Pergamon

Chemical Engineering Science, Vol. 53, No. 11, pp. 2029–2040, 1998

Printed in Great Britain
PII: S0009-2509(98)00025-6
0009-2509/98 $19.00 + 0.00

On the costs of parameter uncertainties. Effects of parameter uncertainties during optimization and design of experiments

José Carlos Pinto*

Programa de Engenharia Química/COPPE, Universidade Federal do Rio de Janeiro, CP:68502 - Cidade Universitária, 21945-970 - Rio de Janeiro - RJ, Brazil

(Received 11 June 1997; accepted 28 January 1998)

Abstract—Mathematical models are of widespread usage for simulating process behavior, designing new processes and equipment and, in a more general sense, decision making. However, as model parameters are uncertain, due to model inaccuracies and experimental errors, all model results are subject to uncertainties. It is shown here that an economical value may be assigned to parameter uncertainties, which can then be used for both process optimization and specially for taking decisions during sequential experimental designs. © 1998 Elsevier Science Ltd. All rights reserved.

Keywords: Parameter estimation; cost; experimental design; optimization; uncertainty.

1. INTRODUCTION

In order to take decisions, engineers and business people usually rely on mathematical models. Such models, no matter whether they are developed within the framework of a certain theory or whether they are completely empirical, are built in order to record experimental observations, particularly, the relationships and interactions which exist among the various variables of interest, in a compact way. When a model is validated, it is generally used for predicting how a certain group of variables will change when other variables are modified (simulation), for finding the conditions or variable settings that will maximize a certain performance index (optimization) or for designing new equipment and/or enterprise (project).

Usually, for a model to be useful, certain numbers or parameters must be either measured or estimated, by obliging the mathematical structure to fit the experimental data. So, knowing that the relationship between two variables may be described by a straight line is of no use unless the angular and linear coefficients are known. The problem is that measurements and experimental observations are subject to errors, caused either by natural fluctuations or by the limitations of real measurement devices. Besides, model and reality are not identical, as many variables are not taken into consideration during the model development (hopefully the least important ones) and because reality is not completely understood (and even if it were understood, the model would probably be too complex to be of practical use). For all these reasons, model predictions are uncertain to some extent. As model predictions depend on model parameters, the measured or estimated parameters may be regarded as uncertain, although this uncertainty actually reflects model inaccuracies and experimental errors and fluctuations.

Parameter estimation procedures for different model structures have been extensively studied in the literature (see Bard, 1974). Some techniques have also been developed for designing experiments for reducing parameter uncertainties (see Pinto *et al.*, 1990, 1991). The impact of parameter uncertainties on the performance of control algorithms has been extensively studied in the literature and has led to the development of a class of process controllers known as robust controllers (for examples, see Keel and Bhattacharyya, 1994; Oldak *et al.*, 1994; Schaper *et al.*, 1992). Similarly, parameter uncertainties are usually taken into consideration to study the sensitivity of process responses to variable changes, which is usually known as sensitivity analysis (for example see Fotopoulos *et al.*, 1994; Reed and Whiting, 1993; Seferlis and Hrymak, 1996; Ganesh and Biegler, 1987; Volin and Ostrovskiy, 1981a, b). The impact caused by parameter uncertainties on variables of economic significance for chemical processes has been largely overlooked though. Besides, to our knowledge, the economics of process variables and parameter uncertainties have not been taken into consideration for proper experimental design for both model discrimination and parameter estimation.

*E-mail: pinto@peq.coppe.ufrj.br.

If one realizes that in a real production environment models are mostly used and developed to maximize profits, it may be concluded that parameter uncertainties lead to profit uncertainties. Moreover, as model parameters are uncertain, optimum variable settings obtained with the model are also uncertain. By not setting the variables to their real optimum values, the entrepreneur loses money. Therefore, an economical value may be assigned to parameter uncertainties.

Seemingly, Weisman and Holzman (1972) were the first to analyze the importance of variable uncertainties during process design. They proposed that the total expected cost of the process be given by

$$E(C_t) = \sum_h E(C_h) + \sum_i p_i E(C_i) \tag{1}$$

where the first term on the right-hand side represents total costs of normal capital and operating items, while the second term on the right-hand side represents the expected failure costs, where p_i are the failure probabilities of the ith process constraint. The failure costs include costs introduced by process variable and parameter uncertainties. Particularly, Weisman and Holzman (1972) showed that cost variances are proportional to variable and parameter variances and that the optimum process design should minimize both process costs and cost variability.

Although developed independently, similar concepts were used by many other researchers in order to develop the set of techniques which are known nowadays as Taguchi's design procedures (for examples, see Taguchi, 1986; Shoemaker *et al.*, 1991; Coleman and Montgomery, 1993; Lucas, 1994). Assuming that linear models may be built to describe process responses and assuming that economical losses are proportional to cost variances, it is possible to provide an experimental design that will allow the proper modeling and optimization of process conditions. Very often, however, the assumptions presented above are not valid for both process and experimental design and final results obtained with Taguchi's designs may not be adequate.

More recently, Becker *et al.* (1994) and Pistikopoulos (1995) presented new algorithms for taking decisions and optimizing process performance in uncertain conditions. Becker *et al.*'s procedure is an extension of the basic procedure developed by Weisman and Holzman (1972), which proposes the simultaneous minimization of process costs and cost variabilities. Pistikopoulos's procedure is based on the minimization of the average process cost, which is computed as a weighed summation of local costs obtained by optimizing process operation conditions at different points inside the domain where variables are allowed to vary. The weight factors may be the joint probability of finding the specified parameter combinations used for computing the local optima.

Our main concern here is showing that an economical value may be assigned to parameter uncertainties, which can then be used for both process optimization and specially for taking decisions during sequential experimental designs. The approach used here is similar to the approach used originally by Weisman and Holzman (1972), which means that process models and objective functions may be linearized in the vicinities of the optimum to allow the computation of the sensitivities of the model responses to parameter perturbations. Although the introduction of equality and inequality constraints into the optimization problem is much more difficult in this case, this approach is appealing because it preserves the basic nonlinear behavior of process responses, allows the derivation of simple mathematical expressions to describe the optimization problem and does not require that extensive computer simulations be carried out for optimum solutions to be found.

2. THEORETICAL FRAMEWORK

Let us assume that a certain objective function (expected profits) $L(z, w, \alpha)$ is to be maximized, where z, w and α are vectors of decision variables (that will be manipulated during optimization), fixed variables (that will remain fixed during the optimization) and model parameters. Thus, optimization here means selecting optimal values for z. If the optimum conditions of the maximization problem are found but are subject to uncertainties, which may be due to experimental errors or model inaccuracies, the actual profits obtained during operation will be different from the expected ones. If variable deviations are not large, the following expression may be written:

$$\begin{aligned}\Delta L &= L(z^* + \varepsilon_z, w + \varepsilon_w, \alpha + \varepsilon_\alpha) - L(z^*, w, \alpha) \\ &= (\nabla_z L)^T \varepsilon_z + (\nabla_w L)^T \varepsilon_w + (\nabla_\alpha L)^T \varepsilon_\alpha \\ &\quad + \frac{1}{2}(\varepsilon_z)^T (\nabla^2_{z,z} L)(\varepsilon_z) + \frac{1}{2}(\varepsilon_w)^T (\nabla^2_{w,w} L)(\varepsilon_w) \\ &\quad + \frac{1}{2}(\varepsilon_\alpha)^T (\nabla^2_{\alpha,\alpha} L)(\varepsilon_\alpha) + (\varepsilon_z)^T (\nabla^2_{z,w} L)(\varepsilon_w) \\ &\quad + (\varepsilon_z)^T (\nabla^2_{z,\alpha} L)(\varepsilon_\alpha) + (\varepsilon_w)^T (\nabla^2_{w,\alpha} L)(\varepsilon_\alpha) \\ &\quad + \cdots\end{aligned} \tag{2}$$

where

$$\nabla_x L = \left[\frac{\partial L}{\partial x_i}\right]_{NX \times 1} \tag{3}$$

$$\nabla^2_{x,y} L = \left[\frac{\partial^2 L}{\partial x_i \, \partial y_j}\right]_{NX \times NY} \tag{4}$$

and the derivatives are computed at the nominal optimum conditions. If ΔL is positive, uncertainties have a positive net effect on the process operation, as profits are larger than expected. The opposite happens when ΔL is negative.

Assuming that uncertainties are centered at zero and that the different groups of variables are not correlated, which is reasonable in most cases as these variables are usually measured and/or evaluated by

independent procedures, then eq. (2) may be averaged to allow the computation of the average profit deviation as

$$\overline{\Delta L} = \sum_{i=1}^{NZ}\sum_{j=1}^{NZ}\frac{1}{2}\frac{\partial^2 L}{\partial z_i\,\partial z_j}\sigma_{zij}^2 + \sum_{i=1}^{NW}\sum_{j=1}^{NW}\frac{1}{2}\frac{\partial^2 L}{\partial w_i\,\partial w_j}\sigma_{wij}^2 + \sum_{i=1}^{N\alpha}\sum_{j=1}^{N\alpha}\frac{1}{2}\frac{\partial^2 L}{\partial \alpha_i\,\partial \alpha_j}\sigma_{\alpha ij}^2 \tag{5}$$

which is often different from zero, due to the nonlinear behavior of the profit function, and may be called the bias of the optimization procedure, as suggested by Becker *et al.* (1994).

The variance of the cost deviation can be computed as

$$\overline{\Delta L^2} = \sum_{i=1}^{NZ}\sum_{j=1}^{NZ}\frac{\partial L}{\partial z_i}\frac{\partial L}{\partial z_j}\sigma_{zij}^2 + \sum_{i=1}^{NW}\sum_{j=1}^{NW}\frac{\partial L}{\partial w_i}\frac{\partial L}{\partial w_j}\sigma_{wij}^2 + \sum_{i=1}^{N\alpha}\sum_{j=1}^{N\alpha}\frac{\partial L}{\partial \alpha_i}\frac{\partial L}{\partial \alpha_j}\sigma_{\alpha ij}^2 \tag{6}$$

$$\sigma_{\Delta L}^2 = \overline{\Delta L^2} - \overline{\Delta L}^2 \tag{7}$$

where σ_{xij}^2 is the covariance of elements i and j of vector x.

Assuming additionally that the uncertainties regarding the expected profits may be described locally by a Gaussian distribution, the following equation may be written as:

$$L_{99\%} = L + \overline{\Delta L} - 2.35\,\sigma_{\Delta L} \tag{8}$$

which represents the minimum profit expected, with a confidence level of 99%, given the uncertainties of the optimization problem. Equation (8) gives a lower boundary for the expected profit, so that profits are not expected to be below this value. In order to take a decision, it may be interesting then to maximize this riskless profit function, as described by eq. (8), instead of maximizing the nominal profit function. In this case, calling L the nominal profit of the enterprise, the risk may be defined as

$$R = \overline{\Delta L} - 2.35\,\sigma_{\Delta L}. \tag{9}$$

It is interesting to observe that eq. (8) implies that both the nominal costs and process variability have to be minimized simultaneously, as described by Weisman and Holzman (1972), Taguchi (1986) and Becker *et al.* (1994), in order to minimize the risk of the enterprise and maximize the actual profits obtained. Eq. (8), however, preserves the nonlinear behavior of both the process model and the objective function, as both $\overline{\Delta L}$ and $\sigma_{\Delta L}^2$ are nonlinear functions of the local variables. For all these reasons, it is proposed here that Equation (8) be used for taking robust decisions and minimizing the risks of the enterprise.

As already discussed, the actual optimum solution is unknown, as both the fixed variables and model parameters are uncertain. At the optimum, however, it is possible to write

$$\nabla_z L\,(z^* + \Delta z^*, w + \varepsilon_w, \alpha + \varepsilon_\alpha) = 0 \tag{10}$$

which may be rewritten as

$$(\nabla_{z,z}^2 L)\,\Delta z^* + (\nabla_{z,w}^2 L)\,\varepsilon_w + (\nabla_{z,\alpha}^2 L)\,\varepsilon_\alpha = 0 \tag{11}$$

so that

$$\Delta z^* = -(\nabla_{z,z}^2 L)^{-1}\{(\nabla_{z,w}^2 L)\,\varepsilon_w + (\nabla_{z,\alpha}^2 L)\,\varepsilon_\alpha\} \tag{12}$$

which shows explicitly how the optimum solution changes when fixed variables and model parameters vary. The value of Δz^* may be added to ε_z in eq. (2), as

$$\begin{aligned}\Delta L^* &= L(z^* + \varepsilon_z + \Delta z^*, w + \varepsilon_w, \alpha + \varepsilon_\alpha) - L(z^*, w, \alpha)\\ &= (\nabla_z L)^T(\varepsilon_z + \Delta z^*) + (\nabla_w L)^T\varepsilon_w + (\nabla_\alpha L)^T\varepsilon_\alpha\\ &\quad + \frac{1}{2}(\varepsilon_z + \Delta z^*)^T(\nabla_{z,z}^2 L)(\varepsilon_z + \Delta z^*)\\ &\quad + \frac{1}{2}(\varepsilon_w)^T(\nabla_{w,w}^2 L)(\varepsilon_w)\\ &\quad + \frac{1}{2}(\varepsilon_\alpha)^T(\nabla_{\alpha,\alpha}^2 L)(\varepsilon_\alpha) + (\varepsilon_z + \Delta z^*)^T(\nabla_{z,w}^2 L)(\varepsilon_w)\\ &\quad + (\varepsilon_z + \Delta z^*)^T(\nabla_{z,\alpha}^2 L)(\varepsilon_\alpha) + (\varepsilon_w)^T(\nabla_{w,\alpha}^2 L)(\varepsilon_\alpha)\\ &\quad + \cdots\end{aligned} \tag{13}$$

allowing the computation of the difference between the new optimum profit that would be obtained, if the variables were allowed to vary and the optimum condition were calculated properly, and the nominal profit expected. This difference represents what is lost for not knowing exactly what the real values are. If it is positive, a certain amount of money is being lost for not operating at the actual optimum conditions. Inserting eq. (12) into eq. (13) and averaging the resulting expression, it is possible to write

$$\overline{\Delta L^*} = \sum_{i=1}^{NZ}\sum_{j=1}^{NZ}\frac{1}{2}\frac{\partial^2 L}{\partial z_i\,\partial z_j}\sigma_{zij}^2 + \sum_{i=1}^{NW}\sum_{j=1}^{NW}\frac{1}{2}g_{ij}\,\sigma_{wij}^2 + \sum_{i=1}^{N\alpha}\sum_{j=1}^{N\alpha}\frac{1}{2}h_{ij}\,\sigma_{\alpha ij}^2 \tag{14}$$

which represents the average loss that is caused by not operating at the actual optimum conditions due to uncertainties. g_{ij} and h_{ij} are

$$G = [g_{ij}]_{NW\times NW} = (\nabla_{w,w}^2 L) - (\nabla_{z,w}^2 L)^T(\nabla_{z,z}^2 L)^{-1}(\nabla_{z,w}^2 L) \tag{15}$$

$$H = [h_{ij}]_{N\alpha\times N\alpha} = (\nabla_{\alpha,\alpha}^2 L) - (\nabla_{z,\alpha}^2 L)^T(\nabla_{z,z}^2 L)^{-1}(\nabla_{z,\alpha}^2 L). \tag{16}$$

Finally, calculating the difference between eqs (14) and (5),

$$C = \overline{\Delta L^*} - \overline{\Delta L} = -\frac{1}{2}\left(\sum_{i=1}^{NW}\sum_{j=1}^{NW}g_{ij}^c\,\sigma_{wij}^2 + \sum_{i=1}^{N\alpha}\sum_{j=1}^{N\alpha}h_{ij}^c\,\sigma_{\alpha ij}^2\right) \tag{17}$$

which represents the total average loss that can be expected for not operating at the unknown optimum conditions. C takes into account the fact that part of

the total loss is caused by not knowing the correct values of process variables (ΔL^*) during the optimization and that a second part is caused by process uncertainties (ΔL), which may have a positive or a negative net effect on the process operation, as discussed previously. If C is positive, a certain amount of money is being wasted for not operating at the actual optimum conditions. g_{ij}^c and h_{ij}^c are

$$G^c = [g_{ij}^c]_{NW \times NW} = (\nabla_{z,w}^2 L)^T (\nabla_{z,z}^2 L)^{-1} (\nabla_{z,w}^2 L) \quad (18)$$

$$H^c = [h_{ij}^c]_{N\alpha \times N\alpha} = (\nabla_{z,\alpha}^2 L)^T (\nabla_{z,z}^2 L)^{-1} (\nabla_{z,\alpha}^2 L). \quad (19)$$

As averaging is a linear operation, eq. (2) may be subtracted from eq. (13) before averaging. After some algebraic manipulation, it is possible to show that C is always positive, as shown in the appendix. This means that uncertainties always have a deleterious net effect on process operation.

Therefore, an economical value (C) may be assigned to the uncertainties, as this is the recoverable loss caused by not knowing the fixed variables and model parameters properly. Notice that the losses caused by the uncertainties of the manipulated variables is not recoverable, as they do not lead to any change of the computed optimum solutions. C is called here the cost of uncertainties. The second summation of eq. (17) is called here the cost of parameter uncertainties and is linked to the parameter estimation procedure used to evaluate the model parameters. Therefore, an economical value may be assigned to any experiment during a sequential experimental design procedure, intended to reduce the uncertainty of model parameters. This will be shown in the examples presented in the next section.

It is important to emphasize that eq. (17) shows that the values assigned to parameter uncertainties and experiments depend heavily on the problem that is being analysed. Unless the objective function is known, which means that it is known precisely what model parameters are used for, the value of model parameters and additional experiments cannot be defined. This is a strong link which may be built between the design procedure and the more fundamental modeling studies and parameter estimation routines.

3. EXAMPLES

Example 1: *The design of a chemical reactor*

Let us assume that a CSTR must be designed in order to carry out the following chemical reaction:

$$A \xrightarrow{K1} B \xrightarrow{K2} C \quad (20)$$

where B is the desired product. The available data are presented in Table 1. It may be seen that kinetic parameters are assumed to be known with a fair amount of uncertainty, as it is not unusual to face kinetic constants with standard deviations that are larger than the absolute value of the kinetic constant. According to the available data, the second kinetic parameter, which is related to the formation of the by-product, is more uncertain than the first one, which is related to the formation of the desired product B, as it usually happens. Besides, kinetic parameters are assumed to be independent of each other, which is not always true, as kinetic parameters are often estimated simultaneously, based on available experimental data (see Bard, 1974).

The optimization problem consists of finding the optimum reactor volume and feed rate (residence time) in order to maximize the profits of the operation in two years, which is assumed to be the investment lifetime. The kinetic model can be described by

$$C_A = \frac{C_{A0}}{(1 + K1\Theta)} \quad (21)$$

$$C_B = \frac{K1\Theta C_{A0}}{(1 + K1\Theta)(1 + K2\Theta)} \quad (22)$$

$$C_C = \frac{K1K2\Theta^2 C_{A0}}{(1 + K1\Theta)(1 + K2\Theta)} \quad (23)$$

while the profit function to be maximized is

$$L(V, \Theta; K1, K2) = \frac{(C_B\,\text{US\$}B + C_C\,\text{US\$}C - C_{A0}\,\text{US\$}A - \text{US\$}Q)\,V\Delta t}{\Theta} - \text{US\$}V\,V^2 \quad (24)$$

Table 1. Available data for example 1

Variable	Value	Meaning
US$$A$$	4.0 US$/Kg	Price of reactant
US$$B$$	20.0 US$/Kg	Price of desired product
US$$C$$	3.0 US$/Kg	Price of by-product
US$$Q$$	0.50 US$/Kg A	Specific operational cost
US$$V$$	20×10^6 US$/(m^3)^2	Specific investment cost
C_{A0}	1000 Kg/m^3	Feed concentration
$K1$	2.5 h^{-1}	Kinetic constant
$K2$	1.0 h^{-1}	Kinetic constant
Δt	2 years	Investment lifetime
σ_{11}^2	0.09 h^{-2}	Variance of $K1$
σ_{22}^2	0.16 h^{-2}	Variance of $K2$
$\sigma_{12}^2, \sigma_{21}^2$	0	Covariances of $K1$ and $K2$

In this problem, $z^T = (V, \Theta)$ and $\alpha^T = (K1, K2)$. All other parameters belong to w.

Figure 1 shows the optimum volume as a function of residence time for different combinations of $K1$ and $K2$, within the range where these parameters are allowed to vary with 99% of confidence. It may be observed that significant variations of the optimum conditions occur. At the nominal value, Figs 1 and 2 show that the optimum is placed at $\Theta = 0.283$ h and $V = 4.23$ m^3, allowing total profits of $L =$ US$ 357×10^6. However, Fig. 2 shows that significant variations may occur. Particularly, if $K2$ is larger and $K1$

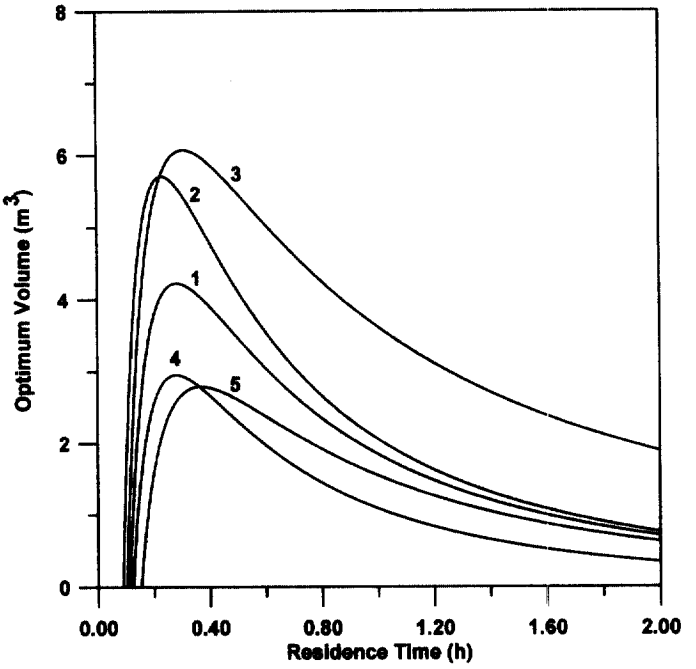

Fig. 1. Optimum reactor volume as a function of residence time. (1) Nominal conditions; (2) $K1 = 3.1$ h^{-1}; (3) $K2 = 0.2$ h^{-1}; (4) $K2 = 1.8$ h^{-1}; (5) $K1 = 1.9$ h^{-1}).

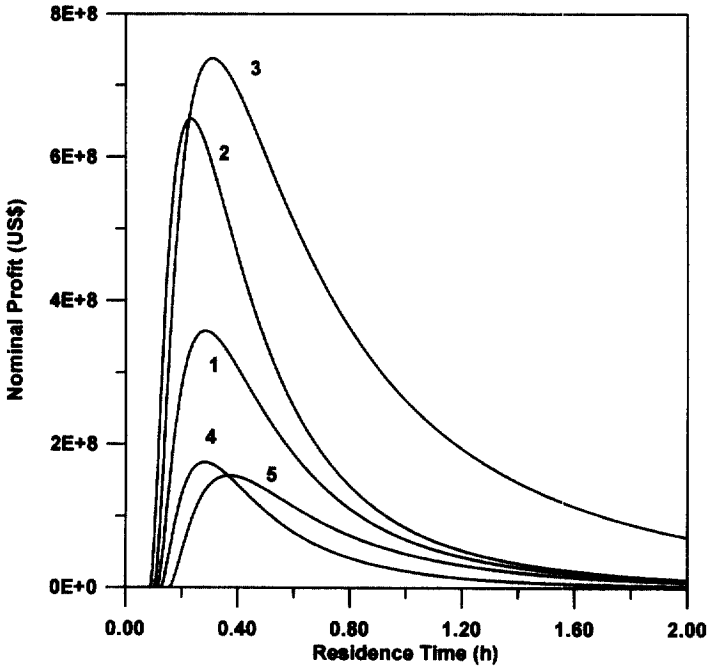

Fig. 2. Maximum profits as a function of residence time. (1) Nominal conditions; (2) $K1 = 3.1$ h^{-1}; (3) $K2 = 0.2$ h^{-1}; (4) $K2 = 1.8$ h^{-1}; (5) $K1 = 1.9$ h^{-1}).

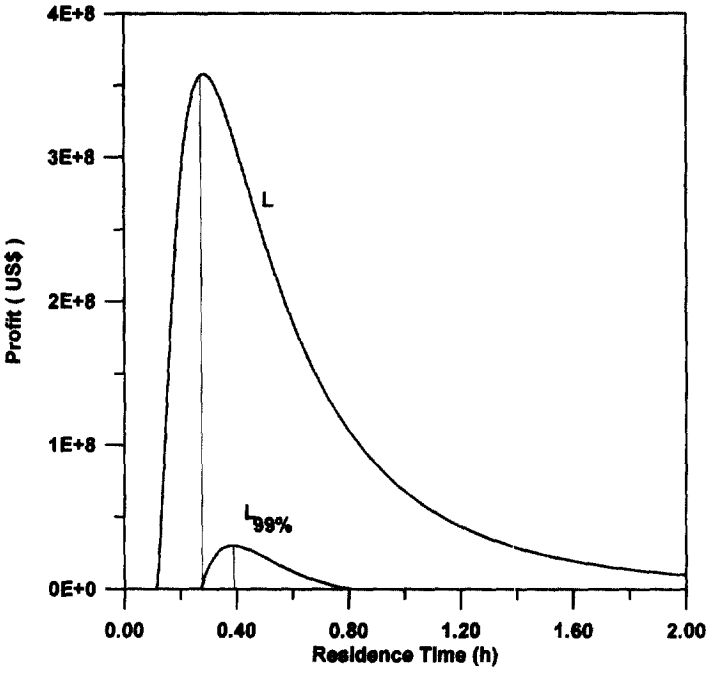

Fig. 3. Maximum nominal and riskless profits as functions of the residence time.

is lower than the nominal values, process operation may lead to a no-profit situation.

Figure 3 shows optimum profits as a function of residence time. If the riskless profit function $L_{99\%}$ is used for reactor design, the optimum condition is placed at $\Theta = 0.388$ h and $V = 3.95$ m^3. In this case, a minimum profit of $L =$ US$ 30×10^6 is guaranteed, although the average profit expected equals $L =$ US$ 340×10^6. This is a much more robust operation condition, as profits cannot be guaranteed with confidence of 99% at the conditions presented before. Comparing the results obtained through maximization of L and $L_{99\%}$, the second set indicates that smaller reactors and feed rates are advised, given the parameter uncertainties.

At the optimum conditions, the cost of parameter uncertainties may then be calculated with eqs (17)–(19). The value obtained is US$ 29×10^6, which means that average losses of 30 million dollars may be expected by not placing the operation conditions at the actual unknown optimum values. This is a small fortune and indicates that 10% of the average profits that may be obtained with the reactor operation may be lost, due to the parameter uncertainties.

In this case, it seems fair to analyze the possibility of evaluating the kinetic parameters more precisely in a research laboratory before designing the final reactor operation conditions. In order to do that, it is assumed here that the financial market pays 12% of bonus per year for money deposits. The financial losses are then computed by calculating the amount of money that is lost by not investing the profits in the financial market during the plant lifetime, due to delaying the building of the reaction plant. In this case, assuming that parameters with variances equal to zero may be obtained in the laboratory (which is not true), the entrepreneur would have a maximum time of approximately two months to obtain the kinetic constants in the laboratory, as shown in Fig. 4. That means that a very well defined time horizon can be established to drive the experimental investigation and improve the process knowledge.

These computations can be made more precise by including the costs of carrying out experiments and considering that parameter variances cannot be made equal to zero. In both cases, however, the time horizon will be even smaller, which means that going on with the reactor design and plant building is probably the best solution in this case, in spite of the cost of parameter uncertainties.

Example 2: A sequential experimental design procedure

A solution of two isomers (A and B) in a natural solvent (S_1) can be obtained as the extract of a certain bio-process. Component B presents optical activity and can be used in many applications, so that its economical value is much higher than the value of isomer A. A second solvent (S_2) can be used to extract component B from the natural solution, as isomer A does not dissolve very well in S_2. The main objective is designing a single stage extractor for separating isomer B from the natural solution, using solvent S_2. Model equations are:

$$x_{A1} = \frac{x_{A0}}{1 + K_A \phi} \tag{25}$$

$$x_{A2} = \frac{K_A x_{A0}}{1 + K_A \phi} \tag{26}$$

$$x_{B1} = \frac{x_{B0}}{1 + K_B \phi} \tag{27}$$

$$x_{B2} = \frac{K_B x_{B0}}{1 + K_B \phi} \tag{28}$$

where indices 0, 1 and 2 mean feed, solvent 1 and solvent 2, respectively, K_A and K_B are the partition coefficients and ϕ is the ratio between the feed rates of solvent 2 and solvent 1. Therefore, the design variable is ϕ. In order to compute the optimal ϕ value, the following profit function must be maximized:

$$\left(\frac{L}{S_1}\right) = x_{A1}\,\text{US\$}A + x_{B2}\,\text{US\$}B - x_{B1}\,\text{US\$}Q - x_{A2}\,\text{US\$}Q - \phi\,\text{US\$}S \tag{29}$$

where the total profit obtained depends on the purity of the output streams, discounted additional necessary purification (with adsorption columns) and solvent costs. The data available is presented in Table 2. In this problem, $z^T = (\phi)$ and $\alpha^T = (K_A, K_B)$. All other parameters belong to w.

Figure 5 shows how the L and $L_{99\%}$ functions depend on ϕ. As it can be seen, the optimum nominal ϕ value is 1.00, although a higher value of ϕ around 1.20 is recommended in order to increase robustness due to parameter uncertainties. At the nominal maximum, the cost of parameter uncertainties is equal to US\$ 0.14/Kg S_1. If 1 m^3/h of S_1 is processed, average losses induced by parameter uncertainties are about US\$ 1.2×10^6 per year. Therefore, an experimental design procedure is to be implemented, in order to improve the accuracy of the partition coefficients and

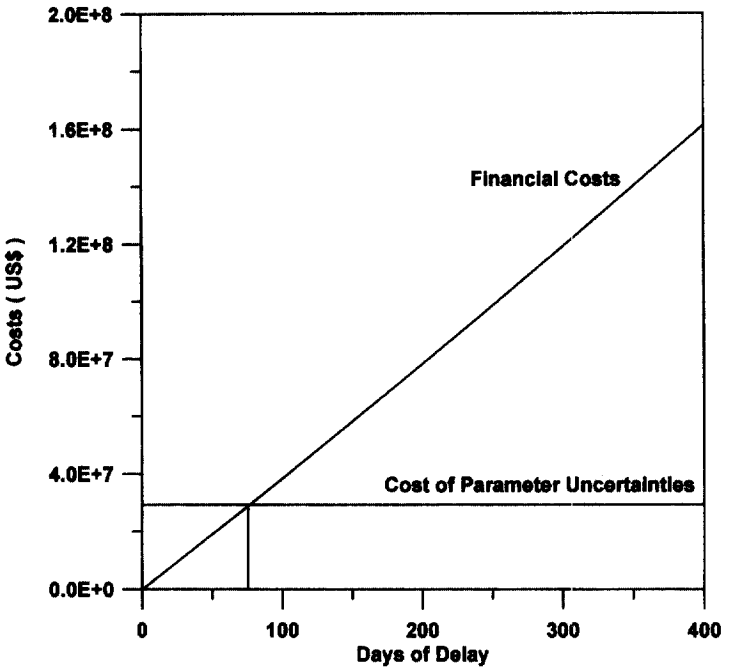

Fig. 4. Parameter and financial costs as functions of project time delay.

Table 2. Available data for example 2

Variable	Value	Meaning
US\$$A$	0.0 US\$/Kg	Market value of A
US\$$B$	20.0 US\$/Kg	Market value of B
US\$$Q$	0.50 US\$/Kg	Additional purification cost
US\$$S$	20×10^6 US\$/$(m^3)^2$	Solvent cost
x_{A0}	0.05	Feed concentration of A
x_{B0}	0.05	Feed concentration of B
K_A	0.10	Partition coefficient of A
K_B	3.0	Partition coefficient of B
σ_{11}^2	0.1	Variance of K_A
σ_{22}^2	1.0	Variance of K_B
σ_{12}^2, σ_{21}^2	0	Covariances of K_A and K_B

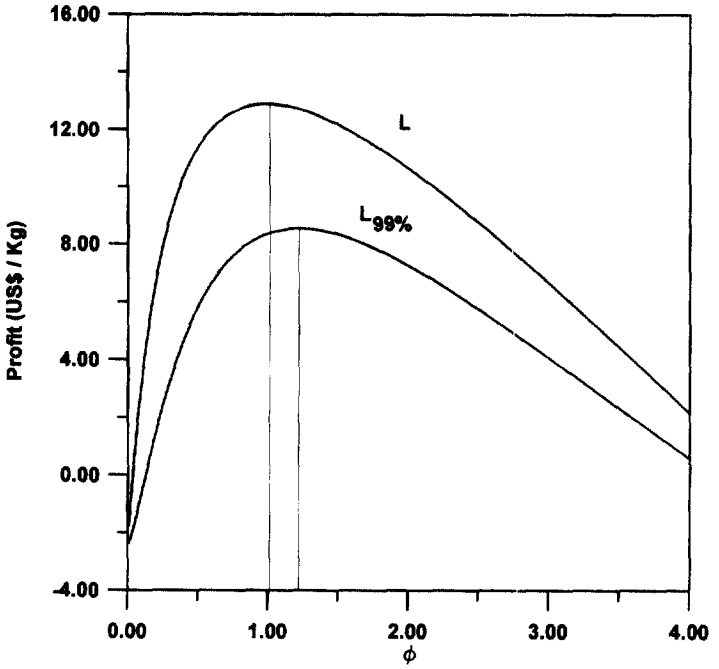

Fig. 5. Maximum nominal and riskless profits as functions of solvent feed ratio.

hopefully diminish the annual losses caused by parameter uncertainties.

Different design criteria may be used in order to improve the accuracy of model parameters (see Pinto *et al.*, 1990,1991). Most experimental design criteria, however, are based on the following equation (see Bard, 1974), which describes how parameter variances are expected to change when an additional experiment is carried out, added to the previous experimental set and used for parameter estimation:

$$V_{\alpha_{k+1}}^{-1} = V_{\alpha_k}^{-1} + B_{\alpha_{k+1}}^{\mathrm{T}} V_{y_{k+1}}^{-1} B_{k+1} \tag{30}$$

where V_{α_k} is the covariance matrix of model parameters after k experiments, $V_{y_{k+1}}$ is the covariance matrix of model output variables at experiment $k+1$ and B_{k+1} is the sensitivity matrix of model output variables at experiment $k+1$, defined as:

$$B_{k+1} = \left[\left. \frac{\partial y_i}{\partial \alpha_j} \right|_{k+1} \right]_{NY \times N\alpha}. \tag{31}$$

Two common experimental design criteria are the β-trace design criterion, which proposes that the best experiment is the one which allows the minimization of the trace of $V_{\alpha_{k+1}}$, and the volume design criterion, which proposes that the best experiment is the one which allows the minimization of the determinant of $V_{\alpha_{k+1}}$. In both cases, the design criteria are based on the idea that the region of parameter uncertainties should be minimized. A full description of these and other experimental design criteria can be found elsewhere (Pinto *et al.*, 1990, 1991). As model parameters will be used for designing the single-stage extractor and as the main objective is reducing the losses caused by parameter uncertainty, we propose here that minimizing the cost function described by eq. (17) be used as the proper experimental design criterion for experimentation and parameter estimation.

In order to choose the best experimental condition for experimentation, additional data are required. First of all, as the sum of the fractions of isomers A and B is equal to 0.1 in the original solution, it is proposed here that the total fraction of A and B in S_1 in the additional experiment be equal to 0.1. Therefore, the experimental grid analysed is composed by synthetic solutions where $(x_{A0} + x_{B0})$ is equal to 0.1. Besides, it is important to say that A and B fractions may be measured experimentally with a precision of $\sigma_y^2 = 1 \times 10^{-4}$. Finally, as the optimum solution seems to be around $\phi = 1.00$, this is the value that will be used in all experiments analysed. Therefore, defining the best experiment for additional experimentation is equivalent to defining the best value of x_{A0}.

Figures 6–8 show how $tr(V_{\alpha_{k+1}})$, $\det(V_{\alpha_{k+1}})$ and C depend on the value of x_{A0} selected. The β-trace design criterion selects the condition where x_{A0} is equal to zero, mostly because the variance of K_B, which is the most uncertain parameter, may be decreased significantly at such conditions (see Fig. 10). The volume design criterion selects the condition where x_{B0} is equal to zero, because the variance of K_A, which has the smallest variance, can be decreased even further at such conditions (see Fig. 9). From an economical point of view, however, the best choice seems to be the experiment where x_{A0} is equal to 0.032, which allows a significant decrease of both parameter variance and the largest decrease of the cost of parameter uncertainties.

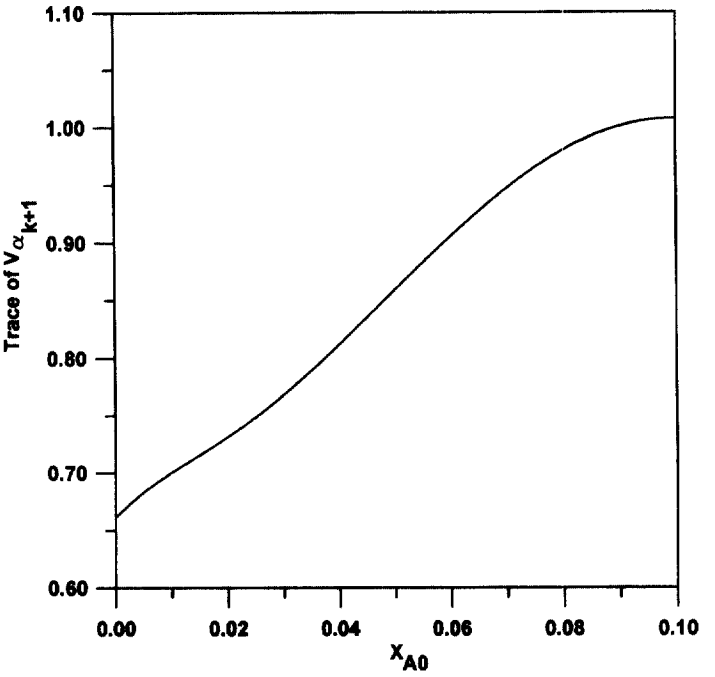

Fig. 6. Using the β-trace design criterion to select the next experiment.

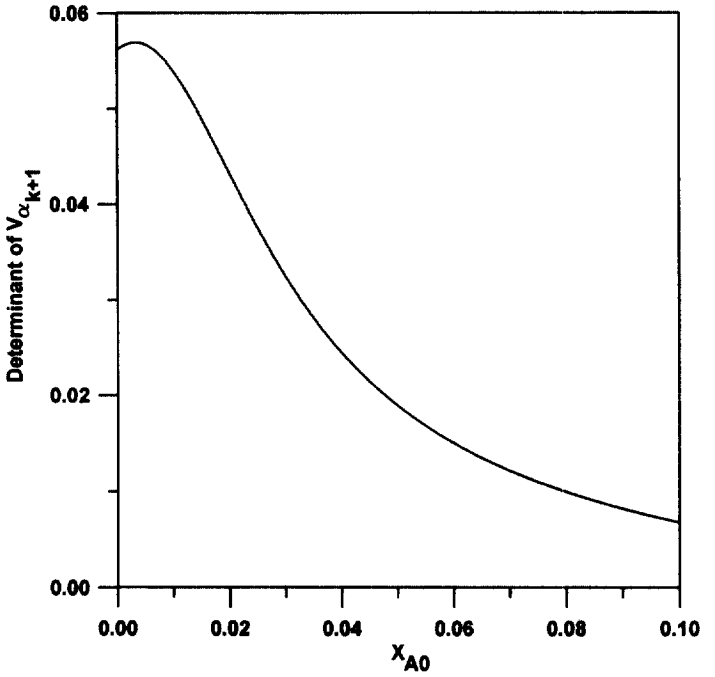

Fig. 7. Using the volume design criterion to select the next experiment.

According to the usual sequential experimental design procedure, the experiment with $x_{A0} = 0.032$ and $x_{B0} = 0.068$ should be carried out and a new set of parameter estimates and variances should be provided. Assuming that the parameter estimates will remain the same and that the new parameter variances will be given by eq. (30), the costs induced by parameter uncertainties will be reduced to US$ 0.73×10^6 per year, leading to additional savings of US$ 0.47×10^6 per year. If the β-trace design criterion

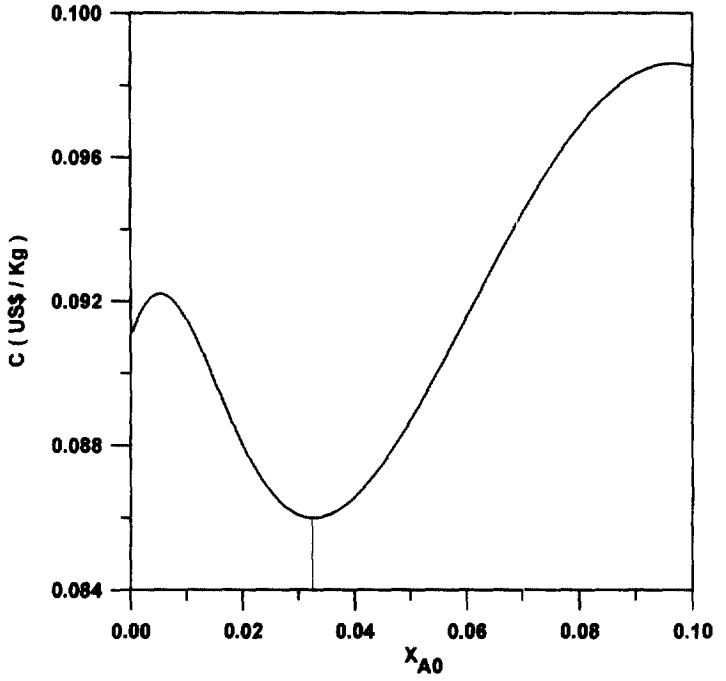

Fig. 8. Using the cost of parameter uncertainties to select the next experiment.

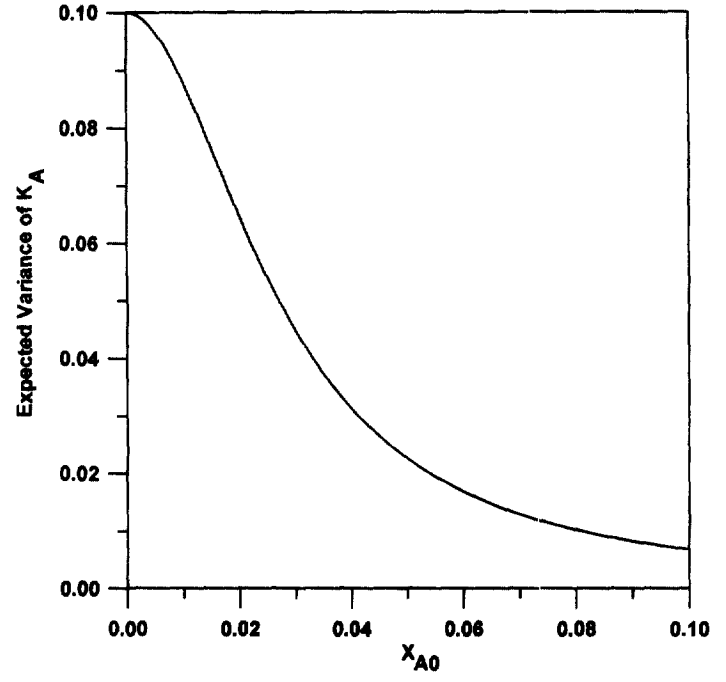

Fig. 9. Expected variance of K_A as a function of the selected experimental condition.

is used, annual savings will be of US$ 0.42×10^6 per year, while if the volume design criterion is used, annual savings will be of US$ 0.36×10^6. Therefore, choosing the right experiment in this case is worth at least US$ 50000,00 a year.

Additional remarks

It is clear that the equations developed and presented here may be used very successfully while designing process conditions and taking decisions, specially those related to the design of experiments. More

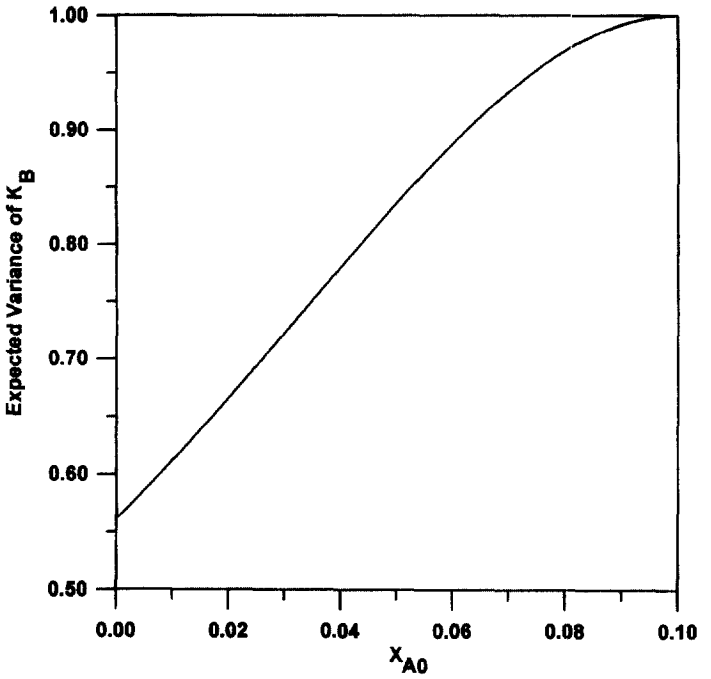

Fig. 10. Expected variance of K_B as a function of the selected experimental condition.

important, the equations developed and the examples presented provide a clear link between the process economics, the process modeling and experimentation and parameter estimation. This is very interesting because uncertainties of parameter estimates are usually much more significant than the uncertainties of the other process variables. However, it is important to say that design variables are not alike, so that it may be useful to separate them in different groups based on the easiness of manipulation at actual process conditions, as proposed originally by Taguchi (see Taguchi, 1986). For instance, in Example 1 V and Θ present very different characteristics, as Θ may be manipulated at the actual plant site according to an EVOP—evolutionary operation procedure—technique (see Himmelblau, 1970), which means that errors may be corrected as time goes on. This is not possible with V, unless another reactor is bought and installed, which means that design errors are difficult to correct. The theoretical framework is being improved in order to allow the inclusion of EVOP and other stochastic optimization techniques in the analysis.

4. CONCLUSIONS

A theoretical framework was developed in order to allow the analysis of economical losses introduced in design problems by parameter uncertainties. It was shown here that an economical value may be assigned to parameter uncertainties, which can then be used for both process optimization and specially for taking decisions during sequential experimental designs. The examples show that taking parameter uncertainties into consideration during the design stage can improve the robustness of process economics and provide a link to process modeling and experimentation for parameter estimation.

Acknowledgments

We thank CNPq - Conselho Nacional de Desenvolvimento Científico e Tecnológico -, Brazil, for supporting this research.

NOTATION

A, B, C	chemical species
C	cost of uncertainties
C_i	concentration of species i
K_i	model parameter (kinetic constant of reaction i, partition coefficient of species i)
L	profit function
$L_{99\%}$	riskless profit function
R	risk of the enterprise
S_1, S_2	solvents, solvent feed rates
US$	value
V	volume
V_y	covariance matrix of model output variables
V_α	covariance matrix of parameters
x	mass fraction
w	vector of fixed variables
y	model output variables
z	vector of manipulated (decision) variables

Greek letters

α	vector of model parameters
Δ	deterministic variable deviation

$\overline{\Delta L}$ bias of the profit function
Δt lifetime of the enterprise
ε random variable deviation
ϕ ratio of solvent feed rates
σ_{ij}^2 covariance of variables i and j
Θ residence time

Superscript
* nominal condition
Subscript
0 feed

REFERENCES

Bard, Y. (1974) *Nonlinear Parameter Estimation.* Academic Press, New York, U.S.A.

Becker, R., Hall, S. and Rustem, B. (1994) *J. Econom. Dynam. Control* **18**, 125–147.

Coleman, D. E. and Montgomery, D.C. (1993) *Technometrics*, **35**, 1–24.

Fotopoulos, J., Georgakis, C. and Stenger Jr., H. G. (1994) *Chem. Engng Sci.* **49**, 5533–5547.

Ganesh, N. and Biegler, L. T. (1987) *A.I.Ch.E J.* **33**, 282–296.

Himmelblau, D. M. (1970) *Process Analysis by Statistical Methods.* John Wiley & Sons, New York, U.S.A.

Keel, L. H. and Bhattacharyya, S. P. (1994) *Int. J. Rob. Nonl. Control* **4**, 87–100.

Lucas, J. M. (1994) *J. Qual. Technol.* **26**, 248–260.

Oldak, S., Baril, C. and Gutman, P. O. (1994) *Int. J. Rob. Nonl. Control* **4**, 101–117.

Pinto, J. C., Lobão, M. W. and Monteiro, J. L. (1990) *Chem. Engng Sci.* **45**, 883–892.

Pinto, J. C., Lobão, M. W. and Monteiro, J. L. (1991) *Chem. Engng Sci.* **46**, 3129–3138.

Pistikopoulos, E. N. (1995) *Comp. Chem. Engng* **19**, S553–S563.

Reed, M. E. and Whiting, W. B. (1993) *Chem. Engng Commun.* **124**, 39–48.

Russell, T. W. F. and Bogaert, R. J. (1980) *Ind. Engng Chem. Process Des. Dev.* **19**, 282–289.

Schaper, C. D., Seborg, D. E. and Mellichamp, D. A. (1992) *Ind. Engng Chem. Res.* **31**, 1694–1704.

Seferlis, P. and Hrymak, A. N. (1996) *Comput. Chem. Engng* **20**, 1177–1200.

Shoemaker, A. C., Tsui, K.-L. and Jeff Wu, C. F. (1991) *Technometrics* **33**, 415–427.

Taguchi, G. (1986) *Introduction to Quality Engineering: Designing Quality Into Products and Processes.* Asian Productivity Organization, Tokyo.

Volin, Y. M. and Ostrovskiy, G. M. (1981a) *Comp. Chem. Engng* **5**, 21–30.

Volin, Y. M. and Ostrovskiy, G. M. (1981b) *Comput. Chem. Engng* **5**, 31–40.

Weisman, J. and Holzman, A. G. (1972) *Ind. Engng Chem. Process. Des. Dev.* **3**, 386–397.

APPENDIX A

If eq. (2) is subtracted from eq. (13) before averaging, it is possible to write:

$$\Delta L^* - \Delta L = (\nabla_z L)^T \Delta z^* + \frac{1}{2}\left[\begin{array}{l} 2(\varepsilon_z)^T (\nabla^2_{z,z} L)\,\Delta z^* + (\Delta z^*)^T (\nabla^2_{z,z} L)\,\Delta z^* \\ + 2(\Delta z^*)^T (\nabla^2_{z,w} L)\,\varepsilon_w + 2(\Delta z^*)^T (\nabla^2_{z,\alpha} L)\varepsilon_\alpha \end{array}\right]. \tag{A1}$$

It may be noted that the first two terms on the right-hand side of eq. (A1) will be nullified by the averaging operation, as they contain first-order terms and uncorrelated uncertainties. Therefore, by neglecting these two terms and inserting eq. (12) into eq. (A1), it is possible to write

$$E\{\Delta L^* - \Delta L\} = \frac{1}{2} E\left\{\left[\begin{array}{l} (\varepsilon_w)^T (\nabla^2_{z,w} L)^T (\nabla^2_{z,z} L)^{-1} (\nabla^2_{z,w} L)\,\varepsilon_w \\ + (\varepsilon_\alpha)^T (\nabla^2_{z,\alpha} L)^T (\nabla^2_{z,z} L)^{-1} (\nabla^2_{z,\alpha} L)\,\varepsilon_\alpha \\ - 2(\varepsilon_w)^T (\nabla^2_{z,w} L)^T (\nabla^2_{z,z} L)^{-1} (\nabla^2_{z,w} L)\,\varepsilon_w \\ - 2(\varepsilon_\alpha)^T (\nabla^2_{z,\alpha} L)^T (\nabla^2_{z,z} L)^{-1} (\nabla^2_{z,\alpha} L)\,\varepsilon_\alpha \end{array}\right]\right\} \tag{A2}$$

where $E\{\ \}$ stands for the averaging. After summing up the terms on the right hand-side of eq. (A2),

$$E\{\Delta L^* - \Delta L\} = -\frac{1}{2} E\left\{\left[\begin{array}{l} (\varepsilon_w)^T (\nabla^2_{z,w} L)^T (\nabla^2_{z,z} L)^{-1} (\nabla^2_{z,w} L)\varepsilon_w \\ + (\varepsilon_\alpha)^T (\nabla^2_{z,\alpha} L)^T (\nabla^2_{z,z} L)^{-1} (\nabla^2_{z,\alpha} L)\,\varepsilon_\alpha \end{array}\right]\right\}. \tag{A3}$$

As the kernel $(\nabla^2_{z,z} L)$ is negative definite (otherwise it is not possible to find an optimum for the problem), the terms inside the brackets on the right-hand side are guaranteed to be negative. Therefore, it may be concluded that C, as presented in eq. (17), is always positive, which means that a certain amount of money is always lost due to the parameter uncertainties.