Voin Kohlenwasserstoffen aus Kohlenoxid und Wasserstoff," Brennst Chem., 48(3), 78 (1967).

Pikios, C. A., and D. Luss, "Isothermal Concentration Oscillations on Catalytic Surfaces," Chem. Eng. Sci., 32, 191 (1977).

Ponec, V., "Some Aspects of the Mechanism of Methanation and Fischer-Tropsch Synthesis," Cat. Rev. Sci. Eng., 18(1), 151 (1978).

Rao, V. U. S., and R. J. Gormley, "Make Olefins from Syn-Gas," *Hydr. Proc.*, 139, (Nov., 1980a).

thesis Gas Conversion to Gasoline Range Hydrocarbons over Medium Pore Zeolite Catalysts Containing 3d-Metals and Bimetallics," ACS, Div. of Fuel Chemistry, Houston, 25, 2 (1980b).

Raupp, G. B., and W. N. Delgass, "Mossbauer Investigation of Suppored Fe Catalysts. III In Situ Kinetics and Spectroscopy during Fischer-Tropsch Synthesis," J. Cat., 58, 361 (1979).
Schmitz, R. A., and G. T. Renola, and A. P. Zioudas, "Dynamic Complex-

Schmitz, R. A., and G. T. Renola, and A. P. Zioudas, "Dynamic Complexities in the Catalytic Oxidation of Hydrogen on a Nickel Film," Kin. of Phys. Osci., Aachen, 1, 221 (1979).

Sexton, B. A., and G. A. Samorjai, "The Hydrogenation of CO and CO₂ over Polycrystalline Rhodium; Correlation of Surface Composition Kinetics and Product Distributions," J. Cat., 46, 167 (1977).

Sheintuch, M., and R. A. Schmitz, "Oscillations in Catalytic Reactions," Cat. Rev. Sci. Eng., 15, 107 (1977). Slinko, M. G., and M. M. Slinko, "Self-Oscillations of Heterogeneous Catalytic Reaction Rates," Cat. Rev. Sci. Eng., 17, 119 (1978).

Spencer, M. S., and T. V. Whittam, "Reactions of Methanol over Highly Siliceous Zeolite," Proc. Symp. on Zeolites, Szeged, Hungary, 307 (1978)

I. L. Tsitovskaya, O. V. Altshuler, and O. V. Krylov, "Certain Characteristics of Oxidation of Cyclohexane on NaY Zeolites," *Dokl. Akad. Nauk*, SSSR, 212 (6), 1400 (1973).

Uppal, A., W. H. Ray, and A. B. Poore, "On the Dynamic Behavior of Continuous Stirred Tank Reactors," Chem. Eng. Sci., 29, 967 (1974).

Vannice, M. A., "The Catalytic Synthesis of Hydrocarbons from Carbon Monoxide and Hydrogen," Cat. Rev. Sci. Eng., 14 (2), 153 (1976).

Varghese, P., J. J. Carberry, and E. E. Wolf, "Spurious Limit Cycles and Related Phenomena during CO Oxidation on Supported Platinum," J. Cat., 55, 76 (1978).

Vayenas, C., B. Lee and J. Michaels, "Limit Cycles and Mechanism of the Ethylene Oxidation on Platinum," J. Cat., 66, 36 (1980).

Whittam, T. V., U.S. Patent 4,060,590 (1977).

Manuscript received April 20, 1981; revision received August 5, and accepted August 24, 1981

Bubble Formation in a Vertically Vibrated System—Tate's Law Region

C. T. BARKER and NOEL de NEVERS

Department of Chemical Engineering University of Utah Salt Lake City, UT 84112

Bubbles forming at small, single, circular orifices at constant, low flow rates obey "Tate's Law." When vertical oscillations are added to the system, low-viscosity fluids show modified behavior with oscillational plus gravitational acceleration influencing bubble release; high-viscosity fluids produce bubbles of the same size as predicted by Tate's law.

THEORY

Tate's law is derived by equating the surface tension force (assuming an angle of attachment of 90°) and the buoyant force and solving for the bubble volume.

$$V_t = \frac{2\pi R_0 \sigma}{\Delta \rho g} \tag{1}$$

Experiments show that Tate's law is a fair predictor (±20%) of bubble size for small orifices with vanishingly small gas flow and negligible chamber volume (Guyer and Peterhans, 1943; Datta et al., 1950; van Krevelen and Hoftijzer, 1950; Coppock and Meiklejohn, 1951; Benzing and Myers, 1955; Hughes et al., 1955; Soo, 1967; Blanchard and Syzdek, 1977; Park et al., 1977).

When the entire bubble-forming apparatus is placed on a vibratory table and placed in sinusoidal vertical motion, it experiences a sinusoidal acceleration, which can be expected to add to that of gravity. Thus, one would expect Tate's law to take the form

$$V_b = \frac{2\pi R_0 \sigma}{\Delta \rho (g + a\omega^2 \sin \omega t)}$$
 (2)

0001-1541-82-5663-0851-\$2.00 © The American Institute of Chemical Engineers, 1982.

Tate's law is static; it says that the bubble releases when the upward buoyant force exceeds the downward surface tension force. Equation 2 shows that the predicted volume for release is sinusoidally varying. This makes more sense if one considers that the downward surface force is constant while the upward, buoyant force is the product of a linearly increasing component (due to bubble growth) and a sinusoidally varying one. Thus, Eq. 2 indicates the bubble volume for which the peak of the sinusoidally varying upward force exceeds the constant downward surface tension force. If there were no resistance to fluid motion due to viscosity, the bubble would be expected to release at this instant; and its size would be given by Eq. 2 If there were considerable viscous resistance, then the bubble would be expected to release only when the upward force, averaged over the entire cycle, exceeded the downward force.

The logical way to express the effect of viscosity is in terms of some form of Reynolds number. The appropriate one for this system appears to be

$$Re = \frac{D_b a \omega \rho}{\mu} \tag{3}$$

This choice of Reynolds number for vibrating bubble systems goes back at least as far as Jameson (1966).

Previous authors have shown that Eq. 1 is only a fair predictor of the observed bubble volumes; they have also discussed the causes of this variation. To eliminate this variability and focus on the effect of vibrations, all results in this note are presented in the form of the ratio of the observed bubble volume in the vibrated case (V_b) to the observed bubble volume for the same apparatus and conditions without vibrations (V_t) . From the preceding theory for bubbles growing slowly, one would expect this ratio to be unity for small Reynolds numbers and to be $[C_o = (1/(1+a\omega^2/g))]$ for large Reynolds numbers.

EXPERIMENTAL APPARATUS AND PROCEDURE

The entire system, comprised of an orifice and liquid chamber above it, was vibrated vertically in a sinusoidal manner at preset amplitudes and frequencies by a vibratory table (All American Machine Company, Model 100 VA).

A thin, porous polycarbonate membrane (Nucleopore N003) with average pore diameter of 0.03 µm, located on the bottom surface of the orifice plate, produced a constant flow rate independent of pressure oscillations in the liquid and the bubble.

The liquids used were water, cyclohexane, baby oil, industrial oil (Amoco Industrial Oil No. 51) and 60, 85, and 96 percent glycerol-in-water solutions. The gas was air except for the flammable liquids, with which nitrogen was used. The ambient laboratory conditions were nearly constant at 8.613 X 10⁴ Pa and 24°C. The range of values of the following parameters defines the region tested.

Vibrational Frequency, Hz	7-49
Vibrational Amplitude, mm	0.368 - 1.59
Gas Flow Rate, mm ³ /s	1.13-11.5
Liquid Head, mm	50.8-152
Orifice Diameter, mm	0.553-2.004
Reynolds Number	0.17 - 549
$a\omega^2/g$	0–6

EXPERIMENTAL RESULTS

Figure 1 summarizes the experimental results. As expected, the ratio of vibrated bubble volume to unvibrated bubble volume is unity for small Reynolds numbers and approaches C_o as an asymptote for large ones. Figure 1 shows only the experimental values for one particular C_0 (0.5). The agreement between experiment and the curves shown is comparable for the other values of Co. For Reynolds numbers less than 1, the experimental values of the bubble volume ratio differed from unity by an average of 3%. For Reynolds numbers greater than 10, the experimental values for all C_o 's differed from C_o by an average of 5%. For the transition region with Reynolds numbers between 1 and 10, the data were satisfactorily curve fit by

Volume Ratio =
$$\frac{V_b}{V_t} = \frac{\alpha Re^{-\gamma} + C_o}{\alpha Re^{-\gamma} + 1}$$
 (4)

where
$$\alpha = 10^{(-3.651 \log_{10} C_o + 0.242)}$$
 (5)

and
$$\gamma = -2.381 C_o + 4.497$$
 (6)

Equation 4 fits the experimental data for all values of C_o with a maximum error of 40% and an average error of 6.6%. Equation 4 has no theoretical basis and should be viewed simply as a datafitting equation. The solid curves on Figure 1 are computed from

All of the data in this project were taken at low flow rates such that the number of vibratory cycles per bubble was greater than 9.0.

Some of the tests with highly vicous fluids and $a\omega^2/g > 9.0$ resulted in dimensionless bubble volumes greater than 20.0. This is due to the compressibility of the gas in the bubble (Buchanan et al., 1962; Jameson and Davidson, 1966; Jameson, 1966; Foster et

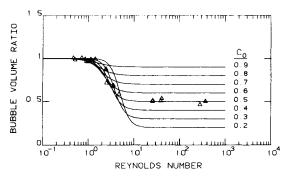


Figure 1. Summary of experimental data: The data points are for $C_o = 0.5$; the curves are from Eq. 4.

al., 1968; Rubin, 1968). Values of $a\omega^2/g$ greater than 9.0 are probably the range of values farthest removed from common industrial situations.

Complete experimental details, tables or data, and much more discussion are presented by Barker (1981).

ACKNOWLEDGMENT

This note is based upon work supported by the National Science Foundation under Grant ENG-7909104.

NOTATION

= amplitude, mm а

= bubble volume ratio calculated by $1/(1 + a\omega^2/g)$ C_o = bubble diameter assuming spherical shape, mm

= gravitational acceleration, m/s²

Re = Reynolds number, Eq. 3

 R_o = orifice radius, mm

= time, s ŧ

= bubble volume, vibrated conditions, mm³ V_b

= bubble volume, static conditions, mm³

Greek Symbols

α = curve fitting parameter

= curve fitting parameter γ

= viscosity, Pa·s μ = density, kg/m^3

 $\Delta \rho$ = gas-liquid density difference, kg/m³

= surface tension, N/m

= angular velocity, rad/s

LITERATURE CITED

Barker, C. T., "The Formation of Bubbles at Vibrated Orifices," Ph.D. Thesis, University of Utah (1981).

Benzing, R. J., and J. E. Myers, "Low Frequency Bubble Formation at Horizontal Circular Orifices," *Ind. Eng. Chem.*, 47, 2087 (1955).

Blanchard, D. C., and L. D. Syzdek, "Production of Air Bubbles of a

Specified Size," Chem. Eng. Sci., 32, 1109 (1977).

Buchanan, R. H., G. J. Jameson, and D. Oedjoe, "Cyclic Migration of Bubbles in Vertically Vibrating Liquid Columns," Ind. Eng. Chem., 1, 82 (May, 1962)

Coppock, P. D., and G. T. Meiklejohn, "The Behavior of Gas Bubbles in Relation to Mass Transfer," Trans. Inst. Chem. Engrs., 29, 75 (1951).

Datta, R. L., D. H. Napier, and D. M. Newitt, "The Properties and Behavior of Gas Bubbles Formed at a Circular Orifice," Trans. Inst. Chem. Engrs., 28, 14 (1950).

Foster, J. M., J. A. Botts, A. R. Barbin, and R. I. Vachon, "Bubble Trajectories and Equilibrium Levels in Vibrated Liquid Columns," Trans. ASME J. Basic Eng., 90, 125 (1968)

Guyer, A., and E. Peterhans, "On Bubble Size. I. Evolution at Single Capillaries," Helv. Chim. Acta, 26, 1099 (1943).

Hughes, R. R., A. E. Handlos, H. D. Evans, and R. L. Maycock, "The Formation of Bubbles at Simple Orifices," Chem. Eng. Prog., 51, 557

Jameson, G. J., and J. F. Davidson, "The Motion of a Bubble in a Vertically Oscillating Liquid: Theory for an Inviscid Liquid and Experimental Results," Chem. Eng. Sci., 21, 29 (1966).

Jameson, G. J., "The Motion of a Bubble in a Vertically Oscillating Viscous Liquid," Chem. Eng. Sci., 21, 35 (1966).

Park, Y., A. L. Tyler, and N. de Nevers, "The Chamber Orifice Interaction in the Formation of Bubbles," Chem. Eng. Sci., 32, 907 (1977).

Rubin, R., "Behavior of Gas Bubbles in Vertically Vibrating Liquid Col-umns," Can. J. Chem. Eng., 46, 145 (1968).

Soo, S. L., Fluid Dynamics of Multiphase Systems, Blaisdell Publishing Co., 95 (1967)

van krevelen, D. W., and P. J. Hoftijzer, "Calculation of Interfacial Area in Bubble Contactors," Chem. Eng. Prog., 46, 29 (1950).

Manuscript received June 22, 1981, and accepted August 7, 1981.