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CLOSURES FOR STOCHASTICALLY DISTRIBUTED REACTANTS

by

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ABSTRACT

Closures at the third and fourth order moment are presented for the problem of the decay of reactants which obey an arbitrary R^{th} order equation (where $1<R\leq3$) and whose initial description is given stochastically. The closures are shown to satisfy prescribed realizability and asymptotic conditions for certain initial assignments of the mean, the mean square fluctuations, third order moments, and fourth order moments of the concentration field.

From theoretical considerations, it was found that non-second order reactions required setting limits on the values of the initial data (in terms of dimensionless ratios that are independent of the form of the closure chosen) in order to obtain physically acceptable results. Thus, they are not as generally applicable as the second order model, which may be applied to any initially realizable values of the mean, mean square, third, and fourth order moments fluctuations. In addition, the fourth order moment closure required a more stringent set of initial conditions than that for the third order moment closure.

I. INTRODUCTION

O'Brien has presented[1,5] a closure at the third order moment for the problem of the decay of reactants which obey a second order equation. The closure has been shown to satisfy prescribed realizability and asymptotic conditions for both the general infinite series form[5] and the simpler two term form of the expansion[1]. O'Brien has also shown[1] that this simpler form can quite closely model the exact solutions for the case of second order reactions.

The objective of this report is two-fold: 1) to extend the analysis to non-second order reactions, and 2) to find a simple fourth order moment closure that satisfies prescribed realizability and asymptotic conditions.

II. THE STATISTICAL PROBLEM

The system is described by the equation

$$\frac{d\Gamma(t)}{dt} = -\Gamma^{R}(t), \tag{2.1}$$

where Γ is the concentration which will be a random variable bounded between $0\leq\Gamma<\infty$, R is the order of the reaction which is bounded by $1<R\leq 3$, and t is the time which has been normalized by a constant reaction rate.

The problem is made stochastic by assigning initial conditions in a statistical manner[1]. For example, if $P[\Gamma(0)]$ is a prescribed initial probability density for the concentration field, then the exact solution for any order moment exists in the following form[2]:

$$\overline{\Gamma^{N}(t)} = \int_{0}^{\infty}\left[\frac{X^{(R-1)}}{1+(R-1)X^{(R-1)}t}\right]^{\frac{N}{R-1}} P(X)dX, \tag{2.2}$$

where the overbar denotes an ensemble average.

There are some asymptotic properties of (2.2) which will play a role in determining the forms of the moment closures to be suggested:

$$\lim_{t\to\infty} \overline{\Gamma(t)} = [(R-1)t]^{-\frac{1}{R-1}}, \tag{2.3}$$

$$\lim_{t\to\infty} \overline{\Gamma^{2}(t)} = C_1[(R-1)t]^{-\frac{2R}{R-1}}, \tag{2.4}$$

$$\lim_{t\to\infty} \overline{\Gamma^{3}(t)} = C_2[(R-1)t]^{-\frac{3R}{R-1}}, \tag{2.5}$$

$$\lim_{t\to\infty} \overline{\Gamma^{4}(t)} = C_3[(R-1)t]^{-\frac{4R}{R-1}}, \tag{2.6}$$

where $\gamma = \Gamma - \overline{\Gamma}$. These follow simply from an asymptotic expansion of (2.2) and the assumption that

$$\int_0^{\infty} X^{-N} P(X) dX = \overline{\Gamma^{-N}(0)}$$

exist for N = 1, 2, 3, 4.

However, in trying to form moment equations, we find that a binomial expansion of (2.1) for R not an integer leads to an infinite series representation. Thus we are now confronted with two formidable problems. First, we do not have moments of all orders readily available; and second, an infinite series is convergent only for $|\overline{\gamma^N}/\overline{\Gamma}^N| \leq 1$, where N is an integer[3]. This second problem must be overcome since a goal of this work is to have the capability to handle cases where $|\overline{\gamma^N}/\overline{\Gamma}^N| > 1$. Therefore, we must form acceptable criteria to decide which terms are to be retained in those moment equations so affected.

The following three criteria were felt to be reasonable means of deciding the number of terms to be retained in the first three lowest order moment equations: First, since the highest order moment closure scheme to be attempted is at the fourth order moment, the moment equations for the derivatives of $\overline{\Gamma}$ and $\overline{\gamma^2}$ were truncated to include terms up to and including $\overline{\gamma^4}$; second, since assuming R = 3 in (2.1) leads to a $\overline{\gamma^5}$ term in the moment equation for the derivative of $\overline{\gamma^3}$, the corresponding infinite series moment equation is truncated to include the effect of $\overline{\gamma^5}$; and third, the first three lowest order moment equations formed from the infinite series expansion and truncated as indicated, must collapse to the same expressions that can be obtained by substituting R = 1, 2, and 3 into (2.1) and forming the moment equations in the usual manner.

With these requirements in mind, the first three moment equations of an infinite unclosed hierarchy were found to be:

$$-\frac{d\overline{\Gamma}}{dt} = \overline{\Gamma}^{R} + \frac{R(R-1)}{2}\overline{\Gamma}^{R-2}\overline{\gamma^2} + \frac{R(R-1)(R-2)}{6}\overline{\Gamma}^{R-3}\overline{\gamma^3} + \\ + \frac{R(R-1)(R-2)(R-3)}{24}\overline{\Gamma}^{R-4}\overline{\gamma^4}, \tag{2.7}$$

$$-\frac{d\overline{\gamma^2}}{dt} = 2R\overline{\Gamma}^{R-1}\overline{\gamma^2} + R(R-1)\overline{\Gamma}^{R-2}\overline{\gamma^3} + \\ + \frac{R(R-1)(R-2)}{3}\overline{\Gamma}^{R-3}\overline{\gamma^4}, \tag{2.8}$$

$$-\frac{d\overline{\gamma^3}}{dt} = 3R\overline{\Gamma}^{R-1}\overline{\gamma^3} - \frac{3}{2}R(R-1)\overline{\Gamma}^{R-2}\left[\overline{\gamma^2}^2 - \overline{\gamma^4}\right] + \\ + \frac{R(R-1)(R-2)}{2}\overline{\Gamma}^{R-3}\left[\overline{\gamma^5} - \overline{\gamma^3}\,\overline{\gamma^2}\right] + \\ - \frac{R(R-1)(R-2)(R-3)}{8}\overline{\Gamma}^{R-4}\overline{\gamma^4}\,\overline{\gamma^2}. \tag{2.9}$$

Note that it is the fractional orders that significantly introduce the effect of $\overline{\gamma^4}$. At the integral orders, (2.7) and (2.8) are independent of $\overline{\gamma^4}$.

The resulting equations suppose that $\overline{\Gamma}(0)$, $\overline{\gamma^2}(0)$, and $\overline{\gamma^3}(0)$ are prescribed. We will require that $\overline{\gamma^3}$ and $\overline{\gamma^4}$ be replaced by specified functions of $\overline{\Gamma}$ and $\overline{\gamma^2}$ whose forms do not depend on the initial data (though the magnitude of $\overline{\gamma^2}(0)\ \overline{\Gamma}(0)^{-2}$ is restricted) and are such that (2.7) and (2.8) yield physically acceptable descriptions of the first three moments. Note that since $\overline{\gamma^5}(0)$ is <u>not</u> prescribed, we cannot use (2.9), even to the extent of evaluating $\frac{d\overline{\gamma^3}}{dt}(0)$.

The following realizability conditions are imposed to specify a certain degree of physical reasonableness to the solution:

$$0 \leq \overline{\Gamma}(t) < \infty \ , \tag{2.10}$$

$$0 \leq \overline{\gamma^2(t)} < \infty , \tag{2.11}$$

$$\overline{\gamma^3(t)} \geq \overline{\gamma^2(t)}^2 / \overline{\Gamma(t)} \quad - \overline{\gamma^2(t)}\ \overline{\Gamma(t)} , \tag{2.12}$$

$$\overline{\gamma^4(t)} \geq \frac{\overline{\gamma^2(t)}^3}{\overline{\Gamma(t)}^2} + 3\overline{\gamma^2(t)}^2 - 3\overline{\gamma^2(t)}\ \overline{\Gamma(t)}^2 - 4\overline{\gamma^3(t)}\ \overline{\Gamma(t)} . \tag{2.13}$$

The inequalities (2.12) and (2.13) arise from a restriction[4] on the skewness and kurtosis of any probability density which is zero for values of the random variable that are less than or equal to zero.

In the next section, closures are proposed whose results satisfy the realizability conditions (2.10), (2.11), (2.12) and (2.13) for all values of $\overline{\Gamma}(0)$, $\gamma^2(0)$, $\overline{\gamma^3}(0)$, and $\overline{\gamma^4}(0)$ which themselves do not violate (2.10), (2.11), (2.12) and (2.13). We will also require that the asymptotic behaviors given by (2.3), (2.4), (2.5), and (2.6) are satisfied. The closures are to be simple functional forms that do not vary for different initial data.

III. PROPOSED CLOSURES

O'Brien has shown[1] that the following simple third order moment closure satisfies asymptotic and realizability conditions for the second order equation. For simplicity, we assume that this form is applicable to non-second order reactions without any modification.

The third order moment closure

$$\overline{\gamma^3(t)} = A_1 \overline{\gamma^2(t)}^2 \, \overline{\Gamma(t)}^{-1} - A_0 \overline{\gamma^2(t)}^{3/2} \tag{3.1}$$

is a combination of simple, dimensionally correct, simultaneous functions.

In the same spirit, the following simple closure for the fourth order moment is proposed:

$$\overline{\gamma^4(t)} = B_1 \overline{\gamma^2(t)}^2 - B_2 \overline{\gamma^2(t)}^3 \, \overline{\Gamma(t)}^{-2}. \tag{3.2}$$

In (3.1), the constant A_1 is obtained from A_o, $\overline{\Gamma}(0)$, $\overline{\gamma^2}(0)$, and $\overline{\gamma^3}(0)$. We will show that there exists a maximum positive value of A_o and of B_1 (say A_{MAX} and B_{MAX}, respectively) such that for $A_o < A_{MAX}$ and $B_1 < B_{MAX}$, the closures (3.1) and (3.2) satisfy the asymptotic conditions (2.3), (2.4), (2.5), and (2.6) and the realizability conditions (2.10), (2.11), (2.12), and (2.13) for certain allowable ranges of the values of $\overline{\Gamma}(0)$, $\overline{\gamma^2}(0)$, $\overline{\gamma^3}(0)$, and $\overline{\gamma^4}(0)$. This differs from [1] in which the use of a second order reaction equation enabled the proof that (3.1) could be applied to all realizable values of $\overline{\Gamma(0)}$, $\overline{\gamma^2}(0)$, and $\overline{\gamma^3}(0)$.

As suggested by O'Brien[5], (3.1) and, analogously, (3.2) have more general closure forms in terms of an infinite series. However, the purpose here is to explore the relevance of these simpler closures by showing that for certain values of the initial data, they satisfy the previously mentioned conditions.

For convenience in constructing subsequent proofs, the following definitions are made:

$$Y_1(t) = \overline{\Gamma(t)} \tag{3.3}$$

$$Y_2(t) = \overline{\gamma^2(t)}\;\overline{\Gamma(t)}^{\,-2} \tag{3.4}$$

$$T = \int_0^t \Big[1 + \frac{R(R-1)}{2}Y_2 + Y_2^2\,\frac{R(R-1)(R-2)}{6}\Big(A_1 + \frac{\{R-3\}}{4}B_1\Big) + \\ - \frac{R(R-1)(R-2)}{6}\Big(A_0 Y_2^{3/2} + B_2 Y_2^{3}\frac{\{R-3\}}{4}\Big)\Big]\,dt' \tag{3.5}$$

$$\tau = 2(R-1)\int_0^t Y_1^{R-1}(t')\,dt' \tag{3.6}$$

When (3.1) through (3.6) are incorporated into (2.7) and (2.8), they become:

$$\frac{dY_1}{dt} = -Y_1^R\Big[1 + \frac{R(R-1)}{2}Y_2 + \frac{\overline{\gamma^3}}{Y_1^3}\,\frac{R(R-1)(R-2)}{6} + \\ + \frac{R(R-1)(R-2)(R-3)}{24}\,\frac{\overline{\gamma^4}}{Y_1^4}\Big],$$

OR

$$\frac{dY_1}{dt} = -Y_1^R\Big[1 + \frac{R(R-1)}{2}Y_2 + \Big(A_1 + \frac{R-3}{4}B_1\Big)\frac{R(R-1)(R-2)}{6}Y_2^2 + \\ - \frac{R(R-1)(R-2)}{6}\Big(A_0 Y_2^{3/2} + B_2 Y_2^{3}\frac{\{R-3\}}{4}\Big)\Big], \tag{3.7a}$$

OR

$$\frac{dY_1}{dT} = -Y_1^R . \tag{3.7b}$$

$$\frac{dY_2}{dt} = -2(R-1)Y_1^{R-1}\left[Y_2 + \frac{R}{2}\left\{1+\frac{(2-R)}{3}Y_2\right\}\frac{\overline{Y^3}}{Y_1^3} + \right.$$

$$\left. + \frac{R(R-2)}{6}\left\{1+\frac{(3-R)}{4}Y_2\right\}\frac{\overline{Y^4}}{Y_1^4} - \frac{R}{2}Y_2^2\right],$$

$$\frac{dY_2}{dt} = -2(R-1)Y_1^{R-1}\left[Y_2 + \frac{R}{2}\left\{A_1 - 1 + \frac{(R-2)}{3}B_1\right\}Y_2^2 + \right.$$

$$- \frac{R(R-2)}{6}\left\{A_1 + \frac{(R-3)}{4}B_1 + B_2\right\}Y_2^3 - \frac{R}{2}A_0Y_2^{3/2} +$$

$$\left. + \frac{R(R-2)}{6}\left\{A_0Y_2^{5/2} + B_2Y_2^4\frac{(R-3)}{4}\right\}\right], \tag{3.7c}$$

or

$$\frac{dY_2}{d\tau} = -\left[Y_2 + \frac{R}{2}\left\{A_1 - 1 + \frac{(R-2)}{3}B_1\right\}Y_2^2 + \right.$$

$$- \frac{R(R-2)}{6}\left\{A_1 + \frac{(R-3)}{4}B_1 + B_2\right\}Y_2^3 - \frac{R}{2}A_0Y_2^{3/2} +$$

$$\left. + \frac{R(R-2)}{6}\left\{A_0Y_2^{5/2} + B_2Y_2^4\frac{(R-3)}{4}\right\}\right]. \tag{3.7d}$$

An immediate consequence of the forms of (3.7a, b, c, d) and the fact that $Y_1(0)>0$ and $Y_2(0)>0$ is that if $Y_1(t_1) = 0$ for some $t = t_1$, then so is

$$\frac{d^N Y_1(t_1)}{dt^N} \quad \text{for all } N.$$

Similarly, if $Y_2(t_2) = 0$, then so is

$$\frac{d^N Y_2(t_2)}{dt^N} \quad \text{for all } N.$$

This implies that if T and τ are increasing functions, then:

$$Y_1(t) \geq 0 \qquad \text{for all } t\,, \tag{3.8a}$$

and

$$Y_2(t) \geq 0 \qquad \text{for all } t\,. \tag{3.8b}$$

However, as can be seen from (3.5) and (3.6), certain restrictions on the values of Y_2, A_1, A_o, B_1, and B_2 must be made before $T \geq 0$ can be assured. If the necessary restrictions are made then it is clear that T and τ are increasing functions of time.

Thus, in order that T and τ be non-decreasing functions of time and that (3.7a) and (3.7b) satisfy (2.3) and (2.4) respectively, it is necessary that, from (3.7a):

$$1 + \frac{R(R-1)}{2} Y_2 + \frac{R(R-1)(R-2)}{6}\left\{A_1 + \frac{(R-3)}{4} B_1\right\} Y_2^2 + \\ - \frac{R(R-1)(R-2)}{6}\left(A_0 Y_2^{3/2} + B_2 Y_2^3 \frac{(R-3)}{4}\right) \geq 0$$

$$\text{FOR } 0 \leq Y_2(t) \leq Y_2(0) \text{ AND } 1 < R \leq 3\,, \tag{3.9}$$

and from (3.7c):

$$Y_2 + \frac{R}{2}\left(A_1 - 1 + \frac{\{R-2\}}{3} B_1\right) Y_2^2 - \frac{R(R-2)}{6}\left(A_1 + \frac{\{R-3\}}{4} B_1 + B_2\right) Y_2^2 + \\ - \frac{R}{2} A_0 Y_2^{3/2} + \frac{R(R-2)}{6}\left(A_0 Y_2^{5/2} + B_2 Y_2^4 \frac{\{R-3\}}{4}\right) \geq 0$$

$$\text{FOR } 0 \leq Y_2(t) \leq Y_2(0) \text{ AND } 1 < R \leq 3. \tag{3.10}$$

IV. THEORETICAL RESULTS

A. General Considerations

Since the terms in (3.9) and (3.10) are explicit functions of "R", while the realizability conditions do not contain "R" explicitly, we find that we will not be able to prove for the whole range of "R" considered, $1<R\leq 3$, that using the assumed closures to satisfy (3.9) and (3.10) is tantamount to satisfying the realizability conditions. In fact, as shown in Ref. [1], only for the particular case of R = 2 is it true that employing the assumed closures in (3.9) and (3.10) leads exactly to the realizability conditions.

In the general case, different limits on the values of $\overline{\gamma^3}$ and $\overline{\gamma^4}$ are required by realizability than by (3.9) or by (3.10). Simultaneous satisfaction of realizability, (3.9), and (3.10) requires limiting the values of $Y_2(0)$, $\overline{\gamma^3}(0)$ and $\overline{\gamma^4}(0)$ to be within ranges amenable to them all. (See Appendix I for details.) Thus, for arbitrary R, not only must the constants used in the closures be limited by certain relations, as in Ref. [1]; but also the value of the initial data must be restricted if one is to obtain physically meaningful results.

The restrictions may be divided into two major divisions: those applicable for $1<R<2$ and those for $2<R\leq 3$. Within each division, different restrictions are required by a third order moment closure than by a fourth order moment closure.

B. Fourth Order Moment Closure

Using the assumed third and fourth order moment closures and requiring that realizability, (3.9) and (3.10) be simultaneously satisfied, yields the following requirements:

For 1<R<2

$$0 \leq Y_2(0) \leq (-3+\sqrt{21})/2 \approx 0.79, \tag{4.1a}$$

$$\frac{\overline{Y^3(0)}}{Y_1^3(0)} \leq \frac{3[2+R(R-1)Y_2(0)]}{R(R-1)(2-R)}, \tag{4.1b}$$

$$\frac{\overline{Y^3(0)}}{Y_1^3(0)} \geq Y_2^2(0) - Y_2(0), \tag{4.1c}$$

$$\frac{\overline{Y^4(0)}}{Y_1^4(0)} \leq 4[6Y_2(0) - 3RY_2^2(0) + R\{3+(2-R)Y_2(0)\}\times \\ \times \frac{\overline{Y^3(0)}}{Y_1^3(0)}][R(2-R)(4+\{3-R\}Y_2(0))]^{-1}, \tag{4.1d}$$

$$\frac{\overline{Y^4(0)}}{Y_1^4(0)} \geq Y_2^3(0) + 3Y_2^2(0) - 3Y_2(0) + \\ -4\frac{\overline{Y^3(0)}}{Y_1^3(0)}. \tag{4.1e}$$

By substituting closures (3.1) and (3.2) into (4.1b) through (4.1e), it can be shown that the constants used in the closures must be such that:

$$A_0 \geq -\{Y_2(0)[3/(2-R) - A_1] + [R(R-1)(2-R)]^{-1}\}Y_2^{-3/2}(0), \tag{4.1f}$$

$$A_0 \leq 2(A_1-1)^{\frac{1}{2}}, \text{ FOR } A_1 > 1, \tag{4.1g}$$

$$B_1 \leq 4[6 + R\{3+(2-R)Y_2(0)\}\{A_1Y_2(0) - A_0Y_2^{\frac{1}{2}}(0)\} - 3RY_2(0)]\times \\ \times[Y_2(0)R(2-R)(4+\{3-R\}Y_2(0))]^{-1} + \\ + B_2Y_2(0), \tag{4.1h}$$

$$B_1 \geq Y_2(0) + 3 - 3Y_2^{-1}(0) - 4(A_1 - A_0 Y_2^{-\frac{1}{2}}(0)) + B_2 Y_2(0); \tag{4.1i}$$

AND FOR $2 < R \leq 3$,

$$0 \leq Y_2(0) \leq (-3 + \sqrt{21})/2 \approx 0.79, \tag{4.1j}$$

$$\frac{\overline{Y^3(0)}}{Y_1^3(0)} < \infty, \tag{4.1k}$$

$$\frac{\overline{Y^3(0)}}{Y_1^3(0)} \geq \frac{-Y_2(0) + \left(\frac{R}{2}\right) Y_2^2(0)}{\frac{R}{6}[3 + (2-R)Y_2(0)]}, \tag{4.1l}$$

$$\frac{\overline{Y^4(0)}}{Y_1^4(0)} \leq \frac{24\left[1 + (R(R-1)/2)Y_2(0) + (R(R-1)(R-2)/6)\frac{\overline{Y^3(0)}}{Y_1^3(0)}\right]}{R(R-1)(R-2)(3-R)}, \tag{4.1m}$$

$$\frac{\overline{Y^4(0)}}{Y_1^4(0)} \geq Y_2^3(0) + 3Y_2^2(0) - 3Y_2(0) - 4\frac{\overline{Y^3(0)}}{Y_1^3(0)}. \tag{4.1n}$$

And by following the procedure previously indicated, it can be shown that the constants used in the closures must be such that:

$$-\infty < A_0 \le \frac{3[Y_2(0)(A_1-1) + \{A_1(2-R)/3\}Y_2^2(0) + 2/R]}{Y_2^{\frac{1}{2}}(0)(5-R)} . \tag{4.1o}$$

However, if R is very close to 2, or if

$$\frac{(R/6)(2-R)Y_2(0)\dfrac{\overline{Y^3(0)}}{Y_1^{\,3}(0)}}{Y_2(0) - \frac{R}{2}Y_2^2(0) + \frac{R}{2}\dfrac{\overline{Y^3(0)}}{Y_1^{\,3}(0)}} \ll 1, \tag{4.1p}$$

then it can be shown that (4.1o) may be simplified to yield the requirement that A_0 be such that:

$$-\infty < A_0 \le 2\left[\frac{2}{R}(A_1-1)\right]^{\frac{1}{2}}; \quad A_1 > 1 . \tag{4.1q}$$

Returning to generally applicable results, we find that:

$$B_1 \le \frac{4[A_1 - A_0 Y_2^{-\frac{1}{2}}(0)]}{(3-R)} + \frac{12[2+R(R-1)Y_2(0)]}{Y_2^2(0)R(R-1)(R-2)(3-R)} + B_2 Y_2(0), \tag{4.1r}$$

and

$$B_1 \ge Y_2(0) + 3 - 3Y_2^{-1}(0) - 4[A_1 - A_0 Y_2^{-\frac{1}{2}}(0)] + B_2 Y_2(0) . \tag{4.1s}$$

Using the same procedures, the third order moment closure problem was also considered.

C. Third Order Moment Closure

If it is assumed that $\overline{Y^4}$ terms can be neglected from (3.9) and (3.10), and thus only a third order moment closure is required, then $\overline{Y^3}$ realizability, (3.9), and (3.10) require that:

For $1 < R < 2$

$$0 \leq Y_2(0) \leq \frac{R(R-1)(5-R) + \{[R(R-1)(5-R)]^2 + 24R(R-1)(2-R)\}^{\frac{1}{2}}}{2R(R-1)(2-R)}, \tag{4.2a}$$

$$Y_2^{\,2}(0) - Y_2(0) \leq \frac{\overline{Y^3(0)}}{Y_1^{\,3}(0)} \leq \frac{3[2 + R(R-1)Y_2(0)]}{R(R-1)(2-R)}, \tag{4.2b}$$

$$A_0 \geqslant -\frac{[Y_2(0)\{3/(2-R) - A_1\} + \{R(R-1)(2-R)\}^{-1}]}{Y_2^{3/2}(0)}, \tag{4.2c}$$

$$A_0 \leq 2\,(A_1 - 1)^{\frac{1}{2}}\,;\; A_1 > 1, \tag{4.2d}$$

and for $2 < R \leq 3$

$$0 \leq Y_2(0) < \frac{3}{R-2}\,, \tag{4.2e}$$

$$\frac{-Y_2(0) + \frac{R}{2}Y_2^{\,2}(0)}{\frac{R}{6}[3 + (2-R)Y_2(0)]} \leqslant \frac{\overline{Y^3(0)}}{Y_1^{\,3}(0)} < \infty, \tag{4.2f}$$

$$-\infty < A_0 \leq \frac{3[Y_2(0)(A_1 - 1) + A_1\{(2-R)/3\}Y_2^{\,2}(0) + 2/R]}{Y_2^{\frac{1}{2}}(0)\,(5-R)}, \tag{4.2g}$$

and if

$$\frac{\frac{R}{6}(2-R)\,Y_2(0)\,\frac{\overline{Y^3(0)}}{Y_1^{\,3}(0)}}{Y_2(0) - \frac{R}{2}Y_2^{\,2}(0) + \frac{R}{2}\frac{\overline{Y^3(0)}}{Y_1^{\,3}(0)}} \ll 1, \tag{4.2h}$$

then, as before, (4.2g) is simplified to:

$$-\infty < A_0 \le 2\left[\frac{2}{R}(A_1-1)\right]^{\frac{1}{2}};\quad A_1 > 1. \tag{4.2i}$$

In general, therefore, the major advantage of the third order moment closure over the fourth order moment closure is that it is applicable to many more initial values of $Y_2(0)$. A plot of (4.2a) and (4.2e) versus R (see Fig. 1) shows that the maximum allowable value of the ratio $\overline{y^3(0)}/Y_1^3(0)$ for the third order moment closure is at least three times that allowed by the fourth order moment closure.

Estimates of the value of the constants A_o and B_1 may also be obtained from an asymptotic expansion of (2.2). By comparing the resultant time series with (2.4), (2.5), and (2.6), one finds that:

$$C_1 = (R-1)^2\left(\overline{\Gamma^{-(2R-2)}(0)} - \overline{\Gamma^{-(R-1)}(0)}^{\,2}\right), \tag{4.3}$$

$$C_2 = (R-1)^3\left(\overline{\Gamma^{-(3R-3)}(0)} + 2\,\overline{\Gamma^{-(R-1)}(0)}^{\,3} + {} - 3\,\overline{\Gamma^{-(R-1)}(0)}\;\overline{\Gamma^{-(2R-2)}(0)}\right), \tag{4.4}$$

$$C_3 = (R-1)^4\left(\overline{\Gamma^{-(4R-4)}(0)} - 4\,\overline{\Gamma^{-(3R-3)}(0)}\;\overline{\Gamma^{-(R-1)}(0)} + {} + 6\,\overline{\Gamma^{-(2R-2)}(0)}\;\overline{\Gamma^{-(R-1)}(0)}^{\,2} - 3\,\overline{\Gamma^{-(R-1)}(0)}^{\,4}\right). \tag{4.5}$$

Thus, from the closures (3.1) and (3.2), together with the asymptotic condition (2.3) through (2.6) it is possible to identify A_o and B_1 as follows:

$$A_0 = C_2\, C_1^{-3/2}, \tag{4.6}$$

or

$$A_0 = \frac{\left[\overline{\Gamma^{-(3R-3)}(0)} + 2\,\overline{\Gamma^{-(R-1)}(0)}^{\,3} - 3\,\overline{\Gamma^{-(R-1)}(0)}\;\overline{\Gamma^{-(2R-2)}(0)}\right]}{\left[\overline{\Gamma^{-(2R-2)}(0)} - \overline{\Gamma^{-(R-1)}(0)}^{\,2}\right]^{3/2}}, \tag{4.7}$$

$$B_1 = C_3 C_1^{-2}, \tag{4.8}$$

OR

$$B_1 = \left[\overline{\Gamma^{-(4R-4)}(0)} - 4\,\overline{\Gamma^{-(3R-3)}(0)}\;\overline{\Gamma^{-(R-1)}(0)} + 6\,\overline{\Gamma^{-(2R-2)}(0)}\;\overline{\Gamma^{-(R-1)}(0)}^{2} + -3\,\overline{\Gamma^{-(R-1)}(0)}^{4}\right] / \left[\overline{\Gamma^{-(2R-2)}(0)} - \overline{\Gamma^{-(R-1)}(0)}^{2}\right]^{2}, \tag{4.9}$$

which, for R=2 reduce

$$A_o = \overline{\gamma^{-3}(0)}\;\overline{\gamma^{-2}(0)}^{-3/2} \tag{4.10}$$

and

$$B_1 = \overline{\gamma^{-4}(0)}\;\overline{\gamma^{-2}(0)}^{-2}, \tag{4.11}$$

where $\gamma^{-1} \equiv \Gamma^{-1} - \overline{\Gamma^{-1}}$

If only $\overline{\Gamma^{-1}(0)}$ and $\overline{\gamma^{-2}(0)}$ exist, then it is possible to obtain a lower bound to A_o and B_1 in the following fashion. Since Γ^{-1} is a non-negative random variable, it too, simplifies an inequality such as (2.12) or (2.13). Thus, we can write that

$$\overline{\gamma^{-3}(0)} \geq \overline{\gamma^{-2}(0)}^{2}\;\overline{\Gamma^{-1}(0)}^{-1} - \overline{\gamma^{-2}(0)}\;\overline{\Gamma^{-1}(0)} = A_{MIN}, \tag{4.12}$$

or $$A_o = O\left(A_{MIN}\,\overline{\gamma^{-2}(0)}^{-3/2}\right) \tag{4.13}$$

and $$\overline{\gamma^{-4}(0)} \geq \overline{\gamma^{-2}(0)}^{3}\;\overline{\Gamma^{-2}(0)} + 3\,\overline{\gamma^{-2}(0)}^{2} - 3\,\overline{\gamma^{-2}(0)}\;\overline{\Gamma^{-1}(0)}^{2} - 4\,A_{MIN}\,\overline{\Gamma^{-1}(0)} = B_{MIN}, \tag{4.14}$$

or $$B_1 = O\left(B_{MIN}\,\overline{\gamma^{-2}(0)}^{2}\right). \tag{4.15}$$

Because of the approximate nature of our analysis, the preceding should be considered as order of magnitude estimates for A_o and B_1. However, in the cases of distributions for which negative moments do not exist, there appears to be no analytical way in which the constants A_o and B_1 can be estimated.

D. Asymptotic Conditions

As indicated in sections B and C, if the initial statistics fall within the ranges outlined, one can then say that $\overline{\gamma^3(t)}$ realizability (2.12), $\overline{\gamma^4(t)}$ realizability-(2.13), (3.9) and (3.10) are simultaneously satisfied.

If (3.9) is satisfied, then this implies from (3.5) that T is a non-decreasing function of time. Using this fact, one can find from (3.7b) that

$$Y_1(T) = [Y_1^{R-1}(0) / \{1+(R-1)Y_1^{R-1}(0)T\}]^{\frac{1}{R-1}}. \tag{4.16}$$

With T increasing, (4.16) is a monotonically decreasing function of time. Changing the time scale, we can write that (4.16) indicates that

$$\lim_{t \to \infty} Y_1(t) = [(R-1)t]^{-\frac{1}{R-1}}. \tag{4.17}$$

Thus, asymptotic condition (2.3) is satisfied.

Further, use of (3.7c) and (3.6) with the results of (4.16) imply that

$$\lim_{t \to \infty} Y_2(t) = O[t^{\frac{2-2R}{R-1}}]. \tag{4.18}$$

This with (2.3) implies that

$$\lim_{t \to \infty} \overline{\gamma^2(t)} = O(t^{-\frac{2R}{R-1}}). \tag{4.19}$$

Thus, asymptotic condition (2.4) is satisfied.

Using these results, (3.8a and b), i.e., $\overline{\Gamma}(t) \geq 0$, $\overline{\gamma^2}(t) \geq 0$, together with the forms of the closures, (3.1) and (3.2), suggested for $\overline{\gamma^3}(t)$ and $\overline{\gamma^4}(t)$ indicate that asymptotic conditions (2.5) and (2.6) are also satisfied, i.e.:

$$\lim_{t \to \infty} \overline{\gamma^3(t)} = -A_0 \overline{\gamma^2(t)}^{3/2},$$

$$\lim_{t \to \infty} \overline{\gamma^3(t)} = -A_0 \left(t^{-\frac{2R}{R-1}}\right)^{3/2},$$

$$\lim_{t \to \infty} \overline{\gamma^3(t)} = -A_0 \left[t^{-\frac{3R}{R-1}}\right], \tag{4.20}$$

which satisfies (2.5), and

$$\lim_{t \to \infty} \overline{\gamma^4(t)} = B_1 \overline{\gamma^2(t)}^2,$$

$$\lim_{t \to \infty} \overline{\gamma^4(t)} = B_1 \left(t^{-\frac{2R}{R-1}}\right)^2,$$

$$\lim_{t \to \infty} \overline{\gamma^4(t)} = B_1 \left[t^{-\frac{4R}{R-1}}\right], \tag{4.21}$$

which satisfies (2.6).

Thus, in sections B, C, and D we have shown that when the initial data are found to be within the limits outlined, use of the closure forms (3.1) and (3.2) do indeed satisfy the prescribed realizability and asymptotic conditions for the probability distribution under consideration.

V. CONCLUSIONS

In the previous sections, a method has been presented that will yield satisfactory closure solutions at the third and fourth order moments provided that certain requirements on initial data are met. In addition, this method has been shown to be directly applicable to reactions of other than second order (though the order is herein limited to be between one and three).

An interesting result of proceeding from a binomial expansion for the moment equations is that the effect of the $\overline{\gamma^4(t)}$ dependence clearly appears only for $R \neq 2$. However, in accounting for its presence, the fourth order moment closure by its very nature requires a much more stringent set of limitations on the values of the initial moments than those imposed by a third order moment closure. Thus, since use of a third order closure eliminates accounting for the $\overline{\gamma^4(t)}$ dependence, it is probably the more useful of the two for purposes of application.

Quite interestingly, for a closure at either the third or the fourth order moment, the second order reaction is the only one that allows any realizable value of $\overline{\Gamma(0)}$, $\overline{\gamma^2(0)}$, $\overline{\gamma^3(0)}$, and $\overline{\gamma^4(0)}$ to be used. This seems to be due to the nature of the form of the third order moment closure used. This form not only makes the moment equations automatically satisfy the third order realizability condition, but is also of a functional form that requires relatively minor constraints on the constants so that quite accurate solutions[1] can be obtained for any initially realizable value of $\overline{\Gamma(0)}$, $\overline{\gamma^2(0)}$, and $\overline{\gamma^3(0)}$.

As pointed out in Section IV, the fact that the realizability conditions are not explicit functions of R, while the general moment equations are, makes it very unlikely that a closure form can be devised that will automatically satisfy realizability and the physical requirements on the moment equations.

In general, it seems that restricting the number of terms and their forms forces restrictions on the range of allowable initial data; whereas, attempting to allow for any realizable initial value forces one to adopt much more complicated closures. Of course, increasing the number of terms hinders the ease of applicability of the closure.

Besides, as Orszag has pointed out[6], a distribution function is not determined by the specification of a few lowest order moments and it is thus clearly untenable to expect that a specific statistical description will be accurate for all possible distribution functions that display the same initial values of the three lowest moments.

Therefore, since there is no guarantee that increasing the number of terms by a finite amount will produce more accurate closures and since practical utility makes a many term closure awkward to use this paper has been devoted to the demonstration that simple-two-term closure forms can indeed satisfy realizability and asymptotic conditions and are thus very useful for practical applications. Reiterating the remarks in[1] on the closure and realizability problems, we emphasize that the use of realizability conditions will be relevant to any problem involving a statistical description of reactions since, by nature, concentrations are non-negative quantities. This special nature of the concentration as a random variable can be ignored only in the case of first order reactions; since the mean concentration and the fluctuations do not interact for this order. Of course, initial moments must still be properly specified.

For turbulently mixed reactions, closures of the kind presented here can be usefully applied to moments that consist only of the concentration variables. Moments involving both velocity and concentration will require the input of a closure suited to them; for example, the results obtained from the Lagrangian History Direct Interaction Hypothesis[7].

Questions pertaining to the accuracy of the proposed closures are presently being investigated. Results are scheduled to be presented as College of Engineering Report No. 124.

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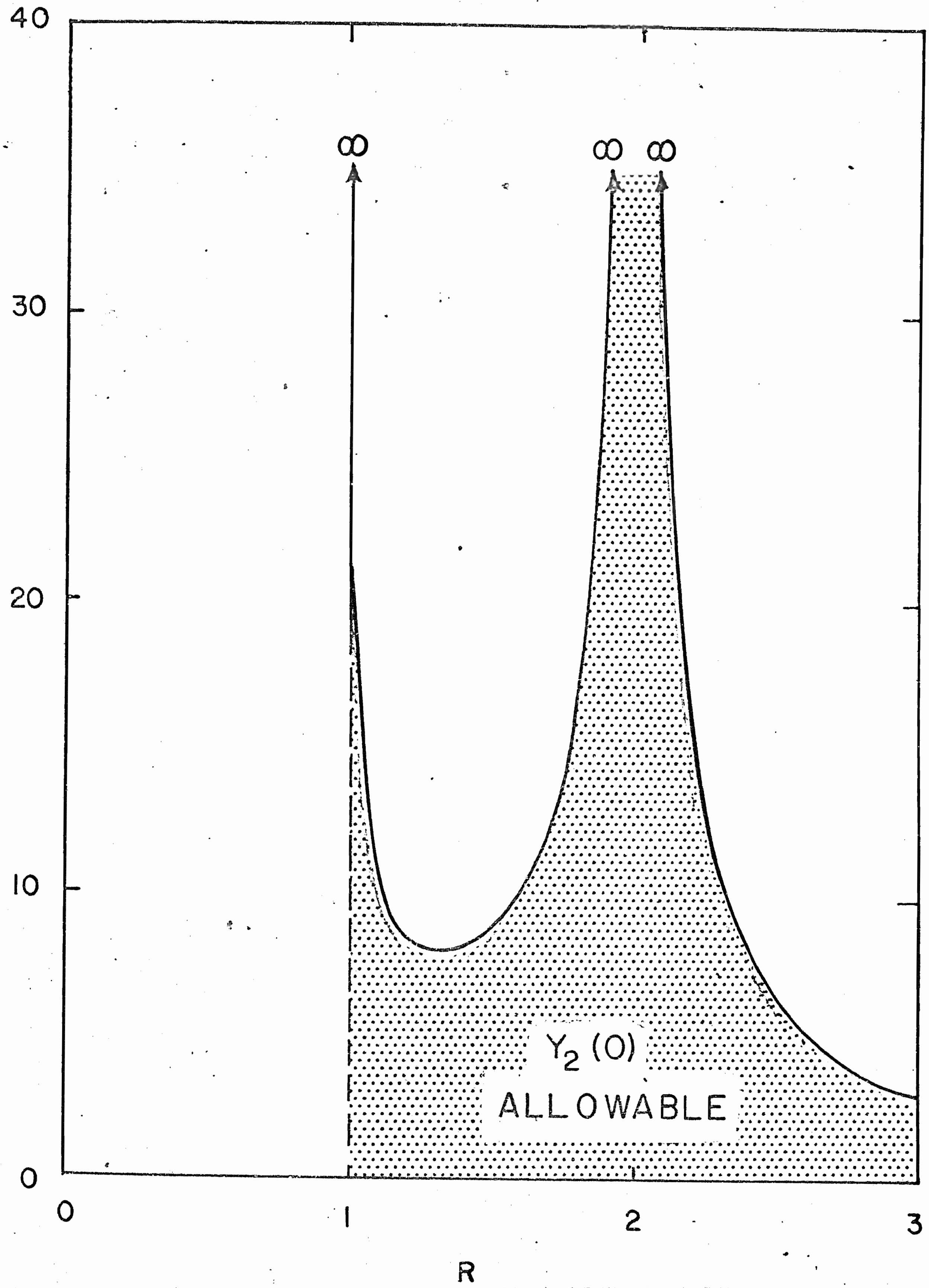

Fig. 1. $Y_2(0)_{max}$ Allowable Vs. R From Third Order Closure Theory

APPENDIX I

A. For $1<R<2$:

1. Y_2 upper bound for fourth order closure

From $\overline{\gamma^4}$ realizability we have that

$$\frac{\overline{\gamma^4}}{Y_1^4} \geq Y_2^3 + 3Y_2^2 - 3Y_2 - 4\frac{\overline{\gamma^3}}{Y_1^3}, \tag{A.1}$$

and since $\overline{\gamma^4}$ is by nature ≥ 0, the most stringent requirement on $\overline{\gamma^4}$ for the case $\overline{\gamma^3} \geq 0$ is given by $\overline{\gamma^3} = 0$, i.e.

$$\frac{\overline{\gamma^4}}{Y_1^4} \geq Y_2^3 + 3Y_2^2 - 3Y_2 . \tag{A.2}$$

For (A.2) to be true for all possible $\overline{\gamma^4}$, i.e. that any $\overline{\gamma^4}$ that is ≥ 0 satisfies $\overline{\gamma^4}$ realizability, implies that

$$Y_2^3 + 3Y_2^2 - 3Y_2 \leq 0, \tag{A.3}$$

$$\text{OR} \quad Y_2 \leq \frac{-3+\sqrt{21}}{2} \simeq 0.79. \tag{A.4}$$

2. $\overline{\gamma^3}/Y_1^3$ upper bound

From (3.7a) without $\overline{\gamma^4}$ term we have that:

$$1 + \frac{R(R-1)}{2}Y_2 + \frac{R(R-1)(R-2)}{6}\frac{\overline{\gamma^3}}{Y_1^3} \geq 0, \tag{A.5}$$

$$\text{OR} \quad \frac{\overline{\gamma^3}}{Y_1^3} \leq \frac{3[2+R(R-1)Y_2]}{R(R-1)(2-R)} . \tag{A.6}$$

3. $\overline{\gamma^3}/Y_1^3$ lower bound

(3.7c) without the $\overline{\gamma^4}$ term reads:

$$Y_2 - \frac{R}{2} Y_2^2 + \frac{R}{2}\left[1 + \frac{(2-R)}{3} Y_2\right] \frac{\overline{Y^3}}{Y_1^3} \geq 0, \tag{A.7}$$

$$\text{OR} \quad \frac{\overline{Y^3}}{Y_1^3} \geq \frac{-Y_2 + \frac{R}{2} Y_2^2}{\frac{R}{2}\left[1 + \frac{(2-R)}{3} Y_2\right]} . \tag{A.8}$$

Thus if (A.8) satisfies $\overline{Y^3}$ realizability then use of (A.8) constitutes simultaneous satisfaction of realizability and requirement on $\frac{dY_2}{dt}$. However, if (A.8) does not satisfy realizability, then use of $\overline{Y^3}$ realizability as the lower bound implies that (A.7) should always be greater than zero, which of course also satisfies the requirement on (3.7c).

Since substitution of the range of R and Y_2 allowable into (A.8) does not yield satisfaction of $\overline{Y^3}$ realizability, then by the previous reasoning, the lower bound on $\overline{Y^3}/Y_1^3$ is given by:

$$\frac{\overline{Y^3}}{Y_1^3} \geq Y_2^2 - Y_2 . \tag{A.9}$$

4. $\overline{Y^4}/Y_1^4$ upper bound

From (3.7c) with $\overline{Y^4}$ term we find that

$$\frac{\overline{Y^4}}{Y_1^4} \leq \frac{\left[Y_2 - \frac{R}{2} Y_2^2 + \frac{R}{2}\left\{1 + \frac{(2-R)}{3} Y_2\right\} \frac{\overline{Y^3}}{Y_1^3}\right]}{[R(2-R)/6][1 + \{(3-R)/4\} Y_2]} . \tag{A.10}$$

5. $\overline{Y^4}/Y_1^4$ lower bound

From $\overline{Y^4}$ realizability we have that

$$\frac{\overline{Y^4}}{Y_1^4} \geq Y_2^3 + 3Y_2^2 - 3Y_2 - 4\frac{\overline{Y^3}}{Y_1^3} . \tag{A.1}$$

While from (3.7a) with $\overline{Y^4}$ term we have that:

$$\frac{\overline{Y^4}}{Y_1^4} \geq \frac{-24\left[1 + \frac{R(R-1)}{2} Y_2 + \frac{R(R-1)(R-2)}{6} \frac{\overline{Y^3}}{Y_1^3}\right]}{R(R-1)(R-2)(R-3)} . \tag{A.11}$$

It is obvious that satisfaction of (A.5) within this range of R implies that (A.11) gives:

$$\frac{\overline{\gamma^4}}{Y_1^4} \geq \text{negative number} \tag{A.12}$$

for any $\overline{\gamma^3}$.

However, use of (A.3) and $\overline{\gamma^3} \leq 0$ leaves the possibility that (A.1) can imply:

$$\frac{\overline{\gamma^4}}{Y_1^4} \geq \text{positive number.} \tag{A.13}$$

Thus, since satisfaction of (A.13) automatically satisfies (A.12), $\overline{\gamma^4}$ realizability is chosen as the lower bound on $\overline{\gamma^4}/Y_1^4$.

6. Y_2 upper bound for third order closure

Since (A.6) is for this range always greater or equal to zero it automatically satisfies $\overline{\gamma^3}$ realizability for $Y_2 \leq 1$. However, for $Y_2 > 1$, requiring realizability to be satisfied implies:

$$\frac{3[2+R(R-1)Y_2]}{R(R-1)(2-R)} \geq Y_2^2 - Y_2 \, . \tag{A.14}$$

Solving for Y_2 yields (As $Y_2 \geq 0$):

$$Y_2 \leq \frac{D + (D^2 + 12C)}{C}, \tag{A.15}$$

where: $D = R(R-1)(5-R)$

$C = 2R(R-1)(2-R)$

Note that since the maximum Y_2 allowed by $\overline{\gamma^4}$ realizability is less than the maximum Y_2 allowed by (A.15) for the 3rd order closure, this implies that for the fourth order moment closure (A.14) also applies.

7. $\overline{Y^4}/Y_1^4{}_{max} \geq \overline{Y^4}/Y_1^4{}_{min}$.

From (A.10)

$$\left.\frac{\overline{Y^4}}{Y_1^4}\right|_{MAX} = \frac{\left[Y_2 - \frac{R}{2}Y_2^2 + \frac{R}{2}\left\{1+\frac{(2-R)}{3}Y_2\right\}\frac{\overline{Y^3}}{Y_1^3}\right]}{[R(2-R)/6][1+\{(3-R)/4\}Y_2]},$$

and from (A.1)

$$\left.\frac{\overline{Y^4}}{Y_1^4}\right|_{MIN} = Y_2^3 + 3Y_2^2 - 3Y_2 - 4\frac{\overline{Y^3}}{Y_1^3}.$$

Thus we want to show that:

$$\frac{\left[Y_2 - \frac{R}{2}Y_2^2 + \frac{R}{2}\left\{1+\frac{(2-R)}{3}Y_2\right\}\frac{\overline{Y^3}}{Y_1^3}\right]}{[R(2-R)/6][1+\{(3-R)/4\}Y_2]} \geq Y_2^3+3Y_2^2-3Y_2-4\frac{\overline{Y^3}}{Y_1^3} \qquad \text{(A.16)}$$

is true for any $\overline{Y^3}$ and Y_2 allowed by (A.4), (A.6), and (A.9).

7a. For $\overline{Y^3} \geq 0$ and $Y_2 \leq 0.79$, (A.10) is always ≥ 0 while (A.1) is always ≤ 0. Thus (A.16) is always satisfied for these conditions.

7b. However, for $\overline{Y^3} \leq 0$, (A.10) is again always ≥ 0, but (A.1) may be less than, equal to, or greater than zero. Thus, for this case (A.16) is not obvious. One way to prove (A.16) is to show that satisfying (A.16) is tantamount to satisfying (A.9), i.e. $\overline{Y^3}$ realizability. From (A.16) one finds that:

$$\frac{\overline{Y^3}}{Y_1^3} \geq \frac{[Y_2^3+3Y_2^2-3Y_2][R(2-R)/6][1+\{(3-R)/4\}Y_2] - Y_2 + \frac{R}{2}Y_2^2}{\frac{R}{2}\left\{1+\frac{(2-R)}{3}Y_2\right\} + 4\left[\frac{R(2-R)}{6}\right]\left[1+\frac{(3-R)}{4}Y_2\right]}. \qquad \text{(A.17)}$$

Thus, since we wish to allow for a full range on $\overline{Y^3}$, for (A.16) to be valid, (A.17) must be related to (A.9), $\overline{Y^3}$ realizability by:

$$\left.\frac{\overline{Y^3}}{Y_1^3}\right|_{(A.17)} \leq \left.\frac{\overline{Y^3}}{Y_1^3}\right|_{(A.9)}. \qquad \text{(A.18)}$$

(A.18), (A.17), and (A.9) can be rearranged to yield:

$$Y_2^3[Y_2-1]\left[\frac{R(2-R)(3-R)}{24}\right]+\frac{Y_2}{6}\left[1-\frac{RY_2}{4}\right]\left[-6+5R-R^2\right]\leq 0. \quad \text{(A.19)}$$

As $Y_2 \leq 0.79$ and $1<R<2$; (A.19) can easily be verified. Thus, any $\overline{\gamma^3}/Y_1^{\ 3}$ that satisfies (A.9), satisfies (A.18), which implies that (A.16) is true, which must be for the terms upper and lower bound on $\overline{\gamma^4}$ to have meaning.

8. Y_2 lower bound

Since $Y_2 = \overline{\gamma^2}/\overline{\Gamma}^2$ physical reality requires that

$$0 \leq Y_2 \quad \text{(A.20)}$$

B. For $2<R\leq 3$:

1. $Y_2|_{max}$ for $\overline{\gamma^4}$ closure same as in Section A.

$$Y_2 \leq 0.79 \quad . \quad \text{(B.1)}$$

2. $\overline{\gamma^3}/Y_1^{\ 3}$ upper bound

As will be seen, the conditions to be satisfied only require a lower bound on $\overline{\gamma^3}/Y_1^{\ 3}$. Thus,

$$\overline{\gamma^3}/Y_1^{\ 3} < \infty \ . \quad \text{(B.2)}$$

3. $\overline{\gamma^3}/Y_1^{\ 3}$ lower bound

From (3.7a) without $\overline{\gamma^4}$ term, (A.5) we now find that:

$$\frac{\overline{\gamma^3}}{Y_1^3} \geq \frac{-3\left[2+R(R-1)Y_2\right]}{R(R-1)(2-R)} \ . \quad \text{(B.3)}$$

However, since (B.3) does not satisfy $\overline{\gamma^3}$ realizability for the full range of allowable Y_2, e.g. let $Y_2 = 0$, the lower bound between these is $\overline{\gamma^3}$ realizability.

Thus

$$\left.\frac{\overline{Y^3}}{Y_1^3}\right|_{MIN} \geqslant Y_2^2 - Y_2 \quad . \tag{B.4}$$

But if in (3.7c) the size of Y_2 is restricted such that the following is true

$$\frac{\overline{Y^3}}{Y_1^3} \geqslant \frac{-Y_2 + \frac{R}{2}Y_2^2}{\frac{R}{6}\left[3+(2-R)Y_2\right]} \quad . \tag{A.8}$$

Then an examination of (B.4) and (A.8) reveals that the following is true

$$\frac{-Y_2 + \frac{R}{2}Y_2^2}{\frac{R}{6}\left[3+(2-R)Y_2\right]} \geqslant Y_2^2 - Y_2 \, . \tag{B.5}$$

Thus satisfaction of (A.8) implies satisfaction of requirements from $\frac{dY_1}{dt}$, $\frac{dY_2}{dt}$, and $\overline{Y^3}$ realizability. Therefore:

$$\left.\frac{\overline{Y^3}}{Y_1^3}\right|_{MIN} \geqslant \frac{-Y_2 + \frac{R}{2}Y_2^2}{(R/6)\left[3+(2-R)Y_2\right]} \, . \tag{B.6}$$

4. $Y_{2\,max}$ for $\overline{Y^3}$ closure. Since no $\overline{Y^4}$ realizability is needed, (A.4) does not hold. However for $\overline{Y^3}$ closure, in order for (A.8) to be true, the following must be true in (A.7) or (3.7c):

$$1 + \frac{2-R}{3} Y_2 \geqslant 0 \, , \tag{B.7}$$

$$\text{OR } \left. Y_2 \right|_{MAX} \leqslant \frac{3}{R-2} \, . \tag{B.8}$$

5. $\overline{Y^4}/Y_1^4$ upper bound

From (3.7a) with $\overline{Y^4}$ term, we find that

$$\frac{\overline{Y^4}}{Y_1^4} \leqslant \frac{24\left[1 + \frac{R(R-1)}{2}Y_2 + \frac{R(R-1)(R-2)}{3}\frac{\overline{Y^3}}{Y_1^3}\right]}{R(R-1)(R-2)(3-R)} \, , \tag{B.9}$$

and since (B.9) is always >0 (see A.5) it at least satisfies the physical reality of $\overline{Y^4}$.

6. $\overline{Y^4}/Y_1^4$ lower bound

From (3.7c) with $\overline{Y^4}$ term

$$\frac{\overline{Y^4}}{Y_1^4} \geq \frac{-\left[Y_2 - \frac{R}{2}Y_2^2 + \frac{R}{2}\left\{1 + \frac{(2-R)}{3}Y_2\right\}\frac{\overline{Y^3}}{Y_1^3}\right]}{[R(R-2)/6][1+\{(3-R)/4\}Y_2]} . \tag{B.10}$$

Thus from (A.7), (B.10) is always negative. However, from $\overline{Y^4}$ realizability

$$\frac{\overline{Y^4}}{Y_1^4} \geq Y_2^3 + 3Y_2^2 - 3Y_2 - 4\frac{\overline{Y^3}}{Y_1^3} , \tag{A.1}$$

which may be a positive or negative number. Thus, since $\overline{Y^4}$ must physically be greater or equal to zero, use of (A.1) as a lower bound automatically satisfies (B.10). Therefore:

$$\left.\frac{\overline{Y^4}}{Y_1^4}\right|_{MIN} \geq Y_2^3 + 3Y_2^2 - 3Y_2 - 4\frac{\overline{Y^3}}{Y_1^3} . \tag{B.11}$$

7. $Y_2|_{min}$ same as (A.20).