PARAMETER ESTIMATION IN MULTIRESPONSE MODELS*

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I. General

In the last decade, many efforts have been devoted to the modelling of the kinetics of heterogeneous chemical reaction systems. Methods have been developed for rigorous parameter estimation, for *a posteriori* discrimination between rival models, for experimental designs of the sequential type aiming at optimal discrimination among rival models, and for sequential designs aiming at obtaining precise estimates of the parameters of the mathematical model. An extensive survey of important contributions in this field has recently been given by KITTRELL (1971).

Application of these advances in mathematical modelling have been restricted almost always to systems consisting of a single dependent variable. In general, however, industrial processes include several response variables.

To obtain maximum information from the system, advantage should be taken of its multi-response character. Several criteria have been suggested in the literature to estimate parameters in multi-response models. In the following paragraph a short survey of the most important methods is presented.

II. Methods for parameter estimation in multi-response models

Let the general representation of the mathematical model be given by the set of v equations, which may be linear or non linear in the parameters:

$$\begin{aligned} y_1 &= f_1(\mathbf{X}, \boldsymbol{\beta}) + \varepsilon_1 = \eta_1 + \varepsilon_1 \\ y_2 &= f_2(\mathbf{X}, \boldsymbol{\beta}) + \varepsilon_2 = \eta_2 + \varepsilon_2 \\ &\vdots \qquad\qquad\qquad \vdots \\ y_v &= f_v(\mathbf{X}, \boldsymbol{\beta}) + \varepsilon_v = \eta_v + \varepsilon_v \end{aligned}$$

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Some criteria which have been used to obtain estimates of the parameters involve the correlation coefficients between the experimentally observed values of the dependent variables and the values predicted by the mathematical model. The correlation coefficient between these quantities, e.g. for the j-th response, is defined as follows:

$$\varrho_j = \frac{\sum_{i=1}^{n} (y_{ij} - \bar{y}_j)(\hat{y}_{ij} - \bar{\hat{y}}_j)}{\sqrt{\sum (y_{ij} - \bar{y}_j)^2 \sum (\hat{y}_{ij} - \bar{\hat{y}}_j)^2}}$$

The $\hat{y}$ values are functions of the unknown parameters. It has been suggested to seek the parameter values which

1. maximize the square of the *smallest* correlation coefficient,
2. maximize the square of the *largest* correlation coefficient,
3. maximize the *sum of the squares* of the v correlation coefficients,
4. maximize the *square of the product* of the v correlation coefficients.

Criterion No. 3 gives the best average multiple correlation coefficient since it weights all the correlation coefficients on an equal basis.

The maximum probability concept which has been shown to be of great value in the case of single response mathematical models, may be extended to multi-response process models.

The observations of the response variables, which are experimental measurements of the v responses at n different settings of the independent controllable variables, can be classified into an $(n \ast v)$ matrix where y_{ij} represents the element in the i-th row and j-th column. Similarly, the experimental errors associated with these $n \ast v$ observations can be grouped into an analog matrix, shown below:

$$\begin{array}{lccccc}
 & \text{Response No.}\ 1 & 2 \ldots\ldots & r \ldots\ldots & v \\
\text{Experiment No.} & & & & \\
1 & \varepsilon_{11} & \varepsilon_{12} \ldots & \varepsilon_{1r} \ldots & \varepsilon_{1v} \\
2 & \varepsilon_{21} & \varepsilon_{22} \ldots & \varepsilon_{2r} \ldots & \varepsilon_{2v} \\
3 & \varepsilon_{31} & \varepsilon_{32} \ldots & \varepsilon_{3r} \ldots & \varepsilon_{3v} \\
\cdot & \cdot & \cdot & \cdot & \cdot \\
\cdot & \cdot & \cdot & \cdot & \cdot \\
\cdot & \cdot & \cdot & \cdot & \cdot \\
n & \varepsilon_{n1} & \varepsilon_{n2} \ldots & \varepsilon_{nr} \ldots & \varepsilon_{nv}
\end{array}$$

The following assumptions are made:

1. the n errors belonging to the r-th response are normally and independently distributed with zero mean and constant variance σ_{rr};

2. the errors within one experiment and associated with the different responses of that experiment are statistically dependent with variance–covariance matrix the symmetric $v \ast v$ matrix $\mathbf{\Lambda}$:

$$\mathbf{\Lambda} = \begin{bmatrix} \sigma_{11} & \sigma_{12} \ldots & \sigma_{1v} \\ \sigma_{12} & \sigma_{22} \ldots & \sigma_{2v} \\ \cdot & \cdot & \cdot \\ \cdot & \cdot & \cdot \\ \cdot & \cdot & \cdot \\ \sigma_{1v} & \sigma_{2v} \ldots & \sigma_{vv} \end{bmatrix}$$

3. the errors associated with measurements of different responses and having unequal experiment number are uncorrelated.

The probability function of the parameters then has the form HUNTER [9]:

$$\mathrm{L}\,(\boldsymbol{\beta} \,|\, \mathbf{y}_1, \mathbf{y}_2, \ldots, \mathbf{y}_v; \mathbf{X}; \mathbf{\Lambda}) = \frac{1}{(\sqrt{2\pi})^{nv} (\det \mathbf{\Lambda})^{\frac{n}{2}}} \exp \left\{ -\frac{1}{2} \sum_{i=1}^{n} [\mathbf{y}^{(i)} - \boldsymbol{\eta}^{(i)}]^T \mathbf{\Lambda}^{-1} \times \right.$$
$$\left. \times [\mathbf{y}^{(i)} - \boldsymbol{\eta}^{(i)}] \right\}$$

or equivalently

$$L = \frac{1}{(\sqrt{2\pi})^{nv} (\det \mathbf{\Lambda})^{\frac{n}{2}}} \exp. \left\{ -\frac{1}{2} \sum_{r=1}^{v} \sum_{s=1}^{v} \sigma^{rs} \left[\sum_{i=1}^{n} (y_{ir} - \eta_{ir})(y_{is} - \eta_{is}) \right] \right\}$$

where σ^{rs} is the appropriate element of $\mathbf{\Lambda}^{-1}$ and η the predicted value of the response variable.

The maximum probability parameter estimates are thus obtained by minimizing

$$\sum_{r=1}^{v} \sum_{s=1}^{v} \sigma^{rs} \sum_{i=1}^{n} (y_{ir} - \eta_{ir})(y_{is} - \eta_{is})$$

From this expression two frequently encountered criteria for estimating parameters in multi-response problems are readily derived:

1. The responses are uncorrelated but have unequal variances. This means that $\sigma^{rs} = 0$ for all r and s with $r \neq s$, so that the matrix $\mathbf{\Lambda}$ and also its inverse $\mathbf{\Lambda}^{-1}$ are reduced to diagonal matrices. The expression which has to be minimized simplifies to a sum of weighted sums of squares:

$$\sum_{r=1}^{v} \sigma^{rr} \sum_{i=1}^{n} (y_{ir} - \eta_{ir})^2$$

2. The responses are uncorrelated and have equal variance. The expression to be minimized further reduces to

$$\sum_{r=1}^{v} \sum_{i=1}^{n} (y_{ir} - \eta_{ir})^2$$

which is nothing but a straight-forward extension of the least squares principle.

It is generally not allowed to assume uncorrelated responses and/or equal variances for all the responses, so that the full expression should be minimized. However, a difficulty arises hereby, since in many cases the matrix Λ and its inverse Λ^{-1} are unknown. Estimates of the σ^{rs} elements may be obtained from replicated experiments. From a practical point of view however, these may be difficult to perform or turn out to be too expensive. A way out of this impasse is to replace the unknown variance–covariance matrix Λ by its maximum probability estimate. Box and Draper [1] showed that the maximum probability estimates of the parameters are then obtained by the minimization of

$$\det \begin{bmatrix} \Sigma(y_1 - \eta_1)^2 & \Sigma(y_1 - \eta_1)(y_2 - \eta_2) \ldots & \Sigma(y_1 - \eta_1)(y_v - \eta_v) \\ \Sigma(y_1 - \eta_1)(y_2 - \eta_2) & \Sigma(y_2 - \eta_2)^2 \ldots & \Sigma(y_2 - \eta_2)(y_v - \eta_v) \\ \cdot & \cdot & \cdot \\ \cdot & \cdot & \cdot \\ \cdot & \cdot & \cdot \\ \Sigma(y_1 - \eta_1)(y_v - \eta_v) & \Sigma(y_2 - \eta_2)(y_v - \eta_v) \ldots & \Sigma(y_v - \eta_v)^2 \end{bmatrix}$$

with respect to the parameters.

Minimization of the above determinant requires search techniques, even when the mathematical equations for all the responses are linear in the parameters.

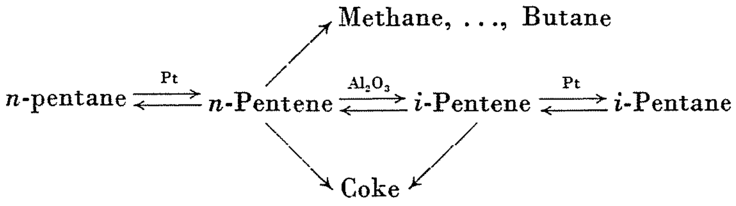
Attention should be given to the fact that the matrix whose determinant is minimized, must not be singular or nearly singular. Singularities arise from linearly dependent responses as encountered e.g. when a so-called 'observed' value is obtained from a mole fraction balance, or when the 'observed' value for one response is nothing but a constant percentage of the truly experimental value of another response. Empirical methods exist to trace such dependences (Box and Hunter [2]).

In practice, it is often not known whether the responses are statistically independent or not. It is therefore recommended to try the determinant criterion first. Only a very limited number of papers has dealt with the application of the determinant criterion to multi-response data. Mezaki and Butt [12] analyzed experimental data concerning the dehydration of ethylalcohol by means of the determinant criterion. In a paper partly dedicated to the use of the determinant criterion (Eakman, [5]), simulated data were employed to compare the performance of this criterion with that of the trivial extension of the least squares principle. The results are, however, of little value, since some basic properties of the error which must be fulfilled in both methods were violated (Erjavec [6]). Finally, Box and (Hunter [2]) recently re-analyzed the data of Fuguitt and Hawkins (1947) concerning the thermal isomerization of α-pinene and presented a method to detect dependences which may cause the determinant to be singular.

III. Estimation of parameters in the complex chemical reaction system: pentane isomerization with catalyst decay

A problem of this kind has been encountered in our laboratory where the kinetics of the isomerization of *n*-pentane on a commercial bifunctional catalyst has been studied.

On the basis of published and own experimental data (Lambrecht, Nussey and Froment [11]); (DePauw and Froment [4]), a tentative mechanism for this complex process has been set up



The deactivation problem has been attacked along the lines suggested by Froment and Bischoff [7], i.e. the coke content on the catalyst has been related to the process variables, such as partial pressures of reactants and reaction products, amount of reactant fed, weight of catalyst etc. ... instead of time as is frequently done. Time is evidently not a true variable from the point of view of the mechanism of coke formation.

A typical set of experimental results (DePauw, Ph. D. Thesis, Gent[3]), is shown in Fig. 1.

The gas samples are taken at several points along the axis of the reactor at time intervals of one hour. The total duration of an experiment is 11 to 12 hours.

At the end of the experiment, the catalyst is unloaded from the reactor and divided in several sections. The coke is burnt off from each section. The difference in weight before and after the burning off operation yields the carbon profile along the bed at the end of the experiment. Fig. 1 shows such

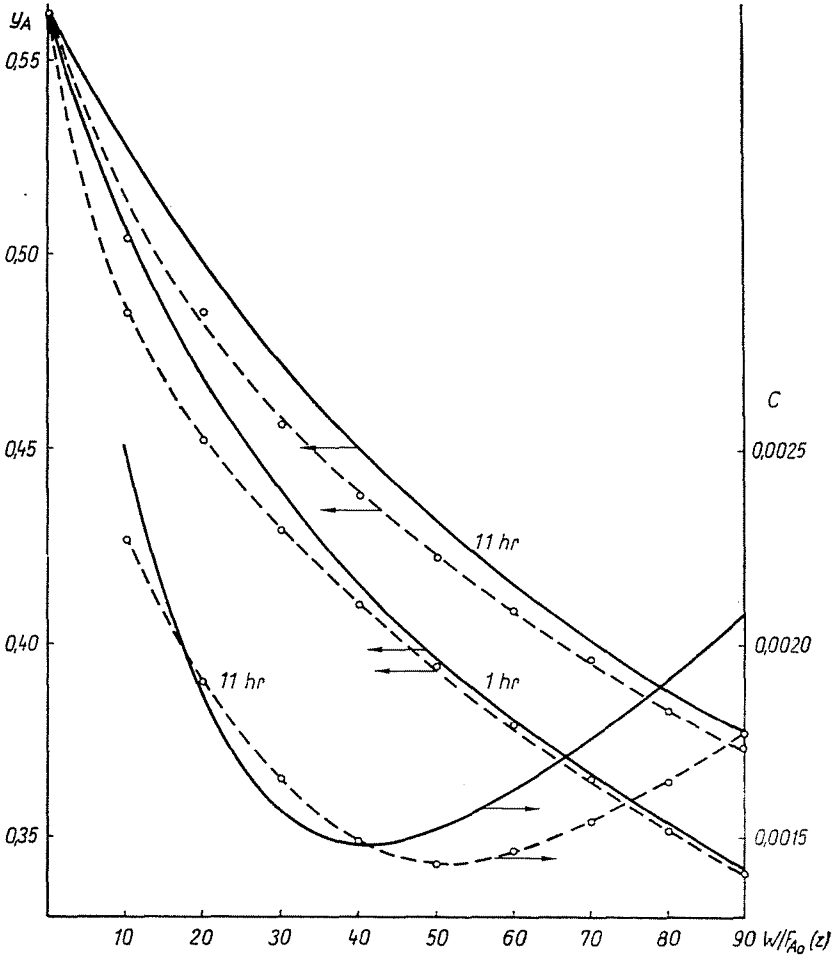


Fig. 1. Experimental (○) and calculated (———) results

an experimental carbon profile. This profile exhibits a descending as well as an ascending part. In terms of the theory set up by FROMENT and BISCHOFF [7] the coke may be considered as deposited by both a parallel and a consecutive mechanism.

In order to describe the process, a mathematical model was set up taking into account the multiresponse character of the system. The reaction rate equations for the three responses were chosen as follows:

Isomerization (main reaction) (HOSTEN and FROMENT [8])

$$r_I = \frac{k_I(y_A - y_B/K)}{y_{H_2} + K_B y_B}$$

Hydrocracking (parallel side reaction)

$$r_H = k_H\,(y_A/y_{H_2})^{n_1}$$

Coke formation (parallel and consecutive secondary reactions)

$$r_C = \frac{k_{CP}\,(y_A/y_{H_2})^{n_2}}{1 + K_B\,y_B/y_{H_2}} + \frac{k_{CC}\,(y_B/y_{H_2})^{n_2}}{K_B + y_{H_2}/y_B}$$

To account for the deactivation of the catalyst, the rate coefficients in the three reaction rate equations contain a deactivation function which is only dependent upon the coke content of the catalyst

$$\begin{aligned} k_I &= k_I^\circ\ \varnothing_1 \\ k_H &= k_H^\circ\ \varnothing_2 \\ k_{CP} &= k_{CP}^\circ\ \varnothing_3 \\ k_{CC} &= k_{CC}^\circ\ \varnothing_4 \end{aligned}$$

From separate experiments, k_{CP} was shown to vary exponentially with the coke content. To limit the complexity, all $\varnothing_i$ were assumed to be identical and of the exponential type, so that

$$\varnothing_1 = \varnothing_2 = \varnothing_3 = \varnothing_4 = e^{-\alpha C}$$

With these simplifications the mathematical model for the process contains seven parameters: four rate constants, two reaction orders and one deactivation parameter. These parameters were estimated by means of the determinant criterion discussed in the preceding paragraph. To limit the computations, the differential method of kinetic analysis, involving reaction rates instead of conversions, had to be adopted, so that the appropriate determinant to be minimized was

$$\det \begin{vmatrix} \Sigma(r_I - \hat{r}_I)^2 & \Sigma(r_I - \hat{r}_I)\ (r_H - \hat{r}_H) & \Sigma(r_I - \hat{r}_I)\ (r_C - \hat{r}_C) \\ \Sigma(r_I - \hat{r}_I)\,(r_H - \hat{r}_H) & \Sigma(r_H - \hat{r}_H)^2 & \Sigma(r_H - \hat{r}_H)\,(r_C - \hat{r}_C) \\ \Sigma(r_I - \hat{r}_I)\,(r_C - \hat{r}_C) & \Sigma(r_H - \hat{r}_H)\,(r_C - \hat{r}_C) & \Sigma(r_C - \hat{r}_C)^2 \end{vmatrix}$$

The rates were not observed directly: they were derived from the experimentally measured mole fractions or coke content.

The isomerization reaction rates were obtained by means of a method similar to that proposed by KITTRELL and MEZAKI [10], involving a fit of some logarithmic function of the conversion by means of a truncated polynomial in $\frac{W}{F_{A_0}}$ and subsequent analytical differentiation.

The hydrocracking reaction rates were obtained by direct numerical differentiation of the experimental hydrocracking conversion vs. W/F_{A_0} curves.

A difficulty is encountered in finding the carbon formation reaction rates. This is due to the fact that only one coke profile corresponding to the entire reaction period is available and all intermediate contents, necessary for the derivation with respect to time, are missing. This seriously complicates the data analysis since an iterative procedure has now to be adopted. In the first stage, the intermediate coke profiles are estimated with the aid of a Voorhies-type rule for the coke content:

$$C = \Psi(z)\sqrt{t}$$

where the function $\Psi(z)$ is defined by the final profile. These profiles have to be improved by iteration. The procedure is as follows. From the preliminary estimated coke contents, first estimates of the carbon formation reaction rates are obtained by numerical differentiation. Minimization of the determinant, an iterative procedure in itself, is then started for parameter estimation. However, only a limited number of parameter improvements is allowed because of the temporary character of the current estimates of the carbon formation reaction rates. With the current parameter estimates, an integration of the model equations is performed. The mathematical model of the process consists of the

continuity equation for n-pentane:

$$\frac{\delta y_A}{\delta t} + \frac{\delta y_A}{\delta z} = -\frac{\varrho_B \Omega d_p}{F_t \gamma} r_I ,$$

continuity equation for hydrocracking products:

$$\frac{\delta y_H}{\delta t} + \frac{\delta y_H}{\delta z} = \frac{\varrho_B \Omega d_p}{F_t} r_H ,$$

continuity equation for carbon on the catalyst:

$$\frac{\delta C}{\delta t} = \frac{\varepsilon \Omega d_p p_t}{F_t} r_C .$$

The integration of this set of partial differential equations is carried out numerically along the characteristics z and $t - z$. This integration yields predicted carbon profiles at the end of the run as well as at all intermediate times. The final integrated carbon profile is shifted to correspond to the experimentally measured profile. This shift yields a scaling function $\varphi(z)$ with the aid of which all intermediate predicted coke profiles are corrected. The obtained corrected coke profiles are now considered as improved estimates of the unknown experimental profiles. The operations described constitute one cycle of the iterative procedure. Program control is then transferred to the numerical differentiation procedure to yield improved estimates of the experimental carbon formation reaction rates. The procedure is continued until stable solutions are obtained.

The search procedure for minimizing the appropriate determinant was ROSENBROCK's method. The number of parameter improvements within one of the described cycles was arbitrarily limited to 99.

In Fig. 2 a typical behaviour of the parameters is shown as a function of the number of iteration cycles. In early stages of the procedure, large oscillations in the parameter estimates are observed. A fairly large number of these iterations is required to level out the oscillations.

In Fig. 1 experimental results and model predictions are compared for a typical experiment. Both calculated curves, predicting the n-pentane mole fraction, lie entirely above the experimental ones. This should not be misinterpreted, however. The systematic deviations are due to the fact that the parameter estimation criterion actually involved the rates, rather than the experimentally observed mole fractions. No abnormal behaviour is observed when predicted and experimental rates are compared, as is done in Fig. 3. The largest deviation between experimentally observed and predicted mole fractions of n-pentane does not exceed 5%. The agreement may be considered as excellent. The largest deviation between experimentally measured and predicted coke contents amounts to 18%. The average error is 9.6% which is indeed the accuracy with which the carbon content of the catalyst is measured. The overall agreement is satisfactory.

It is interesting to compare the results obtained by the determinant criterion with those obtained by a more commonly applied procedure, such as the minimization of a sum of weighted sums of squares. For this purpose the following quantity was minimized

$$F = \Sigma(r_I - \hat{r}_I)^2 + 0.2\Sigma(r_H - \hat{r}_H)^2 + 20\Sigma(r_C - \hat{r}_C)^2$$

where the weights were chosen on the basis of common sense. The procedure was started with the initial parameter estimates and coke profiles used in the procedure based upon the determinant criterion. It was observed that the oscillations persisted for a much larger number of iterations. A comparison of both sets of parameter estimates is made in columns 2 and 3 of Table 1. Rather large deviations are seen to exist among both estimates for the parameters

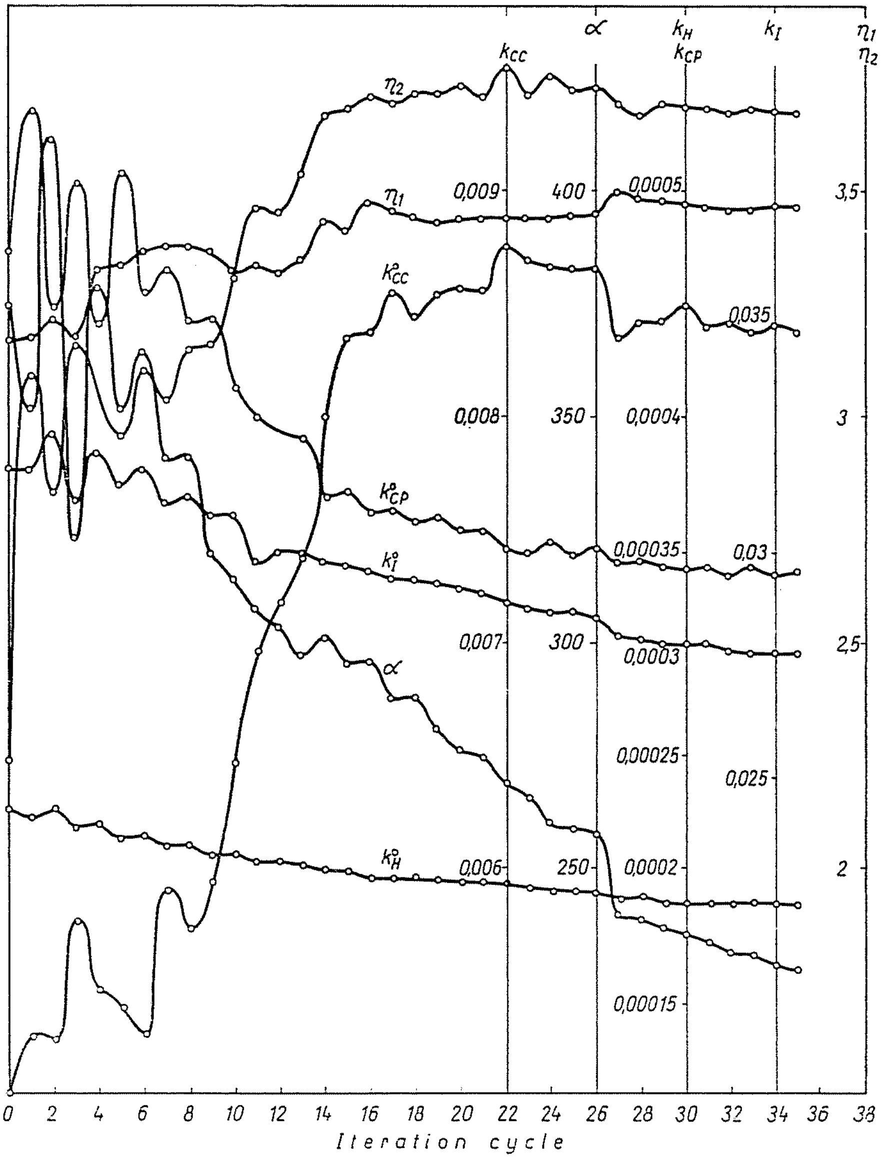


Fig. 2. Behaviour of parameter estimates as a function of the number of iteration cycles

which appear in the carbon formation reaction rate. The deactivation parameter α is also subject to some variation. These discrepancies might be explained to a great extent by the fact that the latter criterion does not account for the correlation between the different responses. Table 2 shows the maximum probability estimates of the correlation coefficients between responses. The

Table 1

Parameter estimates and confidence intervals at the 0.95 probability level

Parameter	Sum of squares criterion	Determinant criterion	Confidence intervals for the parameters from determinant criterion
k_I°	0.0268	0.0278	± 0.00079
α	197	228	± 20.3
n_2	2.46	3.67	± 0.17
k_H°	0.000172	0.000184	± 0.0000052
k_{CP}°	0.000434	0.000333	± 0.000026
k_{CC}°	0.00336	0.00836	± 0.00058
n_1	3.64	3.46	± 0.11

Table 2

Maximum probability estimates of correlation coefficients between the responses

	Isomerization	Hydrocracking	Carbon formation
Isomerization	1	0.419	0.938
Hydrocracking		1	0.183
Carbon formation			1

carbon formation reaction rate seems to be highly correlated with the isomerization reaction rate. Besides the correlation, also the magnitude of the weighting factors in the least squares criterion affects the parameter estimates. The parameters related to the main reaction and the hydrocracking are fairly insensitive with respect to the estimation criterion. For k_I°, this is not surprising since the isomerization rates vary over the widest range and are accurately determined, so that almost any criterion would yield comparable estimates.

Any analysis of this type should be accompanied by a statistical analysis of the reliability of the estimated parameters to test if the model does not contain any superfluous parameters. If it does, these parameters should be deleted since they do not significantly contribute to the adequacy of the model, or further experiments may be designed attempting to improve the reliability of the non significant parameters. Such an analysis was performed and the result is shown in the 4th column of Table 1. This column lists the confidence intervals for the parameters corresponding to the 95% probability level. The intervals are seen to be rather narrow. None of them contains the value zero so that all parameter estimates are significantly determined. The results are statistically meaningful. It should be mentioned here that the confidence intervals in Table 1 are individual confidence intervals, i.e. they give the range

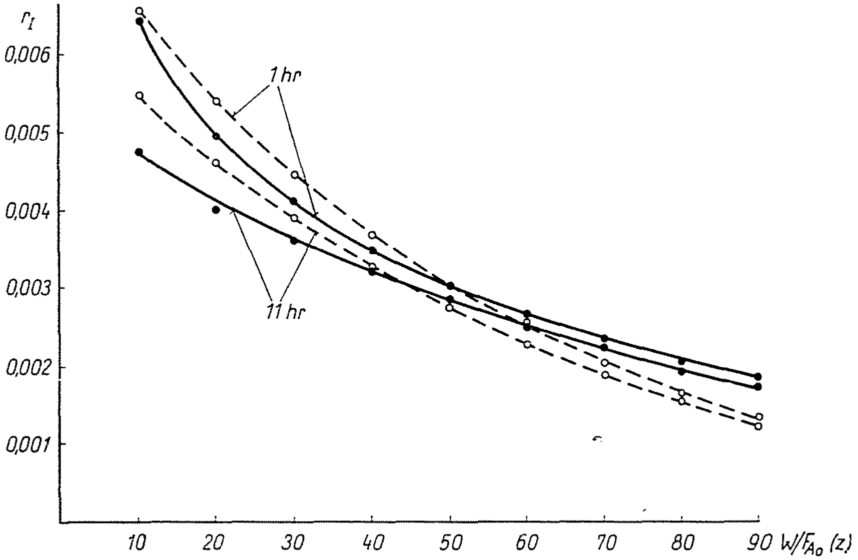


Fig. 3. Experimental (○) and calculated (●) isomerization rates

in which the true but unknown parameter value may be expected to lie 95 out of 100 times when all the remaining parameters are held constant at their optimal value. The construction of the joint confidence region, which accounts for the variability of all parameters simultaneously, is simply out of question here, because of the dimensionality of the problem.

IV. Conclusions

The application of the determinant criterion to the experimental data of a multiresponse problem has been illustrated. The computational labour was found not to increase significantly compared to more commonly used methods which require the choice of the weights of the different responses.

The model contained seven parameters which were significantly estimated as was shown by their individual 95% confidence intervals. The estimates of the parameters related to coke formation were different from those obtained by the least squares approach. This may be explained by the correlation between responses which is taken into account in the determinant criterion and not in the extended least squares method.

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Summary

The paper deals with the estimation of parameters in a multiresponse reaction system, namely the isomerization of n-pentane, accompanied by hydrocracking and coke deposition. The parameters are estimated by the determinant criterion developed by Box and Draper which takes into account the unknown correlation between the responses. A comparison of these estimates with those obtained from a less rigorous extension of the classical least squares principle is made. Significant differences between both sets of estimates for the important coke formation parameters are found. A statistical analysis of the reliability of the parameter estimates obtained by the determinant criterion is also presented and shows that the results are statistically meaningful.

Notation

C	carbon content of the catalyst [g coke/g catalyst];
F_{A0}	molar feed rate of n-pentane at the inlet of the reactor [mole/hr];
F_t	total molar feed rate of n-pentane and hydrogen [mole/hr];
K_B	constant in the reaction rate equations, equal to 10;
W	weight of catalyst [gram];
d_p	catalyst particle diameter [cm];
k_I, k_H	(forward) reaction rate coefficients of isomerization and hydrocracking, respectively [mole/(g catalyst) (hr)];
k_{CP}, k_{CC}	reaction coefficients of coke formation by a parallel mechanism and by a consecutive mechanism, respectively [g coke/(g catalyst) (hr)];
n	number of experiments, i.e. number of settings of the independent variables;
n_1, n_2	exponents in hydrocracking and coke formation reaction rate equations;
p	number of parameters;
p_t	total pressure [atm];
r_I, r_H	reaction rates of isomerization and hydrocracking, respectively [mole/(g catalyst) (hr)];
r_C	reaction rate of coke formation [g coke/(g catalyst) (hr)];
t	time [dimensionless];
v	number of responses;
y_A, y_B, y_{H_2}	mole fractions of n-pentane, i-pentane and hydrogen, respectively;
y_{ij}	experimentally observed value of the j-th response at the i-th setting of the independent variables;
z	axial reactor co-ordinate [dimensionless]

Vectors and matrices

$\beta\ (p \ast 1)$	vector of the true (unknown) parameter values$_9$
$\mathbf{y}^{(i)}\ (v \ast 1)$	vector of experimentally measured values of the v responses at the i-th setting of the independent variables;
$\mathbf{y}_i\ (\mathrm{n} \ast 1)$	vector of observed values of the i-th response for all n experiments
$\eta^{(i)}\ (v \ast 1)$	vector of true (unknown) values of the v responses at the i-th setting of the independent variables;
$\mathbf{X}$	vector of settings of independent variables;
Λ	variance-covariance matrix of the responses, i.e. matrix of σ_{ij} elements;

Other Greek symbols

α	deactivation parameter;
γ	selectivity for isomerization;
ε	void fraction of catalyst bed;
ε_i	unobservable experimental error associated with the i-th response;
η_i	true (unknown) value of the i-th response variable;
ϱ_j	correlation coefficient of experimentally observed and predicted values of the j-th response;
ϱ_B	density of the catalyst bed [g catalyst/cm^3 reactor];
Ω	cross section of the reactor [cm^2];
$\varphi(z)$	function used in shifting the estimated carbon profiles;
Ø	deactivation function;
$\Psi(z)$	function used in the Voorhies law;
σ_{ij}	covariance between responses i and j;

Superscripts

$^\circ$	in the absence of fouling;
$\bar{}$	mean value;
$\hat{}$	predicted value by the mathematical model;
$\bar{\hat{}}$	mean from the predicted values.

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