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AEROSOL PROPAGATION EFFECTS

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MID-CONTRACT REPORT

JUNE 1, 1974

by

G. E. CALEDONIA AND K. L. WRAY

Sponsored By

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ABSTRACT

A model has been developed for the prediction of the time dependent properties of the gas surrounding a water droplet under irradiation by a high intensity laser beam. The basic phenomena included in the model are absorption, heating and vaporization in the droplet and convection, thermal conduction and diffusion in the gas. The model assumes volume absorption by the droplet, a constant pressure gas, and that the initial vapor velocity is much less than the local sound speed. The conditions under which these assumptions are appropriate have been identified. A number of preliminary calculations are presented.

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AEROSOL PROPAGATION EFFECTS

I. INTRODUCTION

The Aerosol Propagation Effects program is concerned with the prediction of laser beam degradation resulting from the interaction between a high power laser beam and atmospheric aerosols. The particular emphasis of this program is on the absorption, scattering and thermal blooming of a DF laser propagating through an atmosphere contaminated by hygroscopic aerosols. The overall program includes a review of the properties of atmospheric aerosols, a study of the atmospheric heating resulting from aerosol absorption of laser energy and first order predictions of the laser beam thermal blooming resulting from this atmospheric heating.

The particular problem addressed in this mid-program progress report is the prediction of the space/time variation in the temperature and water vapor distributions about a pure water droplet irradiated by a laser beam. This problem is relevant to beam propagation through hygroscopic aerosols on humid days. Effects resulting from the varying properties of real aerosol solutes and original Aitken nuclei will be discussed in the final report.

The description of the general problem and derivation of the governing partial differential equations for the droplet and surrounding gas will be given in Section II. Specific "steady state" and time dependent computer solutions of the problem will be presented in Section III. The conclusions and future directions of the study are presented in Section IV.

II. MODELING

In the problem under consideration the aerosol droplet absorbs energy and is heated. As the droplet temperature rises above ambient the droplet conducts heat energy to the surrounding air and undergoes surface vaporization. The vapor leaves the droplet with a directed velocity and a pressure wave is induced in the surrounding gas in response to the mass and heat addition. (If the rate of energy absorption were sufficiently large the droplet would "explode", creating a shock wave; however, the present analysis will be limited to laser intensities and droplet sizes such that the directed velocity of the vapor leaving the droplet surface is much less than the local sound speed.) An additional feature which must be included in the analysis is the fact that the droplet radius decreases with time due to mass loss resulting from vaporization.

A. Gas Equations

A number of approximations have been made to simplify the solution of this problem. The most prominent of these is that the gas is taken to be at a constant pressure. As pointed out above, pressure gradients are induced in the gas because of the mass and heat addition from the droplet. However, if the pressure equilibrates on a time scale fast relative to the times for conduction and diffusion then the heat and mass transfer may be assumed to be occurring in a constant pressure gas. The characteristic time for water vapor to diffuse a distance L in air is approximately

$$\tau_D \approx L^2/D \quad (1)$$

where D is the diffusion coefficient. The characteristic time for heat conduction in air is

$$\tau_C \approx L^2 \rho C_{PA} / k_A \quad (2)$$

where ρ is the density, C_{P_A} the specific heat and k_A the thermal conductivity of air. Lastly, the characteristic time for pressure equilibrium is

$$\tau_P \approx L/c \quad (3)$$

where c is the sound speed in air. Evaluating these expressions with $D = 0.24 \text{ cm}^2/\text{sec}$, $\rho_A = 1.2 \times 10^{-3} \text{ gms/cc}$, $C_{P_A} = 0.24 \text{ cal./gm-}^\circ\text{K}$, $k_A = 6.2 \times 10^{-5} \text{ cal./cm-sec-}^\circ\text{K}$ and $c \approx 3 \times 10^4 \text{ cm/sec}$ results in

$$\tau_D \sim \tau_C \gg \tau_P \quad (4)$$

for $L \gtrsim 1 \text{ }\mu\text{m}$. The characteristic droplet sizes of interest in this work are a few tenths μm to $50 \text{ }\mu\text{m}$, and for the typical atmospheric aerosol distributions of interest the average spacing between droplets, which would be the heating range of interest, is approximately one mm. Thus, it would appear that the approximation of a constant pressure gas is reasonable.

Another simplifying approximation used is that the gas properties are taken to be those of clean air. Although this approximation would suggest that the defining equations would not be valid for high water vapor concentrations this is not the case, as will be demonstrated later in this section. Also, the coefficient of thermal conductivity, k_A and the product of the coefficient of diffusion and number density, DN , were taken to be constant. These quantities vary by $\lesssim 15\%$ over the temperature range of interest, $0 - 100^\circ\text{C}$.

The last simplifications concern the fluid mechanics. The processes considered in the analysis are thermal conduction, diffusion and convection. The phenomena of thermal diffusion (Soret effect) and thermo-diffusion (Dufour effect) have not been included. Generally, these processes are insignificant in flow fields of the type under consideration; nonetheless, the validity of this assertion will be determined from the computer results. Furthermore, consideration must be given to gravitational effects. As the gas surrounding

the droplet is heated it will rise. The characteristic time for the gas to rise a distance L is:

$$\tau_g \approx \left(2L/g \left[\Delta\rho/\rho \right] \right)^{1/2} \quad (5)$$

where g is the acceleration due to gravity, 980 cm/sec^2 , and $\Delta\rho/\rho$ is the fractional difference in density between the heated gas and the surrounding medium. A strong lower bound on τ_g corresponds to $\Delta\rho/\rho \sim 0.3$. Even in this limit it can be shown that

$$\tau_D \sim \tau_C \ll \tau_g$$

for L less than one mm. Of course this effect might have to be included for long time ($t \gtrsim 0.1 \text{ sec}$) irradiation. However, in this case it would perhaps be more appropriate to treat the aerosols as an aggregate rather than individually.

Within the framework of the above-mentioned approximations, the general relationships describing the system are

a) Conservation of mass

$$\partial\rho/\partial t = -\vec{\nabla} \cdot (\rho \vec{V}) \quad (6)$$

where $\vec{V}$ is the gas velocity

b) Euler's equation

$$\partial\vec{V}/\partial t = -\vec{V} \cdot \vec{\nabla} \vec{V} \quad (7)$$

c) Conservation of energy

$$\partial(1/2 \rho V^2 + \rho C_V T)/\partial t = -\vec{\nabla} \cdot \left[\rho \vec{V} (1/2 V^2 + C_P T) - k_A \vec{\nabla} T \right] \quad (8)$$

where C_V is the specific heat at constant volume

and

d) The diffusion equation

$$\partial \rho_v / \partial t = \vec{\nabla} \cdot (D \vec{\nabla} \rho_v - \rho_v \vec{V}) \quad (9)$$

where ρ_v is the water vapor density and D is the H_2O -air diffusion coefficient.

If the droplet is heated uniformly (see Section B), the system is spherically symmetric and it can be shown that the ordered kinetic energy terms drop out of the equations under the assumption of constant pressure. The relevant gas equations for $r > R$, where R is the droplet radius, are

$$\partial T / \partial t = -V \partial T / \partial r + \frac{k k_A A T}{r^2 C_P P M_A} \frac{\partial}{\partial r} (r^2 \partial T / \partial r), \quad (10)$$

$$\partial X_v / \partial t = -V \partial X_v / \partial r + \frac{k T (DN)}{P r^2} \frac{\partial}{\partial r} (r^2 \partial X_v / \partial r), \quad (11)$$

$$\partial V / \partial r = -2 V / r + \frac{k k_A A}{r^2 C_P P M_A} \frac{\partial}{\partial r} (r^2 \partial T / \partial r), \quad (12)$$

where k is Boltzmann's constant, A is Avogadro's number, P is the total pressure taken as one atm., M_A is the molecular weight of air, N is the total number density, and X_v is the mole fraction of water vapor.

These equations have been formulated in terms of number density rather than mass density, since in a constant pressure flow of two gases with different molecular weights the quantity NT , rather than ρT , remains constant. As

mentioned earlier, these equations were derived under the assumption $X_v \ll 1$. In reality, the quantity $C_{P_A} M_A$ should be replaced by the expression

$$C_{P_A} M_A (1 - X_v) + C_{P_v} M_v X_v$$

where the subscript v refers to water vapor, and the diffusion coefficient should be replaced by that for full binary diffusion. However, over the temperature and water vapor ranges of interest in this problem these latter two quantities are reasonably constant and well represented by $C_{P_A} M_A$ and D , respectively. Thus Eqs. (10)-(12) are approximately valid for water vapor concentrations approaching unity.

B. Droplet Equations

In the present analysis the temperature field across the droplet has been taken to be uniform. As long as the laser radiation is absorbed volumetrically by the droplet, this approximation is reasonable until the droplet temperature approaches the boiling point of water (see Section III). The absorption coefficient of a water droplet is a complex function of droplet radius, laser wavelength and the complex index of refraction (see for example Ref. 1). However, for $4 \mu\text{m}$ radiation liquid water has a bulk absorption coefficient,² α , of $\sim 100 \text{ cm}^{-1}$, and it can be shown that for these conditions water droplets of radius $\lesssim 50 \mu\text{m}$ undergo volume absorption. (The general relationship for the absorption coefficient of a water droplet may be found in Ref. 3 and references therein.)

The basic equation connecting the droplet and gas dynamics is conservation of energy in the droplet.

$$\begin{aligned}
4/3 \pi R^3 I \alpha &= 4/3 \pi R^3 \rho_D C_{P_D} dT_D/dt \\
&+ 4 \pi R^2 k_A (-\partial T/\partial r)|_R \\
&+ 4 \pi R^2 \Delta H_v \frac{M_v}{A} (NV)|_R
\end{aligned} \tag{13}$$

The LHS of Eq. (13) is the rate of absorption of energy, where I is the laser intensity, and the RHS represents the energy disposition into droplet heating, conduction and vaporization respectively. The quantity ΔH_v is the change in enthalpy required to proceed from liquid to gaseous state and the subscript D refers to droplet properties. Note that the temperature is continuous across the droplet surface; i.e. $T_D = T(R)$. The terms involving the ordered kinetic energy of the vapor are negligible for the present considerations and have been left out of Eq. (13).

The vapor concentration at the surface of the droplet has been related to the droplet temperature by the Clausius-Clapeyron relationship,

$$\left. \frac{d \ln X_v}{d T} \right|_P = \frac{\Delta H_v}{R_v T^2} \tag{14}$$

where R_v is the ideal gas constant per unit mass of water vapor. It has been demonstrated⁴ that this relationship properly describes the surface vapor pressure of a water droplet when the vapor velocity is less than the sound speed. In the present analysis Eq. (14) has been used in its integral form

$$X_v(R) = X_{v_\infty} \exp \left[\frac{\Delta H_v (T_D - T_\infty)}{R_v T_D T_\infty} \right] \tag{15}$$

where the subscript ∞ refers to ambient conditions. Eq. (15) is derived under the assumption that ΔH_v is a constant. Indeed ΔH_v varies by less than 10% between 0 - 100° C and has been taken to have the constant value of 585 cal/gm.

The last equation connecting the droplet and gas behavior is the conservation of mass of the vapor. If a spherical shell of thickness ΔR is constructed about the droplet, then it is required that the rate of mass loss of the droplet be balanced by the rate of increase of vapor within the shell plus the rate of vapor leaving the exterior of the shell. With some straightforward mathematical manipulation this leads to the relationship

$$(NV) \Big|_R = - (1 - X_v(R))^{-1} DN \partial X_v / \partial r \Big|_R \quad (16)$$

C. Effect of Varying Droplet Size

So far in the analysis the effect of decreasing droplet size has not been considered. This effect can be uncoupled because the rate of change of droplet size is small compared to the rate of change of the gas parameters (because of the large difference between liquid and gas densities). Of course, it has been implicitly assumed that the time varying, rather than initial, value of R will be used in the solution of the equations.

The governing equations have been recast in terms of the variable

$$Z = R(t)/r \quad (17)$$

In terms of this variable the equations may be written as

$$\begin{aligned} \partial T / \partial t = & R^{-1} (QZ^4 - Z dR/dt) \partial T / \partial Z \\ & + \frac{k_A}{C_P} \frac{k_A}{PM_A} TR^{-2} Z^4 \partial^2 T / \partial Z^2 \end{aligned} \quad (18)$$

$$\begin{aligned} \partial X_v / \partial t &= R^{-1} (QZ^4 - Z dR/dt) \partial X_v / \partial Z \\ &+ \frac{k_A DN}{P} T R^{-2} Z^4 \partial^2 X_v / \partial Z^2 \end{aligned} \quad (19)$$

$$\partial Q / \partial Z = - \frac{k_A k_A A}{C_P M_A} R^{-1} \partial^2 T / \partial Z^2 \quad (20)$$

where

$$dR/dt = - \frac{P M_v}{A \rho_D k T} Q (Z = 1) \quad (21)$$

and

$$Q = V/Z^2 \quad (22)$$

The transformed boundary conditions at $r = R$ ($Z = 1$) are

$$\begin{aligned} I \alpha &= \rho_D C_{P_D} \left. \partial T / \partial t \right|_{Z=1} + 3 R^{-2} k_A \left. \partial T / \partial Z \right|_{Z=1} \\ &+ 3 R^{-1} \frac{M_v \Delta H_v P}{A k T} Q (Z = 1), \end{aligned} \quad (23)$$

$$Q (Z = 1) = R^{-1} \frac{k T DN}{P} \left[1 - X_v (Z = 1) \right]^{-1} \left. \partial X_v / \partial Z \right|_{Z=1} \quad (24)$$

and the Clausius-Clapeyron relationship, Eq. (15).

III. PRELIMINARY CALCULATIONS

No analytic solution has been found for the system of Eqs. (15), (18) - (24). A computer program has been developed for their numerical evaluation which employs a fully implicit Crank-Nicolson finite difference scheme. A complication in using the Z co-ordinate is that a constant grid size in Z space results in a limited number of grid points at large r , where the solution may be most interesting. This complication has been circumvented by modifying the finite difference scheme to allow for a change in grid spacing at small Z . In the present mode of operation the grid spacing for $0.0 \leq Z \leq 0.05$ is an order of magnitude finer than that for $0.05 \leq Z \leq 1.00$.

The computer code is now operational and a number of preliminary calculations have been performed. However, these are insufficient in a number to provide an overview of the problem. A "steady state" analysis of the equations has simultaneously been performed in order to predict the effects of varying fundamental parameters such as particle radius and laser intensity.

A. Steady State Solutions

In the limit where the velocity term in Eqs. (18) and (19) can be neglected, the equations effectively uncouple, and it can be readily demonstrated that the steady state solutions for $\Delta T (= T - T_\infty)$ and

$\Delta X_v (= X_v - X_{v\infty})$ scale inversely with r , i.e.,

$$\Delta T(r) = (R/r) \Delta T(R) \quad (25)$$

and

$$\Delta X_v(r) = (R/r) \Delta X_v(R) \quad (26)$$

Indeed, even the time dependent solutions, which may be found in standard texts (for example, Ref. 5) vary approximately as r^{-1} for $r \sim R$. Thus,

in the limit of low velocity the gradients appearing in the equation for droplet energy conservation, Eq. (23), may be evaluated by use of Eq. (25) and (26).

If this is done, use of Eqs. (15), (23) and (24) results in

$$\frac{d(\Delta T_D)/dt}{R^2 \rho_D C_{PD}} = \left\{ \frac{R^2 I \alpha}{3} - k_A \Delta T_D - \frac{\Delta H_v M_v}{A} DN(1 - X_v(R))^{-1} (X_v(R) - X_{v\infty}) \right\} \quad (27)$$

where

$$X_v(R) - X_{v\infty} = X_{v\infty} \left\{ \exp \left[\frac{\Delta H_v \Delta T_D}{R_v (\Delta T_D + T_\infty) T_\infty} \right] - 1 \right\}$$

Although this differential equation is separable, it affords no simple analytic solution. One feature of interest is the maximum value of ΔT_D which is reached when the time derivative is zero. Equation (27) in this limit reduces to a transcendental equation for $\Delta T_{D-\max}$ which may be evaluated as a function of the parameter $R^2 I \alpha$. The solution to this equation is shown in Fig. 1 over a range of 5 orders of magnitude in the parameter $R^2 I \alpha$. Shown for comparison are the predicted maximum droplet temperatures for the related cases of "heat conduction only" and "vaporization only". These calculations were performed for initial conditions of $T_\infty = 293^\circ K$, $X_{v\infty} = 2.1 \times 10^{-2}$.

Note that for the full range of $R^2 I \alpha$ shown the $\Delta T_{D-\max}$ predicted for the case of heat conduction alone is significantly higher than that predicted when both heat conduction and vaporization were included. On the

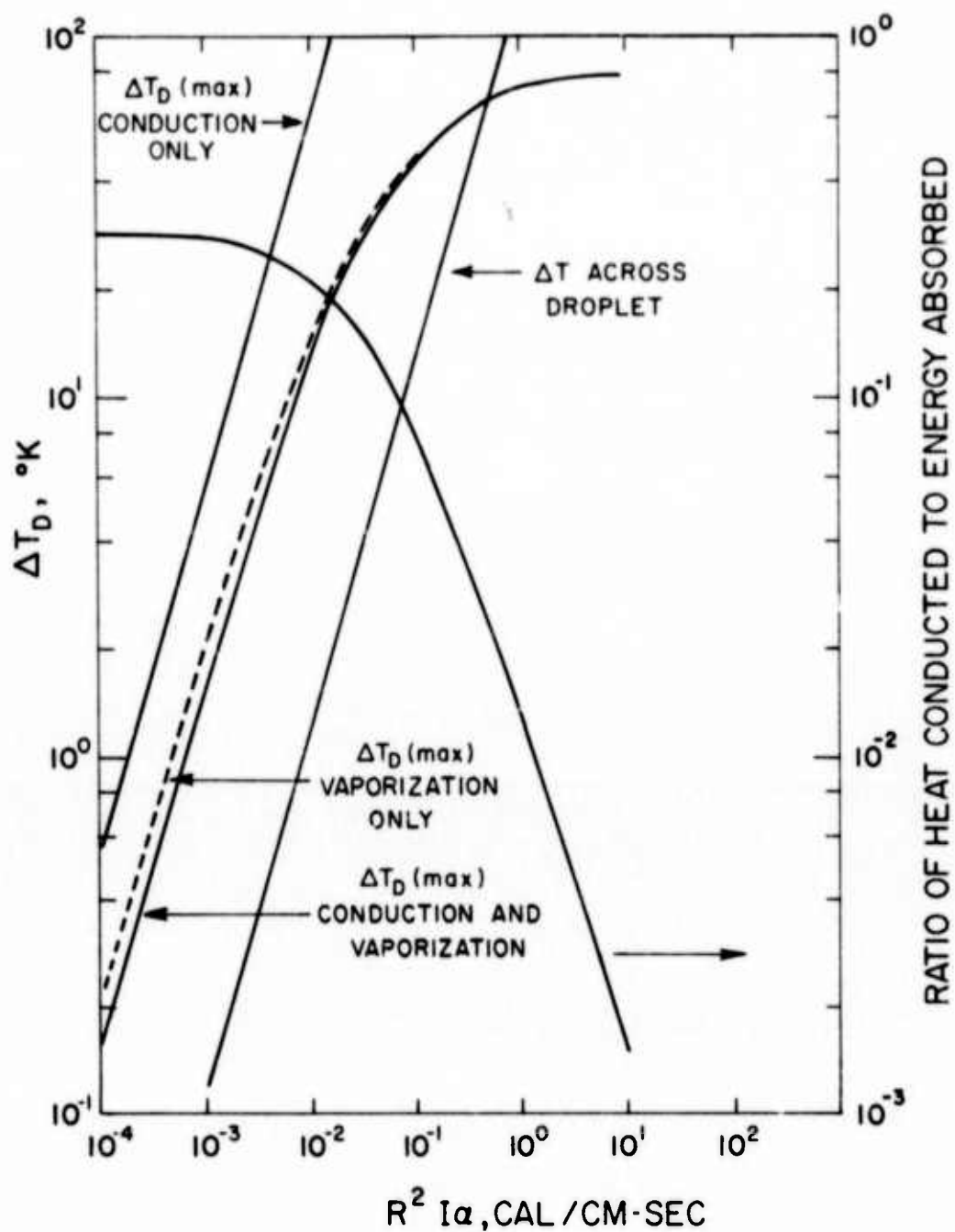


Fig. 1 Maximum Increase in Droplet Temperature vs. $R^2 I \alpha$. Also shown is the fraction of absorbed energy lost to heat conduction and the predicted variation in temperature across the droplet.

other hand, the predictions for the case of "vaporization only" are relatively close to the full solution. The reason for this is that the major portion of the absorbed energy goes into vaporization rather than heat conduction. This is shown explicitly in Fig. 1 where the fraction of the absorbed energy which is lost to heat conduction is plotted vs. $R^2 I \alpha$. For low $R^2 I \alpha$ this fraction asymptotes to ~ 0.3 , and falls off rapidly with increasing $R^2 I \alpha$. This rapid fall-off occurs as the droplet temperature approaches the boiling point. This is because the vaporization rate increases exponentially in this region, whereas the rate of heat conduction approaches a constant. The relative value of this fraction is determined by the ratio of the last two terms on the RHS of Eq. (27). For small $\Delta T_{D-\max}$ this ratio reduces to

$$\frac{\text{Power into heat conduction}}{\text{Power into vaporization}} = \frac{k_A R_v T_\infty^2 A (1 - X_{v\infty})}{X_{v\infty} \cdot DN (\Delta H_v)^2 M_v} \approx 0.43 \quad (28)$$

in agreement with the asymptotic value shown in Fig. 1 (i.e., $0.43/1.43 \approx 0.3$).

It should be pointed out that as the droplet temperature approaches the boiling point the convective terms left out of the steady state analysis become more important and convective heat transfer might dominate conductive heat transfer. The steady state equations including convection are more complicated, but can be analyzed. A discussion of convective effects will be included in the final report.

The fact that vaporization dominates heat conduction as an energy loss mechanism allows an approximate analytic formulation for several of the variables of interest in the problem. In particular, in steady state it can be readily shown from Eq. (27) that the rate of mass loss from the

droplet can be expressed as

$$dm/dt = -4/3 \pi R^3 I \alpha / \Delta H_v \quad (29)$$

or in terms of the droplet radius

$$dR/dt = -RI \alpha / (3 \Delta H_v \rho_D) \quad (30)$$

From Eq. (30) it can be seen that the characteristic e-folding time for droplet radius is

$$\tau_R = 3 \Delta H_v \rho_D / I \alpha \quad (31)$$

which is inversely proportional to $I \alpha$ and independent of initial particle size. Lastly, the vapor velocity at the droplet surface is given by

$$v(R) \approx M_A RI \alpha / (3 M_v \rho \Delta H_v) \quad (32)$$

Note this latter quantity scales as $RI \alpha$ rather than $R^2 I \alpha$, and thus is less sensitive to changes in radius than the droplet temperature.

A last point before leaving the discussion on steady state solutions concerns the importance of temperature gradients within the droplet. If one considers volume heating followed by heat conduction as the only phenomenon occurring within the droplet, the defining steady state energy equation becomes

$$4/3 \pi r^3 I \alpha = -4 \pi r^2 k_D \partial T / \partial r \quad (r < R) \quad (33)$$

where k_D is the coefficient of thermal conductivity of liquid water, taken as 1.4×10^{-3} cal/cm-sec-°K. Eq. (33) requires that

$$T_C - T_S = \frac{R^2 I \alpha}{6 k_D} \quad (34)$$

where the subscripts C and S refer to conditions at the droplet center and surface, respectively. This temperature variation is also shown in Fig. 1 and can be seen to be significantly lower than $\Delta T_{D-\max}$ until the droplet approaches the boiling point. Of course Eq. (34) is not realistic in that region since it would require the droplet to superheat. In reality, vaporization would occur within the droplet and original nuclei effects, etc. could be quite important. In any event, it is clear that the approximation of a constant temperature field across the droplet is not appropriate at large values of $R^2 I \alpha$.

B. Time Dependent Solutions

The full finite difference scheme has been exercised for a limited number of cases. As an example of the results, droplet temperature histories vs. time are shown in Fig. 2 for an $I \alpha$ of 2.5×10^5 cal/cc-sec and two initial particle sizes, $R = 3, 10 \mu\text{m}$. Note that the droplet temperatures rapidly rise to a peak value and then slowly decay. This decay results from the decrease in droplet radius with time. The radii of both the 3 and $10 \mu\text{m}$ particles decrease by $\sim 20\%$ in 2 msec and exhibit a characteristic e-folding time as given by Eq. (31). The calculated droplet temperatures from their peak values onward are in good agreement with the steady state predictions shown in Fig. 1.

Examples of the gas radial temperature distributions for several configurations are shown in Fig. 3. Curves C and D are for the same case, $R = 3 \mu\text{m}$, $I \alpha = 2.5 \times 10^5$ cal/cm³-sec, at two different times. Note that the profiles scale approximately inversely with r until large r , where they drop off more rapidly. At small r the temperature profile is

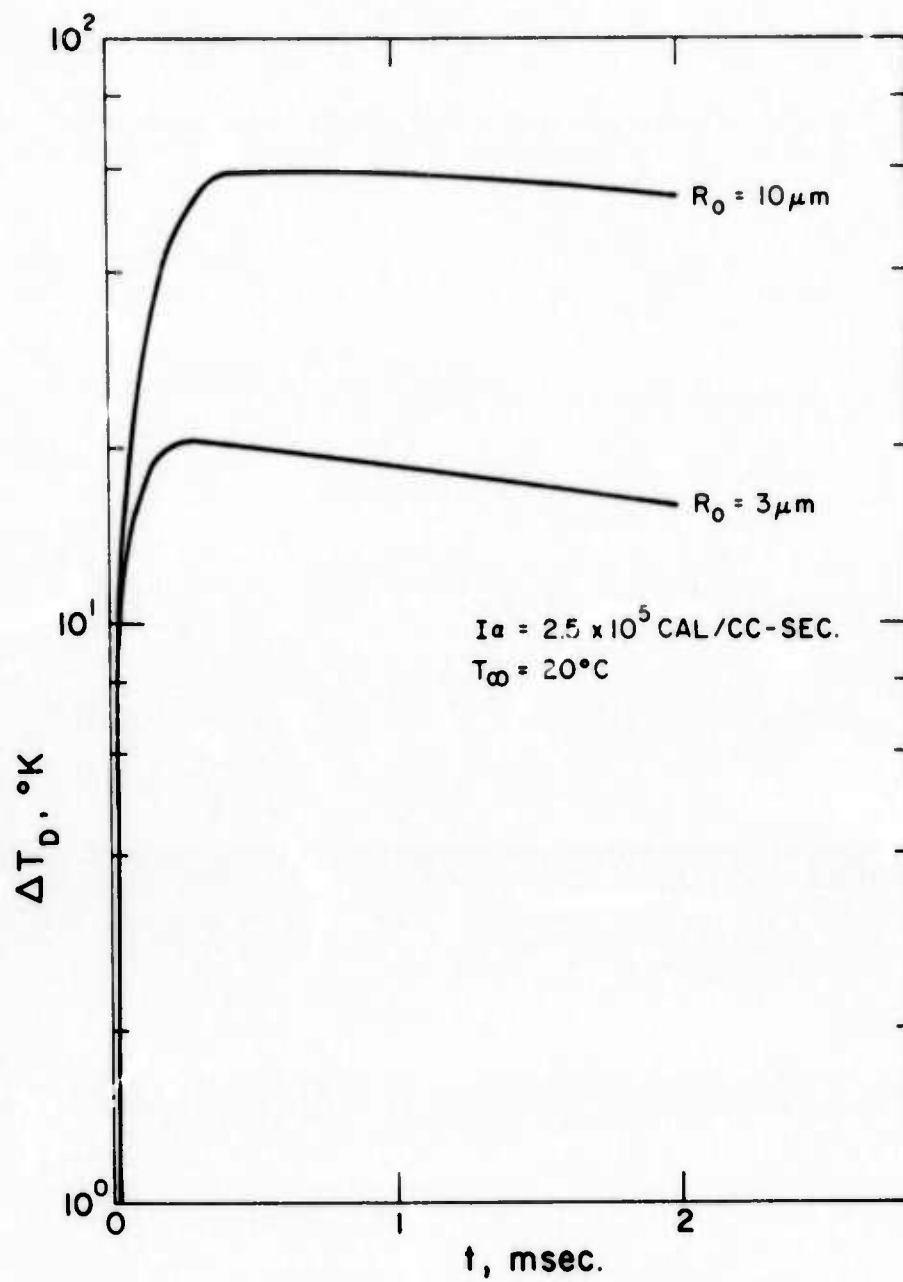


Fig. 2 The Rise in Droplet Temperature Over Ambient vs. Time for Two Different Droplet Sizes.

CURVE	Ia	R	t
	CAL/CC-SEC.	μm	mSEC.
A	2.5×10^6	10	0.2
B	2.5×10^5	10	2.0
C	2.5×10^5	3	0.5
D	2.5×10^5	3	2.0

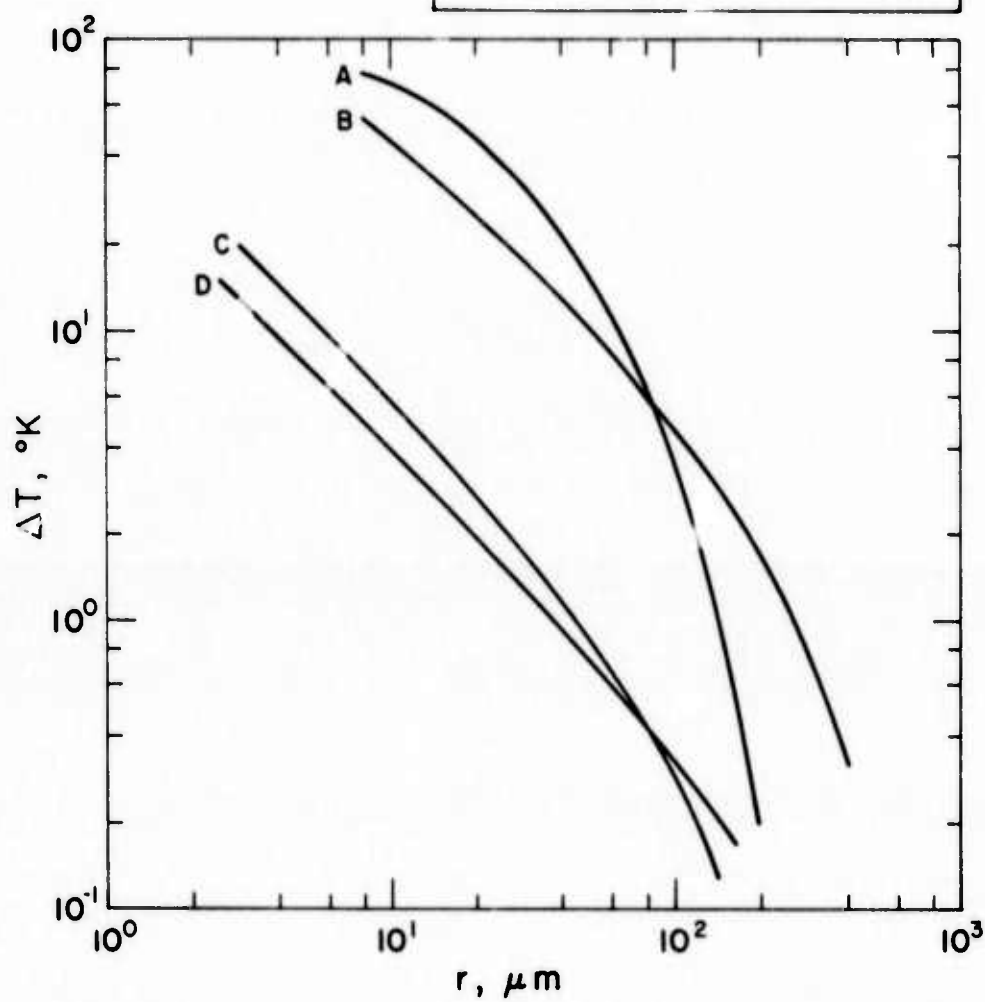


Fig. 3 Gas Temperature Profiles vs. Radius for Several Configurations.

lower at the longer time. This is because of the droplet cooling which occurs as a result of decreasing droplet size. At large r the situation is reversed since the gas temperature in that region is still being driven upwards towards an r^{-1} distribution. The characteristic time to approach this distribution at any distance r is given approximately by Eq. (2), and is independent of the intensity of the incident irradiation.

Curve B is a similar calculation for a $10\text{ }\mu\text{m}$ particle, $I_0 = 2.5 \times 10^5$ cal/cc-sec, at a time of 2 msec. Once again, the profile scales approximately as r^{-1} for small r . Shown for comparison as curve A is another calculation for a $10\text{ }\mu$ droplet with an I_0 of 2.5×10^6 cal/cc-sec at a time of 0.2 msec. Curves A and B correspond to two cases with the same energy input ($I_0 \Delta t$). However, they are radically different. Curve A is sharply bowed away from an r^{-1} distribution, demonstrating the effect of the convection term in Eq. (18). It is recalled from Eq. (32) that the initial vapor velocity scales linearly with I_0 and is thus an order of magnitude larger in case A than in case B. On the other hand, curve A falls below B at large r , since the "conduction length" of the gas scales as $t^{1/2}$, Eq. (2).

No calculations of the radial water vapor (ΔX_v) distributions are presented. However, these are quite similar in shape to the temperature distributions. This is to be expected since it was demonstrated earlier that the characteristic time for diffusion was approximately the same as that for heat conduction.

IV. SUMMARY AND FUTURE DIRECTIONS

A model has been developed for the prediction of the time dependent behavior of the gas surrounding a water droplet under irradiation by a high intensity laser beam. The basic phenomena included in the model are absorption, heating and vaporization in the droplet and convection, thermal conduction and diffusion in the gas. The model assumes volume absorption by the droplet, a constant pressure gas, and that the initial vapor velocity is much less than the local sound speed. The conditions under which these assumptions are appropriate have been identified.

A computer code employing a fully implicit Crank-Nicolson finite difference scheme has been developed for the numerical evaluation of the governing differential equations and a number of preliminary calculations has been performed. Simultaneously, a steady state analysis of the system has been performed in order to identify the salient features of the modeled phenomena over a wide range of the problem parameters. From this analysis it was demonstrated that the model was not strictly appropriate at large values of the quantity $R^2 I \alpha$, where the droplet temperature approaches the boiling point.

A number of tasks are presently in process on this program and results will be included in the final report. These include:

- A) A review of the properties of atmospheric aerosols, including particle size distribution functions, absorption coefficients, original nuclei, etc.
- B) An extension of the steady state analysis to include convection effects.
- C) A matrix of computer calculations of the time dependent radial temperature profiles about a droplet. Parameters to include particle radius, laser intensity, etc.
- D) First order estimates of the thermal blooming arising in typical scenarios.

E) A study of the effects of original Aitken nuclei, phase changes associated with original nuclei material and production of new chemical species. Possible internal boiling effects with accompanying superheating, fragmentation, mass transport and shockheating of the air surrounding the droplet will also be examined.

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