STATE UNIVERSITY OF NEW YORK AT STONY BROOK

COLLEGE OF ENGINEERING

Report No. 124

ACCURACY OF CLOSURES FOR STOCHASTICALLY DISTRIBUTED ARBITRARY ORDER REACTANTS

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Edward E. O'Brien

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Ronald M. Eng.

Department of Mechanics
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ABSTRACT

A comparison between the exact and approximate theoretical solutions for closures at the third and fourth order moment is presented for the problem of the decay of reactants which obey an arbitrary order equation (the order being limited to be between 1 and 3) and whose initial description is given stochastically. The closures satisfy prescribed realizability and asymptotic conditions for certain ranges of initial values of the mean, mean square fluctuations, third order moments, and fourth order moments of the concentration field. The restrictions on initial values are given in terms of dimensionless ratios that are independent of the form of the closures.

The closures were applied to two different probability distributions to determine the accuracy of the closures. It was found that: (1) Violation of the prescribed limits on the initial values of the moments led to severely unphysical, unacceptable behavior in the time history of some or all of the moments considered, and (2) When initial moments were within the allowable limits, proper qualitative behavior was exhibited in all cases. However, quantitative accuracy as determined by comparison with exact stochastic solutions was greatest for the second order reaction and was reduced somewhat as the order diverged from 2. It was further found for the case when all initial moments were such that a fourth order closure was possible that the results were not significantly more accurate than those predicted by a third moment closure.

I. INTRODUCTION

O'Brien and Eng[1] have presented simple two term closure forms at the third and fourth order moments for the problem of the decay of reactants that obey an equation whose order is between 1 and 3.

The forms do not vary for various order equations or for various initial data. However, certain limits have been found which deliniate the ranges within which realizability and proper physical behavior of the evolution of the quantity under consideration can be obtained. These limits have been given[1] in terms of dimensionless ratios that are independent of the closure forms used.

O'Brien has already shown[2] that a simple two term third order closure form can closely model the exact solutions for the case of second order reactions. The objective of this report is to determine the accuracy of similar simple closure forms for the case of arbitrary order reactions.

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 were used to determine the forms of the moment closures to be presented:

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

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

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

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

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 $N = 1, 2, 3, 4$.

By keeping certain requirements in mind[1], 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 not prescribed, we cannot use (2.9), even to the extent of evaluating $\frac{d\overline{\gamma^3}}{dt}(0)$.

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

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

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

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

$$\overline{\gamma^4(t)} > \overline{\gamma^2(t)}^3/\overline{\Gamma(t)}^2 + 3\overline{\gamma^2}^2 + - 3\overline{\gamma^2}\;\overline{\Gamma}^2 - 4\overline{\gamma^3}\;\overline{\Gamma}. \tag{2.13}$$

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 presented that have been shown[1] to satisfy the realizability conditions (2.10), (2.11), (2.12), and (2.13) for all values of $\overline{\Gamma}(0)$, $\overline{\gamma^2}(0)$, $\overline{\gamma^3}(0)$, and $\overline{\gamma^4}(0)$ which themselves do not violate (2.10), (2.11), (2.12), and (2.13). We have also required that the asymptotic behaviors given by (2.3), (2.4), (2.5), and (2.6) are satisfied. The closures are simple functional forms that do not vary for different initial data.

III. CLOSURES AND THEORETICAL RESULTS

The third order moment closure

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

and the fourth order moment closure

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

have been shown[1] to satisfy realizability, physical behavior, and asymptotic requirements if certain conditions are met. The results presented employ the following definitions:

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

$$Y_2 = \overline{\gamma^2(t)} \, \overline{\Gamma(t)}^{-2}, \tag{3.3b}$$

$$S = \overline{\gamma^3(t)} \, \overline{\gamma^2(t)}^{-3/2}. \tag{3.3c}$$

A. Fourth Order Moment Requirements

Using the assumed third and fourth order moment closures and requiring that realizability and physical requirements on the Y_1 and Y_2 moment equations are satisfied, yields the following requirements:

For $1 < R < 2$:

$$0 \leq Y_2(0) \leq (-3+\sqrt{21})/2 \approx 0.79, \tag{3.4a}$$

$$\frac{\overline{\gamma^3(0)}}{Y_1^3(0)} < \frac{3[2+R(R-1)Y_2(0)]}{R(R-1)(2-R)}, \tag{3.4b}$$

$$\frac{\overline{Y^3(0)}}{Y_1^3(0)} \geq Y_2^2(0) - Y_2(0), \tag{3.4c}$$

$$\frac{\overline{Y^4(0)}}{Y_1^4(0)} \leq 4\left[6Y_2(0) - 3RY_2^2(0) + R\{3 + (2-R)Y_2(0)\} \times \right.$$
$$\left. \times \frac{\overline{Y^3(0)}}{Y_1^3(0)}\right]\left[R(2-R)\{4 + (3-R)Y_2(0)\}\right]^{-1}, \tag{3.4d}$$

$$\frac{\overline{Y^4(0)}}{Y_1^4(0)} \geq Y_2^3(0) + 3Y_2^2(0) - 3Y_2(0) - 4\frac{\overline{Y^3(0)}}{Y_1^3(0)}. \tag{3.4e}$$

By substituting closures (3.1) and (3.2) into (3.4b) through (3.4e), it can be shown that the constants used in the closures must be such that:

$$A_0 \geq -\{Y_2(0)(3/(2-R) - A_1) +$$
$$+ [R(R-1)(2-R)]^{-1}\} Y_2(0)^{-3/2}, \tag{3.4f}$$

$$A_0 \leq 2(A_1 - 1)^{\frac{1}{2}} \text{ FOR } A_1 > 1, \tag{3.4g}$$

$$B_1 \leq 4[6 - 3RY_2(0) + R\{3 + (2-R)Y_2(0)\} \times$$
$$\times \{A_1 Y_2(0) - A_0 Y_2^{\frac{1}{2}}(0)\}][Y_2(0)R(2-R) \times$$
$$\times (4 + [3-R]Y_2(0))]^{-1} + B_2 Y_2(0), \tag{3.4h}$$

$$B_1 \geq Y_2(0) + 3 - 3Y_2^{-1}(0) - 4[A_1 - A_0 \times$$
$$\times Y_2^{-\frac{1}{2}}(0)] + B_2 Y_2(0), \tag{3.4i}$$

and for $2 < R \leq 3$:

$$0 \leq Y_2(0) \leq (-3 + \sqrt{21})/2 \approx 0.79, \tag{3.4j}$$

$$\frac{\overline{Y^3(0)}}{Y_1^3(0)} < \infty, \tag{3.4k}$$

$$\frac{\overline{Y^3(0)}}{Y_1^3(0)} \geq \frac{-Y_2(0) + (R/2)Y_2^2(0)}{(R/6)[3 + (2-R)Y_2(0)]}, \tag{3.4l}$$

$$\frac{\overline{Y^4(0)}}{Y_1^4(0)} \leq 24\Big[1 + (R(R-1)/2)Y_2(0) + (R(R-1)(R-2)/6) \times \times \frac{\overline{Y^3(0)}}{Y_1^3(0)}\Big]\Big[R(R-1)(R-2)(3-R)\Big]^{-1}, \tag{3.4m}$$

$$\frac{\overline{Y^4(0)}}{Y_1^4(0)} \geq Y_2^3(0) + 3Y_2^2(0) - 3Y_2(0) - 4\frac{\overline{Y^3(0)}}{Y_1^3(0)}. \tag{3.4n}$$

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

$$-\infty < A_0 \leq \frac{3[Y_2(0)(A_1-1) + A_1(2-R)/3\, Y_2^2(0) + 2/R]}{Y_2^{\frac{1}{2}}(0)\,(5-R)}. \tag{3.4o}$$

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

$$\frac{(R/6)(2-R)\,Y_2(0)\,\overline{Y^3(0)}/Y_1^3(0)}{Y_2(0) - (R/2)Y_2^2(0) + (R/2)\overline{Y^3(0)}/Y_1^3(0)} \ll 1, \tag{3.4p}$$

then it can be shown that (3.4o) may be simplified to yield the requirement that A_o be such that:

$$-\infty < A_0 \leq 2[(2/R)(A_1-1)]^{\frac{1}{2}}\ ;\ A_1 > 1. \tag{3.4q}$$

Returning to generally applicable results, we find that:

$$B_1 \leq 4[A_1 - A_0 Y_2^{-\frac{1}{2}}(0)](3-R)^{-1} + 12[2 + R(R-1)Y_2(0)] \times \times [Y_2^2(0)R(R-1)(R-2)(3-R)]^{-1} + B_2 Y_2(0), \tag{3.4r}$$

and

$$B_1 \geq Y_2(0) + 3 - 3Y_2^{-1}(0) - 4[A_1 + \\ - A_0 \cdot Y_2^{-\frac{1}{2}}(0)] + B_2 Y_2(0). \qquad (3.4)$$

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

B. Third Order Moment Closure

If the $\overline{\gamma^4}$ terms are dropped from the Y_1 and Y_2 moment equations, and thus only a third order moment closure is required, then $\overline{\gamma^3}$ realizability and physical requirements on Y_1 and Y_2 require that:

For $1 < R < 2$:

$$0 \leq Y_2(0) \leq [R(R-1)(5-R) + \{[R(R-1)(5-R)]^2 + \\ + 24R(R-1)(2-R)\}^{\frac{1}{2}}][2R(R-1)(2-R)]^{-1}, \qquad (3.5a)$$

$$Y_2^2(0) - Y_2(0) \leq \overline{\gamma^3(0)}\, Y_1^{-3}(0) \leq 3[2 + \\ + R(R-1)Y_2(0)][R(R-1)(2-R)]^{-1}, \qquad (3.5b)$$

$$A_0 \geqslant -[Y_2(0)\{3(2-R)^{-1} - A_1\} + \{R(R-1) \times \\ \times (2-R)\}^{-1}]\, Y_2^{-3/2}(0), \qquad (3.5c)$$

$$A_0 \leq 2(A_1 - 1)^{\frac{1}{2}}\ ;\ A_1 > 1, \qquad (3.5d)$$

and for $2 < R \leq 3$:

$$0 \leq Y_2(0) \leq 3(R-2)^{-1}, \qquad (3.5e)$$

$$\frac{-Y_2(0)+(R/2)Y_2^2(0)}{(R/6)[3+(2-R)Y_2(0)]} < \frac{\overline{Y^3(0)}}{Y_1^3(0)} < \infty \,, \tag{3.5f}$$

$$-\infty < A_0 \leq 3[Y_2(0)(A_1-1)+A_1\{(2-R)/3\}\times \times Y_2^2(0)+2R^{-1}]\,Y_2^{-\frac{1}{2}}(0)(5-R)^{-1}, \tag{3.5g}$$

and if ;

$$[(R/6)(2-R)Y_2(0)\overline{Y^3(0)}\,Y_1^{-3}(0)]\times \times[Y_2(0)-(R/2)Y_2^2(0)+(R/2)\times \times \overline{Y^3(0)}\,Y_1^{-3}(0)]^{-1} \ll 1. \tag{3.5h}$$

Then, as before, (3.5g) is simplified to:

$$-\infty < A_0 \leq 2[(2/R)(A_1-1)]^{\frac{1}{2}};\; A_1 > 1. \tag{3.5i}$$

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. 19) shows that the maximum allowable value of the ratio for the third order moment closure is at least three times that allowed by the fourth order moment closure.

IV. APPLICATIONS OF THE CLOSURES

Since the major interest of this paper is to determine the accuracy of closures which have already been shown[1] to give at least physically acceptable results for certain ranges of initial values, it is of interest to demonstrate their behavior by a few typical cases. Also, in view of the relationship (2.2), it is possible to compare the results predicted by the closures with those obtained exactly when explicit initial probability distributions, $P(X)$, of the concentration field are prescribed.

The comparisons were carried out using the following distributions:

$$P(x) = (3.5!)^{-1} x^{3.5} e^{-x}, \tag{4.1}$$

$$P(x) = 60 x^2 (1+x)^{-7}. \tag{4.2}$$

These are the same distributions O'Brien used[2]. In particular: (4.1) had initial values that satisfied all third order moment closure and all fourth order moment closure requirements, and (4.2) had initial values that satisfied all third order, but hardly any of the fourth order requirements. The exact details are contained in Tables I and II.

Thus the end result of this procedure is:

1. To highlight the more restrictive nature of the fourth order moment closure, and
2. To emphasize that although the third order moment closure for non-second order reactions is not as generally applicable as for the second order,

it still has enough flexibility to be of fairly general use.

Exact solutions were obtained by substituting (4.1) and (4.2) into (2.2) and numerically integrating on a IBM 360 computer.

Approximate solutions were obtained from (2.7) and (2.8) (suitably recast[2]) with initial conditions specified by the exact solutions.

For each distribution, the quantities Y_1, Y_2 and S are plotted as functions of time with the order of reaction, R, as a parameter. For each quantity, there are three graphs arranged as follows: the exact solution, third order moment closure solution, and fourth order moment closure solution. The order of appearance of distributions is: Fig. 1-9 for (4.1), and Figs. 10-18 for (4.2).

A comparison of the results presented for the particular distributions considered shows:

1. The third order moment closure requirements allow one to handle a distribution whose $Y_2(0)$ is at least three times that allowed by a fourth order moment closure. Thus, the third order closure is demonstrably applicable to many more cases than the fourth order closure (see Fig. 19).
2. However, it was found that for the third order closure, the time histories for $R<2$ (for the distribution where $Y_2(0)\leq 1$) could not be made as accurate as those for $R>2$. Further, the $R\neq 2$ solutions were inaccurate compared to the $R=2$ solutions. Details are presented in Table III for (4.2) only.
3. Violation of any of the requirements set forth in

(3.4) and (3.5) led to serious, quite unacceptable unphysical behavior in some or all of the variables.

4. For (4.1), which does satisfy all requirements set by (3.4) and (3.5), it seems that use of a fourth order moment closure does not significantly improve the accuracy of the results in comparison to those from the third order moment closure.

V. CONCLUSIONS

The results of the preceding section indicate that for the cases considered a simple two term expansion form for closures at the third and fourth order moment does yield solutions that are globally satisfactory when all prescribed limits are satisfied. However, it is also evident that solutions for the non-second order reaction equation are not as accurate as those for the second order equation. The inaccuracies are due to: 1) the truncation of what were originally infinite series moment equations and, 2) the number and types of terms used in the closure forms.

The first source of error would seem to be alleviated by accounting for more terms in the moment equations. However, this leads to the problems of specifying the closure forms and of even having these higher moments accurately available from experiments for comparison with the theoretical results. Besides, as shown by our results, increasing the number of moments accounted for in the moment equations not only does not seem to markedly increase the accuracy of results but also introduces additional restrictions, which reduce the general applicability of the method.

The second source of error may seem to be most easily suppressed by using more terms in the suggested closure forms. Thus, since very good accuracy was obtained in the second order case by forming a closure that automatically satisfied realizability[2], a plausible scheme would be to force this condition for the non-second order reaction. This implies having a closure whose functional form varies explicitly with R. However, as remarked in Ref. [1], the nature of the realizability conditions precludes the formulation of such a closure in its exact form. This coupled with the fairly wide range of values possible for the arbitrary constants involved, makes it very difficult to predict a priori the number and kinds of terms that best fit a particular distribution function.

However, as has been shown, quite satisfactory, globally correct results may be obtained by using a simple two term closure in truncated infinite series moment equations.

ACKNOWLEDGMENT

The authors are grateful to the National Science Foundation for the support of this work under Grant GK 1715.

The authors also thank the Computing Center of the State University of New York at Stony Brook for the aid of their equipment and personnel.

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TABLE I.

ALLOWABLE RATIOS AS A FUNCTION OF R FOR $P(X) = (3.5!)^{-1} x^{3.5} e^{-x}$

Ratio \ R	1.25	1.50	2.00	2.50	3.00
a	-0.173	-0.173	-0.173	-0.133	-0.107
b	0.099	0.099	0.099	0.099	0.099
c	26.49	17.33	∞	∞	∞
d	-0.903	-0.903	Any	-0.903	-0.903
e	0.214	0.214	Any	0.214	0.214
f	1.49	1.93	Any	37.06	∞

a = Minimum allowable value of $\overline{Y^3(0)}\ Y_1^{-3}(0)$

b = Value of $\overline{Y^3(0)}\ Y_1^{-3}(0)$

c = Maximum allowable value of $\overline{Y^3(0)}\ Y_1^{-3}(0)$

d = Minimum allowable value of $\overline{Y^4(0)}\ Y_1^{-4}(0)$

e = Value of $\overline{Y^4(0)}\ Y_1^{-4}(0)$

f = Maximum allowable value of $\overline{Y^4(0)}\ Y_1^{-4}(0)$

TABLE II.
ALLOWABLE RATIOS AS A FUNCTION OF R FOR $P(X) = 60x^2(1+x)^{-7}$

Ratio \ R	1.25	1.50	2.00	2.50	3.00
a	0.0	0.0	0.0	0.24	0.50
b	5.88	5.88	5.88	5.88	5.88
c	29.6	22.0	∞	∞	∞
d	-22.5	-22.5	Any	-22.5	-22.5
e	196.2	196.2	Any	196.2	196.2
f	22.1	31.4	Any	120.7	∞

a = Minimum allowable value of $\overline{Y^3(0)}\,Y_1^{-3}(0)$

b = Value of $\overline{Y^3(0)}\,Y_1^{-3}(0)$

c = Maximum allowable value of $\overline{Y^3(0)}\,Y_1^{-3}(0)$

d = Minimum allowable value of $\overline{Y^4(0)}\,Y_1^{-4}(0)$

e = Value of $\overline{Y^4(0)}\,Y_1^{-4}(0)$

f = Maximum allowable value of $\overline{Y^4(0)}\,Y_1^{-4}(0)$

TABLE III.
INITIAL SLOPES FOR $P(X) = 60x^2(1+x)^{-7}$

A. Exact Expression

$$\left.\frac{dY_2}{dt}\right]_{t=0} = -360(R-1)\left\{\frac{\Gamma(R+3)\Gamma(3-R)}{\Gamma(7)}\right\}$$

R	$\left.\frac{dY_2}{dt}\right]_{t=0}$
1.00	0.0
1.25	-1.25
1.50	-2.67
2.00	-12.00
2.50	-69.54
3.00	Undefined

B. Approximate Expression

$$\left.\frac{dY_2}{dt}\right]_{t=0} = -2(R-1)\left[1+(R/2)\{(A_1-A_0)(5-R)/3-1\}\right]$$

WHERE $A_1 = 7.88$, $A_0 = 2.0$

R	$\left.\frac{dY_2}{dt}\right]_{t=0}$
1.00	0.0
1.25	-2.48
1.50	-5.40
2.00	-11.76
2.50	-17.63
3.00	-21.52

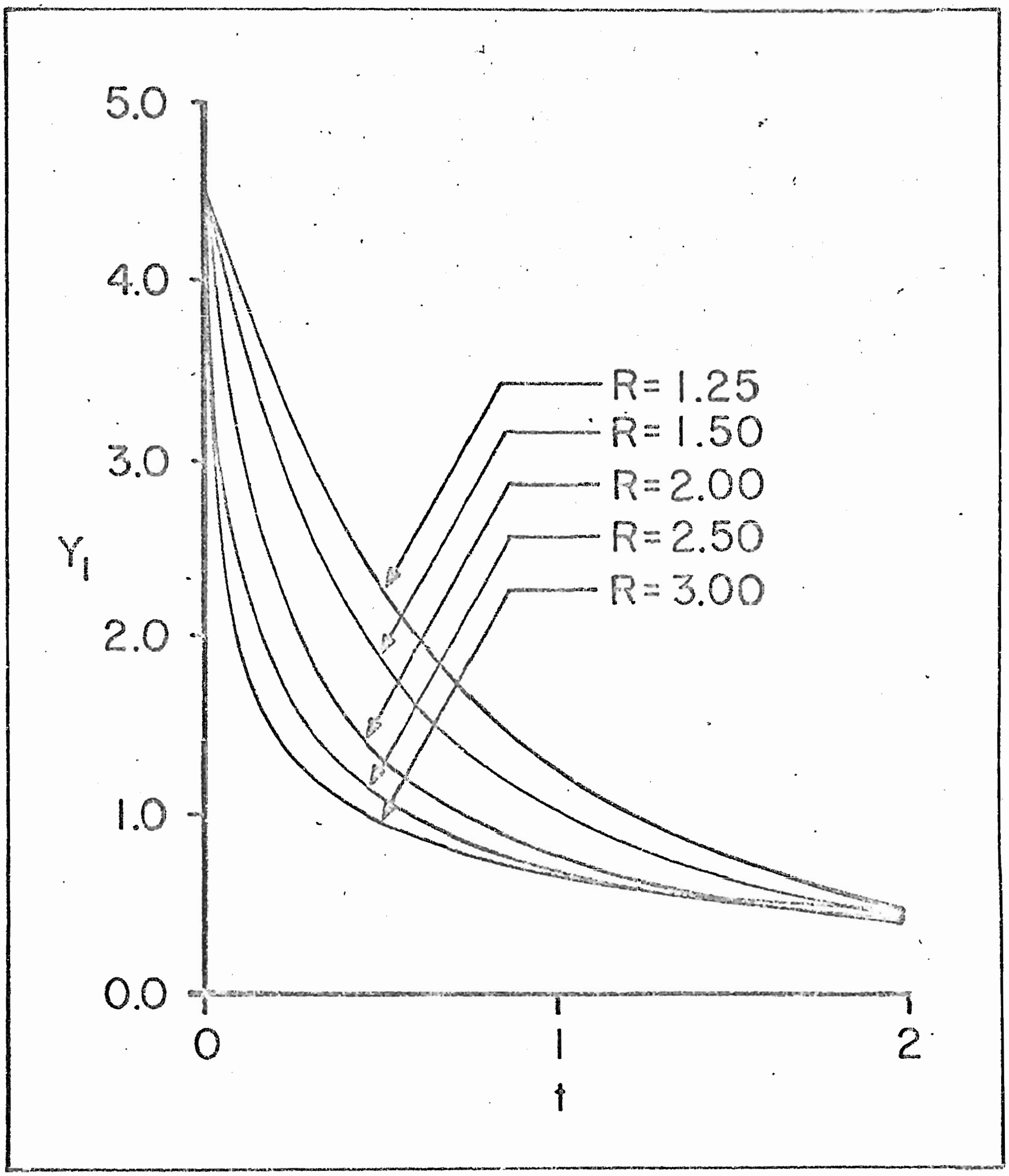

Fig. 1. Exact Solution for Dependence of Mean Concentration on Dimensionless Time for Probability Distribution (4.1).

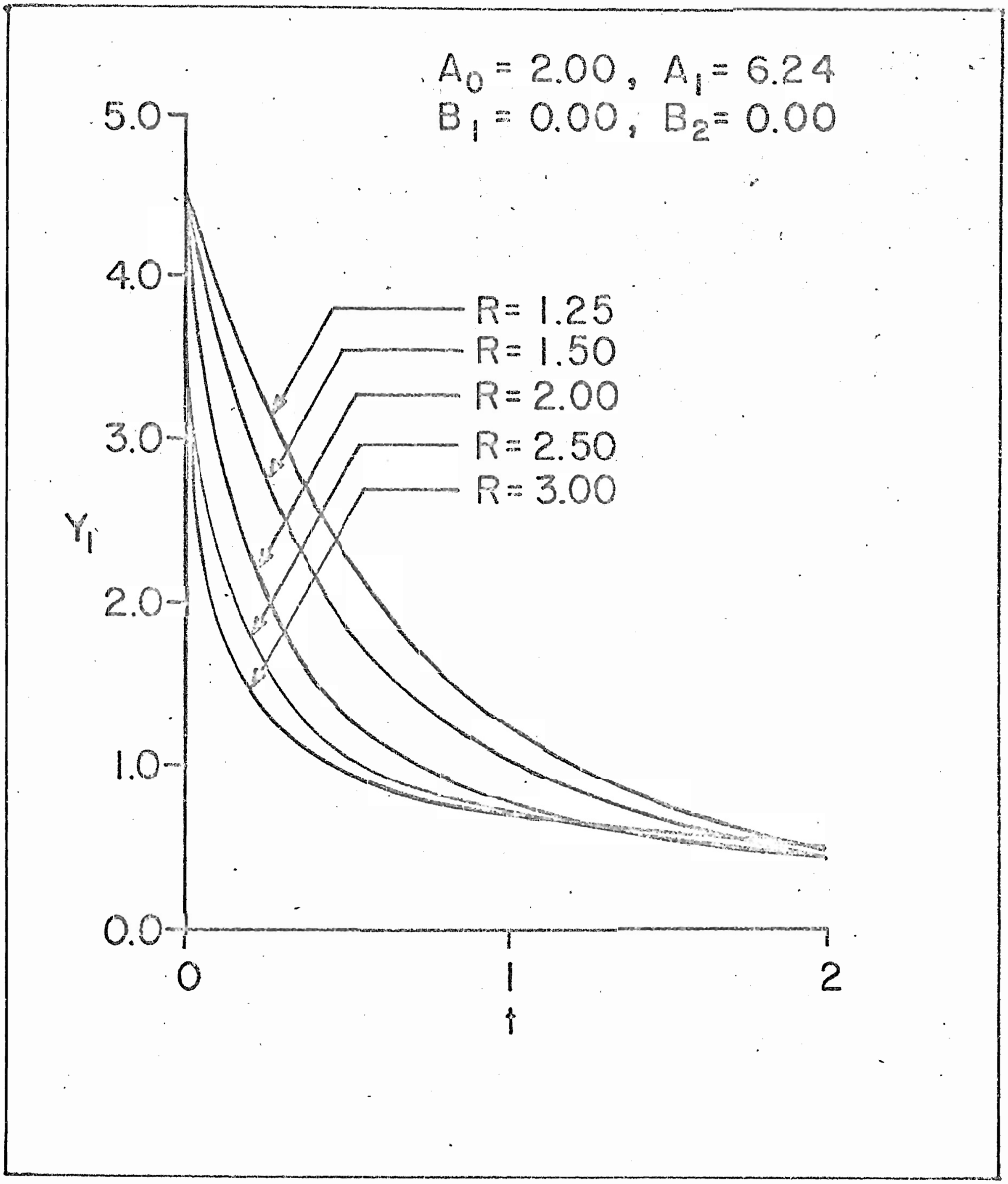

Fig. 2. Third Order Moment Closure Solution for Dependence of Mean Concentration on Dimensionless Time for Probability Distribution (4.1).

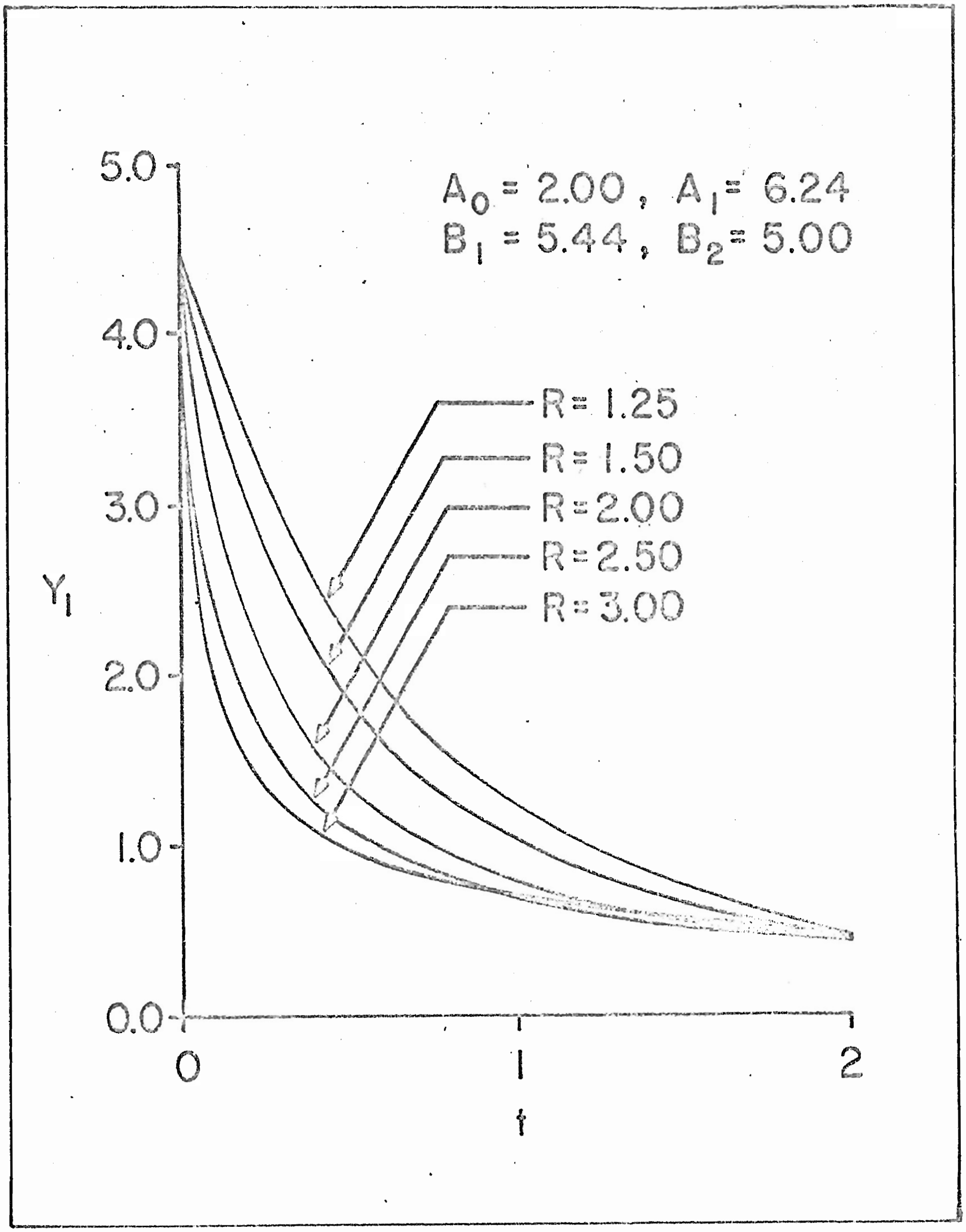

Fig. 3. Fourth Order Moment Closure Solution for Dependence of Mean Concentration on Dimensionless Time for Probability Distribution (4.1).

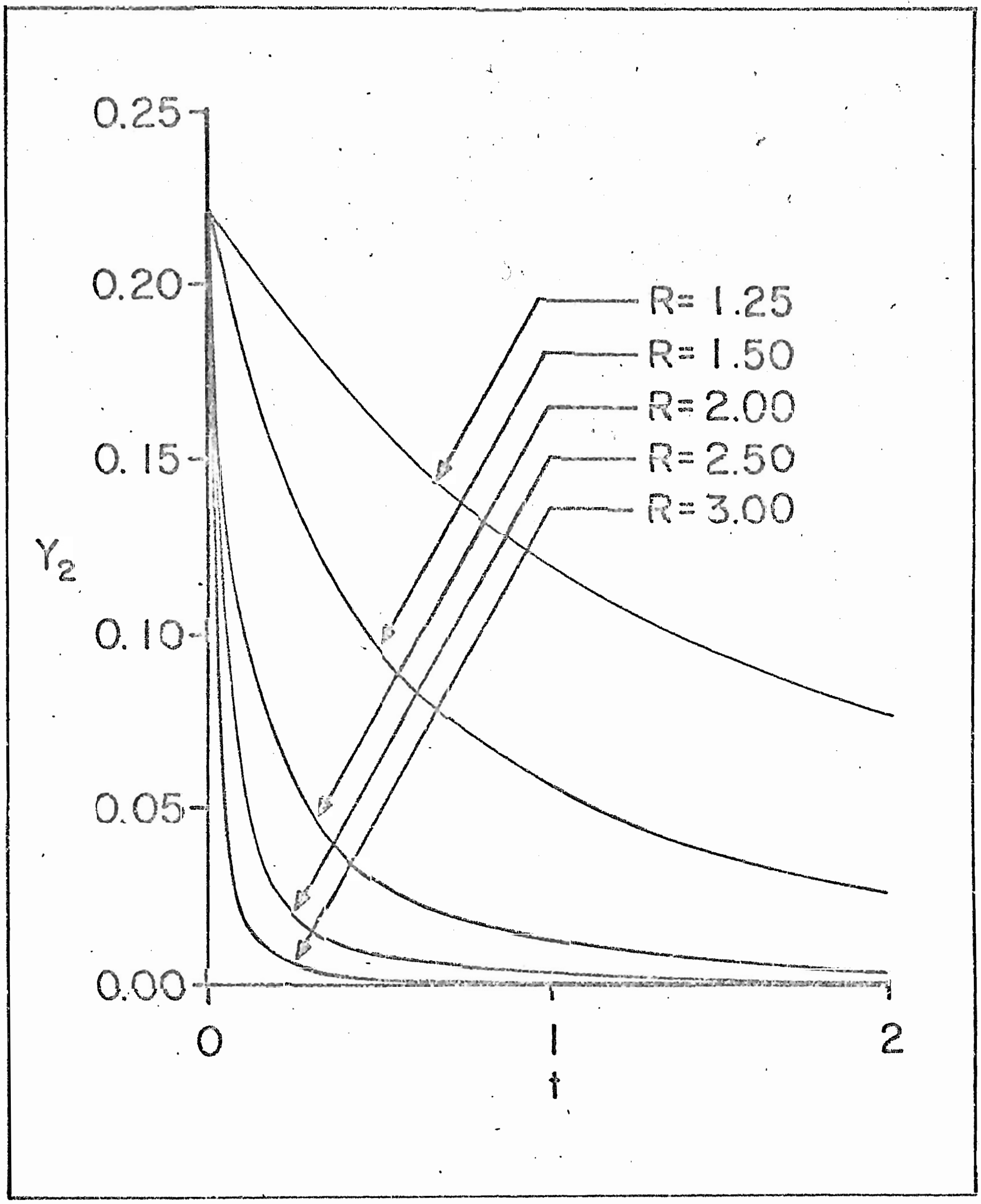

Fig. 4. Exact Solution for Dependence of Concentration Relative Intensity on Dimensionless Time for Probability Distribution (4.1).

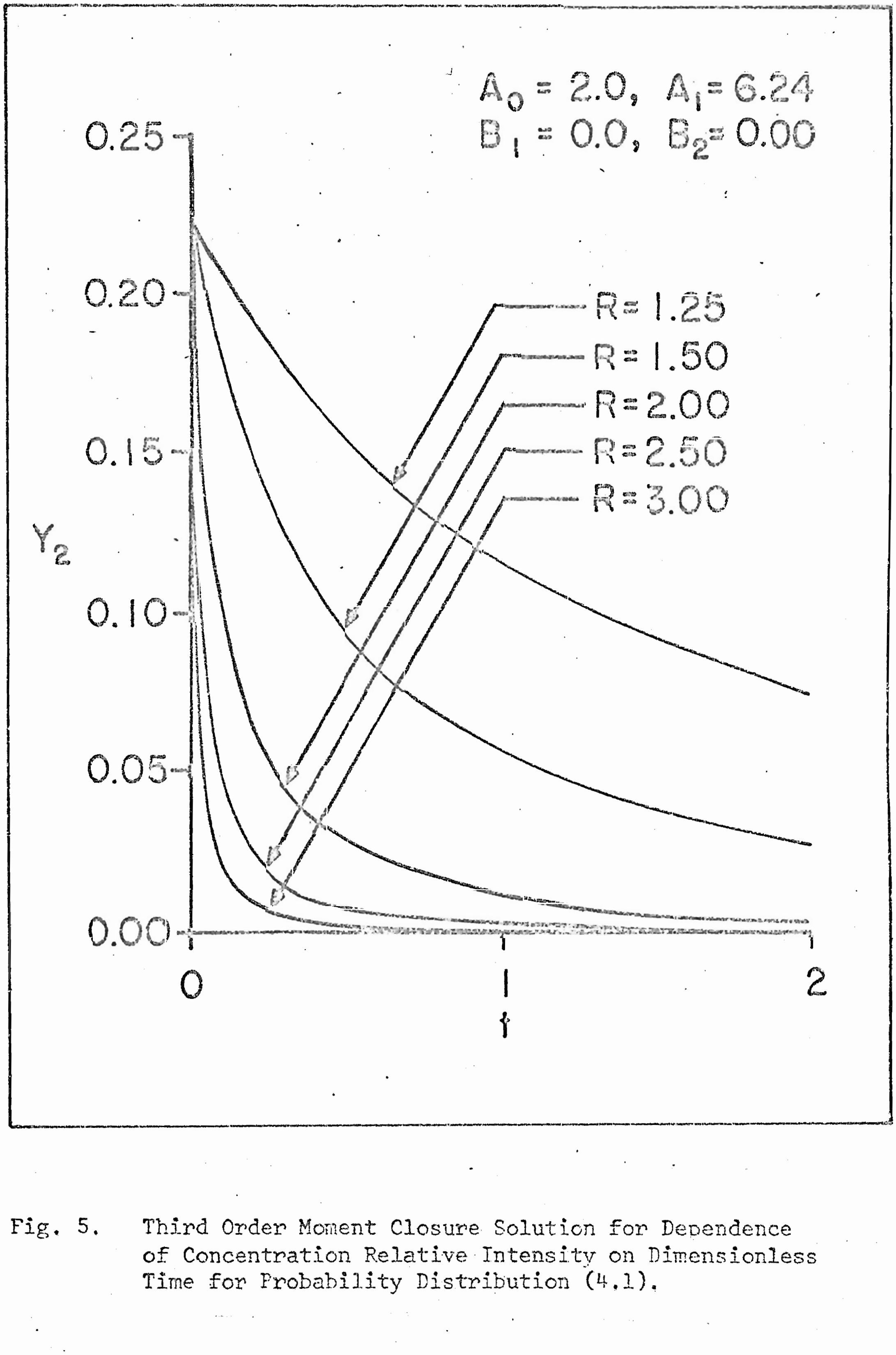

Fig. 5. Third Order Moment Closure Solution for Dependence of Concentration Relative Intensity on Dimensionless Time for Probability Distribution (4.1).

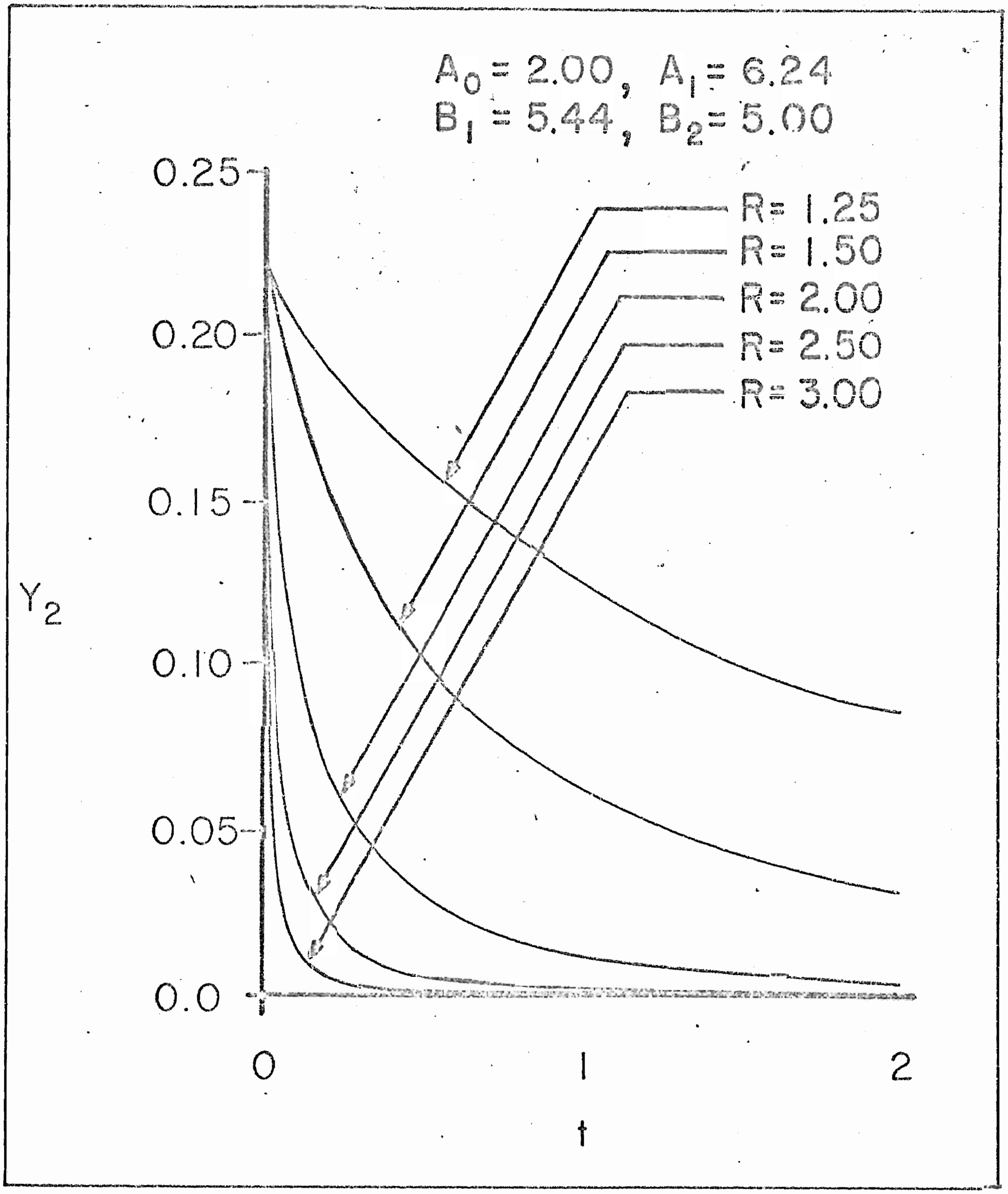

Fig. 6. Fourth Order Moment Closure Solution for Dependence of Concentration Relative Intensity on Dimensionless Time for Probability Distribution (4.1).

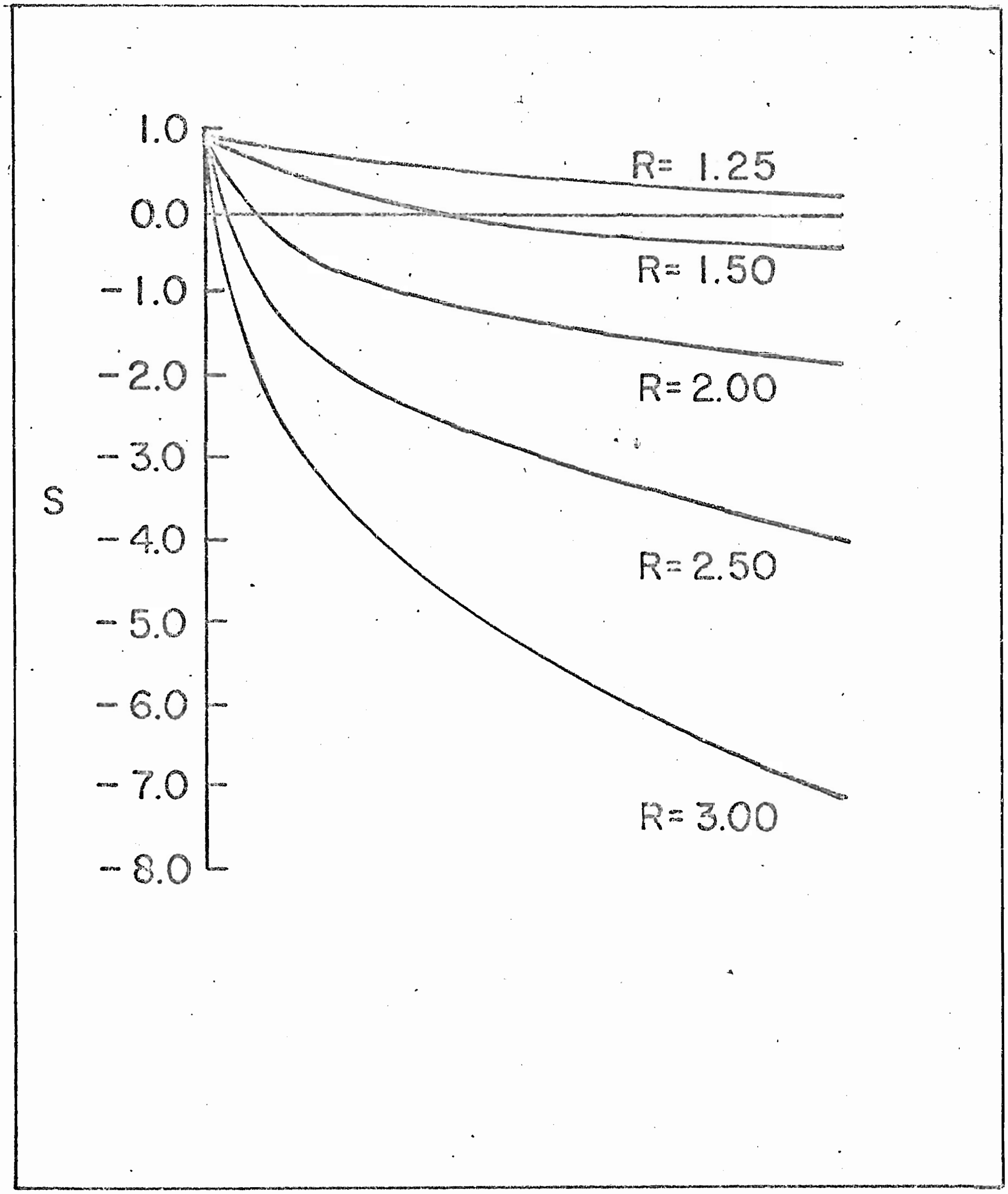

Fig. 7. Exact Solution for Dependence of Concentration Skewness on Dimensionless Time for Probability Distribution (4.1).

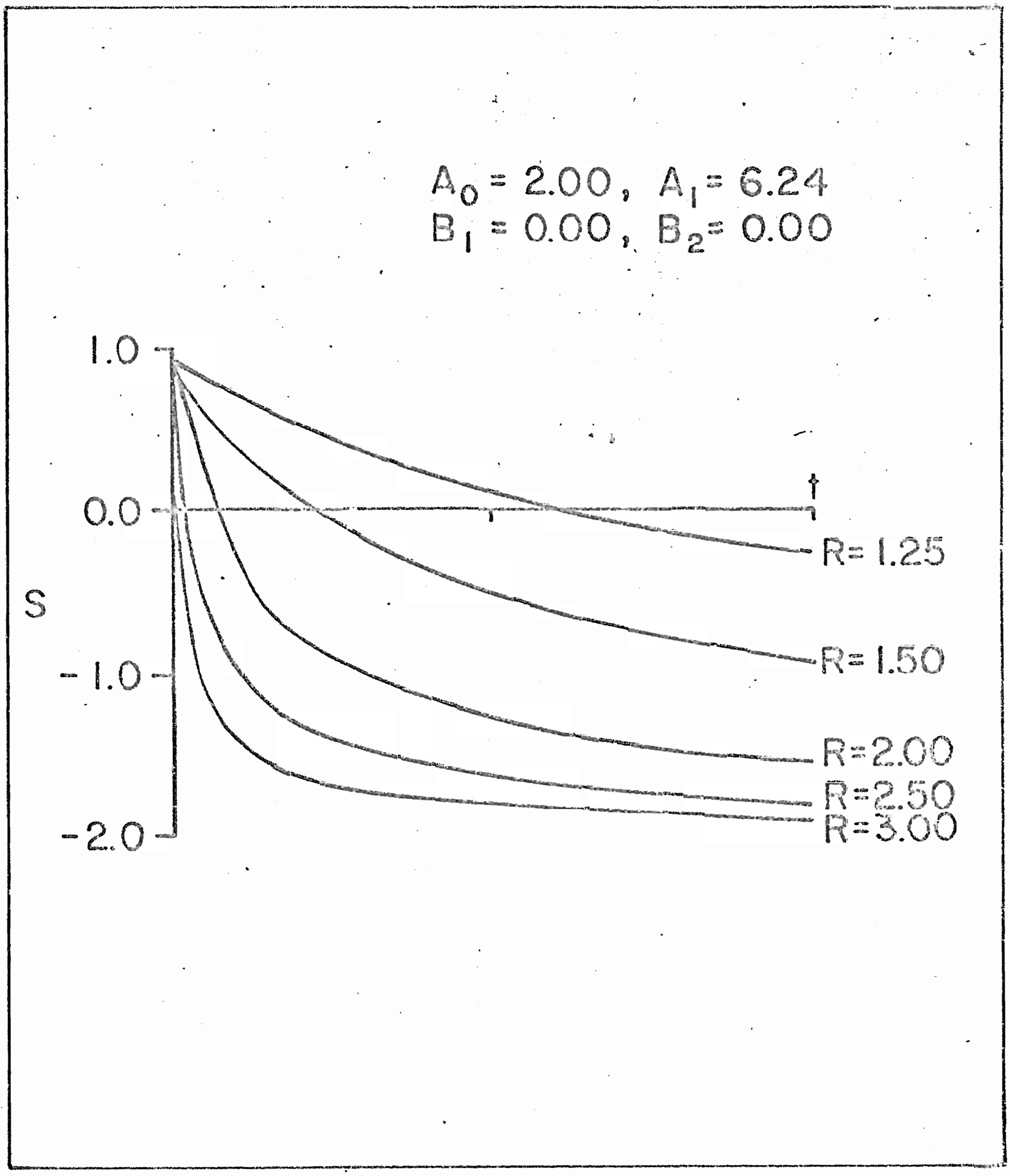

Fig. 8. Third Order Moment Closure Solution for Dependence of Concentration Skewness on Dimensionless Time for Probability Distribution (4.1).

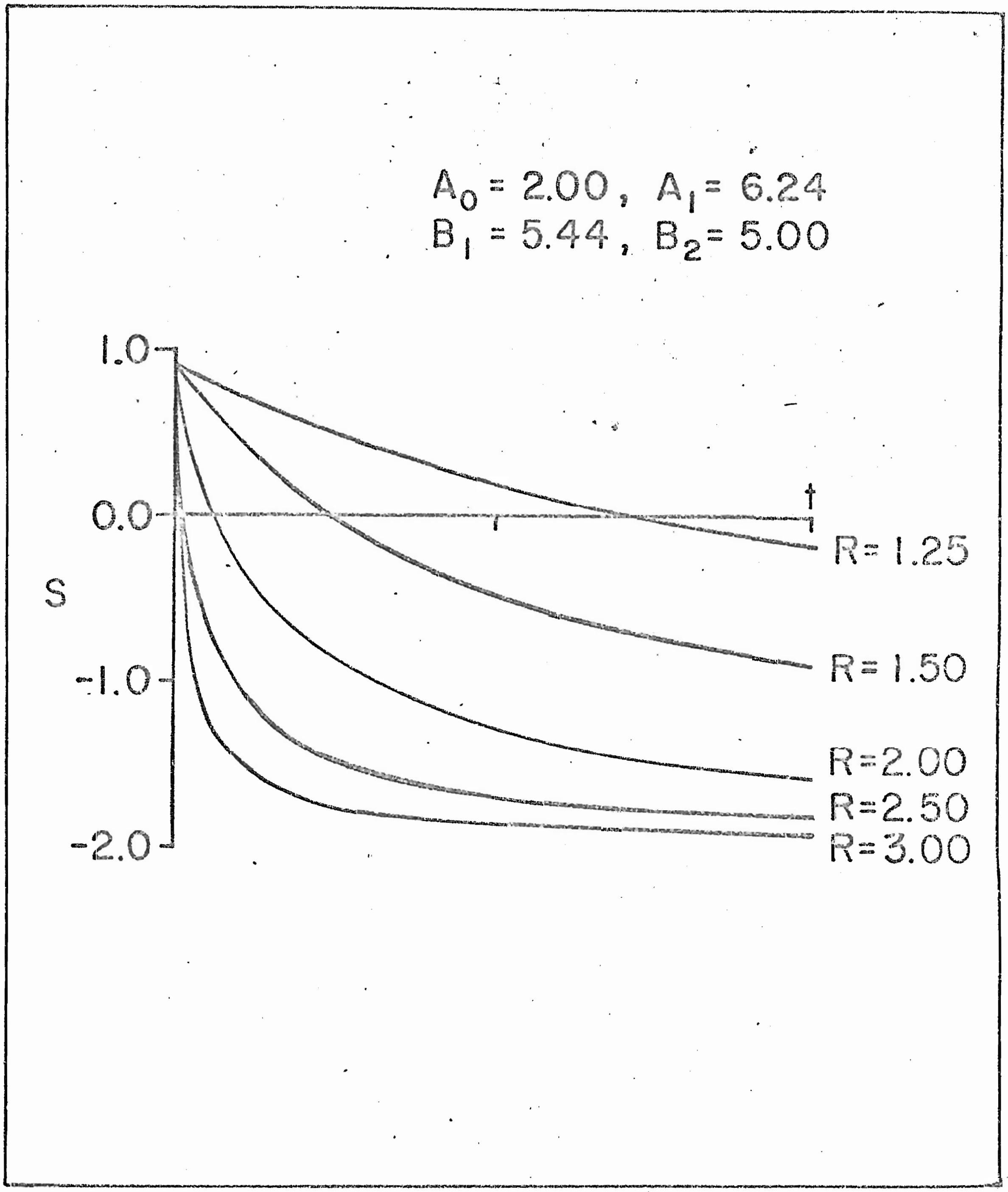

Fig. 9. Fourth Order Moment Closure Solution for Dependence of Concentration Skewness on Dimensionless Time for Probability Distribution (4.1).

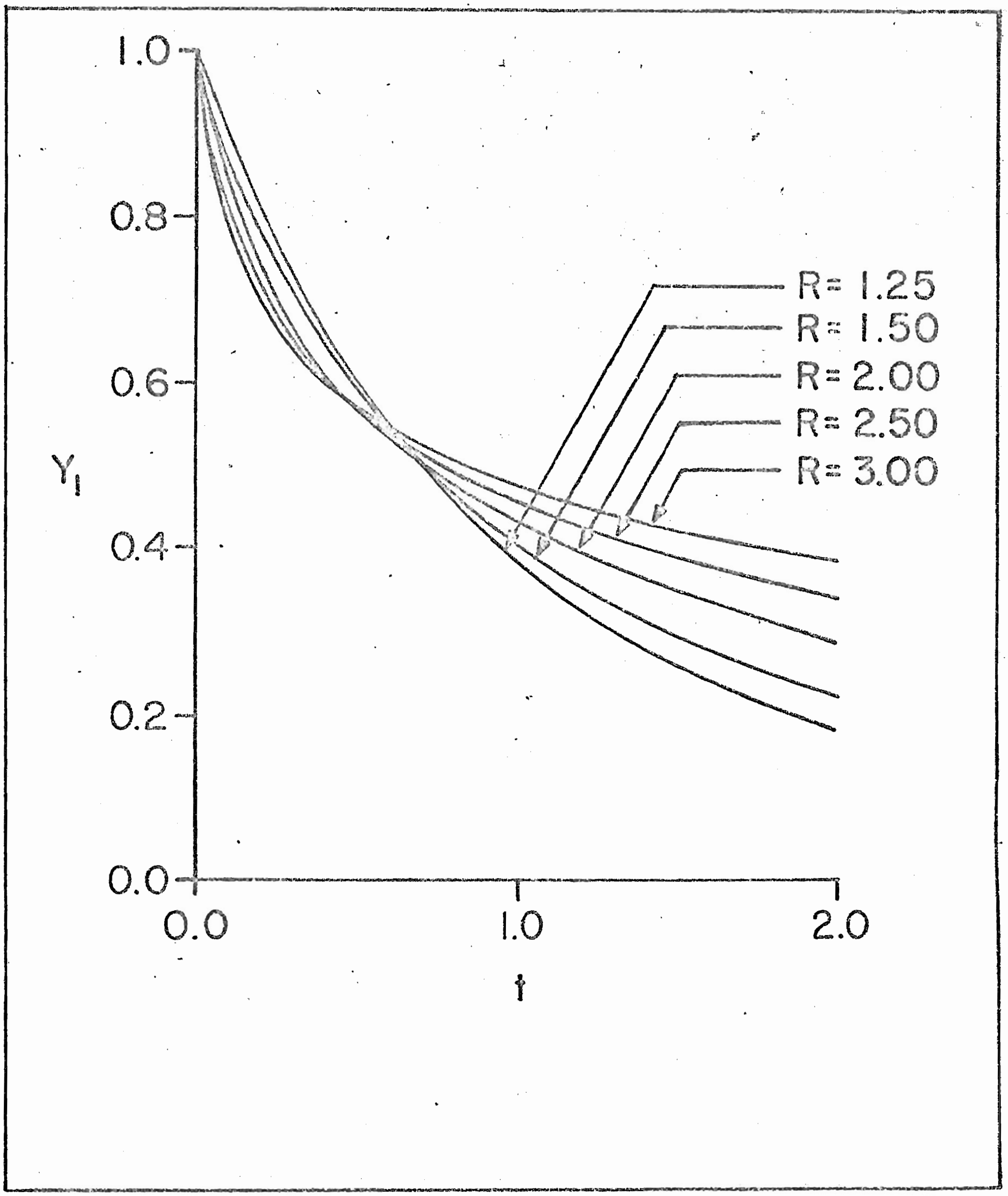

Fig. 10. Exact Solution for Dependence of Mean Concentration on Dimensionless Time for Probability Distribution (4.2).

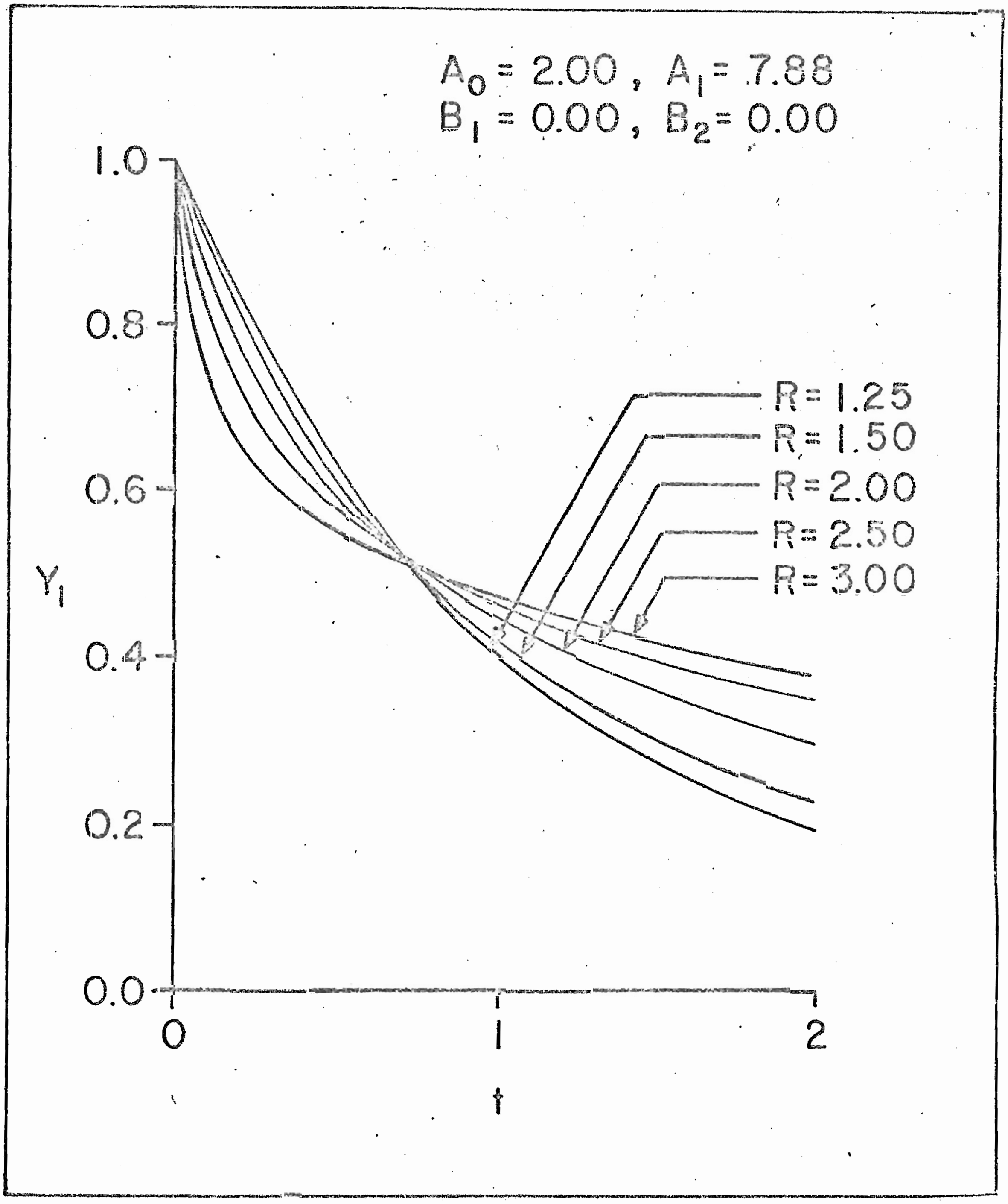

Fig. 11. Third Order Moment Closure Solution for Dependence of Mean Concentration on Dimensionless Time for Probability Distribution (4.2).

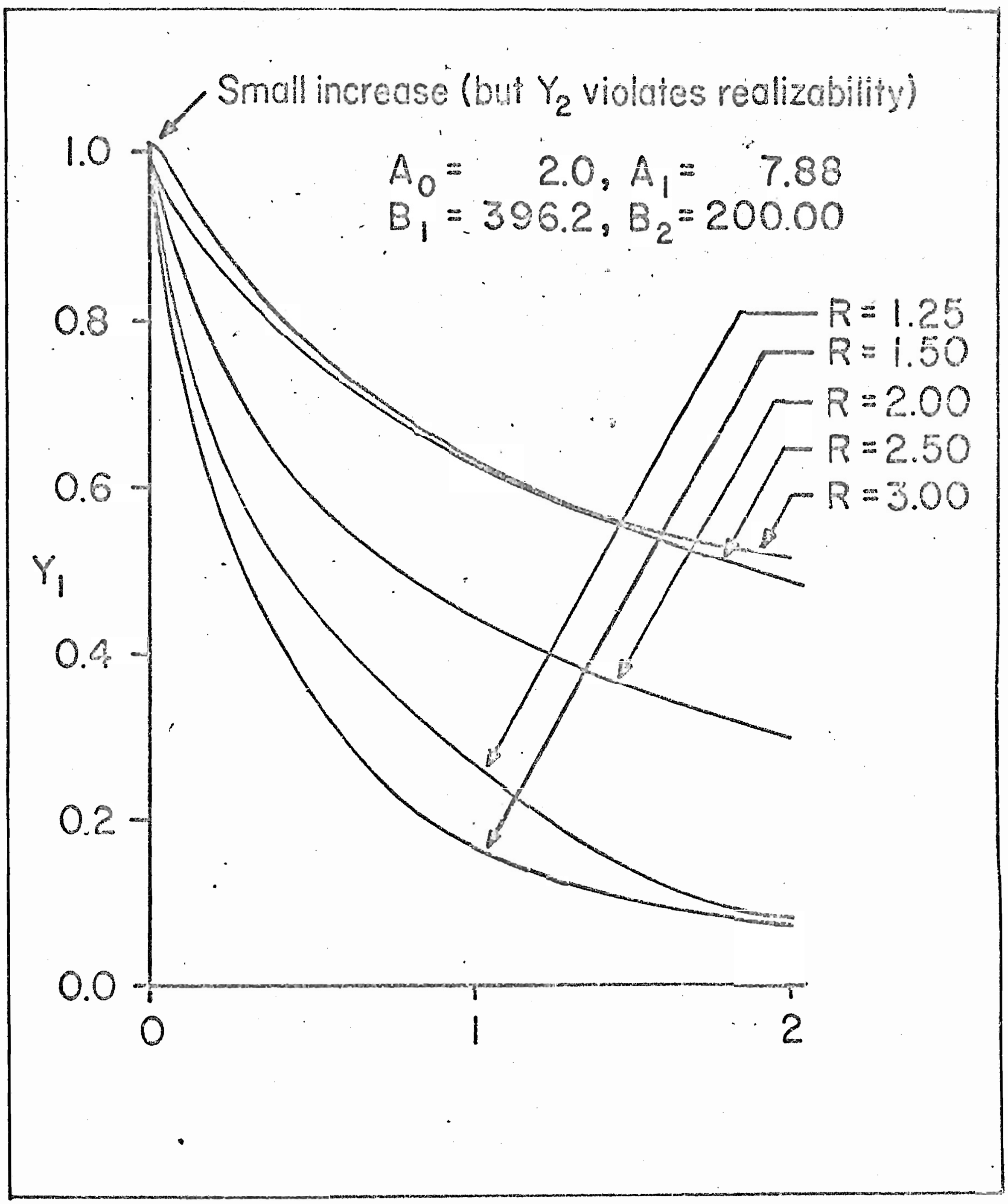

Fig. 12. Fourth Order Moment Closure Solution for Dependence of Mean Concentration on Dimensionless Time for Probability Distribution (4.2).

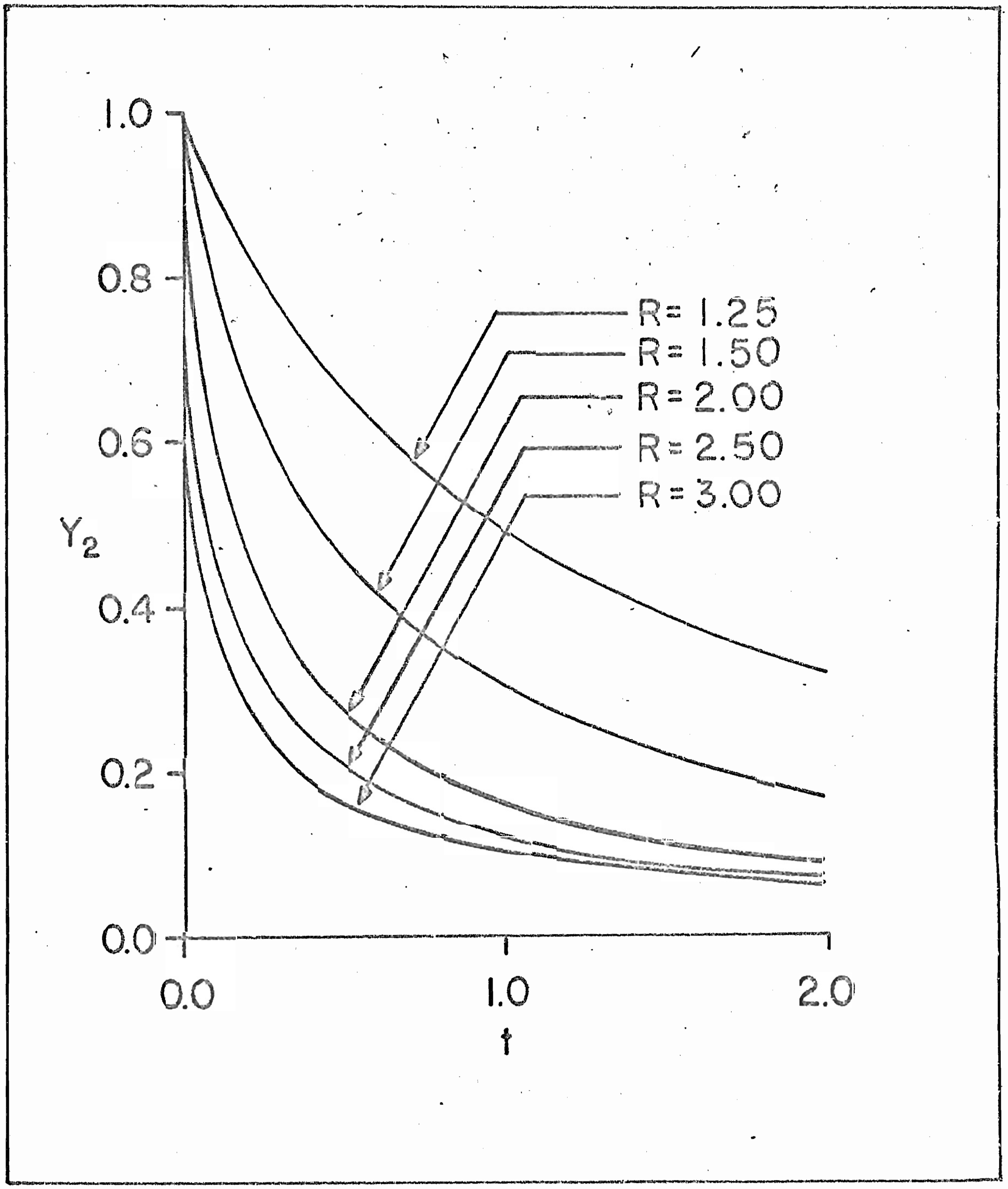

Fig. 13. Exact Solution for Dependence of Concentration Relative Intensity on Dimensionless Time for Probability Distribution (4.2).

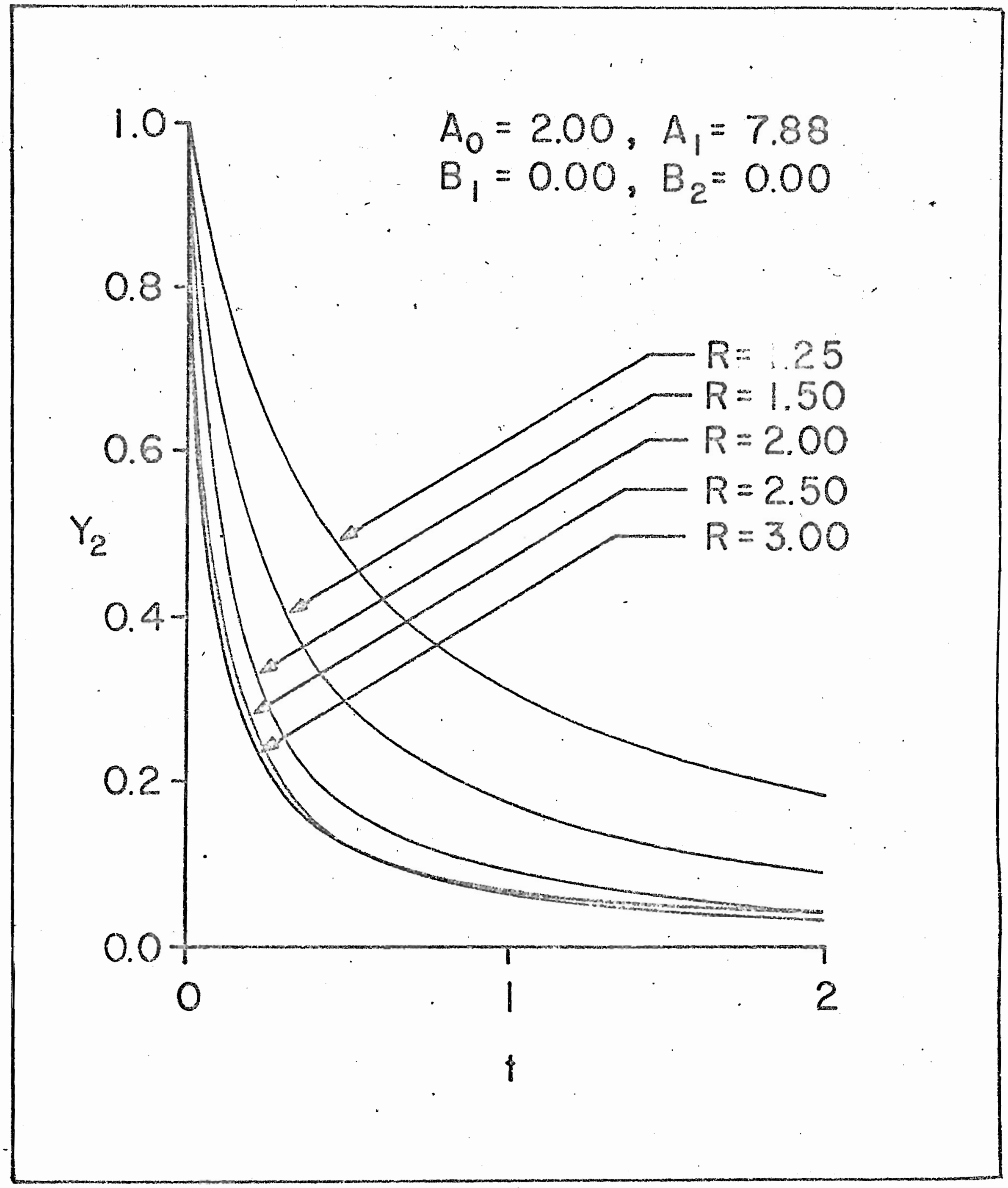

Fig. 14. Third Order Moment Closure Solution for Dependence of Concentration Relative Intensity on Dimensionless Time for Probability Distribution (4.2).

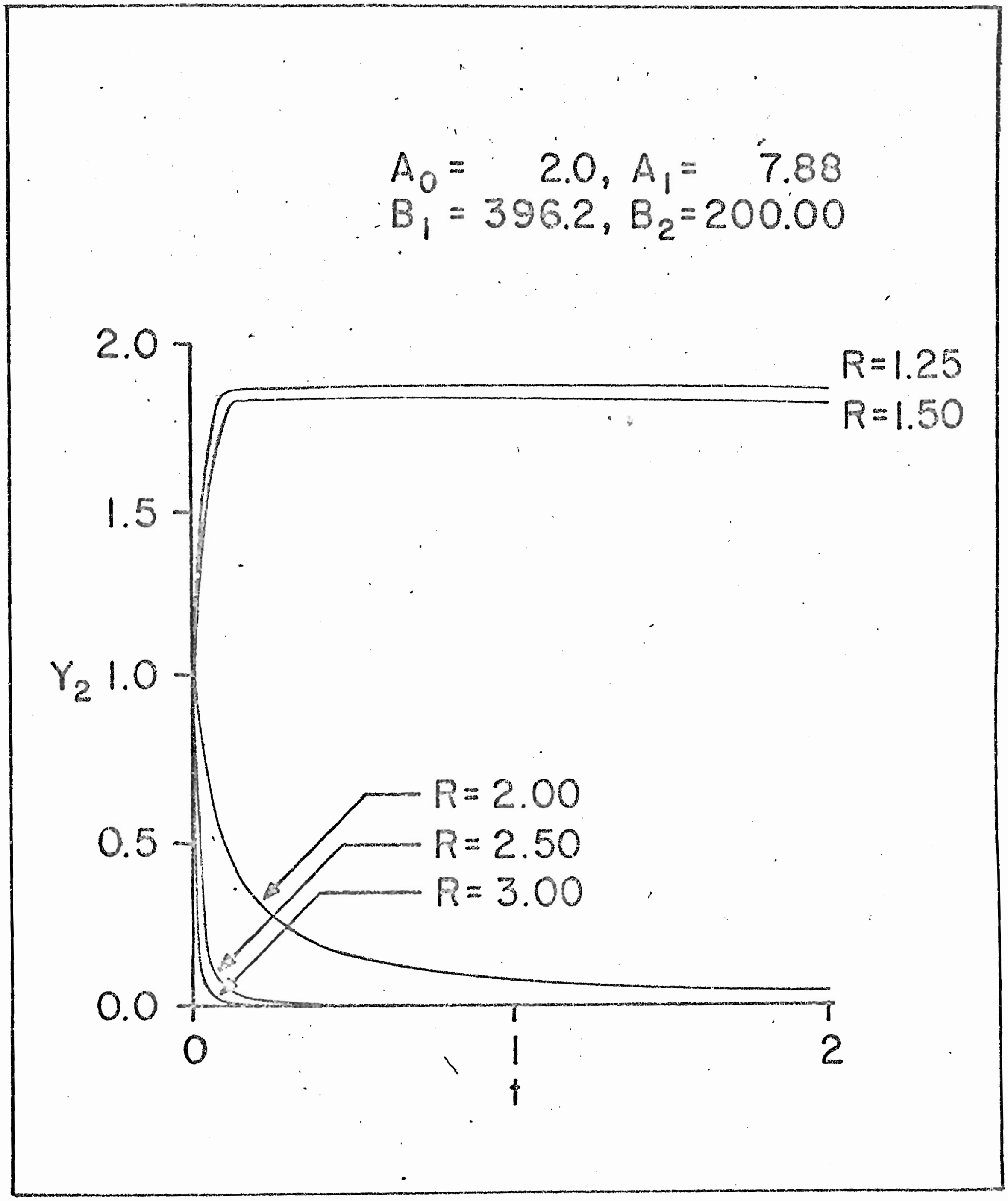

Fig. 15. Fourth Order Moment Closure Solution for Dependence of Concentration Relative Intensity on Dimensionless Time for Probability Distribution (4.2).

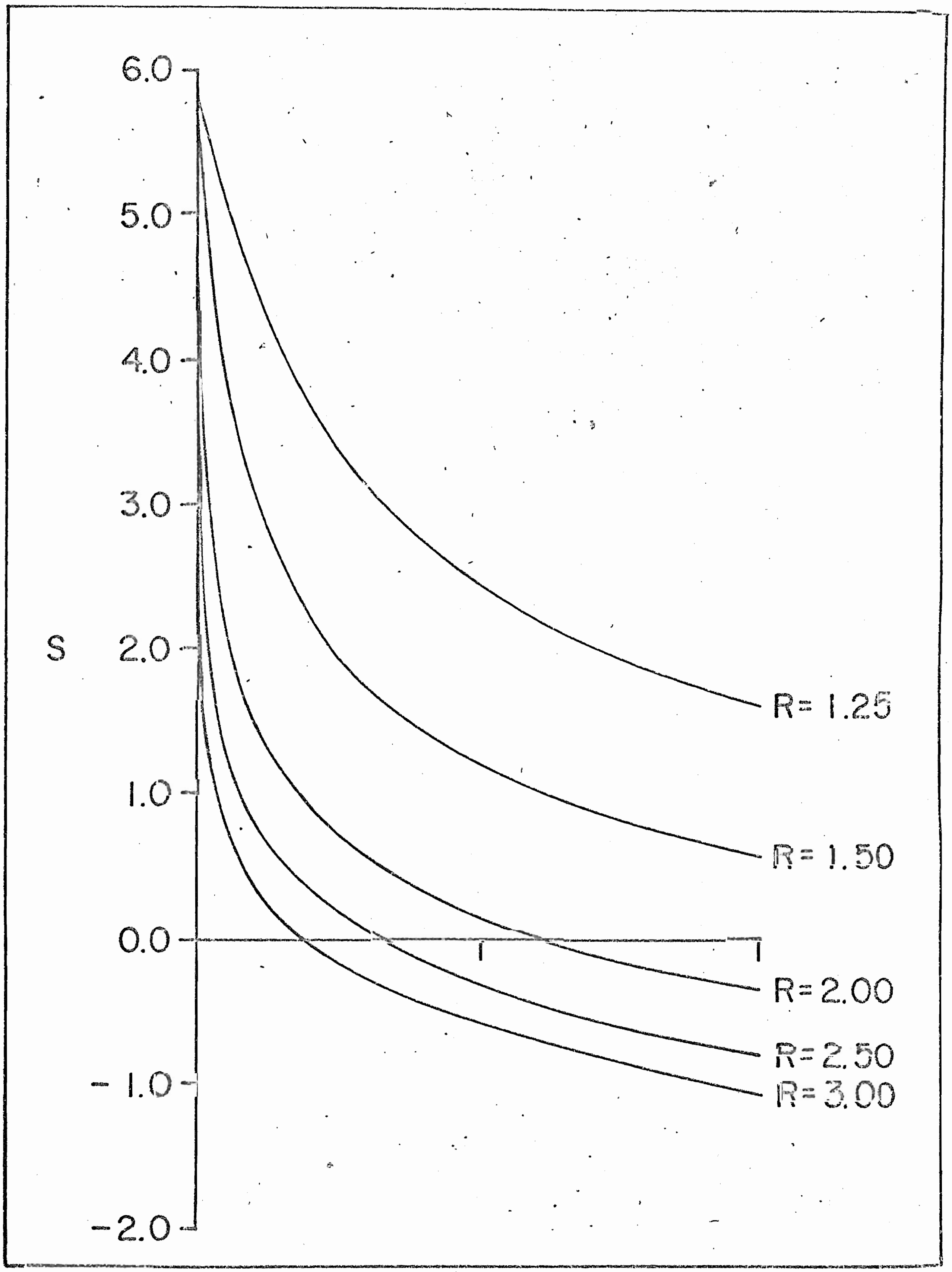

Fig. 16. Exact Solution for Dependence of Concentration Skewness on Dimensionless Time for Probability Distribution (4.2).

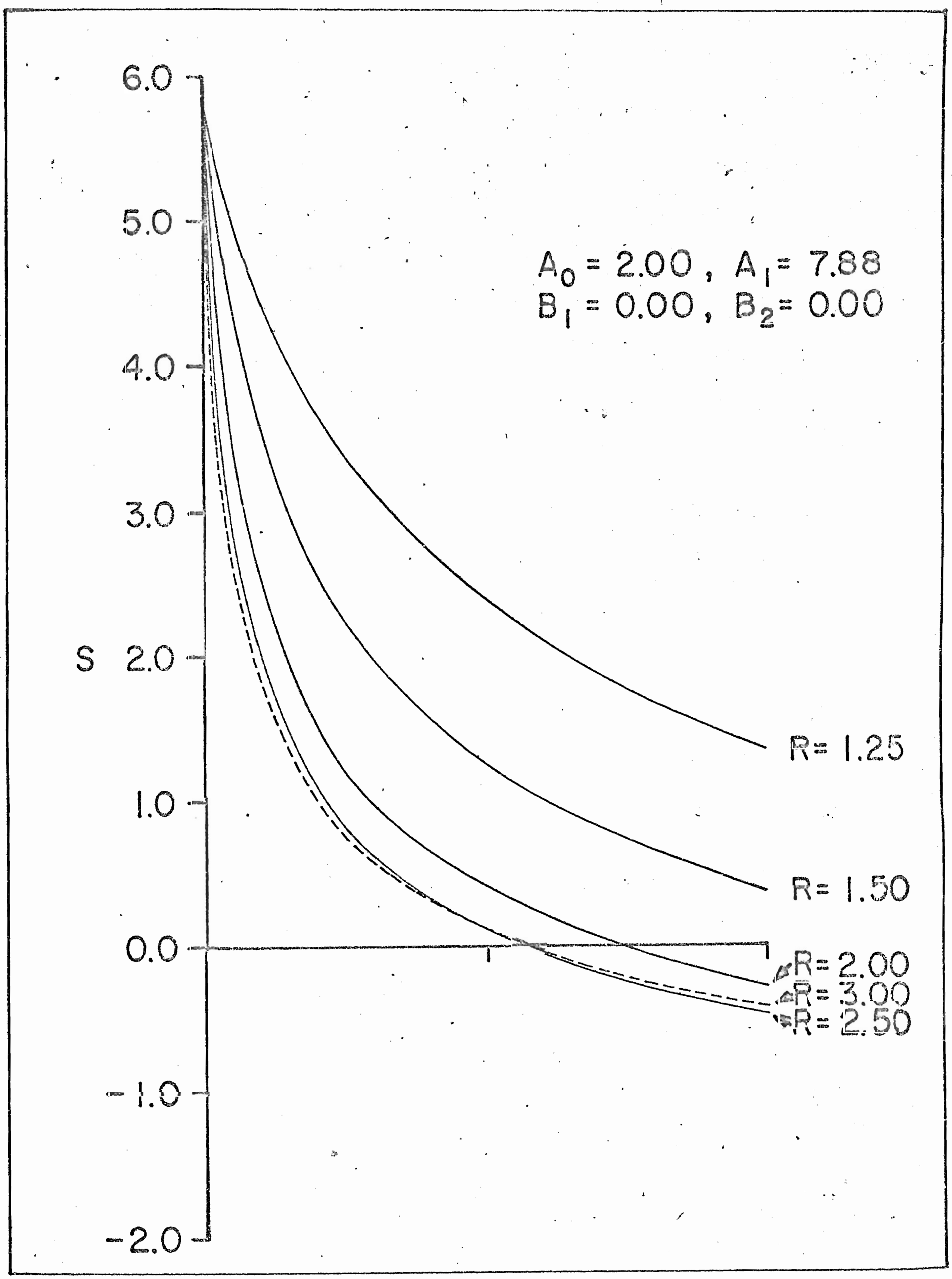

Fig. 17. Third Order Moment Closure Solution for Dependence of Concentration Skewness on Dimensionless Time for Probability Distribution (4.2).

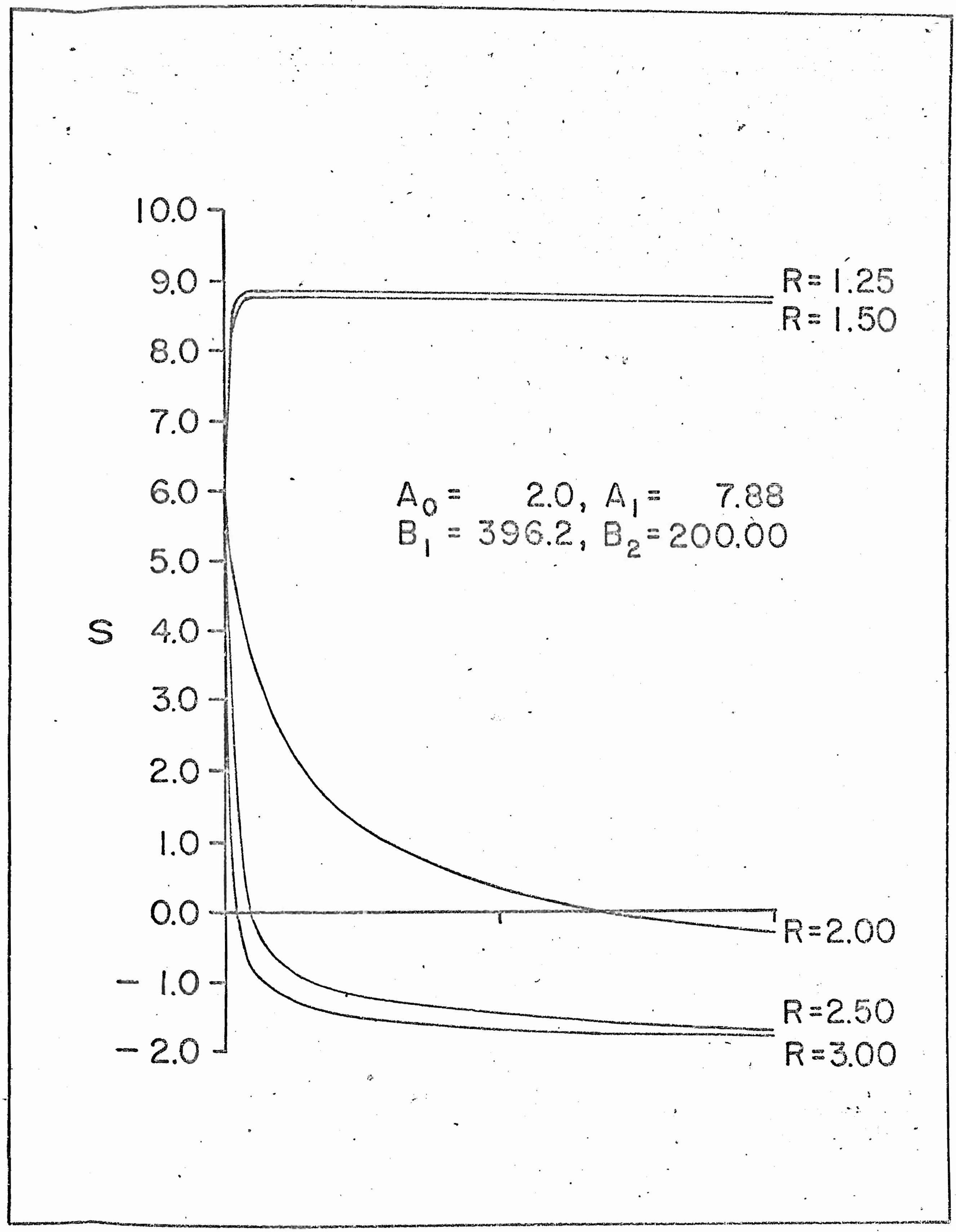

Fig. 18. Fourth Order Moment Closure Solution for Dependence of Concentration Skewness on Dimensionless Time for Probability Distribution (4.2).

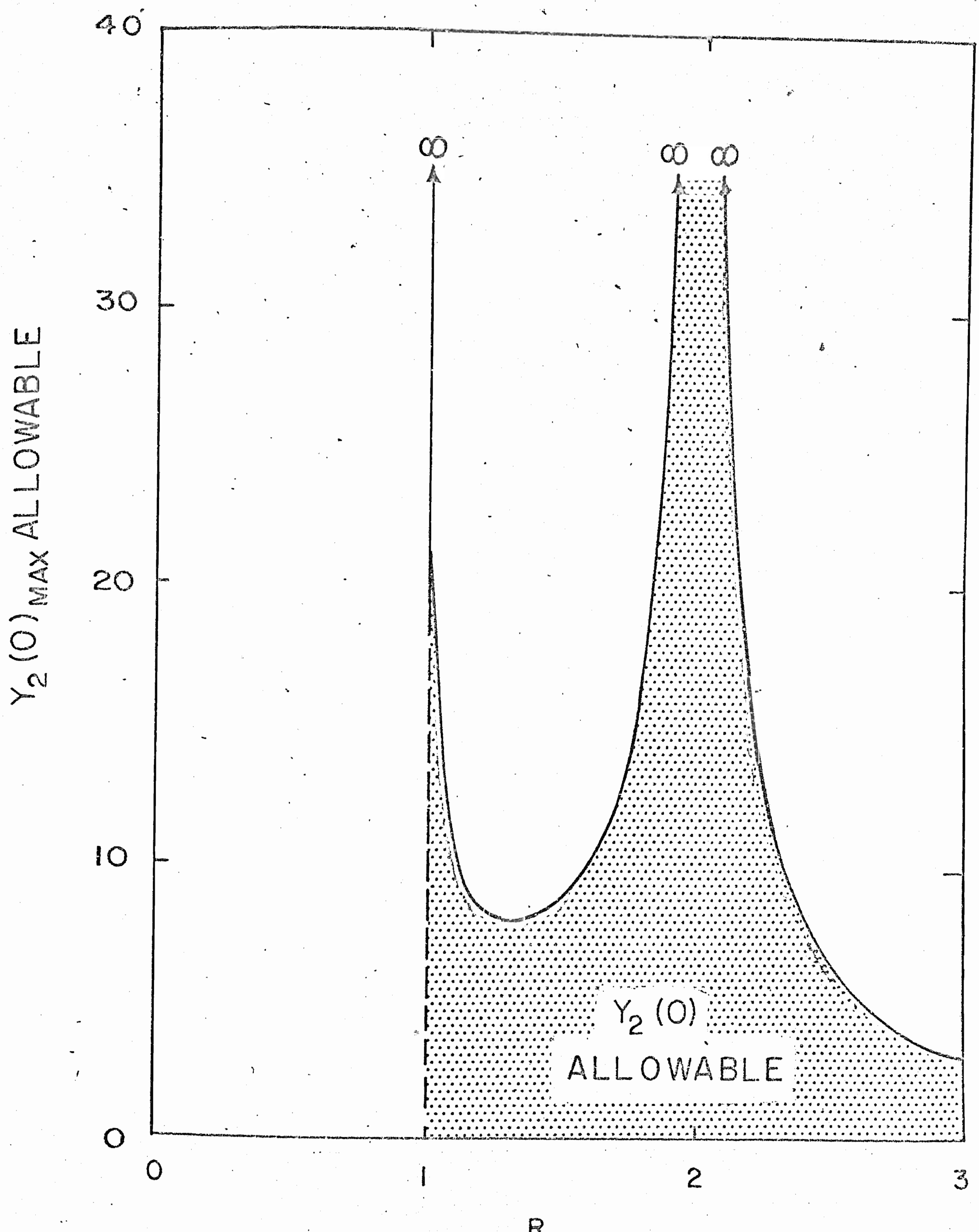

Fig. 19. Maximum Allowable Initial Concentration Relative Intensity as a Function of Order of Reaction From Third Order Moment Closure Theory.