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Evaluation of "Parameter Design" Methodologies for the Design of Chemical Processing Units

by

Sami Boudriga

A thesis submitted to
the school of Graduate Studies
in partial fulfillment of the requirements for the
degree of Master of Applied Science
in
Chemical Engineering

DEPARTMENT OF CHEMICAL ENGINEERING
UNIVERSITY OF OTTAWA
OTTAWA, ONTARIO,
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Sami Boudriga, Ottawa, Canada, 1990



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ABSTRACT

'Parameter design' is a procedure to identify settings of design variables that minimize variation in performance of a processing unit.

Application of such procedures in the manufacturing industries has met with considerable success in improving quality of the product. However their application to the design of chemical processing units has not been widely reported. The objective of this study was to investigate the feasibility of using 'parameter design' methodologies to determine values of the design variables for a non-isothermal Continuous Stirred Tank Reactor which minimized the sensitivity of product quality to disturbances. The methodologies included: (i) Monte Carlo simulations, (ii) Response Surface Methodology, (iii) Error Transmission Analysis, (iv) Taguchi Method and (v) Sequential Elimination of Levels.

The relative merits of each approach and the potential for application of 'parameter design' to chemical unit design are discussed. In summary, Response Surface Methodology associated with Monte Carlo simulations was found to be very efficient. Error Transmission Analysis did not perform well because of the inaccuracy of the first-order approximation adopted in the transmitted variation function. The use of the Taguchi Method was not always justified due to the universal logarithmic transformation, and the use of the marginal means. Sequential Elimination of Levels was found to be generally unreliable.

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Terminology

Design constraints	: additional equations specified by the designer which describe operational restrictions or desires.
Design Region	: the space spanned by all physically realistic values of the design variables.
Design Variables	: those input variables whose values are assigned by the designer.
Design vector	: a unique set of design variables.
Error Variables	: those input variables which are subject to random variation, and transmit error to the response or process output.
Error vector	: a unique set of error variables.
Inner Array	: an array of design vectors, possibly a subset of a complete array which spans properly the design region.
Input Variables	: all the system variables that can be changed freely.
Outer Array	: an array of error vectors, possibly a subset of a complete array which covers the interval of uncertainty.
Performance Statistic	: the objective function which typifies the magnitude of the transmitted error.
Process output	: the variable that typifies the process performance.
Target Value	: the value of the process output required by the designer.

Nomenclature

C_A	: outlet concentration of A.
C_B	: outlet concentration of B.
C_{A_i}	: inlet concentration of A.
C_{B_i}	: inlet concentration of B.
C_p	: molar heat capacity.
d	: derivative.
$D_{\underline{x}}$: matrix containing the derivatives.
E_A	: activation energy for the first reaction.
E_B	: activation energy for the second reaction.
E_N	: expected value.
ETA	: abbreviation for Error Transmission Analysis.
F	: flow rate.
$g_{\underline{x}}$: gradient.
H	: specific enthalpy.
H_l	: heat loss.
H_{RA}	: molar heat for the first reaction.
H_{RB}	: molar heat for the second reaction.
$H_{\underline{x}}$: transfer function.
IMSL	: International Mathematics and Statistics Library.
k_A^0	: pre-exponential constant for the first reaction.
k_B^0	: pre-exponential constant for the second reaction.
k_A	: rate constant for the first reaction.
k_B	: rate constant for the second reaction.
K	: coefficient of proportionality for the loss function.
L	: loss function.
MCS	: abbreviation for Monte Carlo Simulations.
Q	: heat exchanged with the system.
R	: universal gas constant.
$R(\theta)$: average loss.
r_A	: rate of the first reaction.
r_B	: rate of the second reaction.
RSM	: abbreviation for Response Surface Methodology.
SEL	: abbreviation for Sequential Elimination of Levels.
SEL1	: abbreviation for the first application of Sequential Elimination of Levels.
SEL2	: abbreviation for the second application of Sequential Elimination of Levels.
SEL3	: abbreviation for the third application of Sequential Elimination of Levels.
SN	: signal-to-noise ratio.
SN_T	: signal-to-noise ratio with response on target.

T	: reactor temperature.
t	: target value.
T_i	: inlet temperature.
TAG	: abbreviation for Taguchi Method.
V	: reactor volume.
$V_{\underline{x}}$: transmitted variation function.
x_a	: adjustment variables.
x_a^*	: optimal adjustment variables.
x_d	: design variables.
x_d^*	: optimal design variables.
x_e	: error variables.
x_v	: design variables that affect the variance.
y	: response.

Greek letters

β	: regression coefficient.
ϵ	: error term.
η	: performance measure.
μ	: mean.
ξ	: heat loss fraction.
ρ	: density of the reacting mixture.
ρ_i	: density of the inlet mixture.
σ	: standard deviation.
Σ	: variance-covariance matrix.
τ	: space time.

Chapter 1

Introduction

Chemical plants rarely operate at their nominal design conditions. Any deviation in the input variables affecting a processing unit, such as a reactor or distillation column, will be transmitted through the unit causing a deviation in the output variables. Variation in the output variables from their design specifications increases operating costs for the unit. Unfortunately, there has been little attention paid to the transmission of variation during the early stages of design. Traditional considerations in the optimal design of a processing unit have included costs associated with yields and production rates of desirable products, costs for utilities and capital costs but have generally excluded costs related to variability.

To minimize deviation from product specifications, the usual approach has been to design an automatic control system to keep the process output specifications on target, despite changes in the process inputs. However, this has normally been treated as a separate design problem, to be carried out after the unit itself has been designed. In such a two-stage approach, the designer of the control system is limited to selecting an appropriate variable to manipulate, choosing a control

loop configuration (e.g. feedback, feedforward, etc.) and developing an appropriate control algorithm. The effectiveness of the control system is, however, highly dependent upon the nominal values of the operating variables (e.g. flow rates and temperatures) and the mechanical design (e.g., height and diameter) which, regrettably, have already been set by the designer of the processing unit. The high degree of interaction between these design problems is now widely recognized and investigations are currently being made into coupling the design of the unit with that of its control system (Shinsky [29]). A major difficulty has been the immense complexity of the integrated design problem; the number of alternatives is greatly increased and the need to consider the dynamic behavior of the process is introduced .

In this thesis an alternative to the integrated design approach is investigated. It involves consideration of transmitted variation at the initial steady-state design stage. If knowledge of how input variation affects the outputs can be determined at the steady-state design stage, then the unit could be designed to be insensitive to input variations. This would reduce the costs associated with deviations from product specifications and potentially simplify the design of the control system.

Design engineers in the manufacturing industries have tried to build quality into products and processes by choosing values for the design variables that make the process or the product more robust to environmental variations. In other words, they have attempted to design the process to minimize the transmission of variation from input variables to the controlled or target variables.

The Japanese have been very successful in building quality into products and processes through the use of so-called Parameter Design methodologies. In particular, Genichi Taguchi has focused attention on:

- minimizing variation with target on mean,
- making products and processes robust to environmental conditions,
- making products and processes insensitive to input variables variation.

Despite success in these industries, such an approach has not been widely adopted for the design of chemical processing units.

Several methodologies could be used to solve this parameter design problem, including the use of orthogonal arrays to maximize a signal-to-noise ratio as proposed by Taguchi, or the use of nonlinear programming coupled with Monte Carlo simulations.

Assuming that the value of an input fluctuates about its nominal design value according to a known distribution and using Monte Carlo simulations, a number of points according to this distribution can be generated and the design equations solved to yield a distribution for the values of the process output. The variance of the output can then be calculated without any approximation for the error transmission. Application of a multivariable optimization routine gives the settings for the design variables which minimize the transmission of error. This approach can accurately handle all kinds of error structures but, computationally, it is very demanding.

To reduce the amount of computation involved, Error Transmission Analysis may be used instead of Monte Carlo simulations. This method approximates the transmitted variation function by the first-order error propagation formula. For simple systems, an analytical expression for the transmitted variation function can be derived. The optimal setting is obtained by minimizing this transmitted variation function with respect to the design variables. For more complicated systems, where no analytical expression can be obtained, the function can only be evaluated by numerical calculation of its derivatives with respect to the error variables. Although the computational load is greatly reduced using this approach, the reliability of the first-order error propagation formula is questionable.

A popular method that has been used in Japan and is gaining significant attention in North America, called the Taguchi Method [30], consists of the analysis of marginal means of a performance index. Taguchi relates the loss due to variation to the logarithm of so-called "signal-to-noise ratios" and assumes the loss function to be quadratic and to be such that there is no correlation between the mean and the variance of the process output. To maximize these ratios Taguchi used a multiplication of orthogonal arrays for the design variables and the error variables which transmit variation to the process output. Here the distribution of input errors is approximated by a three-point distribution. Moreover, marginal means of the performance measure are used to determine the optimal values of the design variables. Unless the loss function is quadratic and the logarithm is the right transformation, the analysis of marginal means can be very misleading. On

the other hand, the method is computationally straightforward and easy to use.

For large design regions the levels of the design variables are widely spaced and the values of the performance measures are very different. In these cases use of the marginal means is especially unreliable. A method called Sequential Elimination of Levels has been proposed by Wu [38] to deal with this situation. Like the Taguchi Method, it uses orthogonal arrays and signal-to-noise ratios, but tries to overcome the problem of the widely spaced levels by sequentially decreasing the size of the orthogonal array, eliminating the levels associated with the worst process performance for each design variable.

Response Surface Methodology is an optimization technique that can be used to locate an optimal condition of some quality characteristic, such as a high mean value, low variance or both. It is normally carried out in two phases. In phase I, the objective is to move from poor conditions to better ones using successive planar approximations to the objective function coupled with a steepest descent algorithm. In phase II, when near optimal conditions have been found, a more detailed study is carried out using a quadratic approximation to the response function. The method not only gives the optimal setting for the design variables but also gives an empirical model that predicts the value of the objective function (e.g., variation of the process output as a function of the design variables). This model could be a useful tool for the designer especially when multiple design objectives are considered.

The objectives of this study are:

- to critically review each approach;
- to apply each method to design of a simple processing unit, namely a non-isothermal Continuous Stirred Tank Reactor (CSTR) and to evaluate the effectiveness of each approach;
- to determine the potential benefits from using such a design strategy;
- to investigate the compatibility of this design strategy with other more traditional strategies.

Chapter 2

Background and Literature Review

Parameter Design is a process in which design variables under the control of the product designer are varied in a scientific fashion to determine optimal settings for these variables. This method has been popularized in Japan by G. Taguchi for the design of robust and controllable manufacturing processes. The concept of quality improvement through Parameter Design has been discussed by Taguchi [34], Box and Draper [7,10] and Kachar [19]. In this thesis five different approaches to parameter design are examined namely: (i) Taguchi Method, (ii) Sequential Elimination of Levels, (iii) Response Surface Methodology, (iv) Error Transmission Analysis and (v) Monte Carlo simulations. Background for each approach and a review of the pertinent literature on each are presented in this chapter in order to establish the strengths, limitations and potential for each.

2.1 Taguchi Method

Taguchi [30,34] has emphasized the use of fractional factorial designs and other orthogonal arrays in order to

- make products robust to environmental conditions;
- make products insensitive to component variation;
- minimize the mean square error about a target value.

2.1.1 Problem Definition

As the objective function to be maximized, Taguchi chooses the measure $10\log_{10}(SN_T)$ when it is desired to keep a response on target. SN_T is a "signal-to-noise ratio" defined as

$$SN_T = \frac{\mu^2}{\sigma_y^2} - \frac{1}{n} \quad (2.1)$$

where y is the process output, n is the number of runs, μ is the mean value of the process output, and σ_y^2 its variance. Maximizing SN_T is equivalent to maximizing

$$SN = \frac{\mu^2}{\sigma_y^2} \quad (2.2)$$

For convenience Taguchi took $H = \log_{10}(SN)$ and considered the equivalent problem of maximizing H . The numerator μ is obtained directly, the denominator σ_y^2 may be obtained from the usual error transmission formula, employing the first partial derivatives of y with respect to the error variables. Thus the problem is to

maximize a known function H with respect to the design variables within a given region R .

Taguchi employs the terminology "design" or "control" variables for those variables whose nominal levels are chosen by the designer. The term "noise" or "error" variables refers to all the input variables that transmit error to the output variable. He classified the design variables as:

- design variables that affect process variability;
- adjustment variables which have a negligible effect on the variability but alter the mean;
- variables that do not affect the variability or the process mean.

The problem therefore requires definition of

- a d -dimensional vector of design variables \underline{x}_d , whose elements can be chosen in a given region R ;
- a k -dimensional vector of error variables \underline{x}_e , some or all of whose elements may be identical to those of \underline{x}_d and whose known covariance matrix is Σ_e . It is assumed that the elements of \underline{x}_e are statistically independent so that Σ_e is diagonal with the i^{th} diagonal element equal to σ_i ;
- an objective function, H , that measures the relative variation of the quantity of interest, y , whose functional relationship to \underline{x}_e is known.

For a given setting of the design variables, noise N produces an output y . The transfer function is given by $f(N, \theta)$, where θ is the vector of design variables. The noise is supposed random and so is the output. The ideal output corresponds to the target. The average loss is

$$R(\theta) = E_N[L(y, t)]. \quad (2.3)$$

$L(y, t)$ is the loss function. The loss function proposed by Taguchi [33] is quadratic and given by

$$L(y, t) = K(y - t)^2 \quad (2.4)$$

where t is the target value and K is a constant. The objective is then to minimize this loss function.

2.1.2 Classification of Design Problems

Taguchi classified the Parameter Design problems as follows:

1. Smaller the better: The response is continuous and positive, in most cases the desired value is zero and the loss function increases with the response value. In this case Taguchi recommended the use of the logarithm of the mean squared response

$$\eta = 10 \log_{10} (\text{Mean Squared Response}). \quad (2.5)$$

2. Response on target: The response is continuous and has a non-extreme target value, where μ and σ_y represent the predictable and the unpredictable parts

of response y . The loss function increases as the response deviates from the target in either direction. Taguchi suggested

$$\eta = 10 \log_{10} \frac{\mu^2}{\sigma_y^2} \quad (2.6)$$

3. Larger the better: The response is continuous and desired to be as large as possible. This is a particular application of the smaller the better case, where the reciprocal of the response is treated as the performance characteristic. Taguchi uses the mean squared of the reciprocal response

$$\eta = 10 \log_{10} (\text{Mean Squared of the Reciprocal Response}) \quad (2.7)$$

4. Ordered categorical response: The response is categorized into ordered categories such as very bad, bad, acceptable, good, very good.

Taguchi employs designs obtained by a multiplication of an inner array for the design variables and an outer array for the error variables. The arrays that have been used were orthogonal and are available in Taguchi [33] and Day [14]. For each experimental run in the inner array another array of runs is conducted, varying environmental variables that may affect the product's performance. He assumed that the range of possible values for the nominal value of each control variable is known. The allowable region is defined as the design or control region R .

It is possible to compute Taguchi's SN_T ratio using outer arrays at any point \underline{x} and find the settings of \underline{x}_d which maximize SN_T . Taguchi used a global exploration with a three-level orthogonal array. He supposed no interaction between

the variables and used graphs of marginal averages of SN_T to find the maximum performance. The method works well if the objective function is a general second-degree expression with no interaction terms. The second-order expression employed by Taguchi is

$$H_{\underline{x}} = \beta_0 + \sum_{i=1}^k \beta_i x_i + \sum_{i=1}^k \beta_{ii} x_i^2. \quad (2.8)$$

Minimizing the loss is equivalent to minimizing the mean square deviation from target. Taguchi advocated a two-stage procedure involving the following steps:

1. find the optimal setting of the design variables, $\underline{x}_v = \underline{x}_v^*$, that maximize the "signal-to-noise ratio";
2. set the adjustment vector \underline{x}_a to \underline{x}_a^* while the design variables vector \underline{x}_v is fixed at \underline{x}_v^* , in order to bring the mean of the process output on target.

The variables, x_a , are fine-tuning adjustments and can be optimized once the optimal setting for the design variables is established. A change in the product or in the process requirements can be accommodated by adjusting the vector \underline{x}_a while the setting $\underline{x}_v = \underline{x}_v^*$ remains optimal. Leon and Shoemaker [20,21] showed that, by using the signal-to-noise ratio, a logarithmic transformation was performed on the raw data. The objective of this transformation is to make the mean and the variance independent. However, in some situations no transformation is required, or another transformation is needed to eliminate or at least reduce the coupling of these two statistics. Box [2,3,4] and Kachar [18] also noted that direct application

of the SN ratios may be misleading. Recently a method of determining an appropriate transformation to eliminate the unnecessary coupling of dispersion and location effects using a "lambda plot" was described by Atkinson [1].

The use of the orthogonal arrays has been reported to be efficient in improving the quality of the product by Phadke [28], but these arrays do not describe the true situation. The probability that the variable is at its nominal value is not the same as for the low or high level.

Taguchi's signal-to-noise ratio can be used only when the objective function is a quadratic form with no interaction. However, Box and Hunter [9] have mentioned that the need for second-order terms of one kind implies the likely need of other second-order terms such as two factor interactions.

Unless the logarithm is the right transformation and the loss function is quadratic with no interaction terms, the signal-to-noise ratio will be inappropriate.

Based on the reported work in the literature it appears that the Taguchi method has the following advantages:

- the method is easy to use,
- no derivatives need to be generated,
- little computation is required,
- the function does not have to be expressed in an analytical form.

However, there are several weaknesses, namely,

- excessive amount of computation for systems with many variables,
- universal use of logarithmic transformation which may be inappropriate.

2.2 Sequential Elimination of Levels

To deal with widely spaced levels, Wu et al. [38] proposed a variation of the Taguchi Parameter Design method called Sequential Elimination of Levels (SEL). They employed orthogonal arrays for the design and error variables and used the same signal-to-noise ratio as Taguchi. He also used the analysis of marginal means to maximize the performance statistic. The difference arises from the sequential use and the smaller size of the orthogonal arrays employed. Starting with an initial inner array for the design variables the performance measure is determined using an outer array for the error variables. The levels in the inner array that give the worst marginal mean of the performance measure are eliminated. Then a new orthogonal array is constructed for the remaining levels. The procedure is repeated until the number of levels is reduced to two. The optimal setting is the one corresponding to levels associated with the best performance. Other alternatives to using the marginal mean of the signal-to-noise ratios to determine the worst settings of the design variables were introduced by Wu et al. [38], such as using the minimum and the median of the SN_T .

The elimination of poor variable levels enables one to focus on the design region of promise. Starting with a relatively large design region, its size may be considerably reduced by using SEL. When the size of the arrays or the number of

iterations is to be reduced, several levels may be eliminated at a time.

The sequential design problem consists of predicting variable levels with low variance from the data. The technique can be made more powerful if the orthogonal array is applied iteratively to different regions of the variable space as determined by the previous experiments.

Advantages of the method:

- those associated with Taguchi Method,
- the method can quickly focus on the design region of promise,
- less number of runs is required,
- the number of runs can be potentially reduced by eliminating more than one level at a time.

Some possible disadvantages are:

- those associated with Taguchi Method,
- the choice for the descriptive statistic.

2.3 Response Surface Methodology

Response Surface Methodology is a collection of mathematical and statistical techniques used to study the influence of several independent variables

(x_1, x_2, \dots, x_k) , on a response or dependent variable, y , and to optimize this response. The method is described in detail by Box [6,8], Chocran [12] and Thomas [35].

Initially the response surface is approximated by a plane or hyperplane given by

$$y = \beta_0 + \sum_{i=1}^p \beta_i x_i \quad (2.9)$$

from which the direction of steepest descent is given by the vector $\underline{\delta} = (-\beta_1, -\beta_2, \dots, -\beta_p)$. To obtain estimates of the parameters the response is evaluated at various points in the local region.

Orthogonal first-order designs (e.g., 2-level factorial and fractional factorial designs) are used to develop this first-order model because they yield parameter estimates having minimum variance and require small numbers of functions evaluations. Using such experimental designs also provides information about interaction effects indicated by terms such as $\beta_{ij} x_i x_j$. If these terms are important then it indicates that the planar model is not a good approximation to the response surface. More details can be found in Douglas [15].

If there is some evidence that the surface is not planar, a second-order equation,

$$y = \beta_0 + \sum_{i=1}^p \beta_i x_i + \sum_{i,j=1}^p \beta_{ij} x_i x_j \quad (2.10)$$

is used.

The β 's are to be determined using the Least-Squares method.

Curvature is also indicated when the measured response at the center point

is significantly different from the intercept, β_0 . Experimental designs required to estimate the quadratic parameters need at least three levels for each variable. The preferred class of second-order designs is the class of rotatable designs of which the central composite designs are mostly used.

They consist of a 2^k factorial design augmented by $2 \times k$ axial points $(\pm\alpha, 0, \dots, 0)$, $(0, \pm\alpha, 0, \dots, 0), \dots, (0, \dots, \pm\alpha)$, and n_0 central points $(0, \dots, 0)$, where α is equal to $2^{\frac{k}{4}}$ and k is the number of variables.

These designs can be found in more detail in Douglas [15], Box [6], and Draper [16]. The location of the optimum can be found analytically from the quadratic model. In addition, the quadratic model provides a useful description of the response surface around the optimum. A potential disadvantage is that the second-order polynomials do not describe well the sum squares surfaces.

2.4 Error Transmission Analysis

According to Box et al. [4], the Error Transmission method consists of approximating the transmitted variation function $V_{\underline{x}} = var(y_{\underline{x}})$ using the error propagation formula which gives, locally and approximately,

$$V_{\underline{x}} = d_1^2(\underline{x})\sigma_1^2 + d_2^2(\underline{x})\sigma_2^2 + \dots + d_k^2(\underline{x})\sigma_k^2 = d_{\underline{x}}' \sum d_{\underline{x}} \quad (2.11)$$

where k is the number of variables x that transmit error to the process output y , $d_{\underline{x}}$ is the vector $(d_1(\underline{x}), d_2(\underline{x}), \dots, d_k(\underline{x}))'$ of partial derivatives

$$d_i(\underline{x}) = \frac{\partial F(\underline{x})}{\partial x_i}, \quad i = 1, 2, \dots, k \quad (2.12)$$

and Σ is the $k \times k$ diagonal covariance matrix for the errors in \underline{x} , whose i^{th} element is σ_i^2 . The errors in the x 's were assumed to be independent.

The object was to minimize the transmitted variation function $V_{\underline{x}}$ using an optimization routine such as steepest descent. To do this, the gradients of $V_{\underline{x}}$ are required. Taking derivatives of Equation 2.12 yields

$$g_{\underline{x}} = 2D_{\underline{x}} \sum d_{\underline{x}} \quad (2.13)$$

where $D_{\underline{x}}$ is a matrix whose ij^{th} element, $d_{ij}(\underline{x}) = \frac{\partial^2 F(\underline{x})}{\partial x_i \partial x_j}$. In practice the derivatives can only be approximated numerically.

Advantages of the method:

- when the system is not very complicated, the variance of the output variable can be calculated directly from Equation 2.12 without numerical evaluation of derivatives.
- for complicated systems numerical derivatives are required but in either case the computational effort will be much less than Monte Carlo simulations.

A potential disadvantage is that a first-order error propagation formula may not accurately approximate the true variance.

2.5 Monte Carlo Simulations

Simulation has long been an important tool for designers, for three reasons:

- (i) data collection is generally very expensive, (ii) occasionally analytical solutions

are not possible to obtain due to system complexity, and (iii) the formulation of a mathematical model does not guarantee its analytical solution. In all these cases simulation is the only practical solution. Naylor et al. [26] have suggested that simulation might be appropriate to:

- study an experiment with complex internal interactions;
- study environmental changes on the operation by making alteration in the model;
- observe more in detail the system and have a better understanding of the process in order to make some suggestions to improve the process;
- investigate the effect of the different variables and their interactions;
- experiment new conditions for which we have little information about what may happen;
- break down the complicated systems into subsystems.

Monte Carlo simulations are now the most powerful and commonly used technique to analyze complex problems. Applications can be found in many fields. One of these fields is the sampling of random variates from probability distributions, which Morgenthaler [23] calls "model sampling". However, the use of Monte Carlo simulations to minimize the transmission of error from input variables to the output, has not been reported.

Some of the common distributions are the normal and the log-normal distributions.

Normal Distribution: A random variable X has a normal distribution if the probability density function is

$$f_X(x) = \frac{1}{\sigma\sqrt{2\pi}} \exp\left[-\frac{(x-\mu)^2}{2\sigma^2}\right], \quad -\infty \leq x \leq +\infty \quad (2.14)$$

and is denoted by $N(\mu, \sigma^2)$, where μ is the mean and σ^2 is the variance.

The standard normal variable Z is denoted by $N(0,1)$ and $X = \mu + \sigma Z$. More about generation from normal distribution can be found in Fishman [17].

Log-normal Distribution:

Let X be from $N(\mu, \sigma^2)$, then $Y = e^X$ has the log-normal distribution with the probability density function

$$f_Y(y) = \begin{cases} \frac{1}{\sigma y \sqrt{2\pi}} \exp\left[-\frac{(\ln y - \mu)^2}{2\sigma^2}\right] & \text{if } 0 \leq y \leq +\infty \\ 0 & \text{otherwise} \end{cases} \quad (2.15)$$

Once the assumption for the error distribution is made, a number of points are generated according to this distribution, the response is determined for each one of these points, the mean and the variance may be computed, and the distribution for the output can be determined using fitting techniques.

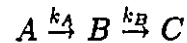
This method combined with some optimization routines gives the optimal settings that have the minimum value for the variance. Using Response Surface Methodology we can have a model that describes the variance as a function of the design variables.

The advantage of the method is that no approximation has been done about the error transmission. The major disadvantage is the large amount of computation required.

Chapter 3

Design Equations for the Nonisothermal CSTR

The system investigated in this work consisted of a first-order sequential reaction,



taking place in a nonisothermal Continuous Stirred Tank Reactor (CSTR).

The process and the associated variables are illustrated in Figure 3.1.

3.1 Design Equations

Application of conservation laws to the total mass, energy, component A and component B for the liquid in the reactor yields the following state equations:

$$\frac{dV}{dt} = F_i - F \quad (3.1)$$

$$\frac{dC_A}{dt} = \frac{F_i}{V}(C_{A_i} - C_A) - r_A \quad (3.2)$$

$$\frac{dC_B}{dt} = \frac{F_i}{V}(C_{B_i} - C_B) + r_A - r_B \quad (3.3)$$

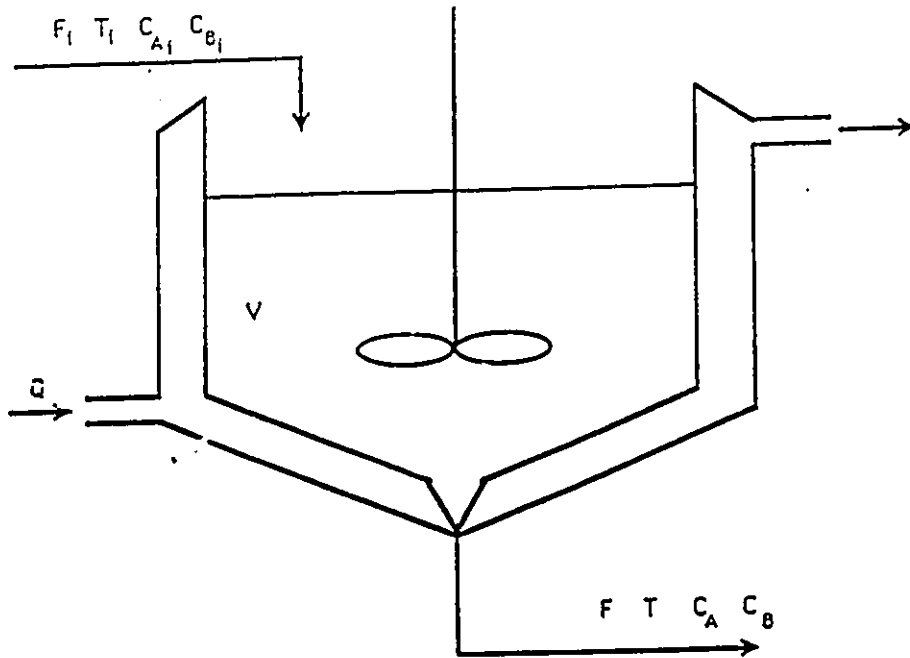


Figure 3.1: The nonisothermal CSTR.

$$\frac{dT}{dt} = \frac{F}{V}(T_i - T) - \frac{H_{RA}r_A}{\rho C_p} - \frac{H_{RB}r_B}{\rho C_p} + \frac{Q}{\rho C_p V} + \frac{H_l}{\rho C_p V} \quad (3.4)$$

where V is the volume of the reactor, F_i and F are the inlet and the outlet volumetric flow rates, C_{A_i} and C_{B_i} are the inlet concentrations of A and B, respectively, C_A and C_B are the bulk concentrations of A and B, $-r_A$ and $-r_B$ are the rates of disappearance of A and B by reaction, ρ_i is the density of the inlet mixture, ρ is the density of the reacting mixture, Q is the rate of heat removal from the system by cooling and H_l the heat loss per unit time.

At steady state the rates of change of all state variables are zero, yielding the steady state equations for the system. Combining these steady-state equations with the expressions for the kinetics yields the following set of six design equations:

$$-r_A = k_A^0 e^{(-\frac{E_A}{RT})} C_A \quad (3.5)$$

$$-r_B = k_B^0 e^{(-\frac{E_B}{RT})} C_B - k_A^0 e^{(-\frac{E_A}{RT})} C_A \quad (3.6)$$

$$\tau = \frac{V}{F} \quad (3.7)$$

$$C_A = \frac{C_{A_i}}{1 + k_A \tau} \quad (3.8)$$

$$C_B = \frac{C_{B_i} + k_A \tau C_A}{1 + k_B \tau} \quad (3.9)$$

$$Q = F \rho C_p (T - T_i) + V (r_A H_{RA} + r_B H_{RB}) (1 - \xi) \quad (3.10)$$

k_A^0 , k_B^0 are the pre-exponential Arrhenius constants for the reactions $A \rightarrow B$ and $B \rightarrow C$, E_A and E_B are the activation energies, R is the universal gas constant, τ is the space time, C_p is the heat capacity of the various streams, T_i is the inlet temperature of the reactants, T is the temperature of the material in the reactor, H_{RA} and H_{RB} are the molar heats of the reactions $A \rightarrow B$ and $B \rightarrow C$, respectively, and ξ is the percentage heat loss ($\xi = \frac{H_i}{V(\tau_A H_{RA} + \tau_B H_{RB})} \times 100\%$).

In this development it was assumed that:

- the reaction mixture and inlet stream were incompressible liquids,
- the densities and heat capacities of all processing streams were equal and constant.

- the molar heats of the reactions were constant.

More details on modelling such a system can be found in Stephanopoulos [30].

3.2 Classification of Terms

The terms in the design equations were classified as parameters (constants) or variables.

The system parameters and the values assigned to them for this study were:

$$\begin{array}{ll}
 k_A^0 & = 8.4 \times 10^5 \text{ min}^{-1} & k_B^0 & = 7.6 \times 10^4 \text{ min}^{-1} \\
 H_{RA} & = -2.12 \times 10^4 \text{ J/mol} & H_{RB} & = -6.36 \times 10^4 \text{ J/mol} \\
 E_A & = 3.64 \times 10^4 \text{ J/mol} & E_B & = 3.46 \times 10^4 \text{ J/mol} \\
 C_p & = 3.2 \times 10^3 \text{ J/kg K} & \rho & = 1180 \text{ kg/m}^3 \\
 R & = 8.314 \text{ J/mol K} & &
 \end{array}$$

The thirteen system variables and the units assigned were:

$$\begin{array}{ll}
 C_{A_i} & (\text{mol/m}^3) & C_{B_i} & (\text{mol/m}^3) \\
 C_A & (\text{mol/m}^3) & C_B & (\text{mol/m}^3) \\
 r_A & (\text{mol/m}^3 \text{ min}) & r_B & (\text{mol/m}^3 \text{ min}) \\
 T_i & (\text{K}) & T & (\text{K}) \\
 V & (\text{m}^3) & F & (\text{m}^3/\text{min}) \\
 \tau & (\text{min}) & \xi & (\%) \\
 Q & (\text{J/min}) & &
 \end{array}$$

3.2.1 Degrees of Freedom

The nominal value for the heat loss was taken as 5% of the total heat generated by the reactions, leaving 12 unspecified system variables. Thus, with the six design equations, there are six degrees of freedom associated with the system. The design objective was to produce 60 mol/min of the component B. With this additional constraint the number of degrees of freedom of the system is reduced

to five.

With five degrees of freedom the designer must specify five of the free system variables.

3.2.2 Selection of the Design and Error Variables

What are the design variables ?

The design variables chosen for this study were:

- the inlet concentration of A, C_{A_i} ,
- the inlet concentration of B, C_{B_i} ,
- the inlet temperature, T_i ,
- the reactor temperature, T,
- and the volume of the reactor, V.

What are the error variables ?

The selected error variables (that transmit error to the production rate were:

- the inlet concentration of A, C_{A_i} ,
- the inlet concentration of B, C_{B_i} ,
- the inlet temperature, T_i ,
- the flow rate, F,
- and the heat loss, H_l .

Two systems will be studied. The first one has a two-dimensional design region, where the inlet concentration of A, the inlet concentration of B and the inlet temperature were kept on their central values. Only the temperature and the volume of the reactor were under study. With two design variables the variance surfaces may be examined. The second system consisted of the complete five design variables.

Chapter 4

Monte Carlo Simulations

The approach of Monte Carlo simulations is the most rigorous, it allows for testing a variety of distributional forms for the error variables and provides a means of examining the resulting errors in the output variables without any simplifying assumptions.

The method was used in conjunction with two different optimization routines, namely, a nonlinear optimization routine and Response Surface Methodology.

4.1 Methodology

The major steps required to perform this approach are the following: computation of the nominal design, generation of the random deviates, determination of the actual variable values, computation of the variance and performance optimization.

The corresponding flowchart of the FORTRAN program written for this method is presented in Figure 4.1.

4.1.1 Initial Guesses for Design Variables

For any optimization routine, initial guesses are required to start the procedure. To ensure convergence to global optimum, different initial guesses were tried. These initial guesses were determined using a complete factorial design, with three levels for each design variable. In summary 9 initial guesses were used in the case of two design variables and 243 for the complete set of five design variables. The design region considered was the following:

- $1000 \leq C_{A_i} \leq 5000 \text{ mol/m}^3$
- $100 \leq C_{B_i} \leq 500 \text{ mol/m}^3$
- $290 \leq T_i \leq 330 \text{ K}$
- $290 \leq T \leq 330 \text{ K}$
- $10 \leq V \leq 90 \text{ l}$

4.1.2 Computation of Nominal Design

Given the values of all the design variables, the rate constants, the space time, the flow rate, and the heat removed from the reactor, the output concentrations were computed using the design equations listed in Chapter 3.

4.1.3 Generation of the Deviations

Deviations from the nominal values for the error variables which are C_{A_i} , C_{B_i} , T_i , F , and H_i were generated.

The seed for the random number generator, and the sample size were required. The distributions considered were normal and log-normal distributions.

The standard deviation for each error variable was expressed in terms of the percentage of the nominal value. For log-normal distribution the standard deviation used was the original standard deviation divided by the nominal value of the error variable. The corresponding IMSL subroutines employed were GGNML and GGNLG. The sample size was varied from 50 to 2000, to investigate its effect, and the seed value was changed from 1 to 10^{21} .

The increase in the sample size yielded more computations. For simple systems this is not a major problem, but for more complicated systems this could lead to a large amount of computation.

4.1.4 Solution of System Equations

Once the deviations were generated, the system of design equations had to be solved. The problem consisted of solving a nonlinear equation in terms of the reaction temperature. A subroutine called ZREAL was used to achieve this.

The validity of the solution was verified by the supplier subroutine WRRRN from IMSL library.

4.1.5 Computation of Variance

The variance of the production rate was the performance index for this design. It was computed using the mean value of the production rate and the set

of response values corresponding to the simulations performed

$$V_P = \frac{\sum_{i=1}^n (P_i - \bar{P})^2}{n - 1} \quad (4.1)$$

where $P_i = FC_B$ is computed at each simulation, \bar{P} is the mean value for the production rate and n the sample size.

4.1.6 Optimization

Two subroutines were used: a nonlinear multivariable optimization routine and Response Surface Methodology. The nonlinear optimization subroutines were NCONF, BCONF and BCPOLE taken from the IMSL. A tolerance of 10^{-8} was used, and verification of the global optimum was done by WRRRN, a supplier subroutine for the optimization software. To obtain an empirical model for the production rate variance in terms of the design variables, Response Surface Methodology was applied. The method also gives the optimal setting of design variables by minimizing the expression obtained using an analytical derivation or a constrained optimization.

The first-order model was fitted using an orthogonal first-order design. For the second-order model central composite design was applied. An IMSL subroutine called RGIVN was used to provide the regression coefficients.

The central composite design employed for the system of two design variables (temperature and reactor volume) was a 2^2 factorial design augmented by 4 axial points $(\pm\alpha, 0)$, $(0, \pm\alpha)$ and five central points $(0,0)$, where α is equal to 1.414.

In the case of five design variables, the central composite design employed consisted

of a 2^5 factorial design augmented by 10 axial points $(\pm\alpha, 0, 0, 0, 0)$, $(0, \pm\alpha, 0, 0, 0)$, $(0, 0, \pm\alpha, 0, 0)$, $(0, 0, 0, \pm\alpha, 0)$, $(0, 0, 0, 0, \pm\alpha)$, and 10 central points $(0, 0, 0, 0, 0)$, where α is equal to 2.378.

The program flowchart for Response Surface Methodology is shown in Figures 4.2 and 4.3.

4.2 Results and Discussion

The effect of the sample size required for Monte Carlo simulations on the variance and the mean of the production rate was examined. The results of this investigation are presented in Appendix E. The routines for generating both normal and log-normal errors were found to yield reliable results for sample size of 1600 or greater. Thus for each set of design conditions simulation using 1600 points was carried out in order to evaluate the mean and the variance of the target variable.

To investigate the sensitivity of the production rate to each one of the error variables, Monte Carlo simulations were performed generating deviations for one error variable at a time. The remaining error variables were kept at their central values. Normally and log-normally distributed errors were considered (see Figures 4.4 and 4.5, respectively). The results for both cases were similar except that the variance was always slightly smaller for log-normally distributed errors. It was observed that a change in the inlet concentration of B and the heat loss do not affect the output variance. These variables were then considered as adjustment variables, where their effect is on the mean value of the production rate only. On

the other hand, a small change in the remaining variables affected the output variance substantially, especially for error levels greater than 4% .

Monte Carlo simulations were performed using a sample of 1600 observations for both normally and log-normally distributed errors. The mean of the production rate was observed to decrease with the input variable error level for both cases (see Figure 4.6). In fact, the mean value was reduced from almost 60 to 54 for a 10% error level.

On the other hand, the system output variance was observed to increase with the input variables error level (see Figure 4.7). The mean was also plotted against the variance (see Figure 4.8) to examine any possibility of correlation between the two statistics. Indeed, they were observed to be highly correlated as any increase in the variance produced a decrease in the mean value for both normally and log-normally distributed errors.

In the case of two design variables, contours of the variance surfaces over the design region were drawn for different error levels (Figure 4.9). The design variables were observed to affect considerably the system output variance. Therefore they have to be properly selected for optimum performance. The optimal setting of the design variables was also observed to depend on the considered error level. In fact, the reactor temperature for optimum performance was increased from 313 to 326 degrees when the noise error increased from 2 to 6% .

To investigate the effect of the error level on the output distribution, Monte Carlo simulations were used to generate values of the production rate associated

with the different sets of input variables. The five design variables were kept at their central values. It was found that the distribution of the production rate was similar to that of the input variables. It is also important to note that for high error levels (7 and 8%) the distribution deviates from its normal or log-normal behavior (see Figures 4.10 to 4.14). The typical histograms for the production rate distribution were drawn using an error level of 5% for each error variable. They are shown in Appendix E. At this error level the normal or log-normal structure of the errors for the input variables was conserved.

With the use of Response Surface Methodology, no major improvement in performance with regard to the variance value, was achieved (see Figures 5.4 and 5.5).

A constrained optimization was carried out using NCONF, BCONF and BCPOLE from the IMSL library in order to get the optimal settings of the design variables.

Even though the empirical model obtained needs a constrained optimization routine, the method involves fewer computations. It required about 120 cpu seconds instead of 200 cpu seconds for Monte Carlo simulations in University of Ottawa AMDAHL mainframe computer.

4.3 Conclusions

The conclusions that can be drawn are:

- the method was found to be reliable for large sample sizes;

- the inlet concentration of B and the heat loss were found to be adjustment variables,
- the levels of the design variables affect considerably the process performance (up to 10 times);
- The inlet temperature was found to have the greatest effect on the variance of the production rate;
- Response Surface Methodology associated with Monte Carlo simulations was found to be efficient in determining the optimal setting of design variables.

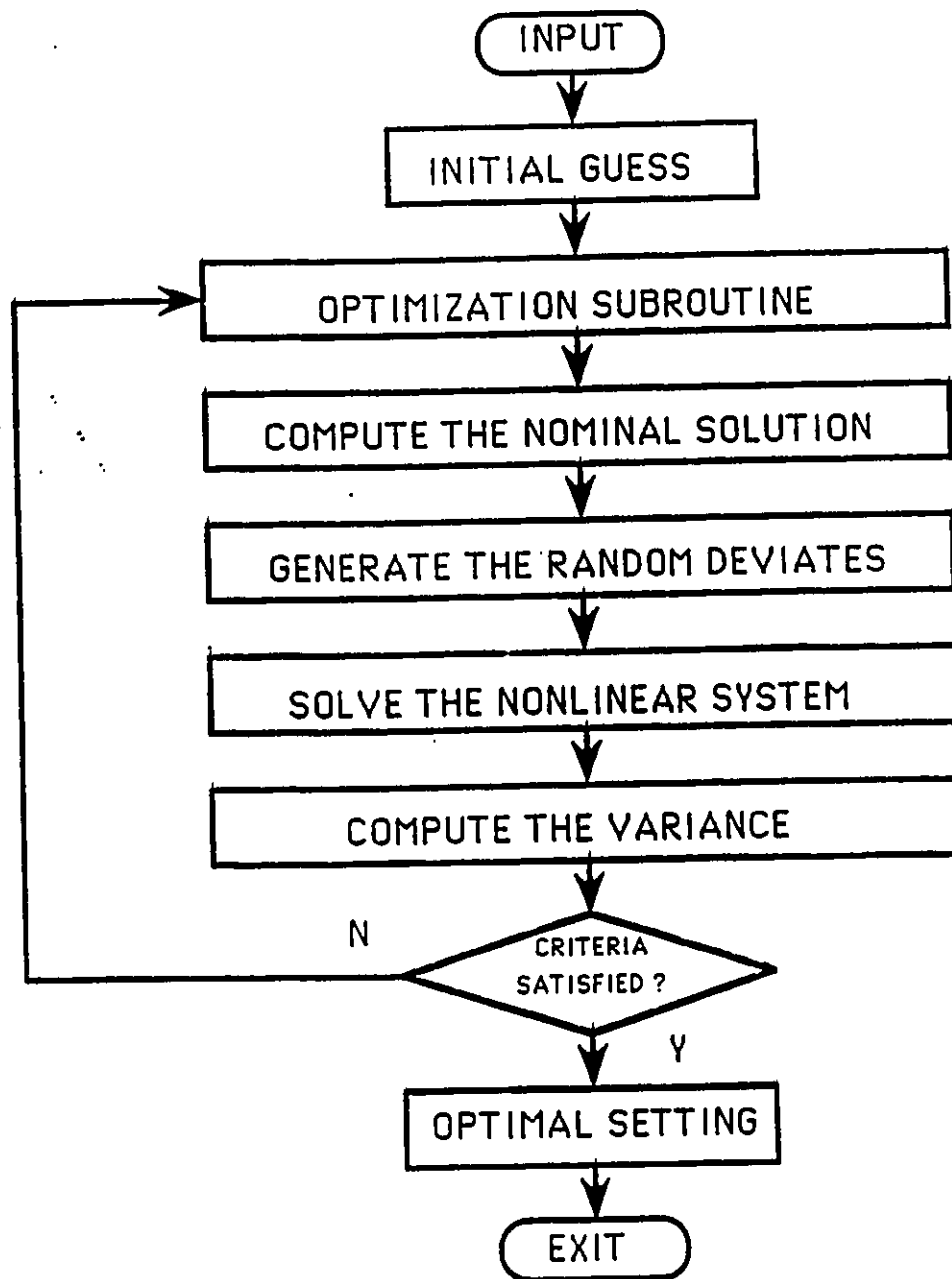


Figure 4.1: The program flowchart for Monte Carlo simulations.

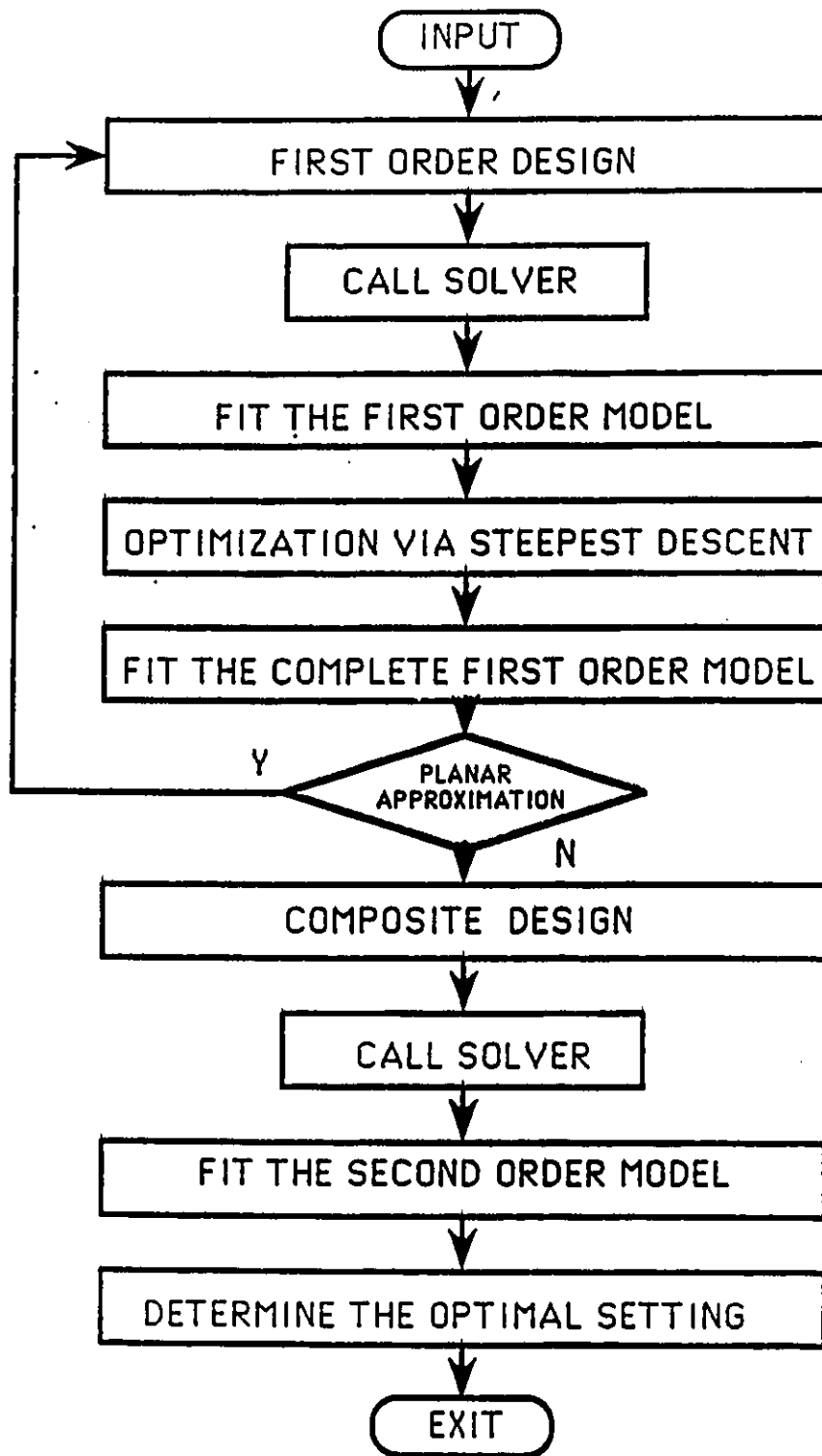


Figure 4.2: The program flowchart for Response Surface Methodology.

SUBROUTINE SOLVER

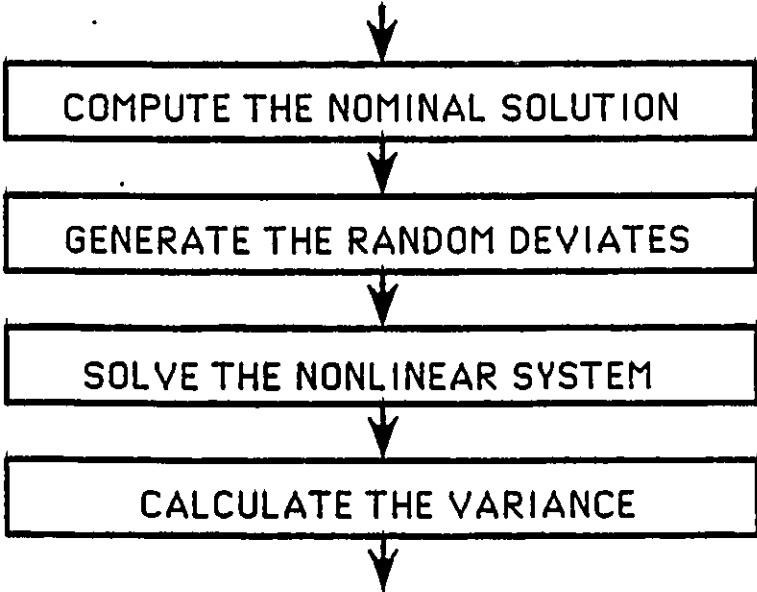


Figure 4.3: The SOLVER flowchart.

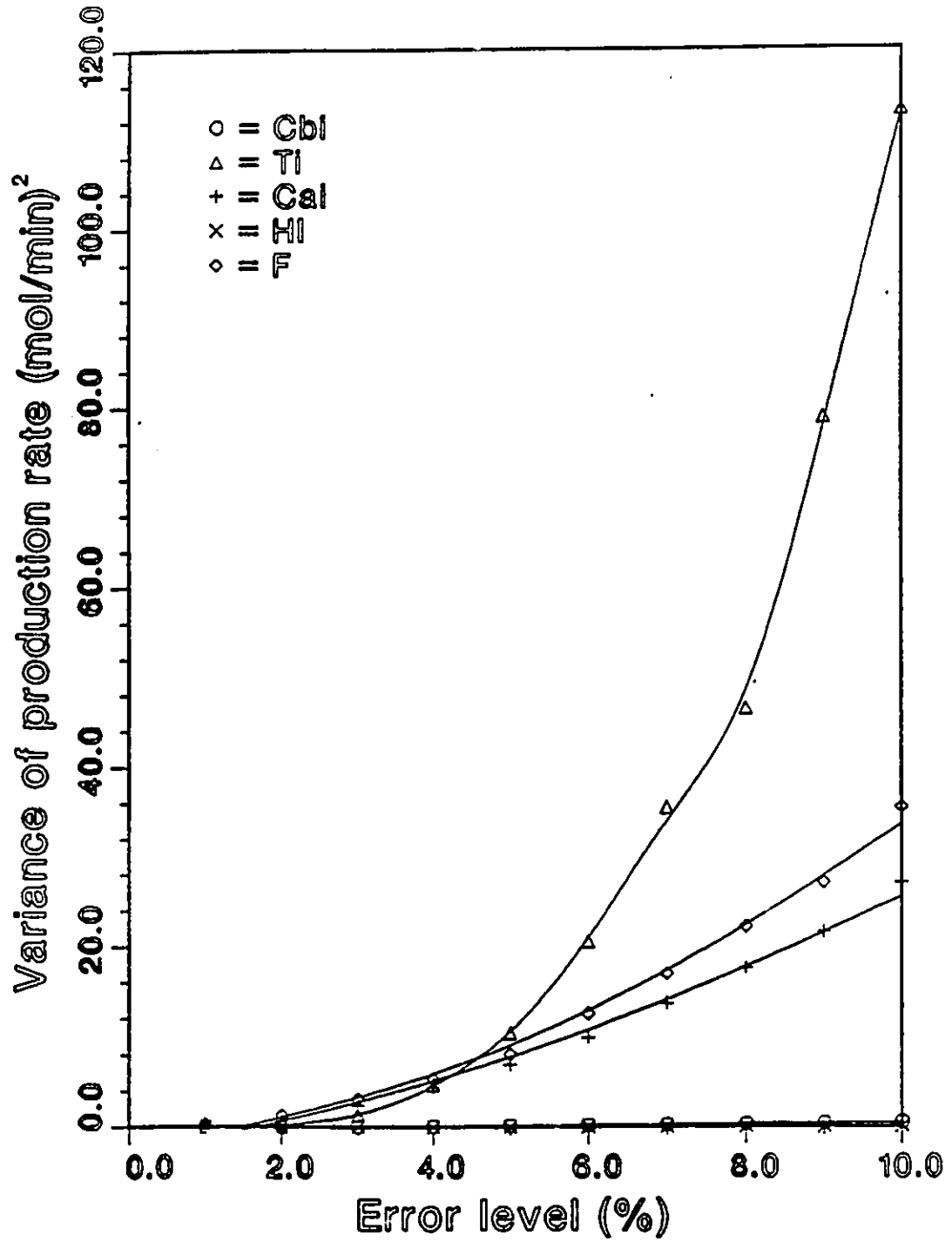


Figure 4.4: The effect of error level for the various error variables on the performance measure (variance of production rate) using normally distributed errors for the error variables. Error level= $[(\text{Std. Dev. } 100\%)/\text{Nominal Value}]$.

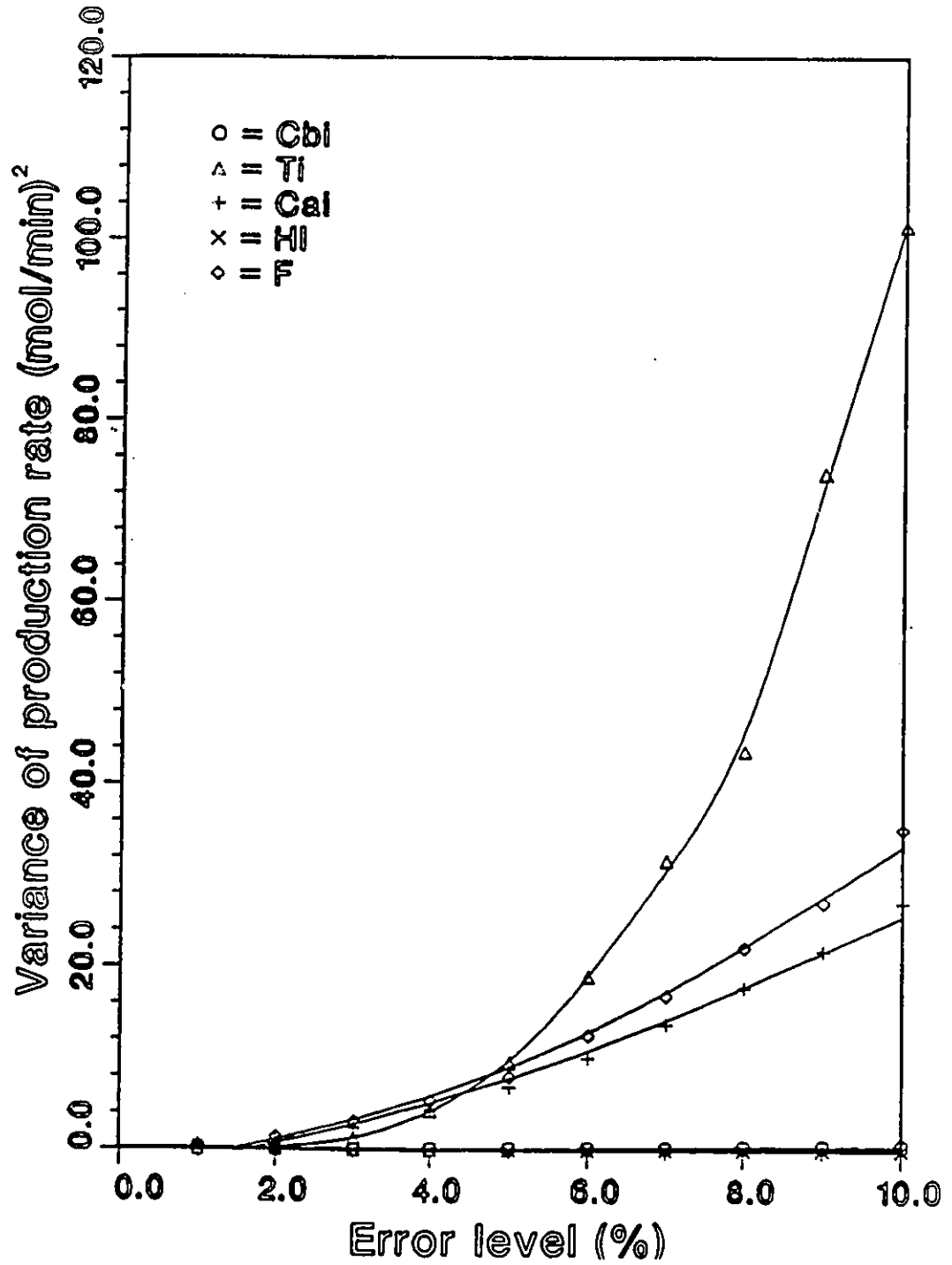


Figure 4.5: The effect of error level for the considered error variables on the performance measure (variance of production rate) using log-normally distributed errors for the error variables. Error level= $[(\text{Std. Dev. } 100\%)/\text{Nominal Value}]$.

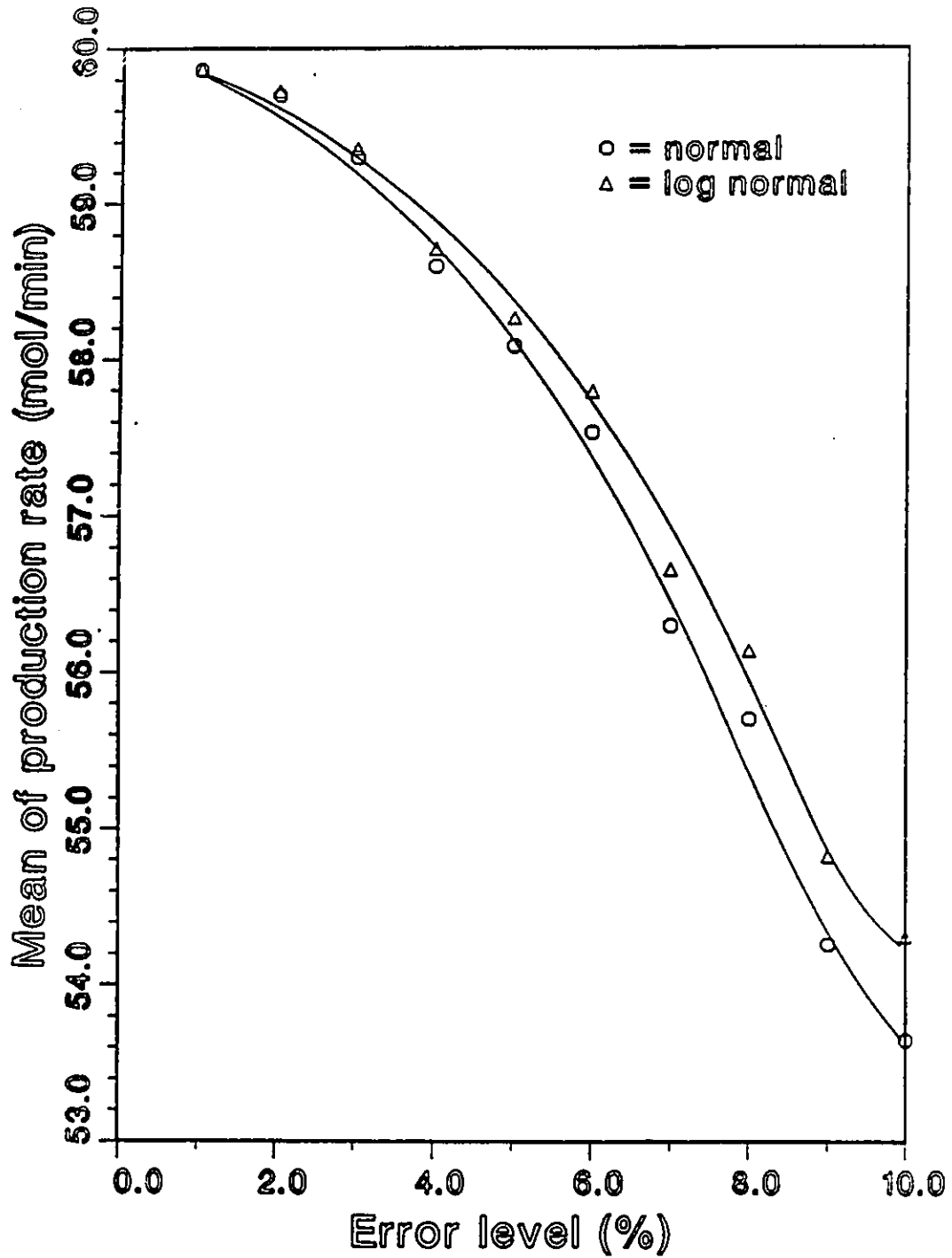


Figure 4.6: The effect of the error level on the mean of the production rate using normally and log-normally distributed errors. Error level= $[(\text{Std. Dev. } 100\%)/\text{Nominal Value}]$.

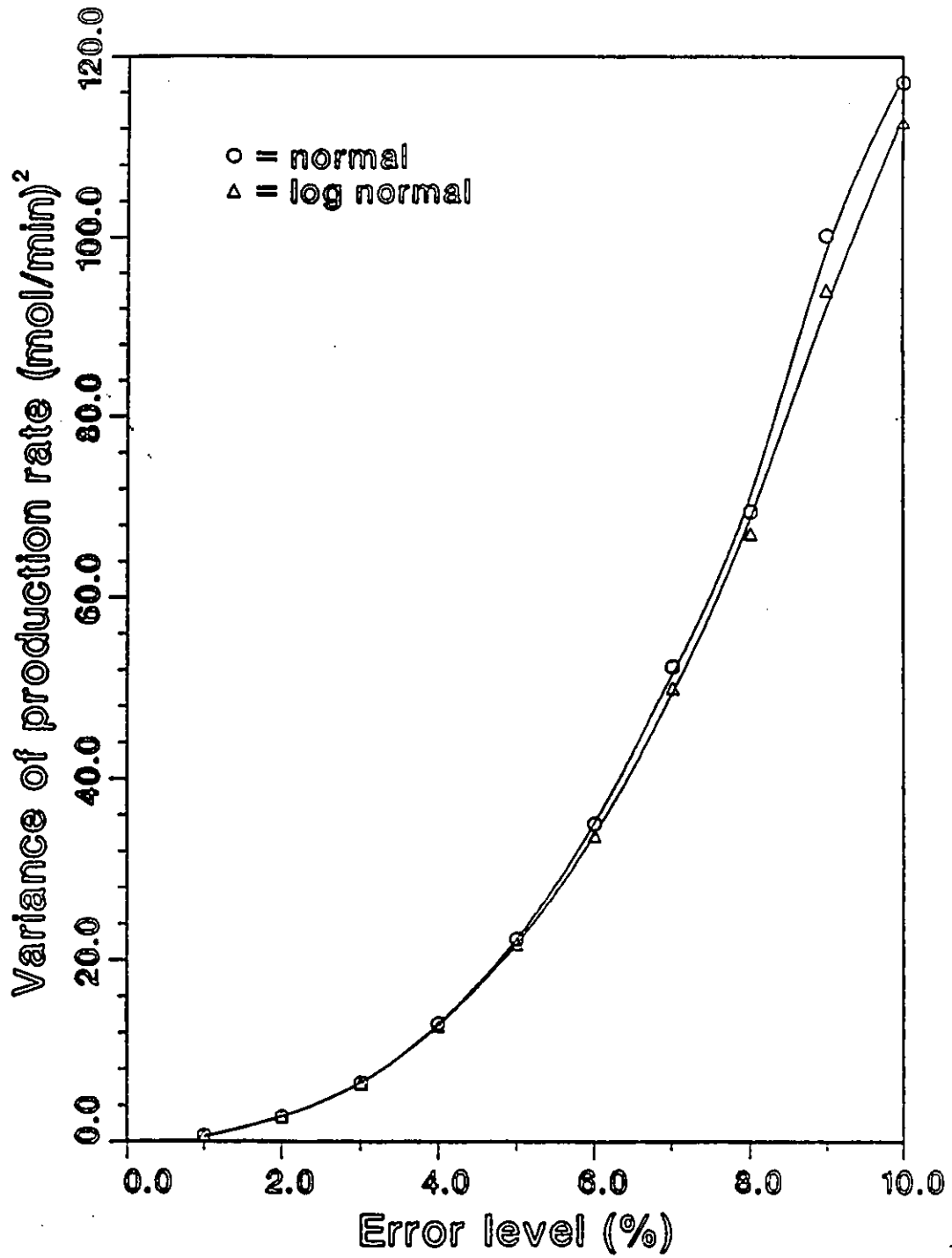


Figure 4.7: The effect of the error level on the variance of the production rate using normally and log-normally distributed errors. Error level= $[(\text{Std. Dev. } 100\%)/\text{Nominal Value}]$.

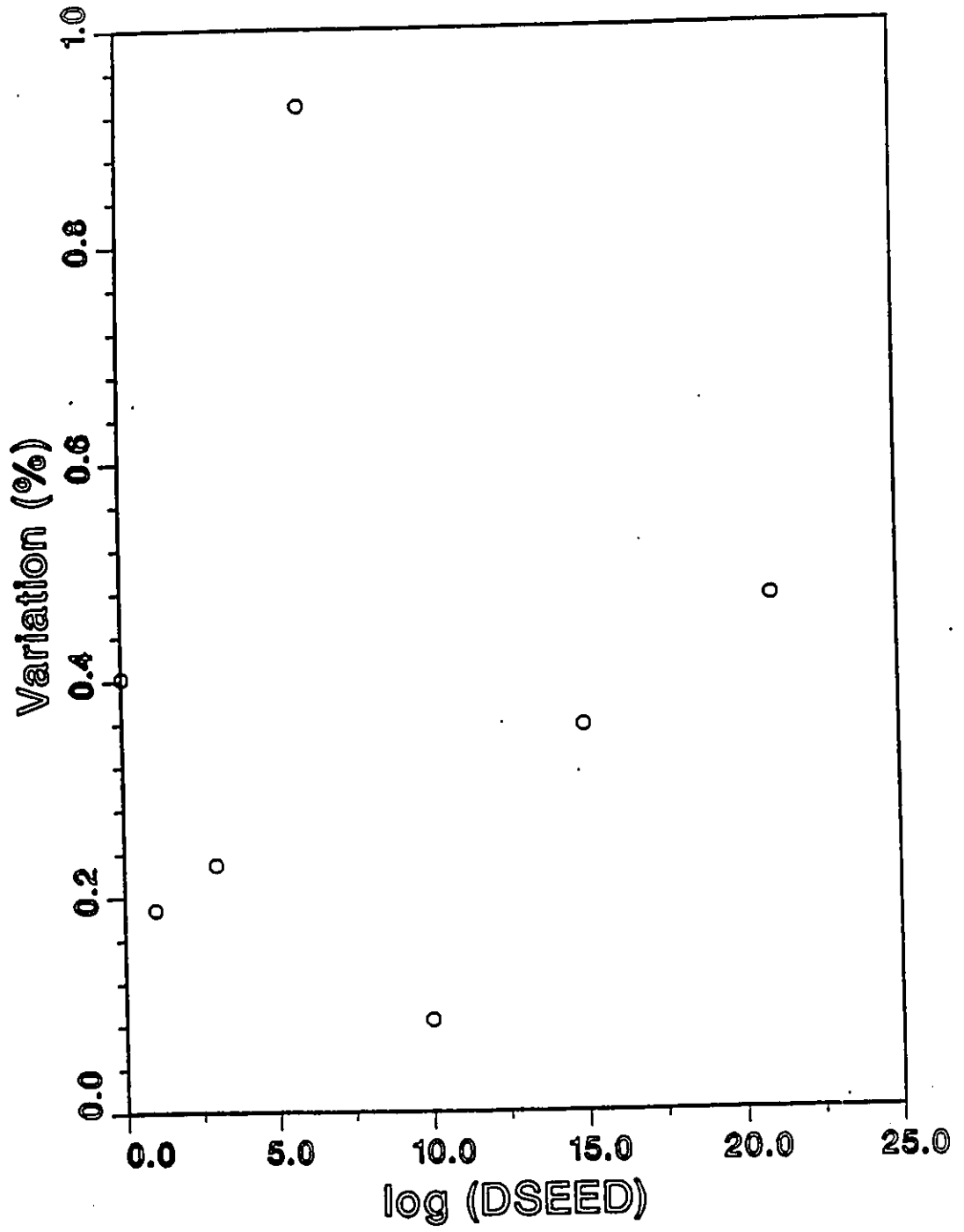


Figure 4.8: The sensitivity of the variance with the seed value (normally distributed errors are considered).

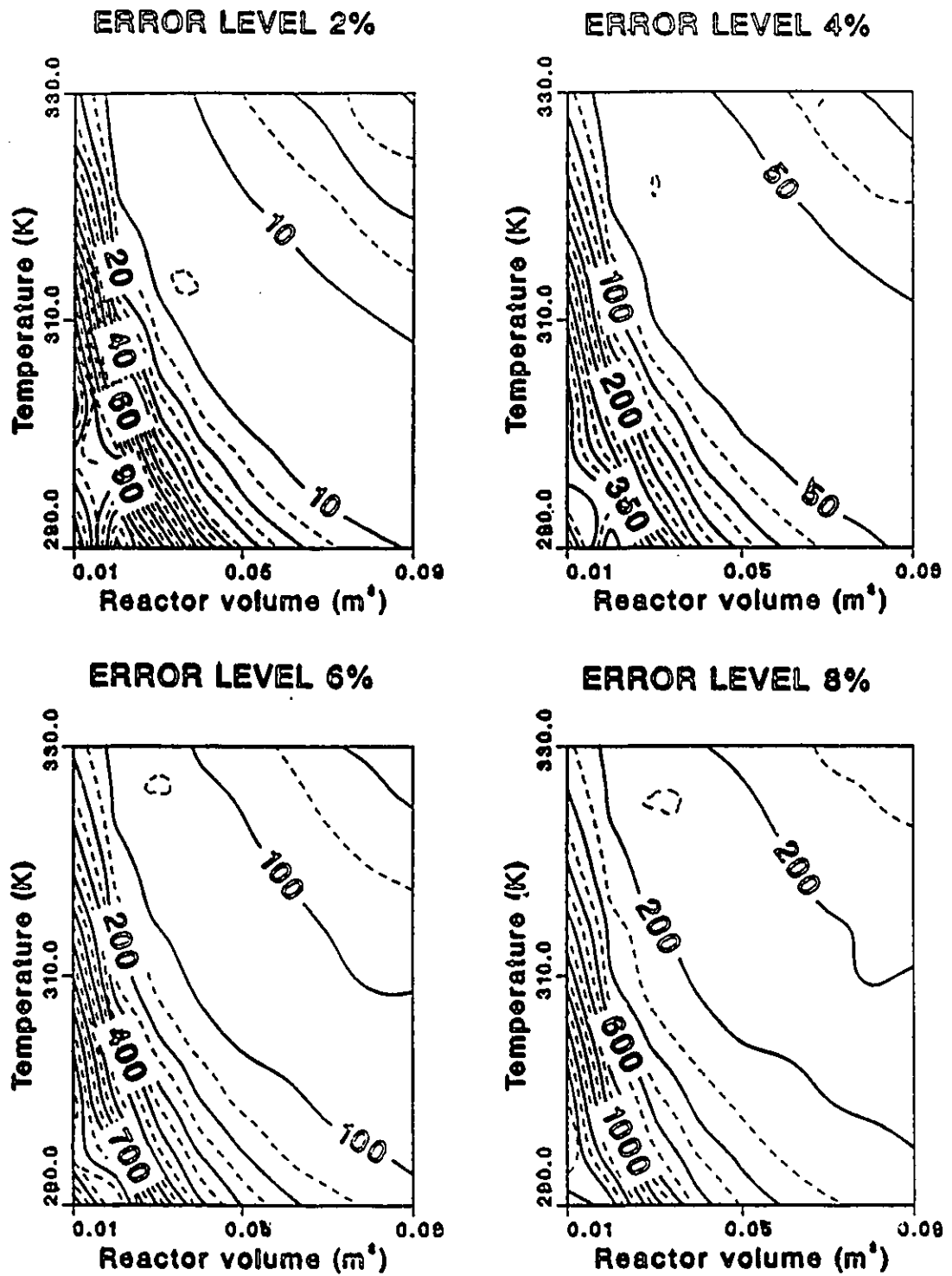


Figure 4.9: Variance contours generated by Monte Carlo simulations using two design variables and normally distributed errors. Error level= $[(\text{Std. Dev. } 100\%)/\text{Nominal Value}]$.

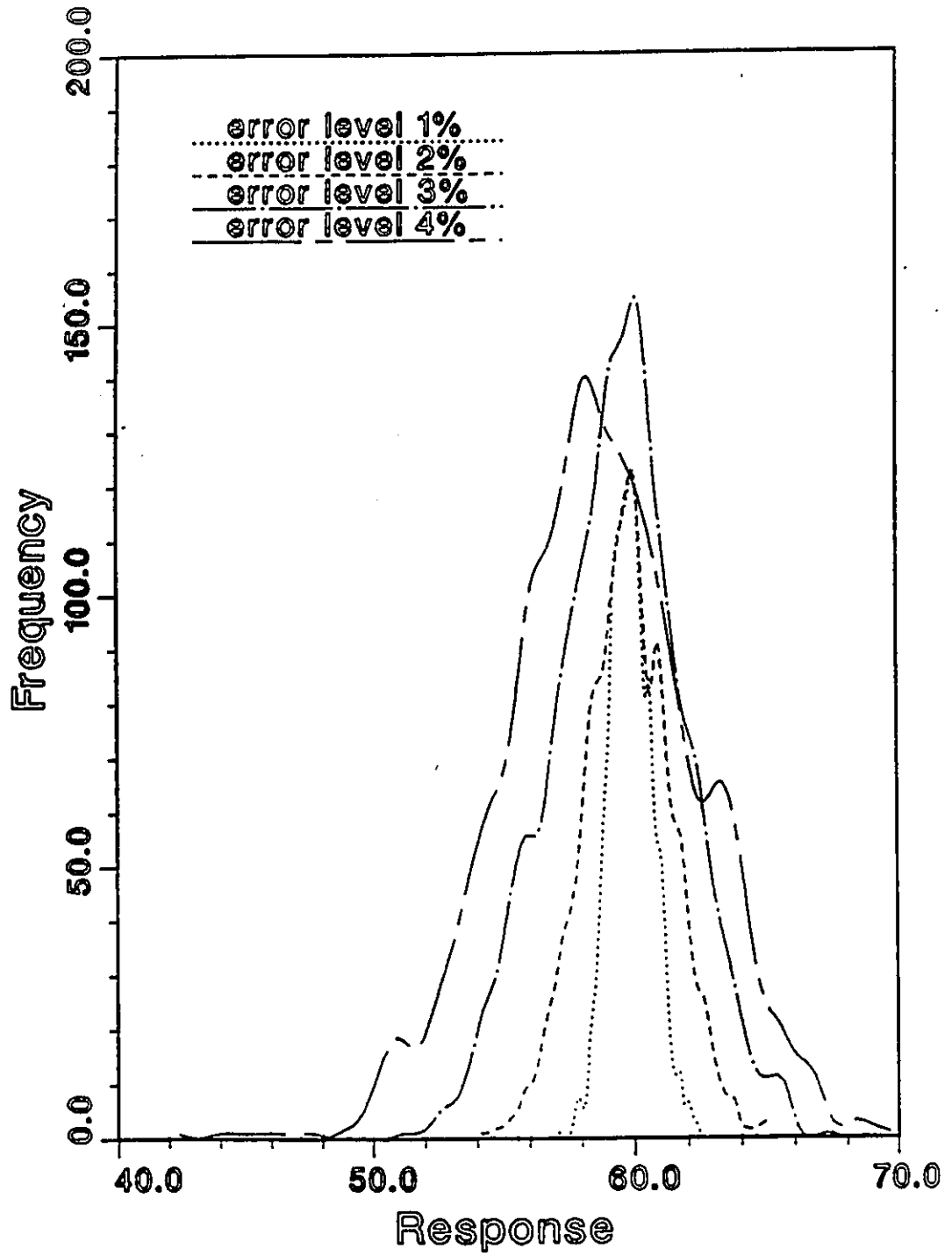


Figure 4.10: Production rate distribution for four different error levels (normally distributed errors).

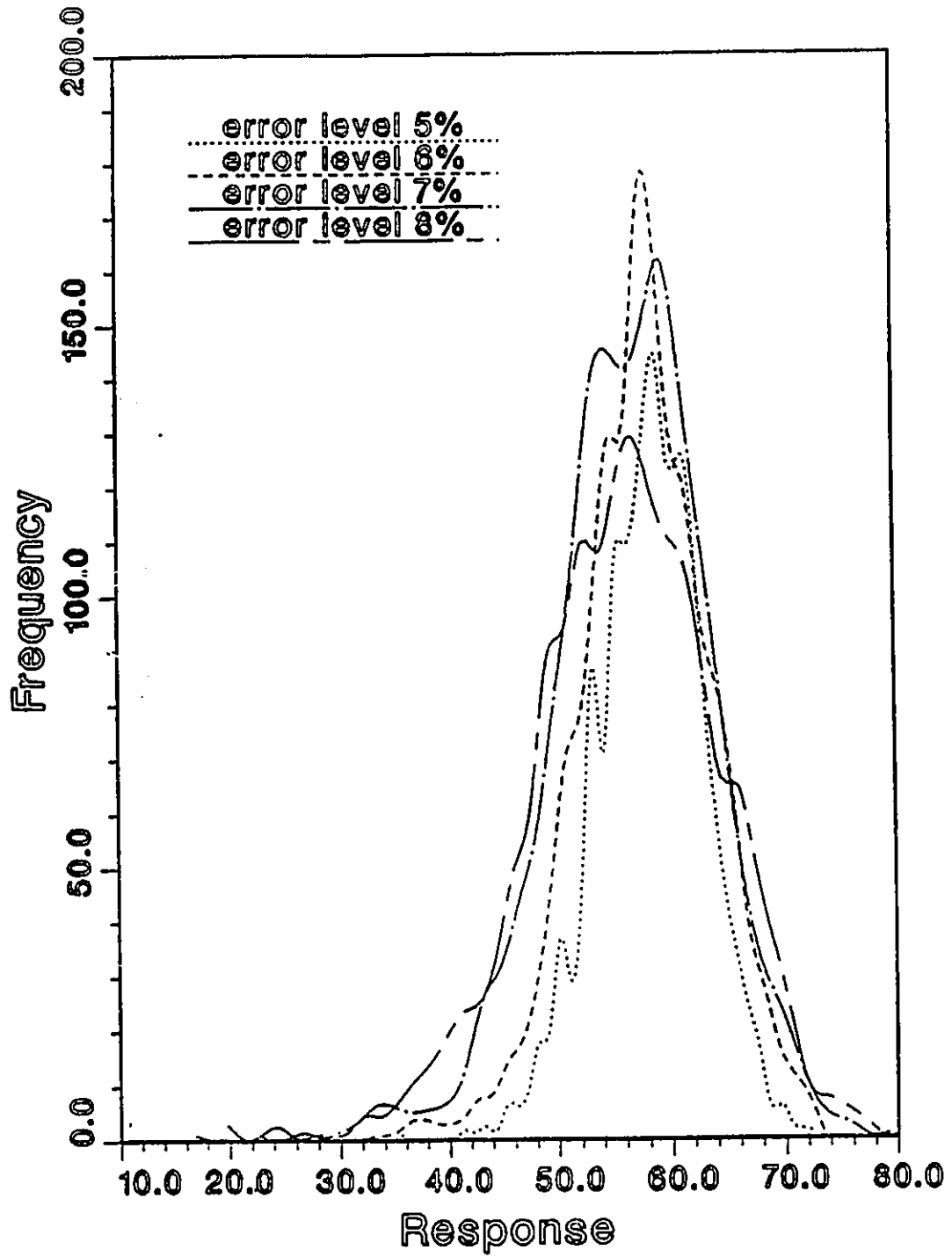


Figure 4.11: Production rate distribution for four different error levels (normally distributed errors).

Table 4.1: Optimal settings using Monte Carlo simulations with five design variables for normally distributed errors.

error level (%)	inlet concentration of A (mol/m^3)	inlet concentration of B (mol/m^3)	inlet temperature (K)	temperature (K)	volume (l)
1	3083.8	355.3	313.5	318.7	44.9
2	3118.0	342.5	314.4	314.0	39.1
3	3044.1	372.2	310.6	319.5	48.8
4	3827.6	424.5	307.9	308.3	44.1
5	4537.0	370.3	308.0	304.3	35.0
6	4905.6	457.7	307.2	302.1	36.3
7	4992.8	400.0	308.5	300.6	33.7
8	4800.4	456.1	314.2	290.8	29.9
9	4907.7	359.3	313.3	294.7	30.0
10	3762.4	425.0	324.8	314.5	25.9

Table 4.2: Optimal settings using Monte Carlo simulations and Response Surface Methodology with five design variables and normally distributed errors.

error level (%)	inlet concentration of A (mol/m^3)	inlet concentration of B (mol/m^3)	inlet temperature (K)	temperature (K)	volume (l)
1	3145.5	354.2	319.8	312.3	45.8
2	3180.4	341.4	320.7	307.7	39.9
3	3105.0	371.1	316.8	313.1	49.8
4	3904.1	423.2	314.1	302.1	45.0
5	4627.7	374.7	314.1	298.2	35.7
6	5000.0	463.5	313.3	296.2	37.0
7	5000.0	408.0	314.7	294.6	34.4
8	4896.4	465.2	320.2	290.1	30.5
9	5000.0	358.2	319.6	290.0	30.6
10	3837.6	430.2	330.0	308.3	26.4

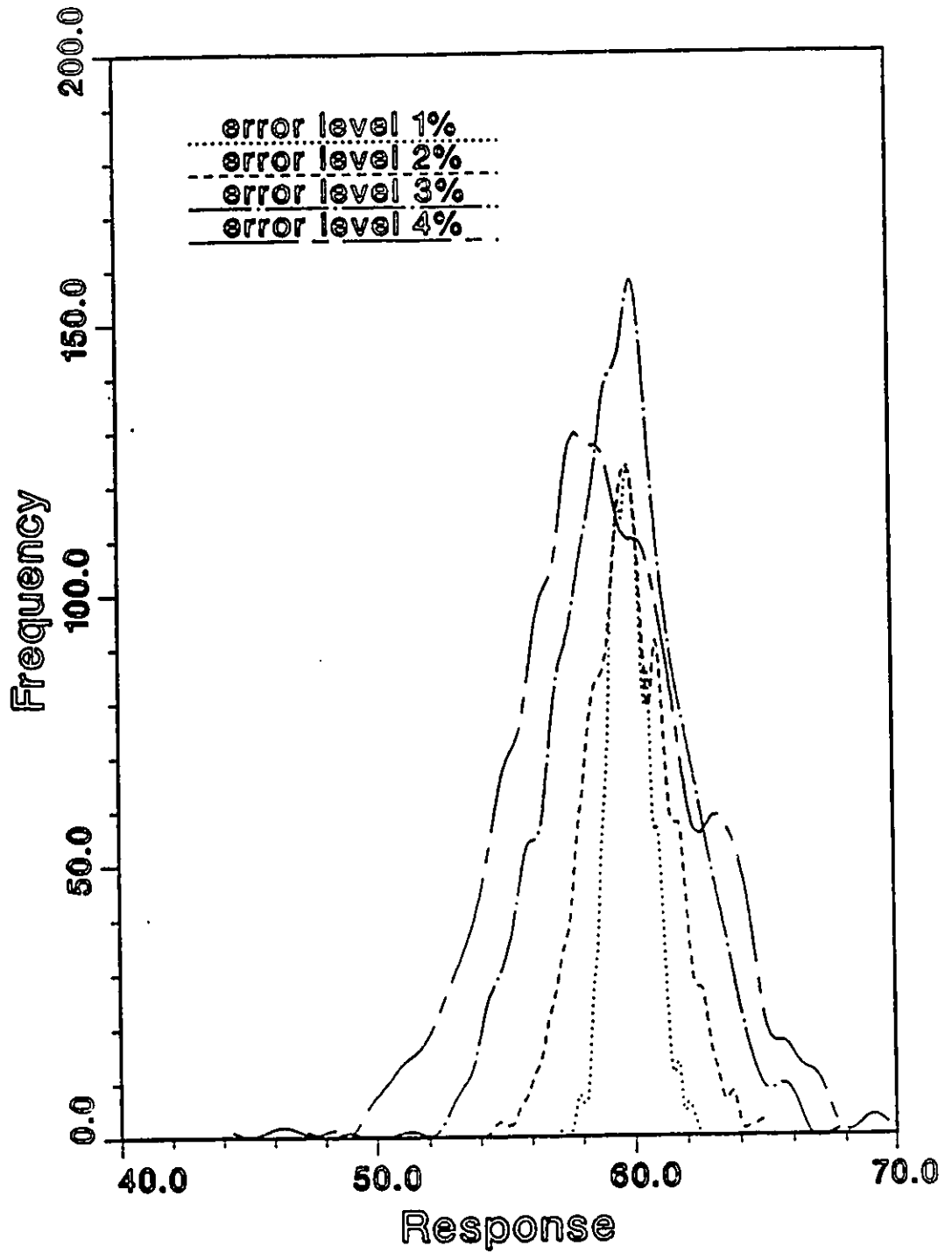


Figure 4.12: Production rate distribution for four different error levels (log-normally distributed errors).

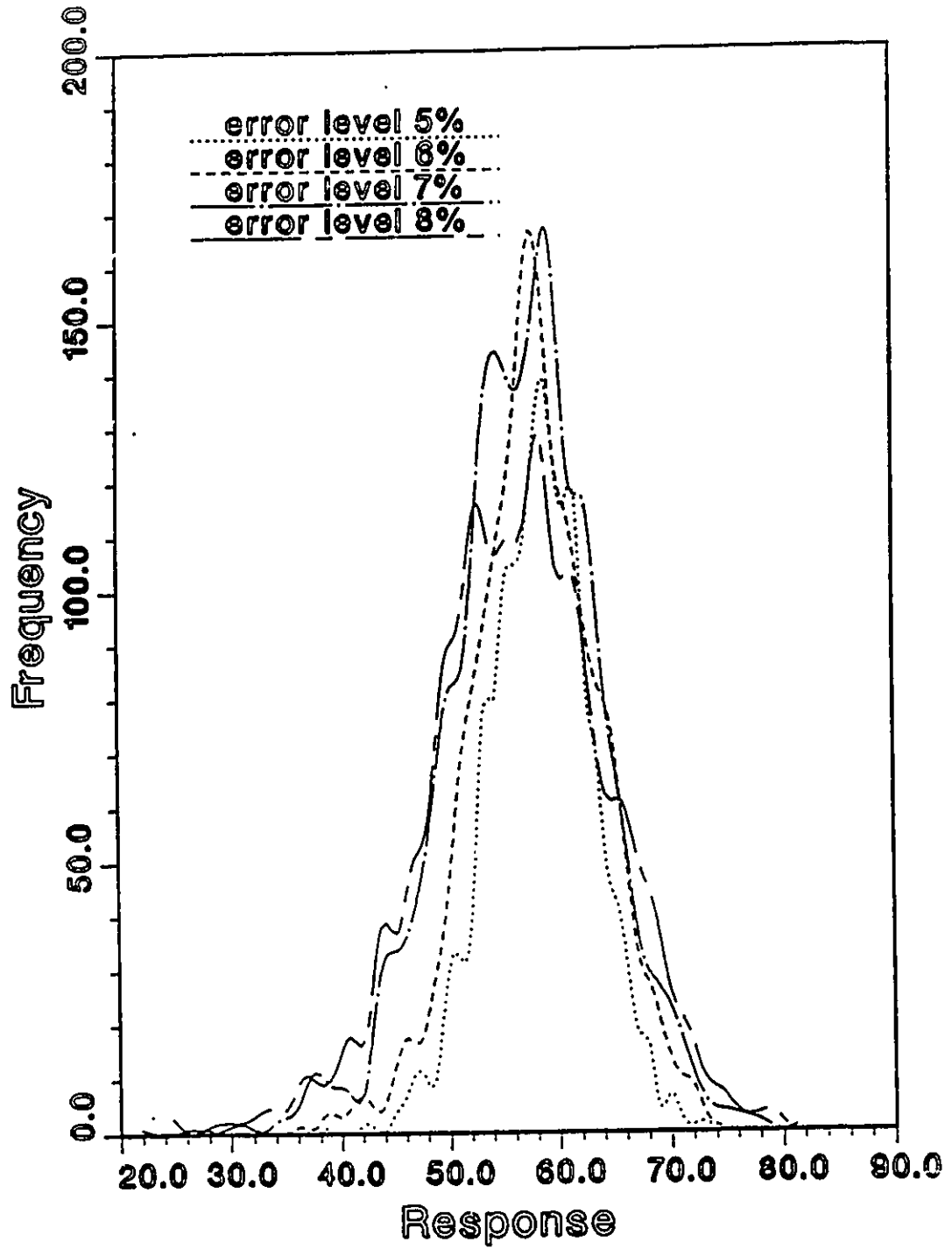


Figure 4.13: Production rate distribution for four different error levels (log-normally distributed errors).

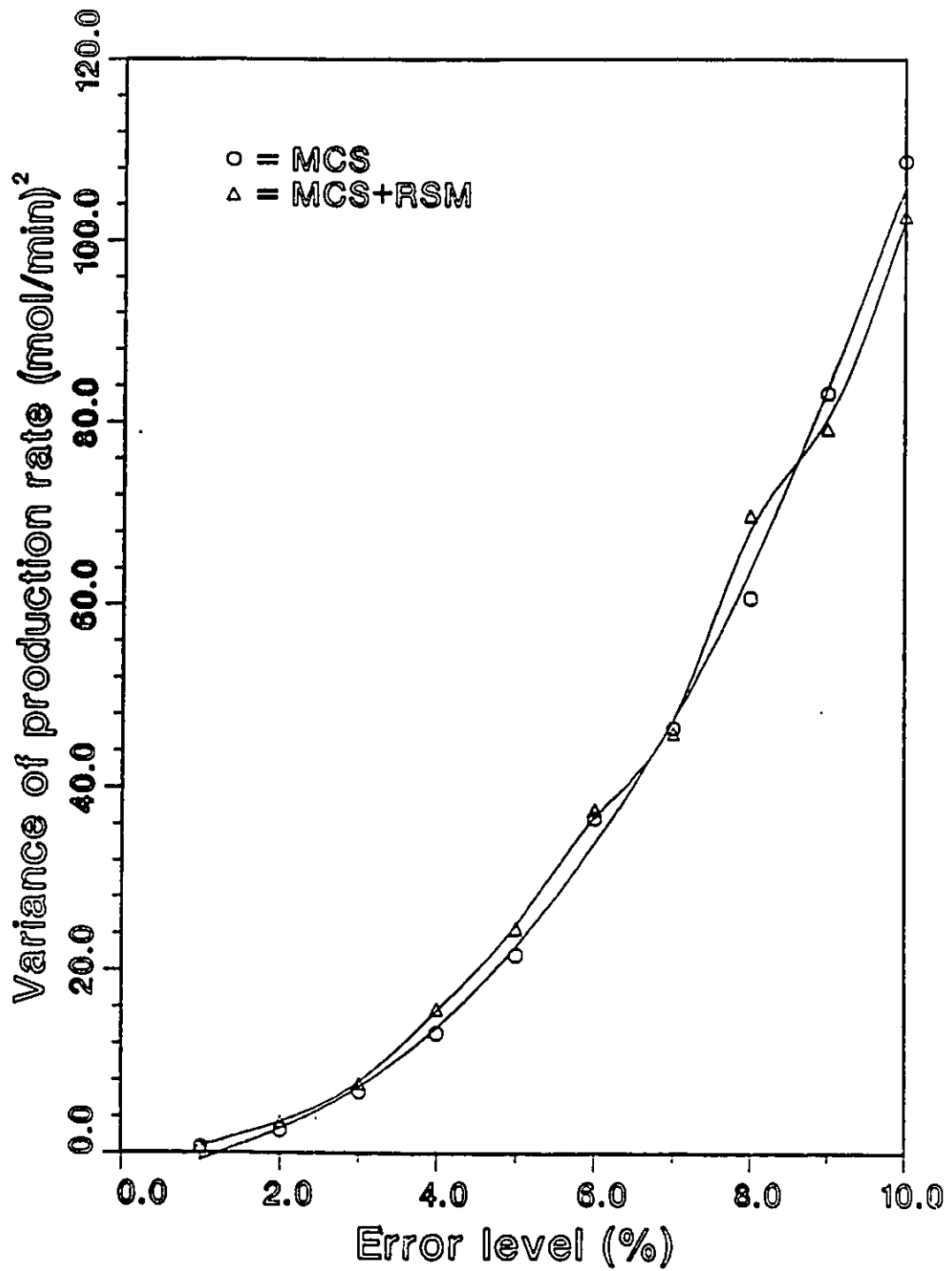


Figure 4.14: The effect of incorporating the Response Surface Methodology in Monte Carlo simulations considering two design variables and normally distributed errors. Error level=[(Std. Dev. 100%)/Nominal Value].

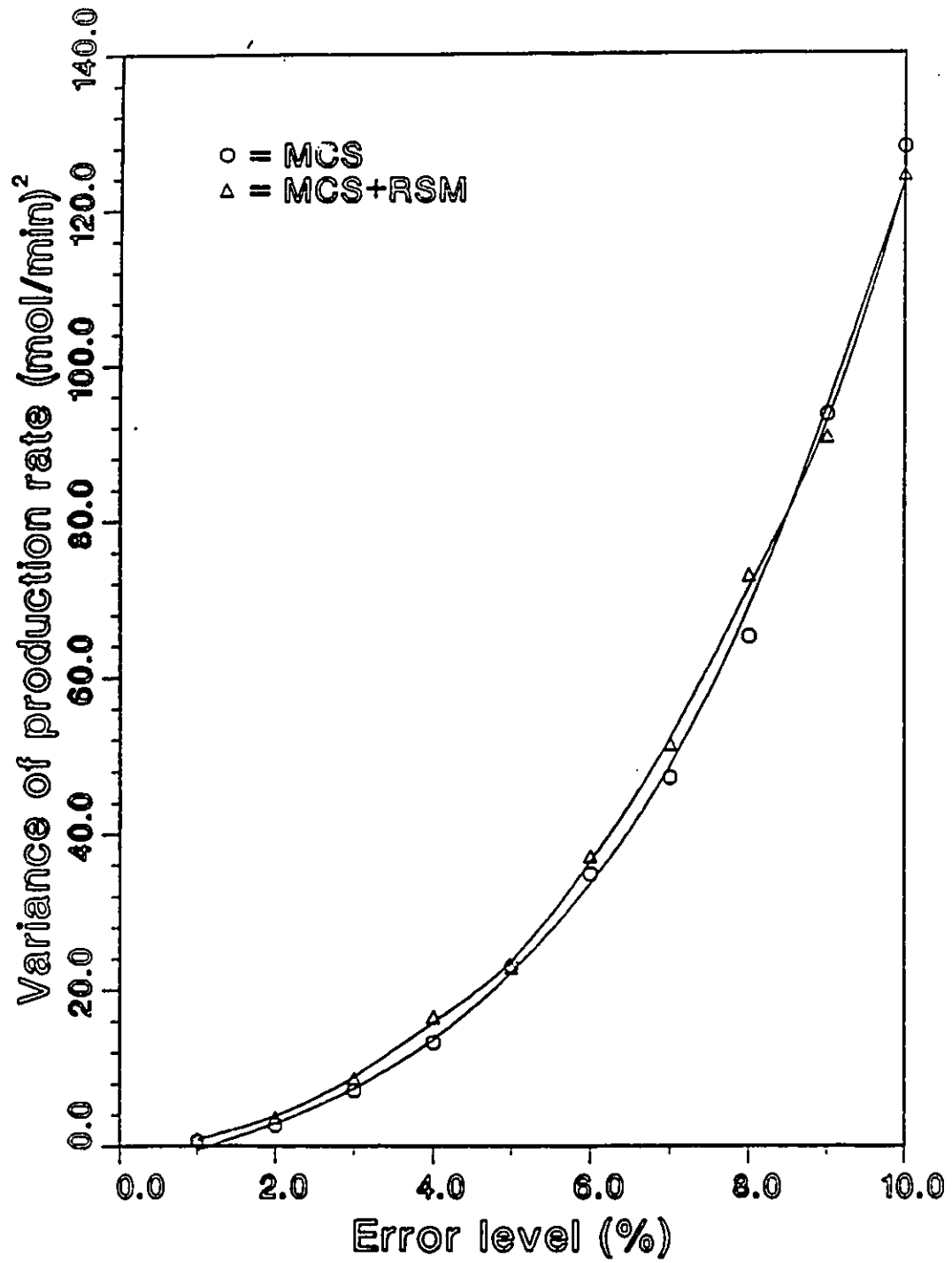


Figure 4.15: The effect of incorporating the Response Surface Methodology in Monte Carlo simulations considering two design variables and log-normally distributed errors. Error level= $[(\text{Std. Dev. } 100\%)/\text{Nominal Value}]$.

Chapter 5

Error Transmission Analysis

In order to reduce the large computational effort required for Monte Carlo simulations, a first-order error propagation formula was used to evaluate the variance of the process output. This formula uses a first-order Taylor series approximation to the design equations and results in a straight forward means to evaluate the output variance.

5.1 Methodology

The major steps involved in this approach were : computation of the nominal design, determination of the derivatives, computation of the transmitted variation function and optimization of the performance measure. The flowchart for the FORTRAN program is given in Figure 5.1.

5.1.1 Preliminary Steps

The initial guesses for the design variables and computation of the nominal design were already discussed in sections 4.1.1 and 4.1.2. The same procedures were employed for tests using Error Transmission Analysis.

Systems of two and five design variables were studied. For each one, the settings of design variables that minimize the transmission of error from input variables to the output were determined.

5.1.2 Computation of the Derivatives

Even though the system considered (CSTR) was not very complicated, no analytical expression could be obtained for the derivatives of the production rate with respect to all error variables. An IMSL subroutine (DERIV) was used to provide numerical derivatives of the production rate with respect to all design variables. The step size was automatically changed to enhance accuracy.

5.1.3 Transmitted Variation Function

The propagation formula used was:

$$V_{\underline{x}} = d_1^2(\underline{x})\sigma_1^2 + d_2^2(\underline{x})\sigma_2^2 + d_3^2(\underline{x})\sigma_3^2 + d_4^2(\underline{x})\sigma_4^2 + d_5^2(\underline{x})\sigma_5^2 \quad (5.1)$$

where σ_i and $d_i(\underline{x})$ are the standard deviation and the derivative of the response with respect to the error variable x_i evaluated at the design condition. The standard deviations were given by the error level for each error variable.

5.1.4 Optimization

Several nonlinear multivariable optimization routines were used: NCONF, BCONF and BC POL from the IMSL library. A tolerance of 10^{-8} was used. A verification of the global optimum was done by checking the comments of WRRRN, a supplier subroutine for the optimization software.

5.2 Results and Discussion

The optimal setting of design variables obtained are listed in Tables 5.1 and 5.2. From Table 5.1 the optimal temperature seems to be independent from the error level. Compared with the results reported earlier for Monte Carlo simulations, the method was not successful. Even though the variance was minimized using an optimization routine, the variability of the optimal design obtained was still large especially when the number of design variables increases. This is explained by the use of the first-order approximation of the transmitted variation function. The linearization of the model is not valid.

The amount of computation is considerably reduced compared to Monte Carlo simulations; 5 to 10 cpu seconds were required.

5.3 Conclusions

The conclusions that can be drawn are the following:

- The method was not successful in locating the optimal conditions,
- ETA performs even worse when the number of variables increases,
- The first order-approximation of the transmitted variation function does not describe the nonlinearity of the system considered.

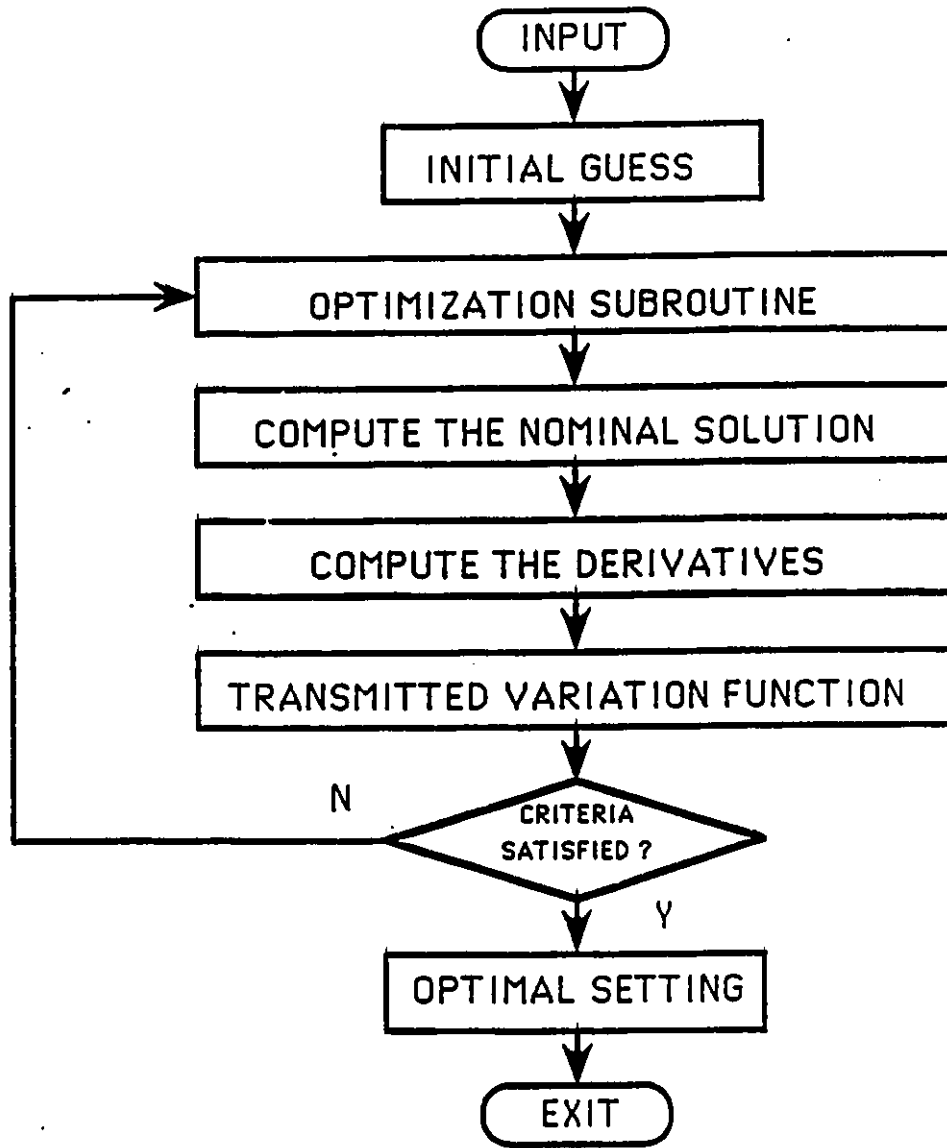


Figure 5.1: The program flowchart for Error Transmission Analysis

Table 5.1: Optimal settings using Error Transmission Analysis with two design variables.

error level (%)	temperature (K)	volume (l)
1	300.0	90.0
2	300.0	90.0
3	300.0	90.0
4	300.0	87.1
5	300.0	87.2
6	300.0	87.2
7	300.0	87.2
8	300.0	87.5
9	300.0	87.0
10	300.0	87.8

Table 5.2: Optimal settings using Error Transmission Analysis with five design variables.

error level (%)	inlet concentration of A (mol/m ³)	inlet concentration of B (mol/m ³)	inlet temperature (K)	temperature (K)	volume (l)
1	4131.3	233.8	328.4	327.8	85.2
2	4319.8	286.4	329.9	315.7	78.5
3	4353.4	343.9	330.0	328.3	74.3
4	4374.2	195.7	330.0	325.9	79.9
5	4775.7	289.0	330.8	310.5	89.7
6	4768.1	307.9	326.7	309.0	88.5
7	2455.2	330.2	329.8	329.6	86.9
8	3819.9	208.4	328.2	319.6	90.0
9	3834.2	290.9	328.4	321.1	81.1
10	4837.8	279.5	329.2	306.6	86.5

Chapter 6

Taguchi Method

Orthogonal arrays were constructed for the design and error variables. The signal-to-noise ratios were computed as a measure of performance. They are maximized by using the analysis of marginal means.

6.1 Methodology

The method consisted of: (i) computation of the nominal solution, (ii) solution of the system equations, (iii) computation of the performance and (iv) selection of the optimal setting.

The flowchart program is shown in Figure 6.1.

6.1.1 Preliminary Steps

The computation of the nominal design and the solution of system equations was discussed earlier in sections 4.2.2 and 4.2.4.

6.1.2 Computation of the Performance Statistic

For each vector in the inner array, a set of error vectors is selected from the outer array and the values for all the variables were determined by solving a nonlinear equation using ZREAL. Once the outer array was exhausted the corresponding signal-to-noise ratio is computed using

$$SN_T = \log\left[\frac{\mu^2}{\sigma_y^2} - \frac{1}{n}\right] \quad (6.1)$$

where y is the production rate and n the size of the outer array. Two levels were studied, an inner array of three levels and five levels. An alternative to the use of marginal averages was examined. Both the sum squares of errors and the signal-to-noise ratio were fitted with a quadratic model.

The inner and the outer arrays employed are listed in Appendix C.

6.1.3 Selection of the Optimal Setting

The optimal setting corresponds to setting of design variable levels that has the maximum marginal mean for the signal-to-noise ratio.

6.2 Results and Discussion

Figure 6.2 shows that the signal-to-noise ratio of Taguchi Method does not vary much with the error level for all error variables. This would imply that the SN ratio is not an appropriate measure of variance. The effect of the error level for the error variables on the mean and the variance of the logarithm of the production rate was examined. The mean was found to decrease with the

error level, and the variance increases as the error level increases (see Figures 6.3 and 6.4). To investigate any correlation between the variance and the mean of the production rate, the mean was plotted against the variance (see Figure 6.5). Unlike what the method assumes, the mean and the variance were shown to be highly correlated when logarithmic transformation is performed. On the other hand, Taguchi assumes equal probability for different levels of error variables, which can explain the inefficiency or poor performance of Taguchi Method.

The number of levels used in the orthogonal arrays was observed to affect substantially the system output variance. In fact, using 5 levels yielded a different setting of design variables and a lower variance for both normally and log-normally distributed errors (see Figures 6.6 and 6.7, respectively).

To avoid the use of marginal averages, the sum squares of errors and the signal-to-noise ratio were fitted to a quadratic model in terms of the design variables and then constrained optimization was performed to yield the optimal setting. The results are presented in Tables 6.1 and 6.2. For both cases, the variance was reduced compared to the original Taguchi Method. However, the incorporation of the sum squares of errors yielded a slightly better performance (see Figure 6.6).

6.3 Conclusions

The following conclusions can be made:

- due to the inappropriate logarithmic transformation and the use of marginal means, the signal-to-noise ratio was not an efficient measure for the variance of the production rate;
- unlike what the method assumes, no adjustment variables have been identified;
- the accuracy of the Taguchi method was found to be sensitive to the number of levels employed, which is due to the use of marginal means;
- the method was found to perform better by modelling the sum squares of errors rather than the signal-to-noise ratio.

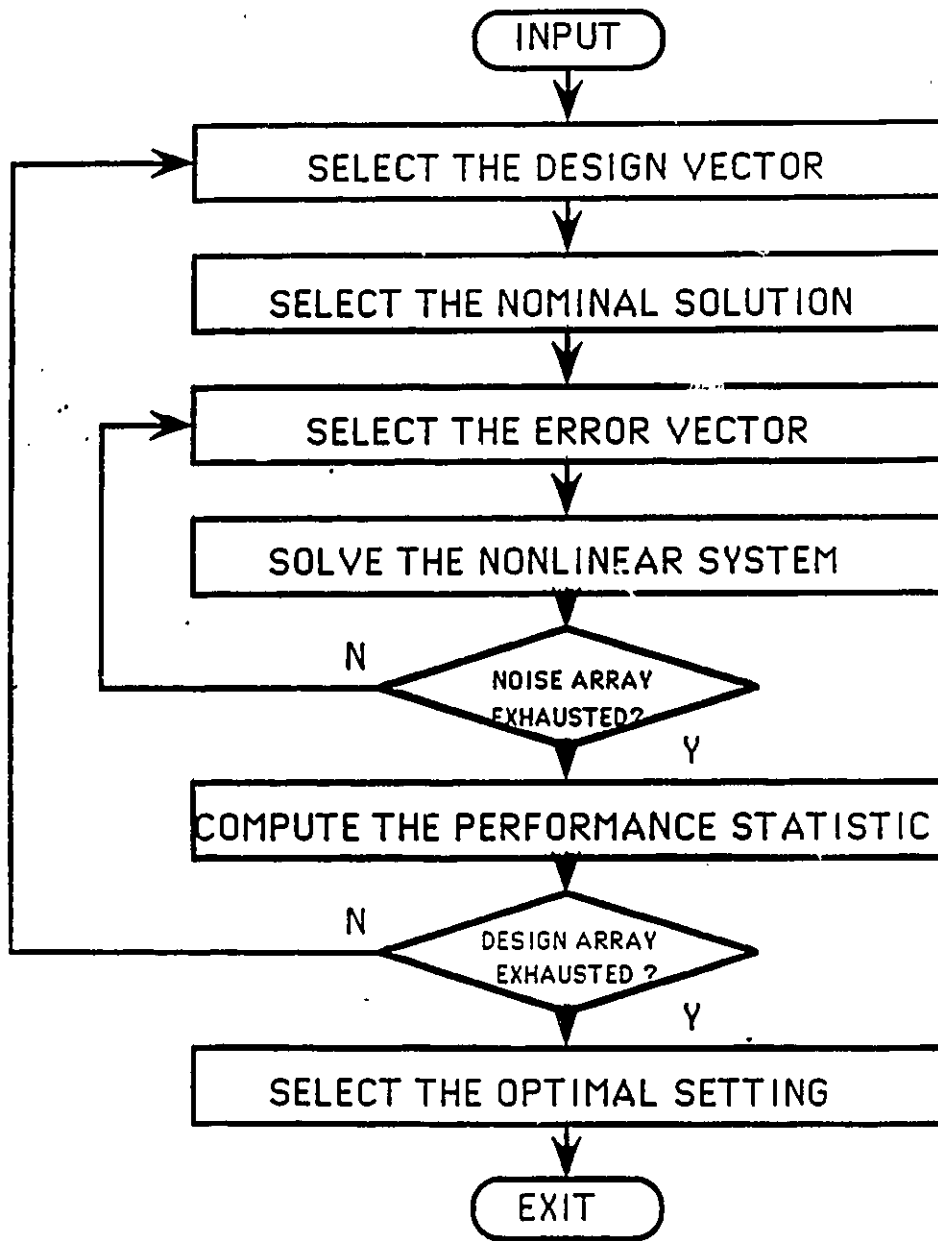


Figure 6.1: The program flowchart for Taguchi Method

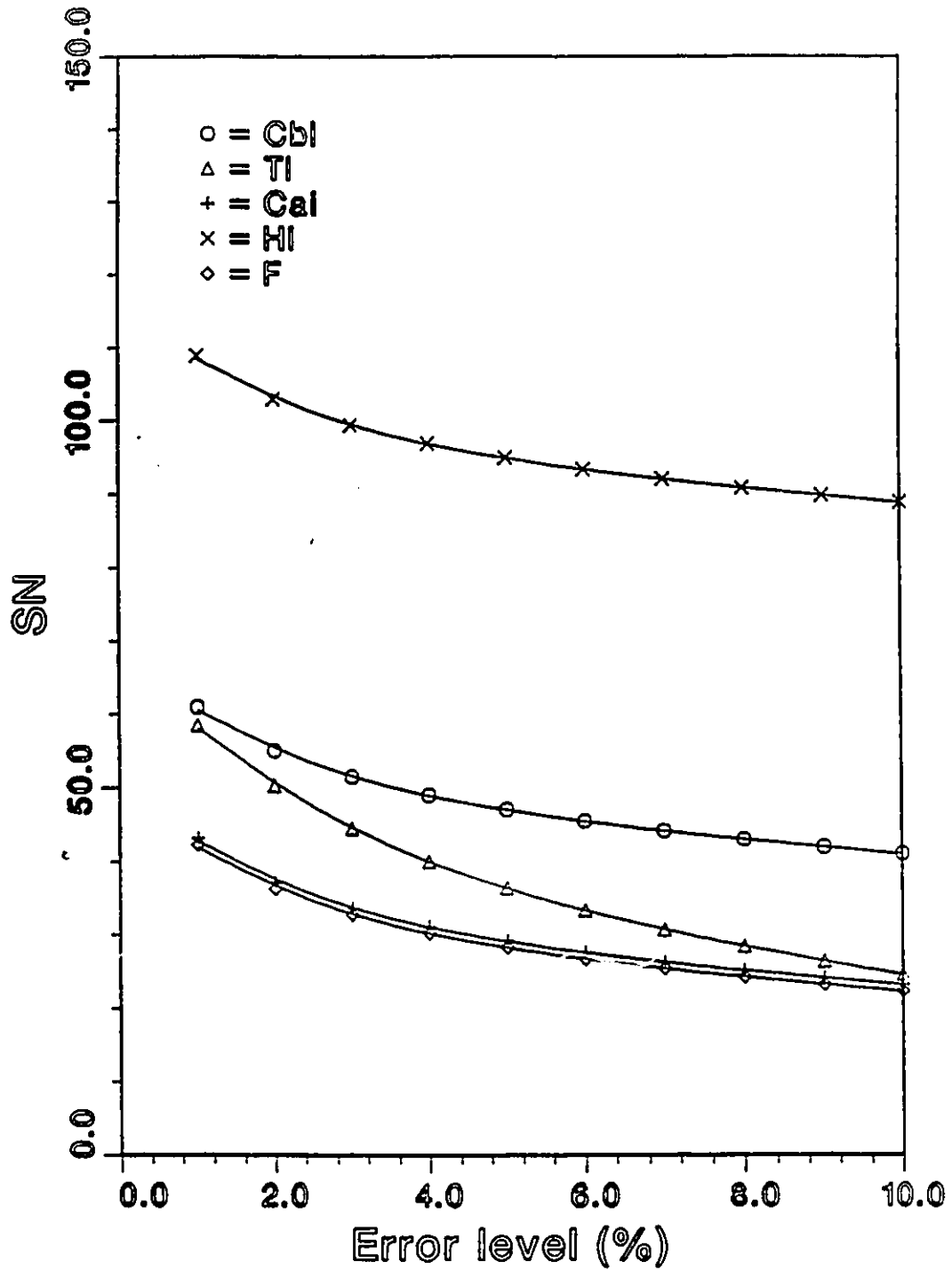


Figure 6.2: The effect of error level on the signal-to-noise ratio for the various error variables. Error level= $[(\text{Std. Dev. } 100\%)/\text{Nominal Value}]$.

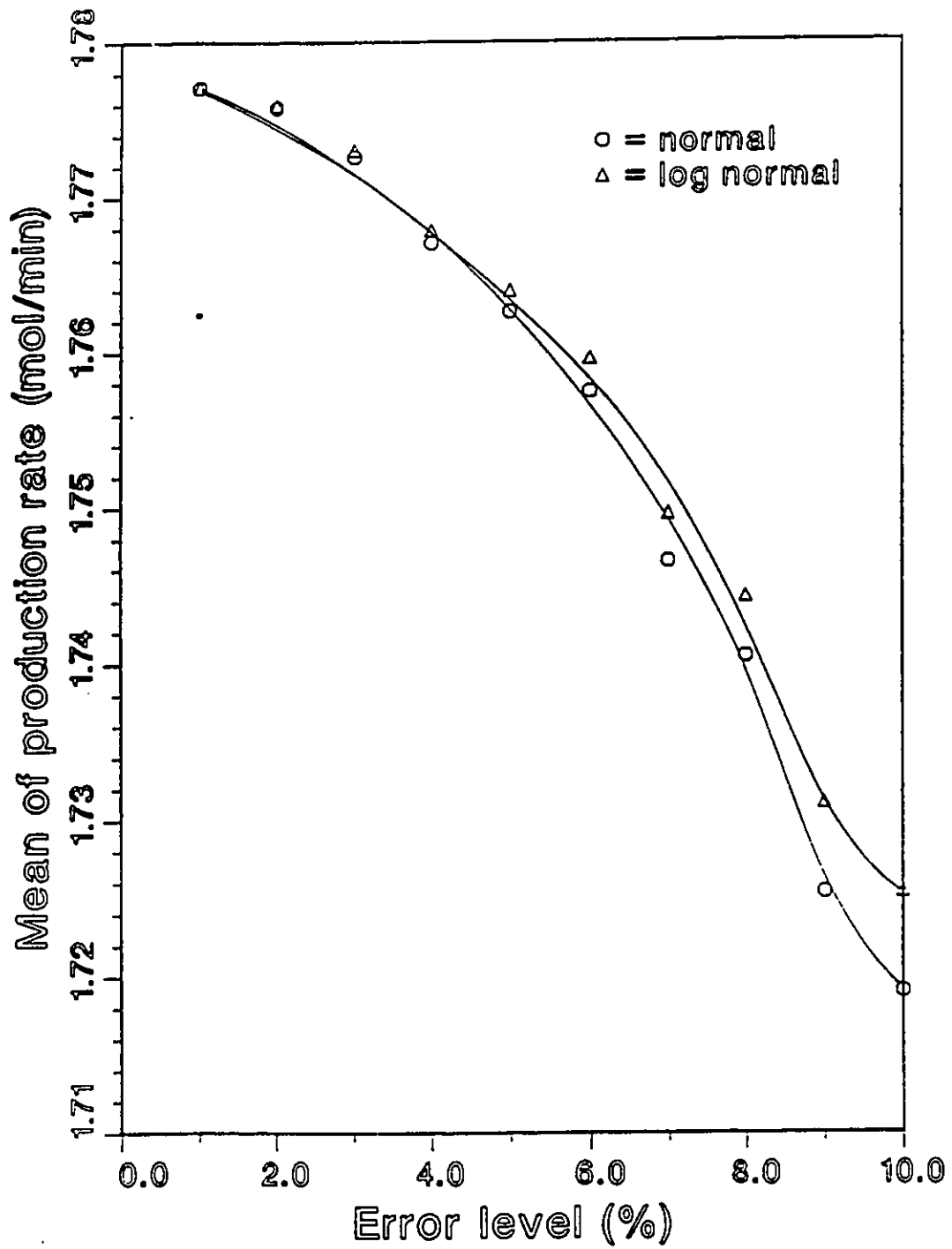


Figure 6.3: The effect of the error level on the mean of the logarithm of the production rate using normally and log-normally distributed errors. Error level= $[(\text{Std. Dev. } 100\%)/\text{Nominal Value}]$.

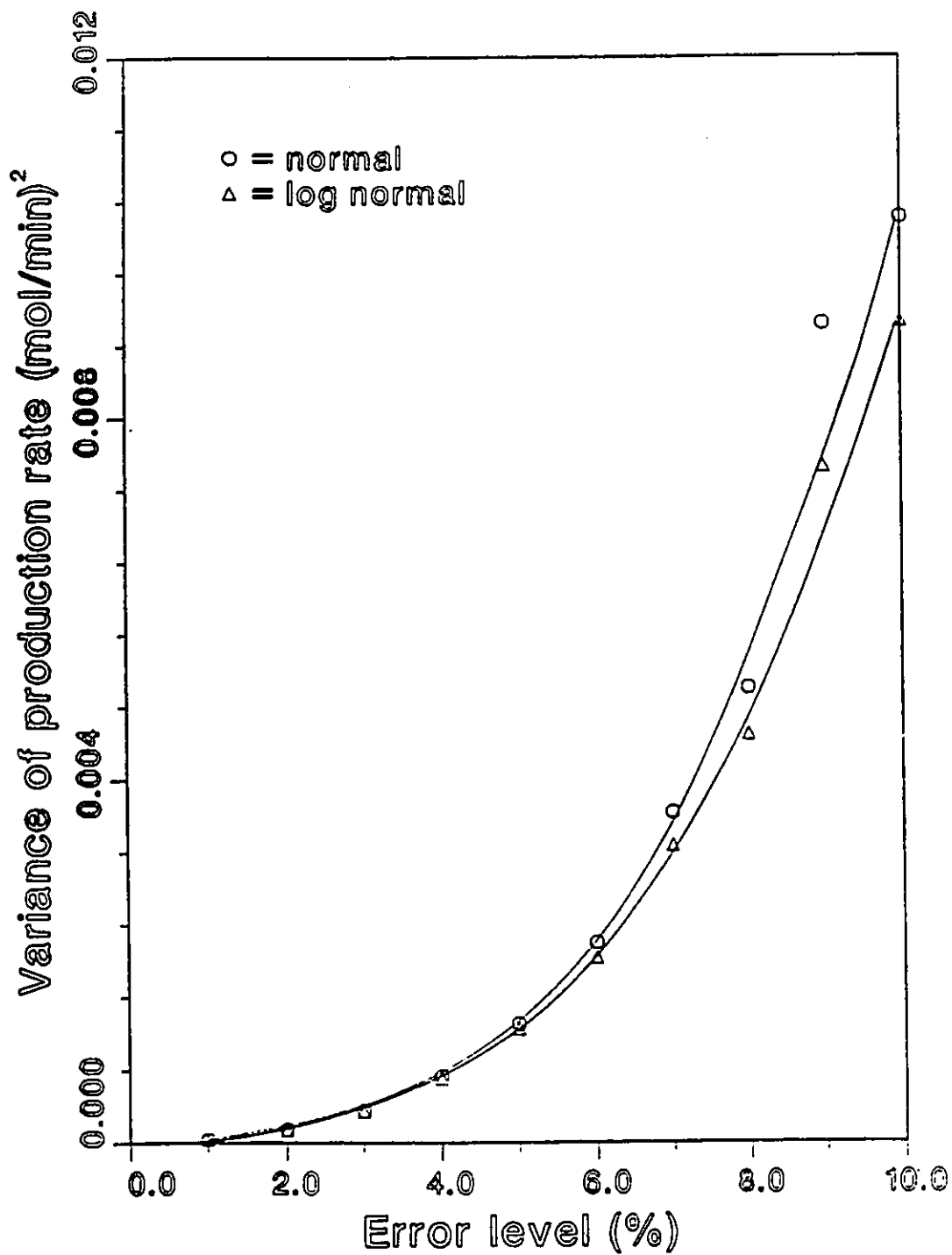


Figure 6.4: The effect of the error level on the variance of the logarithm of the production rate using normally and log-normally distributed errors. Error level= $[(\text{Std. Dev. } 100\%)/\text{Nominal Value}]$.

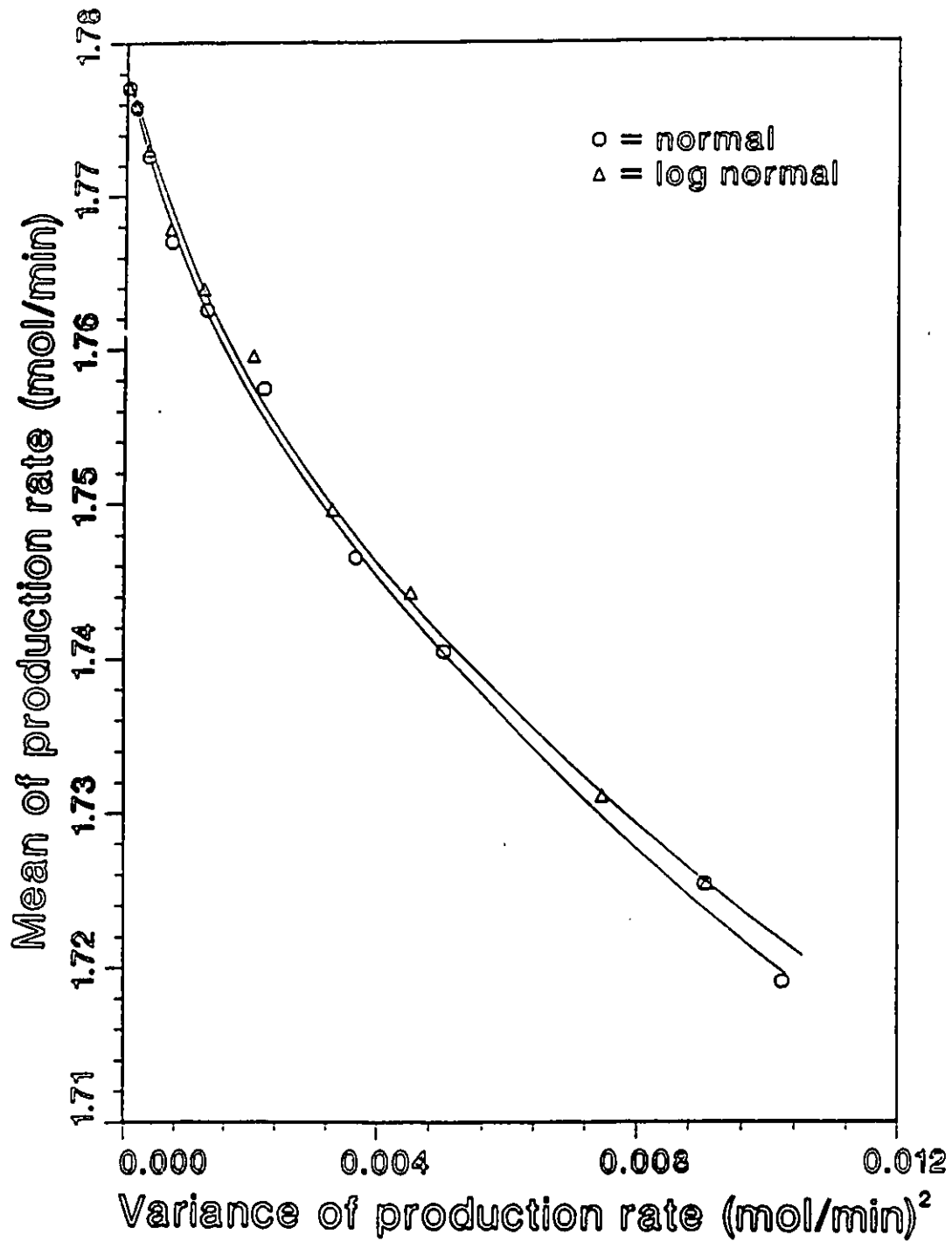


Figure 6.5: Correlation between the variance and the mean of the logarithm of the production rate using normally and log-normally distributed errors. Error level= $[(\text{Std. Dev. } 100\%)/\text{Nominal Value}]$.

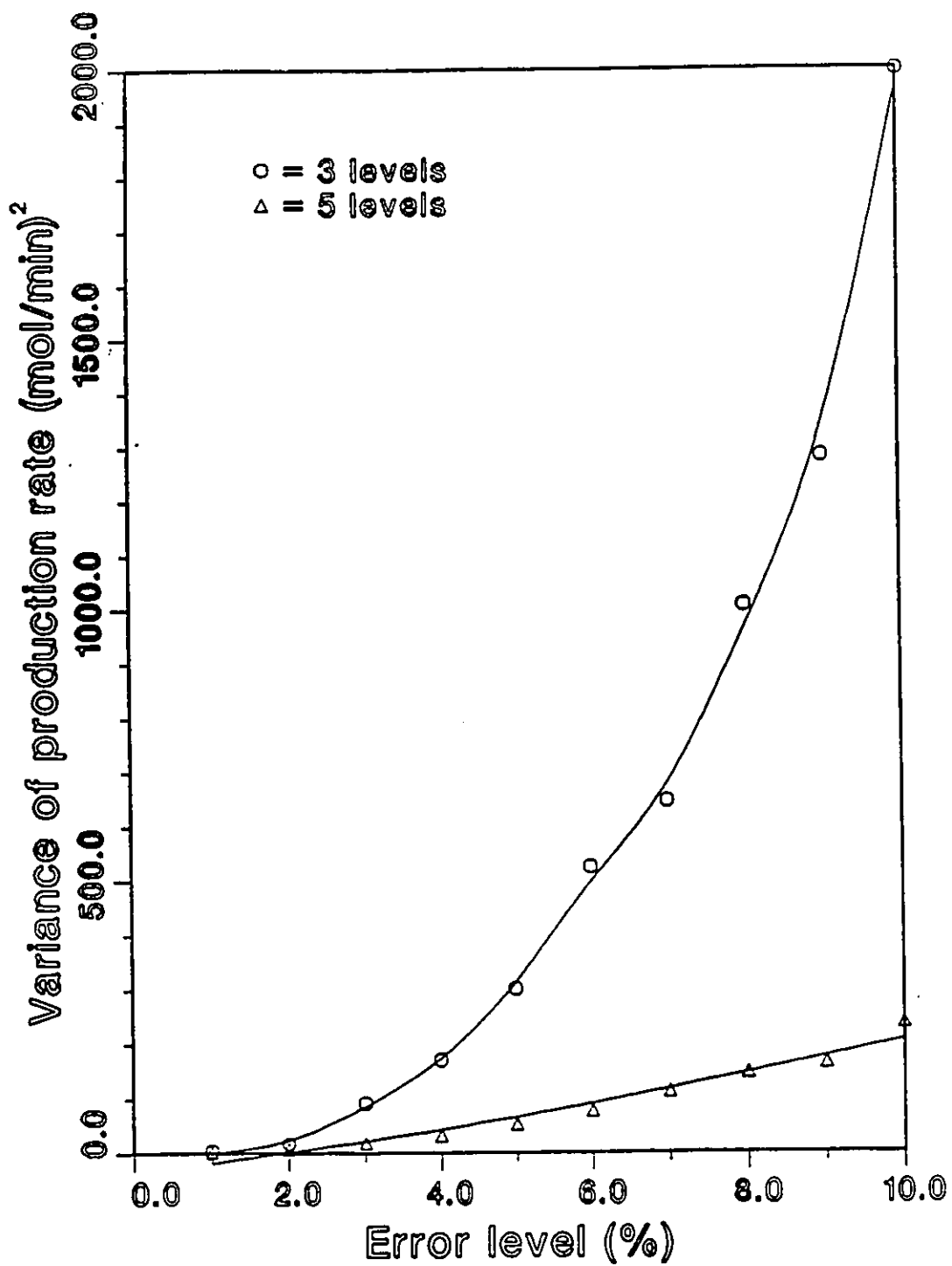


Figure 6.6: The effect of the number of levels on the efficiency of Taguchi method using normally distributed errors. Error level= $\{(\text{Std. Dev. } 100\%)/\text{Nominal Value}\}$.

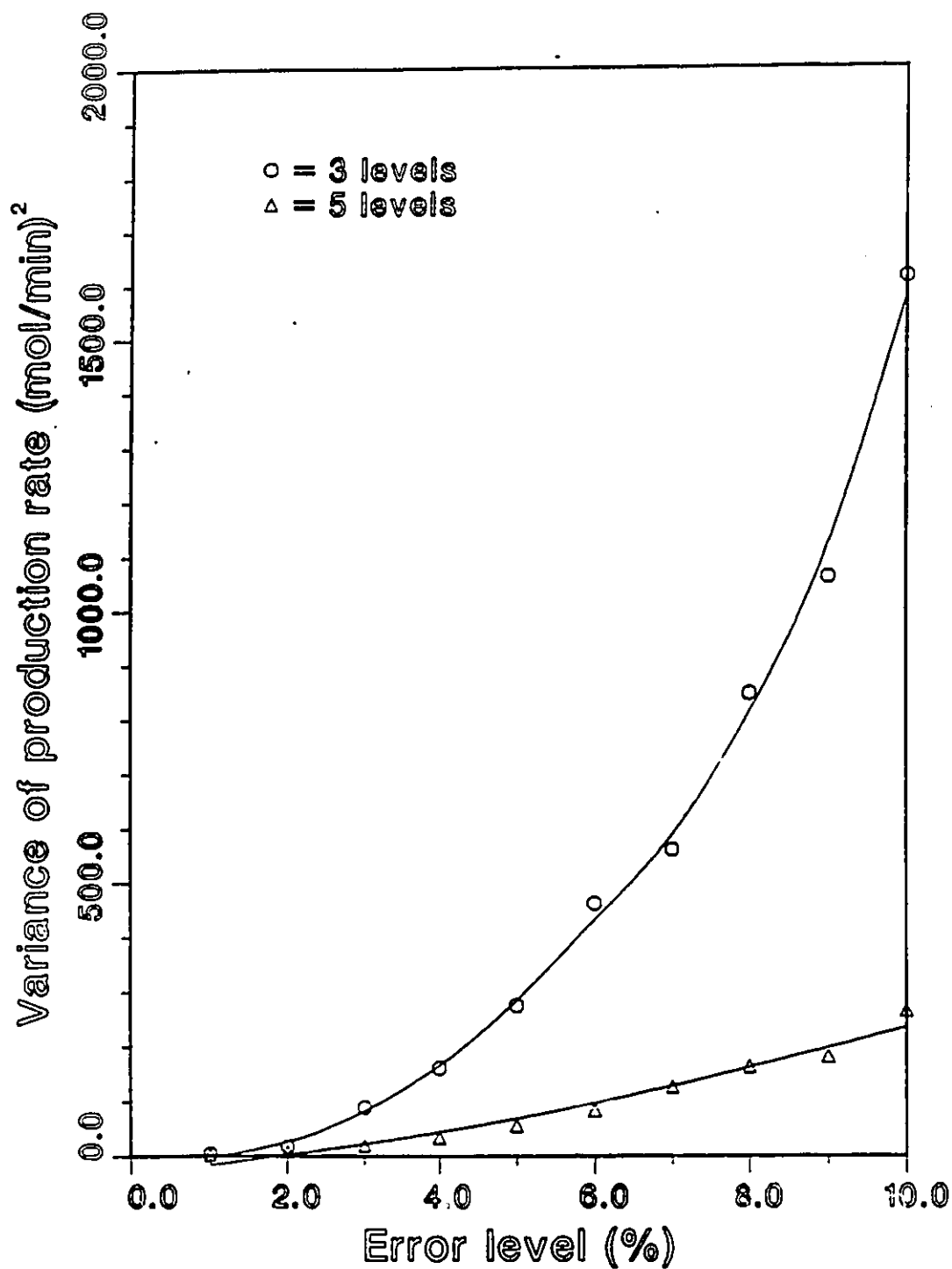


Figure 6.7: The effect of the number of levels on the efficiency of Taguchi method using log-normally distributed errors. Error level=[(Std. Dev. 100%)/Nominal Value].

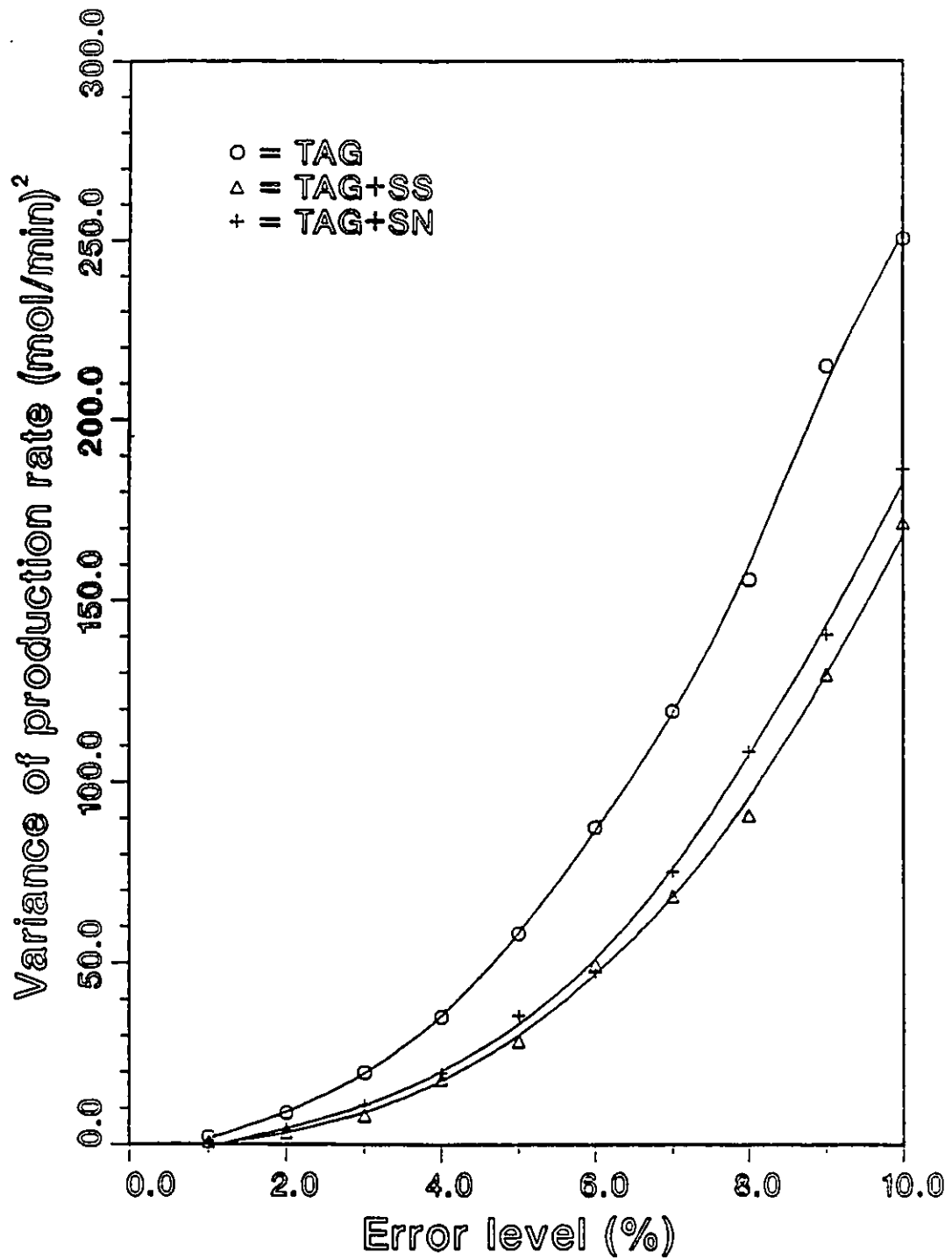


Figure 6.8: The effect of modelling the sum squares of errors and the signal-to-noise ratio on variance reduction (two design variables and normally distributed errors are considered). Error level= $\{(\text{Std. Dev. } 100\%)/\text{Nominal Value}\}$.

Table 6.1: Optimal settings using a quadratic model for the sum squares of errors with five design variables at five levels each.

error level (%)	inlet concentration of A (mol/m ³)	inlet concentration of B (mol/m ³)	inlet temperature (K)	temperature (K)	volume (l)
1	4645.9	482.9	330.0	328.8	83.6
2	1000.0	500.0	330.0	325.1	90.0
3	1131.8	221.0	330.0	325.4	90.0
4	5000.0	500.0	330.0	330.0	90.0
5	1000.0	500.0	330.0	323.4	70.6
6	1000.0	120.4	330.0	330.0	71.7
7	4753.7	447.1	330.0	328.3	89.9
8	2089.3	100.0	298.9	330.0	85.4
9	1009.4	333.3	329.5	326.8	86.8
10	5000.0	482.6	330.0	322.4	90.0

Table 6.2: Optimal settings using a quadratic model for the signal-to-noise ratio with five design variables at five levels each.

error level (%)	inlet concentration of A (mol/m ³)	inlet concentration of B (mol/m ³)	inlet temperature (K)	temperature (K)	volume (l)
1	1000.3	100.3	290.0	290.0	87.4
2	1000.0	100.3	290.0	330.0	10.1
3	1000.0	500.0	330.0	290.0	63.5
4	1031.7	100.0	291.2	291.4	19.0
5	1000.0	100.0	290.0	330.0	10.0
6	1000.0	500.0	290.0	290.0	90.0
7	1000.0	500.0	330.0	290.0	61.3
8	1000.0	395.6	290.0	290.7	90.0
9	1000.0	100.0	290.0	330.0	10.0
10	1000.0	500.0	330.0	290.0	53.3

Chapter 7

Sequential Elimination of Levels

The method employs the analysis of marginal means and Taguchi's signal-to-noise ratios, and tries to reduce the number of runs by eliminating the levels associated with the worst marginal mean.

7.1 Methodology

The different steps involved were: (i) computation of the nominal solution, (ii) solving the system equations, (iii) selection of the worst levels and (iv) determination of the optimal setting.

The program flowchart is shown in Figure 7.1.

7.1.1 Preliminary Steps

The computation of the nominal solution, the solution of system equations and the computation of performance statistic were already discussed in sections 4.2.2, 4.2.4 and 5.2.2. It is important to mention that the orthogonal arrays employed were of smaller size because of the successive application of the method. They are shown in Appendix C.

7.1.2 Elimination of the Worst Levels

The design variable level corresponding to the worst marginal mean for the signal-to-noise ratio was eliminated. Another orthogonal array of smaller size is constructed with the remaining levels.

7.1.3 Selection of the Optimal Setting

When the number of levels was reduced to 2, the optimal level for each design variable is the one that has the best performance.

7.2 Results and Discussion

To overcome the problem of widely spaced levels, the Sequential Elimination of Levels was considered. The performance of this latter technique was observed to be much better than Taguchi Method for the case of two design variables. In the case of five design variables, the method did not perform better than Taguchi method. This is due to the smaller size of the orthogonal arrays employed. To improve convergence to the optimal setting, SEL was applied repeatedly in three cycles. The total number of runs was kept the same as that of Taguchi Method.

While no improvement was observed for the case of two design variables (Figures 7.2 and 7.3), The variance was slightly reduced when five design variables were considered (Figures 7.4 and 7.5). Occasionally, the variance was observed to decrease with an increase in error level, which is obviously inconsistent with the result reported earlier. The use of orthogonal array and marginal averages

explains such an inconsistency.

Using the minimum value for the signal-to-noise ratio as a performance criterion was also examined. No major changes in the optimal settings of the design variables were observed, and the performance of these optimal settings was still poor.

7.3 Conclusions

One can conclude that:

- by eliminating the worst levels at each set of orthogonal arrays for the design variables, Sequential Elimination of Levels reduces the total number of runs;
- the method may be misleading because of the relative small size of the orthogonal arrays employed;
- successive application of the method to smaller design region may lead to a setting of design variables that gives a lower production rate variance;
- even with the successive application of the method, the results were still unreliable.

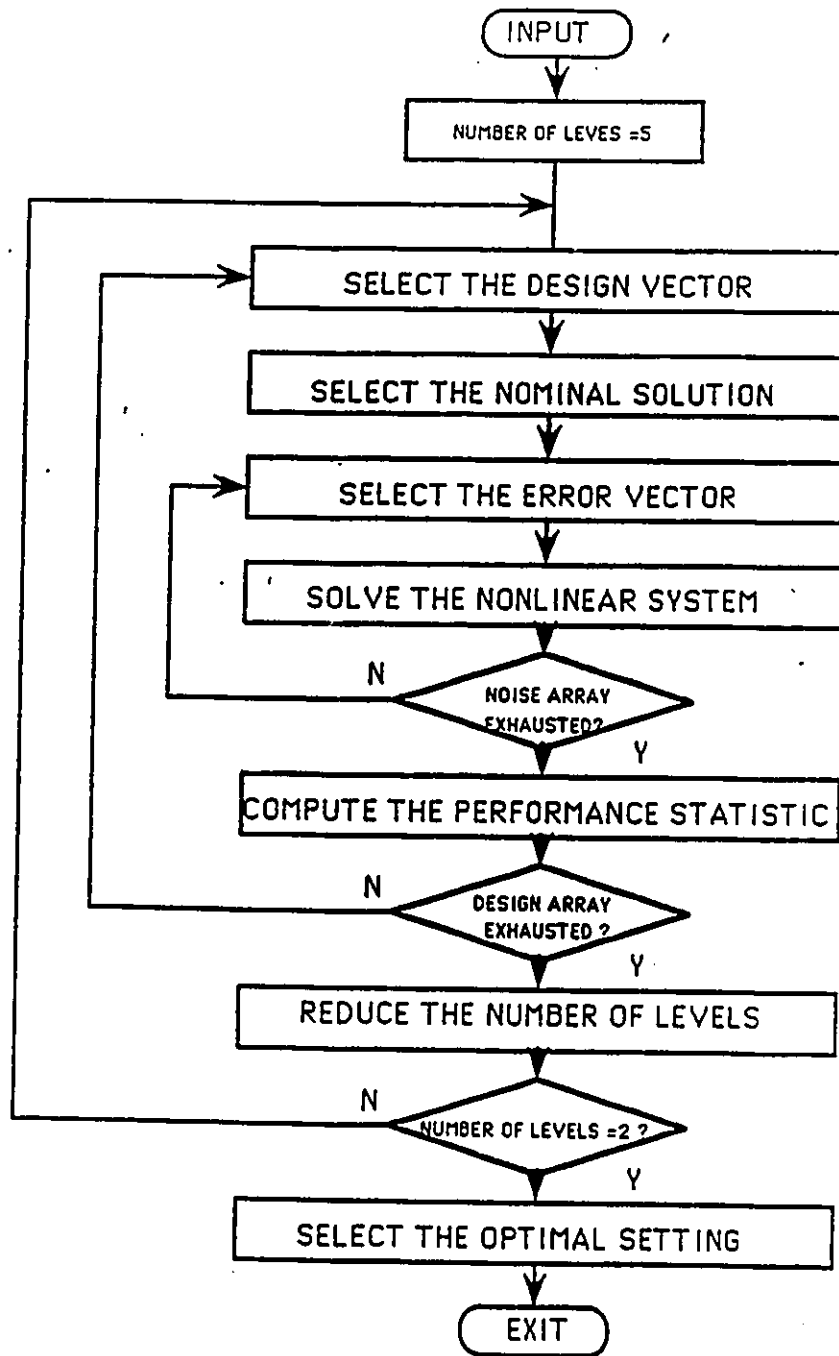


Figure 7.1: The program flowchart for Sequential Elimination of Levels

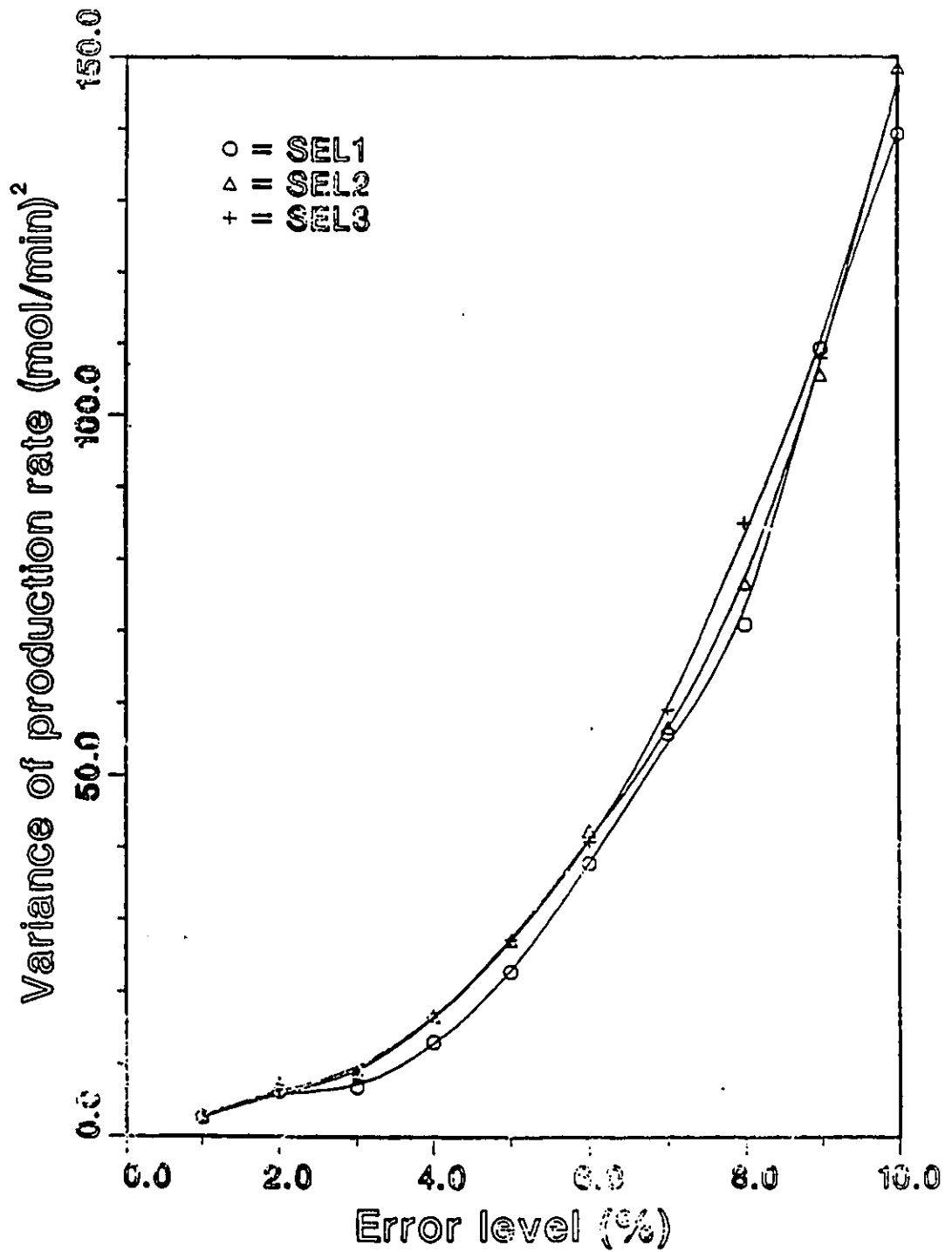


Figure 7.2: The effect of the successive use of the method of Sequential Elimination of Levels in improving performance (two design variables and normally distributed errors). Error level= $[(\text{Std. Dev. } 100\%)/\text{Nominal Value}]$.

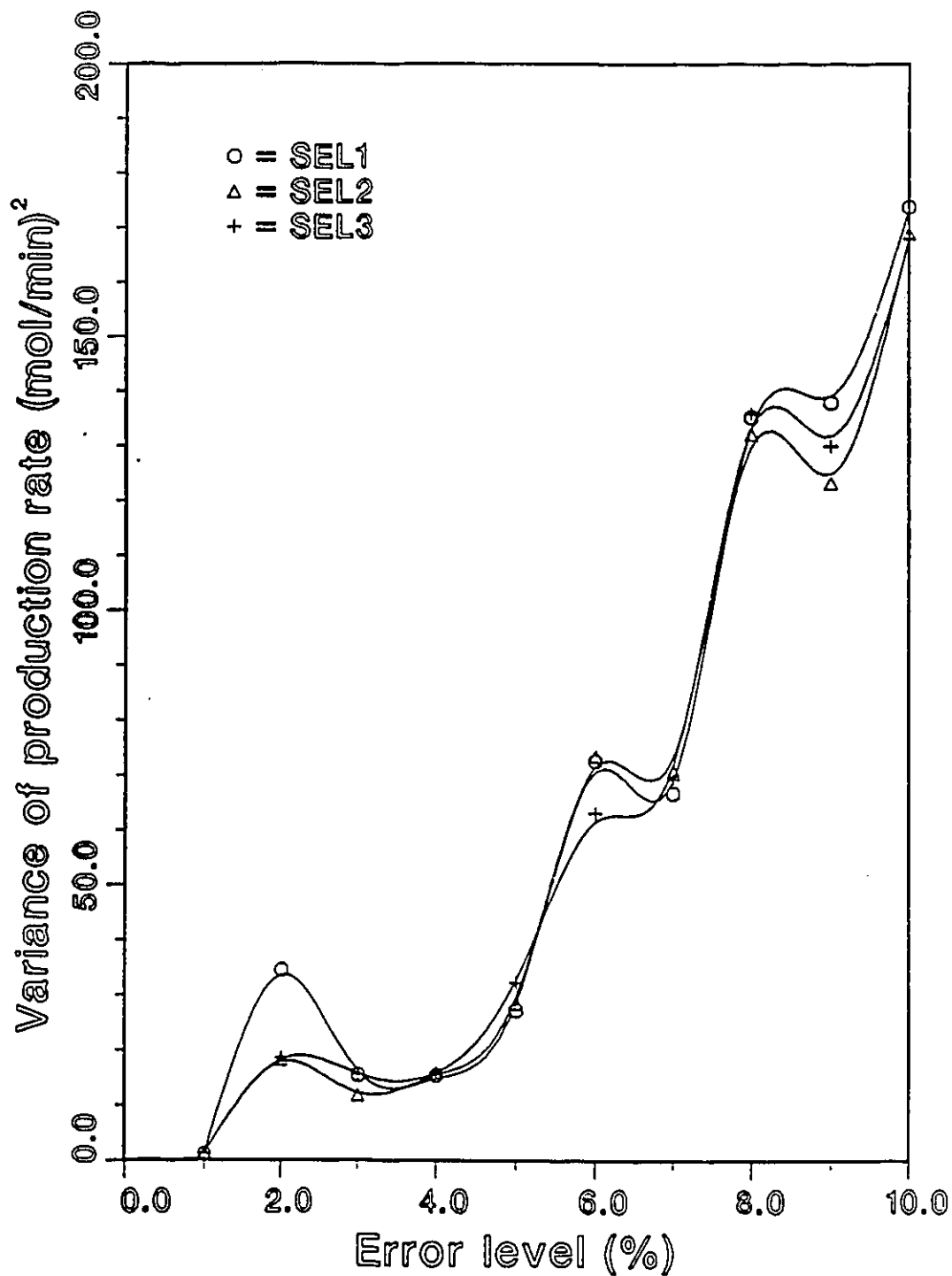


Figure 7.4: The effect of the successive use of the method of Sequential Elimination of Levels in improving performance (five design variables and normally distributed errors). Error level= $[(\text{Std. Dev. } 100\%)/\text{Nominal Value}]$.

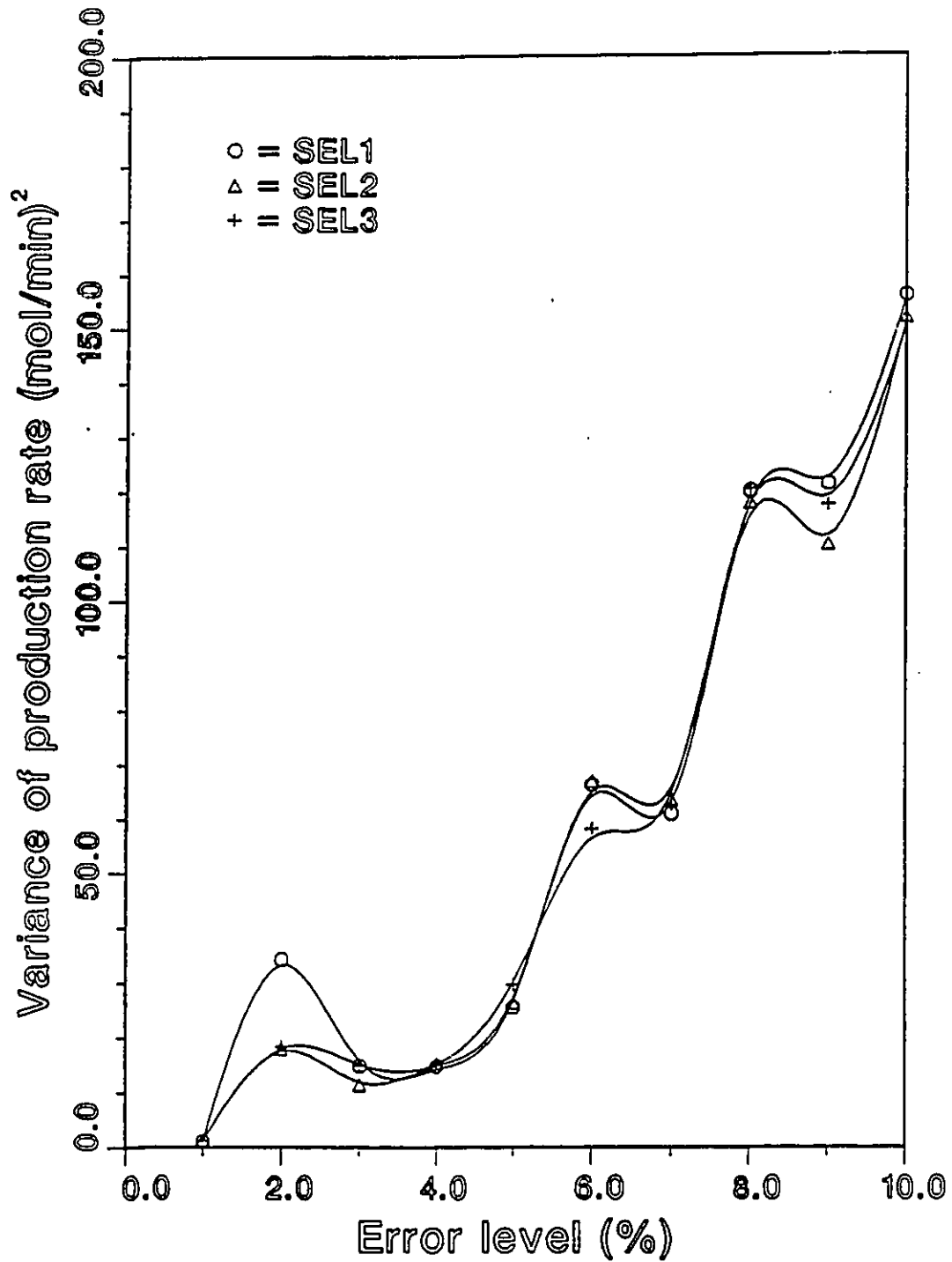


Figure 7.5: The effect of the successive use of the method of Sequential Elimination of Levels in improving performance (five design variables and normally distributed errors). Error level= $[(\text{Std. Dev. } 100\%)/\text{Nominal Value}]$.

Table 7.1: Optimal settings using Sequential Elimination of Levels with five design variables at five levels each and a total of 16 runs.

error level (%)	inlet concentration of A (mol/m^3)	inlet concentration of B (mol/m^3)	inlet temperature (K)	temperature (K)	volume (l)
1	3000.0	400.0	310.0	330.0	70.0
2	3000.0	100.0	290.0	310.0	50.0
3	5000.0	200.0	290.0	310.0	50.0
4	5000.0	200.0	290.0	310.0	70.0
5	5000.0	200.0	290.0	310.0	70.0
6	5000.0	200.0	290.0	310.0	50.0
7	5000.0	200.0	290.0	310.0	70.0
8	5000.0	200.0	290.0	310.0	50.0
9	5000.0	200.0	290.0	310.0	70.0
10	5000.0	200.0	290.0	310.0	70.0

Table 7.2: Optimal settings using Sequential Elimination of Levels with five design variables at five levels each and a total of 32 runs.

error level (%)	inlet concentration of A (mol/m^3)	inlet concentration of B (mol/m^3)	inlet temperature (K)	temperature (K)	volume (l)
1	2200.0	320.0	318.0	328.0	70.0
2	3400.0	100.0	292.0	308.0	50.0
3	5000.0	200.0	292.0	310.0	50.0
4	4800.0	240.0	292.0	310.0	70.0
5	4600.0	160.0	292.0	310.0	70.0
6	4800.0	120.0	292.0	302.0	50.0
7	4600.0	160.0	292.0	310.0	70.0
8	4800.0	120.0	292.0	302.0	50.0
9	4600.0	280.0	292.0	310.0	70.0
10	4600.0	280.0	292.0	310.0	70.0

Table 7.3: Optimal settings using Sequential Elimination of Levels with five design variables at five levels each and a total of 48 runs.

error level (%)	inlet concentration of A (mol/m ³)	inlet concentration of B (mol/m ³)	inlet temperature (K)	temperature (K)	volume (l)
1	2300.0	320.0	317.0	328.0	70.0
2	3400.0	120.0	292.0	308.0	50.0
3	4900.0	200.0	292.0	312.0	50.0
4	4800.0	240.0	291.0	312.0	70.0
5	4600.0	170.0	291.0	312.0	70.0
6	4800.0	130.0	291.0	302.0	50.0
7	4600.0	160.0	291.0	312.0	70.0
8	4800.0	120.0	292.0	302.0	50.0
9	4600.0	290.0	292.0	310.0	70.0
10	4500.0	280.0	292.0	312.0	70.0

Chapter 8

Comparison of the Different Methodologies

The variances of the production rate for the optimal designs generated using the five different Parameter Design techniques are compared in Figures 9.1 to 9.4. The variances in each case were determined using Monte Carlo simulations with a sample size of 1600. The coupling of nonlinear optimization with Monte Carlo simulations provides the most rigorous means of determining the variance of the production rate. Consequently, the results obtained using this approach represented the optimal results. Although the method is rigorous, as a result of the necessary large sample size it is computationally demanding. The incorporation of Response Surface Methodology with Monte Carlo simulations resulted in a reduced number of runs, without changing much the variance of the production rate for the optimal setting. The number of simulations involved at each design point was reduced compared to the original Monte Carlo simulations combined with a

multivariable optimization routine. The worst scenario occurred using Error Transmission Analysis because of the deficiency of the first-order approximation for the transmitted variance function. This approximation did not describe the nonlinearity of the system studied. The Error transmission Analysis performed even worse in the case of five design variables as can be seen in Figures 9.3 and 9.4. Variances of production rate produced using the Taguchi Method were observed to be much higher than those generated using Monte Carlo simulations. This is due to several factors. First, the signal-to-noise ratio was not an efficient measure of the variance of the production rate. Second, the logarithmic transformation in order to uncorrelate the the location and dispersion effects was not appropriate. Finally, the use of the analysis of marginal means was not justified. The use of Sequential Elimination of Levels decreased the number of runs required and resulted in much better performance compared to Taguchi Method for the case of two design variables. This shows once again the effect of using marginal mean in reducing the efficiency of the latter method.

To compare the performance of the parameter design method to that of a classical design technique, four of the optimal design variables obtained by Monte Carlo simulations were kept the same. The reactor volume was computed by the classical design constraint mentioned in Appendix A. Based on Monte Carlo simulations, the variances were

computed for both designs and their ratio, called "performance ratio", was plotted against the error level for both normally and log-normally distributed errors (Figures 9.5 and 9.6). The performance ratio was observed to be greater than 1 under all circumstances. It was also shown to increase with an increase in error level, especially for log-normally distributed error condition, where the performance was much better than for the case of normally distributed errors. Taking an error level of 5% for instance it was shown that using the parameter design method yielded a reduction in the system output variance of 19 and 20% for normally and log-normally distributed errors respectively for an energy activation of 34.6 kJ/mol. To further compare the effectiveness of parameter "parameter design" in minimizing transmission of variation the same analysis was repeated for a system expected to be more sensitive to disturbances. This system was identical to that used in the rest of the work except that the activation energy was increased from 34.6 kJ/mol to 36.6 kJ/mol. The results given in Figure 9.10 show that the performance was better as a reduction in variance of 23 and 33% was observed.

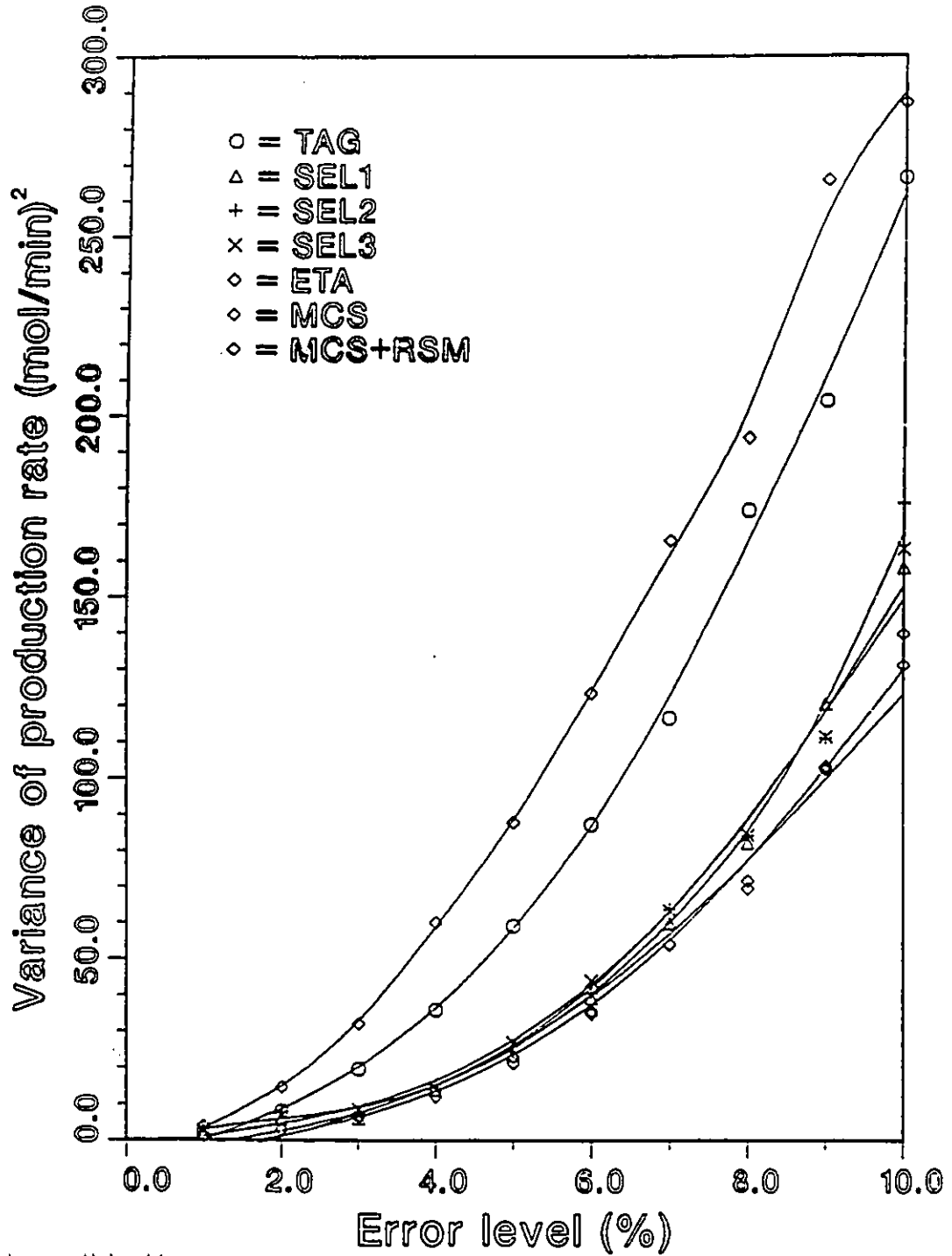


Figure 8.1: Comparison of the different Parameter Design techniques in terms of their minimization of the production rate variance (two design variables and normally distributed errors.) Error level= $[(\text{Std. Dev. } 100\%)/\text{Nominal Value}]$.

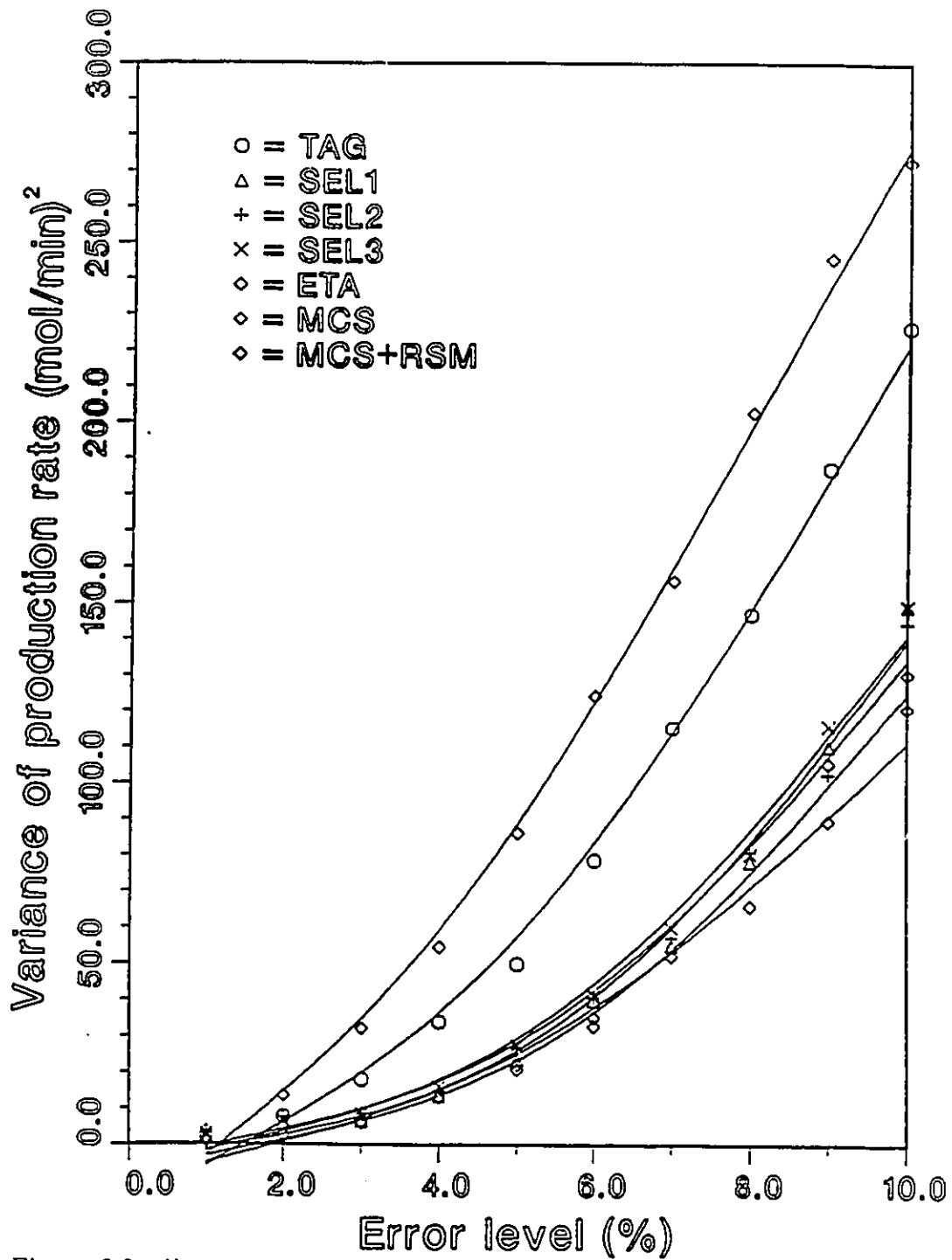


Figure 8.2: Comparison of the different Parameter Design techniques in terms of their minimization of the production rate variance (two design variables and log-normally distributed errors). Error level= $[(\text{Std. Dev. } 100\%)/\text{Nominal Value}]$.

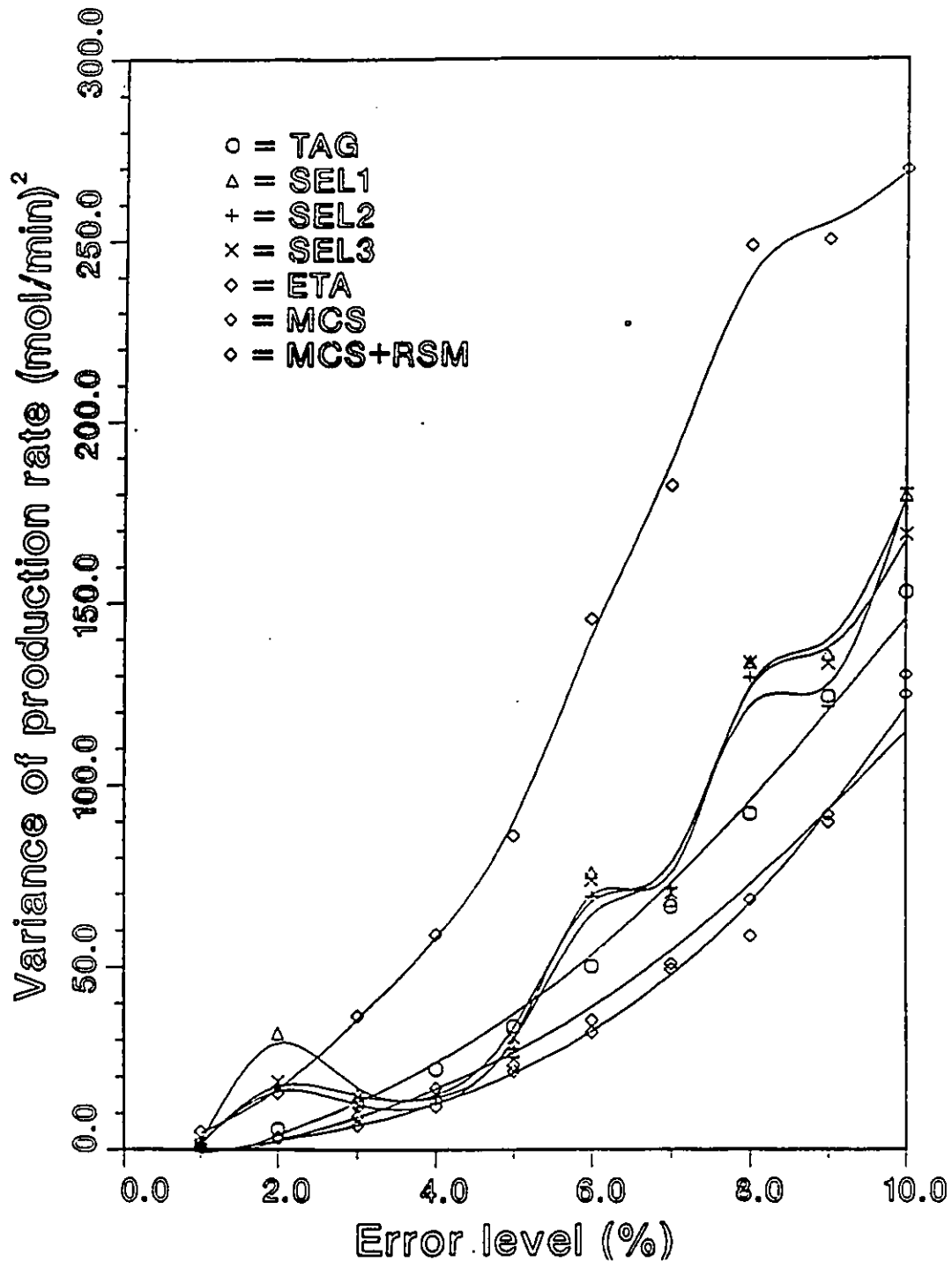


Figure 8.3: Comparison of the different Parameter Design techniques in terms of their minimization of the production rate variance (five design variables and normally distributed errors). Error level= $[(\text{Std. Dev. } 100\%)/\text{Nominal Value}]$.

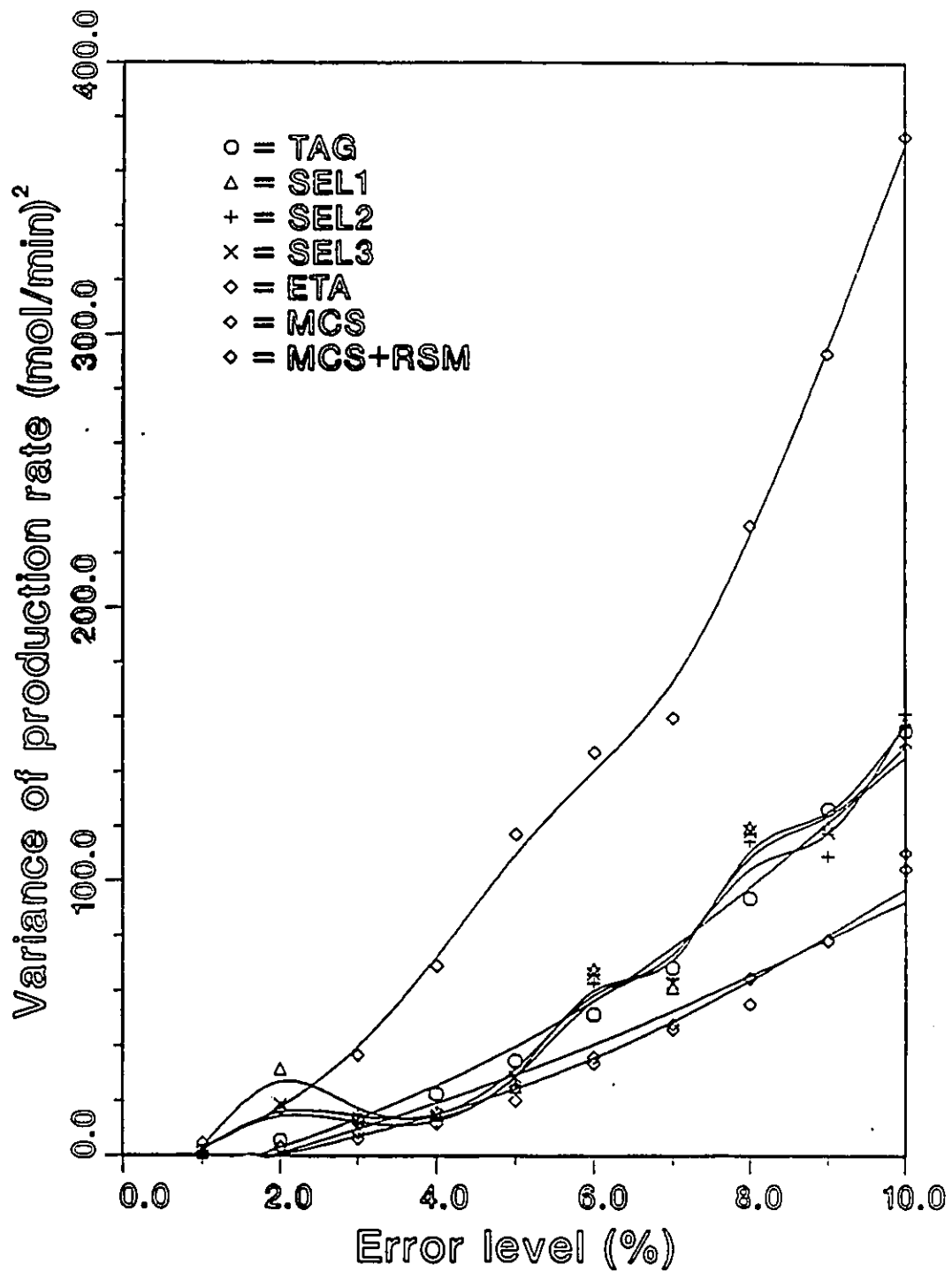


Figure 8.4: Comparison of the different Parameter Design techniques in terms of their minimization of the production rate variance (five design variables and log-normally distributed errors). Error level= $[(\text{Std. Dev. } 100\%)/\text{Nominal Value}]$.

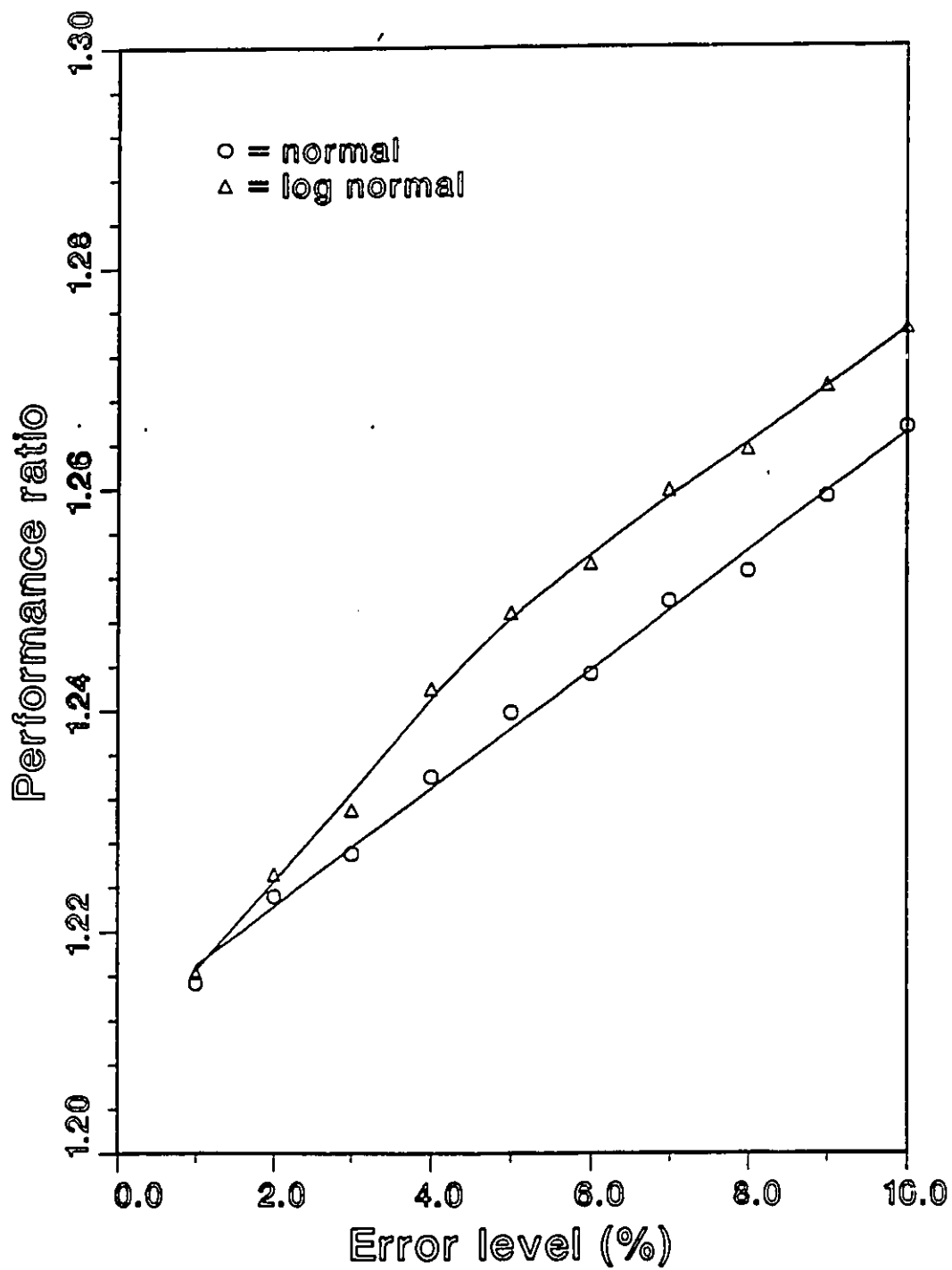


Figure 8.5: Improvement of variance with the use of Parameter Design compared to the Classical Design ($E_B = 34.6$ kJ/mol). Error level= $[(\text{Std. Dev. } 100\%)/\text{Nominal Value}]$.

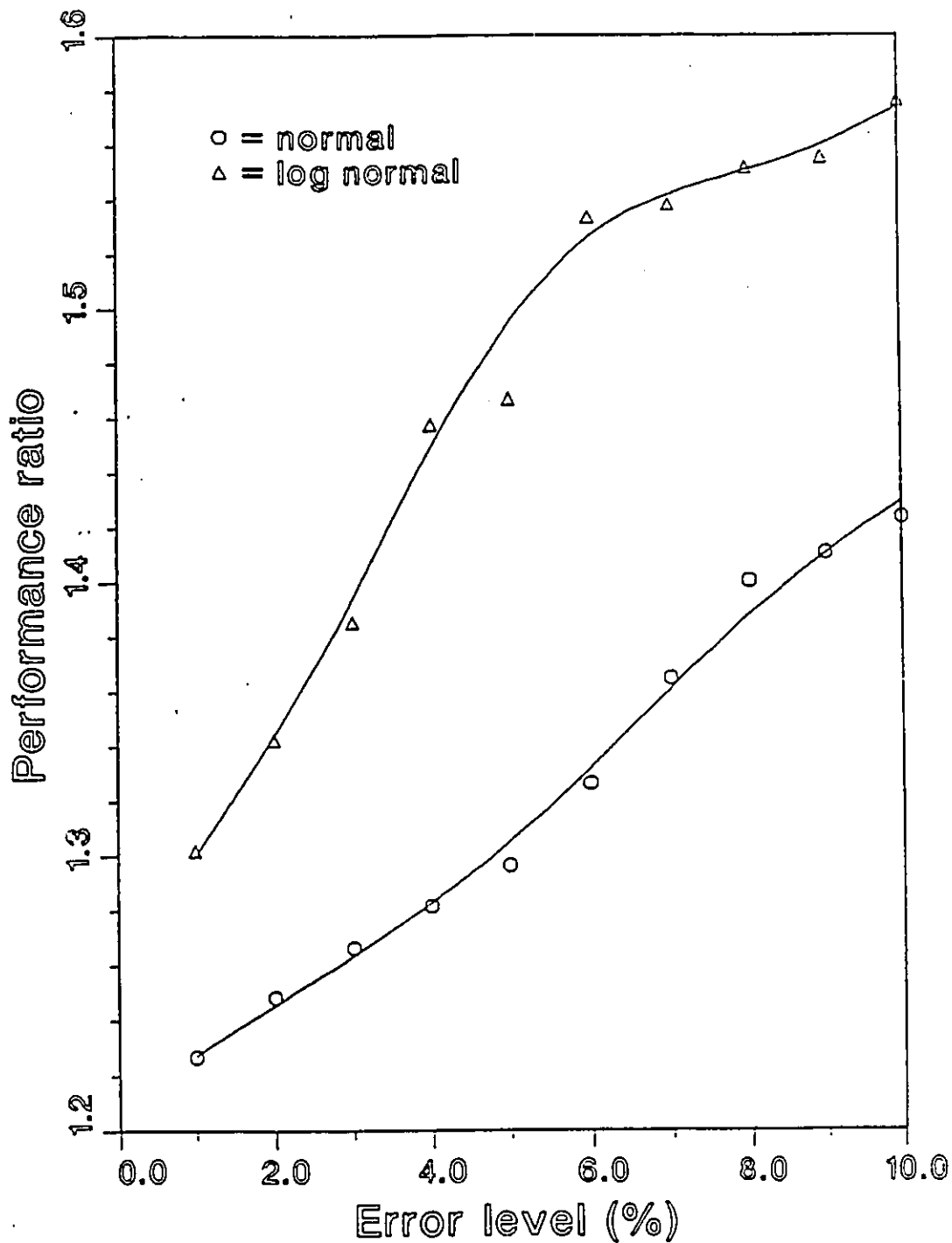


Figure 8.6: Improvement of variance with the use of Parameter Design compared to the Classical Design ($E_B = 36.6$ kJ/mol). Error level= $[(\text{Std. Dev. } 100\%)/\text{Nominal Value}]$.

Chapter 9

Conclusions and Recommendations

9.1 Conclusions

With respect to the critical review and the evaluation of the effectiveness of each approach, the following conclusions can be drawn

- * Response Surface Methodology associated with Monte Carlo simulations was shown to be very efficient. It also resulted in an important design tool, which consists of an empirical model relating the process variability to operating conditions.
- * concerning Error Transmission Analysis, the first order approximation for the transmitted variation function was not sufficient to describe the nonlinearity of the system considered.
- * the direct application of Taguchi Method may be misleading. The logarithmic transformation may be inappropriate and the use of the marginal means is not justified.

- * the Sequential Elimination of Levels was found to be generally unreliable.

With respect to the potential benefits from using Parameter Design it can be concluded that

- * the process performance was observed to depend on the choice of the design variables as well as the error level for the error variables. Hence, the transmission of variation has to be considered at the early stages of the design.

With respect to the compatibility of Parameter Design with other traditional strategies one can conclude that

- * compared to the classical approach, Parameter Design leads to a more robust chemical processing units.

With regard with the applicability of Parameter Design to chemical process industry, there is still much to be done.

9.2 Recommendations

- * Develop performance statistics such as signal-to-noise ratios that consider the appropriate data transformation and include both the mean and the variance of the objective function.
- * The uncertainty of estimating the parameters on which the design is based must be considered.

- * Examine dynamic systems where the process output is very sensitive to error transmission.
- * Quality improvement was achieved through Parameter Design, which implies that the concept of closeness to target rather than variation within specification should be considered at the design stages.
- * In order to eliminate or reduce variable interactions and dispersion effects, empirical methods to identify data transformation have to be adopted.
- * Techniques for estimating reasonable loss function in terms of the process output mean and variability, have to be made available to designers.

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Appendix A

Classical Design Constraint

A classical design objective for the production of an intermediate in a sequence of reactions is to maximize the outlet concentration of the intermediate by operating at the appropriate space time. For the system studied in this thesis the outlet concentration of B is given by

$$C_B = \frac{C_{B_i} + k_A C_A \tau}{1 + k_B \tau}$$

with

$$C_A = \frac{C_{A_i}}{1 + k_A \tau}$$

Then, combining these equations gives

$$C_B = \frac{C_{B_i}(1 + k_A \tau) + k_A C_{A_i} \tau}{(1 + k_A \tau)(1 + k_B \tau)}$$

The optimal space time is obtained when

$$\frac{dC_B}{d\tau} = 0$$

which is equivalent to

$$k_A^2 k_B (C_{A_i} + C_{B_i}) \tau^2 + [2k_A k_B C_{B_i} k_A (C_{A_i} + C_{B_i}) + k_A (k_A + k_B) (C_{A_i} + C_{B_i})] \tau + k_B C_{B_i} - k_A C_{A_i} = 0.$$

Solving this second order equation we obtain the following optimal space time:

$$\tau_{opt} = \frac{-k_A k_B C_{B_i} + \sqrt{k_A^2 k_B^2 C_{B_i}^2 + k_A^2 k_B (C_{A_i} + C_{B_i}) (k_A C_{A_i} - k_B C_{B_i})}}{k_A^2 k_B (C_{A_i} + C_{B_i})}. \quad (\text{A.1})$$

When $C_{B_i} = 0$, the optimal space time becomes

$$\tau_{opt} = \frac{1}{\sqrt{k_A k_B}}. \quad (\text{A.2})$$

Appendix B

IMSL Subroutines

B.1 Optimization Subroutines

B.1.1 NCONF

NCONF is a subroutine that solves a general nonlinear problem using a successive programming algorithm and a finite difference gradient. It is based on a FORTRAN code developed by Schittkowski.

B.1.2 BCONF

The subroutine BCONF minimizes a function of n variables subject to bounds on the variables using a quasi-Newton method and a finite-difference gradient.

B.1.3 BCPOL

The subroutine BCPOL minimizes a function of n variables subject to bounds on the variables using a direct search complex algorithm. The method is based on function comparison without any smoothing. It

starts with $2n$ points. At each iteration a new point is generated to replace the worst point which has the largest function value.

B.2 Simulation Subroutines

B.2.1 GGNML

The subroutine GGNML is a random deviate generator. It consists of generating a pseudo-random normal (0,1) deviates by transforming uniform deviates to normal deviates using the inverse normal routine MDNRIS.

Given the seed DSEED and the number of points N , GGUBS generates N uniform random numbers in the exclusive range (0,1), MDNRIS transforms each of numbers to a normal (0,1) deviate.

B.2.2 GGNLG

The subroutine GGNLG generates log-normal pseudo-random deviates. The algorithm is similar to GGNML.

B.3 Subroutine for Nonlinear Equations

B.3.1 ZREAL

The subroutine computes n real zeros for a real function f . Given f and the initial guess, Muller's method is used to locate the zeros of f .

B.4 Model Fitting Subroutine

B.4.1 RGIVN

The subroutine RGIVN fits a multivariate regression model via fast given transformations.

B.5 Differentiation Subroutine

B.5.1 DERIV

The subroutine DERIV computes the derivative of a user-supplied function. The derivative estimate originates from a centered finite difference approximation, where the step size is adoptively changed to enhance accuracy.

Appendix C

Monte Carlo Simulations

Figures E.1 and E.2, respectively, show the effect of sample size required for Monte Carlo simulations on the variance of the production rate, for normally and log-normally distributed errors. Figures E.3 and E.4 show large fluctuations in the mean value for relatively small sample sizes. This is explained by the fact that a small sample size can hardly represent the population. As the sample size increased, however, the fluctuations were damped out and reasonably stable mean values were obtained for sample sizes exceeding 1600.

To investigate the effect of the seed on the random number generator, the seed was varied from 1 to 10^{21} , the sensitivity of the variance did not exceed 1% under all circumstances (see Figures E.5 and E.6 for normally and log-normally distributed errors, respectively). This showed that the seed had little or no effect on the sample under investigation. This was anticipated because of large sample size employed

for the Monte Carlo simulations.

Histograms for the production rate were drawn in Figures E.7 and E.8 for normally and log-normally distributed errors. The distributions were observed to be similar to that of the input variables.

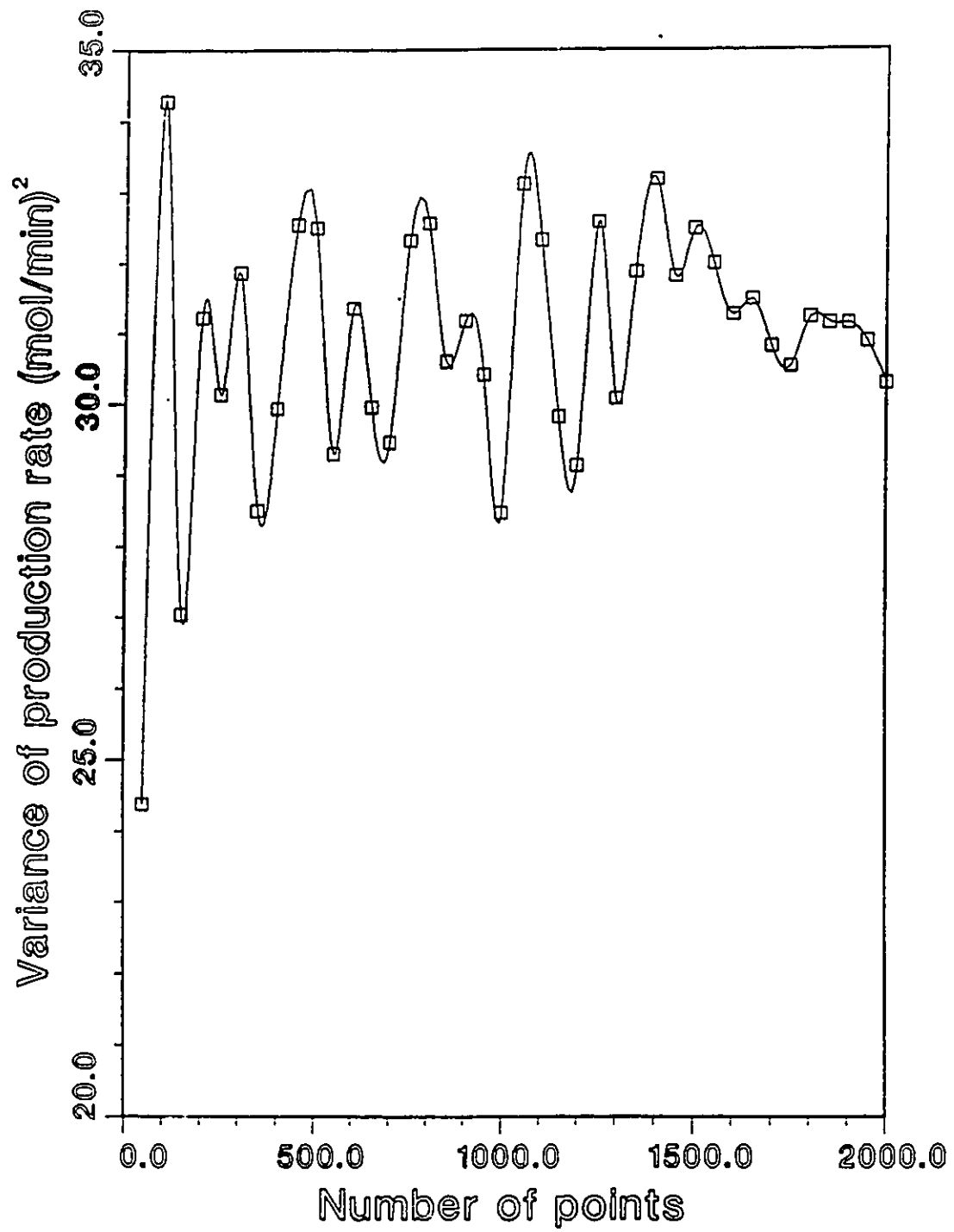


Figure C.1: The effect of sample size on the variance of the production rate for normally distributed errors.

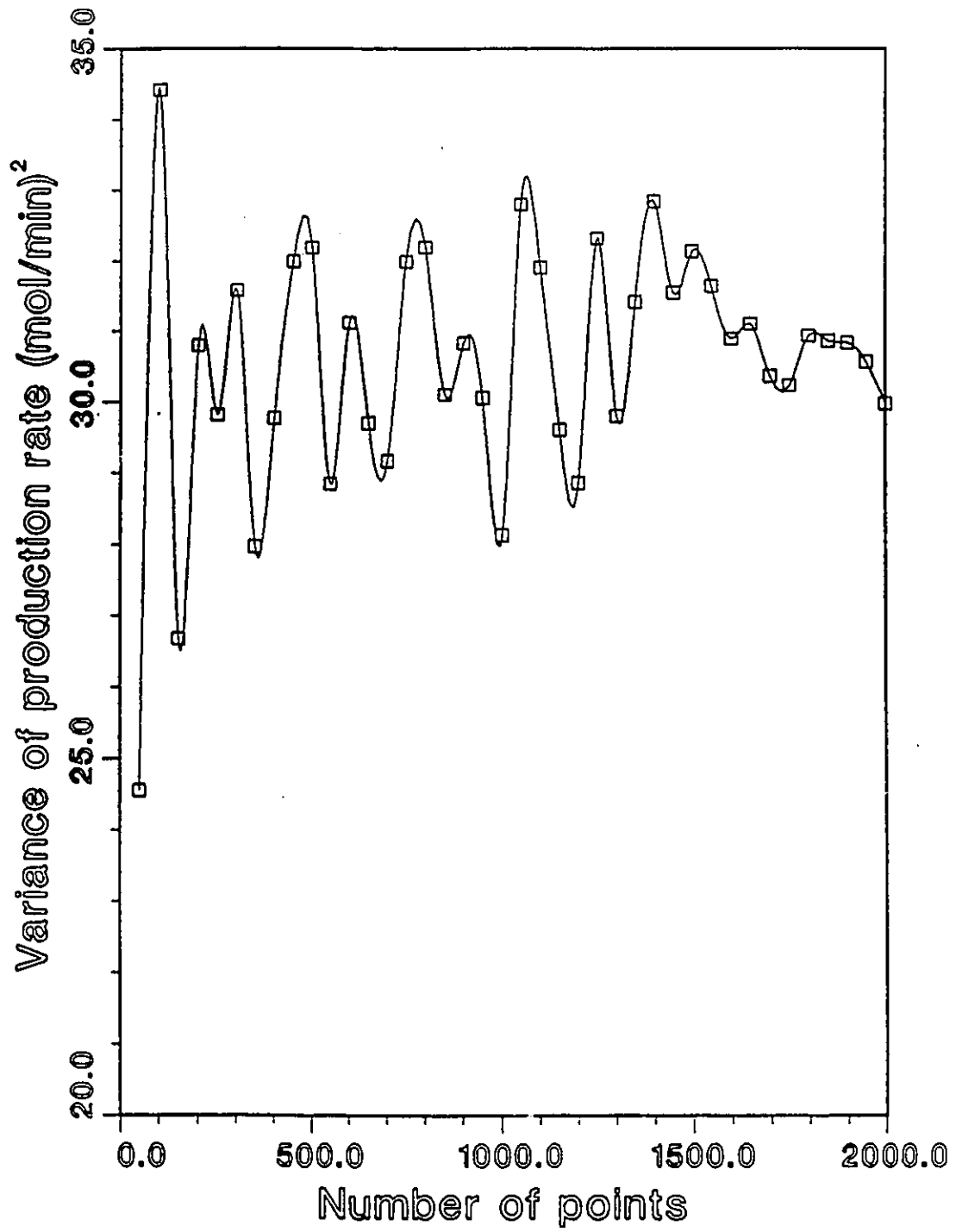


Figure C.2: The effect of sample size on the variance of the production rate for log-normally distributed errors.

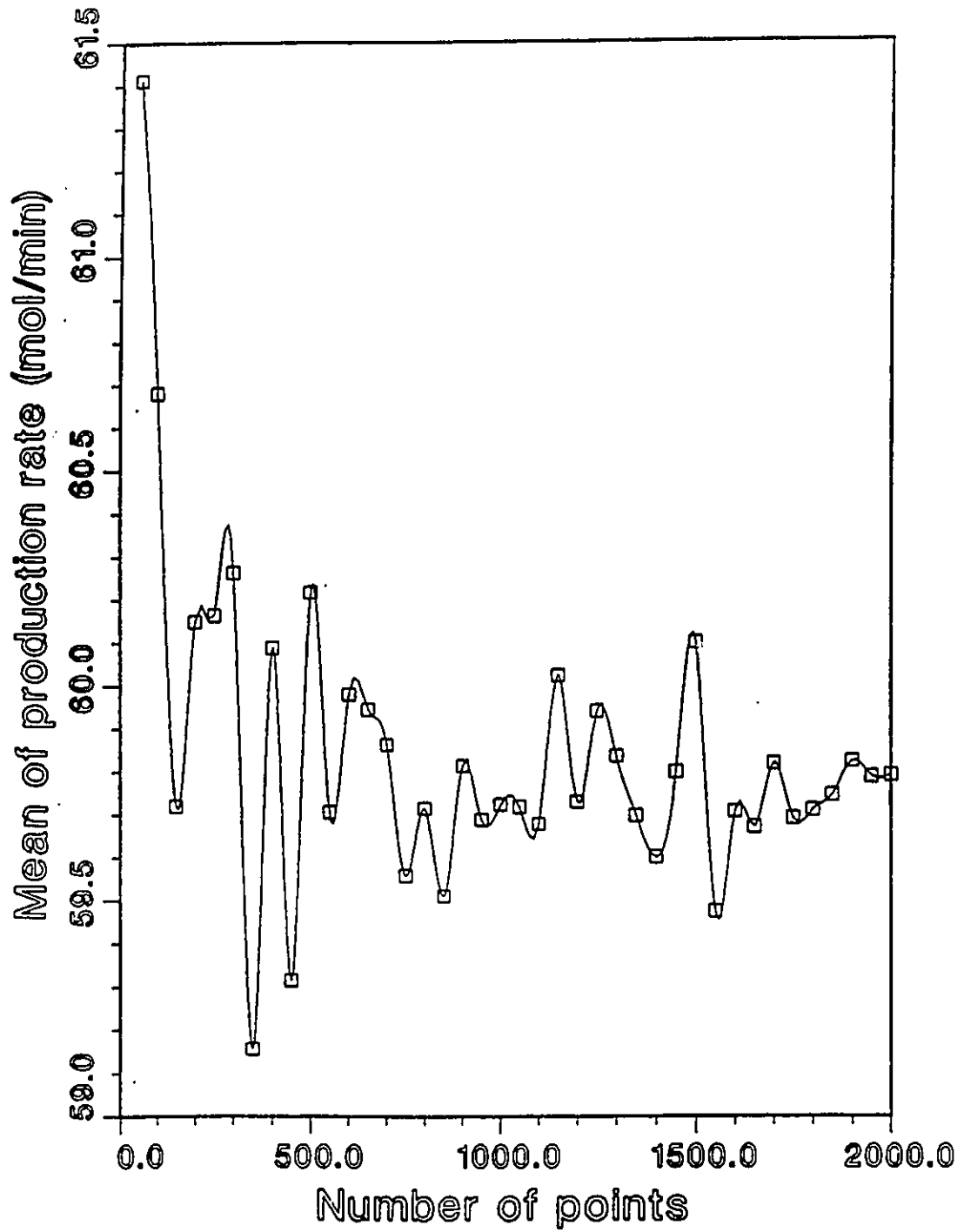


Figure C.3: The effect of sample size on the mean of the production rate for normally distributed errors.

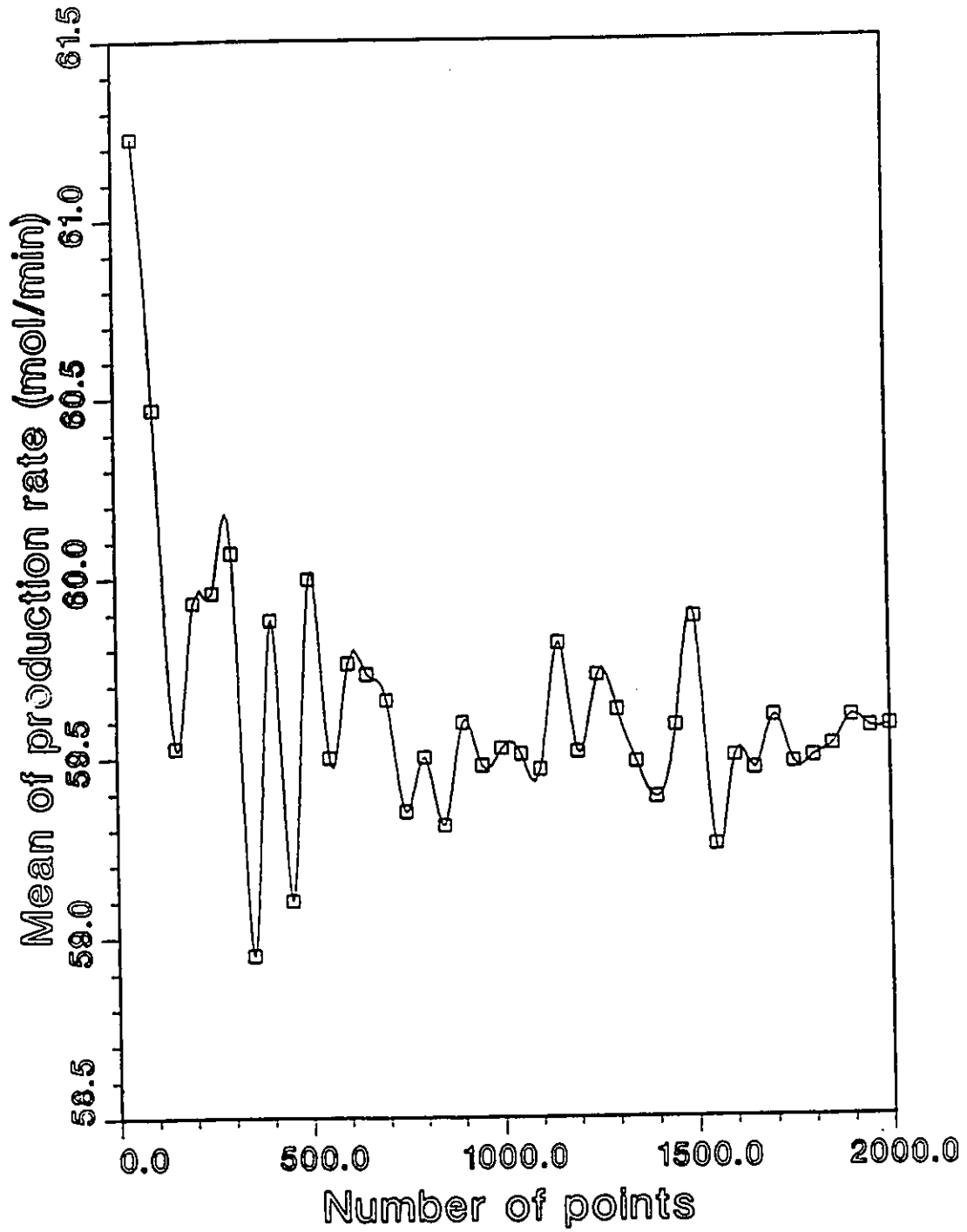


Figure C.4: The effect of sample size on the mean of the production rate for log-normally distributed errors. Error level= $\frac{\text{Std. Dev. } 100\%}{\text{Nominal Value}}$.

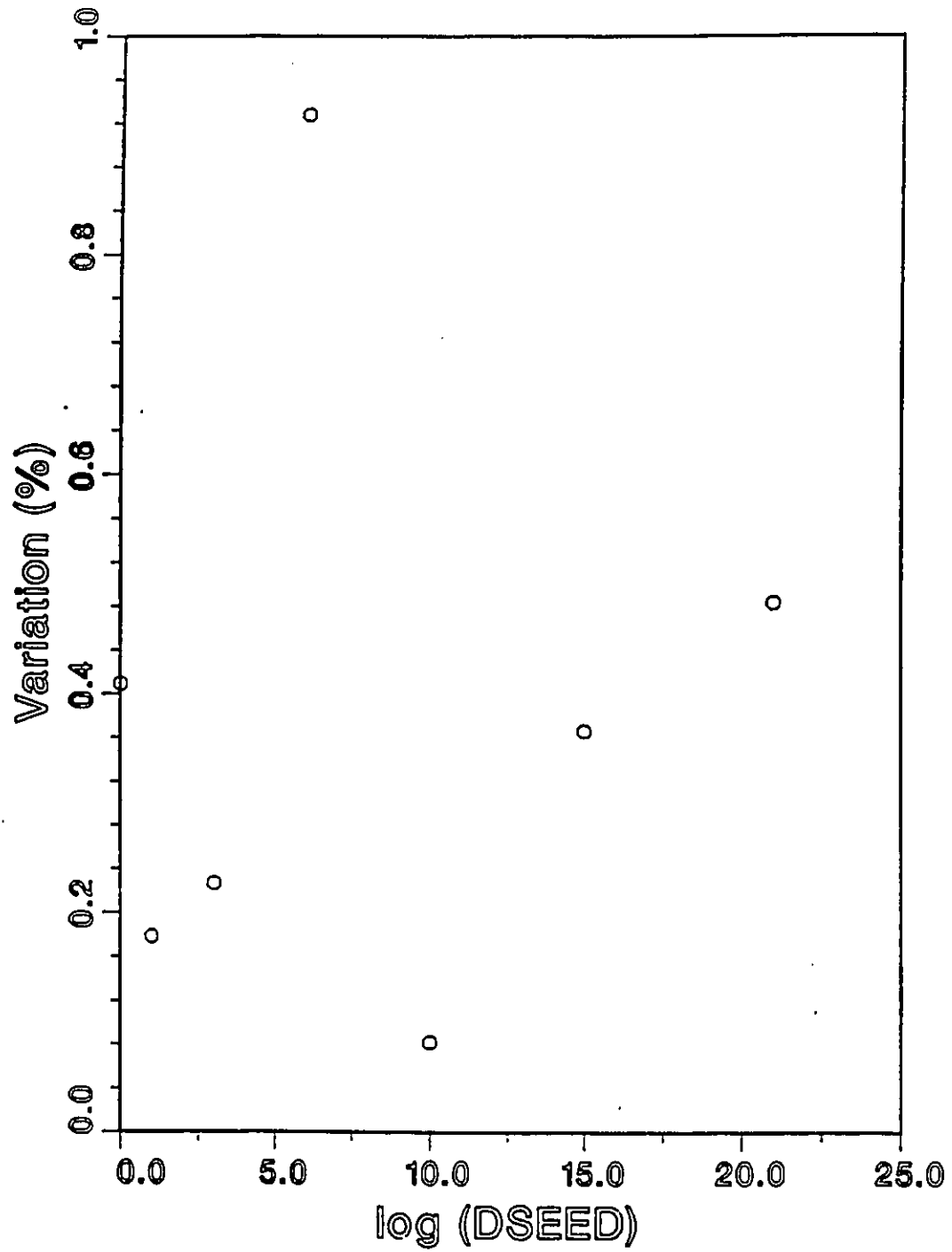


Figure C.5: The sensitivity of the variance with the seed value (log-normally distributed errors are considered).

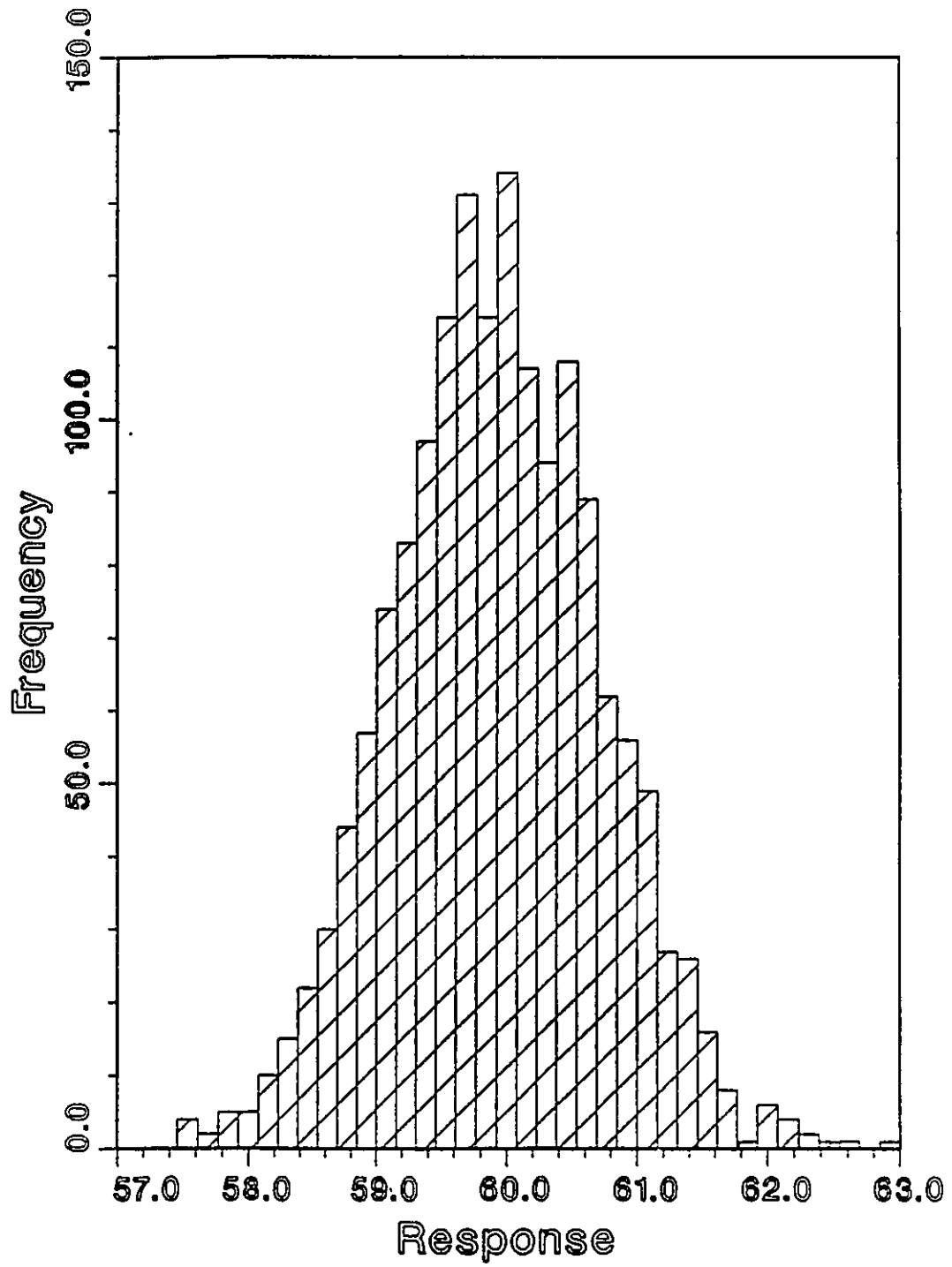


Figure C.6: Frequency Histogram of the production rate (normally distributed errors).

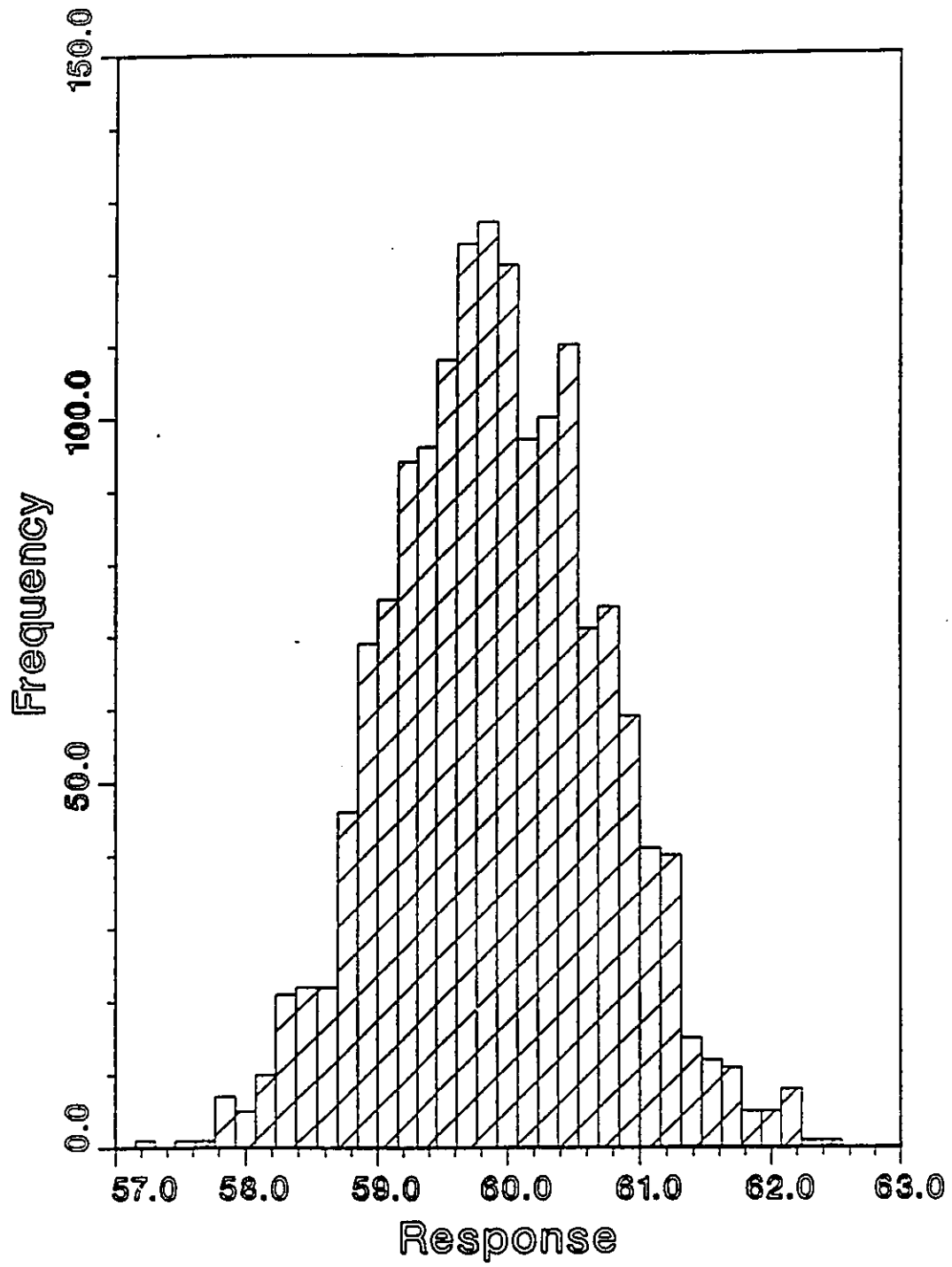


Figure C.7: Frequency Histogram of the production rate (log-normally distributed errors).

Appendix D

Orthogonal Arrays

D.1 Orthogonal Arrays for Taguchi Method

Table D.1: Inner array: two design variables with three levels each.

	1	2
1	1	1
2	1	2
3	2	3
4	2	2
5	3	1
6	3	3

Table D.2: Outer array: five error variables with three levels each.

	1	2	3	4	5
1	1	1	1	1	1
2	2	2	2	2	2
3	3	3	3	3	3
4	1	1	2	2	3
5	2	2	3	3	1
6	3	3	1	1	2
7	1	2	1	3	2
8	2	3	2	1	3
9	3	1	3	2	1
10	1	3	3	2	2
11	2	1	1	3	3
12	3	2	2	1	1
13	1	2	3	1	3
14	2	3	1	2	1
15	3	1	2	3	2
16	1	3	2	3	1
17	2	1	3	1	2
18	3	2	1	2	3

Table D.3: Inner array: two design variables with five levels each.

	1	2
1	1	1
2	2	2
3	3	3
4	4	4
5	5	5
6	2	3
7	3	4
8	4	5
9	5	1
10	1	2
11	3	5
12	4	1
13	5	2
14	1	3
15	2	4

Table D.4: Outer array: five error variables with five levels each.

	1	2	3	4	5
1	1	1	1	1	1
2	2	2	2	2	2
3	3	3	3	3	3
4	4	4	4	4	4
5	5	5	5	5	5
6	1	2	3	4	5
7	2	3	4	5	1
8	3	4	5	1	2
9	4	5	1	2	3
10	5	1	2	3	4
11	1	3	5	2	4
12	2	4	1	3	5
13	3	5	2	4	1
14	4	1	3	5	2
15	5	2	4	1	3
16	1	4	2	5	3
17	2	5	3	1	4
18	3	1	4	2	5
19	4	2	5	3	1
20	5	3	1	4	2
21	1	5	4	3	2
22	2	1	5	4	3
23	3	2	2	5	4
24	4	3	2	1	5
25	5	4	3	2	1

Table D.5: Inner array: five design variables with five levels each.

	1	2	3	4	5
1	1	1	1	1	1
2	2	2	2	2	2
3	3	3	3	3	3
4	4	4	4	4	4
5	5	5	5	5	5
6	1	2	3	4	5
7	2	3	4	5	1
8	3	4	5	1	2
9	4	5	1	2	3
10	5	1	2	3	4
11	1	3	5	2	4
12	2	4	1	3	5
13	3	5	2	4	1
14	4	1	3	5	2
15	5	2	4	1	3
16	1	4	2	5	3
17	2	5	3	1	4
18	3	1	4	2	5
19	4	2	5	3	1
20	5	3	1	4	2
21	1	5	4	3	2
22	2	1	5	4	3
23	3	2	2	5	4
24	4	3	2	1	5
25	5	4	3	2	1
26	1	1	4	5	4
27	2	2	5	1	5
28	3	3	1	2	1
29	4	4	2	3	2
30	5	5	3	4	3
31	1	2	1	3	3
32	2	3	2	4	4
33	3	4	3	5	5
34	4	5	4	1	1
35	5	1	5	2	2
36	1	3	3	1	2
37	2	4	4	2	3
38	3	5	5	3	4
39	4	1	1	4	5
40	5	2	2	5	1
41	1	4	5	4	1
42	2	5	1	5	2
43	3	1	2	1	3
44	4	2	3	2	4
45	5	3	4	3	5
46	1	5	2	2	5
47	2	1	3	3	1
48	3	2	4	4	2
49	4	3	5	5	3
50	5	4	1	1	4

D.2 Orthogonal Arrays for Sequential Elimination of Levels

Table D.6: Inner array: two design variables with five levels each.

	1	2
1	1	2
2	2	3
3	3	4
4	4	5
5	5	1
6	1	3
7	2	4
8	3	5
9	4	1
10	5	2

Table D.7: Inner array: two design variables with four levels each.

	1	2
1	1	2
2	2	3
3	3	4
4	4	1
5	1	3
6	2	4
7	3	1
8	4	2

Table D.8: Inner array: two design variables with three levels each.

	1	2
1	1	2
2	2	3
3	3	1
4	1	3
5	2	1
6	3	2

Table D.9: Inner array: two design variables with two levels each.

	1	2
1	1	1
2	2	2
3	1	2
4	2	1

Table D.10: Inner array: five design variables with five levels each.

	1	2	3	4	5
1	1	2	3	4	5
2	2	3	4	5	1
3	3	4	5	1	2
5	5	1	2	3	4
6	1	3	5	2	4
7	2	4	1	3	5
8	3	5	2	4	1
9	4	1	3	5	2
10	5	2	4	1	3

Table D.11: Inner array: five design variables with four levels each.

	1	2	3	4	5
1	1	2	3	4	1
2	2	3	4	1	2
3	3	4	1	2	3
4	4	1	2	3	4
5	1	2	3	4	1
6	2	3	4	1	2
7	3	4	1	2	3
8	4	1	2	3	4

Table D.12: Inner array: five design variables with three levels each.

	1	2	3	4	5
1	1	2	3	1	2
2	2	3	1	2	3
3	3	1	2	3	1
4	1	2	3	1	2
5	2	3	1	2	3
6	3	1	2	3	1

Table D.13: Inner array: five design variables with two levels each.

	1	2	3	4	5
1	1	2	1	2	1
2	1	2	2	1	2
3	2	1	1	2	2
4	2	1	2	1	1

Table D.14: Outer array: five error variables with five levels each.

	1	2	3	4	5
1	1	1	1	1	1
2	2	2	2	2	2
3	3	3	3	3	3
4	4	4	4	4	4
5	5	5	5	5	5
6	1	2	3	4	5
7	2	3	4	5	1
8	3	4	5	1	2
9	4	5	1	2	3
10	5	1	2	3	4
11	1	3	5	2	4
12	2	4	1	3	5
13	3	5	2	4	1
14	4	1	3	5	2
15	5	2	4	1	3
16	1	4	2	5	3
17	2	5	3	1	4
18	3	1	4	2	5
19	4	2	5	3	1
20	5	3	1	4	2
21	1	5	4	3	2
22	2	1	5	4	3
23	3	2	2	5	4
24	4	3	2	1	5
25	5	4	3	2	1

Appendix E

Optimal Designs for the Different Methods

E.1 Monte Carlo Simulations

Table E.1: Optimal settings using Monte Carlo simulations with two design variables and normally distributed errors.

error level (%)	temperature (K)	volume (l)
1	317.3	36.6
2	313.7	37.7
3	315.6	41.5
4	326.6	28.2
5	326.1	26.8
6	327.2	26.8
7	329.7	24.3
8	326.6	27.3
9	327.8	26.6
10	325.1	30.4

Table E.2: Optimal settings using Monte Carlo simulations with two design variables for log-normally distributed errors.

error level (%)	temperature (K)	volume (l)
1	316.7	35.6
2	327.1	24.9
3	327.2	23.1
4	316.4	35.4
5	325.1	26.4
6	325.0	30.2
7	323.8	31.2
8	329.7	23.8
9	328.6	20.3
10	328.6	20.4

Table E.3: Optimal settings using Monte Carlo simulations with five design variables for log-normally distributed errors.

error level (%)	inlet concentration of A (mol/m ³)	inlet concentration of B (mol/m ³)	inlet temperature (K)	temperature (K)	volume (l)
1	1293.3	455.6	314.5	306.0	65.1
2	1144.2	417.1	321.5	306.9	54.0
3	2633.9	420.0	312.3	308.3	47.4
4	3134.2	293.0	324.3	306.1	27.5
5	3452.4	340.8	320.9	301.2	27.9
6	3859.4	443.2	305.3	301.5	47.2
7	4723.3	460.0	310.7	305.2	27.2
8	4807.1	498.4	310.6	292.6	32.3
9	4995.7	405.9	317.4	292.1	21.6
10	4213.0	369.4	324.6	297.6	19.4

Table E.4: Optimal settings using Monte Carlo simulations and Response Surface Methodology with two design variables and normally distributed errors.

error level (%)	temperature (K)	volume (l)
1	318.7	38.4
2	317.8	39.6
3	318.2	39.4
4	325.8	29.6
5	327.7	28.1
6	328.7	28.1
7	330.0	25.5
8	330.0	28.7
9	330.0	27.9
10	329.9	31.9

Table E.5: Optimal settings using Monte Carlo simulations and Response Surface Methodology with two design variables for log-normally distributed errors.

error level (%)	temperature (K)	volume (l)
1	316.7	35.6
2	327.1	24.9
3	327.2	23.1
4	316.4	35.4
5	325.1	26.4
6	325.0	30.2
7	323.8	31.2
8	329.7	23.8
9	328.6	20.3
10	328.6	20.4

Table E.6: Optimal settings using Monte Carlo simulations and Response Surface Methodology with five design variables for log-normally distributed errors.

error level (%)	inlet concentration of A (mol/m ³)	inlet concentration of B (mol/m ³)	inlet temperature (K)	temperature (K)	volume (l)
1	1319.1	464.7	319.7	299.9	66.4
2	1167.1	424.1	327.9	301.1	55.1
3	2656.6	426.7	317.3	302.2	48.3
4	3196.9	298.7	329.8	300.2	28.0
5	3521.4	339.7	327.3	295.2	28.5
6	3936.6	448.0	310.7	295.5	48.1
7	4817.8	466.6	316.9	299.3	27.7
8	4903.2	499.7	316.1	290.1	32.9
9	5000.0	414.0	323.3	290.1	22.0
10	4297.3	376.8	329.1	291.6	19.7

E.2 Error Transmission Analysis

Table E.7: Optimal settings using Error Transmission Analysis with two design variables.

error level (%)	temperature (K)	volume (l)
1	300.0	90.0
2	300.0	90.0
3	300.0	90.0
4	300.0	87.1
5	300.0	87.2
6	300.0	87.2
7	300.0	87.2
8	300.0	87.5
9	300.0	87.0
10	300.0	87.8

Table E.8: Optimal settings using Error Transmission Analysis with five design variables.

error level (%)	inlet concentration of A (mol/m ³)	inlet concentration of B (mol/m ³)	inlet temperature (K)	temperature (K)	volume (l)
1	4131.3	233.8	328.4	327.8	85.2
2	4319.8	286.4	329.9	315.7	78.5
3	4353.4	343.9	330.0	328.3	74.3
4	4374.2	195.7	330.0	325.9	79.9
5	4775.7	289.0	330.8	310.5	89.7
6	4768.1	307.9	326.7	309.0	88.5
7	2455.2	330.2	329.8	329.6	86.9
8	3819.9	208.4	328.2	319.6	90.0
9	3834.2	290.9	328.4	321.1	81.1
10	4837.8	279.5	329.2	306.6	86.5

E.3 Taguchi Method

Table E.9: Optimal settings using Taguchi Method with two design variables at three levels each.

error level (%)	temperature (K)	volume (l)
1	290	50
2	290	50
3	290	10
4	290	10
5	290	10
6	290	10
7	290	10
8	290	10
9	290	10
10	290	10

Table E.10: Optimal settings using Taguchi Method with two design variables at five levels each.

error level (%)	temperature (K)	volume (l)
1	290	70
2	290	70
3	290	70
4	290	70
5	290	70
6	290	70
7	290	70
8	290	70
9	290	70
10	290	70

Table E.11: Optimal settings using a quadratic model for the sum squares of errors with two design variables at five levels each.

error level (%)	temperature (K)	volume (l)
1	294.1	87.8
2	294.0	87.9
3	294.4	87.7
4	294.6	87.6
5	295.2	87.3
6	296.2	87.3
7	296.3	86.7
8	296.9	86.4
9	296.9	86.4
10	298.1	85.8

Table E.12: Optimal settings using a quadratic model for the signal-to-noise ratio with two design variables at five levels each.

error level (%)	temperature (K)	volume (l)
1	290.6	90.0
2	290.8	90.0
3	290.9	90.0
4	291.0	90.0
5	291.6	90.0
6	291.8	90.0
7	292.5	90.0
8	293.0	90.0
9	293.2	90.0
10	293.7	90.0

Table E.13: Optimal settings using Taguchi Method with five design variables at five levels each.

error level (%)	inlet concentration of A (mol/m^3)	inlet concentration of B (mol/m^3)	inlet temperature (K)	temperature (K)	volume (l)
1	4000	300	310	310	70
2	4000	300	310	310	70
3	4000	300	310	310	70
4	4000	300	310	310	70
5	4000	300	310	310	70
6	4000	300	310	310	70
7	4000	300	310	310	70
8	4000	300	310	310	70
9	4000	300	310	310	70
10	4000	300	310	310	70

E.4 Sequential Elimination of Levels

Table E.14: Optimal settings using Sequential Elimination of Levels with two design variables at five levels each and a total of 16 runs.

error level (%)	temperature (K)	volume (l)
1	320.0	90.0
2	310.0	90.0
3	310.0	50.0
4	310.0	50.0
5	310.0	50.0
6	310.0	50.0
7	310.0	50.0
8	310.0	50.0
9	310.0	50.0
10	310.0	50.0

Table E.15: Optimal settings using Sequential Elimination of Levels with two design variables at five levels each and a total of 32 runs.

error level (%)	temperature (K)	volume (l)
1	328.0	74.0
2	318.0	74.0
3	310.0	66.0
4	310.0	66.0
5	310.0	66.0
6	310.0	66.0
7	310.0	66.0
8	310.0	66.0
9	310.0	58.0
10	306.0	58.0

Table E.16: Optimal settings using Sequential Elimination of Levels with two design variables at five levels each and a total of 48 runs.

error level (%)	temperature (K)	volume (l)
1	328.0	70.0
2	318.0	70.0
3	312.0	62.0
4	312.0	62.0
5	312.0	62.0
6	312.0	62.0
7	312.0	62.0
8	312.0	62.0
9	312.0	54.0
10	305.0	60.0