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MEASUREMENT OF DIFFUSION COEFFICIENTS FROM SOLUTION RATES OF BUBBLES

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The rate of solution of a stationary bubble is limited by the diffusion of dissolved gas molecules away from the bubble surface. Diffusion coefficients computed from measured rates of solution give mean values higher than accepted literature values, with standard errors as high as 10% for a single observation. Better accuracy is achieved with sparingly soluble gases, small bubbles, and highly viscous liquids. Accuracy correlates with the Grashof number, indicating that free convection is the major source of error. Accuracy should, therefore, be greatly increased in a gravity-free environment. The fact that the bubble will need no support is an additional important advantage of Spacelab for this measurement.

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by

Irvin M. Krieger[a]

A stationary gas bubble immersed in a liquid will dissolve at a rate limited by the outward transport of solute molecules from the bubble surface. In the absence of convection, the rate of transport is controlled by diffusion, and can readily be calculated from Fick's law. A closed-form solution for the rate of disappearance of the bubble is available for the isothermal case with concentration-independent diffusion coefficient and spherical symmetry. Diffusion coefficients can be calculated by fitting the theoretical equation to the experimental data. If the experiment can be conducted under conditions where the assumptions are valid, one should obtain better reproductibility and accuracy in the measured diffusion coefficients than is achievable by the best methods currently available. Given accurate diffusion coefficients for probe molecules of various sizes in pure liquids and solutions, valuable inferences can be drawn about the structures of the liquids and the mechanisms of mass transport.

Due to buoyancy effects, a stationary free bubble can not be achieved in a gravitational field. Various artifices have been invoked to hold the bubble....attachment to flat plates or fibers, even confinement by a centrifugal field. Use of a flat plate destroys the spherical symmetry, so that either approximate integrations or empirical calibrations are needed. Attachment of the bubble to a single fiber, of diameter far less than that of the bubble, was achieved in the author's laboratory (J. Phys. Chem. 71, 1173 (1967); 78, 2516 (1974)). Statistical analysis of a large body of data showed that

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average diffusion coefficients obtained by this method were consistently higher than those obtained by such accepted methods as the liquid jet technique. The relative standard deviations of the measurements in pure liquids were typically from 5 to 9%; only for measurements in viscous polymer solutions were the relative standard deviations appreciably below 5%. The standard deviations correlated with the dimensionless Grashof Number

$$Gr = \frac{\rho_\ell g d^3 \Delta C \frac{\partial \rho}{\partial c}}{\eta^2},$$

where ρ_ℓ is the density of the liquid, g the gravitational acceleration, d the bubble diameter, c the solute concentration, ΔC the difference between concentrations at the bubble surface and in the bulk, and η the liquid's viscosity.

What is proposed, then, is a Spacelab experiment in which a small bubble is injected into a liquid, and its rate of solution followed by time-lapse photography. The gravity-free environment would confer two very important benefits: (1) absence of appreciable buoyancy would allow the free bubble to remain stationary; and (2) the very low g-value would effectively eliminate gravity-driven convection. The accurate diffusion coefficients that should be obtainable from these data should permit determination of the effects of such variables as solute concentration, temperature and size of probe molecules.

Ground-based experiments in preparation for those in Spacelab should concentrate on reducing the Grashof number. The most sensitive variables are bubble diameter and liquid viscosity. Use of small bubbles and viscous polymer solutions should allow reductions in the Grashof number equivalent to reduction of g by two or three orders of magnitude. With liquids of sufficiently high viscosity, it should be possible to slow the bubble's rise to such an extent as to achieve an effectively stationary bubble without a supporting fiber.