Reconciliation of Process Flow Rates by Matrix Projection

Part I: Linear Case

Flow rate measurements in a steady-state process are reconciled by weighted least squares so that the conservation laws are obeyed. A projection matrix is constructed which can be used to decompose the linear problem into the solution of two subproblems, by first removing each balance around process units with an unmeasured component flow rate. The remaining measured flow rates are reconciled, and the unmeasured flow rates can then be obtained from the solution of the conservation equations. The basic case contains constraints which are linear in the component and the total flow rates. The method is extended to cases with bilinear constraints, involving unknown parameters such as split fractions.

Chi-square and normal statistics are used to test for overall gross measurement errors, for gross error in each node imbalance which is fully measured, and for each measurement adjustment.

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SCOPE

To monitor the performance of a chemical process, we require balanced component and total flow rates in the process streams. These flows can be calculated from judiciously chosen measurements of concentrations, temperatures and total flow rates; but since these measurements are subject to random error, the conservation laws will in general be violated.

The basic case considered here is linear in that it is assumed that whenever a concentration or temperature in a stream is measured, so is the total flow rate. Then the component or enthalpy flow can be calculated and used as the raw measurement data. These data are adjusted (reconciled), and the unmeasured flow rates are estimated so that the weighted sum of squares of the adjustments is a minimum and the conservation laws are obeyed. This restriction is then relaxed to allow the inclusion of bilinear constraints, which contain unknown parameters, multiplied by measured quantities.

The computational effort can be minimized if a reduced set of balance equations can be obtained from the original balances, such that the reduced set involves no unmeasured flow rate but a maximum number of measured flow rates. This was accomplished originally by Vaclavek (1969b), and later by Mah et al. (1976) and Romagnoli and Stephanopoulos (1981) by algorithms which iteratively eliminate balances involving unmeasured feed or product flow rates and merge two balances with a common unmeasured flow rate. These workers assumed that a stream was either unmeasured or completely measured, an assumption not made here.

The approach here is to define a projection matrix which can be directly constructed and which effectively blanks out the unmeasured quantities in producing this reduced set of balances.

If any of the measured flow rates or the balances is grossly in error as a result of instrument malfunction of miscalibration, sampling errors, unsuspected leaks or departures from steady state, such gross errors will affect all of the adjustments made and should be detected. Previous workers, notably Reilly and Carpani (1963), Almasy and Sztano (1975) and Mah et al. (1976), have presented statistical tests for the errors in the reduced set of balances, either collectively (chi-square test) or individually (normal distribution test). However, it was necessary to establish algorithms to find gross errors in the measurement adjustments (Mah et al., 1976; Romagnoli and Stephanopoulos, 1981). One objective then was to define test statistics which would directly reveal gross errors in both imbalances and in adjustments and would avoid trial and error deletion in turn of suspect measurements as was done by Ripps (1965).

CONCLUSIONS AND SIGNIFICANCE

A projection matrix has been used to obtain a reduced set of balance equations from the original component balances. This approach is equivalent to the algorithm of Vaclavek (1969b) for the case where a stream is completely measured or is unmeasured. It includes cases with partial measurement of streams, chemical reactions, and even sets of linear constraint equations with arbitrary matrices.

The approach has been extended to allow the inclusion of nonlinear constraints which contain unknown parameters, such as split fractions of a flow splitter or an unknown total flow rate in a stream with measured concentration.

A statistical test, based on the chi-square distribution, can be chosen to detect gross error collectively in a set of linear combinations of the errors in the reduced balance equations. Any one linear combination of those errors can be tested against the normal distribution. One such combination, which is of interest, tests any particular imbalance error as proposed by Reilly and Carpani (1963). Another effectively tests any particular adjustment in a measurement. As is illustrated, these two normal statistics together with the chi-square statistic point clearly in these simple cases to the offending measurements or balances.

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INTRODUCTION

The calculation of the steady-state mass and energy balances in the streams of a chemical process is a basic tool for monitoring its performance. To calculate these balances, one collects as many measurements as practicable of flow rates, concentrations and temperatures in the process streams. These raw measured data are in fact subject to random and possibly gross errors so that they may not be consistent with the conservation of mass and energy. The data must then be adjusted so that the adjusted values obey the conservation laws and some weighted measure of the total amount of adjustment is minimized.

Unfortunately, some of the measurements may also be subject to gross error because of instrument malfunction or miscalibration, because of departures from steady state through accumulation or depletion in process units or because of unsuspected leaks from pipes or vessels. Before the data can be adjusted, one must try to identify and correct or eliminate such gross errors; otherwise, the adjustments made would be seriously distorted by their inclusion.

The reconciliation problem was first considered by Kuehn and Davidson (1961) who solved for the optimal adjustments, using Lagrange multipliers for the case where all component flow rates are measured. They also considered energy balances where the product of total flow rate and specific enthalpy makes the equations nonlinear (more specifically, bilinear). Much more work has subsequently been done, notably by Ripps (1962, 1965), Reilly and Carpani (1963), Swenker (1964), Vaclavek (1969a,b,c, 1975, 1976), Mah et al. (1976), Romagnoli and Stephanopoulos (1981), Stanley and Mah (1981a,b), and Mah and Tamhane (1982). Excellent reviews of the published work have been presented by Hlavacek (1977) and Mah (1981). Ham et al. (1979) have reported on industrial applications of data reconciliation.

The problems considered have until recently been restricted to cases where either all the concentrations in a stream are measured or none are, although Stanley and Mah (1981a,b) have given an analysis free of this restriction. Chemical reactions have usually been taken into account by adding fictitious feed and product streams to a process unit where reactions occur.

Vaclavek (1969a,b) introduced the "reduced balance scheme" (RBS) to reduce the magnitude of the computations by eliminating iteratively any node with an unmeasured feed or product stream and by combining any pair of nodes connected by an unmeasured stream.

Vaclavek (1975, 1976), Mah et al. (1976), and Romagnoli and Stephanopoulos (1981) have extended the RBS to cases where a stream may have all or none of its concentrations measured, and may or may not have its total flow rate measured. We propose a general approach based on matrix projection.

The detection of gross errors in measurements was studied by Reilly and Carpani (1963) who established a chi-square test for the errors in the balance equations. They also showed that the individual imbalance errors could be tested against the univariate normal distribution. Ripps (1965) suggested that the problem be solved with and without the inclusion of a suspect measurement. This was computationally cumbersome, and the decision to reject a measurement was not justified by an accepted statistical test. Nogita (1972) proposed a test statistic for gross error which was shown by Mah et al. (1976) to be unsatisfactory. Almasy and Sztano (1975) proposed several test statistics, including ones for the case where the variances of the measurements were unknown in advance.

The basic problem to be treated here will be restricted to balances which are linear. The linear case is based on the assumption that for any stream in which concentration or temperature is measured, the total flow rate is also measured. The number of component concentrations which are measured in any stream is left arbitrary. The flow rate of enthalpy or of a component in a stream may then be calculated, if the data are measured, from the product of the total flow rate and the specific enthalpy or concentration, and used as the measurement to be adjusted. Because the total flow rate is a common factor in all measured flow rates of components or enthalpy in a stream, there will be covariance among these flow rates. We do assume that measured flow rates in different streams are independent, although this does not impose any restriction on the generality of the theory. The basic problem is then extended to include bilinear constraints.

PROBLEM STATEMENT

Consider a chemical plant in which there are K process units (or nodes), J streams and C components in a stream. The structure of the plant is expressed in the incidence matrix $\boldsymbol{A}$ with rows corresponding to nodes and columns to streams. Then

$$A_{kj} = 1 \text{ if stream } j \text{ enters node } k$$
$$= -1 \text{ if stream } j \text{ leaves node } k$$
$$= 0, \text{ otherwise}$$

for $k = 1,2,\ldots,K$ and $j = 1,2,\ldots,J$.

We replace each $(\pm)$ 1,0 in $\boldsymbol{A}$ by $(\pm)$ the identity and null matrices, respectively, to obtain the balance matrix $\boldsymbol{B}$. Then $\boldsymbol{B}$ is a block matrix, each of whose block rows contains C rows and corresponds to a node and each of whose block columns contains C columns and corresponds to a stream. For every node k in which R_k chemical reactions occur (reaction node) we construct a stoichiometric matrix $\boldsymbol{S}_k$.

Each row of $\boldsymbol{S}_k$ corresponds to a chemical reaction $r(1,2,\text{--}R_k)$ and each column to a compound $c(=1,2,\ldots,C)$. The entry $S_{k,rc}$ is the stoichiometric coefficient of compound c in reaction r occurring in node k, following the usual convention of a negative, positive or zero coefficient for a reactant, product or inert, respectively. A vector ϵ_k of extents of reaction is defined for each reaction node. Then we define a master stoichiometric matrix $\boldsymbol{S}$ with a nonzero $(R_k x C)$ block $\boldsymbol{S}_k$ only for a reaction node, so that each block row corresponds to a reaction node. Each of the K block columns contains C columns and corresponds to a node. For example, if

$$\boldsymbol{S} = \begin{bmatrix} S_1 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & S_3 & 0 & 0 & 0 \\ \cdot & \cdot & \cdot & S_4 & 0 & 0 \end{bmatrix}, \tag{1}$$

reactions occur in nodes 1, 3, 4, but not in node 2, 5 or 6.

If $\boldsymbol{x}_j$ is the vector of component flow rates in steam j and $\boldsymbol{x}^T = [\boldsymbol{x}_1^T \boldsymbol{x}_2^T \ldots \boldsymbol{x}_J^T]$, then the material balances for the plant can be written as

$$\boldsymbol{B}x + \boldsymbol{S}^T\epsilon = \boldsymbol{0} \tag{2}$$

with ϵ as the composite vector of extents of reaction for all reaction nodes. Vector ϵ is not measured nor known in advance.

Energy balances can be added to the problem definition by considering enthalpy flow rate as the $(C + 1)$th component. Then $\boldsymbol{B}$ will have K block rows, one for each node, with $(C + 1)$ rows. The matrix $\boldsymbol{S}_k$ is modified by adding a column containing the standard enthalpy change for each reaction (based on a product with unit stoichiometric coefficient). In addition, the energy balance for each node may have a heat flow q_k (usually unmeasured).

When all components in a stream are to be balanced, we must guarantee, if the total flow rate is measured, that the adjusted total flow rate equals the sum of the adjusted component flow rates. If x_{tj} is the total flow rate in stream j,

$$\boldsymbol{1}^T \boldsymbol{x}_j = x_{tj} \tag{3}$$

where $\boldsymbol{1}^T$ is a row vector of 1's. The matrix $\boldsymbol{B}$ is then augmented by adding a row and a column for each such stream, and the vector x_t of total flow rates is appended to x so that Eq. 2 becomes

$$\left[\begin{array}{cccc|c} \multicolumn{4}{c|}{B} & 0 \\ \hline I^T & 0 & \cdots & -I & \\ 0 & I^T & 0 & \cdots & \\ \cdot & \cdot & \cdots & & \end{array}\right]\left[\begin{array}{c} x \\ \hline x_t \end{array}\right] + \left[\begin{array}{c} S^T \\ \hline 0 \end{array}\right]\epsilon = 0 \tag{4}$$

We shall retain the balance Eqs. 2 for simplicity but if necessary will represent the modified B by B^+ and modified x by x^+. It is important not to alter the originally measured data so that Eq. 3 is forced to be valid for them. This will destroy statistical information and introduce dependencies among the data (Box et al., 1973).

Note that one common constraint, namely equality of concentrations and temperatures among streams leaving a flow splitter whose split is unknown, would strictly require the equating of the ratios of component to total flow rates in two streams, resulting in a nonlinear constraint. Such constraints could be rewritten to contain unknown split fractions which must be found by iteration. Almasy et al. (1969) proposed a method to iterate on all of the unknowns together. Below, a method is proposed which iterates only on the split fractions or other similar parameters.

The vector $\tilde{x}$ of measurements of component flow rates is again composed of vectors $\tilde{x}_j$ for each stream but which contain only as many elements as measured values. The flow rate of any component, whose concentration is not measured, is included in u, the vector of unknown flow rates.

The columns of $\mathbf{B}$ are then partitioned so that

$$B = [B_0 | B_1 | B_2].$$

B_0, B_1, B_2 contain respectively the columns corresponding to the exactly known flow rates c, to the measured component flow rates $\tilde{x}$ and to the unmeasured component flow rates u.

The measurements $\tilde{x}$ are to be adjusted by amounts a so that the material balances, as given in Eq. 2, hold for the adjusted flow rates, that is

$$B_0c + B_1(\tilde{x} + a) + B_2u + S^T\xi = 0 \tag{5}$$

It is assumed that the balances are linearly independent.

The estimated flow rates, $\hat{x}$, are then given by

$$\hat{x} = \tilde{x} + \mathbf{a} \tag{6}$$

For simplicity, we define

$$P \equiv [B_2 \vdots S^T] \tag{7}$$

$$\text{and } v^T \equiv [u^T \vdots \epsilon^T] \tag{8}$$

so that Eq. 5 becomes

$$B_0c + B_1(\tilde{x} + a) + Pv = 0 \tag{9}$$

Let us define

Problem P1

$$\min_{a,u,\xi} F(a) \triangleq a^T\Sigma^{-1}a \tag{10}$$

subject to Eq. 9. Σ is generally the inverse of a positive-definite weighting matrix, but here is the variance-covariance matrix of measurements $\tilde{x}$. It is usually assumed to be block diagonal with each diagonal block corresponding to a stream, although this assumption is not necessary. Any measured flow rates which are considered exactly known should be separated out in a vector of constant values to avoid making Σ singular. We shall now proceed to construct a solution to problem P1.

While problem P1 can readily be solved using Lagrange multipliers, we shall define a second problem whose solution is also a solution of P1.

Define Y as a matrix whose columns span the null space of P^T, that is

$$P^TY = 0 \tag{11}$$

Any vector $w(\neq 0)$, such that $P^Tw = 0$, is then a linear combination of the columns of Y. Then from Eq. 9,

$$Y^T[B_0c + B_1(\tilde{x} + a)] = 0 \tag{12}$$

This leads to the definition of

Problem P2

(a) $\min_a F(a)$ subject to Eq. 12.

(b) If a_* is the minimal solution to P2(a), solve

$$Pv_* = -B_1(\tilde{x} + a_*) - B_0c \tag{13}$$

for v_*

It is clear that any (a_*, v_*) which satisfy Eq. 13 also satisfy Eq. 9 and conversely both equations imply Eq. 12. Then the minimum (a_*) must be the same for both problems. We note that problem P2 is an extension of the reduced balance scheme of Vaclavek (1969b) in that the balance Eqs. 12 which remain have only measured flow rates but the remaining combinations of nodes are different in general for different components. The RBS was called a reconciliation graph by Mah et al. (1976). Indeed, the use of matrix Y here is a direct extension of the work of Mah et al. (1976) on partitioning the incidence matrix for the single-component flow rate problem. It will be seen that the exact choice of the matrix Y does not affect a_* but that one possible choice of Y for the case of no reactions is equivalent to deleting and merging nodes as suggested by Vaclavek (1969a), but for each component separately. Stanley and Mah (1981) have recently presented a general method of decomposition, which differs from the present one in finding a suitable transformation of the variables rather than of the constraint equations.

A minimal solution to P2(a) exists because the variance matrix, Σ, is positive definite, and thence the value of a_* which minimizes $F(a)$ is unique. We note that the minimal vector of adjustments, a_*, can be found even if there is no unique v_*.

Vector v_* which satisfies Eq. 13 exists, and it is unique, as shown by Mah et al. (1976), if and only if the columns of matrix P are linearly independent. This means that

$$Pv = 0 \Rightarrow v = 0 \tag{14}$$

and leads to a direct computational test of the uniqueness of v_*.

From the definitions of P and v, Eq. 14 becomes

$$B_2u + S^T\xi = 0 \tag{15}$$

Thus, nonuniqueness of v_* is equivalent to being able to satisfy material balances involving only unmeasured parameters. Note that the values of elements of u and ξ could be of either sign so that such balances can be made without regard to the actual directions of flow or of reaction. The streams in which u has nonzero values satisfying Eq. 15 form a particular example of the perturbation subgraph of Stanley and Mah (1981b).

Vaclavek (1969b) classified the unmeasured variables as determinable or indeterminate (observable or unobservable) and the measured variables as overdetermined or critically determined (redundant or non-redundant). When the variance matrix Σ is diagonal, a nonredundant measurement is not adjusted but in our case, it is in general adjusted if it has a nonzero covariance with a redundant measurement.

Stanley and Mah (1981a) generalized Vaclavek's classification of measurements and unmeasured variables in terms of observability. Following Stanley and Mah's definitions, the unobservable variables here correspond to the set of linearly dependent columns of matrix P and could be identified by column reduction of P. The remaining unmeasured variables are observable. An observable variable v_i is barely observable if there is a nonredundant measurement $\tilde{x}_j$ whose removal would render v_i unobservable. Such a variable would correspond to a column of P which is linearly

independent of other columns of P, but which is dependent on those columns together with the column of B_1 corresponding to $\tilde{x}_j$.

The nonredundant measurements correspond to zero columns of $(Y^T B_1)$ and thus do not contribute directly to the calculation of the adjustments. The remaining measurements are redundant.

DETERMINATION OF ADJUSTMENTS

Using Lagrange multipliers, we can solve problem P2 by finding a stationary point (a_*,λ_*) of the Lagrangian

$$L(a,\lambda) \equiv \frac{1}{2}F(a) - \lambda^T Y^T[B_0 c + B_1(\tilde{x} + a)] \tag{16}$$

This gives

$$a_* = \Sigma B_1^T Y \lambda_* \tag{17}$$

and

$$H\lambda_* = -Y^T[B_0 c + B_1 \tilde{x}] \tag{18}$$

with

$$H \triangleq Y^T B_1 \Sigma B_1^T Y \tag{19}$$

Equation 18 can be solved for λ_* and then a_* can be calculated, provided $(Y^T B_1)$ has full row rank. Equation 13 is then solved for v_*.

The matrix Y can be constructed as follows:

(a) Column reduce P to obtain P_r with linearly independent columns.

$$[P_r : 0] = PF \tag{20}$$

where F represents the nonsingular matrix which performs the necessary operations on the columns of P.

(b) Partition P_r so that

$$P_r = \begin{bmatrix} P_1 \\ \hline P_2 \end{bmatrix} \tag{21}$$

with P_1 square and nonsingular. Then

$$Y^T = [-P_2 P_1^{-1} : I] \tag{22}$$

For cases in which there is no chemical reaction, the use of Y will be equivalent to Vaclavek's (1969b) reduced balance scheme applied to each component separately.

DETECTION OF GROSS ERRORS

Let us assume that each concentration and flow rate measurement is independent and normally distributed with unknown mean and unknown variance. Although the product of two independent normal variables is not normally distributed, such a product is approximately normally distributed if the two variables take only positive values and if their coefficients of variation (relative standard deviations) are small enough, say less than 5%. This is not overly restrictive since the 95% confidence interval for a variable would then be written ±10% of its value. We will thus assume that the vector of measurements $\tilde{x}$ is a sample of a multivariate normal distribution with unknown mean x and known variance-covariance matrix Σ.

Measurements in different streams are usually assumed to be statistically independent as are measurements of concentrations and of the total flow in the same stream. Neither assumption is required by the following theoretical treatment. However, the flow rates of components in the same stream are not independent because of the common total flow rate. The matrix Σ is then block diagonal, in its simplest form, provided only that the matrix $\mathbf{B}^+$ and the vector $\mathbf{x}^+$ are rearranged so that the total flow in a stream is placed after the component flows in that stream. Madron et al. (1977) have discussed the computation of the matrix Σ.

Define the residuals of the balance equations, the imbalance vector e, as

$$e \equiv B_0 c + B_1 \tilde{x} + P v_* \tag{23}$$

where v_* are the estimated values of v. Then, with Eqs. 11 and 18,

$$Y^T e = Y^T[B_0 c + B_1 \tilde{x}] = -H\lambda_* \tag{24}$$

In order to establish the statistical distribution of $Y^T e$, we note t the true value of v must satisfy

$$B_0 c + B_1 x + P v = 0 \tag{25}$$

Then

$$E(Y^T e) = 0 \tag{26}$$

from Eqs. 11 and 25.

The variance of $Y^T e$ is given by

$$\begin{aligned} \text{var}(Y^T e) &= E(Y^T e e^T Y) \\ &= Y^T B_1 \Sigma B_1^T Y = H \end{aligned} \tag{27}$$

Then, as was first pointed out by Reilly and Carpani (1963), and later by Almasy and Sztano (1975), the quantity

$$\chi_Y^2 = e^T Y H^{-1} Y^T e \tag{28}$$

has a chi-square distribution with the number of degrees of freedom equal to the rank of Y. Thus the imbalances can be collectively tested against tabulated values of χ^2.

It is interesting to note that from Eqs. 17, 19 and 24,

$$\chi_Y^2 = a_*^T \Sigma^{-1} a_* = F(a_*) \tag{29}$$

This fact provides a statistical basis for arguments, such as those of Ripps (1965), for rejecting a measurement when its deletion results in a significant reduction in $F(a)$.

Furthermore, any set of linear combinations of $Y^T e$ can be similarly tested. For example, all the remaining balances for a particular node, or all the remaining balances for a particular component, could be selected by multiplying $Y^T e$ by a matrix whose rows are the appropriate unit vectors. In general, if W is a conformable matrix with full column rank, then corresponding to $W^T Y^T e$, the quantity

$$\chi_{YW}^2 \equiv e^T Y W (W^T H W)^{-1} W^T Y^T e \tag{30}$$

also has a chi-square distribution, but with degrees of freedom equal to the rank of W (< rank of Y). Note that the transformation $Y \rightarrow YQ$, where Q is any conformable square non-singular matrix, has no effect on the value of χ_Y^2 in Eq. 28, nor on the adjustments a_* in Eq. 17.

If in particular, instead of a matrix W, we use a vector w,

$$z \triangleq \frac{w^T Y^T e}{(w^T H w)^{1/2}} \sim N(0,1) \tag{31}$$

so that any linear combination of the elements of $Y^T e$ can be tested against the univariate normal distribution.

There are two specific choices of w which offer direct tests respectively of any particular imbalance and of any particular adjustment. Thus, if

$$w = \delta_k \tag{32}$$

where δ_k is the unit vector with unity in the k^{th} position,

$$z_k^e \triangleq (Y^T e)_k / H_{kk}^{1/2} \tag{33}$$

This test was first suggested by Reilly and Carpani (1963) and later by Mah et al. (1976).

Thus z_k^e can be calculated for every element of $Y^T e$ and tested against the normal distribution. If the cause of a gross error were a leak or unsteady-state accumulation in a process unit, one might expect all envelopes which contained that node would give rise to

large values of $|z_k^e|$. Those envelopes which did not contain that node would be expected to lead to normal values. It should be noted that the calculation of the analogous test statistic for a simple combination of the envelopes is an easy computation. The appropriate sum of elements of $(\boldsymbol{Y}^T \boldsymbol{e})$ is divided by the square root of the sum of the corresponding rows *and* columns of $\boldsymbol{H}$. We also note then that the values of z_k^e obtained will depend on the particular choice of $\boldsymbol{Y}$.

A second choice of $\boldsymbol{w}$ is to set

$$\boldsymbol{w}^T = -\delta_j^T \boldsymbol{\Sigma} \boldsymbol{B}_1^T \boldsymbol{Y} \boldsymbol{H}^{-1} \tag{34}$$

Then

$$\boldsymbol{w}^T \boldsymbol{Y}^T \boldsymbol{e} = -\delta_j \boldsymbol{\Sigma} \boldsymbol{B}_1^T \boldsymbol{Y} \boldsymbol{\lambda}_* \tag{35}$$

from Eqs. 17 and 24.

$$\boldsymbol{w}^T \boldsymbol{H} \boldsymbol{w} = (\boldsymbol{\Sigma} \boldsymbol{B}_1^T \boldsymbol{Y} \boldsymbol{H}^{-1} \boldsymbol{Y}^T \boldsymbol{B}_1 \boldsymbol{\Sigma})_{jj} \tag{36}$$
$$\triangleq \boldsymbol{Q}_{jj}$$

and

$$z_j^a \triangleq (a_j / \boldsymbol{Q}_{jj}^{1/2}) \sim N(0,1) \tag{37}$$

This allows a direct test of each adjustment against the normal distribution. This test has been independently proposed by Mah and Tamhane (1982).

One notes that the test statistic z_k^e could be calculated before solving for the adjustments but that both the χ_Y^2 and z_j^a test statistics require inversion of the matrix $\boldsymbol{H}$ as does calculation of the adjustment, $\boldsymbol{a}$.

ADDITION OF NONLINEAR CONSTRAINTS

As stated above, the linear case strictly excludes constraints on flow splitters and concentrations which are measured without the measurement of the corresponding total flow rates. These two exclusions are particularly important examples of bilinear constraints in which unknown parameters (split fractions, total flow rates, etc.) multiply the quantities to be adjusted or determined. If the unknown parameters are assigned a specific set of values, the problem becomes linear and can be solved as above. The questions remain, what initial set of values should be assigned and how should a new set of values of the parameter be obtained from the old one?

The bilinear constraints can be included by considering that the matrices, $\boldsymbol{B}_0$, $\boldsymbol{B}_1$, $\boldsymbol{P}$ and $\boldsymbol{Y}$ are functions of the parameters α. For a particular component of α, say α_k, the criterion of optimality is, from Eq. 16

$$-\frac{\partial L}{\partial \alpha_k} = \boldsymbol{\lambda}^T \frac{\partial}{\partial \alpha_k} [\boldsymbol{Y}^T (\boldsymbol{B}_0 \boldsymbol{c} + \boldsymbol{B}_1 (\tilde{\boldsymbol{x}} + \boldsymbol{a}))] \tag{38}$$
$$= 0 (k = 1,2, \ldots, S)$$

However, when the α_k are not optimal, the values of $(-\partial L / \partial \alpha_k)$ are elements of the steepest descent direction for L and can be used to obtain new values, $\alpha_k^\dagger$, from α_k using

$$\alpha_k^\dagger = \alpha_k - \epsilon (\partial L / \partial \alpha_k) \tag{39}$$

with ϵ suitably chosen to reduce the objective function. Alternatively, the secant method could be used to find the zeroes of $(\partial L / \partial \alpha_k)$.

In general, with $\boldsymbol{Y}^T$ given by Eq. 22,

$$\frac{\partial \boldsymbol{Y}^T}{\partial \alpha_k} = \left[\left\{ \boldsymbol{P}_2 \boldsymbol{P}_1^{-1} \frac{\partial \boldsymbol{P}_1}{\partial \alpha_k} - \frac{\partial \boldsymbol{P}_2}{\partial \alpha_k} \right\} \boldsymbol{P}_1^{-1} \middle| \boldsymbol{0} \right] \tag{40}$$

and the derivatives in Eq. 38 can be readily computed. In most cases, the matrices $(\boldsymbol{Y}^T \boldsymbol{B}_0)$ and $\boldsymbol{Y}^T \boldsymbol{B}_1)$ will be linear in the parameters and their derivatives are constant matrices, which are obtained with little effort. Thus, the iteration consists in solving the problem with a given set of α_k and then obtaining new values $\alpha_k^\dagger$ by the secant method, steepest descent or other appropriate methods. Once a satisfactorily converged solution is obtained, the statistical tests, *for fixed* α, can be done as before.

The set of starting values of the α_k could be obtained, if possible, from a solution in which the parameters are absent, that is without the splitter constraints and only using measurement of concentrations where flow rates are also measured. Alternatively, reasonable values could be obtained from experience with the process in question.

The addition of constraints on a splitter, to a problem originally without them, would add new constraints equal in number to the number of components entering the splitter times one less than the number of exit streams. The constraints, in terms of true flow rates, are of the form

$$x_j - \alpha_k x_l = 0 \qquad (0 \le \alpha_k \le 1) \tag{42}$$

for stream l entering and stream j leaving splitter node k. There is no change in the column partitions among $\boldsymbol{B}_0$, $\boldsymbol{B}_1$ and $\boldsymbol{P}$ and if $\boldsymbol{P}_1$ existed before imposing the splitter constraints, it exists after their imposition and is independent of the α_k. Then $(\boldsymbol{Y}^T \boldsymbol{B}_0)$ and $(\boldsymbol{Y}^T \boldsymbol{B}_1)$ are linear in the α_k. In example 3, the use of splitter constraints is illustrated.

The addition of measured concentrations in streams without measured total flow rates requires at least two concentrations in any given stream. Otherwise, the adjustment of the single concentration in a stream is chosen as zero and the balance achieved by choosing the total flow rate. The addition of such measured concentrations would transfer the respective columns of $\boldsymbol{P}$ into $\boldsymbol{B}_1$ and replace the unknown flow rate, u_{jc}, of component c in stream j, by

$$\alpha_j (\tilde{x}_{jc} + a_{jc})$$

Here, $\tilde{x}_{jc}$ is the measured concentration, a_{jc} its adjustment and α_j the unknown total flow rate. Note that the variance matrix $\boldsymbol{\Sigma}$ should be augmented, for each of these streams by a diagonal block which contains the variances of the measured concentrations.

ILLUSTRATIVE EXAMPLES

Several simple examples are presented to demonstrate the application of the theory. The calculations were programmed in the APL language, which is particularly suited for multiplication, transposition and inversion of matrices. It will be seen that not all of the tests necessarily indicate that some gross error exists in a given example. It is prudent to use the three tests, namely chi-square, z_k^e and z_j^a, jointly on a problem to obtain as much evidence as possible.

Example 1

This example (Ripps, 1965) represents a single process unit with two input streams and two output streams. It is special in that the four total flows are measured but the mole fractions are taken to be exactly known.

The data are as follows:

$$\boldsymbol{B}_1 = \begin{bmatrix} 0.1 & 0.6 & -0.2 & -0.7 \\ 0.8 & 0.1 & -0.2 & -0.1 \\ 0.1 & 0.3 & -0.6 & -0.2 \end{bmatrix}$$

$$\tilde{\boldsymbol{x}} = [0.1850 \quad 4.7935 \quad 1.2295 \quad 3.880]^T$$

$\boldsymbol{Y}^T = \boldsymbol{I}$ since $\boldsymbol{B}_2$ and $\boldsymbol{S}$ are absent

$\boldsymbol{\Sigma}$ = diag (2.89E-4 2.50E-3 5.76E-4 4.0E-2)

The results of the calculations are shown in Table 1. There, in column 1, we see that the existence of gross error is indicated by the chi-square and adjustment tests (in $\tilde{x}_2$ and $\tilde{x}_3$) but not by the imbalance tests. Without the adjustment test, Ripps deleted measurements in turn until the greatest reduction in the chi-square value was found.

Here, we can immediately delete $\tilde{x}_2$, since it has the largest $|z_j^a|$, the adjustments as shown in the second column. All tests are passed.

TABLE 1. RIPPS' (1965) EXAMPLES RESULTS

		Deleted Variable		
	None	$\tilde{x}_2$	$\tilde{x}_3$	
B_2^T	—	[0.6 0.1 0.3]	[−0.2 −0.2 −0.6]	
Y^T	I	$\begin{bmatrix}1 & -6 & 0\\ 0 & 3 & -1\end{bmatrix}$	$\begin{bmatrix}1 & -1 & 0\\ 0 & 3 & -1\end{bmatrix}$	
$\lvert z^e\rvert$	0.47	0.33	0.49	
	0.24	0.85	0.85	
	1.26			
χ^2_Y	8.36+	0.91	1.46	
$\chi^2_{d.f.,0.95}$	7.81	5.99	5.99	
a	−0.0174	−0.0099	−0.0194	
	0.0658	—	0.0093	
	−0.0566	−0.0041	—	
	−0.0261	0.1463	−0.0708	
$\lvert z^a\rvert$	1.03	0.60	1.15	
	2.73	—	0.86	
	2.63	0.42	—	
	0.13	0.78	0.36	
$\hat{x} = \tilde{x} + a$	0.1676	0.1751	0.1656	
	4.8593	5.0766*	4.8028	
	1.1730	1.2254	1.1592*	
	3.8538	4.0263	3.8092	
$[\Sigma_j(\hat{x}_j - x_j^2]^{1/2}$	0.239	0.043	0.313	

* Value needed to satisfy balances.
+ Underlined values exceed tabulated values at 95% confidence level.

For comparison, the results of deleting $\tilde{x}_3$ are shown in the third column. Again all tests are passed but the chi-square value and thus the objective function is greater than for the case with $\tilde{x}_2$ deleted. We would conclude that $\tilde{x}_2$ is in gross error. Ripps generated the measurements based on "true" values

$$x = [0.1739 \quad 5.0435 \quad 1.2175 \quad 4.000]^T$$

We see that deletion of $\tilde{x}_2$ gives $\hat{x}$ very close to the "true" values and that deletion of $\tilde{x}_3$ actually pushed the original estimates in the wrong direction. Some slight differences in our calculated results from those of Ripps (1965) and of Romagnoli and Stephanopoulos (1981) may have been caused by the ill conditioning of the matrix H for this case. The inverse of H used here was verified to be correct to 9 significant digits.

Example 2

This example is designed to illustrate how the theory can be used to detect an unsuspected leak. The process consists of three units in series, with an "unknown" leak in the second unit, as shown in Figure 1. Only total mass balances are considered.

$$B_1 = \begin{bmatrix}1 & -1 & & \\ & 1 & -1 & \\ & & 1 & -1\end{bmatrix} \qquad Y^T = I$$

$$x = [100 \quad 100 \quad 95 \quad 95]^T \qquad \Sigma = I$$

$$\tilde{x} = [98.5 \quad 101. \quad 96.5 \quad 95.5]^T$$

$$\therefore H = B_1 B_1^T = \begin{bmatrix}2 & -1 & 0\\ & 2 & -1\\ \text{sym.} & & 2\end{bmatrix}.$$

The values of the imbalance and the corresponding test statistic for all possible envelopes are:

Envelope:	1	2	3	(1 + 2)	(1 + 3)	(2 + 3)	(1 + 2 + 3)
e:	−2.5	4.5	1.0	2.0	−1.5	5.5	3.0
$\lvert z^e\rvert$:	1.77	3.18	0.71	1.41	0.75	3.89	2.12

We see that imbalances in all but one of the envelopes which include the second unit are excessive compared to the 95% confidence level of the normal distribution, 1.96. This might in itself be taken as evidence of a leak. However, if we proceed to determine the other statistics, we find the results shown in Table 2.

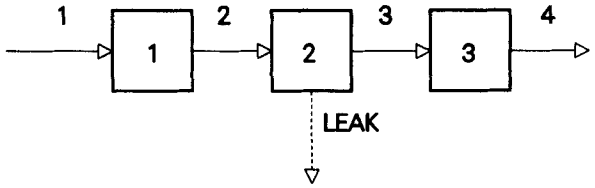

Figure 1. Flow diagram, example 2.

In particular, deletion of either $\tilde{x}_2$ or $\tilde{x}_4$ leads in each case to an adjustment being excessive where it was not originally. The only choice which contains no evidence of gross error is the assumption of a leak in unit 2, that is the addition of an unmeasured product stream to unit 2. This choice also has the smallest value of the objective function.

Example 3

Consider the ammonia synthesis system shown in Figure 2, with the measured species flow rates shown in Table 3. All other flows are unmeasured.

Then the matrices B_1, P and Σ are, with periods or blanks indicating zeroes.

B_1 =

	Stream 1	2	3	4	5
N_2	1	−1			
H_2	1				
Ar	1	−1			
N_2		1	−1		
H_2					
NH_3					
Ar		1			
N_2			1		
H_2					−1
NH_3				−1	
Ar					
N_2					
H_2					1
Ar					
N_2					
H_2					$-\alpha$
Ar					

P =

	Stream 2	3	5	6	7	S^T
					1	
	−1				1	
						1
						−1
	1	−1				−3
		−1				2
			−1			
			−1			
	1					
		1				
			1	−1		
			1	−1	−1	
				−1	−1	
			1		−1	−1	
			$-\alpha$	1		
				1		
				$-\alpha$	1	

TABLE 2. RESULTS OF EXAMPLE 2

	None	Variable Deleted $\tilde{x}_2$	$\tilde{x}_4$	Assumed Leak in Unit 2
$e(\lvert z^e \rvert)$	−2.5(1.77)		−2.5(1.77)	−2.5(1.77)
		2.0(1.41)		
	4.5(3.18)		4.5(3.18)	—
	1.0(0.71)	1.0(0.71)		1.0(0.71)
$\chi^2_Y(\chi^2_{2,f,.0.95})$	17.7(7.81)	4.67(5.99)	10.17(5.99)	3.63(5.99)
$a(\lvert z^a \rvert)$	−0.62(0.72)	−1.67(2.05)	0.17(0.20)	1.25(1.77)
	−3.12(3.61)	—	−2.33(2.85)	−1.25(1.77)
	1.38(1.59)	0.33(0.40)	2.17(2.65)	−0.50(0.71)
	2.38(2.74)	1.33(1.63)	—	0.50(0.71)
$\hat{x}$	97.88	96.83	98.67	99.75
	97.88		98.67	99.75
	97.88	96.83	98.67	96.0
	97.88	96.83		96.0

TABLE 3. RESULTS OF EXAMPLE 3

		$N_2^{(1)}$	$H_2^{(1)}$	$Ar^{(1)}$	$N_2^{(2)}$	$Ar^{(2)}$	$N_2^{(3)}$	$NH_3^{(4)}$	$H_2^{(5)}$	$X^2_y(X^2_{df,95})$
Measured Flows										
mol/s		33	89	0.4	101	20.2	69	62	205	
Without	Flows+	33	89	0.4	100.36	20.14	69.30	62.12	205	0.078(3.84)
Splitter	mol/s									
Straints	z^a	—	—	—	−0.28	−0.28	0.28	0.28	—	
				$z^e = 0.28$						
With	Flows,	31.68	94.94	0.40	100.05	20.12	69.76	60.57	203.95	15.78(9.49)
Splitter	mol/s									
Constraints										
($\alpha_* = 0.02003$)	z^a	−1.81	3.09	0.36	−0.41	−0.18	0.66	−1.04	−3.92	
				$z^e = 0.28$	−0.11	−2.22	−0.29			
Delete	Flows,	32.70	98.04*	0.40	100.56	20.15	69.23	62.66	205.0	0.29(7.81)
H_2 Value	mol/s									
In Feed	z^a	−0.45	—	−0.21	−0.19	−0.11	0.20	0.52	—	
($\alpha_* = 0.01976$)				$z^e = 0.28$	−0.10	0.05				

+ Requires negative H_2 purge.
* Needed to satisfy balances.

$$\Sigma = \begin{bmatrix} 0.82 & 1.14 & 5.12\text{E-}3 & & & & & & \\ 1.14 & 6.34 & 0.0142 & & & & & & \\ 5.12\text{E-}3 & 0.0142 & 1.28\text{E-}4 & & & & & & \\ & & & 8.16 & 0.816 & & & & \\ & & & 0.816 & 0.326 & & & & \\ & & & & & 3.81 & & & \\ & & & & & & 3.08 & & \\ & & & & & & & & 32.0 \end{bmatrix}$$

The matrix Y can be chosen as

$$Y^T = \begin{bmatrix} \cdot & \cdot & \cdot & 1 & \cdot & \frac{1}{2} & \cdot & \cdot & \cdot & \frac{1}{2} & \cdot & \cdot & \cdot & \cdot & \cdot & \cdot \\ 1 & \cdot & \cdot & \cdot & \cdot & \cdot & \cdot & (1-\alpha) & \cdot & \cdot & \cdot & 1 & \cdot & \cdot & 1 & \cdot \\ \cdot & 1 & \cdot & \cdot & 1 & \frac{3}{2} & \cdot & \cdot & 1 & \frac{3}{2} & \cdot & \cdot & 1 & \cdot & \cdot & 1 \\ \cdot & \cdot & 1 & \cdot & \cdot & (1-\alpha) & \cdot & \cdot & \cdot & (1-\alpha) & \cdot & \cdot & 1 & \cdot & \cdot & 1 \end{bmatrix}$$

Then

$$Y^T B_1 = \begin{bmatrix} \cdot & \cdot & \cdot & 1 & \cdot & -1 & -\frac{1}{2} & \cdot \\ 1 & \cdot & \cdot & -1 & \cdot & (1-\alpha) & \cdot & \cdot \\ \cdot & 1 & \cdot & \cdot & \cdot & \cdot & -\frac{3}{2} & -\alpha \\ \cdot & \cdot & 1 & \cdot & -\alpha & \cdot & \cdot & \cdot \end{bmatrix}$$

The rows correspond respectively to an elemental N balance on the reactor, the split condition for N_2, and overall elemental balances on H and Ar.

The results of the calculations are given in Table 3. If the splitter constraints are ignored, that is the last three rows of Y^T are deleted, the reconciliation obtained appears acceptable.

However, the flow of hydrogen out the purge stream 6 is negative, which is unacceptable.

With the splitter constraints imposed, the value of α_* is found to be 0.02003, with a very sharp minimum.

The flows are all reasonable but the three statistical tests all show excessive values. The third imbalance (H_2) is excessive as are the adjustments of the two H_2 measurements.

If we delete the H_2 measurement in stream 5, we find negative flows of H_2 in streams 2, 3, 5, 6 and 7. Deletion of the H_2 measurement in the feed causes column 2 of B_1 to move into matrix P and the matrix Y^T to lose its third row. This makes the last column of $(Y^T B_1)$ zero so that the H_2 measurement in stream 5 is nonredundant. The results are reasonable throughout and the value of α, 0.01976, is changed only slightly from the previous case.

The iterations to find the optimal split fraction, α_*, used the

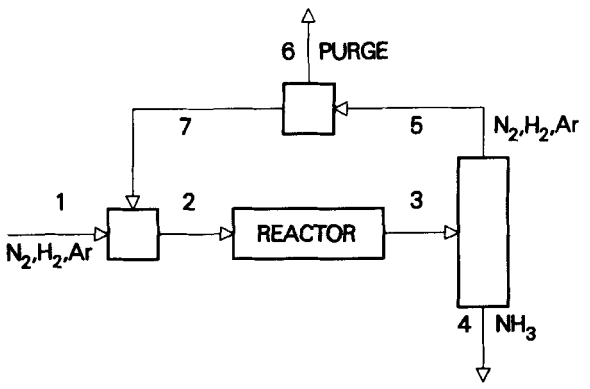

Figure 2. Flow diagram, example 3.

secant method to locate the zero of $\partial L/\partial \alpha$. In this case, this method was more effective than single-variable minimization of the objective function.

It is prudent to check all three statistical tests since as seen in example 1, the imbalance showed no gross error while the adjustments and chi-square value did. The pattern of excessive values of z^e and z^a could lead, as in example 3, to a ready diagnosis of the source of gross error.

SUMMARY

A theoretical approach has been presented for solving the problem of reconciling flow measurement data in streams of a chemical process. The essence is to construct a matrix which is orthogonal to the matrix in the balance equations which corresponds to unmeasured quantities. The problem can then be divided into a minimization problem to reconcile redundant measurements and then equation solution for the unmeasured variables. Bilinear constraints involving unknown parameters such as split fractions, can be included but their values must be found by iteration.

A class of test statistics is defined which can be used to detect gross errors in measurements or leaks or accumulations in process units. Two particular statistics test individual imbalances and measurement adjustments, respectively.

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NOTATION

$\boldsymbol{a}$ = vector of adjustments to measured flowrates
$\boldsymbol{A}$ = incidence matrix of process
$\boldsymbol{B}$ = balance matrix of process
$\boldsymbol{B}_0, \boldsymbol{B}_1, \boldsymbol{B}_2$ = columns of $\boldsymbol{B}$ corresponding to fixed, measured and unmeasured flow rates
$\boldsymbol{c}$ = vector of constant flow rates
C = number of components
$\boldsymbol{e}$ = imbalance vector, Eq. 23
$F(\boldsymbol{a})$ = objective function, Eq. 10
$\boldsymbol{H}$ = $\triangleq \boldsymbol{Y}^T \boldsymbol{B}_1 \boldsymbol{\Sigma} \boldsymbol{B}_1^T \boldsymbol{Y}$
$\boldsymbol{I}$ = identity matrix
j = stream number index
J = number of streams
k = node number index
K = number of process units (nodes)
$\boldsymbol{P}$ = = $[\boldsymbol{B}_2 : \boldsymbol{S}^T]$
R_k = number of reactions in reaction node k
$\boldsymbol{S}_k$ = stoichiometric matrix for reaction node k
$\boldsymbol{S}$ = stoichiometric matrix for process
$\boldsymbol{u}$ = vector of unmeasured flow rates
$\boldsymbol{v}$ = = $[\boldsymbol{u}^T \boldsymbol{\xi}^T]^T$
$\boldsymbol{w}$ = vector for normal test statistic, Eq. 31
$\boldsymbol{W}$ = matrix for chi-square test statistic, Eq. 30
$\boldsymbol{x}$ = vector of true flow rates
$\tilde{\boldsymbol{x}}$ = vector of measured flow rates
$\hat{\boldsymbol{x}}$ = = $\tilde{\boldsymbol{x}} + \boldsymbol{a}$, estimated flow rates
$\boldsymbol{Y}$ = defined by $\boldsymbol{Y}^T \boldsymbol{P} = 0$
z = test statistic, Eq. 31

Greek Letters

$\boldsymbol{\delta}_j$ = unit vector j
$\boldsymbol{\lambda}$ = vector of Lagrange multipliers
$\boldsymbol{\xi}$ = vector of extents of reaction
$\boldsymbol{\Sigma}$ = variance matrix of $\tilde{\boldsymbol{x}}$

LITERATURE CITED

Almasy, G. A., T. Sztano, P. Csillag, G. Veress, and J. Holderith, "Complex Material and Heat Balances as a Tool of Computer Control in Continuous Processes," 3rd CHISA Congress, Paper D3.10, Marianske Lazne, Czechoslovakia (1969).

Almasy, G. A., and T. Sztano, "Checking and Correction of Measurements on the Basis of Linear System Model," *Problems of Control and Information Theory*, **4**, 57 (1975).

Box, G. E. P., W. G. Hunter, J. F. MacGregor, and J. Erjavec, "Some Problems with the Analysis of Multiresponse Data," *Technometrics*, **15**, 33 (1973).

Ham, P. G., G. W. Cleaves, and J. K. Lawlor, "Operation Data Reconciliation: An Aid to Improved Plant Performance," Proc. 10th World Petroleum Congr., Bucarest, **4**, 281 (1979).

Hlavacek, V., "Analysis of a Complex Plant—Steady State and Transient Behavior," *Comput. Chem. Eng.*, **1**, 75 (1977).

Kuehn, D. R., and H. Davidson, "Computer Control. II Mathematics of Control," *Chem. Eng. Prog.*, **57** (6), 44 (1967).

Madron, F., V. Veverka, and V. Vanecek, "Statistical Analysis of Material Balance of a Chemical Reactor," *AIChE J.*, **23**, 482 (1977).

Mah, R. S. H., "Design and Analysis of Process Performance Monitoring Systems," Proc. Engng. Foundation Conf. on Chemical Process Control 2, Sea Island, GA (1981).

Mah, R. S. H., G. M. Stanley, and D. M. Downing, "Reconciliation and Rectification of Process Flow and Inventory Data," *Ind. Eng. Chem. Process Des. Dev.*, **15**, 175 (1976).

Mah, R. S. H., and Tamhane, "Detection of Gross Errors in Process Data," *AIChE J.*, **28**, 828 (Sept., 1982).

Nogita, S., "Statistical Test and Adjustment of Process Data," *Ind. Eng. Chem. Process Des. Dev.*, **11**, 197 (1972).

Reilly, P. M., and R. E. Carpani, "Application of Statistical Theory of Adjustment to Material Balances," 13th Can. Chem. Eng. Conf., Montreal, Que. (Oct., 1963).

Ripps, D. L., "Adjustment of Experimental Data," *Chem. Eng. Prog.*, **58**, (10), 120 (1962).

Ripps, D. L., "Adjustment of Experimental Data," *Chem. Eng. Prog. Symp. Ser.*, **61**, No. 55, 8 (1965).

Romagnoli, J. A., and G. Stephanopoulos, "Rectification of Process Measurement Data in the Presence of Gross Errors," *Chem. Eng. Sci.*, **36**, 1849 (1981).

Stanley, G. M., and R. S. H. Mah, "Observability and Redundancy in Process Data Estimation," *Chem. Eng. Sci.*, **36**, 259 (1981a).

Stanley, G. M., and R. S. H. Mah, "Observability and Redundancy Classification in Process Networks, Theorems and Algorithms," *Chem. Eng. Sci.*, **36**, 1941 (1981b).

Swenker, A. G., "Ausgleichung von Messergenbnissen in der Chemischen Industrie (Adjustment of Measurements in the Chemical Industry)," Proc. 3rd Int. Measurement Conf., Budapest, **1**, 29 (1964).

Vaclavek, V., "Studies on System Engineering II. On the Application of the Calculus of Observations in Calculations of Chemical Engineering Systems," *Coll. Czech. Chem. Commun.*, **34**, 364 (1969a).

Vaclavek, V., "Studies on System Engineering. III: Optimal Choice of the Balance Measurements in Complicated Chemical Systems," *Chem. Eng. Sci.*, **24**, 947 (1969b).

Vaclavek, V., "A Note to the Problem of Adjustment of Material Balance of Chemical Reactor," *Coll. Czech. Chem. Commun.*, **34**, 2662 (1969c).

Vaclavek, V., M. Kubicek and M. Loucka, "Calculation of Material Balances for Chemical Engineering Systems with Due Allowance for Measurement Errors," *Theor. Found. Chem. Eng.*, **9**, 242 (1976).

Vaclavek, V., M. Kubicek, and M. Loucka, "Calculation of Material Balances in Chemical Engineering Systems with Allowance for Errors in Measurement Classification of Stream Parameters," *Theor. Found. Chem. Eng.*, **10**, 256 (1976).

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