they can give very different values of

d^2y/dx^2. Thus, in an experiment involving such a second derivative, the experimental result is a strong function of the method used to calculate the second derivative from the x-y data, as will be illustrated. The common methods are:

1. Plot the data, draw a smooth curve through it, and read the slopes from this curve (*8*).
2. Fit the entire set of data with some fitting function and then differentiate this function.
3. Fit small segments of the data with some fitting function and then obtain local derivatives by differentiating the segmented functions (*15*).
4. Difference locally via a difference table, plot the differences, and smooth the differences, either graphically or numerically (*13*).

Each of these procedures involves smoothing processes, that is, the replacement of a measured value with another smoothed value. The derivatives are then based on the smoothed values. How these methods smooth data will be illustrated in several examples.

FIRST EXAMPLE

Smoothing the Data

In columns 1 and 2 of Table 1 are shown the observed angular positions of a particle moving outward in a centrifugal force field at equal time increments. The measurements were taken from an enlarged, multiple-exposure photograph of the particle trajectory (*10*).

The first step in analyzing any such data is to plot it as θ vs. t. On this plot (not shown), it appears that a straight line represents the data fairly well. One way of drawing that line is via the least squares fit. This has been done, and the resulting residuals are shown in column 3 of Table 1. The residuals form a consistent (nonrandom) pattern, suggesting that a better fit can be obtained with some curve rather than a straight line.

If method 1 is to be used, a french or ship's curve is selected and a smooth curve drawn through the data points. Unfortunately, there is no reproducible way of doing this; two engineers will draw different curves, and a single engineer will draw two different curves if he smooths the same set of data twice. Methods are available (*8*) for making the best estimate of the slope of such a curve once it is drawn, but no reproducible method is available for drawing the curve.

To use method 2, one normally selects a power series as his fitting function because of the convenient availability of computer programs for fitting such power series by least squares fit. The data in columns 1 and 2 of Table 1 were fit by such a program (*3*) by using five arbitrary constants; the residuals are shown in column 4 of Table 1. The constants for the fit are shown in Table 3.

To use method 3, some fitting procedure is needed. Examples are the Douglas-Avakian (*8*) and Whitaker-Pigford (*15*) methods. The Whitaker-Pigford method determines the first and second derivatives at a point by fitting a three-constant power series (a parabola) to five consecutive data points, with the point in question as the midpoint, by least squares. From the resulting power series constants, the local values of the first and second derivatives are computed. The calculated value of the fitting function at any one point has up to five different values, one for the fit where it is the midpoint, one for the fit where it is one point lower, etc. In column 5 of Table 1 are listed the residuals calculated from the averages of these representations by

$$r_i = \frac{\sum_{j=1}^{j=n} |r_{ij}|}{n} \tag{1}$$

Here r_i is the average residual at point i, r_{ij} is the value of r_i calculated by using the j^{th} curve fit which includes point i, and n is the number of curve fits that include point i. N is 1 for the first and last points, 2 for the second and next to last, etc., up to a maximum of five for interior points.

Method 3 (the difference table method) may be construed as a variant of the third. The use of the difference table in numerical analysis is well known (*4, 6, 9, 18*), but its use to test and differentiate engineering data is ignored by most textbooks (*4, 6, 9, 18*), treated lightly by others (*11*), and argued against by some (*5, 12*). A method of data graduation, based on the difference table method shown here, has been proposed (*2, 13*). A difference table of the data in Table 1 is shown in Table 2.

The connection between methods 3 and 4 is that both fit short sections of the data by arbitrary functions. In the Whitaker-Pigford method, a parabola is fitted to five points. In the difference table method, the first divided differences ($\Delta\theta/\Delta t$ in Table 2) are obtained by fitting a straight line exactly to two consecutive points. The second divided differences ($\Delta^2\theta/\Delta t^2$ in Table 2) are obtained by

TABLE 2. DIFFERENCE TABLE FOR DATA SHOWN IN TABLE 1

θ, rad.	t, sec.	$\Delta\theta$, rad.	Δt, sec.	$\Delta\theta/\Delta t$, rad./sec.	$\Delta(\Delta\theta/\Delta t)$, rad./sec.	Δt, sec.	$\Delta^2\theta/\Delta t^2$, rad./sec.^{2}
0.251	0.0798						
		0.433	0.0798	5.42			
0.684	0.1596				−0.02	0.0798	−0.25
		0.431	0.0798	5.40			
1.115	0.2394				−0.22	0.0798	−2.75
		0.413	0.0798	5.18			
1.528	0.3192				−0.14	0.0798	−1.75
		0.402	0.0798	5.04			
1.930	0.3990				−0.28	0.0798	−3.50
		0.380	0.0798	4.76			
2.310	0.4788				+0.02	0.0798	+0.25
		0.382	0.0798	4.78			
2.692	0.5586				−0.12	0.0798	−1.50
		0.368	0.0798	4.62			
3.060	0.6384				−0.02	0.0798	−0.25
		0.367	0.0798	4.60			
3.427	0.7182						

TABLE 1. DATA OF EXAMPLE 1

Observed and correlated angular positions of a spherical particle moving outward in a centrifugal force field (10). Residual = θ Observed − θ Calculated.

(1) Observed angle θ, rad.	(2) Observed time t, sec.	(3) Residual from linear fit	(4) Residual from polynomial fit	(5) Residual from *W-P* fit	(6) Residual from data difference fit
0.251	0.0798	−0.0530	0.0027	0.0012	—
0.684	0.1596	−0.0162	−0.0050	0.0016	—
1.115	0.2394	0.0187	0.0002	0.0010	+0.0041
1.528	0.3192	0.0356	0.0012	0.0010	—
1.930	0.3990	0.0414	0.0042	0.0020	+0.0030
2.310	0.4788	0.0253	−0.0034	0.0027	—
2.692	0.5586	0.0112	0.0007	0.0016	+0.0046
3.060	0.6384	−0.0170	−0.0014	0.0006	—
3.427	0.7182	−0.0461	0.0009	0.0001	—
Avg. residual $= \sum_n \frac{\lvert r \rvert}{n}$		0.0293	0.0022	0.0013	0.0012
Root-mean-square residual $= \sqrt{\frac{\Sigma r^2}{n}}$		0.0369	0.0028	0.0015	0.0023

fitting a parabola exactly to three consecutive points. The distinction between the two methods is that method 3 does the smoothing in this fitting procedure (three constants, five points), whereas method 4 uses the results of this exact fitting (three constants, three points) as the starting point for the smoothing process. To smooth the data by using the difference table, the divided differences $\Delta\theta/\Delta t$ shown in Table 2 are plotted vs. t in Figure 1. Each line represents the average value of $(d\theta/dt)_{\text{obs.}}$ over the t interval. The area under each line equals the change in θ for this Δt. Some of the properties of this type of plot have been published (*11, 13*).

In Figure 1 a straight line has been drawn through the various data lines. The lines scatter above and below it, suggesting experimental error. This is also obvious from the right-hand column of Table 2, in which the second divided differences are seen to vary erratically. For physical reasons it appears unlikely that the velocity of the particle is discontinuous, so it seems reasonable to smooth the data in Figure 1. Note that up to now no smoothing has been made at all in this method. In Figure 1 the value of $\Delta\theta/\Delta t$ from $t = 2$ to $t = 3$ is high; from $t = 3$ to $t = 4$, $\Delta\theta/\Delta t$ is slightly low relative to the rest of the data. If the measured value of θ at $t = 3$ were smaller, this would bring these more into line. Similar comments apply to the values of θ_5 and θ_7. A table similar to Table 2 is obtained by dropping these three readings completely out of the table and by repeating tabular differentiation. The resulting values of $\Delta\theta/\Delta t$ are plotted in Figure 2 and a smooth curve drawn through them. Thus, there are two smoothing steps in this method: (1) dropping the most unlikely data from the difference table and (2) drawing a smooth curve through the resulting first divided differences.

After drawing the smooth curve in Figure 2, one can now back-calculate the smoothed values of the data points which were dropped from this table. For example, the smoothed value of θ_3 must be

$$\theta_3 = \theta_2 + \left(\frac{d\theta}{dt}\right)_{\text{avg } t_2 - t_3} (t_3 - t_2) \tag{2}$$

The average values of the derivatives are read from Figure 2 and the correction obtained. These are shown as residuals in column 6 of Table 2.

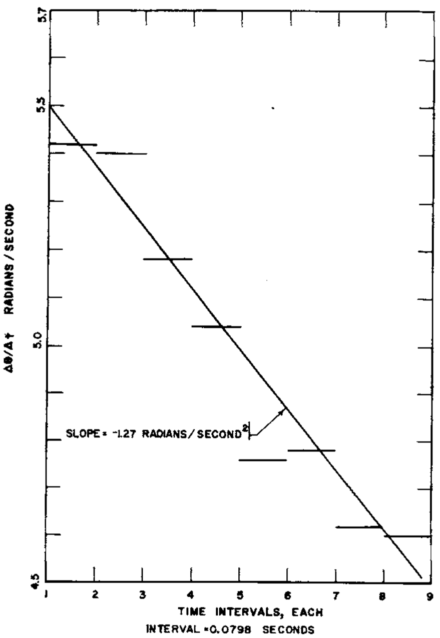

Fig. 1. $(\Delta\theta/\Delta t)$ vs. time from differenced data in Table 2.

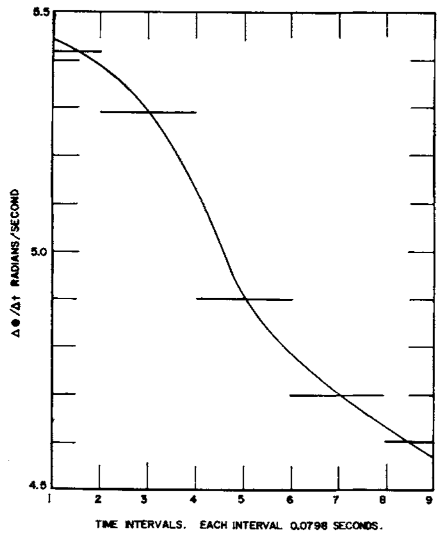

Fig. 2 $(\Delta\theta/\Delta t)$ vs. time from difference table made by dropping readings at t_3, t_5, and t_7 from Table 1.

Now the difference table is constructed again and the first and second derivatives are plotted to see if they form a smooth sequence. If not, a new curve is drawn in Figure 2 and the process repeated until smooth first and second derivatives are obtained.

Finding the Derivatives

After smoothing the data, as discussed, one can then find the derivatives from these smoothed data. If a smooth curve was drawn through the data (method 1), the slopes could be read by well-known methods (*8*). From the curve fit (method 2), by using a fourth-order power series, the derivatives are calculated from

$$\frac{d\theta}{dt} = a_1 + 2a_2 t + 3a_3 t^2 + 4a_4 t^3 \tag{3}$$

$$\frac{d^2\theta}{dt^2} = 2a_2 + 6a_3 t + 12a_4 t^2 \tag{4}$$

Here a_n are the constants in the power series fit. These curves are shown in Figures 3 and 4.

From the segmented polynomial fit, by using the Whitaker-Pigford method (method 3), the local values of the first two derivatives at various points are calculated from equations of the form

$$\left(\frac{d\theta}{dt}\right)_j = A_{1j} + 2A_{2j} t \tag{5}$$

$$\left(\frac{d^2\theta}{dt^2}\right)_j = 2A_{2j} \tag{6}$$

Here A_{ij} are the i^{th} constants in the parabolic fit for the five points, including point j as the midpoint. These values are shown as point values in Figures 3 and 4.

From the difference table method (method 4), a new difference table is made of the smoothed data the resulting first and second derivatives shown in Figures 3 and 4. A smooth curve has been drawn through the data differences in Figures 3 and 4.

In Figure 3 it is clear that methods 2, 3, and 4 give about the same first derivative. Note that if the straight line through the data on the $\theta - t$ plot were accepted as the best representation of the data, then the values of $d\theta/dt$ corresponding to it would be a straight horizontal line ($d\theta/dt = 5.0$) in Figure 3.

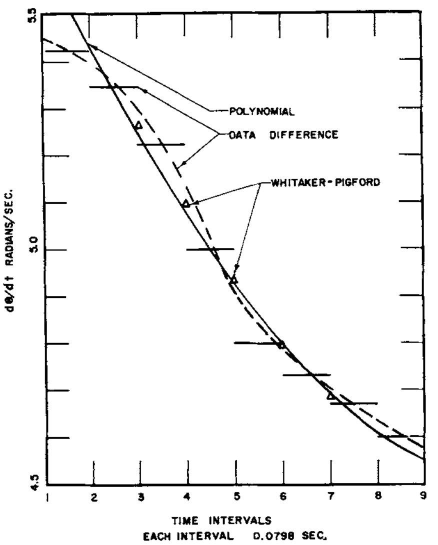

Fig. 3. Calculated first derivatives for first example.

In Figure 4, however, the second derivatives calculated by methods 2, 3, and 4 are quite different. Methods 3 and 4 give about the same derivatives, but the power series representation gives a much different derivative, particularly at low values of t. If the straight line on the θ—t plot were accepted as the best representation, then all values in Figure 4 would be zero.

Which Is Best?

In this case, the information actually sought was the second derivative, $d^2\theta/dt^2$. Figure 4 shows that by using different methods of smoothing the data to calculate this derivative from integral data, one can obtain results which differ significantly. Which should he believe, if any?

It seems clear that the straight-line representation on the θ—t plot is not the best. The average residual can be reduced tenfold by going from that representation to any of the others. Of the three remaining methods, the polynomial fit gives the largest average and root-mean-square residuals. Of all the methods, the polynomial method makes the most severe advance conditions on what form the data must have. If, as was done in this example, a fourth-degree polynomial is used, then, as shown in Equation (4), the second derivative, over the entire range of the data, is constrained to be a parabolic segment. The other two methods indicate that the second derivative does not have this shape.

On these two criteria, it appears that the polynomial fit is the worst of the three remaining methods. The other two give practically the same result; the choice between them should probably be based on convenience of application or on personal preference.

Do any of the methods give true derivatives? Given the data in Table 1 alone, there is no way to find true derivatives. All available methods smooth the data to find derivatives. Those which are most faithful to the data are those which change it least in smoothing and which make the fewest assumptions in advance about the form of the resulting smoothed data table. In smoothing these data, all methods indicate that changes larger than the experimental uncertainty are needed to produce a table with smooth derivatives. Thus, the experimental data must be significantly in error; only approximate derivatives are possible.

SECOND EXAMPLE

The first example showed that for the data presented, four significant figures are not enough to provide an unambiguous second derivative. From Figure 4 it is also obvious that, starting with these data and using the different smoothing methods, one would calculate third derivatives which differ from each other in sign and in absolute value by several orders of magnitude.

If very precise data are smoothed by the various methods, no such ambiguity results. An example is shown in Figure 5. Here the P-V (pressure-volume) data of Michels and Nederbragt (7) for methane at 0°C. have been differenced three times and the values of $\Delta^3P/\Delta V^3$ plotted

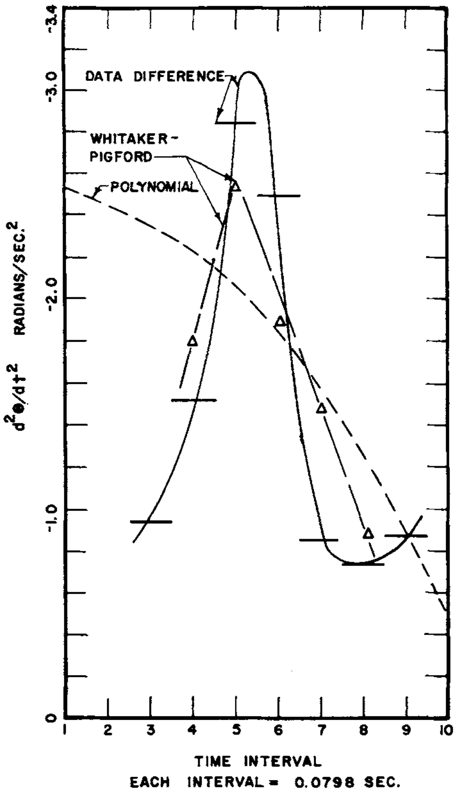

Fig. 4. Calculated second derivatives for first example.

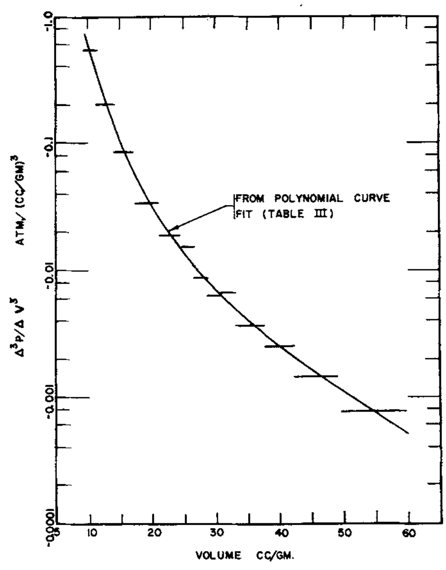

Fig. 5. ($\Delta^3P/\Delta V^3$) of methane at 0°C. Data of Michels and Nederbragt (7).

vs. V. The results of random experimental errors (or round-off error) show up slightly in this plot. They are more apparent in the $\Delta^4 P/\Delta V^4$ plot (not shown), but that plot also can be readily interpreted by drawing an average curve through the data differences.

Michels and Nederbragt (7) fitted their experimental data by a truncated power series. The constants are also shown in Table 3. By using this equation, d^3P/dV^3 was calculated, and is also shown in Figure 5. The agreement between the derivatives obtained by data differencing and by power series representation is very good. (The agreement is also satisfactory for the power series and data differenced values of d^4P/dV^4.) This example shows that if data of great precision are available, either method is acceptable (if the data can be accurately represented as a truncated power series). Data of this precision could also probably be plotted on huge graph paper and graphically differentiated with good accuracy. Notice that in order to obtain data precise enough to stand three differencings, it is necessary to use data from thermodynamics. Rate data of this precision are rare, if they exist at all.

Discontinuous Derivatives

Discontinuous first derivatives are common in thermodynamics (for example, the break in the cooling curve of a pure component, which corresponds to a phase change), but they are less common in rate processes. For example, a discontinuous velocity would require an infinite acceleration, which is physically unlikely. However, if measurements were made of temperature–distance in a double-pipe cooler-condenser in which a multicomponent mixture was being cooled and condensed, then the temperature–distance curve would be continuous but the dT/dx vs. distance curve would have a jump discontinuity at the point where condensation began.

Discontinuous second derivatives are more common in problems involving motion. For example, if one measured the distance–time behavior of an auto over the time span during which the driver took his foot off the accelerator and shoved it quickly onto the brake, the distance–time curve would be continuous, the velocity–time curve would be continuous, but the acceleration–time curve would have a jump discontinuity.

Customarily, mathematicians assume that all natural phenomena are representable by analytic functions. But analytic functions have continuous derivatives of all orders, whereas the previous examples show that significant physical phenomena have discontinuous derivatives. If one uses a derivative finding method, which assumes in advance that the data can be represented by an analytic function, then he has guaranteed in advance that, if such jump discontinuities in derivatives exist, they will be smoothed over. Tyler (*14*) has shown that the acceleration of spheres in a moving gas stream can be discontinuous. If he had used an analytic function method of differentiating his position–time data, he would never have observed this behavior.

TABLE 3. CONSTANTS IN DATA FITTING EQUATIONS
$y = a_0 + a_1x + a_2x^2 + a_3x^3 + a_4x^4 + a_5x^5$

Data set	Table 1 linear fit	Table 1 polynomial fit	Michel's data (7)
y	θ	θ	PV
x	t	t	ρ
a_0	−0.09211	−2.074934	1.00242
a_1	4.964	5.808533	-2.4144×10^{-3}
a_2	0	−1.208626	5.7541×10^{-6}
a_3	0	0.0	-3.550×10^{-9}
a_4	0	0.3208876	1.218×10^{-11}

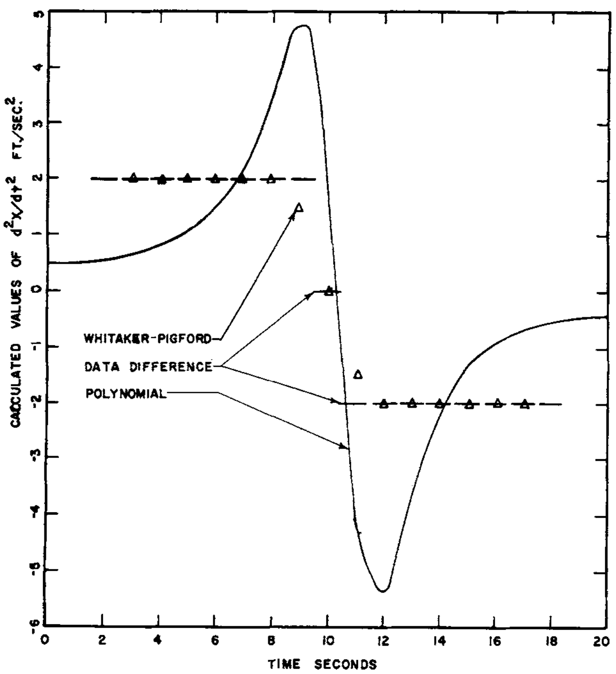

Fig. 6. Calculated second derivatives for discontinuous derivative example.

THIRD EXAMPLE

To illustrate the problem of obtaining second derivatives from integral data when the second derivative is discontinuous, a set of values was generated from the equations

$$\text{at } t = 0,\ x = 0, \text{ and } dx/dt = 0$$

$$\frac{d^2x}{dt^2} = 2\,\frac{\text{ft.}}{\text{sec.}} \text{ for } 0 \leq t \leq 10 \text{ sec.}$$

$$= -2\,\frac{\text{ft.}}{\text{sec.}} \text{ for } 10 < t \leq 20 \text{ sec.} \qquad (8)$$

The set of values gives x at twenty equal time intervals from 1 to 20 sec.

These values were then fit with a fifth-order polynomial of the form

$$t = a_o + a_1x \ldots + a_5x^5 \qquad (9)$$

(This gave a better fit, that is, lower residuals than $x = a_o + a_1t$, etc.) These values were also represented by the Whitaker-Pigford method. The second derivatives calculated by the polynomial, Whitaker-Pigford, and data difference methods are shown in Figure 6.

All three methods indicate a sharp change in the second derivative at $t = 10$. The data difference and Whitaker-Pigford methods do so without distorting the derivative in the rest of the data range. The polynomial fit, which asserts in advance that a jump discontinuity in the second derivative does not exist, distorts the derivative over the entire data range to make its assertion come true.

STATISTICAL APPROACHES

As shown above, all methods of calculating derivatives from integral data involve some smoothing before the calculation of derivatives. There are well-known statistical methods for deciding when one of a series of measurements can be rejected (*17*); it is pertinent to inquire if these can guide the above smoothing process. These statistical methods are based on comparing several measurements of the same value; they are not directly applicable to a table of x-y data, because in such a table each value is measured only once. Any comparison to be made is not of multiple measurements of a single value but of one measurement relative to preceding and following measurements of some other value.

Whitaker and Pigford (*15*) showed with their method that, if one assumes: (1) that the fitting function used was the correct functional form of the data, (2) that the differences between the fitting function and the experimental data were due to random errors which follow a Gaussian distribution, and (3) that this distribution was the same for each point, then one can calculate a standard deviation of the first and second derivatives at every point. Then, using Student's t table, one may calculate the uncertainty in the derivatives corresponding to any required confidence level. They showed that for the example in their paper, the uncertainty in the first derivative at the 99% confidence level is $\pm$ 6%. If one continues their example to compute the second derivative, he sees that at the same confidence level the second derivative has an uncertainty of $\pm$ 200%.

Although this method of estimating the uncertainties of derivatives is useful as a caution against placing too much reliance on such a derivative, it is limited by the strong assumptions which are required to make such a calculation.

CONCLUSIONS

The problem of obtaining first and second derivatives from integral data is important in many rate process studies. Various methods of determining first and second derivatives from such data have been examined by means of three examples. Based on these examples, it appears:

1. Given a table of x-y data, the values of the derivatives computed from this table are a function of the method used to compute them. There is no method which automatically gives correct derivatives.
2. The first derivatives obtained by any of the methods are approximately equal. The second derivatives obtained by the various methods agree only if the data are very precise (that is, free from random or round-off error) and if the various methods fit the data within the limits of its precision.
3. If discontinuities exist in the second derivatives, the power series method will indicate this but will distort the entire second derivative in so doing. The other two methods will also indicate the discontinuity but without distorting the remaining part of the data.

THE PHILOSOPHY OF TREATING EXPERIMENTAL DATA

Experimental data like those in Table 1 are generally obtained at great effort and expense. As White and Churchill (*16*) have pointed out, the tendency of the experimenter is to use a data manipulation method which smooths over any experimental errors. The power series representation does this. On the other hand, the user of the data would like to know just how bad the data may be. The data difference method shows this.

Most workers prefer data interpretation schemes which are mechanical; that is, in which one feeds in the integral data and reads out the derivatives without further human intervention. The polynomial and segmented polynomial schemes fulfill this requirement. It is widely believed that these methods are objective and unbiased because the user makes no personal intervention in their application. However, in another sense, they are quite subjective and biased because, in applying them, the user has made strong assumptions (often without knowing it) as to the functional form of his data. If, as in example 3, there is a discontinuous second derivative and the polynomial method is used, the results will be very strongly influenced by the assumption of continuity of the second derivative.

In contrast, in the data difference method, the subjective, arbitrary parts of the procedure are all in plain view and must be made by the conscious intervention of the data interpreter. No a priori assumptions are made about the functional form of the data. The assumptions are made only after the data have been subjected to the mathematical manipulations which reveal its flaws.

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NOTATION

$a_0 \ldots a_n$ = constants in power series fitting equation
$A_1 \ldots A_3$ = constants in Whitaker-Pigford fitting equations
P = pressure
r = residual = (observed value − calculated value)
t = time
V = volume per mole
x = distance or some unspecified coordinate
y = some unspecified coordinate
θ = angular distance

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