

(11) is fairly satisfactory for the stirred cell at 437 rev./min. and for water flowing over the smooth plate.

For the wake-interference flow, however, with its apparently anomalously efficient mass transfer, the reduction of  $k_L$  by surface-active agent is particularly marked, and is considerably more pronounced than is expected from the theory of Equation (11). This is evidently due to some inhibition of the wake-interference flow pattern by the local surface tension gradients. Indeed, the surface becomes appreciably smoother. However,  $k_L$  for *clean* surfaces in the wake-interference flow is about 3 times greater than for flow over the smooth plate if  $Re$  is the same. Hence  $k_L$  for wake-interference flow is still about twice as great in the presence of surface-active agent at  $\Pi = 5$  dynes/cm., that is, though the surface-active material reduces the advantage of the eddy-promoting roughness, it does not eliminate this advantage.

In the present experiments the adsorbed film behaves as if insoluble, because there is no large-scale renewal and removal of the surface, but only local elastic clearings.

#### NOTATION

$B_f$	= film thickness
$d$	= internal diameter of tube
$D$	= molecular diffusivity
$D_E$	= eddy diffusivity
$e$	= height of roughness element
$g$	= gravitational acceleration
$k_L$	= mass transfer coefficient in liquid phase
$R$	= radius of curvature
$Re$	= Reynolds number
$s_1$	= separation between ridges
$s_2$	= thickness of ridge
$Sc$	= Schmidt number
$Sh$	= Sherwood number ( $= k_L d/D$ )
$v_0$	= characteristic shear stress velocity of turbulent flow
$v_m$	= mean linear flow velocity

$v'$	= eddy fluctuation velocity
$V_b$	= volumetric flow rate per unit width of plate
$We$	= Weber number ( $We = v_m(\rho d/\sigma)^{1/2}$ )

#### Greek Letters

$\delta_1$	= thickness of mathematically equivalent viscous layer near surface
$\delta_2$	= thickness of mathematically equivalent diffusion layer near surface
$\theta$	= angle of inclination to horizontal
$\mu$	= viscosity of liquid
$\nu$	= kinematic viscosity of liquid
$\Pi$	= lowering of surface tension
$\rho$	= density
$\sigma$	= surface tension
$\tau_0$	= wall stress

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# Thermodynamics of Multicomponent Reactive Nonuniform Systems (Adsorption)

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Certain systems are characterized by a nonuniform distribution of matter in space. Examples of such systems are liquid-fluid (1 to 3) and the solid-fluid interfaces (4). It is the main purpose of this paper to extend the thermodynamics of nonuniform systems to multicomponent reactive mixtures in the presence of an external field and to apply the above formalism to physical adsorption. This amounts to an extension of Hart's original analysis (5 to 7) as well as an improved analysis of the solid-fluid interface presented recently by Metiu and Ruckenstein (8). Furthermore, various approximate solutions for the solid-fluid interface are considered, all of which reduce properly to known results.

The characterization of the thermodynamic state of an inhomogeneous system in which nonuniformity is associated with the distribution of mass over space can be done

in two different ways. In the first approach one can calculate a potential  $\varphi(r)$  created by a variation of density with position and deduce in this fashion a relationship be-

tween  $\varphi(r)$  and density gradients at  $r$ . This is an attempt to go from distribution functions to density gradients. Such a methodology has been discussed recently by Metiu and Ruckenstein (8). An alternate procedure would be to postulate the existence of density gradients and derive from such formalism distribution functions (9). While the former method is statistical in essence, the latter is purely phenomenological. Here we shall examine only the latter type of structure. This type of thermodynamic treatment has been formalized rigorously by Hart (5 to 7). Hart considered a one component system in the absence of an external field. The necessary modification to account for the presence of an external field has already been partly examined (9) in a particular application of the present type of theory.

## THERMODYNAMIC TREATMENT

Consider a multicomponent nonuniform fluid system enclosed within a rigid boundary. Following the methodology of Hart (5 to 7) we first postulate the existence of equilibrium states which are characterized completely by a set of extensive variables. These are: the internal energy density  $eU$ , strains  $e_{jk}$ , mole number densities  $e\rho_i$ , and density gradients  $e\rho_{i,j}$ , where  $\rho_{i,j} = \frac{\partial \rho_i}{\partial x_j}$ . We shall also admit the existence of entropy and denote by  $eS$  its value at a point. In the above definitions  $e$  is the dilatation

$$e = \text{Det } ||e_{jk}|| \quad (1)$$

The properties  $U$ ;  $S$ ;  $\rho_i$ ;  $\rho_{i,j}$  are per unit volume in the strained state. It follows that  $eU$ ;  $eS$ ;  $\dots$  are properties per unit volume in the reference state.

The assumed validity of the above postulates implies that in the presence of an external field ( $\psi_i$  acting on species  $i$ ) the fundamental equation in the energy representation is given by

$$\delta eU = T\delta eS - P_{jk}\delta e_{jk} + (\mu_i + \psi_i)\delta e\rho_i + \mu_{ij}\delta e\rho_{i,j}^* \quad (2)$$

$j, k = 1, 2, 3$   
 $i = 1, 2, \dots, n$

where  $T$ ;  $P_{jk}$ ;  $\mu_i$ ;  $\mu_{i,j}$  are intensive properties—respectively, temperature, stresses, density potentials and density gradient potentials. Because of the assumed homogeneity of the field properties with respect to the extensive properties, it follows from Equation (2)

$$eU = TeS - P_{jk}e_{jk} + (\mu_i + \psi_i)e\rho_i + \mu_{ij}e\rho_{i,j}^{**} \quad (3)$$

It would be worthwhile to mention at this stage that a more general case would be one taking into account all spatial derivatives of  $\rho_i$  (that is,  $\rho_{i,j}$ ;  $\rho_{i,jk}$ ;  $\rho_{i,jkl}$   $\dots$ ). Furthermore, it should be stressed that the concept of local equilibrium and local thermodynamic properties are features inherent in approximate theories. Hence, even though the above methodology is a rigorous analysis it is still an approximate theory.

We shall not consider thermal and mechanical equilibrium as these have already been considered by Hart and the results are unaltered when one considers a multicomponent reactive system in an external field. These are respectively

$$T = T_0 = \text{constant}^\dagger, \text{ everywhere in } V \quad (4)$$

$$P_{jk} = p\delta_{jk}, \text{ at } \underline{r} \quad (5)$$

## CHEMICAL EQUILIBRIUM

Consider a multicomponent ( $B_1, B_2, \dots, B_n$ ) isolated system in which chemical transformations are taking place

$$\nu_{i\alpha} B_i = 0 \quad \alpha = 1, 2, \dots, t \quad (6)$$

Since in the present discussion we are merely concerned with the distribution of the various species at equilibrium, we shall take  $\delta eS = \delta e_{jk} = \delta e = 0$  ( $e = 1$ ). In accordance with the internal energy minimum principle, we seek an extremum of  $\mathcal{U}$  the total internal energy of the system

$$\mathcal{U} = \text{MIN} \int U dV \quad (7)$$

subject to  $n$  constraints given by the law of definite proportions

$$N_i^* = \int [\rho_i - \nu_{i\alpha} \xi_\alpha] dV \quad i = 1, 2, \dots, n \quad (8)$$

In the above  $N_i^*$  stands for the initial molar mass of component  $i$  while  $\xi_\alpha$  is the extent of the  $\alpha$ th reaction. It should be pointed out that Equations (8) imply the existence of  $t$  stoichiometric equations; these are obtained using the principle of conservation of mass together with Equations (8)

$$\nu_{i\alpha} M_i = 0 \quad \alpha = 1, 2, \dots, t \quad (9)$$

In order to solve the above variational problem we introduce CONSTANT Lagrange multipliers  $\mu_i^0$  and construct the combined extremal (10)

$$\delta \mathcal{U} = \int [(\mu_i - \mu_i^0 + \psi_i)\delta \rho_i + \mu_{ij}\delta \rho_{i,j} + [\nu_{i\alpha} \mu_i^0 \delta \xi_\alpha]] dV \quad (10)$$

Integration by parts yields

$$\delta \mathcal{U} = \int \left[ \mu_i - \mu_i^0 + \psi_i - \frac{\partial \mu_{ij}}{\partial x_j} \right] \delta \rho_i dV + \int \nu_{i\alpha} \mu_i^0 \delta \xi_\alpha dV + \int \mu_{ij} \delta \rho_i dA_j \quad (11)$$

At equilibrium  $\delta \mathcal{U} = 0$ , hence we conclude that

$$\mu_i + \psi_i - \frac{\partial \mu_{ij}}{\partial x_j} = \mu_i^0 \quad (12)$$

$$- \nu_{i\alpha} \mu_i^0 = 0 \quad (13)$$

everywhere in  $V$  while

$$\mu_{ij} \delta \rho_i dA_j = 0 \quad (14)$$

everywhere on  $A$ . As one can see, only two types of boundary conditions are possible (10).

$$\text{Type 1 } \mu_{ij} dA_j = 0 \quad (15)$$

$$\text{Type 2 } \delta \rho_i = 0$$

Previous works (5, 6) have assumed surface conditions of type 1, but those of type 2 often give a better physical picture.

It should be noted at this stage that the chemical potential  $\mu_i$  is an intrinsic property and as such is not observable and that  $\mu_i^0$  is an extrinsic potential.  $\mu_i^0$  is a CONSTANT throughout the system and would be the value observed when the nonuniform system is fully equilibrated with a homogeneous system. In addition, the equilibrium constant

\* For simplicity we shall use throughout Einstein's summation convention (summation over repeated indices).

\*\* In the present paper a uniform system would be defined as one where the gradient variables are neglected. A uniform system with  $\psi_i = 0$  will be called homogeneous.

† 0 indicates an extrinsic (measurable) property.

of each chemical reaction is constant throughout the non-uniform system. This is easily seen from Equation (13).

## MEASURABLE STRESSES

The stresses  $P_{jk}$  we have introduced in Equation (2) are intrinsic properties and are not measurable; at equilibrium they are purely hydrostatic and vary from point to point, that is, Equation (5). Therefore before embarking on a study of the solid-fluid interface we must obtain a general expression for the measurable extrinsic stresses  $P_{jk}^0$  which will be required later. Consider a distortion of the system in which points at  $x_i$  are moved to  $x_i' = x_i + \delta x_i$  while requiring that  $e\rho_i$  and  $eS$  be unaltered by such a transformation.\* We have

$$\delta U = \int [e\rho_i \psi_{i,j} \delta x_j - P_{jk} \delta e_{jk} + \mu_{ij} \delta e\rho_{i,j}] dV \quad (16)$$

It was shown in (5) [Hart's Equations (1), (11), (23)] that

$$\left. \begin{aligned} \delta e_{jk} &= \delta x_{j,k}; & P_{jk} &= p \delta_{jk} \\ \delta e\rho_{i,j} &= -[\rho_{i,k} \delta x_{k,j} + \rho_i \delta x_{k,kj}] \end{aligned} \right\} \quad (17)$$

Substitution of Equations (17) into Equation (16) yields

$$\delta U = - \int \left[ -(\rho_i \psi_{i,j}) \delta x_j + (p \delta_{jk} + \mu_{ik} \rho_{i,j}) \delta x_{j,k} + \frac{\rho_i}{2} (\mu_{il} \delta_{jk} + \mu_{ik} \delta_{jl}) \delta x_{j,kl} \right] dV \quad (18)$$

If Equation (18) is identified with Hart's Equation (8) in reference 7, one obtains directly from Hart's Equations (14) and (18) the measurable stresses and the conditions they must satisfy at equilibrium.

$$p_{jk}^0 = \left[ p - \left( \frac{\rho_i}{2} \mu_{il} \right)_{,l} \right] \delta_{jk} + \frac{\rho_{i,j}}{2} \mu_{ik} - \frac{\rho_i}{2} \mu_{ik,j} \quad (19)$$

$$p_{jk,k}^0 = -\rho_i \psi_{i,j} \quad (20)$$

The last point in a general treatment would be to include an analysis of the thermodynamic stability. At present there is no solution available for such a general problem (11); this is largely due to our insufficient mathematical knowledge of  $n$ th order partial differential equations. In certain restricted cases the analysis can be performed (12). Such a case is discussed in the next section.

## ONE DIMENSIONAL NONUNIFORM SYSTEMS

This case is important in many applications, the most important being the solid-fluid interface (finite or semi-infinite systems) and the liquid-fluid interface (infinite systems). In such cases an integrated expression for the internal energy would be useful. In this case the total energy is given by

$$U = A \int [TS - p + (\mu_i + \psi_i) \rho_i + \mu_{i,z} \rho_{i,z}] dZ \quad (21)$$

using the conditions for mechanical, chemical, and thermal equilibria, Equations (4), (5), and (12), one obtains

$$U = T_0 S - P_0 V + \mu_i^0 V \rho_i^0 + A \mu_i^0 \Gamma_i + \sigma A \quad (22)$$

where

$$\Gamma_i = \int (\rho_i - \rho_i^0) dZ \quad (23)$$

$$\sigma = \int (P_0 - p) dZ \quad (24)$$

A more useful expression for the surface tension can easily be obtained using the extrinsic mechanical stresses. Using Equations (19) and (20) it is easily shown

$$P_{zz}^0 = p - \rho_i \mu_{i,z,z} \quad (25)$$

$$P_{zz}^0 = P^0 - \int \rho_i \psi_{i,z} dZ \quad (26)$$

Equating Equations (25) and (26) and eliminating  $(P_0 - p)$  in Equation (24) yield after integration by parts

$$\sigma = \int [\rho_{i,z} \mu_{i,z} + \int \rho_i \psi_{i,z} dZ] dZ \quad (27)$$

Before pursuing the present analysis, a word on the calculation of the various quantities presented so far is in order. In order to obtain the density profiles, Equations (8), (12), and (13) must be solved simultaneously. This is a closed problem with  $(2n + t)$  equations and unknowns ( $t$  values of  $\xi_\alpha$ ,  $n$  of  $\mu_i^0$  and  $n$  of  $\rho_i$ ).\*\* We notice that Equations (12) are second order differential equations in  $\rho_i$  since  $\mu_{ij}$  is in general a function of  $\rho_{i,z}$ . The necessary boundary conditions ( $2n$  of them) are restricted to two possible types given by Equations (15). From the calculated density profile one could obtain easily  $\Gamma_i$  and  $\sigma$  using Equations (23) and (27).

Equation (27) deserves more discussion. First let us examine an infinite system (liquid-fluid interface without end effects). The first case to be considered is a nonuniform system in the absence of an external field. In such instances Equation (27) reduces to

$$\sigma = \int \rho_{i,z} \mu_{i,z} dZ \quad (28)$$

This equation has been obtained previously for a one component system (5). Secondly, suppose that an external field exists across the interface; in such instances Equation (28) would predict an infinite value for  $\sigma$  which is erroneous, and hence we see that the purpose of the second term in Equation (27) is to deduct the uniform contribution due to an external field. It should be emphasized that the presence of an external field affects the interfacial tension; this contribution is contained implicitly in the first term of Equation (27) as seen from Equation (12). The same type of discussion could be applied to the solid-fluid interface.

We see that not only is the present treatment an alternate way of examining multicomponent reactive system but that in some instances it provides us with a better description of certain phenomena such as the solid-fluid interface, disjoining pressure, chemical reaction in the presence of a solid surface. This is so because in some instances Metiu and Ruckenstein have made approximations equivalent to the neglect of the gradient extensive variables (that is  $\rho_{i,z} = 0 \rightarrow \varphi = 0$ ). It should also be pointed out that the analysis of adsorption and that of reacting mixture on a solid surface presented in reference 8 are inconsistent model-wise.

In view of the above facts we shall present in the next section a detailed analysis of the solid-fluid interface.

Before concluding the present section, it would be appropriate to investigate the stability against diffusion in a multicomponent system. Such an analysis is feasible in a very general manner in a one-dimensional system. Calculus of variations tells us (10, 12) that for Equation (12) to yield a minimum of Equations (7) and (8) one of the conditions to be satisfied is the so-called Legendre condition. The Legendre condition states that the following matrix should be positive definite

\* The author is grateful to Dr. E. W. Hart for indicating to him how one could obtain a better set of measurable stresses than those presented in references 5 and 6.

\*\* Assuming of course that  $N_i^*$ ,  $\gamma_i^*$  and  $\psi_i$  are known as well as the necessary equations of state for  $\mu_i$ ,  $\mu_{ij}$ .

$$\left\| \frac{\partial^2 U}{\partial \rho_{i,z} \partial \rho_{m,z}} \right\| > 0 \quad (29)$$

It should be noted that this condition is different from that of a uniform system since then one would require rather that

$$\left\| \frac{\partial^2 U}{\partial \rho_i \partial \rho_m} \right\| > 0 \quad (30)$$

### SOLID-FLUID INTERFACE-ADSORPTION

We have presented a general thermodynamic treatment; for computational purposes a thermodynamic model is required. For simplicity we shall restrict ourselves to the one component system and the square gradient theory.

$$U = U_H + \frac{C}{2} \left( \frac{d\rho}{dZ} \right)^2 \quad (31)$$

Using the stability condition, Equation (29), it is seen that  $C > 0$ . The equations describing the system are now given by

chemical equilibrium

$$C \frac{d^2 \rho}{dZ^2} = RT \ln f/f_0 + \psi^* \quad (32)$$

measurable stresses

$$P_{zz} = p - C\rho \left( \frac{d^2 \rho}{dZ^2} \right) \quad (33)$$

surface properties

$$\Gamma = \int (\rho - \rho_0) dZ \quad (34)$$

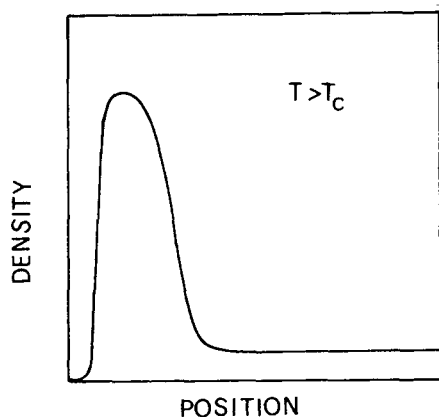


Fig. 1. Density profile; refined model ( $T > T_c$ ) or crude model ( $T > T_c$ ).

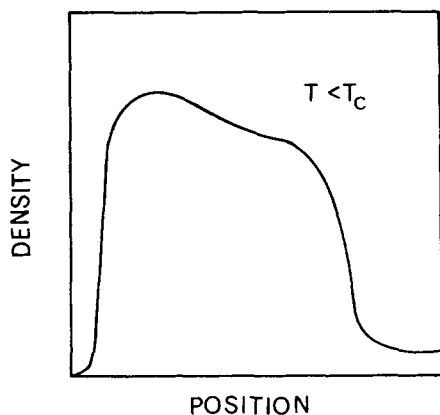


Fig. 2. Density profile; refined model ( $T < T_c$ ).

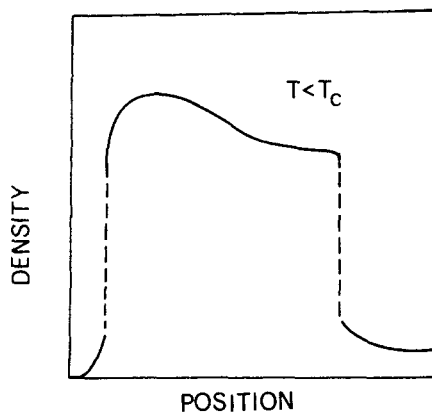


Fig. 3. Density profile; crude model ( $T < T_c$ ).

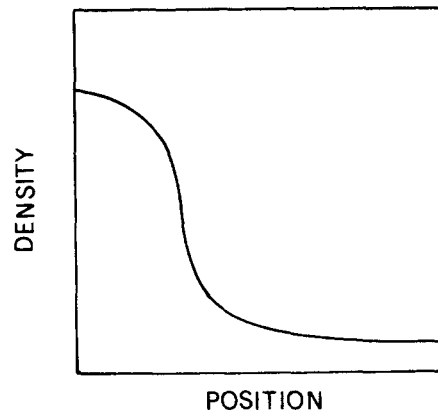


Fig. 4. Density profile; crude model, reference 8.

$$\sigma = \int \left[ C \left( \frac{d\rho}{dZ} \right)^2 + \int \rho \psi_{,z} dZ \right] dZ \quad (35)$$

In the present case the external field  $\psi$  corresponds to the so-called Van der Waal forces (13) ( $\psi(\infty) = 0$ ,  $\psi(0) = \infty$ ). In the case of a finite system,  $\rho_0$  is the average density of the system while  $f_0$  is determined using the conservation of mass [that is, Equation (8)]. The boundary conditions associated with Equation (32) are of type 2,  $\rho(0) = 0$ ,  $\rho(\infty) = \rho_*$ .<sup>\*\*</sup> In the case of a semifinite system it is easily shown that  $\rho_0$ ,  $f_0$  are respectively the density and fugacity of the bulk fluid ( $Z = \infty$ ). Should Equation (32) be solved one would obtain a density profile such as that shown in Figure 1 for  $T > T_c$  and as that shown in Figure 2 for  $T < T_c$ . The model proposed by Metiu and Ruckenstein is equivalent to set  $C = 0$  in Equation (32). In such instances we have

$$RT \ln \frac{f_1}{f_0} + \psi = 0 \quad (36)$$

Using this type of equation one would obtain a density profile similar to that presented in Figure 1 for  $T > T_c$  and one such as in Figure 3 if  $T < T_c$ . This case has already been presented in reference 4. We first note that these are different from that presented by Metiu and Ruckenstein (Figure 4) and we feel that the density profiles presented here are more realistic close to the wall and are consistent with available statistical mechanical calculations (14). We notice in Figure 3 two sharp phase transitions for the crude model [Equation (36)] and two continuous phase

\* Here we have introduced a fugacity for convenience.

\*\* This type of boundary condition is in agreement with available statistical theories.

transitions (Figure 2) for the more refined model [Equation (32)]. The net result is that the crude model does not have a surface tension and as a result cannot describe capillary condensation.

It should be noted that Equation (36) is very similar to the Polanyi potential model (15). In the case of a perfect gas, Equation (36) reduces properly to the Halsey equation

$$\rho = \rho_\infty \exp \left( - \frac{\psi}{RT} \right) \quad (37)$$

which was obtained using statistical arguments (16). If one considers Equation (36) as a first-order approximation one could obtain from Equation (32) using a method of successive approximation better and better approximations. This is possible since both Equations (32) and (36) satisfy the same surface conditions. Differentiation of Equation (36) with respect to  $Z$  and elimination of the second derivative in Equation (32) yield a second-order approximation.

$$RT \ln \frac{f_2}{f_0} = - \left[ \psi + C \left[ \frac{\psi_{,zz} + \frac{1}{RT} \left[ \frac{\psi_{,z}}{\frac{\partial \ln f_1}{\partial \rho}} \right]^2 \frac{\partial^2 \ln f_1}{\partial \rho^2}}{RT \frac{\partial \ln f_1}{\partial \rho}} \right] \right] \quad (38)$$

It should be mentioned that Equation (38) is very similar in structure to those developed by Steele for dense fluids (14). The method used by Steele was statistical mechanics. One can also see from Equation (38) that the higher the temperature the more accurate is Equation (36).

## OBJECTIONS TO THE CONTINUUM APPROACH

As pointed out by Metiu and Ruckenstein (8) one of the objections to the continuum approach raised by Widom (2) is that in equations such as (32) one must use  $\mu_H(f_H)$  in a range where  $\partial \mu_H / \partial \rho$  is negative, that is, in the unstable portion of an homogeneous fluid. Metiu's reply is that  $\mu_H$  is a virtual chemical potential, not a real one. In the discussion following Equation (15) we saw that this was indeed the case. Furthermore, we have seen [Equations (29) and (30)] that in the case of nonuniform systems the stability criterion would not be  $\partial \mu / \partial \rho$  but rather  $\partial \mu_Z / \partial \rho_Z$  and as a result empirical equations such as Equation (31) must be evaluated on their own using the proper stability criterion, Equation (29). Even though the above two arguments attempt to answer Widom's objection we feel they are incomplete since whenever one wants to formulate models such as Equation (31) one must start from equations of state available for uniform fluids. The real question then is: do homogeneous equations of state, showing continuous isotherms as one goes from the vapour to the liquid phases on a phase diagram, exist? We feel that such equations do exist; probably the most striking evidence for their existence lies in the computer experiments of Verlet (17) where this type of equation of state has been obtained using molecular dynamics. We feel that the latter result may provide a possible answer to Widom's objection. Finally, it should be pointed out that the present theory is still approximate since it does not include effects of density gradients of all orders. In addition to this, theories based upon a continuous approach can only provide us with a smeared description of nonuniform systems.

## CONCLUSIONS

The thermodynamic treatment of Hart has been extended to multicomponent reactive systems. The theory has been applied to adsorption and provides us with a better description of the solid-fluid interface than that provided in reference 8. It is obvious that the alternate theory of reference 8 could be corrected easily to provide one with results as general as those presented here.

## ACKNOWLEDGMENT

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## NOTATION

$A_j$	= element of boundary surface in the $j$ direction
$B_i$	= chemical compound
$C$	= constant in the square gradient theory
$e$	= dilatation
$e_{jk}$	= strain
$f$	= fugacity
$M_i$	= molecular weight
$n$	= number of components
$N$	= molar mass
$N^\circ$	= initial molar mass
$p$	= pressure
$p_{jk}$	= stresses
$\underline{r}$	= position vector
$R$	= gas constant
$S$	= entropy density
$\mathcal{S}$	= total entropy
$t$	= number of chemical transformations
$T$	= temperature
$U$	= internal energy density
$\mathcal{U}$	= total internal energy
$V$	= total volume
$x_j$	= coordinate number
$Z$	= coordinate number

## Greek Letters

$\delta$	= variational operator
$\delta_{jk}$	= Kronecker delta
$\xi$	= extent of a reaction
$\mu_i$	= density potential ( $\mu$ -one component system)
$\mu_{ij}$	= gradient density potential ( $\mu_j$ -one component system)
$\nu_{ia}$	= stoichiometric coefficient
$\rho$	= density
$\rho_{,j}$	= density gradient
$\sigma$	= interfacial tension
$\varphi$	= field created by a density gradient
$\psi$	= external field

## Subscripts

$c$	= critical point
$H$	= homogeneous contribution
$i, m$	= a chemical species of type $i, m$
$j, k, l$	= in the $j, k, l$ direction
$0$	= extrinsic and measurable property
$1, 2$	= first- and second-order approximation
$,$	= differentiation ( $\rho_{,z} = \partial \rho / \partial Z$ )
$\infty$	= conditions at an infinite distance away from a solid surface

## Superscript

$0$	= extrinsic and measurable property
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# The Blending Efficiencies of Some Impellers in Batch Mixing

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Mixing times for the blending of viscous liquids with tracer materials have been measured in a mechanically agitated tank using two conventional agitator types (turbine, helical ribbon) and a series of novel tubular-type agitators. The range of conditions studied were: viscosity = 1 to 70,000 cP, agitator Reynolds number = 5 to  $10^5$ , tank diameter = 11.5 in. It was found that in terms of agitator power requirement, the tubular agitators were generally more efficient for viscosities greater than 100cP than the others for the blending process.

The effects of pseudoplastic non-Newtonian fluid behavior, of tank-baffles, and of tank size on mixing times have also been examined in several of the above cases.

"What is most needed in agitation research at the present time is not extensive correlation of power consumption of inefficient mixing devices, but intensive study directed toward design and selection of impellers which will utilize the power invested more efficiently" (1). "For economy of agitation it is, however, important to ensure that the impeller operates for the shortest time under such conditions which permit the required agitation. When judging the power consumption of an impeller it is then necessary to take into account the total energy consumption during the time necessary for the accomplishment of the required mixing effect" (2).

Relatively little work has been done on the comparative evaluation of various impellers with regard to the efficiency with which they utilize power. Within this context, an attempt has been made to compare a few conventional impeller types with some novel nonconventional ones. Specifically, the relationship between mixing time and power consumption (as the two basic parameters characterizing mixing efficiency of the impellers) was experimentally examined. The experiments were limited to the blending of miscible liquids of the same viscosity and density.

sel, 11½ in. in diameter, with standard or almost standard geometric configurations (2). For scaling up inferences, a few runs were also performed in a 16-in. diameter vessel. The liquid height was fixed at one tank diameter in all experiments, and the impellers were operated in the usual central position. For baffled conditions, the common arrangement of four wall baffles each of width equal to 1/10 the tank diameter, were used.

The following impeller types were tested (for additional details, see Figure 1 and Notation).

1. 6-bladed flat turbine:  $D = 4$  in.,  $C = 1/3T$ ,  $w = 1$  in.,  $h = 0.75$  in.

2. helical ribbon with single flight:  $D = 8$  in.,  $w = 0.75$  in.,  $h = 11.5$  in., pitch = 4 in.

3. single tubular agitator I:  $D = 6.25$  in., I.D. of main tube = 1.03 in. (O.D. = 1.2 in.), I.D. of arms = 0.63 in. (O.D. = 0.75 in.),  $C = 2.5$  in.,  $C' = 8.5$  in.

4. single tubular agitator II:  $D = 7.0$  in., I.D. of main tube = 0.69 in. (O.D. = 0.75 in.), I.D. of arms = 0.44 in. (O.D. = 0.50 in.),  $C = 1.5$  in.,  $C' = 8$  in.

5. double tubular agitator I:  $D$  (upper) = 7.0 in.,  $D$  (lower) = 4.5 in., I.D. of main tube = 0.69 in. (O.D. = 0.75 in.), I.D. of arms (upper) = 0.44 in. (O.D. = 0.50 in.), I.D. of arms (lower) = 0.31 in. (O.D. = 0.375 in.),  $C = 1.5$  in.,  $C_1 = 8$  in.,  $C_2 = 4$  in.

6. double tubular agitator II:  $D$  (upper and lower) = 7.0 in., I.D. of main tube = 0.69 in. (O.D. = 0.75 in.), I.D. of arms (upper and lower) = 0.44 in. (O.D. = 0.50 in.),  $C = 1.5$  in.,  $C_1 = 8$  in.,  $C_2 = 4$  in.

## EXPERIMENT

Most of the experiments were conducted in a cylindrical ves-