COMMUNICATIONS TO THE EDITOR

An Example of the Use of Combined Models: Mixing in a Tubular Reactor with Return Bends

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Recently, some data on axial mixing in a tubular high-pressure reactor with a large proportion of return bends were presented by Carter and Bir (*4*). They found, as would be qualitatively expected, that the axial mixing was somewhat greater than that observed for straight pipe flow. Thus, they concluded that more experimenetal work is needed for systems of this type.

It would seem that a reactor of this type could be broken up naturally into two sections. First, the straight, tubular part, and second, the return bends. The axial mixing in the straight portion could be found then from correlations such as Levenspiel's (*6*), and the bends might be approximately treated as perfect mixers. This latter approximation is probably a rather poor one, since the bends would not act as perfect mixers under all flow conditions. A better representation could be found by experimentally measuring mixing in bends, but this information is not available at the present time. Thus the results of the analysis to be presented will be limited to relatively crude estimates of the total axial dispersion and will serve more as an indication of what could be done if more data were available.

Models of this type, which were given the name of *mixed models* or *combined models* (*9*), and organized and generalized by Levenspiel (*7*) have been found to be useful in other types of complex flow situations. Thus, one of the earlier applications was to the flow in fluidized beds as discussed, for example, by May (*10*). Another application to flow in real stirred tanks was by Cholette and Cloutier (*5*). The advantages of such a procedure are mainly that the mixing characteristics of each individual section may be obtained from standard correlations such as those for empty pipes, perfectly mixed regions, etc., and then combined for the total system. Thus, the very extensive experimental testing of every possible type of system made up of combinations of the single elements, which would be almost impossible, is avoided. The correlation of mixing data then becomes less voluminous.

DEVELOPMENT OF MODELS

The model that lumps the straight-pipe and return-bend mixing into one effective axial dispersion coefficient, as used by Carter and Bir, is represented

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Kihara and Lennard-Jones Parameters for the Isomeric Hexanes

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Precise methods of estimating physical data are quite useful and are often the sole source of such information for the process engineer. One such method is the estimation of transport properties (*1* to *4*) using the parameters of the Lennard-Jones intermolecular potential (*5*):

$$U(r) = 4\epsilon\left[\left(\frac{\sigma}{r}\right)^{12} - \left(\frac{\sigma}{r}\right)^{6}\right] \tag{1}$$

Another technique is the estimation of vapor-liquid equilibrium constants (*6*) using parameters of the Kihara intermolecular potential (*7*):

$$U_p = U_o\left[\left(\frac{\rho_o}{\rho}\right)^{12} - 2\left(\frac{\rho_o}{\rho}\right)^{6}\right] \tag{2}$$

The Lennard-Jones potential assumes a spherical molecule with *r*-to-*r* spacing between molecules. Although specifically designed to describe spherical molecules, it is applicable to nonspherical molecules, especially over short temperature ranges.

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TABLE 1. KIHARA AND LENNARD-JONES PARAMETERS FOR THE ISOMERIC HEXANES

Compound	l_1	l_2	l_3	V_o	S_o	M_o	ρ_o	U_o/k	σ(Å)	ϵ/k (°K.)
2,2-dimethylbutane	0.862	0.543	2.51	3.35	39.6	17.65	2.50	1060	7.614	266
2,3-dimethylbutane	0.783	0.788	1.57	4.02	30.8	19.65	2.50	1070	6.751	412
2-methylpentane	0.783	0.684	2.51	3.35	36.7	18.4	2.50	1130	8.065	267
3-methylpentane	0.862	0.543	2.51	3.35	35.3	17.6	2.50	1178	8.587	265

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The Relationship between the Froude and Reynolds Numbers in Falling Vertical Films

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Numerous workers in the field of falling liquid films have accepted the Froude number as a very appropriate dimensionless group for correlating their experimental results (*1*, *2*, *14*). Jackson (*14*) used the Froude number in the form

$$N_{Fr}' = \frac{V}{(gm)^{1/2}} \tag{1}$$

as a criterion for wave inception on films. His definition of the Froude number in this way must have been influenced by the fact that long gravity waves in a shallow, horizontal, open

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channel propagate with a velocity (*18*)

$$V_w = (gz)^{1/2} \qquad (2)$$

if the amplitude of the wave is considerably smaller than the depth of the water at that point.

Jackson has argued therefore that a Froude number, N_{Fr}', of unity represents a critical condition at which the average velocity of the falling liquid is equal to the velocity of wave propagation, and that it might be expected, therefore, that this condition would be a critical point for the appearance of waves in film flow.

Belkin et al. (*1*), using the same definition of the Froude number, have expressed a dimensionless film thickness parameter as a function of both the Reynolds and Froude numbers (for laminar flow), thus

$$m\, g^{1/3}\, \nu^{-2/3} = 0.397\ (N_{Re}/N_{Fr}')^{2/3} \qquad (3)$$

Brauer (*2*) has used another dimensionless criterion including the Weber group and both the Froude and Reynolds groups, that is the reduced Weber number, $N_{We}/N_{Re}\ N_{Fr}$, for correlation of film turbulence inception data. He has deduced that the critical Reynolds number of turbulence inception is given by

$$N_{Ret} = 9 \left[\frac{N_{Re} \cdot N_{Fr}}{N_{We}} \right]^3 \cdot N_{KF}^{-1} \qquad (4)$$

where N_{KF} is yet another dimensionless group, known as the *film number* defined by

$$N_{KF} = \frac{\rho\sigma^3}{g\mu^4} \qquad (5)$$

It will be indicated here that one does not gain any advantage by introducing simultaneously both the Reynolds and Froude criteria for the correlation of experimental data on falling film flow, and that in most cases it is sufficient to use one criterion only because the use of either number may be employed to the exclusion of the other for laminar and wavy flow.

STEADY LAMINAR FLOW

The Froude number for film flow is commonly taken as

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Measurements of Slip Velocity in Two-Phase Mercury Flows

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The vapor volume fraction has been commonly used in two-phase systems as a parameter. It is also an important characteristic in evaluating the performance of a forced convection boiling system. For metallic fluids a large portion of the volumetric flow may be in the vapor state even at a relatively low vapor quality (weight percent) because of the large ratio of densities of liquid to vapor ($\rho'/\rho'' = 4{,}000$ for mercury). Consequently, the void fraction depends on the slip velocity ratio, u''/u'. No analytical prediction of void fraction or slip velocity ratio in a two-phase flow system is available. Bankoff suggested a variable-density single-fluid model for turbulent two-phase flow in a pipe (*1*) which predicts the slip velocity ratio for a steam-water system in bubble or slug flow patterns to be

$$u''/u' = (1-\alpha)/(K-\alpha), \qquad 0.5 < K < 1 \qquad (1)$$

Although this simplified equation was substantiated by experimental data in the literature (*2*) for values of vapor volumetric flow concentration, β, up to 0.8, the equation would not be valid beyond this value even for the steam-water system. For two-phase metallic fluids such as mercury, the reported experimental values in Russian literature indicate a considerable deviation from the aforementioned equation. This is shown in Figure 1 where the void fraction is plotted against β.

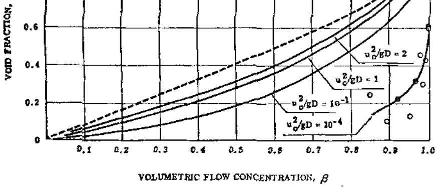

Fig. 1. Correlation of void fraction with volumetric flow concentration.

By definition,

$$u'' = u_o''/\alpha, \quad u' = u_o'/(1-\alpha),$$

and

$$\beta = \frac{u''_o}{u''_o + u_o'}$$

It follows that the relationship between α and β can be expressed by

$$\alpha/\beta = (1-\alpha)\ (u'/u'') + \alpha = K \qquad (2)$$

The mercury vapor-liquid system deviates from the linear relationship at β equals 0.4. This paper reports measurements of void fraction in a mercury-liquid system for high β values ($\beta > 0.8$).

EXPERIMENTAL MEASUREMENTS

A schematic diagram for the experimental apparatus is shown in Figure 2. Mercury is circulated by an a.c. electromagnetic pump through the electrically heated preheater and boiler in a vertical stainless steel 321 circular test section (0.25-in. O.D. with a 0.049-in. thick wall). Mounted above the boiler is a two-millicurie radium source and ion chamber for void fraction detection by the gamma-ray attenuation method. This was accomplished in a single-shot method as described by Richardson (*3*). To obtain accurate readings of the average void fraction, a section of the tube is enlarged through a smooth, insulated transition piece (3 in. long) to a tube

TABLE 1. RESULTS OF VOID FRACTION MEASUREMENTS

Run no.	u_{o6} (ft./sec.)	$x_6 = x_7$	T_{sat} (°F.)	α_7	β_7	(u_{o7}''/u_{o7})	(u_{o7}^2/gD)	ϕ_7
32	1.85	0.008	665	0.325	0.964	26.2	1.2×10^{-4}	63.9
33	1.73	0.004	654	0.23	0.920	11.7	1.1×10^{-4}	50.6
34	1.76	0.009	655	0.45	0.966	27.4	1.1×10^{-4}	40.6
35	1.72	0.021	656	0.43	0.990	78.0	1.1×10^{-4}	109
36	0.36	0.208	652	0.61	0.999	800	0.5×10^{-5}	780
37	0.31	0.257	652	0.62	0.999	990	0.4×10^{-5}	993
40	1.68	0.003	658	0.11	0.894	8.6	1.0×10^{-4}	83.3
41	1.68	0.006	655	0.12	0.951	20.0	1.0×10^{-4}	174
43	2.03	0.003	663	0.25	0.852	5.9	1.5×10^{-4}	30.0
44	2.02	0.019	668	0.29	0.986	68.3	1.5×10^{-4}	178

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$$N_{Fr} = \frac{V^2}{g\,m} \qquad (6)$$

Equations for the viscous flow of liquid films down a vertical plate have been established by Nusselt (*20*). Using his expressions for the mean velocity of the film, *V*, and the average film thickness, *m*, one obtains

$$N_{Fr} = \frac{\rho^2\, g\, m^3}{9\,\mu^2} = \frac{1}{3}\,\frac{Q}{\nu} \qquad (7)$$

There are two definitions of Reynolds number in common use:

$$N_{Re'} = \frac{Q}{\nu} \qquad (8a)$$

and

$$N_{Re} = \frac{4Q}{\nu} \qquad (8b)$$

When Equations (7) and (8*b*) are combined, the Reynolds number may be expressed in terms of the Froude number as

$$N_{Re} = 12\, N_{Fr} \qquad (9)$$

The above expression gives a very simple relationship between the Reynolds and Froude numbers. It has been derived by combining Equations (7) and (8*b*) and is therefore valid when these expressions are applicable. Equa-

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of solids-ductility, flexibility, strength, stress-strain relationships, and related subjects. Chapter 6 treats the various magnetic properties which are exhibited by materials at low temperatures. Chapter 7 is concerned with thermal and transport properties, such as, heat capacity, coefficient of expansion, and thermal and electrical conductivity. Chapter 8 deals with superconductivity, first describing the phenomenon and then presenting the BCS (after authors Bardun, Cooper, and Schrieffer) theory, which appears to offer the best explanation of this amazing phenomenon. The final chapter considers briefly some of the practical applications of cryogenics. The applications discussed cover the fields of rocketry, the life sciences, the use of bubble chambers in high-energy particle physics, the production of high-strength magnetic fields, and the use of low temperatures in infrared detectors, masers, lasers, and cryotrons.

The book is well written, authoritative, and readable and might be recommended to anyone interested in gaining a broad, descriptive picture of the field of cryogenics as it exists today.

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ERRATA

In Equation (14) of the article, "The Application of Boundary-Layer Theory to Power-Law Pseudoplastic Fluids: Similar Solutions" by W. R. Schowalter, which appeared on page 24 of the March, 1960, issue of the *A.I.Ch.E. Journal,* the second term should read

$$\frac{g^{n+1}\, W^o}{(U^o)^n}\,\frac{\partial U^o}{\partial z^o}\,[F'\, G' - 1]$$

The subsequent analysis for three-dimensional boundary layers which possess similar solutions is affected. One is led to the conclusion that if W^o and U^o are proportional, similar solutions are possible for those classes of potential flows which yield similar solutions in two-dimensional flow.

The author is grateful to Professor J. N. Kapur, who brought this error to his attention.

The article, "Behavior of Non-Newtonian Fluids in the Inlet Region of a Channel" by Morton Collins and W. R. Schowalter, which appeared on page 98 of the January, 1963, issue of the *A.I.Ch.E. Journal* contains an error on page 102. The eleventh line of the first column of that page should read, "found to be 0.069 and 0.68 . . . ."

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Application of Benedict-Webb-Rubin equation of state to H_2S-hydrocarbon mixtures, Simon, Ralph and J. E. Briggs, **A.I.Ch.E. Journal, 10,** No. 4, p. 548 (July, 1964).

Key Words: Equation of State-6, 7, 9, Hydrogen Sulfide-8, PVT Relationships-6, 7, 9, Pressure-6, Temperature-6, Density-6, Steepest Descent Method-10, Optimization-10, Gradients-10.

Abstract: The authors have developed a set of hydrogen sulfide constants for the BWR equation of state. The constants are compatible with the hydrocarbon, nitrogen, and carbon dioxide constants previously reported in the literature and are intended to be used for gaseous mixtures containing up to 20 mole % hydrogen sulfide.

Accuracy of calculations made with the hydrogen sulfide constants was tested by comparing observed pressures for gaseous systems with pressures predicted by the BWR equation. This comparison showed that for seventy-one data points for multicomponent systems the average absolute deviation was 2.8% .

The mathematical method used to obtain the hydrogen sulfide constants is fully described.

Numerical and experimental study of damped oscillating manometers: II. non-Newtonian fluids, Biery, John C., **A.I.Ch.E. Journal, 10,** No. 4, p. 551 (July, 1964).

Key Words: A. Manometers (Damped Oscillating)-9, Non-Newtonian Fluids-9, Transient Behavior-8, Viscosity-6, Damping Factor-7, Frequencies-7. B. Simulation (Mathematical)-8, Numerical Integration-10, Equation of Motion-10, Velocity Profiles-7, Computer (Digital)-10, Digital Computer-10, Convergence-7. C. End Effects (Manometer)-8, Flow Reversal-8. D. Fluids (Manometer)-9, Newtonian, Non-Newtonian, Natrosol-Hi-9, Natrosol-Medium-9, Polyox-9. E. Rheology-8, Viscosity-8, Newtonian Fluids-9, Non-Newtonian Fluids-9, Cone and Plate Viscometer-10, Sisko Model-8, Oldroyd Model-8.

Abstract: The time-dependent behavior of non-Newtonian oscillating manometers was studied experimentally and mathematically. The manometers were succssfully simulated by numerically integrating the axial component of the equation of motion to produce the radial velocity profiles. The simulations showed that steady state rheological models can be successfully used in slowly oscillating systems.

Joule-Thomson effects for nitrogen-methane mixtures, Stockett, A. L., and L. A. Wenzel, **A.I.Ch.E. Journal, 10,** No. 4, p. 557 (July, 1964).

Key Words: Joule-Thomson Effect-7, Virial Coefficient-7, Interaction Effect-7, Nitrogen-10, Ethane-10, Mixture-8, Pressure-6, Temperature-6, Phase-5, Rule-9, Enthalpy-8, Predict-10, Isenthalp-8, Valve-10, Cryostat-10, Compressor-10, Composition-6.

Abstract: Joule-Thomson effects were measured for ethane and five nitrogen-ethane gas mixtures ranging in composition from 28 to 95% nitrogen. Pressures ranged from 2,400 to 50 lb./sq. in. gauge and temperatures from 25° to —100°C. The data covered the temperature and pressure region adjacent to the vapor-liquid phase boundary and penetrated into this two-phase region.

Zero-pressure Joule-Thomson coefficients were evaluated and found to be lower by an average of 15.7% than values predicted by a linear combination rule. Use of a combination rule based on the virial method represents the data with an average absolute deviation of 4.6%.

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tion (8*b*) is valid for all rates of flow, but Equation (7) is restricted by the limits of application of Nusselt's equations for the average film velocity, *V*, and the film thickness, *m*, in steady laminar flow. In Nusselt's work the film was assumed to be in perfectly viscous flow where no shear or wave motion existed at the liquid surface, and therefore his equations should not be extended too far into the wavy region of flow (*15, 22, 8, 10, 6, 24*).

Wavy Flow

Since waves and ripples, sometimes of considerable amplitude, are usually present on most falling liquid films at all but the very lowest flow rates, the flow behavior in the wavy or pseudolaminar region is sufficiently different from the steady laminar one that the value of the Froude group is bound to be affected. The resulting change in the value of the Froude number may now be established quantitatively.

When the mean film velocity, *V*, is eliminated from Equation (6) by the use of the expression, $V = Q/m$, one obtains

$$N_{Fr} = \frac{V^2}{g\,m} = \frac{Q^2}{m^3\,g} \qquad (10)$$

The above equation expresses the Froude number as a function of the average film thickness, *m*, which may be evaluated according to Kapitsa's (*15, 22*) theory of the wavy film flow as

$$m^3 = 2.4\,\frac{\nu Q}{g} \qquad (11)$$

When the above expression is substituted for *m* in Equation (10)

$$N_{Fr} = \frac{Q^2}{m^3 g} = \frac{1}{4}\,\frac{Q}{\nu} \qquad (12)$$

By combining Equations (12) and (8*b*), one obtains

$$N_{Re} = 16\,N_{Fr} \qquad (13)$$

The above equation applies in the pseudolaminar and transitional regions of flow (*22, 24, 25*) up to the critical Reynolds number for the inception of turbulence. The values given in the literature for the Reynolds number at which turbulence appears in film flow seem to vary. For water-films the values quoted range between $N_{Ret} = 1{,}000$ and $N_{Ret} = 2{,}400$, where N_{Ret} is the Reynolds number of transition between viscous and turbulent flow (*2 to 4, 6, 7, 9 to 11, 13, 14, 16, 17, 19, 21 to 23, 26*).

The above equations are limited to vertically falling films. It may be worthwhile to mention that the cosine of the angle of inclination with the vertical becomes a factor for nonvertical surfaces (*12*).

Turbulent Flow

Comparatively little theoretical work has been done on turbulent film flow. Dukler and Bergelin (*6*) have presented an expression for the film thickness function, $\psi(m)$, based on the universal velocity profile for full-pipe flow. The author has recently extended this method and developed generalized velocity distribution equations for all regimes of vertical film flow (*22*).

According to this concept

$$m^3 = \frac{\eta^2 \nu^2}{g} \qquad (14)$$

When the above expression is substituted for m in Equation (10), one obtains

$$N_{Fr} = \frac{1}{\eta^2}\left(\frac{Q}{\nu}\right)^2 \qquad (15)$$

Since $\frac{Q}{\nu} = N_{Re}/4$,

$$N_{Fr} = \left(\frac{1}{2\eta}\right)^2 \cdot N_{Re}^2 \qquad (16a)$$

or

$$N_{Fr}' = \frac{N_{Re}}{2\eta} \qquad (16b)$$

A simpler relationship between N_{Re} and N_{Fr} may be obtained by introducing another correlating parameter, that is the Fanning friction factor. This may be expressed in the form (*5, 27, 1*).

$$\frac{f}{2} = C + k\,N_{Re}^n \qquad (17)$$

and since $1/N_{Fr} = f/2$, therefore

$$N_{Fr}^{-1} = C + k\,N_{Re}^n \qquad (18)$$

where C and k are constants and n is a simple index.

NOTATION

f = Fanning friction factor = $\frac{2R}{\rho V^2}$, dimensionless

g = acceleration owing to gravity, cm./sec.^{2}

m = average film thickness, cm.

N_{Fr} = Froude number = $\frac{V^2}{gm}$, dimensionless

N_{Fr}' = reduced Froude number = $\frac{V}{(gm)^{1/2}}$, dimensionless

N_{KF} = film number = $\frac{\rho\sigma^3}{g\mu^4}$, dimensionless

N_{Re} = Reynolds number = $\frac{4Q}{\nu}$, dimensionless

N_{We} = Weber number = $\frac{mV^2\rho}{\sigma}$, dimensionless

Q = volumetric rate of flow per unit wetter perimeter, cc./sec. cm.

R = shear stress in liquid at boundary surface, g./cm. sec.^{2}

V = average film velocity, cm./sec.

INFORMATION RETRIEVAL

(Continued from page 598)

Monte Carlo solution of radiant heat transfer in a nongrey nonisothermal gas with temperature dependent properties, Howell, John R., and Morris Perlmutter, **A.I.Ch.E. Journal, 10,** No. 4, p. 562 (July, 1964).

Key Words: Radiation-8,7, Heat Transfer-8, 2, 7, Monte Carlo-10 8, Variable Properties-6, 8, Radiant Heat Transfer-8, 2, 7, Gas Radiation-8, Hydrogen-5, Radiation Absorption Coefficient-6, Wave Length Dependence-6, 2, Temperature Dependence-6, 2, Approximate Solutions-6, 2, 10, Emissive Power Distribution-2, 7, IBM-7090 Computer-10, Thermal Radiation-8, 7.

Abstract: The Monte Carlo method is applied to the problem of determining the radiant heat transfer and emissive power distribution in an absorbing emitting nongrey gas with temperature dependent properties contained between infinite parallel black walls. The gas emissive power distribution is presented for the cases of no heat source and a parabolic distribution of heat sources in the gas. Calculations are carried out for hydrogen at temperatures in the range 5,000° to 12,000°K. A comparison is made with various approximate methods and with the limiting diffusion and transparent solutions. The results indicate that the approximate solutions may be in considerable error.

Kinetics of the Catalytic Oxidation of Sulfur Dioxide, Davidson, Burton, and George Thodos, **A.I.Ch.E. Journal, 10,** No. 4, p. 568 (July, 1964).

Key Words: Sulfur Dioxide-1, Sulfur Trioxide-2, Oxidation-10, Catalyst-5, Differential Reactor-10, Vanadium Pentoxide-5, Orsat Gas Analyzer-10, Chemisorption-4, Langmuir-Hinshelwood, Hougen-Watson-10.

Abstract: Experimental studies on the catalytic oxidation of sulfur dioxide have been carried out at 643°, 672°, and 722°F. and 1.47 atm. in a differential bed reactor in the presence of nitrogen with a supported vanadium pentoxide catalyst. Interfacial compositions were calculated and were used to show that the apparent rate controlling step involves the surface reaction between chemisorbed sulfur dioxide and chemisorbed atomic oxygen to form chemisorbed sulfur trioxide and a vacant site. The chemical velocity constant and the adsorption equilibrium constants for all reactants and products (including nitrogen) were found to be positive and produced linear relationships with temperature, consistent with the Arrhenius theory.

Reaction rates were calculated and compared with the eight experimental values for each temperature, producing deviations of 6.9% at 643°F., 25.0% at 672°F., and 15.6% at 722°F.

A kinetic study of sulfur dioxide in aqueous solution with radioactive tracers, Wang, J. C., and D. M. Himmelblau, **A.I.Ch.E. Journal, 10,** No. 4, p. 574 (July, 1964).

Key Words: Sulfur Dioxide-1, Reaction Kinetics-1, Reaction Data-1, Kinetic Models-8, Coefficient Estimation-2, Rate Constants-9, Water-5, Aqueous-5, Solution-5, Liquid-5, Temperature-6, Concentration-6, Radioactive-9, Tracer-9, Bisulfite-1.

Abstract: A kinetic study was made of the homogeneous reactions of sulfur dioxide in water with a radioactive tracer technique. Data were collected from 0° to 20°C. and from a pH of 1.2 to 4.3. Analysis of different kinetic models demonstrated that the classical reaction $SO_2 + H_2O \rightleftharpoons H^+ + HSO_3^-$ represented the data best from a macroscopic point of view. Forward and reverse rate constants are reported for the first time for this system.

Heat transfer coefficients for a hot gas oscillating at high amplitudes in a cylindrical Chamber, Horton, M. D., J. L. Eisel, and G. L. Dehority, **A.I.Ch.E. Journal, 10,** No. 4, p. 580 (July, 1964).

Key Words: Oscillations-2, Oscillatory Combustion-2,8, Oscillating Gas-6, Mean Pressure-6, Heat Transfer Coefficient-7, Transient Heat Transfer-7, Combustion-10, T Burner-10.

Abstract: Estimates are made of the average heat transfer coefficient which exists for a high-temperature combustion gas oscillating in a cylindrical steel chamber. The estimates are made from the rate of change of the mean chamber pressure in the sealed system. It is found that the oscillations increase the convective heat transfer coefficient by about 100% at 2,000 cycles/sec., and the effect decreases with frequency until at 8,000 cycles/sec. the oscillations do not change the heat transfer.

V_w = water wave velocity, cm./sec.
z = height of water surface, cm.

Greek Letters

μ = absolute viscosity, poises
ν = kinematic viscosity, stokes
ρ = density, g./cc.
σ = dynamic surface tension, dyne/cm.
η = magnitude of the universal distance parameter at the film surface, dimensionless
$\psi(m)$ = film thickness function = $mg^{1/3} (\rho/\mu)^{2/3}$

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