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A COMPREHENSIVE MODEL TO DETERMINE THE EFFECTS OF TEMPERATURE AND SPECIES FLUCTUATIONS ON REACTION RATES IN TURBULENT REACTING FLOWS*

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1. Summary

The use of probability theory to determine the effects of turbulent fluctuations on reaction rates in turbulent combustion systems is briefly reviewed. Results are presented for the effect of species fluctuations in particular. It is found that turbulent fluctuations of species act to reduce the reaction rates, in contrast with the temperature fluctuations previously determined to increase Arrhenius reaction rate constants.[4,6,8,9] For the temperature fluctuations, a criterion is set forth for determining if, in a given region of a turbulent flow field, the temperature can be expected to exhibit ramp-like fluctuations. Using the above-described results, along with results previously obtained[4,6,8,9], a model is described for testing the effects of turbulent fluctuations of temperature and species on reaction rates in computer programs dealing with turbulent reacting flows. An alternative model which employs three-variable probablity density functions (temperature and two species) and is currently being formulated is discussed as well.

2. General Theory

2.1 Probability Density Functions

A parameter x is said to be a continuous random variable if there exists a probability density function, p(x), which satisfies the following conditions [1]:

$$p(x) \geq 0 \tag{1a}$$

$$\int_{-\infty}^{+\infty} p(x)dx = 1 \tag{1b}$$

The pdf may, of course, be defined on an interval other than $(-\infty,+\infty)$, for example (0,1). Since the pdf is defined on a specific interval, the functional value of p(x) is zero elsewhere.

Equation (1) must be satisfied by a pdf of a one-dimensional continuous random variable. Probability density functions may be written for a multi-dimensional continuous random variable. A two-dimensional continuous random variable, for example, is comprised of two one-dimensional continuous random variables. A probability density function for a two-dimensional continuous random variable, denoted p(x,y), is termed a bivariate or joint pdf. For such a pdf, the conditions corresponding to equation (1) are:

$$p(x,y) \geq 0 \tag{2a}$$

$$\int_{-\infty}^{+\infty}\int_{-\infty}^{+\infty} p(x,y)\,dx\,dy = 1 \tag{2b}$$

The expected, or mean value of a one-dimensional continuous random variable, x, is expressed as:

$$\mu_x = E(x) = \int_{-\infty}^{+\infty} x\,p(x)\,dx \tag{3}$$

Equation (3) is also termed the first moment about the origin. In general, the k^{th} moment about the origin of a one-dimensional continuous random variable, x, is expressed as [1]:

$$\mu_{x,k} = \int_{-\infty}^{+\infty} x^k p(x)\,dx \tag{4}$$

where $k = 1,2,3,...$

For the case of k=1, equation (3) is equal to equation (4).

The variance of one-dimensional continuous random variable, x, is expressed as:

$$\sigma_x^2 = V(x) = \int_{-\infty}^{+\infty} (x - E(x))^2 p(x)\,dx \tag{5}$$

A useful quantity derived from the variance is the standard deviation which is the square root of the variance. Equation (5) is the special case of the more general expression for the k^{th} central moment, or moment about the mean, of a one-dimensional continuous random variable, x [10]:

$$\mu'_{x,k} = \int_{-\infty}^{+\infty} (x - E(x))^k p(x)\,dx \qquad (6)$$

where k = 1,2,3,...

Clearly, equation (6) indicates that the first central moment (k=1) is zero. The second central moment (k=2) is termed the variance. For the case of k=2, equation (6) is equal to equation (5). Higher central moments (k>2) are often used in probability theory to give further descriptions of a particular pdf under consideration. For example, the third central moment (k=3), termed the skewness, is used to describe the symmetry or skewness of a pdf. The fourth central moment (k=4), termed the kurtosis or flatness factor, is used to measure the "flatness" of a pdf.

The concept of "moments" of a one-dimensional continuous random variable may be extended to multi-dimensional continuous random variables. For example, the k^{th} joint moment about the origin of a two-dimensional continuous random variable, (x,y), is expressed as [2]:

$$\mu_{xy,k} = \int_{-\infty}^{+\infty}\int_{-\infty}^{+\infty} x^s y^r p(x,y)\,dx\,dy \qquad (7)$$

where $k = s+r$, the order of the moment
$s = 1,2,3,\ldots$
$r = 1,2,3,\ldots$

By comparison, equation (7) is seen to be an extension of equation (4).

The k^{th} joint moment about the mean of a two-dimensional continuous random variable, (x,y), is expressed as [2]:

$$\mu'_{xy,k} = \int_{-\infty}^{+\infty}\int_{-\infty}^{+\infty} (x-\bar{x})^s (y-\bar{y})^r p(x,y)\,dx\,dy \qquad (8)$$

where $k = s+r$, the order of the moment
$s = 1,2,3,\ldots$
$r = 1,2,3,\ldots$

By comparison, equation (8) is seen to be an extension of equation (6).

For a specified joint pdf, it is possible to examine the distribution of any of the one-dimensional components of a multi-dimensional continuous random variable by consideration of its marginal pdf. For a joint pdf, $p(x,y)$, the marginal pdf of x, for example, is given by:

$$h(x) = \int_{-\infty}^{+\infty} p(x,y)\,dy \qquad (9)$$

The marginal distribution of x may be thought of as the distribution of x, with the simultaneous behavior of the other variable(s) suppressed. In other words, only the behavior of x is being examined.

Utilizing the concept of a marginal pdf, the k^{th} moment about the origin of any of the one-dimensional components of a multi-dimensional continuous random variable may be expressed. For a two-dimensional continuous random variable (x,y), the k^{th} moment about the origin of x, for example, is given by:

$$\mu_{x,k} = \int_{-\infty}^{+\infty}\int_{-\infty}^{+\infty} x^k p(x,y)dx\, dy \tag{10}$$

where k = 1,2,3,...

Examination of equations (9) and (10) reveals that the k^{th} moment about the origin of x is expressed as the integral from $-\infty$ to $+\infty$, with respect to x, of the product of x and its marginal pdf.

Equation (3) may be extended to functions of a continuous random variable. In the case of a function f(x) of a one-dimensional random variable having a pdf p(x), the mean value of f(x) is:

$$\overline{f(x)} = \int_{-\infty}^{+\infty} f(x)p(x)\, dx \tag{11}$$

Similarly, in the case of a function g(x,y) of a two-dimensional random variable, having the joint pdf p(x,y), the mean value of g(x,y) is:

$$\overline{g(x,y)} = \int_{-\infty}^{+\infty}\int_{-\infty}^{+\infty} g(x,y)\, p(x,y)dx\, dy \quad , \tag{12}$$

The correlation coefficient, ρ_{xy}, is a parameter defined for the two-dimensional continuous random variable (x,y) as:

$$\rho_{xy} = \frac{E\{[x - E(x)][y - E(y)]\}}{\sqrt{V(x)V(y)}} \tag{13}$$

The correlation coefficient is a measure of the degree of linearity between x and y. Values of the correlation coefficient near +1 or -1 reflect a high degree of linearity, while values of the correlation coefficient near zero indicate a lack of linearity. Positive values of the correlation coefficient indicate that as y increases, x increases. Negative values of the correlation coefficient indicate that y increases as x decreases.

The numerator of equation (13) is defined as the covariance of x and y. The covariance is denoted by σ_{xy} and expressed as:

$$\sigma_{xy} = \overline{x'y'} = \int_{-\infty}^{+\infty}\int_{-\infty}^{+\infty}(x-\bar{x})(y-\bar{y})p(x,y)dx\,dy \qquad (14)$$

By comparison of equation (10) and (8), the covariance is seen to be the second joint moment about the mean. The significance of the covariance can be ascertained by considering a two-dimensional random variable (x,y). The x and y are termed independent random variables if the value of x has no influence on the value of y (and likewise, the value of y has no influence on the value of x). When x and y are independent random variables, the covariance is zero. Hence, the covariance may be considered as a minimum "criterion" of statistical dependence. This criterion can assure, at the very least, that when the covariance is not zero, the variables are not independent. However, no statement can be made concerning independence, on this basis alone, if the covariance is zero [3].

2.2 Most-Likely Bivariate pdf for Two Species

A model which accounts for the combined effects of temperature and species concentration fluctuations on the mean turbulent reaction rate is reviewed in this section. In this model, the temperature is assumed to be statistically independent of the species concentrations [Model II, ref. 4, p. 22].

Mathematically, this three-variable pdf for temperature and species is expressed as:

$$p(t,r_A,r_B) = f(t)\,g(r_A,r_B), \qquad 0 \leq t \leq 1, \quad 0 \leq r_A \leq 1, \quad 0 < r_B \leq 1 \tag{15}$$

where $f(t)$ = a pdf for temperature
$g(r_A,r_B)$ = a joint pdf for the concentrations of A and B species

Equation (15) is a valid pdf since it satifies the following extension of equations (2a) and (2b) for a three-variable pdf:

$$p(t,r_A,r_B) \geq 0 \tag{16a}$$

$$\int_0^1 \int_0^1 \int_0^1 p(t,r_A,r_B)\,dt\,dr_A\,dr_B = 1 \tag{16b}$$

The most-likely bivariate pdf is utilized as the joint pdf for the concentrations of any two species in this model. This pdf is selected on the basis of the excellent agreement between the most-likely bivariate pdf, based on three moments, and an experimentally-measured pdf of concentrations in turbulent non-reacting flow, as discussed in Reference [5]. The three moments considered are: the first moment about the origin of each species concentration, the second joint moment about the mean (i.e., the covariance, k=2 in equation (8)), and the third joint moment about the mean (k=3 in equation (8)). In addition, this pdf is selected on the basis of its potential for increased accuracy in the modeling of the joint pdf for species through the incorporation of higher moments. As an initial step in the utilization of the most-likely bivariate pdf for two species, the most-likely bivariate pdf based on the first moment about the origin of each species concentration and the covariance of the two concentrations, is selected. The expression for the chosen form of the most-likely bivariate pdf for two species is:

$$g(r_A, r_B) = q \cdot \exp(\lambda_0 + \lambda_1 r_A + \lambda_2 r_B + \lambda_3 r_A r_B) \qquad (17)$$

where q = *a priori* probability
r_A = dimensionless concentration of A
r_B = dimensionless concentration of B

The continuous random variables in equation (17) are treated as passive scalars. With this simplification, the "q" term in equation (17) is a constant, set equal to unity for convenience in this study. The value of this constant "q" term is shown in Appendix C to have no effect on the calculated value of the most-likely bivariate pdf and the values of the mean quantities calculated with it.

The values of the constant coefficients, $\lambda_0,\lambda_1,\lambda_2,\lambda_3$, in equation (17) are obtained from the simultanious solution of the following constraint equations for known values of $\overline{r}_A$, $\overline{r}_B$ and $\overline{r'_A r'_B}$:

$$\int_0^1 \int_0^1 g(r_A,r_B)dr_A\, dr_B = 1 \tag{18}$$

$$\int_0^1 \int_0^1 r_A\, g(r_A,r_B)dr_A\, dr_B = \overline{r}_A \tag{19}$$

$$\int_0^1 \int_0^1 r_B\, g(r_A,r_B)dr_A\, dr_B = \overline{r}_B \tag{20}$$

$$\int_0^1 \int_0^1 (r_A-\overline{r}_A)(r_B-\overline{r}_B)g(r_A,r_B)dr_A\, dr_B = \overline{r'_A r'_B} \tag{21}$$

A typical term in a reaction rate equation for species A would be

$$\dot{w}_A^{(1)} = k(t)(r_A C_{A,max})(r_B C_{B,max}) \tag{22}$$

For the case of statistical independence between temperature and species concentration, the expression for the mean turbulent reaction rate

$$\overline{\dot{w}}_t^{(1)} = \{-A \int_0^1 (k_1 t+k_2)^B \exp[-T_A/(k_1 t+k_2)]f(t)dt\} \cdot\{C_{A,max}C_{B,max} \int_0^1 \int_0^1 r_A r_B\, g(r_A,r_B)dr_A\, dr_B\} \tag{23}$$

The corresponding value of the "laminar" reaction rate is determined by inserting the appropriate values of $\bar{t}$, $\bar{r}_A$ and $\bar{r}_B$ into equation 22. This yields:

$$\dot{w}_\ell^{(1)} = -A\ (k_1\bar{t}+k_2)^B \exp[-T_A/(k_1\bar{t}+k_2)]\bar{r}_F C_{F,max}\bar{r}_O C_{o,max} \qquad (24)$$

The ratio of equations (23) and (24) is

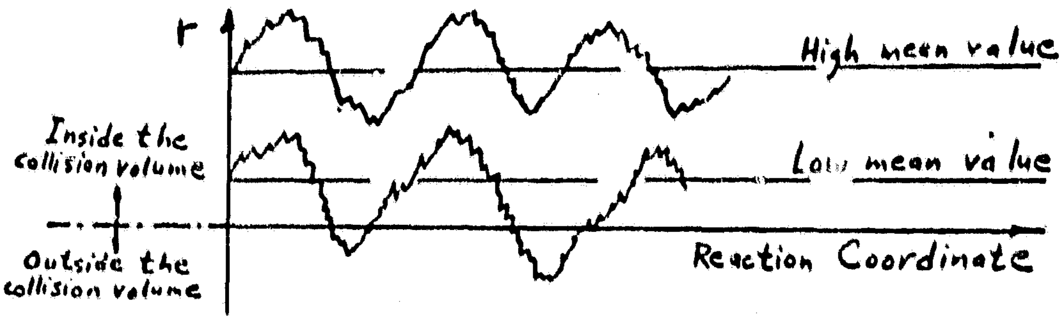
$$Z = \frac{\{{}_0\int^1 (k_1 t+k_2)^B \exp[-T_A/(k_1 t+k_2)]f(t)dt\}\ \{{}_0\int^1{}_0\int^1 r_A r_B\ g(r_A,r_B)dr_A dr_B\}}{(k_1\ \bar{t}+k_2)^B \exp[-T_A/(k_1\bar{t}+k_2)]\ \bar{r}_A\ \bar{r}_B} \qquad (25)$$

This ratio may be expressed as the product of a term which accounts for the effects of temperature fluctuations, Z_t, and a term which accounts for the effects of species concentration fluctuations, Z_r. These terms are expressed as follows:

$$Z_t = \frac{{}_0\int^1 (k_1 t+k_2)^B \exp[-T_A/(k_1\bar{t}+k_2)]f(t)dt}{(k_1\bar{t}+k_2)^B \exp[-T_A/(k_1\bar{t}+k_2)]} \qquad (26)$$

$$Z_r = \frac{{}_0\int^1\ {}_0\int^1\ r_A r_B g(r_A,r_B)dr_A\ dr_B}{\bar{r}_A\bar{r}_B} \qquad (27)$$

The reaction rate "amplication ratio" Z_t has been determined previously[4,6,8,9]. The species "unmixedness factor" Z_r was determined using the numerical methods and considerations described in Appendices A, B, and C. Results are presented in Figures 1a thru 1i assuming a correlation coefficient of -0.9[4]. As may be seen, species unmixedness predominates (low Z_r) when the mean concentrations of the species are low and turbulent fluctuations are high. This is to be anticipated on physical grounds; that is, as shown in the accompanying sketch, lower mean concentrations result in an increased probability that one or both species will be outside the collision volume within which reaction is possible (for the same intensity of fluctuations).



Also as anticipated, increasing fluctuations intensity $\overline{r'^2}$ leads to increased unmixedness.

The pdf selected for temperature is either the beta pdf or the temperature ramp-induced pdf, based upon the criteria discussed in Section 3. The complete model, which incorporates the bivariate pdf formulation discussed in this Section, is detailed in Section 4.

3. Development of a Criterion for the Selection of the Beta or Temperature Ramp-Induced PDF

3.1 Experimental results from the literature

The motivation for developing a criterion for selecting either the ramp pdf or beta pdf lies in observed experimental results. Temperature fluctuations that exhibit a ramp-like structure have been observed experimentally under certain flow conditions, and more importantly, the flatness factor of these temperature fluctuations lies within a narrow range.

Fiedler[10] studied a heated two-dimensional mixing layer; i.e., the higher velocity stream was heated while the external stream had zero velocity and was at ambient temperature. A turbulent heated jet with a coaxial flowing stream was investigated by Antonia, Prabhu, and Stephenson[11]. LaRue and Libby[12], and Gibson, Chen, and Lin[13] independently studied the turbulent flow behind a heated cylinder. Ramp-like temperature fluctuations were observed in all four investigations. Even though these experiments were performed in non-reacting flows, it is assumed in this study that these results can be generalized to reacting flows as a result of the wide diversity of flow conditions under which ramp-like temperature fluctuations were observed. Gibson et al.[13] believe that this type of fluctuation is due to sharp thermal gradients, with the point of highest temperature of the ramp occurring at either the downstream or upstream end of the turbulent/irrotational interface, depending upon the sign of the vorticity of the main flow.

Temperature signals obtained in these experiments are reproduced in figure 2 from the original papers[10,11,13]. Note the similarity of the ramp-like structures even though they were obtained under different flow situations. The flatness factor distributions obtained by three of the researchers as they appeared in the original papers, are shown in figure 3[10,11,12].

The abscissa η of all graphs in figure 3 is a normalized radial distance, with each author using a different normalizing constant.

The general trend of these curves is that the flatness factor has a value of approximately 3 (the Gaussian value) near the centerline, decreases to a value near 2, then sharply increases in the other region of the flowfield.

This trend led Fielder[10] to suggest that the value of 2 is characteristic of the "sawtooth" appearance of the temperature signal. Antonia et al.[11] subsequently agreed with Fiedler's. conclusion. This suggests that the flatness factor may be used as a criterion for the selection of either the beta or ramp pdf.

4.2 An initial selection procedure

It was pointed out in references 4 and 6 that the ramp pdf is only capable of generating flatness factor values between 1.0 and 3.7. Of the four adjustable constants in the ramp pdf, the flatness factor is most sensitive to variations in σ^*. Since it is so sensitive, an assumption (to be tested subsequently) will be to assign σ^* a value based upon physical grounds. Antonia and Atkinson[7] indicate that a value of $\sigma^* = 0.25$ yields good agreement with experimental results. As a consequence of fixing σ^*, varying α', β, and c over the ranges previously indicated[4] generates flatness between 2.0 and 2.6. This suggests that the ramp pdf is applicable when F falls within this range.

Before proceeding any further, one should ascertain whether this criterion agrees with experimental results. Antonia, Prabhu, and Stephenson (ref 11, p. 477) state that ramp-like behavior was observed in the region $0.6 \leq \eta \leq 1.2$. The range of flatness factors suggested by this study, where the ramp pdf is applicable, is 2.0

to 2.6. If the normalized radial distance corresponding to this range of F is obtained from figure 35, one can see that $0.65 \leq \eta \leq 1.2$. This agrees closely with the range within which Antonia et al.[11] observed ramp-like temperature fluctuations. This agreement supports the contention that the ramp pdf is applicable when $2.0 \leq F \leq 2.6$. (This implicitly justifies the assumption of constant σ^*.)

An initial criterion would then be to determine if F, at a point in the flowfield, lies between 2.0 and 2.6. If it does, then the ramp pdf is applicable at that point. If it does not fall within that range, the beta pdf is to be used.

However, recall that the beta pdf generates the values $1.66 < F < 8.64$. Since the beta pdf is continuous, every intermediate value can be obtained by the proper selection of $\bar{t}$ and $\overline{t'^2}$. This implies that the beta pdf generates the values 2.0-2.6 for the flatness factor (as does the ramp pdf). As a result, the flatness factor cannot be used as the sole criterion due to this overlapping.

The skewness also overlaps, but it is distinct when $2.0 \leq F \leq 2.6$. For the ramp pdf, $-1.57 \leq S \leq -1.25$, and for the beta pdf, $-0.82 \leq S \leq +0.82$ (both when $2.0 \leq F \leq 2.6$). See table 1.

The selection procedure is as follows: if the flatness factor lies outside the range 2.0 to 2.6, then always select the beta pdf. If the flatness factor lies within this range, examine the skewness value. If $-1.57 \leq S \leq -1.25$, choose the ramp pdf; otherwise select the beta pdf (see figure 5).

	BETA	RAMP
F	1.66 ⋮ 2.0 ↓ 2.6 ⋮ 8.64	2.0 ↓ 2.6
S	-0.82 ↓ 0.82	-1.57 ↓ -1.25

Table 1: Summary of initial criterion between the beta and ramp pdfs

The skewness alone cannot be used as a primary criterion because the full ranges generated by the two pdf's are not distinct.

4.3 An alternative selection process

The selection procedure outlined in the previous section is inconvenient from the computational point-of-view because of the difficulty in determining the third and fourth moments at every point in a flowfield. It would be preferable to use, if possible the first and second moments (mean and standard deviation) instead. This can be achieved by using the available experimental data as follows:

1. On a F vs. η (axial distance) graph, find the range of η which corresponds to $2.0 \le F \le 2.6$
2. On a $\bar{t}$ vs. η graph, find the range of $\bar{t}$ which corresponds to the range of η determined in step (1).
3. Repeat step (2) for $\overline{t'^2}$.

The same ranges of $\bar{t}$ and $\overline{t'^2}$ result from three independent papers[11,12,13] after taking the different normalizing constants into account. The consensus is that if

$$0.35 \le \bar{t} \le 0.75$$

and

$$\overline{t'^2} > 0.08$$

then the temperature fluctuations exhibit a definite ramp-like character. Outside this range, the beta pdf is to be used.

It is to be anticipated that $\overline{t'^2} \ge 0.08$ since the ramp pdf is applicable when the temperature fluctuations are large. It is also to be anticipated that $0.35 \leq \bar{t} \leq 0.75$. If $\bar{t} < 0.35$, then the mean temperature is low, indicating a low reaction rate. This slow rate of reaction will not induce high temperature fluctuations. If $\bar{t} > 0.75$, the mean temperature is high, implying that the magnitude of the fluctuations will be a smaller percentage of the mean than it is at a lower temperature, effectively reducing the effect of the fluctuations. Hence a pdf which describes low intensity fluctuations (e.g., the beta pdf) would be applicable.

4. A Model for Determining Reaction Rates in Turbulent Reacting Flows

In this section, the completed Model II[4] is set forth employing the elements discussed in Sections 2 and 3, as well as those discussed in previous status reports[4,6,8,9].

Consider a general chemical kinetic mechanism, as shown below:

$$A_1 + A_2 \underset{k_{-1}}{\overset{k_1}{\rightleftarrows}} A_3 + A_4$$

$$A_1 + A_5 \underset{k_{-2}}{\overset{k_2}{\rightleftarrows}} A_6 + A_7$$

$$\vdots$$

$$A_1 + M \underset{k_{-n}}{\overset{f_n}{\rightleftarrows}} A_M + A_M + M$$

The "laminar" reaction rate expression for species A_1 (i.e., the source term in the species transport equation for A_1 would be

$$\dot{w}_{A1,\ell} = -k_1 C_{A_1} C_{A_2} + k_{-1} C_{A_3} C_{A_4} - k_2 C_{A_1} C_{A_5} + k_{-2} C_{A_6} C_{A_7}$$

$$- \ldots - k_n C_{A_1} C + k_{-n} C_{Am}{}^2 C \qquad (28)$$

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In accordance with the model proposed here, this would be rewritten for a turbulent reacting flow

$$\dot{w}_{A1,t} = -(\Xi_t\ k_1)\ (\Xi_{r,1-2}\ C_{A_1}\ C_{A_2}) + (k_c/\Xi_t\ k_1)\ (\Xi_{r,3-4}\ C_{A_3} C_{A_4}) - \ldots \quad (29)$$

where $\Xi_t = \Xi_t\ (\overline{t},\ \overline{t'^2})$ is the temperature "amplification ratio" discussed in references 4, 6, 8, and 9, $\Xi_{r,i-j} = \Xi_{r,i-j}(\overline{r}_i, \overline{r}_j, \overline{r_i'^2}\ \overline{r_j'^2})$ is the species "unmixedness" factor discussed in Section 2 herein, and k_c is the equilibrium constant based upon molar concentrations.

In accordance with the criteria set forth in Section 3, the beta pdf is to be used to determine Ξ_t in all regions of the turbulent flow field <u>except</u> when

$$0.35 \leq \overline{t} \leq 0.75$$

and

$$\overline{t'^2} \geq 0.08 \quad (30)$$

When the conditions of eq. (30) are satisfied, the temperature ramp-induced pdf is to be used as detailed in ref. 9, p. 6.

The species "unmixedness" factor, $\Xi_{r,i-j}$, is determined from the values of $\overline{c}_i$, $\overline{c}_j$, $\overline{c_i'^2}$ and $\overline{c_j'^2}$ obtained from appropriate transport equations. Since $\overline{r}_i = \overline{c}_i/c_{i,max}$, etc., estimates of the maximum molar concentrations are required. For a reactant species (fuel, oxidizer), $c_{i,max}$ is taken to be the initial moler concentration. For a principal product species (eg. H_2O, CO_2, SiO_2), $c_{i,max}$ is taken as the final equilibrium value determined from a preliminary equilibrium calculation. For an intermediate species (eg, OH, O, H), $c_{i,max}$ must be estimated from a preliminary "laminar" kinetics calculation using either the multi-dimensional computer program of interest (eg, CHARNAL, SHIP) or a one-dimensional kinetics program (eg. ref. 14), Once $\overline{r}_i$, $\overline{r}_j$, $\overline{r_i'^2}$ and $\overline{r_j'^2}$ have been determined, $g_{r,i-j}$ is obtained from curve-fits (or table look-up) of the "unmixedness" data presented in Section 2 herein.

The above-described procedure is carried out for all of the terms in eq. (28) and the analogous equations for each species present. It should be noted that this model indicated that the temperature fluctuations and species fluctuations result in opposing effects on the reaction rates. That is, g_t is always greater than unity, while g_r is always less than unity. The behavior of these parameters (Figs. 1a thru 13 herein and, for example, Figs. 6 thru 24 of ref. 8 and Figs. 1 thru 10 of ref. 9) lead to the following conclusions:

1. In regions where the principal constituents are reactant species at relatively low temperature (high $\overline{r}_i$, $\overline{r}_j$, low $\overline{t}$), the principal effect will be through the increased Arrhenius rate constant (beta pdf) due to the temperature fluctuations.
2. In regions of intermediate temperature and species concentrations, the greatly amplified rate constant due to the temperature fluctuations (ramp pdf) competes with the species "unmixedness," which may become substantial.
3. In regions where combustion nears completion (high $\overline{t}$ and $\overline{r}$ for the principal product species), reaction rates will approach their laminar values.

5. An Alternative Three-Variable Model for Determining Reaction Rates in Turbulent Reacting Flows

In this section, a full three-variable model (temperature and two species), currently under development, is discussed. This model was previously referred to as Model III[4]. In this model, individual terms in the reaction rate equations have the form

$$\dot{w}_1^{(1)} = C_{1,max}\, C_{2,max} \int_0^1 \int_0^1 \int_0^1 K(t)\, x_1\, x_2\, p(t_1 x_1, x_2) \cdot dt\, dx_1\, dx_2 \tag{31}$$

where the three-variable most-likely pdf is

$$p(t_1, x_1, x_2) = q \cdot \exp\,(\lambda_0 + \lambda_1 t + \lambda_2 x_1 + \lambda_3 x_2 + \lambda_4 t x_2 + \lambda_5 t x_1 + \lambda_6 x_2 x_1) \tag{32}$$

There are seven constants in this pdf, which can be evaluated by the following seven equations:

$$\begin{aligned}
\int_0^1\!\!\int\!\!\int p(x_1, x_2, t)\, dx_1\, dx_2\, dt &= 1 \\
\int_0^1\!\!\int\!\!\int x_1 \cdot p(x_1, x_2, t)\, dx_1\, dx_2\, dt &= \overline{x}_1 \\
\int_0^1\!\!\int\!\!\int x_2 \cdot p(x_1, x_2, t)\, dx_1\, dx_2\, dt &= \overline{x}_2 \\
\int_0^1\!\!\int\!\!\int t \cdot p(x_1, x_2, t)\, dx_1\, dx_2\, dt &= \overline{t} \\
\int_0^1\!\!\int\!\!\int (x_1 - \overline{x}_1)(x_2 - \overline{x}_2) \cdot p(x_1, x_2, t)\, dx_1\, dx_2\, dt &= \overline{x_1' x_2'} \\
\int_0^1\!\!\int\!\!\int (x_2 - \overline{x}_2)(t - \overline{t}) \cdot p(x_1, x_2, t)\, dx_1\, dx_2\, dt &= \overline{x_2' t} \\
\int_0^1\!\!\int\!\!\int (x_1 - \overline{x}_1)(t - \overline{t}) \cdot p(x_1, x_2, t)\, dx_1\, dx_2\, dt &= \overline{x_1' t'}
\end{aligned} \tag{33}$$

where $\bar{x}$, $\bar{t}$ are the mean values of species concentration and temperature and $\overline{x't'}$, $\overline{x'x'}$ are the covariants of the second central moments. Using Newton's method for systems, the seven non-linear equations can be transformed into seven linear equations and the constants found by iteration. The equations can be rewritten as follows:

$$a(\lambda_0, \lambda_1, \ldots, \lambda_6) = \int_0^1\int\int p(x_1, x_2, t)\, dx_1\, dx_2\, dt - 1 = 0$$

$$b(\lambda_0, \lambda_1, \ldots, \lambda_6) = \int_0^1\int\int x_1 \cdot p(x_1, x_2, t)\, dx_1\, dx_2\, dt - \bar{x}_1 = 0$$

$$c(\lambda_0, \lambda_1, \ldots, \lambda_6) = \int_0^1\int\int x_2 \cdot p(x_1, x_2, t)\, dx_1\, dx_2\, dt - \bar{x}_2 = 0$$

$$d(\lambda_0, \lambda_1, \ldots, \lambda_6) = \int_0^1\int\int t \cdot p(x_1, x_2, t)\, dx_1\, dx_2\, dt - \bar{t} = 0$$

$$e(\lambda_0, \lambda_1, \ldots, \lambda_6) = \int_0^1\int\int (x_1 - \bar{x}_1)(x_2 - \bar{x}_2) \cdot p\, dx_1\, dx_2\, dt - \overline{x_1' x_2'} = 0$$

$$f(\lambda_0, \lambda_1, \ldots, \lambda_6) = \int_0^1\int\int (x_2 - \bar{x}_2)(t - \bar{t}) \cdot p\, dx_1\, dx_2\, dt - \overline{x_2' t'} = 0$$

$$g(\lambda_0, \lambda_1, \ldots, \lambda_6) = \int_0^1\int\int (x_1 - \bar{x}_1)(t - \bar{t}) \cdot p\, dx_1\, dx_2\, dt - \overline{x_1' t'} = 0$$

(34)

The equations to find the lambda's using Newton's method for the three-variable, most-likely pdf are as follows:

$$\lambda_{0_{NEW}} = \lambda_{0_{OLD}} + \frac{\det \begin{vmatrix} -a & b_{\lambda_0} & c_{\lambda_0} & \cdots & g_{\lambda_0} \\ -b & b_{\lambda_1} & \cdots & & \vdots \\ \vdots & \vdots & \vdots & & \vdots \\ -g & b_{\lambda_6} & c_{\lambda_6} & \cdots & g_{\lambda_6} \end{vmatrix}}{\det \begin{vmatrix} a_{\lambda_0} & b_{\lambda_0} & \cdots & & g_{\lambda_0} \\ a_{\lambda_1} & \cdots & & & \vdots \\ \vdots & \vdots & \vdots & & \vdots \\ a_{\lambda_6} & b_{\lambda_6} & \cdots & & g_{\lambda_6} \end{vmatrix}}$$

$$\lambda_{1_{NEW}} = \lambda_{1_{OLD}} + \frac{\det \begin{vmatrix} a_{\lambda_0} & -a & c_{\lambda_0} & \cdots & g_{\lambda_0} \\ a_{\lambda_1} & -b & \vdots & & \vdots \\ \vdots & \vdots & \vdots & & \vdots \\ a_{\lambda_6} & -g & c_{\lambda_6} & \cdots & g_{\lambda_6} \end{vmatrix}}{\det \begin{vmatrix} \text{denominator above} \end{vmatrix}}$$

$$\vdots$$

etc.

(35)

where a_{λ_0} is the partial derivative of equation a with respect to λ_0, etc.

Each of the entries in the matricies are triple integrals, which have to be numerically integrated. The determinant of the matricies can then be evaluated using Gauss elimination, or Gauss-Jordan reduction.

It is shown in Appendix C that for the one-variable and two-variable most-likely pdf's, the turbulent reaction rates are independent of q. As a result, the following two equations for the one- and two-variable pdf's can be written:

$$\int_0^1 k(t)\ p(t)dt = \int_0^1 k(t) \cdot q\ p(t)\ dt \tag{36}$$

$$\int_0^1 k(t)\ p(t,r)dt = \int_0^1 \int_0^1 k(t) \cdot q\ (p(t,r))\ dt \tag{37}$$

Noting that the Arrhenius reaction rate is the same on both sides of equations (36) and (37), if the turbulent reaction rate is constant then the pdf with q=1 must be equal to the pdf with q=q. Writing the constraint equations for the one-variable most-likely pdf for q=1 and q=q alternately, the following pattern results:

$$\begin{aligned}
e^{\lambda_0} \int_0^1 e^{\lambda_1 t + \lambda_2 t^2}\, dt &= 1 \\
q \cdot e^{\lambda_0'} \int_0^1 e^{\lambda_1' t + \lambda_2' t^2}\, dt &= 1 \\
e^{\lambda_0} \int_0^1 t \cdot e^{\lambda_1 t + \lambda_2 t^2}\, dt &= \bar{t} \\
q \cdot e^{\lambda_0'} \int_0^1 t \cdot e^{\lambda_1' t + \lambda_2' t^2}\, dt &= \bar{t} \\
e^{\lambda_0} \int_0^1 (t-\bar{t})^2 e^{\lambda_1 t + \lambda_2 t^2}\, dt &= \overline{t'^2} \\
q \cdot e^{\lambda_0'} \int_0^1 (t-\bar{t})^2 e^{\lambda_1' t + \lambda_2' t^2}\, dt &= \overline{t'^2}
\end{aligned} \tag{38}$$

By numerical solution it was shown for the one-variable most-likely pdf that

$$e^{\lambda_0} = q \cdot e^{\lambda_0'} \;, \quad \lambda_1 = \lambda_1' \;, \quad \lambda_2 = \lambda_2' \quad . \tag{39}$$

This solution indicates that the constants in this pdf are changing in a manner which causes the pdf to remain a constant for all values of q.

A similar pattern emerges when the constraint equations for the two-variable pdf are written with q=1 and q=q alternately:

$$e^{\lambda_0} \int_0^1\!\!\int e^{\lambda_1 t + \lambda_2 x + \lambda_3 tx} \, dt\, dx = 1 \;; \quad e^{\lambda_0} \int_0^1\!\!\int x\, e^{\lambda_1 t + \lambda_2 x + \lambda_3 tx} \, dt\, dx = \bar{x}$$

$$q \cdot e^{\lambda_0''} \int_0^1\!\!\int e^{\lambda_1'' t + \lambda_2'' x + \lambda_3'' tx} \, dt\, dx = 1 \;; \quad q \cdot e^{\lambda_0''} \int_0^1\!\!\int x\, e^{\lambda_1'' t + \lambda_2'' x + \lambda_3'' tx} \, dt\, dx = \bar{x}$$

$$e^{\lambda_0} \int_0^1\!\!\int t\, e^{\lambda_1 t + \lambda_2 x + \lambda_3 tx} \, dt\, dx = \bar{t} \;;$$

$$q \cdot e^{\lambda_0''} \int_0^1\!\!\int t\, e^{\lambda_1'' t + \lambda_2'' x + \lambda_3'' tx} \, dt\, dx = \bar{t} \;;$$

$$e^{\lambda_0} \int_0^1\!\!\int (t-\bar{t})(x-\bar{x})\, e^{\lambda_1 t + \lambda_2 x + \lambda_3 tx} \, dt\, dx = \overline{t'x'} \;;$$

$$q \cdot e^{\lambda_0''} \int_0^1\!\!\int (t-\bar{t})(x-\bar{x})\, e^{\lambda_1'' t + \lambda_2'' x + \lambda_3'' tx} \, dt\, dx = \overline{t'x'} \;. \tag{40}$$

By numerical solution for the two-variable case, it was shown that

$$e^{\lambda_0} = q e^{\lambda_0''} \;, \quad \lambda_1 = \lambda_1'' \;, \quad \lambda_2 = \lambda_2'' \;, \quad \lambda_3 = \lambda_3'' \;. \tag{41}$$

This solution again shows that the constants are varying in a manner which keeps the value of the pdf a constant for any value of q.

To decrease the computer time required to obtain the constants in the three-variable pdf, the triple integrals in the augmented matrix were reduced to double integrals, where the variables are separable. This enables the use of the single variable Simpson's Rule program to evaluate the double integrals, since the integral of one of the variables is a standard function. The augmented matrix used to evaluate the constants in Newton's Method is composed of 49 triple integrals. Of the 49 integrals, 26 are duplicates, leaving 23 integrals that must be evaluated for each iteration.

Because of the cross-product terms in the three-variable pdf equation (32), one change of variables and two Jacobian transforms were utilized to change the 23 triple integrals to double integrals of the following form:

$$\int_{\tau}\int_{\sigma} f(\tau)\,\sigma^{N}\,e^{-(\chi/4)\sigma^{2}}\,d\sigma\,d\tau \qquad (42)$$

where N is an integer between 0 and 5.

When N=0 in equation 42, the integral over σ is a scaled version of the bell-shaped distribution curve. When N=1, this integral over σ can be evaluated explicitly leaving a single integral. When N=3, 4 or 5, the integral over σ can be evaluated by parts. The resulting integrals are a function of the Gaussian distribution. Details will appear in a future status report.

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LIST OF SYMBOLS

c_i	molar concentration of species i
E(x)	expected value of x
F	flatness factor (or kurtosis) of a pdf
h(x)	marginal pdf of x
k	Arrhenius reaction rate constant
k_1	a constant equal to $T_{max} - T_{min}$
k_2	a constant equal to T_{min}
p(x)	probability density function (pdf) of x
q	*a priori* probability in the most-likely pdf
r_i	normalized molar concentration equal to $c_i/c_{i,max}$
S	skewness of a pdf
T_A	activation temperature equal to activation energy divided by the gas constant
t	normalized temperature equal to $(T-T_{min}/(T_{max}-T_{min})$
V(x)	variance of x
$\dot{w}_i$	reaction rate of species i (or a term in the reaction rate expression for species i)
β_r	species "unmixedness" factor
β_t	Arrhenius reaction rate constant amplification ratio
λ_i	constants in the most-likely pdf
ρ_{xy}	correlation coefficient between x and y
μ	Gaussian mean
σ	Gaussian rms fluctuates
η	normalized axial distance

Subscripts

ℓ	laminar
max	maximum value
min	minimum value
t	turbulent

Appendix A

A1 Evaluation of the constant coefficients of the one-variable most-likely pdf.

A1.1 Numerical Procedure

The one-variable most-likely pdf for temperature is given by:

$$p(t) = q \cdot \exp[\lambda_0 + \lambda_1 t + \lambda_2 t^2] \qquad (1A)$$

where q is equal to unity, as discussed in Section 3.1. The constant coefficients of equation (1D), $\lambda_0, \lambda_1, \lambda_2$, are determined from the simultaneous solution of the following constraint equations for known values of $\overline{t}$ and $\overline{t'^2}$:

$$\int_0^1 p(t)dt = 1 \qquad (2A)$$

$$\int_0^1 t\, p(t)dt = \overline{t} \qquad (3A)$$

$$\int_0^1 (t-\overline{t})p(t)dt = \overline{t'^2} \qquad (4A)$$

Since equations (2A), (3A) and (4A) are non-linear, Newton's Method [1A] for a system of non-linear equations is an

1A. Gerald, E. F., Applied Numerical Analysis, Second Edition, Addison-Wesley Publishing Co., Reading, MA, 1978.

appropriate technique of solution. This method transposes the original problem of solving a system of non-linear equations to that of solving a system of linear equations in terms of the "unknown" constant coefficients of equation (1D). This is accomplished as follows:

Equations (2A), (3A), and (4A) are re-written as:

$$F(\lambda_0,\lambda_1,\lambda_2) = F = {}_0\int^1 p(t)dt - 1 = 0 \qquad (5A)$$

$$G(\lambda_0,\lambda_1,\lambda_2) = G = {}_0\int^1 t\, p(t)dt - \bar{t} = 0 \qquad (6A)$$

$$H(\lambda_0,\lambda_1,\lambda_2) = H = {}_0\int^1 (t-\bar{t})^2 p(t)dt - \overline{t'^2} = 0 \qquad (7A)$$

Let $\lambda_0=x_0$, $\lambda_1=x_1$ and $\lambda_2=x_2$ be the solution for the constant coefficients of equation (1A). Let $\lambda_0=\lambda_0^{(1)}$, $\lambda_1=\lambda_1^{(1)}$, and $\lambda_2=\lambda_2^{(1)}$ be a point "near" the solution, (x_0,x_1,x_2). It is possible to expand the functions given by equations (5A), (6A) and (7A) as a Taylor series about the point $(\lambda_0^{(1)},\lambda_1^{(1)},\lambda_2^{(1)})$ in terms of $(x_0-\lambda_0^{(1)})$, $(x_1-\lambda_2^{(1)})$ and $(x_2-\lambda_2^{(1)})$. If $(\lambda_0^{(1)},\lambda_1^{(1)},\lambda_2^{(1)})$ is "near enough" to (x_0,x_1,x_2) it is possible to truncate the Taylor series after the first-derivative terms in order to obtain an approximate solution. This yields:

$$F = F + \sum_{i=0}^{2} F_{\lambda_i}(x_i-\lambda_i^{(1)}) \qquad (8A)$$

$$G = G + \sum_{i=0}^{2} G_{\lambda_i}(x_i-\lambda_i^{(1)}) \qquad (9A)$$

$$H = H + \sum_{i=0}^{2} H_{\lambda_i}(x_i-\lambda_i^{(1)}) \quad (10A)$$

where F_{λ_i} is the partial derivative of F with respect to λ_i. Similarily for G and H. The functions F, G and H, and their associated partial derivatives are evaluated at $(\lambda_0^{(1)}, \lambda_1^{(1)}, \lambda_2^{(1)})$.

Equation (8A), (9A), and (10A) form a system of linear equations. The "unknowns" in these equations are the "improvements" in each approximated variable: $(x_0-\lambda_0^{(1)})$, $(x_1-\lambda_1^{(1)})$, $(x_2-\lambda_2^{(1)})$. These equations may be solved by utilizing any appropriate technique for the solution of systems of linear equations. In the present work, Cramer's Rule is selected. The application of Cramer's Rule to equations (8A), (9A) and (10A) yields the following expressions for the "improvements" in the approximated constants of equation (1A):

$$x_i - \lambda_i^{(1)} = \frac{\det A_j}{\det A}, \qquad \begin{matrix} i = 0,1,2 \\ j = i+1 \end{matrix} \quad (11A)$$

where A = the coefficient matrix of the system of linear equations given by equation (11A)

A_j = the matrix formed by replacing the elements of the j^{th} column of A by $-F$, $-G$, $-H$; $j = 1,2,3$.

det = the determinant of a designated matrix

Matrix A is expressed as:

$$\begin{bmatrix} F_{\lambda_0} & F_{\lambda_1} & F_{\lambda_2} \\ G_{\lambda_0} & G_{\lambda_1} & G_{\lambda_2} \\ H_{\lambda_0} & H_{\lambda_1} & H_{\lambda_2} \end{bmatrix}$$

Solving equations (11A) for the x_i's yields:

$$x_i = \lambda_i^{(1)} + \frac{\det A_j}{\det A^*}, \qquad i = 0,1,2 \quad j = i+1 \tag{12A}$$

For the case of the one-variable most-likely pdf x_0, x_1, x_2 are unknown quantities. Hence, $\lambda_0^{(1)}, \lambda_1^{(1)}, \lambda_2^{(1)}$ are taken as an "initial guess" for the solution. The resulting values for x_0, x_1, x_2 are "improved" solutions. Substitution of x_0, x_1, x_2 for the values of $\lambda_0^{(1)}, \lambda_1^{(1)}, \lambda_2^{(1)}$ yields "further-improved" solutions. The solution process continues in this manner until the desired degree of convergence is achieved.

The expressions for the elements of matrix A are given in Table A1., Section A1.2. All integrations are performed using Simpson's Rule.

*where det A is not equal to zero.

A1.2 Expressions for the elements in matrix A

The expressions for the elements in matrix A are obtained through the application of Liebniz's Theorem for Differentiation of an Integral [2A]. These expressions are given in Table A1.

Table A1. Expressions for the elements of matrix A

Element	Expression
F_{λ_0}	$\int_0^1 p(t)dt$
F_{λ_1}	$\int_0^1 t\, p(t)dt$
F_{λ_2}	$\int_0^1 t^2\, p(t)dt$
G_{λ_0}	$\int_0^1 t\, p(t)dt$
G_{λ_1}	$\int_0^1 t^2\, p(t)dt$
G_{λ_2}	$\int_0^1 t^3\, p(t)dt$
H_{λ_0}	$\int_0^1 (t-\bar{t})^2 p(t)dt$

2A. U. S. Dept. of Commerce, Nat. Bureau of Stand., Handbook of Mathematical Functions with Formulas, Graphs, and Mathematical Tables, AMS 55, Ed. by M. Abramowitz and I. Stegun, December 1972, p. 11.

Table A1. (continued)

Element	Expression
H_{λ_1}	$\int_0^1 t(t-\bar{t})^2 p(t)dt$
H_{λ_2}	$\int_0^1 t^2(t-\bar{t})^2 p(t)dt$

A1.3 Mathematical Limitations

If the "initial guess" for the constant coefficients in equation (1A) is "sufficiently close" to the solution, Newton's Method converges to the solution. If the "initial guess" is not "sufficiently close," this method may diverge from the solution. Difficulty in achieving convergence is frequently encountered with very high and very low values of dimensionless mean temperature, $\bar{t}$ (e.g., $\bar{t}$=0.1, 0.9).

A2. Evaluation of the constant coefficients of the most-likely bivariate pdf

The discussion in this section considers the most-likely bivariate for temperature and species. An analogous discussion applies to the most-likely bivariate pdf for two species.

A2.1 Numerical Procedure

The most-likely bivariate pdf for temperature and species is expressed as:

$$p(t,r) = q \cdot \exp[\lambda_0+\lambda_1 t+\lambda_2 r+\lambda_3 tr] \qquad (13A)$$

where q is equal to unity, as discussed in Section .2. The constant coefficients of equation (13A) are determined from the simultaneous solution of the following constraint equations for known values of $\overline{t}$, $\overline{r}$, and $\overline{t'r'}$:

$$\int_0^1\int_0^1 p(t,r)dt\,dr = 1 \qquad (14A)$$

$$\int_0^1\int_0^1 t\,p(t,r)dt\,dr = \overline{t} \qquad (15A)$$

$$\int_0^1\int_0^1 r\,p(t,r)dt\,dr = \overline{r} \qquad (16A)$$

$$\int_0^1\int_0^1 (t-\overline{t})(r-\overline{r})p(t,r)dt\,dr = \overline{t'r'} \qquad (17A)$$

Since equations (14A) through (17A) are non-linear, Newton's Method for a system of non-linear equations is utilized in the manner of Section A1.1:

Equations (14A) through (17A) are re-written as:

$$J(\lambda_0,\lambda_1,\lambda_2,\lambda_3) = J = \int_0^1\int_0^1 p(t,r)dt\,dr-1 = 0 \qquad (18A)$$

$$K(\lambda_0,\lambda_1,\lambda_2,\lambda_3) = K = \int_0^1\int_0^1 t\,p(t,r)dt\,dr-\overline{t} = 0 \qquad (19A)$$

$$L(\lambda_0,\lambda_1,\lambda_2,\lambda_3) = L = \int_0^1\int_0^1 r\,p(t,r)dt\,dr-\overline{r} = 0 \qquad (20A)$$

$$M(\lambda_0,\lambda_1,\lambda_2,\lambda_3) = M = \int_0^1\int_0^1 (t-\overline{t})(r-\overline{r}) \cdot p(t,r)dt\,dr-\overline{t'r'} = 0 \qquad (21A)$$

Let $\lambda_0=x_0$, $\lambda_1=x_1$, $\lambda_2=x_2$, $\lambda_3=x_3$ be the solution for the constant coefficients of equation (13A). Let $\lambda_0=\lambda_0^{(1)}$, $\lambda_1=\lambda_1^{(1)}$, $\lambda_2=\lambda_2^{(1)}$, $\lambda_3=\lambda_3^{(1)}$ be a point "near" the solution (x_0,x_1,x_2,x_3). It is possible to expand the functions given by equations (18A to (21A) as a Taylor series about the point $(\lambda_0^{(1)},\lambda_1^{(1)},\lambda_2^{(1)},\lambda_3^{(1)})$ in terms of $(x_0-\lambda_0^{(1)})$, $(x_1-\lambda_1^{(1)})$, $(x_2-\lambda_2^{(1)})$, $(x_3-\lambda_3^{(1)})$. If $(\lambda_0^{(1)},\lambda_1^{(1)},\lambda_2^{(1)},\lambda_3^{(1)})$ is "near enough" to (x_0,x_1,x_2,x_3), it is possible to truncate the Taylor series after the first-derivative terms in order to obtian an approximate solution. This yields:

$$J = J + \sum_{i=0}^{3} J_{\lambda_i}(x_i-\lambda_i^{(1)}) \qquad (22A)$$

$$K = K + \sum_{i=0}^{3} K_{\lambda_i}(x_i-\lambda_i^{(1)}) \qquad (23A)$$

$$L = L + \sum_{i=0}^{3} L_{\lambda_i}(x_i-\lambda_i^{(1)}) \qquad (24A)$$

$$M = M + \sum_{i=0}^{3} M_{\lambda_i}(x_i-\lambda_i^{(1)}) \qquad (25A)$$

where J_{λ_i} is the partial derivative of J with respect to λ_i, $i = 0,1,2,3$. Similarly for K, L and M. Functions J, K, L, M and their associated partial derivatives are evaluated at $(\lambda_0^{(1)},\lambda_1^{(1)},\lambda_2^{(1)},\lambda_3^{(1)})$.

Equations (22A), (23A), (24A), and (25A) form a system of linear equations. The "unknowns" in these equations are the "improvements" in each approximated variable: $(x_0-\lambda_0^{(1)})$, $(x_1-\lambda_1^{(1)})$, $(x_2-\lambda_2^{(1)})$, $(x_3-\lambda_3^{(1)})$. These equations are solved by utilizing Cramer's Rule as in Section A1.1. The application of Cramer's Rule to equations (22A), (23A), (24A) and (25A) yields the following expressions for the "improvements" in the approximated constants of equation (13A):

$$(x_i-\lambda_i^{(1)}) = \frac{\det B_j}{\det B^*}, \quad i = 0,1,2,3 \quad j = i+1 \tag{26A}$$

where B = the coefficient matrix of the system of linear equations given by (26A)

B_j = the matrix formed by replacing the elements of the j^{th} column of B by $-J$, $-K$, $-L$, $-M$; $j = 1,2,3,4$.

det = the determinant of a designated matrix

Matrix B is expressed as:

$$\begin{bmatrix} J_{\lambda_0} & J_{\lambda_1} & J_{\lambda_2} & J_{\lambda_3} \\ K_{\lambda_0} & K_{\lambda_1} & K_{\lambda_2} & K_{\lambda_3} \\ L_{\lambda_0} & L_{\lambda_1} & L_{\lambda_2} & L_{\lambda_3} \\ M_{\lambda_0} & M_{\lambda_1} & M_{\lambda_2} & M_{\lambda_3} \end{bmatrix}$$

*where det B is not equal to zero.

Solving equations (26A) for the x_i's yields:

$$x_i = \lambda_i^{(1)} + \frac{\det B_j}{\det B}, \quad \begin{matrix} i = 0,1,2,3 \\ j = i+1 \end{matrix} \tag{27A}$$

For the case of the most-likely bivariate pdf, x_0,x_1,x_2,x_3 are unknown quantities. Hence $\lambda_0^{(1)},\lambda_1^{(1)},\lambda_2^{(1)},\lambda_3^{(1)}$, are taken as an "initial guess" for the true solution. The resulting values for x_0,x_1,x_2,x_3 are "improved" solutions. Substitution of x_0,x_1,x_2,x_3 for the values of $\lambda_0^{(1)},\lambda_1^{(1)},\lambda_2^{(1)},\lambda_3^{(1)}$ yields "further-improved" solutions. The solution process continues in this manner until the desired degree of convergence is achieved.

The expressions for the elements in matrix B are given in Table 2A, Section A2.2. These elements are initially expressed in terms of double integrals. The large amounts of computer time required to solve for the constant coefficients of equation (13A) by utilizing this numerical procedure and the double-integral expressions render this solution impractical. Use of the Monte Carlo integration technique, as opposed to Simpson's Rule, yields no appreciable decrease in the amounts of computer time required. In order to decrease this time requirement, the following method, developed during the course of this study, is utilized:

The double-integral expressions for the elements of matrix B are "reduced" to single integrals by noting, for example, that

$$J_{\lambda_0} = {}_0\int^1 {}_0\int^1 \exp[\lambda_0+\lambda_1 t+\lambda_2 r+\lambda_3 tr]dt\ dr \qquad (28A)$$

may be written as:

$$J_{\lambda_0} = \exp(\lambda_0)\, {}_0\int^1 \exp(\lambda_1 t)\,[{}_0\int^1 \exp(\lambda_2 r+\lambda_3 tr)dr]dt \qquad (29A)$$

Evaluation of the inner integral in equation (29A) yields:

$$J_{\lambda_0} = \exp(\lambda_0)\, {}_0\int^1 \frac{\exp(\lambda_1 t)}{\lambda_2+\lambda_3 t}\,[\exp(\lambda_2+\lambda_3 t)-1]dt \qquad (30A)$$

Similar manipulations are performed with the remaining elements of matrix B. Utilization of the single integral expressions given in Table 3A decreases the amounts of computer time required and renders this solution practical. All integrations are performed with Simpson's Rule.

D2.2 Expressions for the elements in matrix B

The expressions for the elements in matrix B are obtained through the application of Liebniz's Theorem for Differentiation of an Integral [2A]. The double-integral forms of these expressions are given in Table 2A. The single-integral forms of these expressions are given in Table 3A. The following definitions are utilized in Table 2A.

$$X_1 = {}_0\int^1 p(t,r)dr = \exp(\lambda_0+\lambda_1 t)\ \frac{\exp(\lambda_2+\lambda_3 t)-1}{\lambda_2+\lambda_3 t} \qquad (31A)$$

$$X_2 = {}_0\int^1 r\ p(t,r)dr = \exp(\lambda_0+\lambda_1 t) \cdot \frac{(\lambda_2+\lambda_3 t-1)\exp(\lambda_2+\lambda_3 t)+1}{(\lambda_2+\lambda_3 t)^2} \qquad (32A)$$

$$X_3 = {}_0\int^1 r^2\ p(t,r)dt = \frac{\exp(\lambda_0+\lambda_1 t+\lambda_2+\lambda_3 t)}{(\lambda_2+\lambda_3 t)^3} \cdot \left[(\lambda_2+\lambda_3 t)^2 - 2(\lambda_2+\lambda_3 t-1) - \frac{2}{\exp(\lambda_2+\lambda_3 t)}\right] \qquad (33A)$$

Table 2A. Double integral expressions for the elements for matrix B

Element	Expression
J_{λ_0}	${}_0\int^1 {}_0\int^1 p(t,r)dt\ dr$
J_{λ_1}	${}_0\int^1 {}_0\int^1 t\ p(t,r)dt\ dr$
J_{λ_2}	${}_0\int^1 {}_0\int^1 r\ p(t,r)dt\ dr$
J_{λ_3}	${}_0\int^1 {}_0\int^1 tr\ p(t,r)dt\ dr$
K_{λ_0}	${}_0\int^1 {}_0\int^1 t\ p(t,r)dt\ dr$
K_{λ_1}	${}_0\int^1 {}_0\int^1 t^2\ p(t,r)dt\ dr$

Table 2A (continued)

Element	Expression
K_{λ_2}	$\int_0^1 \int_0^1 tr\ p(t,r)dt\ dr$
K_{λ_3}	$\int_0^1 \int_0^1 t^2 r\ p(t,r)dt\ dr$
L_{λ_0}	$\int_0^1 \int_0^1 r\ p(t,r)dt\ dr$
L_{λ_1}	$\int_0^1 \int_0^1 tr\ p(t,r)dt\ dr$
L_{λ_2}	$\int_0^1 \int_0^1 r^2\ p(t,r)dt\ dr$
L_{λ_3}	$\int_0^1 \int_0^1 tr^2\ p(t,r)dt\ dr$
M_{λ_0}	$\int_0^1 \int_0^1 (t-\bar{t})(r-\bar{r})p(t,r)dt\ dr$
M_{λ_1}	$\int_0^1 \int_0^1 t(t-\bar{t})(r-\bar{r})p(t,r)dt\ dr$
M_{λ_2}	$\int_0^1 \int_0^1 r(t-\bar{t})(r-\bar{r})p(t,r)dt\ dr$
M_{λ_3}	$\int_0^1 \int_0^1 tr(t-\bar{t})(r-\bar{r})p(t,r)dt\ dr$

Table 3A. Single integral expressions for the elements in matrix B

Element	Expression
J_{λ_0}	${}_0\int^1 X_1 \, dt$
J_{λ_1}	${}_0\int^1 t \, X_1 \, dt$
J_{λ_2}	${}_0\int^1 X_2 \, dt$
J_{λ_3}	${}_0\int^1 t \, X_2 \, dt$
K_{λ_0}	equal to J_{λ_1}
K_{λ_1}	${}_0\int^1 t^2 \, X_1 \, dt$
K_{λ_2}	equal to J_{λ_3}
K_{λ_3}	${}_0\int^1 t^2 \, X_2 \, dt$
L_{λ_0}	equal to J_{λ_2}
L_{λ_1}	equal to J_{λ_3}
L_{λ_2}	${}_0\int^1 X_3 \, dt$
L_{λ_3}	${}_0\int^1 t \, X_3 \, dt$
M_{λ_0}	$J_{\lambda_3} - \bar{r} \, J_{\lambda_1} - \bar{t} \, J_{\lambda_2} + \bar{t} \, \bar{r} \, J_{\lambda_0}$
M_{λ_1}	$K_{\lambda_3} - \bar{t} \, J_{\lambda_3} - \bar{r} \, K_{\lambda_1} + \bar{t} \, \bar{r} \, J_{\lambda_1}$

Table 3A. (continued)

Element	Expression
M_{λ_2}	$L_{\lambda_3} - \bar{t}\, L_{\lambda_2} - \bar{r}\, J_{\lambda_3} + \bar{t}\, \bar{r}\, J_{\lambda_2}$
M_{λ_3}	${}_0\int^1 t^2\, X_3\, dt - \bar{t}\, L_{\lambda_3} - \bar{r}\, K_{\lambda_3} + \bar{t}\, \bar{r}\, J_{\lambda_3}$

A2.3 Mathematical Limitations

If the "initial guess" for the constant coefficients in equation (13A) is "sufficiently close" to the solution, Newton's Method converges to the solution. If the "initial guess" is not "sufficiently close," this method may diverge from the solution. In this case, no convergence is achieved. Additional difficulties with convergence may arise when, in the denominator of X_1, X_2 or X_3, the value of $(\lambda_2+\lambda_3 t)$ is close to zero

Appendix B

The Uniqueness of the Solutions for the Constant Coefficients of the Most-Likely pdf

The utilization of Newton's Method for the solution of the unknown constant coefficients of the one-variable most-likely pdf and the most-likely bivariate pdf is described in detail in Appendix A. These constants are determined from the solution of the system of non-linear constraint equations for each pdf. The utilization of Newton's Method reduces the problem of solving a system of non-linear equations to that of solving a system of linear equations. The system of linear equations is solved by utilizing Cramer's Rule. Thus, the proof of uniqueness of the solutions obtained by utilizing Cramer's Rule is proof of the uniqueness of the solutions for the constant coefficients of the most-likely pdf. For simplicity, the following discussion demonstrates the uniqueness of the solutions for the constant coefficients of the one-variable most-likely pdf. An analogous discussion applies to the constant coefficients of the most-likely bivariate pdf.

As shown in Appendix A, the solution to the following set of linear constraint equations

$$F = F + \sum_{i=0}^{2} F_{\lambda_i} (x_i - \lambda_i^{(1)}) \qquad (8A)$$

$$G = G + \sum_{i=0}^{2} G_{\lambda_i} (x_i - \lambda_i^{(1)}) \qquad (9A)$$

$$H = H + \sum_{i=0}^{2} H_{\lambda_i} (x_i - \lambda_i^{(1)}) \qquad (10A)$$

is given by

$$x_i - \lambda_i^{(1)} = \frac{\det A_j}{\det A}, \qquad \begin{matrix} i = 0,1,2 \\ j = i+1 \end{matrix} \qquad (11A)$$

for the case where det A is not equal to zero. Equations (11A) are the expressions for the "improvements" in the approximated values of the constant coefficients of the one-variable pdf. Cramer's Rule states that if "det A" is not equal to zero, then the system of linear equations given by equations (8A), (9A), and (10A) has the unique solution given by equations (11A). For a formal proof of this statement, the reader is referred to Reference [1B]. Thus, the solutions for the constant coefficients of both the one-variable most-likely pdf and the most-likely bivariate pdf, obtained with the use of the numerical procedure outlined in Appendix A, are unique.

1B. Gerald, C. F., Applied Numerical Analysis, Second Edition, Addison-Wesley Publishing Co., Reading, MA, 1978.

Appendix C

Variation of the Mean Turbulent Reaction Rate Constant and Mean Turbulent Reaction Rate with the Value of the *a priori* probability, q

The discussion in this appendix utilizes a combination of analytical and numerical calculations to demonstrate that the mean turbulent reaction rate constant obtained with the use of the one-variable most-likely pdf (treating temperature as a passive scalar) is independent of the value of the *a priori* probability, q. An analogous discussion demonstrating that the mean turbulent reaction rate obtained with the use of the most-likely bivariate pdf is independent of the value of the *a priori* probability follows directly from that given for the one-variable most-likely pdf. Hence, the former discussion is not presented.

Let the expression for the one-variable most-likely pdf wherein "q" is a constant, not equal to unity, be given by:

$$f(t) = q\cdot\exp[\lambda_0+\lambda_1 t+\lambda_2 t^2] \tag{1C}$$

Equation (1C) is utilized in Case I of this discussion. Let the expression for the one-variable most-likely pdf wherein q is a constant, equal to unity, be given by:

$$p(t) = \exp[\lambda_0'+\lambda_1' t+\lambda_2' t^2] \quad (2c)$$

Equation (2c) is utilized in Case II of this discussion. The corresponding constraint equations for Cases I and II are given by the following:

<u>Case I</u>

$$\int_0^1 f(t)dt = 1 \quad (3c)$$

$$\int_0^1 t\, f(t)dt = \bar{t} \quad (4c)$$

$$\int_0^1 (t-\bar{t})^2 f(t)dt = \overline{t'^2} \quad (5c)$$

<u>Case II</u>

$$\int_0^1 p(t)dt = 1 \quad (6c)$$

$$\int_0^1 t\, p(t)dt = \bar{t} \quad (7c)$$

$$\int_0^1 (t-\bar{t})^2 p(t)dt = \overline{t'^2} \quad (8c)$$

The following comparison of Case I and Case II is made, utilizing the same values for $\bar{t}$ and $\overline{t'^2}$ for both cases, in order to demonstrate that the one-variable most-likely pdf, the mean turbulent reaction rate constants and, hence, the reaction rate constant amplification ratios determined for each case are the same. Thus:

In order for equations (3c), (4c), and (5c) to be valid simultaneously with equations (6c), (7c) and (8c), then

$$f(t) = p(t) \tag{9c}$$

Equation (9c) is verified by the numerical results obtained with the computer program written for the procedure described in Appendix C. Moreover, equation (9c) demonstrates that the values of the mean turbulent reaction rate constant determined for Cases I and II are identical since both cases utilize identical expressions for the one-variable most-likely pdf. The numerical results of this same computer program indicate that this is so.

The relationships between the constant coefficients of equations (1c) and (2c) may be ascertained from the following:

Equation (9c) indicates that

$$q \cdot \exp[\lambda_0+\lambda_1 t+\lambda_2 t^2] = \exp[\lambda_0'+\lambda_1' t+\lambda_2' t^2] \tag{10c}$$

<u>If</u> $\lambda_1=\lambda_1'$ and $\lambda_2=\lambda_2'$, then

$$q \cdot \exp(\lambda_0)\exp(\lambda_1 t)\exp(\lambda_2 t^2) = \exp(\lambda_0')\exp(\lambda_1' t)\exp(\lambda_2' t^2) \tag{11c}$$

Cancellation of "like terms" in equation (11c) yields:

$$q \cdot \exp(\lambda_0) = \exp(\lambda_0') \tag{12c}$$

Solving equation (12) for λ_0 yields:

$$\lambda_0 = \lambda_0' - \ln(q) \tag{13c}$$

Equation (13C) as well as the fact that $\lambda_1=\lambda_1'$ and $\lambda_2=\lambda_2'$ are verified by the numerical results obtained with the computer program written for the procedure outlined in Appendix A.

Thus, knowledge of the value of the constant *a priori* probability for the most-likely pdf of a passive scalar(s) is not required since the values of both the pdf and the mean quantities calculated with it's use are independent of "q", where "q" is a non-zero constant.

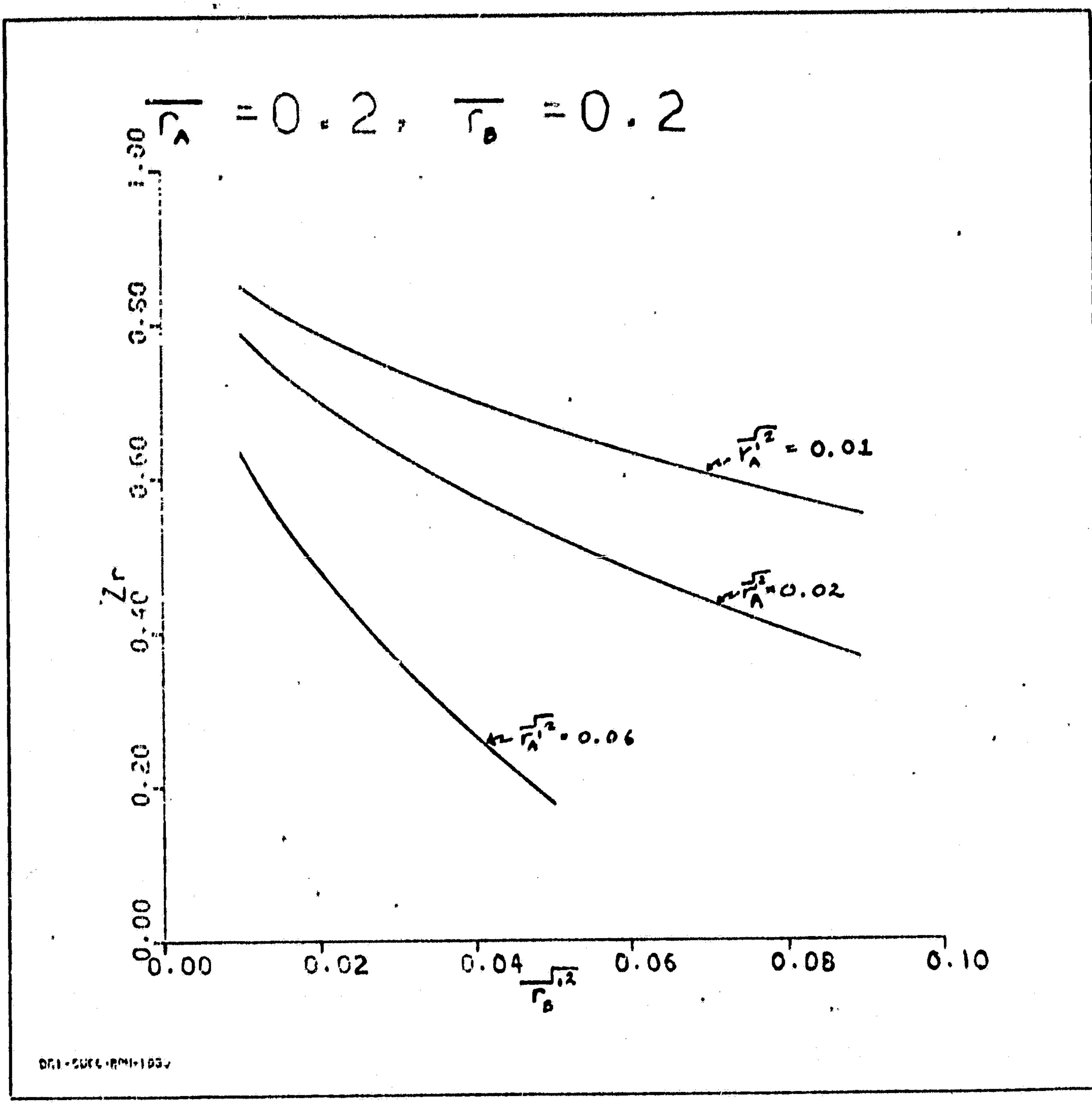

FIG. 1. SPECIES "UNMIXEDNESS" PARAMETER
(a) $\overline{r_A} = 0.2$, $\overline{r_B} = 0.2$

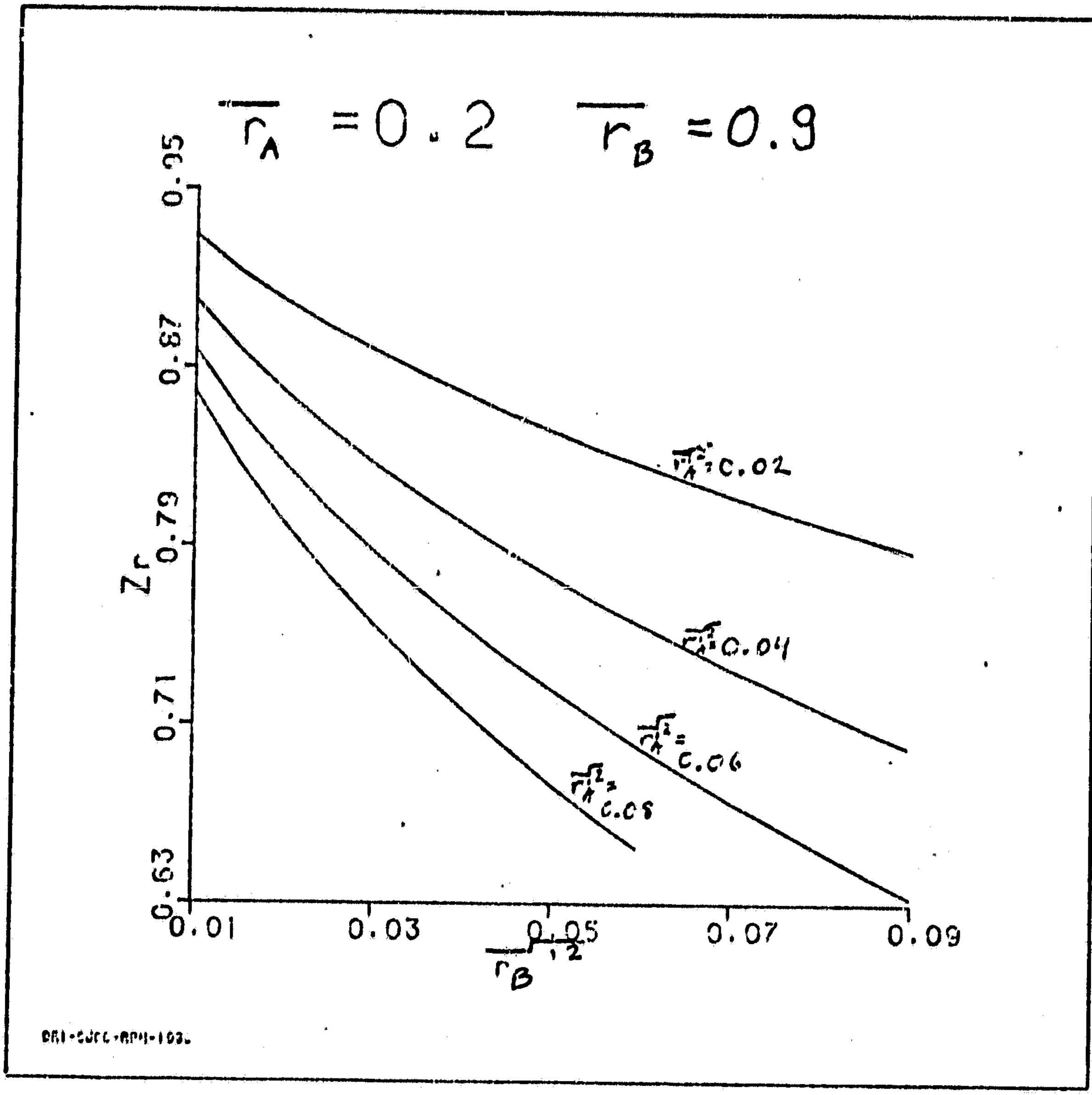

FIG. 1. CONTINUED
(b) $\overline{r_A} = 0.2$, $\overline{r_B} = 0.9$

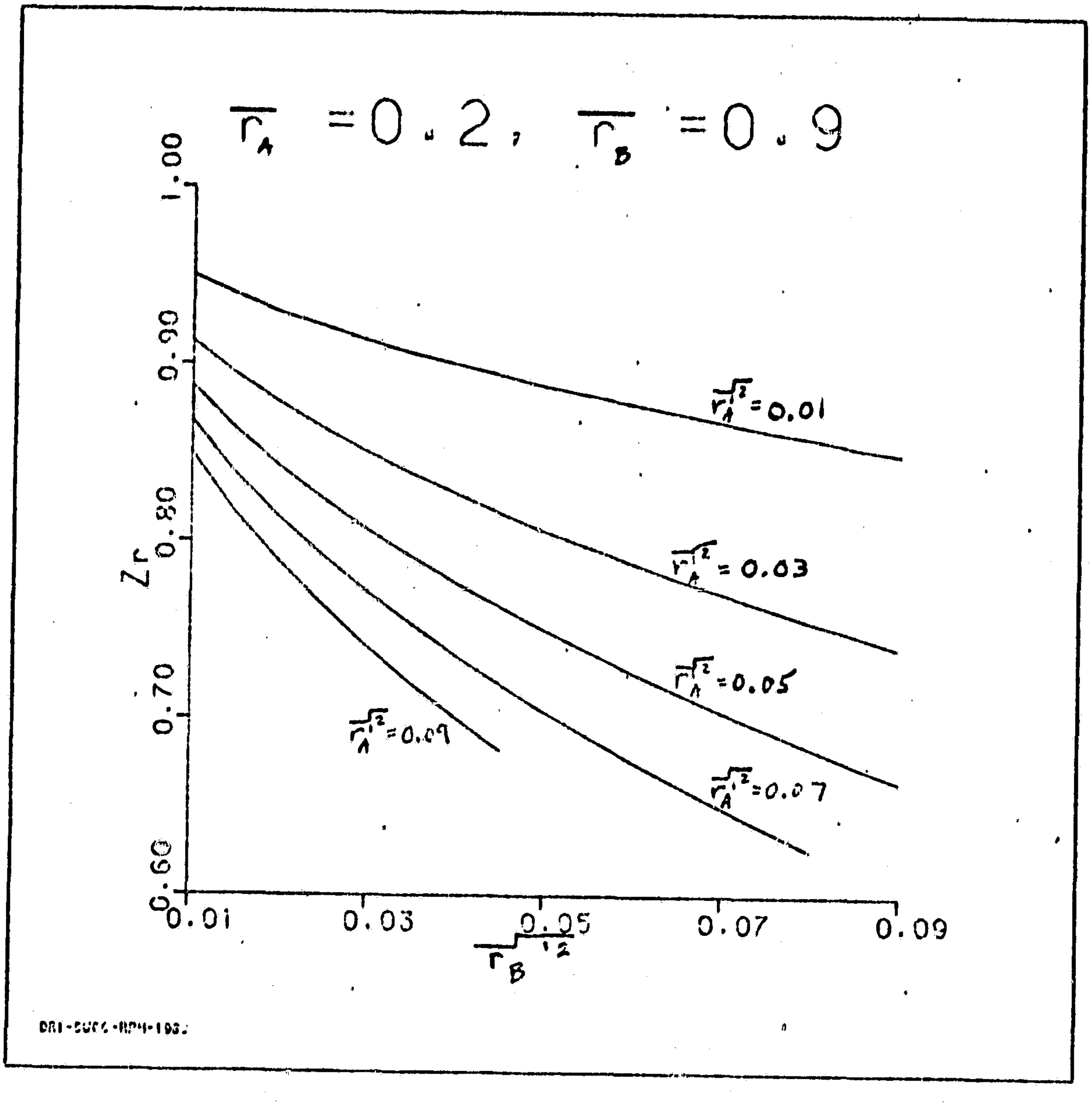

FIG. 1. CONTINUED
(c) $\overline{r_A} = 0.2$, $\overline{r_B} = 0.9$

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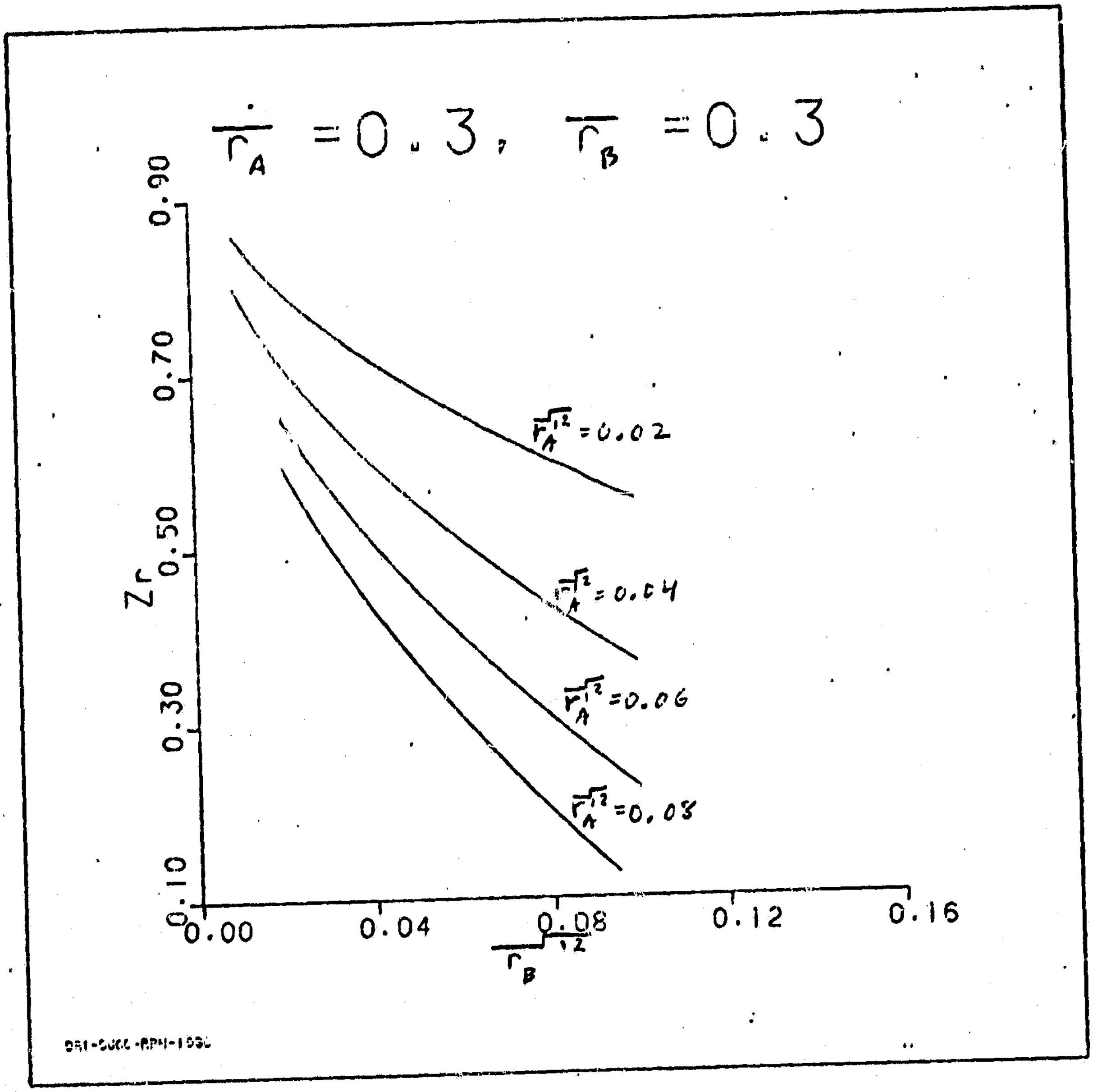

FIG. 1. CONTINUED
(d) $\overline{r_A} = 0.3, \overline{r_B} = 0.3$

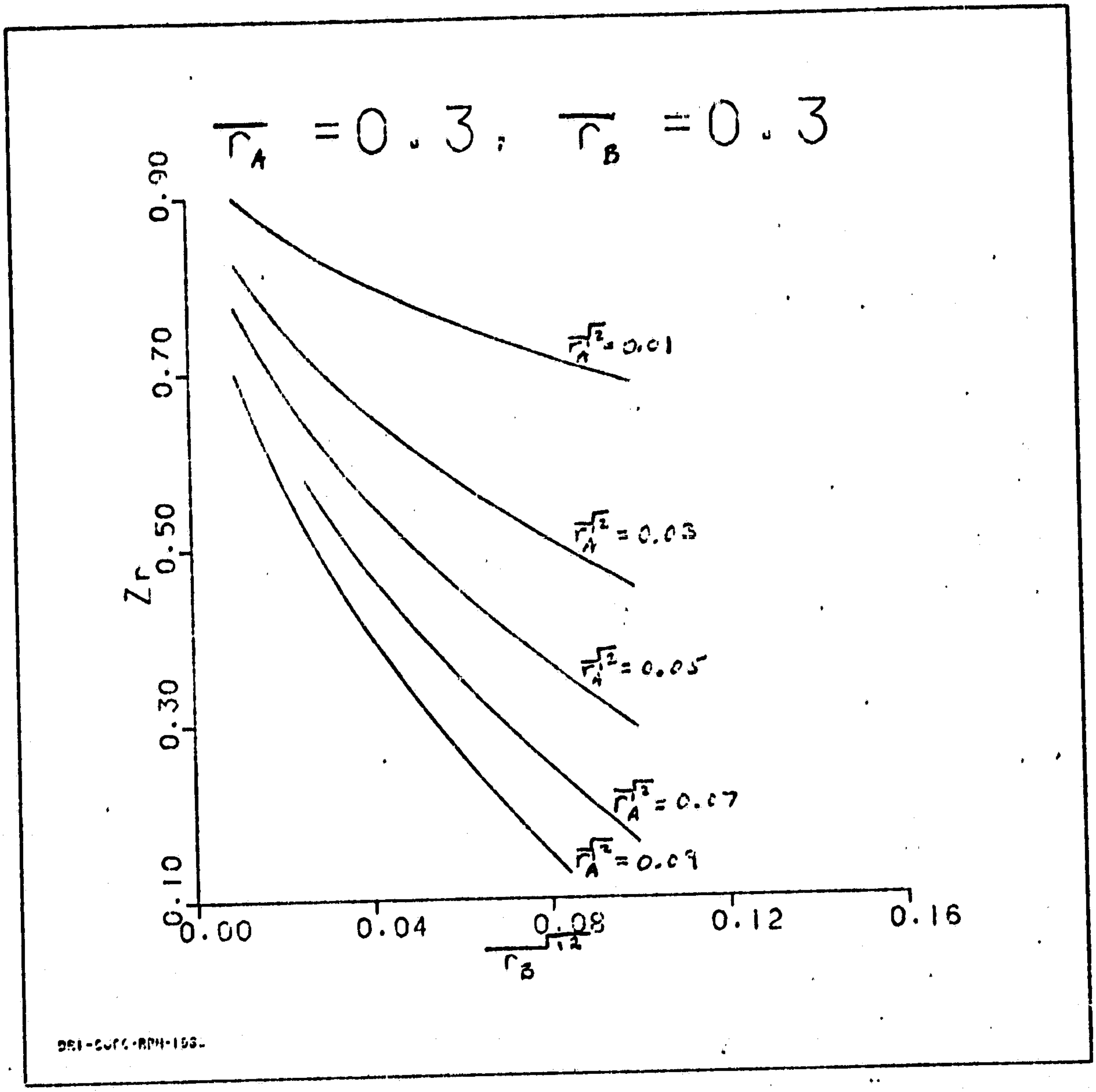

FIG. 1. CONTINUED
(e) $\overline{r_A} = 0.3$, $\overline{r_B} = 0.3$

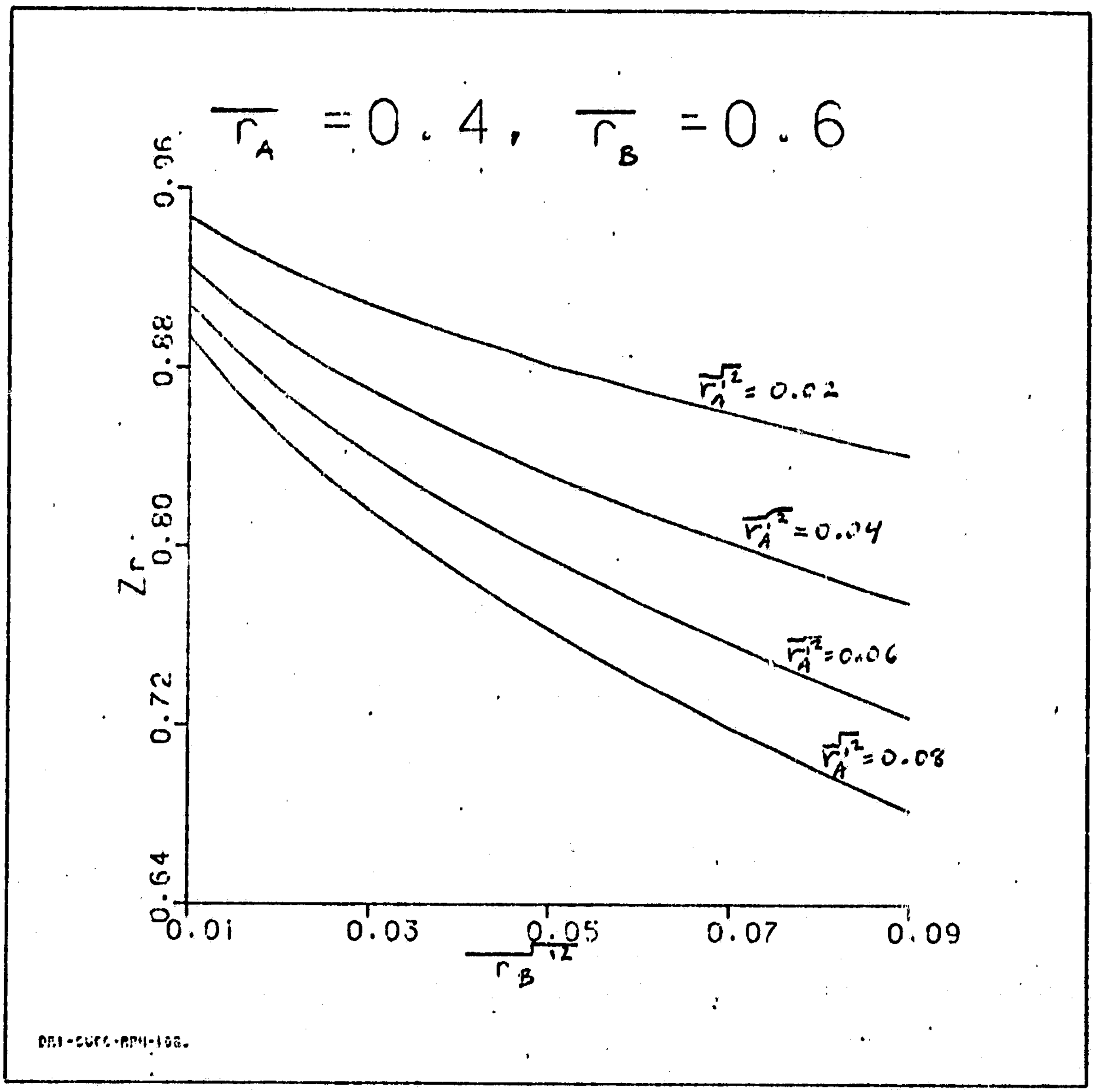

FIG. 1. CONTINUED
(f) $\overline{r_A} = 0.4$, $\overline{r_B} = 0.6$

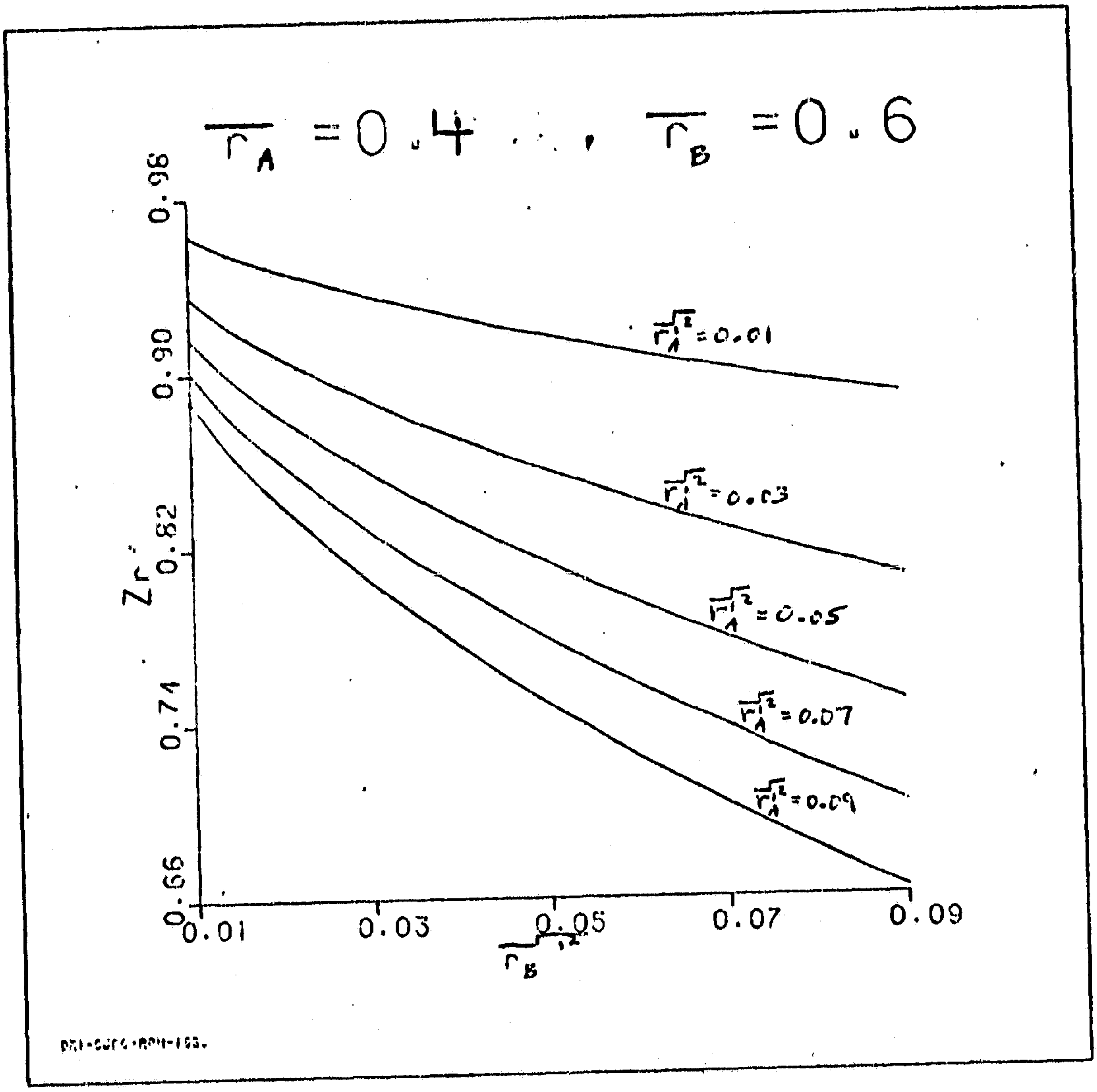

FIG. 1 CONTINUED
(g) $\overline{r_A} = 0.4$, $\overline{r_B} = 0.6$

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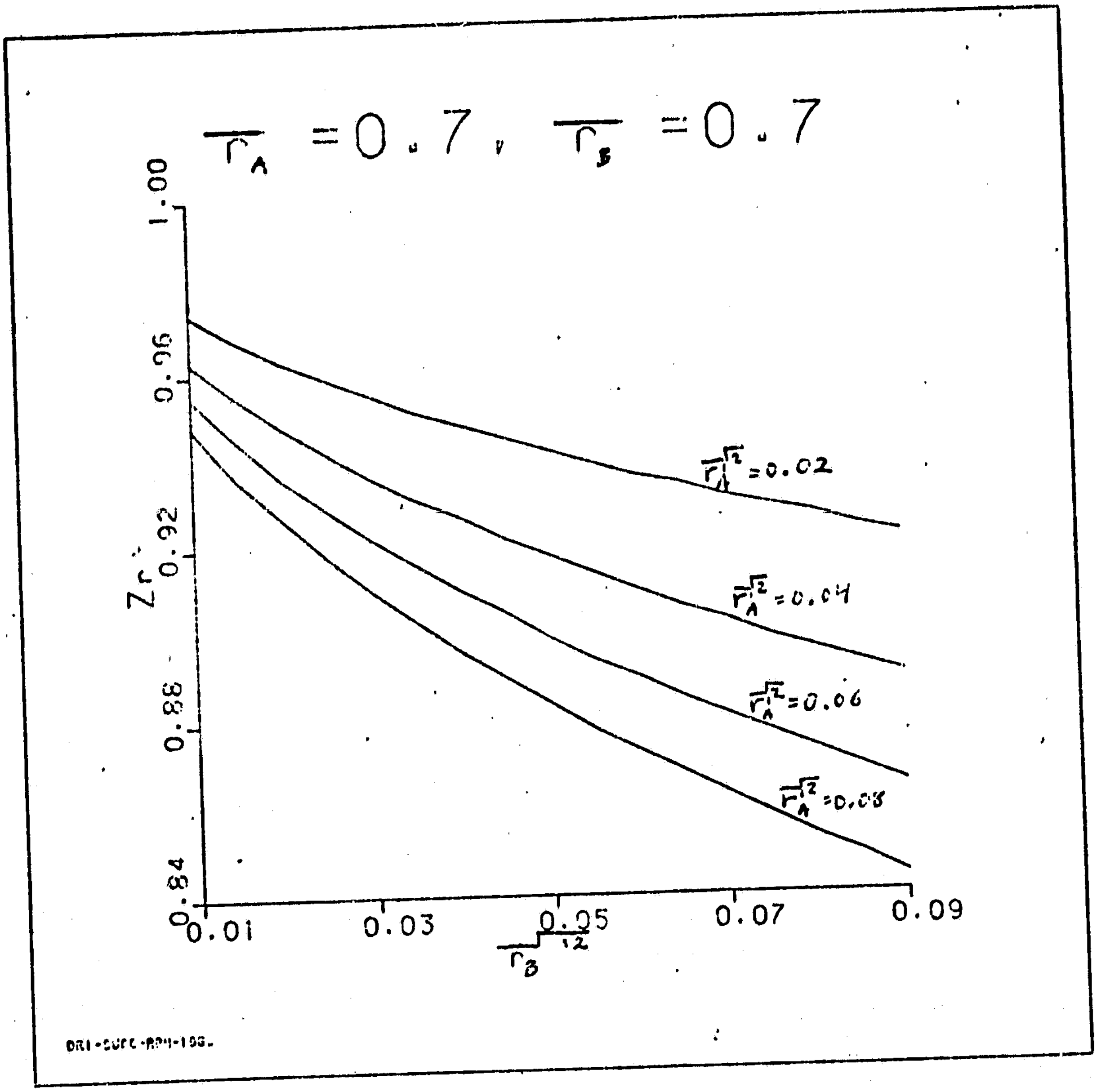

FIG. 1. CONTINUED
(h) $\overline{r_A} = 0.7$, $\overline{r_B} = 0.7$

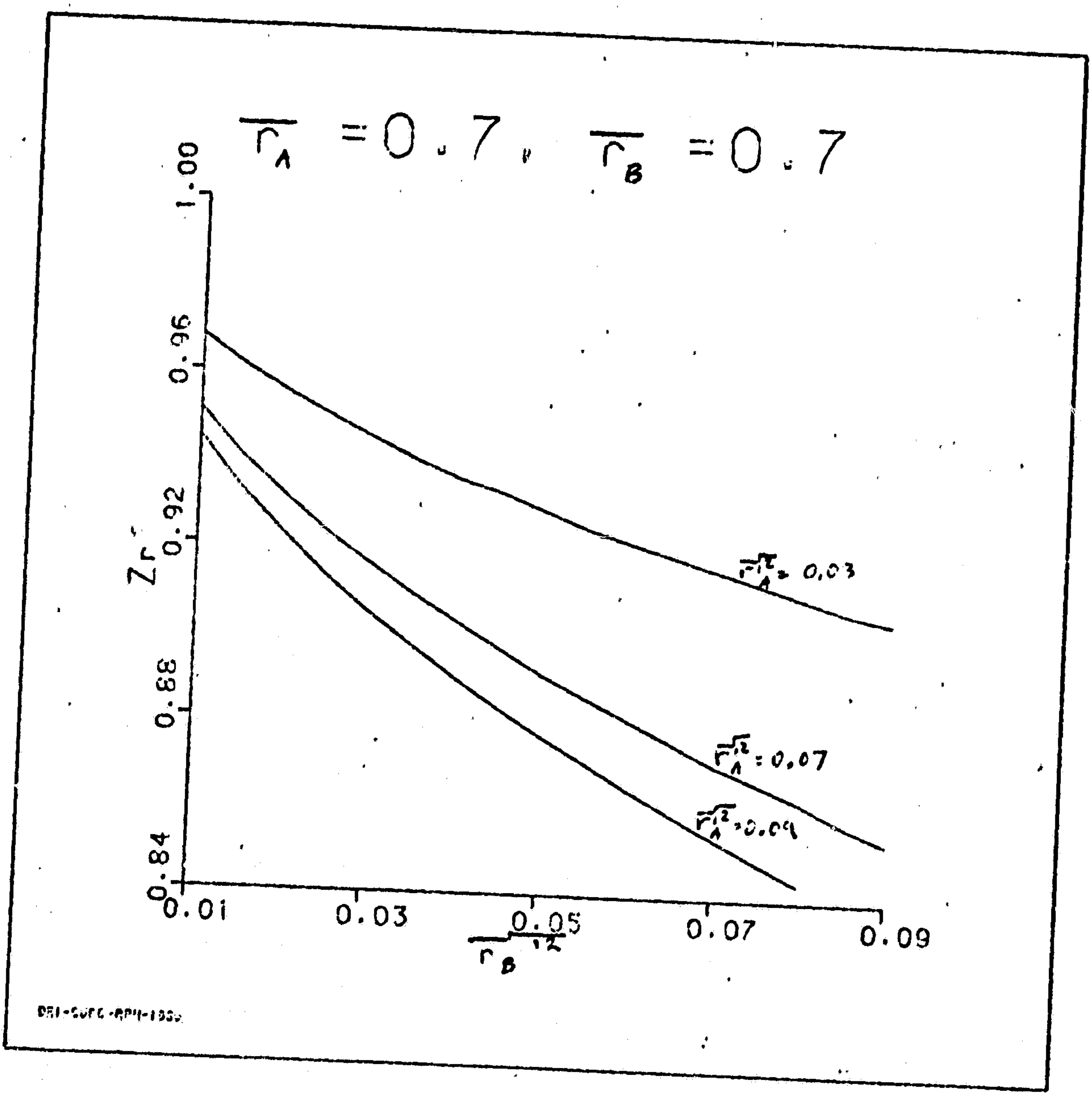

FIG. 1. CONTINUED
(i) $\overline{r_A} = 0.7$, $\overline{r_B} = 0.7$

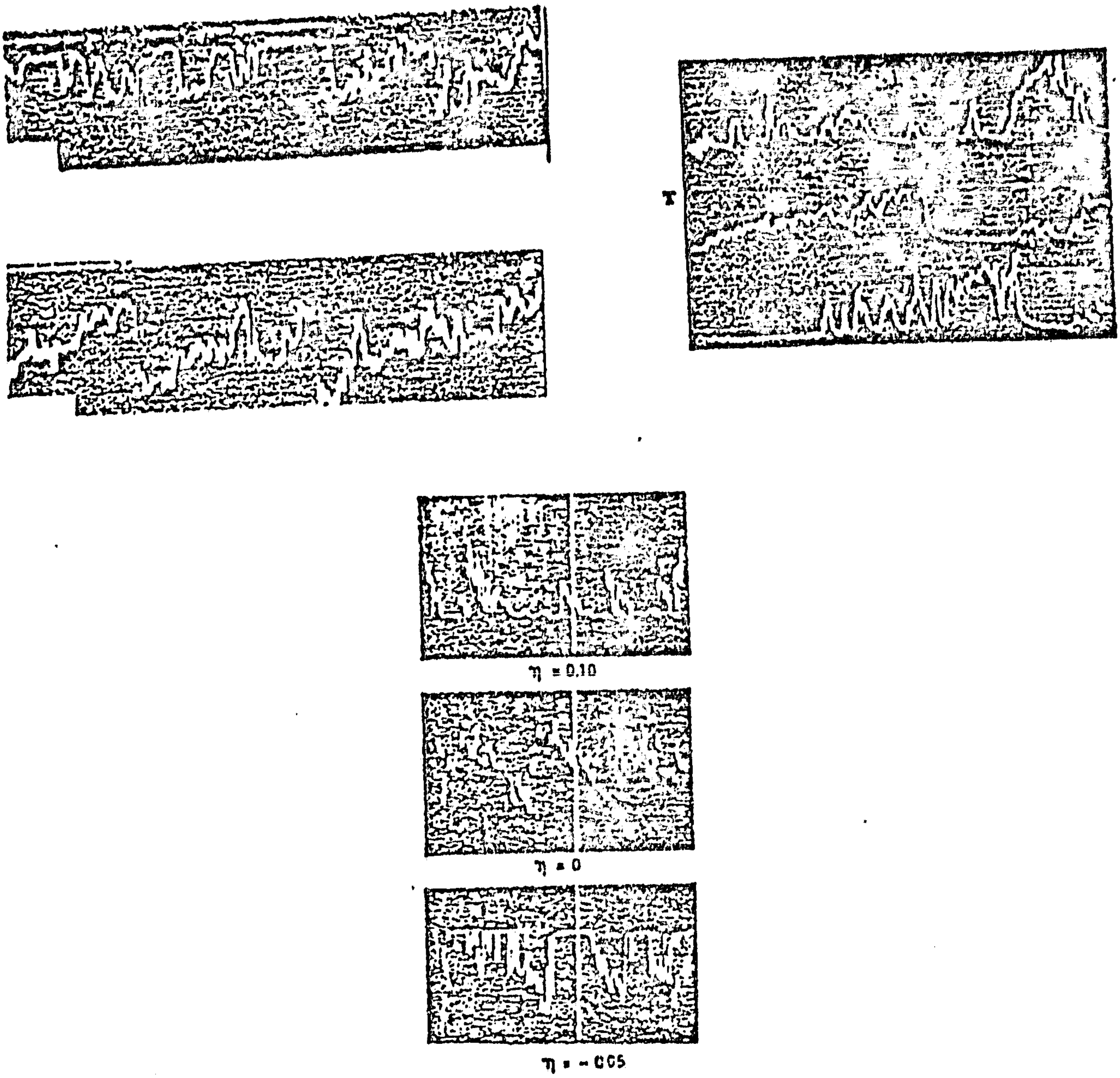

Figure 2 : Experimental temperature signals from Fiedler, Antonia, and Gibson's original papers

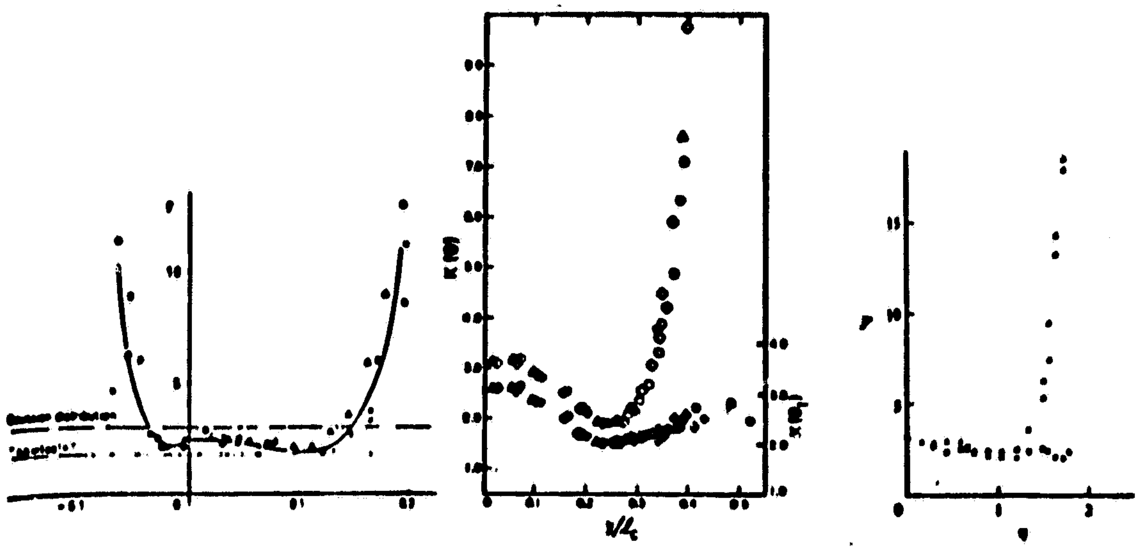

Figure 3, Experimental flatness factor distributions from Fiedler, LaRue and Libby, and Antonia's original publications

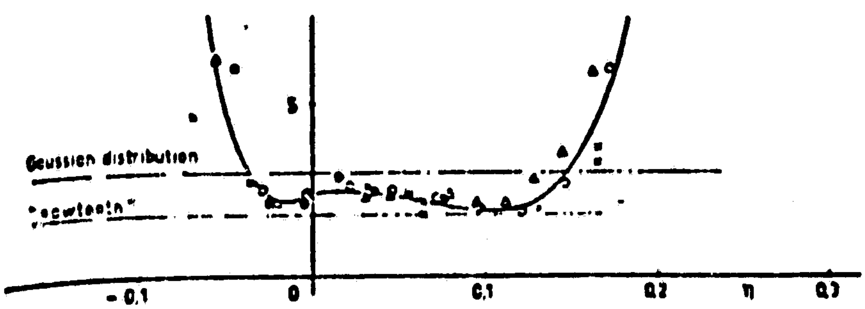

Figure 4: Flatness factor distribution from Antonia's original paper

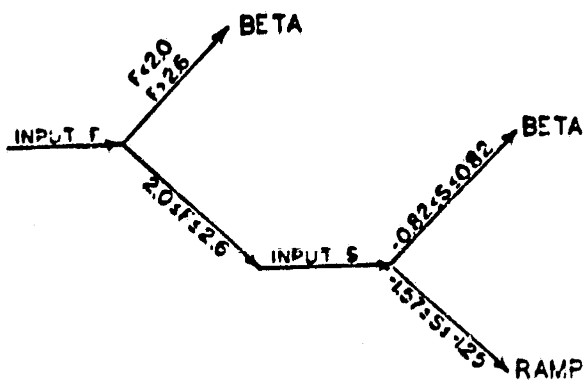

Figure 5: Schematic of selection process