

An Integral Method for Convective Diffusion-Bubble Dissolution

T. G. THEOFANOUS, H. S. ISBIN, and H. K. FAUSKE

Purdue University, Lafayette, Indiana

Diffusion controlled (heat or mass) spherical phase change is an important and challenging problem. The importance is derived from the many engineering operations involving bubble growth or collapse in one- or two-component systems. The challenge stems from the difficulties associated with solving the convective diffusion equation for a region which not only has a moving boundary but also deforms with time.

For bubble growth from a zero initial size in uniformly superheated or supersaturated systems, a well-known similarity solution exists, which solves the problem for the case that diffusion is the controlling mechanism. Further, the Plesset-Zwick temperature integral gives a first-order approximation under the assumption of a thin diffusion layer surrounding the growing bubble. Both solutions are in agreement, and they also agree with recent numerical results (1) indicating the plausibility of the thin layer assumption for the growth problem and the minor effect of the assumed zero initial radius in the similarity solution.

For bubble collapse, in uniformly subcooled or undersaturated systems, the similarity transformation does not apply, and one can easily show the thin diffusion layer assumption to be significantly in error. We wish to discuss the mathematical problem corresponding to this last situation. To be explicit, we consider the gas bubble dissolution by molecular diffusion.

In the past, solutions have been sought in three different ways:

1. Quasistationary approximations, with convective terms neglected.
2. Quasi steady state approximations, with the time derivative neglected.
3. Numerical, finite difference schemes on exact equation for the model.

These previous solutions are discussed by Duda and Vrentas (1), who provide a fourth approach. They devised a perturbation scheme based on the smallness of the physical parameters N_a and N_b . Further, in their numerical work, they immobilized the bubble wall and mapped the infinity of fluid surrounding the gas bubble onto a region of finite extent. Their perturbation results were in very good agreement with their numerical calculations for the range of parameters investigated; however, gross deviations from the previous detailed numerical work of Cable and Evans (2) and other approximate solutions were demonstrated.

Integral methods have been applied extensively and successfully in the main for ordinary conduction problems, and, in particular, with those problems involving temperature dependent transport properties. For nonplanar geometries, appropriate modifications of the usually assumed polynomial profiles contributed to an increased accuracy for the tested cases of heat conduction from a cylinder or a sphere conducting in an infinite stationary medium (3). Information on the applicability of the integral method to the class of problems involving the convective diffusion equation in deforming medium with time dependent boundaries is lacking.

It is the purpose of this note to apply integral techniques to the above described problem of gas bubble dissolution with the following two aims in mind:

1. To illustrate and justify the method for a simple but representative case of the class (just described) for which there is no information in the literature. This would serve as a first step in extending this useful and simple method to other more complicated problems of the same class.
2. To develop simple closed form expressions for the radius-time behavior.

FORMULATION OF THE METHOD

The equations in dimensionless form are given in (1):

$$\frac{\partial \rho^*}{\partial t^*} + \frac{R_v^{*2}}{r^{*2}} R_v^* \frac{\partial \rho^*}{\partial r^*} = \frac{1}{r^{*2}} \frac{\partial}{\partial r^*} \left[r^{*2} \frac{\partial \rho^*}{\partial r^*} \right] \quad (1)$$

$$\text{in } t^* > 0 \quad \text{and} \quad R_v^* < r^* < \infty$$

with

$$\rho^*(r^*, 0) = 0 \quad r^* > 1 \quad (2)$$

$$\rho^*(\infty, t^*) = 0 \quad t^* \geq 0 \quad (3)$$

$$\rho^*(R_v^*, t^*) = 1 \quad t^* > 0 \quad (4)$$

$$R_v^* = N_a \frac{\partial \rho^*}{\partial r^*} \bigg|_{r^*=R_v^*} \quad (5)$$

$$R_v^*(0) = 1 \quad (6)$$

Without loss of generality, we take $N_b = 0$ (that is, gas-liquid systems). The stars and subscript of N_a are suppressed below.

We anticipate the existence of a concentration boundary layer of radius $R_L = R_L(t)$, such that

$$\rho(R_L) = 0 \quad \text{and} \quad \frac{\partial \rho}{\partial r} \bigg|_{R_L} = 0 \quad (7)$$

H. S. Isbin is at the University of Minnesota, Minneapolis, Minnesota.
H. K. Fauske is with Argonne National Laboratory, Reactor Engineering, Argonne, Illinois.

TABLE 1

$G(R, N)$						$F(R, N)$					
$R \backslash N$	0.005	0.5	0.1	0.15	0.2	$R \backslash N$	0.005	0.05	0.1	0.15	0.2
0.98	2.223	0.077	0.0233	0.0111	0.0065	0.8	0.7420	0.5309	0.4887	0.4684	0.4561
0.90	13.998	0.831	0.3109	0.1681	0.1067	0.6	0.7373	0.5171	0.4700	0.4464	0.4314
0.82	24.717	1.675	0.6715	0.3808	0.2505	0.4	0.7338	0.5067	0.4558	0.4294	0.4121
0.74	33.800	2.444	1.0172	0.5925	0.3981	0.2	0.7317	0.5003	0.4468	0.4185	0.3997
						0.05	0.7311	0.4982	0.4439	0.4150	0.3956

Integrating (1) over the liquid volume and utilizing (7) and (5), we obtain the integral mass balance

$$\frac{d}{dt} \int_{R_V}^{R_L} \rho r^2 dr = - R_V^2 \frac{\partial \rho}{\partial r} \bigg|_{R_V} = - R_V^2 \frac{\dot{R}_V}{N} \quad (8)$$

which, in turn, when integrated with $R_L(0) = R_V(0) = 1$, leads to

$$\int_{R_V}^{R_L} \rho r^2 dr = - \frac{1}{3N} (R_V^3 - 1) \quad (9)$$

Now we satisfy conditions (4) and (7) by choosing

$$\rho = \frac{R_V}{r} \left(\frac{r - R_L}{R_V - R_L} \right)^2 \quad (10)$$

Combination of (10) with (9) and (5) gives

$$R_V(R_L + 3R_V)(R_L - R_V) = - \frac{4}{N} (R_V^3 - 1) \quad (11)$$

and

$$\dot{R}_V = N \frac{R_V + R_L}{R_V(R_V - R_L)} \quad (12)$$

Elimination of R_L between (11) and (12) yields

$$R_V^2 \dot{R}_V^2 + 2NA R_V \dot{R}_V + AN^2 = 0 \quad (13)$$

where

$$A = \frac{(1-N)R_V^3 - 1}{R_V^3 - 1} \quad (14)$$

From this the time derivative of R_V can be obtained:

$$R_V = - \frac{NA}{R_V} \left[1 + \sqrt{1 - \frac{1}{A}} \right] \quad (15)$$

SOLUTION

Equation (15) has to be solved with the initial condition (6) in the range $0 \leq R_V \leq 1$. The numerical solution

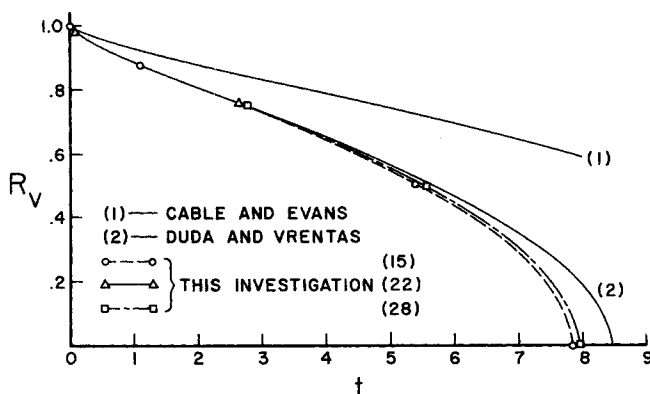


Fig. 1. Bubble dissolution with $N = 0.05$.

of this problem is quite simple, but an analytical solution, however, requires some elaboration.

Initial Stages of Collapse

Multiplying Equation (15) by R_V^2 and rearranging, we obtain upon integration

$$\int_1^{R_V} \frac{dR_V^3}{A \left[1 + \sqrt{1 - \frac{1}{A}} \right]} = - 3N \int_0^t R_V dt \quad (16)$$

With $R_V^3 = x$, the left-hand side becomes

$$\int_1^{R_V^3} \frac{(1-x)dx}{- [(1-N)x - 1] + [Nx(1 - (1-N)x)]^{1/2}} = - 3N \int_0^t R_V dt \quad (17)$$

Now, we let $tx = \sqrt{Nx - N(1-N)x^2}$, and (17) becomes

$$\int_N \frac{-2N(t-N)dt}{[N(1-N) + t^2]^2} = - 3N \int_0^t R_V dt \quad (18)$$

Elementary integration of (18) and the definition of a mean radius

$$\bar{R}_V = \frac{1}{t} \int_0^t R_V dt \quad (19)$$

produce the final result:

$$G(R_V, N) = \frac{1 - R_V^3}{3N} - \frac{1}{3(1-N)} \left[R_V^3 \sqrt{\frac{1}{NR_V^3} - \frac{1-N}{N}} - 1 \right] - \frac{1}{3N(1-N)} \sqrt{\frac{N}{1-N}} \left[\arctg \sqrt{\frac{1}{R_V^3(1-N)}} - 1 - \arctg \sqrt{\frac{N}{1-N}} \right] \quad (20)$$

and

$$\bar{R}_V t = G(R_V, N) \quad (21)$$

But R_V changes almost linearly with time, and the value $(R_V + 1)/2$ is a very good approximation for \bar{R}_V

$$\therefore t = \frac{2G(R_V, N)}{1 + R_V} \quad (22)$$

This solution is expected to be valid for small intervals of R_V , say $1 \leq R_V \leq 0.7$ in which the assumed expression for the mean radius remains valid. It is used only to pro-

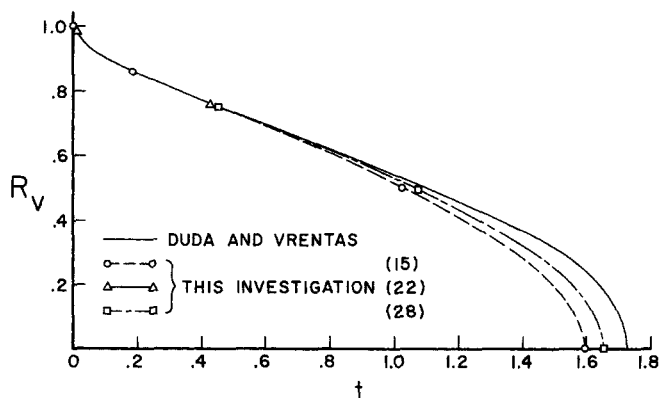


Fig. 2. Bubble dissolution with $N = 0.2$.

vide the initial part of the solution which can be continued.

Continuation of Solution

From (14), one can see that to a first order of approximation ($N < 1$), and for $R_V < 1$

$$A \approx 1 \quad (23)$$

and Equation (15) then gives

$$\dot{R}_V = -\frac{N}{R_V} \quad (24)$$

From the development of (13), and the use of (24) as noted below

$$\begin{aligned} (R_V^3 - 1) [R_V \dot{R}_V + N]^2 &= N^2 R_V^3 (N + 2R_V \dot{R}_V) \\ &= N^2 R_V^3 (N - 2N) = -N^3 R_V^3 \end{aligned} \quad (25)$$

or

$$\begin{aligned} \frac{R_V dR_V}{[1 + \sqrt{N} \sqrt{R_V^3 (1 - R_V^3)^{-1/2}}]} \\ \approx \frac{R_V dR_V}{1 + \sqrt{N} \sqrt{R_V^3}} = -N dt \end{aligned} \quad (26)$$

The added simplification is used:

$$(1 - R_V^3)^{-1/2} \approx 1 + \frac{1}{2} R_V^3 + 0 (R_V^6) \approx 1 \quad (27)$$

With an error less than 5% for $R_V < 0.8$ and $N < 0.2$, and by decreasing rapidly with R_V , integration of (26) between R_0 and R_V for t_0 and t yields

$$t = t_0 + \frac{2}{N\sqrt{N}} [F(R_0, N) - F(R_V, N)] \quad (28)$$

where

$$\begin{aligned} F(R, N) = \sqrt{R} - \frac{N^{-1/6}}{3} \\ \left\{ \frac{1}{2} \ln \frac{(\sqrt{R} + N^{-1/6})^2}{(R - N^{-1/6}\sqrt{R} + N^{-1/3})} \right. \\ \left. + \sqrt{3} \arctg \left[\frac{2\sqrt{R} - N^{-1/6}}{N^{-1/6}\sqrt{3}} \right] \right\} \end{aligned} \quad (29)$$

RESULTS AND DISCUSSION

Equations (22), (28), and (29) provide a closed form solution for the bubble dissolution problem over the entire range of R_V ($0 \leq R_V \leq 1$). Figures 1 and 2 illustrate comparisons of calculations based on these equations with the results of Duda and Vrentas for the two extreme cases of N reported by them. In all cases, (22) was valid at

least down to $R_V = 0.7$, where (28) and (29) become very accurate. Results from the numerical integration of our Equation (15) are also displayed for comparison.

The deviation of our analytical expressions from the solution of (15) is minor for $N = 0.2$ and decreases rapidly with N . In general, the agreement with the numerical results is very good, justifying the analytical approach utilized.

It is noteworthy that our solutions deviate somewhat from the exact solution (numerical results) at the very end of the collapse, retaining though a similar trend. The authors of (1) do not report numerical results for the very last part of the collapse, where also their perturbation results began to deviate (in the same way as our solutions) from their finite difference results. Further, the perturbation solution of (1) as well as our analytical expressions may be expected to break down as N increases. It would be interesting to see which one breaks down first and to what extent the numerical results from our simple ordinary differential Equation (15) agree with the finite difference results as N increases. Unfortunately, these comparisons cannot be made now, since there are no reliable numerical results for $N > 0.2$. For the convenience of others, the functions $G(R, N)$ and $F(R, N)$ are given in Table 1 for values of n and R .

ACKNOWLEDGMENT

The Purdue University Computer Center furnished the time for the numerical computations.

NOTATION

D	= binary diffusion coefficient
R_V	= radius of the bubble, $\dot{R}_V = \frac{dR_V}{dt}$
R_0	= initial bubble radius
R_L	= radius of the concentration boundary layer
r	= radial coordinate
t	= time
\hat{V}	= partial specific volume of the gas
ρ	= mass density of gas in solution
ρ_0	= initial concentration
ρ_E	= equilibrium concentration at the liquid side of the interface
$\hat{\rho}$	= density of gas in the bubble

Dimensionless Quantities

Given in Equations (1) through (6) and henceforth not designated with the *

$$\begin{aligned} r^* &= \frac{r}{R_0} \\ R_V^* &= \frac{R_V}{R_0} \text{ and } R_L^* = \frac{R_L}{R_0} \\ t^* &= \frac{Dt}{R_0^2} \\ \rho^* &= \frac{\rho - \rho_0}{\rho_E - \rho_0} \\ N_a &= N = \frac{\rho_E - \rho_0}{\hat{\rho}(1 - \hat{V}_{\rho_E})} \text{ and } N_b = \frac{\rho_E - \rho_0}{1/\hat{V} - \rho_E} \\ &\quad \text{(used as zero in this paper)} \end{aligned}$$

LITERATURE CITED

1. Duda, J. L., and J. S. Vrentas, *AIChE J.*, **15**, 351 (1969).
2. Cable, M., and D. J. Evans, *J. Appl. Phys.*, **38**, 2899 (1967).
3. Lardner, T. J., and F. V. Pohle, *Trans. Am. Soc. Mech. Engrs.*, **E28**, 310 (1961).