# Optimization by the Method of Contour Tangents

**DOUGLASS J. WILDE**

**University of Texas, Austin, Texas**

The chemical engineer is constantly concerned with improving the performance of chemical manufacturing processes. He is in fact charged with finding those values of the pertinent variables which render the chosen criterion of performance as nearly optimal as possible, while respecting whatever constraints there may be on the independent variables.

When the dependence of the criterion of performance $y$ on the independent variables $x_i$ is known quantitatively, the optimum can be sometimes found by mathematical techniques. But in many practical situations this dependence is so complicated mathematically, so approximate owing to simplifying assumptions, or so obscured by imprecise physical and economic data, that only by a direct experiment on the full size system itself can the effects of any particular conditions be evaluated adequately. In such circumstances one is forced to search for the optimum, using the results of past experiments to decide where to locate future ones.

Search problems are quite widespread and so difficult to handle in any rational manner that a large part of present chemical engineering theory has been developed precisely in order to avoid direct experimentation on the system. But it is not often that one can predict, without complicated calculations, the exact consequences of any set of operation conditions. To find the optimum one usually has to search for it, so this article is devoted to the study of efficient search for an optimum.

Extremely effective techniques for finding a peak when there is only one independent variable and no experimental error have been discovered independently by Kiefer (*1*) and Johnson (*2*). The basic idea is to perform a sequence of experiments in such a way that each trial eliminates a portion of the line segment being searched. In this manner the exploration is confined to successively smaller sections until the final interval of uncertainty is quite small. The one-dimensional character of this problem has made it easy to define plausible measures of effectiveness for comparing search procedures. This in turn has led to identification of the best (minimax) searching plan, which, as Kiefer and Johnson have shown, is based on the Fibonacci sequence.

It has not yet been possible to extend the elegant results for the one-dimensional case to problems with two or more independent variables. Research on this more complicated problem has in fact taken an entirely different tack. Box and Wilson (*3*) suggested directing searches along rising paths until the summit is reached. The pattern search technique of Hooke and Jeeves (*4*) also involves climbing an ascending path, although the method for determining the direction and step size of the search is quite different from that of Box and Wilson. Unlike the one-dimensional problems, measures of effectiveness have not yet been defined for these multidimensional procedures, though this should not discourage their use. They are certain to converge eventually to the optimum, and they can be used even in the presence of considerable experimental error.

It will be helpful in subsequent discussion to use Box's response surface concept (*5*), in which one pictures the dependent variable as plotted vertically above a (hyper) plane whose points represent possible experiments. With this geometric representation one may classify search techniques according to whether they use global properties of the surface or not. A global property, as distinct from a local one, is a characteristic shared by all points on the response surface. For example slope would be a local property because it varies from point to point, while convexity would be a global characteristic.

When there is but one independent variable, the response surface becomes a plane curve. The powerful Kiefer-Johnson technique is based on the global property of unimodality, which means, roughly speaking, that the curve has only one hump. On the other hand the multidimensional procedures cited use only local properties measured in the neighborhood of each successive block of experiments. This is not surprising because the global concept of unimodality, although it can be extended to the multidimensional case, is not strong enough to be used to eliminate regions on a multidimensional response surface.

It would seem promising to develop multidimensional elimination techniques taking advantage of whatever global properties one might reasonably assume. It has been shown previously (*6*) that if a response surface is unimodal on every vertical cutting plane, then when experimental error is small one may indeed devise an elimination technique. Since the suggested procedure involved the same local measurements as did the climbing methods described above, it was pointed out that with little effort one could combine ascent with elimination. The procedure depended on the fact that no matter where the local measurements were taken, the summit could not be below the tangent to the contour passing through the point.

In that article the author's attitude towards multidimensional elimination was rather cautious, and recommendations extended only to the two-dimensional case. Since then how to make fuller use of the elimination concept has been found, and the techniques to be described here are applicable in principle to any multidimensional search problem which is relatively free from experimental error. In this article the author shall make more precise the strong unimodality conditions under which elimination is possible and suggest effective locations for successive blocks of local experiments. As its name implies the contour tangent method uses (hyper) planes tangent to the response surface to cut down the size of the region being searched. The contour tangent, which will be defined precisely here, is not affected by changes of scale. Hence the experimenter is not faced with the sometimes perplexing problem of selecting scales of measurement (*3*, p. 4, *6*, *7*, *8*). Moreover since the contour tangent technique is based on global properties, no extrapolation of local characteristics is involved in going from one block of experiments to another.

Contour tangent methods locate the next block of experiments according to the shape of the remaining experimental region, since nothing is assumed known about the behavior of the dependent variable itself. For this reason the contour tangent methods are definite, always giving a precise location for the next block. There are however

Douglass J. Wilde is with Stanford University, Stanford, California.

many different rules for locating a block, each rule being the most effective according to some different measure of searching efficiency. Hence although each technique is well-defined, there is more than one technique, none of which can claim uncontestable superiority over the others. Four plausible rules and the measures of effectiveness on which they are based will be discussed. Because the contour tangent method does not involve climbing, there could be temporary setbacks in which a block of experiments called for gives a yield lower than for preceding experiments. The use of pilot experiments to avoid this situation and still reduce the region of uncertainty effectively is shown.

Since writing the preliminary version of this article, the author has learned of the work of Buehler, Shah, and Kempthorne (7, 8, 9) which also involves contour tangents and global properties. Their technique, which they call the *method of parallel tangents*, involves neither climbing nor elimination. They show how to reach the top of a response surface having concentric elliptical contours after a small fixed number of experiments. The method of parallel tangents is quite effective, although its optimality property of course limited by the requirement that the contours be concentric ellipses.

The method of contour tangents on the hypothetical and rather complicated response surface, whose contours are shown in Figure 1, will be illustrated. In the example the final area of uncertainty is, after five blocks of three experiments, about 3.2% of the original area, which is close to the reduction factor of $2^5$ which one might expect. An area this small could be explored quite effectively with the quadratic methods proposed by Box (3).

It is customary for the proponent of a new technique to compare it with others already in use. Unfortunately the method of steepest ascent, which is the principal existing multidimensional search technique, is at present a controversial subject among statisticians. Therefore this paper shall confine itself to simply describing the contour tangent method and leave all comparisons to the reader.

## THE CONTOUR TANGENT

Let $y$ be a continuous function of $n$ independent variables $x_1, x_2, \ldots, x_n$, and consider any particular point $\mathbf{x}$ at which $y$ has the value $Y$. The partial derivatives $\partial y/\partial x_i$ $(i = 1, 2, \ldots n)$ evaluated at the point are constants whose values will be written $m_i$. The contour passing through $\mathbf{x}$ is the surface on which $y$ maintains the same value $Y$ as at $\mathbf{x}$. The hyperplane tangent to this contour at $x$ has the equation

$$y - Y = \Sigma m_i \Delta x_i \qquad (1)$$

where

$$x_i = x_i - X_i \qquad (2)$$

$X_i$ is the value of $x_i$ at $\mathbf{x}$. This hyperplane is a linear approximation to the contour surface in the neighborhood of $\mathbf{x}$. The values of the $x_i$ for which $\Sigma m_i \Delta x_i > 0$ are such that $y > Y$ on the tangent hyperplane. Thus the contour tangent, defined as the set of points for which

$$\Sigma\, m_i\, \Delta\, x_i = 0 \qquad (3)$$

separates higher values of $y$ from lower ones on the tangent plane. Geometrically speaking the contour tangent is the intersection of the tangent hyperplane with the horizontal hyperplane $y = Y$. When $n = 2$ the contour tangent is a straight line as in Figure 1.

Both the Box-Wilson method of steepest ascent and the Hooke-Jeeves pattern search technique require the measurement, or at least estimation, of the slope coefficients $m_i$ at the point $(X_1, \ldots, X_n)$ by a block of local experiments. Thus either method is based on obtaining the information needed to describe the contour tangent, although neither technique uses the contour tangent itself to locate the next block of experiments. The procedure to be described here employs the contour tangent directly.

The set of all possible points $\mathbf{x}$ at which it is possible to perform an experiment will be called the *experimental region*. This region is usually bounded by known physical limitations on the range of the independent variables. Often this large region can be reduced further by eliminating parts of it known in advance not to contain the peak. Any contour tangent separates the experimental region into two subregions, one for which $\Sigma m_i \Delta x_i$ is greater than zero and one where the sum is less than zero. It will be shown that for certain commonly occurring functions one may eliminate the latter subregion from all further consideration and thus reduce the size of the experimental region to be explored further. To do this one must define certain global properties characteristics of the response surface which hold for every point on it.

## STRONG UNIMODALITY

Let $\mathbf{r}$ represent any point on a response surface, and designate the summit of the surface by $\mathbf{r}^*$. Any continuous curve on the surface connecting $\mathbf{r}$ to $\mathbf{r}^*$ will be called a *path* from $\mathbf{r}$ to $\mathbf{r}^*$. A path will be described as strictly rising if and only if the value of the criterion $y$ always increases as one moves along the path toward the summit $\mathbf{r}^*$.

Suppose that from every point on the response surface there is at least one strictly rising path to the peak. Then it is said that the surface is unimodal, that is has but one hump. Unimodality is a global property because its definition is based on a characteristic shared by every point on the surface. Since the method of steepest ascent and the pattern search technique both involve climbing rising paths, their ultimate success in reaching the summit can be guaranteed only on unimodal surfaces.

The contour tangent technique proposed will not work on all unimodal surfaces; it requires a slightly stronger global property. A surface is said to be strongly unimodal if the straight line from any point $\mathbf{r}$ to the peak $\mathbf{r}^*$ is a strictly rising path. Although strong unimodality is not as general a property as ordinary unimodality, most unimodal response surfaces are also strongly unimodal. This is fortunate because strong unimodality is necessary to establish the following theorem on which the contour tangent technique is based.

## ELIMINATION THEOREM

For every point $\mathbf{r}$ on a strongly unimodal response surface the summit $\mathbf{r}^*$ must be on the high side of the contour tangent where

$$\Sigma\, m_i\, \Delta\, x_i > 0 \qquad (4)$$

To prove this consider the straight line between $\mathbf{r}$ and the peak $\mathbf{r}^*$. This line must be a strictly rising path because the surface has been assumed strongly unimodal. In the immediate vicinity of $\mathbf{r}$ the change in $y$ is given by $dy = \Sigma m_i dx_i$, since the $m_i$ are the partial derivatives at $\mathbf{r}$. This change must be positive on a strictly rising path. Hence all strictly rising paths and therefore the peak $\mathbf{r}^*$ itself can only be on the high side of the contour tangent. This fact permits us to eliminate from further exploration the entire region in which $\Sigma m_i \Delta x_i \leq 0$.

In general, as in Figure 1, there will be points in the region eliminated that are higher than $\mathbf{r}$, just as there may be lower points in the region remaining. The important thing is that the summit $\mathbf{r}^*$ be in the reduced experimental region.

## EXPERIMENTAL ERROR

If there is no uncertainty in the derivatives $m_i$, which is the case when experimental error is absent, Equation

(3) will bound the region to be explored further. When on the other hand experimental error cannot be neglected, one must choose a confidence level (say 99%) and only eliminate those points where there is less than one chance in a hundred that Equation (3) will not be satisfied. Experimental error will certainly complicate the mathematics and markedly decrease the efficiency of the search, and only the case where there is no uncertainty in the derivatives $m_i$ will be considered here.

## LOCATING SUBSEQUENT BLOCKS

Each time a region has been eliminated it is necessary to decide where to put the next block of experiments. Clearly the next block should be somewhere well inside the eligible region, so that the next contour tangent will eliminate a significant portion. In fact if successive searches are always made at interior points, the region of uncertainty can be made as small as desired, and the strong unimodality of $y$ guarantees that eventually the maximum will be reached.

Although it is comforting to know that any search sequence using interior points will converge in the long run, one is usually more interested in getting a high value of the response $y$ as early as possible. Since nothing is known about $y$ except that it is strongly unimodal, past values of $y$ cannot guide the search. However something is known about the independent variables $x_i$, namely the region of uncertainty remaining to be explored. Thus one may either try to get as close as possible to the point $\mathbf{r}^*$ at which $y$ is maximum or else attempt to make the region of uncertainty as small as possible. Either criterion leads one to locate each new block somewhere in the middle of the remaining region of uncertainty, because a point on the boundary risks either being quite far from $\mathbf{r}^*$ or giving a contour tangent with which only a tiny region can be eliminated.

The block location problem will be illustrated with an example. Figure 1 shows yield from a mathematical model of a hypothetical chemical reactor as a function of operating temperature and pressure. The response surface is strongly unimodal. Notice that the contours are not necessarily oval and are in fact rather complicated. It is assumed that the experimenter knows nothing about the response surface except that the peak lies somewhere between 400° and 600°F. and between 4 and 9 atm. Experimental error is neglected, a reasonable procedure in the study of a mathematical model too complicated to be optimized by such direct methods as the calculus.

The first block of experiments, denoted in Figure 1 by the circled numeral 1, is located in the center of the original rectangular experimental region. The block consists of enough experiments (at least one more than the number of independent variables, three in this case) to determine the equation of the contour tangent in the vicinity of the block. This tangent, together with the region it eliminates, is shown in Figure 1. The trapezoidal area remaining is of course only half as large as that of the original rectangle.

The second block is located at the center of volume of the trapezoid, where a new contour tangent is determined. Location of the center of volume, as well as three other possible center points, will be described in the sections following. For the moment it is only necessary to notice that again approximately half of the remaining region has been eliminated, leaving only about a quarter of the original area. In this example the procedure is continued until five blocks (at least fifteen experiments) are used. Since each block reduces the area of uncertainty by a factor of approximately 2, one would expect the final area to be about one thirty-second (3.1%) of that of the original region. The final region actually remaining is 3.2% of the original, even though the contour tangents lie at many different angles.

Notice that blocks 4 and 5 give yields of less than that at block 3. This is not surprising because the blocks have been placed so as to cut down the region of uncertainty as rapidly as possible, not at all taking account of the yields obtained. It is possible, at the cost of a few extra experiments, to have each block give a yield at least as good as that obtained in previous blocks while still reducing the region of uncertainty in an effective manner. The details of locating successive blocks in the middle of the experimental region will now be discussed.

Four interior points, all more or less in the middle of the region of uncertainty, will be discussed. None of the four points are affected by changes of scale of the independent variables. In the special case where the region of uncertainty is rectangular, all the points will in fact coincide. These points will be in the interior of any convex region, that is one which has the property that any straight line connecting two points on the boundary will lie entirely inside or on the boundary of the region. Since any region bounded entirely by hyperplanes is convex, after several blocks of experiments the remaining region of uncertainty will tend to be convex. And even when the region is not convex, in practice the four points more often than not will be in the interior.

The four points will be referred to respectively as the *midpoint* $\mathbf{x}^{\cdot}$, the *minimax point* $\mathbf{x}^m$, the *center of volume* $\hat{\mathbf{x}}$, and the *centroid* $\bar{\mathbf{x}}$. Each of them can be considered optimal according to some criterion of effectiveness. By the same token however none of them can claim to be uncontestably the best. This discussion will be limited therefore to their definitions and to the exposition of their various advantages and disadvantages.

### The Midpoint

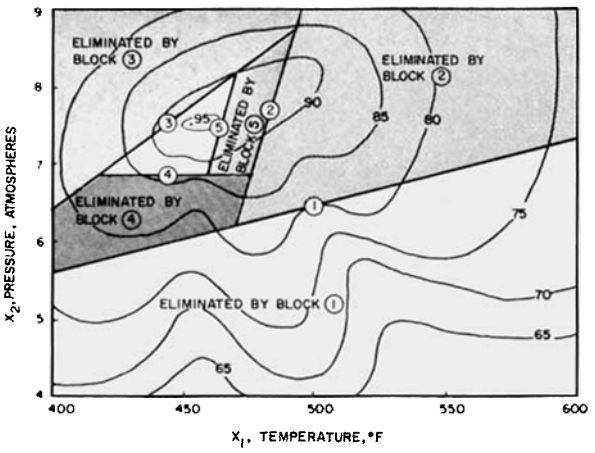
The midpoint $\mathbf{x}$, by far the most easy of the four to calculate, is at the same time based on the most cautious measure of effectiveness. Let $s_i$ and $t_i$ respectively be the minimum and maximum values assumed by the variable $x_i$ in the region of uncertainty $s_i = \min x_i$ and $t_i = \max x_i$. The midpoint $\mathbf{x}^{\cdot}$ is halfway between the extremes. That is for all $i$ one obtains

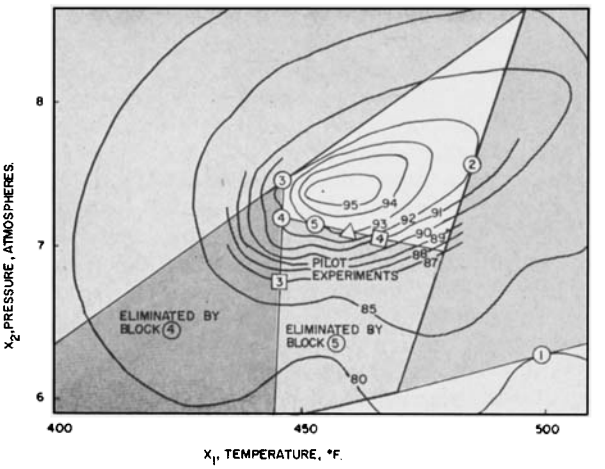
$$x^{\cdot}_i = (s_i + t_i)/2 \qquad (5)$$

The midpoint is quite easy to calculate and seems a reasonable place to locate the next block when the region has no long protuberances. It is however perhaps too middle-of-the-road, being unduly influenced by the extreme points and not at all by points in the middle of the region. Although the midpoint is invariant under changes of scale, it will in general move if the coordinate system is rotated. Even though rotations are not well-defined unless the variables all have the same dimensions, the sensitivity of the midpoint to rotations is unsatisfying, if only from an aesthetic viewpoint.

### The Minimax Point

Suppose that instead of ignoring all but the extreme points, as in the preceding analysis, one considers all points to be of equal importance regardless of their location. Then the hypervolume of the region of uncertainty remaining after the next block of experiments should be as small as possible. To make this idea precise let $\mathbf{x}$ be a typical interior point, and consider an arbitrary hyperplane passing through this point so as to divide the region of uncertainty into two subregions whose respective hypervolumes will depend on the location of $x_i$ and on the constants $m_i$. Let these two hypervolumes be represented by $v_1(\mathbf{x}, m_i)$ and $v_2(\mathbf{x}, m_i)$ with $v_1(\mathbf{x}, m_i) \leq v_2(\mathbf{x}, m_i)$. The minimax point $\mathbf{x}^m$ is where the




largest possible hypervolume is as small as possible:

$$v(\mathbf{x}^m) \equiv \min_{\mathbf{x}} \max_{m_i} \{v_2(\mathbf{x}, m_i)\} \quad (6)$$

The minimax point is insensitive not only to scale changes, but unlike the midpoint $\bar{\mathbf{x}}$, to rotations of the axes as well. Furthermore it is possible in principle to predict with certainty just what the largest possible volume remaining would be after a given number of blocks located at successive minimax points. This remainder would be an a priori measure of the effectiveness of a minimax search in a given region. There is no such a priori measure available for any of the other three points discussed in this article, nor for any of the search techniques already considered.

The minimax point has a major disadvantage, it would be quite difficult to calculate for regions of even mildly complicated shape. At present then the minimax point is mainly of academic rather than of practical interest.

#### The Center of Volume

The first measure of effectiveness involved distances only to the extreme points of the region, while the second did not use the concept of distance at all. A point is now proposed where the mean distances to every point of the region, as measured parallel to each of the axes, are minimum. Let $A_i(\mathbf{x})$ be the hyperarea of the intersection of the region with any hyperplane for which $x_i$ is constant for one of the components. The components $x_i$ of the point $x$ must satisfy

$$\int_a^{\hat{x}_i} A_i(x_i)\, dx_i = \int_{\hat{x}_i}^b A_i(x_i)\, dx_i \quad (7)$$

Since the two sides of this equation are the hypervolumes of the subregions bounded by the hyperplane for which $x_i = \hat{x}_i$, the point $\hat{\mathbf{x}}$ will be called the *center of volume*.

Although the center of volume usually will be easier to calculate than the minimax point, the computations will still be difficult, often requiring trial and error. This is because the $\hat{x}_i$ appear as limits of a definite integral. In spite of the computation problems the center of volume is not entirely devoid of interest. For most configurations occurring in practice the contour tangent passing through the center of volume will divide the region into two nearly equal subregions. Thus it often should be pretty close to the minimax point itself, which has the advantages already discussed. Although the center of volume is not invariant to rotation of the axes, it does remain insensitive to scale changes. Each contour tangent in Figure 1 passes through the centers of volume of the remaining region of uncertainty.

#### The Centroid

The components $\bar{x}_i$ of the centroid $\bar{\mathbf{x}}$ are given by

$$\bar{x}_i = \int_a^b x_i A_i(x_i)\, dx_i \Big/ \int_a^b A_i(x_i)\, dx_i \quad (8)$$

It is well-known that the centroid is invariant, not only to scale changes but to rotations as well. The calculations needed to find the centroid, involving as they do only evaluations of definite integrals, are much easier than for either the minimax point or the center of volume.

### CONTINUATION OF THE SEARCH

Suppose now that a point **b** in the middle of the region has been selected from among the four suggested, and let **a** represent the point where the old block of experiments was located. The next thing to do is conduct a single experiment, not an entire block, at **b**. If the value of $y$ at **b** is greater than that at **a**, one feels safe in locating the next block in the vicinity of **b**, determining a new contour tangent, and continuing the exploration.

But what should be done if $y$ turns out to be less at **b** than at **a**? In Figure 1 for example the yield drops from 92 to 87% between blocks 3 and 4. Fortunately the fact that the line between **a** and **b** starts out as a rising path permits one to locate the next block at an improved level of $y$ and still be sure that the new contour tangent will contain the point **b**, thus preserving whatever advantages **b** might have for reducing the size of the region of uncertainty. Since there must be a point between **a** and **b** giving a higher response than at **b**, it seems reasonable to try another experiment inside this interval before one determines a new contour tangent. If the new response is still less than at **a**, one can move closer to **a** and increase $y$ even further. Eventually by this procedure a point will be found with a higher response than the original one at **a**.

There is in fact an advantage in seeking out a point which is at least locally maximum on the line between **a** and **b**. Let **c** represent such a local maximum. Along the line the $x_i$ simultaneously change in such a way that at **c**, $dy = 0$. Now the contour tangent at **c** is precisely the hyperplane on which $dy = 0$ at **c**. Hence the line between **a** and **b** must lie in the contour tangent hyperplane at **c**, which of course implies that the contour tangent passes through **b**. One sees then that by searching out a local maximum on the line one finds not only a point with high response but one whose contour tangent will contain **b** and be therefore highly effective in cutting down the remaining region of uncertainty.

In summary if the response at **b** is higher than at **a**, locate the next block at **b** without further ado. If not then the author suggests searching out a local maximum on the line so that the next block will give good values of this response and still be effective in reducing the region of uncertainty.

This procedure is illustrated in Figure 2 in which the center of the area of uncertainty remaining after block 3 is indicated by the square marked 3'. The first pilot experiment, marked by a circle 4, gives a yield of 92%, the same as at 3. Since this suggests immediately that there must be a high point in the portion between 3 and this experiment, the fourth block is located at point 4. Since there are in this case only two independent variables, the new contour tangent is a line, identical with that between 3 and 3'. The pilot experiment, shown as point 5, gives an increase (93%), indicating the need for a second, indicated by a triangle. Its yield being also 93% there must be a high point between these two pilot experiments. Block 5 is located arbitrarily at the first.

## CONCLUSIONS

The contour tangent search method is an elimination technique of the Kiefer-Johnson type rather than a climbing method of the sort proposed by Box and Wilson. The proposed method can be modified so that it will climb, or at least not descend. Fundamental to the practical success of the contour tangent method is that the response surface be strongly unimodal as defined in the article and that the experimental error be small. The technique is not affected by the scales of measurement selected, and the need for extrapolation is avoided by exploiting the global property of strong unimodality.

## NOTATION

$A_i(x_i)$ = hyperarea (element of integration)
$\mathbf{a}, \mathbf{b}$ = fixed points in experimental region
$m_i$ = $i$th slope coefficient
$\mathbf{r}$ = point on response surface
$\mathbf{r}^*$ = summit of response surface
$s_i$ = minimum value of $i$th independent variable
$t_i$ = maximum value of $i$th independent variable
$v(\mathbf{x})$ = maximum hypervolume
$v(\mathbf{x}^m)$ = minimax hypervolume
$V$ = hypervolume
$\mathbf{x}$ = variable point in experimental region
$\mathbf{x}^c$ = midpoint
$\mathbf{x}^m$ = minimax point
$\hat{\mathbf{x}}$ = center of volume
$\bar{\mathbf{x}}$ = centroid
$y$ = dependent variable

## LITERATURE CITED


1. Kiefer, J., *Proc. Am. Math. Soc.*, **4**, 502 (1953).
2. Johnson, S. M., "Rand Corporation Report P-856," Santa Monica, California (1956).
3. Box, G. E. P., and K. B. Wilson, *J. Roy. Stat. Soc.*, **B13**, 1 (1951).
4. Hooke, R., and T. A. Jeeves, *J.A.C.M.*, **8**, 2, 212-229 (April, 1961).
5. Box, G. E. P., *Biometrics*, **10**, 16 (1954).
6. Wilde, D. J., "Advances in Chemical Engineering," Vol. 3, pp. 286-292, T. B. Drew, J. W. Hoopes, and T. Vermeulen, ed., Academic Press, New York (1962).
7. Buehler, R. J., B. V. Shah, and O. Kempthorne, "Statistical Laboratory Technical Report No. 1," Office of Naval Research Contract Nonr-530(05) NR-042-207, Iowa State Univ., Ames, Iowa (April, 1961).
8. *Ibid.*, Report No. 2 (April, 1961).
9. *Ibid.*, Report No. 3 (September, 1961).



*Manuscript received January 2, 1962; revision received July 25, 1962; paper accepted July 30, 1962. Paper presented at A.I.Ch.E. Los Angeles meeting.*


# Dispersion in Laminar Flow by Simultaneous Convection and Diffusion


**M. A. FARRELL and E. F. LEONARD**

**Columbia University, New York, New York**


Recent interest in residence time distributions has developed from a recognition of the effects which these distributions can have on the performance of chemical reactors and contacting units. Efficient operation of such equipment often requires that the holding times be confined to a narrow range. Knowledge of the way in which established fluid-mechanical conditions affect residence time distributions is needed for improvement of current design methods.

This paper considers one of a class of residence time distribution function models which has not previously received attention, multidimensional diffusion-convection models which cannot be simplified to equivalent one-dimensional models. The specific model chosen for study is the transient convection-diffusion encountered during laminar flow in round pipes, but the method of treatment is applicable to any flow-diffusion situation in ducts of constant cross section through which a fluid is flowing with a known, steady velocity profile. The attack differs from earlier residence time distribution function studies in retaining the inherent two-dimensional nature of the diffusion-convection process. The results, unlike those of previous studies, are not always equivalent to simple one-dimensional models, and they are capable of explaining differences between observed residence time distribution functions in similar systems and the predictions of these simple models.

The residence time or holding time distribution function is perhaps best defined as the fraction of material introduced into a system at a time $t = 0$ which appears at the outlet of the system between $t$ and $t + dt$ (8). The residence time distribution function may also be regarded as a probability distribution or as the response of a flow system to a unit impulse input. When a system is not unidimensional, the fraction of material leaving the system at any time must generally be determined by integration over a surface normal to the direction of outflow as indicated in Equation (1):

$$f(t) = \frac{\int_A [V(A)C(A) - D_m \nabla C] dA}{Q} = \frac{\overline{V} A C_{mo}}{Q} \quad (1)$$

## PREVIOUS RESIDENCE TIME DISTRIBUTION FUNCTION STUDIES

Studies of the relationship between fluid mechanical conditions and the residence time distribution function have taken two opposite points of view: either known fluid mechanical conditions have been used to predict the residence time distribution function, experimental measurements of which have been used to confirm the analysis, or residence time distribution function measurements have been used