Analysis of Activation Energy of Grouped Parallel Reactions

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The effect of temperature on the reaction rate of grouped species which are consumed by parallel nth order irreversible reactions is investigated. It is shown that when the activation energies of the individual reacting species is about equal the temperature effect can be described by an Arrhenius expression. However, when the activation energies of the various reactions are widely spread the Arrhenius dependence may not be adequate unless one assumes that the activation energy of the lump may be temperature and conversion dependent. Under these conditions the Arrhenius temperature dependence is at best a rough approximation and it is very important to define exactly the experimental method of determining the activation energy. Widely different activation energies for the pseudocomponent may be obtained from different experimental techniques which yield the same value for a single reactant.

In many important processes the reaction mixtures contain a large number of reacting species. Analytical difficulties and costs usually prevent the determination of the detailed kinetics of these complex reaction networks and it is customary to lump the species into independent reactive groups and treat each one as a pseudo component. The lumped models have to accomplish an adequate balance between the requirements of acceptable accuracy, compactness in form and sufficient generality in describing the behavior of the grouped species.

Wei and Kuo (*1*) presented a formal general lumping analysis for monomolecular systems and their work has been extended by Bailey (*2*) for a continuum of reacting species. The main practical disadvantage of this approach is that it requires detailed knowledge of the various reaction rate constants and this information is usually not available.

A special interesting network is that of many independent parallel reactions. This case is encountered in several important processes such as hydrodesulfurization of crude oils in refineries (*3*, *4*), the parallel reactions of organic isomers in many chemical processes, the oxidation of unburned hydrocarbon in catalytic mufflers (*5*) and the decomposition of organic pollutants in water. Aris (*6*) has shown that the rate expression of the grouped species depends on the initial composition. Several basic behavioral features of these isothermal reacting mixtures have been discussed in (*7*, *8*) while the effect of diffusional disguise was presented in (*9*).

When a single component is consumed by an n^{th} order reaction, the temperature dependence of the rate constant can be described by an Arrhenius expression whose activation energy can be determined by several techniques. These include plotting the logarithm of either the time required to attain a certain fractional conversion or of the reaction rate at a fixed conversion level versus $1/T$. In practice it is often customary to describe the reaction rate of the grouped species by an empirically fitted simple kinetic model in conjunction with an Arrhenius temperature dependence (*5*, *10*, *11*). This phenomenological approach has the advantage that it requires a minimal number of adjustable parameters and that it usually yields a rather good approximation to the concentration of the grouped species.

In this work we explore the effect of temperature on the behavior of the grouped species and examine the validity of describing it by an Arrhenius type expression as well as the relationship between the activation energies of the lump and the individual species.

EFFECT OF TEMPERATURE ON THE REACTION RATE OF THE GROUPED SPECIES

We consider a system in which M parallel independent and irreversible n^{th} order reactions occur ($n \geqq 0$). The reaction rate of species i is given by

$$-\frac{dc(k_i,t)}{dt} = k_i\, c^n(k_i,t) \qquad i = 1, 2, \ldots, M \quad (1)$$

Hence, the concentration of species i is

$$c(k_i,t) = c(k_i,0)\, f(k_i,t)^{\frac{1}{1-n}} H(f(k_i,t)) \quad (2a)$$

where

$$f(k_i,t) = 1 + (n-1)\, c(k_i,0)^{n-1} k_i t \quad (2b)$$

and H is the Heaviside function

$$H(X) = \begin{Bmatrix} 0 & x \leqq 0 \\ 1 & x > 0 \end{Bmatrix} \quad (2c)$$

The concentration of the grouped species is defined as

$$C(t) = \sum_{i=1}^{M} c(k_i,t) \quad (3)$$

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$C(t)$ is a monotonic decreasing function of time hence its reaction rate can always be expressed by

$$-\frac{dC}{dt} = f(C(t)) \tag{4}$$

where the function f depends on the initial composition and distribution of the individual rate constants (*6, 7, 8*). It was shown in (*8*) that only if the conversion of all the individual species is identical for all times, is the reaction rate of $C(t)$ that of an n^{th} order reaction. In all other cases the rate of $C(t)$ cannot be fitted exactly by an n^{th} order rate expression.

When the temperature of the mixture changes, so do all the individual rate constants. Hence, a temperature variation requires in general a change in the *functional form* of $C(t)$ as well as of the various *parameters* which are included in it. This situation is rather undesirable, since the determination of a new rate expression at a number of different temperatures requires an extensive experimental effort and causes extreme difficulties in the extrapolation of the rate.

In order to avoid this difficulty, it is customary to describe the rate of the grouped species by the expression

$$-\frac{dC}{dt} = k(T, C)\, R(C(t)) = f(C(t)) \tag{5}$$

where $R(C(t))$ is an expression whose functional form and parameters are temperature independent and where all the temperature effects are accounted for by variation in the value of k. Thus, at a given conversion level and temperature, Equation (5) can be considered to be a definition of $k(T, C)$. The rate expression $R(C(t))$ may not describe exactly the relationship between the lumped concentration and rate expression over the entire temperature range. Hence, k may depend on $C(t)$ even though this dependence is often ignored in applications (*5, 10, 11*) and is usually not stated explicitly. This situation is analogous to the common procedure of describing the diffusion in concentrated solutions by Fick's law even though the diffusivity may be concentration dependent, or of computing the stress for a non-Newtonian fluid by a shear dependent viscosity.

In practice, it is common to neglect the concentration dependence of $k(T, C)$ and further to assume that its functional dependence can be described by an Arrhenius expression. A similar approximation is often used to describe the effect of temperature on the reaction rate of a catalyst with decaying activity (*12*). We will now examine the adequacy of this approximation and the various pitfalls which may exist in determining the activation energy of the grouped species.

DETERMINATION OF THE ACTIVATION ENERGY

The time required to obtain a certain fractional conversion can be determined by integration of Equation (5) to yield

$$t = -\int_{C(0)}^{(1-a)C(0)} \frac{dC}{k(T, C)\, R(C)} = \frac{Q(a)}{k^*(T)} \tag{6}$$

where we have used the mean value theorem and

$$Q(a) = -\int_{C(0)}^{(1-a)C(0)} \frac{dC}{R(C)} \tag{7}$$

while $k^*(T)$ is the value of $k(T, C)$ at some concentration in the range $(C(0), (1-a)C(0))$. Clearly if $k(T, C)$ is independent of C then $k^*(T) = k(T, C)$. Assuming that the temperature dependence of $k^*(T)$ is given by

$$k^*(T) = Ae^{-E'/RT} \tag{8}$$

we obtain by combining Equations (6) and (8)

$$E'/R = -\left(\frac{\partial \ln k^*(T)}{\partial(1/T)}\right)_a = \left(\frac{\partial \ln t}{\partial(1/T)}\right)_a \tag{9}$$

Differentiation of Equation (3) yields

$$\frac{dC}{d(1/T)} = \sum_{i=1}^{M} z_i \left(t\frac{E_i}{R} - \frac{\partial t}{\partial(1/T)}\right) \tag{10}$$

where

$$z_i = k_i\; c(k_i, t)^n = -\frac{dc(k_i, t)}{dt} \tag{11}$$

At a constant conversion level the left-hand side of Equation (10) vanishes and we obtain

$$E' = \sum_{i=1}^{M} z_i E_i \bigg/ \sum_{i=1}^{M} z_i \tag{12}$$

Thus, E' is the weighted average of the activation energy of all the individual reactants with the weighting function being z_i — the reaction rate of the i^{th} species at the specific level of conversion. Clearly if the activation energies of all the reactants are equal, E' will be independent of either the conversion level or temperature. In all other cases, E' may vary with both temperature and conversion, however its value is always bounded by the maximal and minimal activation energies of the various reactions. By differentiation of Equation (12) it is found that

$$\left(\frac{\partial E'}{\partial(1/T)}\right)_a = \sum_{i=1}^{M} \left(\frac{\partial z_i}{\partial(1/T)}\right)_a (E_i - E') \bigg/ \sum_{i=1}^{M} z_i \tag{13}$$

which can be rearranged to

$$R\left(\frac{\partial E'}{\partial(1/T)}\right)_a = \frac{\left[\sum_{i=1}^{M} z_i E_i\right]^2 - \sum_{i=1}^{M} z_i E_i^2 \sum_{i=1}^{M} z_i}{\left[\sum_{i=1}^{M} z_i\right]^2} + \frac{t \sum_{i=1}^{M} D_i (E_i - E')^2}{\sum_{i=1}^{M} z_i} \tag{14}$$

where

$$D_i = \frac{d^2 c(k_i, t)}{dt^2} = n k_i^2 c^{2n-1}(k_i, t) \tag{15}$$

It should be noted that according to Schwartz's inequality the first term of the right-hand side of Equation (14) is nonpositive, while the second one is nonnegative. When all the E_i's are equal the right-hand side of Equation (14) vanishes. Similarly

$$\left(\frac{\partial E'}{\partial t}\right)_T = \sum_{i=1}^{M} \left(\frac{\partial z_i}{\partial t}\right)_T (E_i - E') \Bigg/ \sum_{i=1}^{M} z_i$$

$$= \sum_{i=1}^{M} D_i (E' - E_i) \Bigg/ \sum_{i=1}^{M} z_i \qquad (16)$$

Equations (14) and (16) indicate that the constancy of the activation energy or in other words the adequacy of describing $k(T, C)$ by an Arrhenius expression depends on the spread of the activation energies as well as of the composition and the rate constants of the reacting species. When all the individual activation energies are equal $k(T, C)$ can be described exactly by an Arrhenius expression. When the spectrum of the activation energies is rather narrow the activation energy of the grouped species should be rather insensitive to variations in temperature and conversion. However, when the E_i's are widely spread the activation energy of the grouped species may be sensitive to changes in temperature or conversion. These effects occur since the exact functional form of $k(T)$ differs from the Arrhenius expression.

It is not possible to obtain general rules about the magnitude or even the sign of the changes in the activation energy of the grouped species due to the interaction between a large number of independent parameters. However, certain rules may be developed for special cases. For example, suppose the species are arranged in order of decreasing initial rate of conversion, that is

$$k_i c^{n-1}(k_i, 0) \geq k_{i+1} c^{n-1}(k_{i+1}, 0) \qquad \text{for} \quad i = 1, 2, \ldots, M-1 \qquad (17)$$

Here it can be shown (see Appendix) that if

$$E_i \leq E_{i+1} \qquad \text{for} \quad i = 1, 2, M-1 \qquad (18)$$

then

$$\left(\frac{\partial E'}{\partial t}\right)_T \geq 0 \qquad \text{for} \quad t \geq 0 \qquad (19)$$

For a single n^{th} order reaction the activation energy can be determined also from the effect of temperature on the reaction rate at a given conversion level. If we assume that for the grouped species

$$k(T, C) = Ae^{-E''/RT} \qquad (20)$$

then we obtain

$$\frac{-E''}{R} = \left[\frac{\partial \ln\left(-\dfrac{dC}{dt}\right)}{\partial(1/T)}\right]_\alpha = \left[\frac{\partial \ln\left[\sum_{i=1}^{M} z_i\right]}{\partial(1/T)}\right]_\alpha \qquad (21)$$

Substitution of Equation (12) into (21) and algebraic manipulations yield

$$E'' = E' + t\left(\frac{\partial E'}{\partial t}\right)_T \qquad (22)$$

Thus, unless E' is independent of temperature and conversion changes, E'' will also depend on the temperature and conversion level. E' and E'' are equal only when initial rate information is used to determine E'' or when $\left(\frac{\partial E'}{\partial t}\right)_T$ vanishes. While E' is a certain weighted average of the E_i's, E'' is not an average of the E_i's and its value may deviate largely from that of the individual activation energies. Thus, while

$$\text{Min } E_i \leq E' \leq \text{Max } E_i \qquad (23)$$

E'' is not bounded by the E_i's. This discrepancy will be especially marked at high conversion levels (large values of t) for which E'' might in special cases have a negative value even though the activation energies of all the individual reactions are positive. This pathological effect is due to the fact that in this case $k(T, C)$ cannot be properly described by the Arrhenius form.

TABLE 1. COMPOSITION AND RATE CONSTANTS DISTRIBUTION IN EXAMPLE 1

$c(k_i, 0)$, g.-mole/liter	A_i, 1/hr.	E_i, cal./mole
1	1,000	4,000
1.5	5,000	6,000
2	25,000	8,000
2.5	125,000	10,000
3	625,000	12,000

TABLE 2. COMPOSITION AND RATE CONSTANTS DISTRIBUTION IN EXAMPLE 2

$c(k_i, 0)$, g.-mole/liter	A_i, 1/hr.	E_i, cal./g.-mole
3.75	1.336	5,000
0.031	1.1×10^3	7,000
0.175	1.63×10^6	15,000
0.206	5.04×10^{10}	25,000
1.125	3.172×10^{15}	35,000

NUMERICAL EXAMPLES

We report here two numerical examples which demonstrate the trends that have been predicted by the analysis in the previous section. A much more comprehensive set of simulations is reported in (*13*). Both examples describe mixtures in which several parallel first-order reactions occur. The corresponding initial compositions and rate constants are reported in Tables 1 and 2.

Figure 1 describes the logarithm of the time required to obtain a certain level of conversion versus $1/T$ for example 1. The simulated computer experiments were carried out at increments of 20°C. over the temperature range of 460° to 660°K. for each level of conversion. The curves through the computed points have a slight curvature which is most noticeable for high and low conversion levels.

The activation energy E' is usually determined from the slope of curves such as Figure 1 (*5*). A more accurate determination of E' was carried out by use of Equation (12) and the corresponding values are reported in Figures 2 and 3 as a function of the temperature and conversion of the grouped species. Under certain conditions such as $T = 620$°K. or 55% conversion, E' is essentially a constant indicating that the Arrhenius dependence is adequate here. However, under other conditions such as $T = 460$°K. or a 95% conversion, E' is strongly affected by variations of the temperature or conversion indicating that the Arrhenius dependence is not adequate for these conditions.

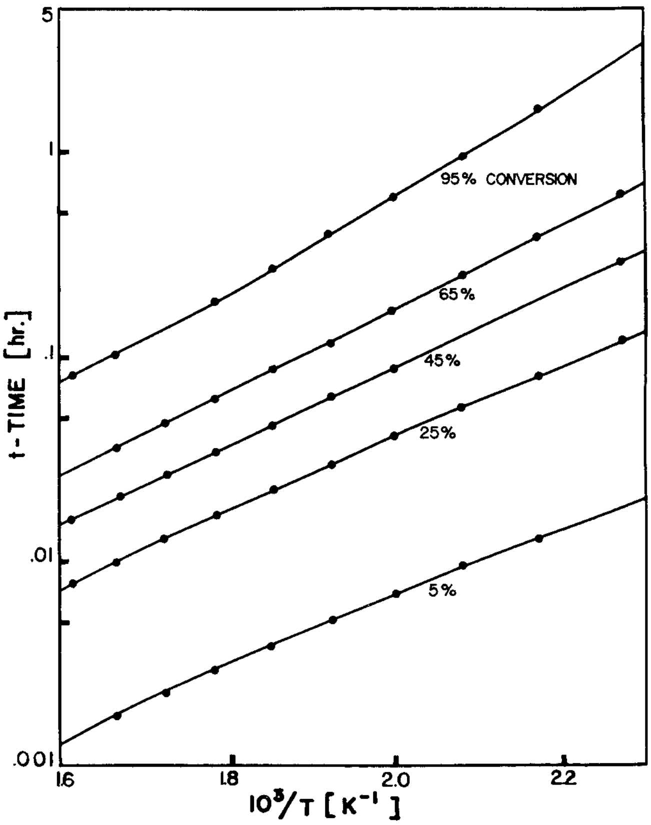

Fig. 1. The time required to reach a certain conversion level at various temperatures for the grouped species of example 1.

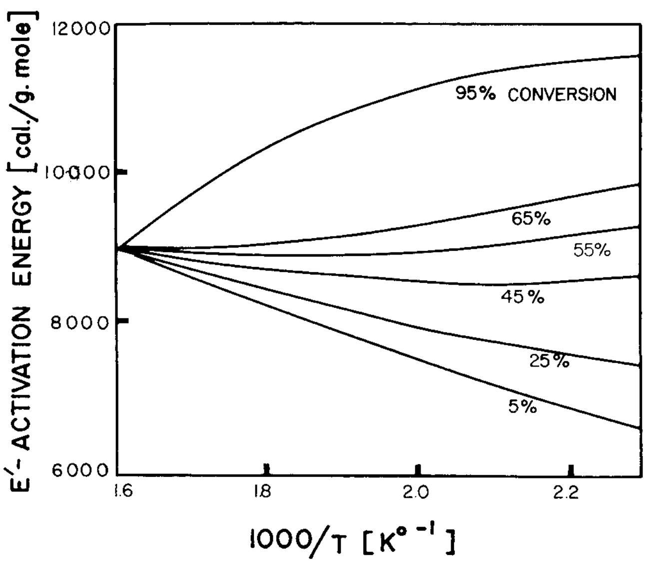

Fig. 2. E'—the activation energy of the grouped species (example 1) at various temperatures and conversion levels.

Figure 3 indicates that at very high conversion levels the activation energy of the grouped species is most influenced by that of the species with the slowest conversion rates since the fast reacting species are the first to be consumed. In all the cases E' is bounded by the maximal and minimal activation energy of the reacting species as predicted in the previous section. The example demonstrates that it is impossible to make any general a priori rules even about the direction of change of E' with temperature or conversion.

Figure 4 describes the logarithm of the reaction rate of the grouped species versus $1/T$ at several conversion levels. The corresponding compositions and rate constants are reported in Table 2. For a single irreversible n^{th} order reaction the slopes of all the curves should be identical and equal to E/R. However, Figure 4 clearly demonstrates that E'' — the activation energy obtained by this procedure assuming that grouped species act as a single component, will be strongly affected by changes of conversion or temperature.

Under certain conditions, such as 35% conversion, the reaction rate of the grouped species is decreasing with

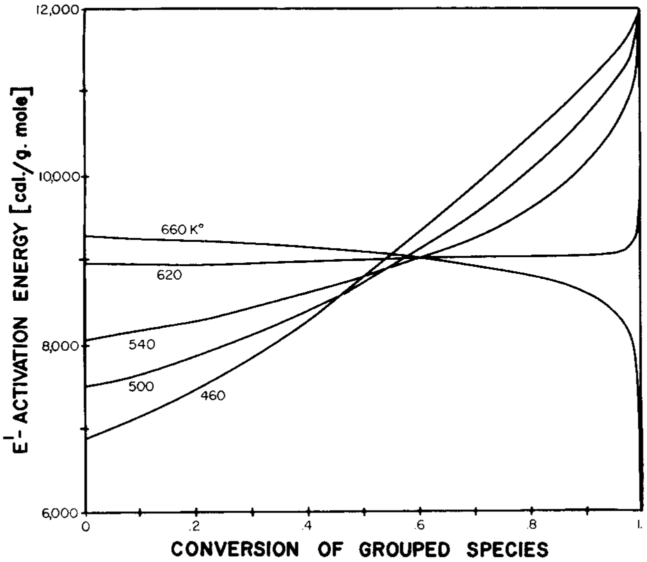

Fig. 3. E'—the activation energy of the grouped species (example 1) at various temperatures and conversion levels.

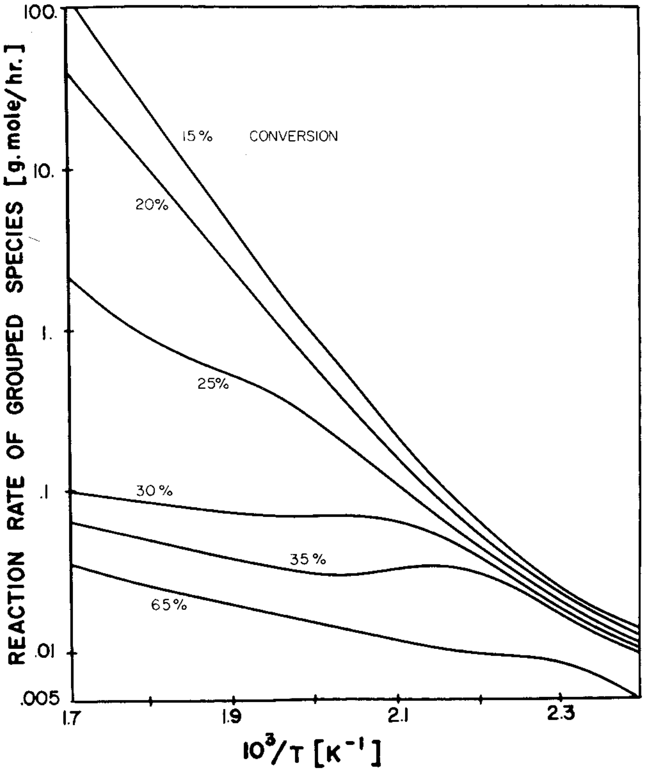

Fig. 4. The reaction rate of the grouped species (example 2) at various temperatures and conversions.

increasing temperatures even though the activation energy of all the individual reactions is positive. The pathological behavior may lead one to conclude that E'' has a negative value. The example demonstrates that computation of an apparent activation energy of the grouped species from the overall reaction rate at various temperatures is a most dangerous procedure. In any design application it is preferable to use E' which is at least a weighted average of the activation energies of the various species.

CONCLUSIONS AND REMARKS

When chemical reacting species are grouped together, the behavior of this "pseudocomponent" may be rather different that that of the individual components. In the previous work (8) we have shown that only if the conversion of all the species is identical for all times then the functional form of the rate expression for the grouped species is equal to that of the individual components. The failure of an n^{th} order rate expression to fit exactly the behavior of a reacting mixture is an indication of a wide spectrum of conversion rates.

In this work it was shown that the adequacy of describing the temperature effect on the grouped species by an Arrhenius expression depends on the spectrum of the activation energies of the reacting species. When all the E_i's are equal the temperature dependence is described exactly by the Arrhenius expression. However, when the individual activation energies are widely spread the temperature dependence may not be properly described by an Arrhenius expression and in order to obtain an adequate representation it may be necessary to assume that the activation energy is both concentration and temperature dependent. The discrepancy between the activation energy as determined from the two different experiments (E' and E''), which does never occur for a single reaction, is another manifestation of the inadequacy of the Arrhenius expression when the various E_i's are widely different. Thus, in order to obtain an appropriate description of the temperature effects on the reaction rate of the lump with an Arrhenius expression it is essential to group together species which have about the same activation energies.

In practice it is usually impossible to compute the exact value of E' from Equation (12) due to lack of sufficient information about the initial composition and rate constants. Hence, E' is most often computed from the slopes of experimentally determined curves similar to those shown in Figure 1 (5). These curves are most valuable for design and control purposes over the region in which the data has been obtained. However, any extrapolation of these experimental curves is a rather dangerous procedure due to the possible variation of the slopes with temperature and the inherent inaccuracies of the experimental points. The numerical examples point out that determination of the activation energy at one level of conversion may lead to severe pitfalls when this value is applied to another level of conversion.

ACKNOWLEDGMENT

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NOTATION

a = fractional conversion
A = pre-exponential factor
$c(k_i, t)$ = concentration of the i^{th} reactant at time t
$C(t)$ = total reactant concentration at time t
D_i = quantity defined by Equation (15)
E_i = activation energy of i^{th} reactant
E' = activation energy of the lump as defined by Equation (8)
E'' = activation energy of the lump based on the reaction rate at a given conversion level
f = function defined by Equation (2b)
H = Heaviside function
k_i = rate constant of the i^{th} reaction
$k(T, C)$ = rate constant for grouped species
M = number of reactants
n = reaction order
R = universal gas constant
t = time
T = temperature
z_i = reaction rate of i^{th} reaction

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APPENDIX

Proof of Equation (19)

Any two species which satisfy Equations (17) and (18) have to satisfy the inequality

$$(E_j - E_i)(k_jc^{n-1}(k_j, 0) - k_ic^{n-1}(k_i, 0)) \leq 0 \qquad (24)$$

Assuming for the moment that for all species $f(k_i, t) > 0$ we obtain by dividing Equation (24) by $f(k_j, t)f(k_i, t)$ and rearranging

$$(E_j - E_i)(k_jc^{n-1}(k_j, 0)/f(k_j, t) - k_ic^{n-1}(k_i, 0)/f(k_i, t)) \leq 0 \qquad (25)$$

Multiplication by $nk_ic^n(k_i, t)k_jc^n(k_j, t)$ and rearrangement yields

$$z_iD_jE_j + z_jD_iE_i \leq z_iD_jE_i + z_jD_iE_j \qquad (26)$$

Writing up inequalities similar to Equation (26) for every pair of species and adding them up yields

$$\sum_{i=1}^{M} \sum_{\substack{j=1\\ j\neq i}}^{M} D_iz_jE_i \leq \sum_{i=1}^{M} \sum_{\substack{j=1\\ j\neq i}}^{M} D_iz_jE_j \qquad (27)$$

Adding $\sum_{i=1}^{M} D_iz_iE_i$ to both sides of inequality (27) and rearrangement yields

$$\sum_{i=1}^{M} D_iE_i \sum_{i=1}^{M} z_i \leqq \sum_{i=1}^{M} D_i \sum_{i=1}^{M} z_iE_i \qquad (28)$$

which can be rewritten as

$$\sum_{i=1}^{M} \left(\frac{\partial z_i}{\partial t}\right) E_i \sum_{i=1}^{M} z_i - \sum_{i=1}^{M} \left(\frac{\partial z_i}{\partial t}\right) \sum_{i=1}^{M} z_iE_i$$

$$= \sum_{i=1}^{M} z_i \sum_{i=1}^{M} \left(\frac{\partial z_i}{\partial t}\right) (E_i - E') \geqq 0 \qquad (29)$$

Comparison of Equations (29) and (16) indicates that in this case

$$\left(\frac{\partial E'}{\partial t}\right)_T \geqq 0 \qquad Q.E.D. \qquad (30)$$

Similarly, it can be shown that if

$$E_i \geqq E_{i+1} \quad \text{for} \quad i = 1, 2, \ldots, M-1 \qquad (31)$$

then

$$\left(\frac{\partial E'}{\partial t}\right)_T \leqq 0$$

When $f(k_j, t)$ vanishes for some of the species inequality (26) becomes an equality since in this case $z_j = D_j = 0$. However, this does not affect the proof.

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Optimal Feedback Control of a Class of Linear Tubular Processes

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Optimal output feedback control law requiring measurement of the output only has been considered for a class of tubular processes. The advantage of this formulation, which requires a single measurement of the output only, over a previous result which requires an infinite number of measurements, has been demonstrated through a heat exchanger example. However, when the terminal time is greater than the residence time of fluid, a time-delay element is needed.

Processes involving transport of fluids or solids have transportation lags associated with trem, or they can be approximated by models with time delays. Frequently, the control of the output (at the exit) of these tubular processes can be described by differential-difference equations. Open-loop controls of processes described by differential-difference equations have been considered by a number of investigators (*1* to *4*). Koppel (*1*) considered the time-optimal control of processes described by a differential-difference equation. Ray (*2*) treated the optimal open-loop control of processes with pure time delay using a variational approach. Seinfeld and Lapidus (*3*) considered the open-loop control of a distributed-parameter process described by differential-difference equations. Using a direct programming approach Lim (*4*) presented and solved time optimal control problems.

For control systems governed by linear differential-difference equations, Koepcke (*5*) developed a synthesis technique particularly suited for a digital computer. The optimum feedback control of a class of processes described by differential-difference equations was considered by Koppel et al. (*6*) as a special case of more general distributed-parameter process control, and they obtained near optimal feedback solution which, when applied to a heat exchanger, requires the measurement of the entire temperature profile along the length (hence requires an infinite number of sensors). More recently, Shih (*7*) considered a special case of the terminal time being less than one, $t_f < 1$. However, in this case the process reduces to the usual lumped-parameter process and truly distributed nature of the distributed-parameter processes appears only when $t_f > 1$. We present here a different approach and show that for the output transfer from one steady state to another the optimal control law requiring the measurement of the output alone, can be obtained for all time, that is, both cases $t_f \leqq 1$ and $t_f > 1$.

GENERAL CONTROL PROBLEM

Many tubular processes can be represented by differential-difference Equations (1) to (3), for example,

$$\sum_{i=0}^{n} a_i\, d^iy(t)/dt^i = k_1\, m(t) - k_2\, m(t-\tau)$$

where τ is the time delay. As shown by Lim (*4*) it is convenient to represent the input-output relationship by state and output equations of the type $\dot{x}(t) = A\,x(t) + b\,m(t)$ and $y(t) = k_1x_1(t) - k_2x_2(t-\tau)S(t-\tau)$. Therefore, we consider a general class of processes with multiple inputs, multiple outputs, and a constant time delay in the state vector

$$\dot{x}(t) = Ax(t) + Bu(t) \qquad (1)$$