

Molecular-Flow Effects in Evaporation and Condensation Phenomena

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Content

- Remarks on the validity of continuum fluid dynamic descriptions
- Boltzmann Equation and kinetic boundary layers
- Moment solution of Boltzmann Eq. for evaporation/condensation
- Matching to external continuum flow
- Three cases
 1. Evaporation into shear flow
 2. Explosive boiling of liquid droplet
 3. Laser ablation of carbon
- Concluding remarks

Validity of continuum fluid dynamics on Navier-Stokes level

- $$\frac{\lambda}{c} \left| \frac{\partial u_i}{\partial x_j} \right|_{\max} \ll 1, \quad \frac{\lambda}{T} \left| \frac{\partial T}{\partial x_j} \right|_{\max} \ll 1$$

from Chapman-Enskog expansion of the Boltzmann Equation, where

- $$\lambda \approx \frac{\mu}{\rho c} : \text{Molecular mean free path}$$

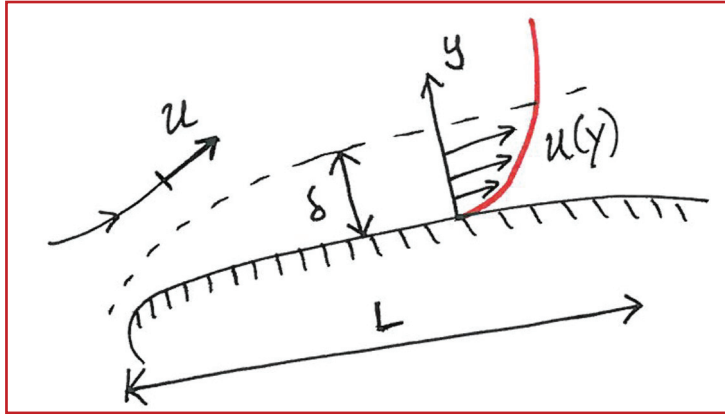
- $$c \approx \sqrt{RT} : \text{Molecular random thermal velocity}$$

- Also:
$$\text{Kn} = \frac{\lambda}{L} \approx \frac{\mu}{\rho c L} = \frac{\mu}{\rho U L} \cdot \frac{U}{c} \approx \frac{M}{Re}$$

Knudsen number



Macroscopic gradients in Boundary Layer



- Thickness of Prandtl boundary layer: $\delta \approx \frac{1}{\sqrt{\text{Re}}}$
- $\frac{\lambda}{c} \left| \frac{\partial u}{\partial y} \right| \approx \frac{\lambda U}{c \delta} \approx \frac{\lambda U}{c L} \sqrt{\text{Re}} \approx \frac{\lambda}{L} M \sqrt{\text{Re}} \approx \frac{M^2}{\sqrt{\text{Re}}}$
- since $\frac{\lambda}{L} \approx \frac{M}{\text{Re}}$ (von Karman)

Limiting cases for which $\frac{\lambda}{c} \left| \frac{\partial u_i}{\partial x_j} \right| \approx \frac{M^2}{\sqrt{\text{Re}}} \not\ll 1$:

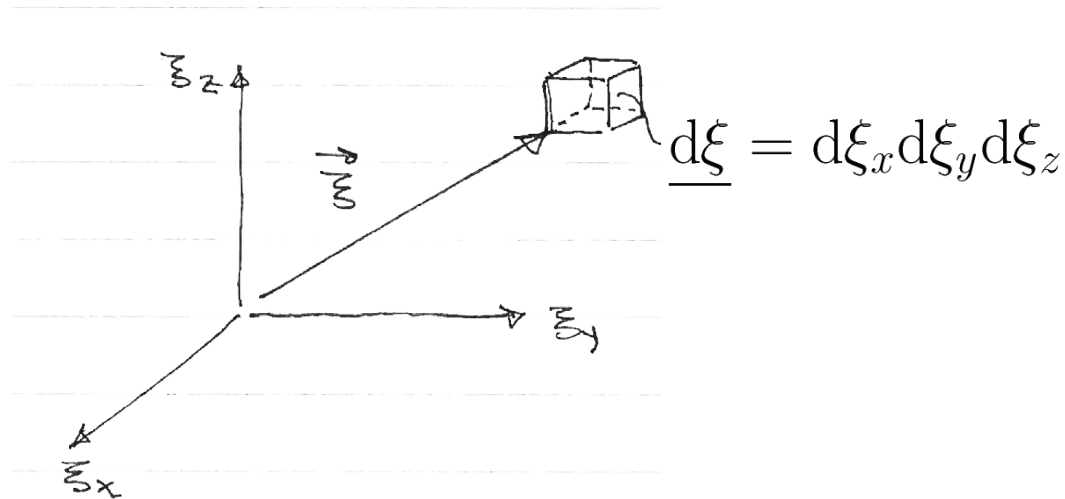
- Rarefied Gas Dynamics: $\frac{\lambda}{L} \not\ll 1$
(1950 → Grad, Cercignani, Kogan ...)
- Hypersonic flight: $M \gtrsim 7$
(1960 → Hays & Probstein ...)
- Shock wave structure: $\left| \frac{\partial U}{\partial x} \right| \sim \frac{U}{\lambda}$
(1950 → Gilbarg & Paulucci, Mott-Smith ...)
- Evaporation/condensation in single component systems: $\left| \frac{\partial V}{\partial y} \right| \sim \frac{c}{\lambda}$
(1970 → Sone, Kogan, Aoki ...)

The Boltzmann Equation (1867) for molecular distribution function

- $$\frac{\partial f}{\partial t} + \xi_j \frac{\partial f}{\partial x_j} = \int (f' f'_1 - f f_1) |\xi_1 - \xi| d\Sigma \underline{d\xi}_1 \quad (1)$$

binary collisions

- $f \underline{d\xi} \stackrel{\text{def}}{=} \text{number of molecules per unit volume with velocities in the range } \xi, \xi + d\xi$



- $$\int_{\xi} f \underline{d\xi} = n(t, \mathbf{x}) \quad ; \text{ number density of molecules}$$

Evaporation/Condensation Knudsen layer

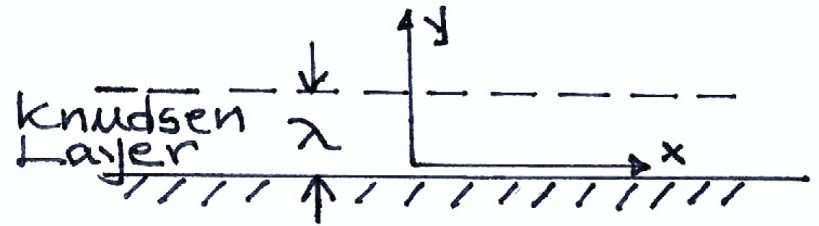
- Mathematics: Nondimensional Boltzmann Equation for 2D steady flow:

$$\epsilon \left(\xi_x \frac{\partial f}{\partial x} + \xi_y \frac{\partial f}{\partial y} \right) = \mathcal{C}(f f_1)_{\text{coll}}$$

$\mathcal{O}(1/\epsilon)$

Singular $\rightarrow$ Regular perturbation $\epsilon \rightarrow 0$:

$$\xi_y \frac{\partial f}{\partial y} = \mathcal{C}(f f_1)_{\text{coll}} \quad (2)$$



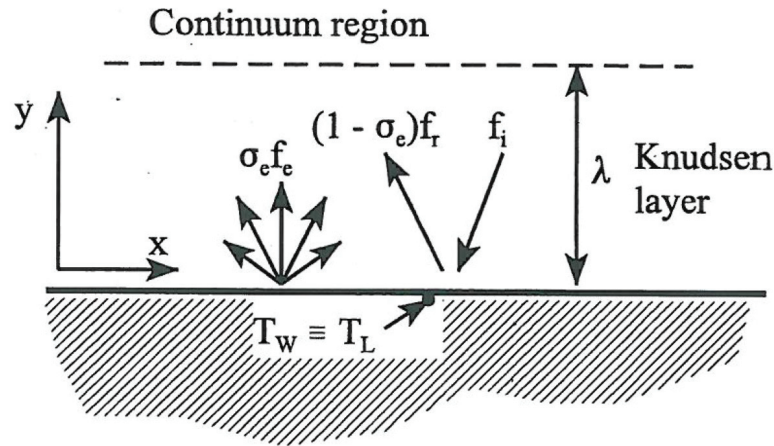
$$\epsilon = \frac{\lambda}{L}$$

: Knudsen number

Sone & Onishi (1978), Kogan (1992), Cercignani (2000)

Evaporation/Condensation Knudsen layer

- Physics:



- Two-sided molecular distribution

$$f_e(\boldsymbol{\xi}, T_L) \text{ for } \xi_y > 0$$

$$f_i(\boldsymbol{\xi}, T_\infty) \text{ for } \xi_y < 0$$

- Typically:

$$f_e = \frac{n_e(T_L)}{(2\pi RT_L)^{\frac{3}{2}}} \exp\left(-\frac{\xi^2}{2RT_L}\right) \quad (3)$$

- $n_e(T_L) \equiv n_{\text{sat}}(T_L)$

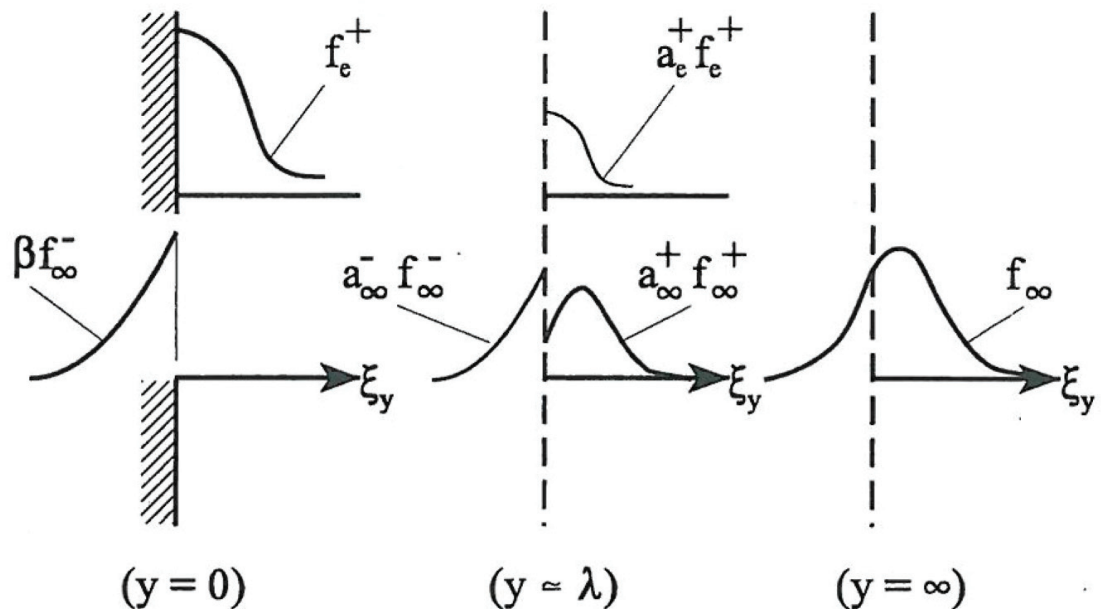
Solution of Boltzmann equation for evaporation

- Basic "Ansatz" for distribution function:

$$f(y, \xi) = \sum_{k=1}^3 a_k(y) f_k(\xi, T_k) \quad (4)$$

- Boundary conditions:

$$y = 0 : \quad \begin{aligned} a_e^+ &= 1 \\ a_\infty^+ &= 0 \\ a_\infty^- &= \beta \end{aligned} \quad y \rightarrow \infty : \quad \begin{aligned} a_e^+ &= 0 \\ a_\infty^+ &= 1 \\ a_\infty^- &= 1 \end{aligned} \quad (5)$$



Anisimov (1968),
Ytrehus (1977, 1997)

Conservation equations

- Moments of the Boltzmann Equation for the collision invariant m , $m\xi_y$, $\frac{1}{2}m\xi^2$ over the velocity space:

$$\frac{\partial}{\partial y} \int \xi_y \begin{pmatrix} m \\ m\xi_y \\ \frac{1}{2}m\xi^2 \end{pmatrix} f d\xi = \int \begin{pmatrix} m \\ m\xi_y \\ \frac{1}{2}m\xi^2 \end{pmatrix} \mathcal{C}(ff_1) d\xi = 0$$

- Leads to the classical conservation eqs., but with τ_{ij} (stresses) and q_i (heat flux) determined by the distribution function.

- For instance the 1D continuity equation reads:

$$\rho_e \sqrt{\frac{RT_L}{2\pi}} a_e^+ + \rho_\infty \sqrt{\frac{RT_\infty}{2\pi}} F^+ a_\infty^+(y) - \rho_\infty \sqrt{\frac{RT_\infty}{2\pi}} F^- a_\infty^-(y) = \rho_\infty v_\infty \quad (6)$$

$$F^\pm = \sqrt{\pi} S_\infty (\pm 1 + \operatorname{erf} S_\infty) + e^{-S_\infty^2}$$

Gas Kinetic Connection Problem

- Evaluated between the interphase surface ($y=0$) and external equilibrium ($y \rightarrow \infty$) according to Eq. (5), the conservation eqs. Read:

$$\rho_e \sqrt{\frac{RT_L}{2\pi}} - \rho_\infty \sqrt{\frac{RT_\infty}{2\pi}} \beta F^- = \rho_\infty v_\infty \quad (7a)$$

$$\frac{1}{2} \rho_e RT_L + \frac{1}{2} \rho_\infty RT_\infty \beta G^- = \rho_\infty v_\infty^2 + \rho_\infty RT_\infty \quad (7b)$$

$$2 \rho_e RT_L \sqrt{\frac{RT_L}{2\pi}} - 2 \rho_\infty RT_\infty \sqrt{\frac{RT_\infty}{2\pi}} \beta H^- = \rho_\infty v_\infty \left(\frac{1}{2} v_\infty^2 + \frac{5}{2} RT_\infty \right) \quad (7c)$$

with F^- , G^- , H^- being transcendental functions of the speed ratio

$$S_\infty = \frac{v_\infty}{\sqrt{2RT_\infty}} = \sqrt{\frac{\gamma}{2}} M_\infty$$

as given in Ytrehus (1977, 1997).

Solution of connection problem

- Solution of conservation Eqs (7) is expressed in terms of the single free control parameter

$$z = \frac{p_e}{p_\infty} \equiv \frac{p_{\text{sat}}(T_L)}{p_\infty} \quad (8)$$

as follows:

$$\sqrt{\frac{T_\infty}{T_L}} = -\frac{\sqrt{\pi}}{8} S_\infty + \sqrt{1 + \frac{\pi}{64}} S_\infty^2 \quad (9a)$$

$$z_e = \frac{2e^{S_\infty^2}}{F^- + \sqrt{\frac{T_\infty}{T_L}} G^-} \quad (9b)$$

$$\beta = \frac{2(2S_\infty^2 + 1) \sqrt{\frac{T_\infty}{T_L}} - 2\sqrt{\pi} S_\infty}{F^- + \sqrt{\frac{T_\infty}{T_L}} G^-} \quad (9c)$$

Tabulated results

Table 1 Gas dynamic parameters in evaporation

S_∞	p_e / p_∞	ρ_∞ / ρ_e	T_∞ / T_L	β	$\dot{m} / \dot{m}_e$
0.0	1.000	1.000	1.000	1.000	0.000
0.1	1.231	0.849	0.957	1.020	0.294
0.2	1.500	0.728	0.915	1.060	0.494
0.3	1.812	0.630	0.876	1.135	0.627
0.4	2.170	0.550	0.838	1.271	0.714
0.5	2.577	0.484	0.802	1.511	0.768
0.6	3.037	0.429	0.767	1.928	0.800
0.7	3.553	0.383	0.734	2.644	0.815
0.8	4.127	0.345	0.703	3.862	0.820
0.9	4.764	0.312	0.673	5.932	0.816
0.907*	4.813	0.310	0.671	6.132	0.816

* Critical value.

- Evaporation: $\left(\frac{p_e}{p_\infty}\right)_{\text{crit}} = 4.813$ Nozzle: $\left(\frac{p_0}{p_\infty}\right)_{\text{crit}} = \left(\frac{\gamma + 1}{2}\right)^{\frac{\gamma}{\gamma-1}} = 2.053$

- Linearized result:

$$\frac{v_\infty}{\sqrt{2RT_L}} = \frac{1}{2\sqrt{\pi}} \frac{32\pi}{32 + 9\pi} \frac{p_e - p_\infty}{p_e} \simeq 0.4705 \frac{p_e - p_\infty}{p_e} \quad (10)$$

Spatial Structure

Introducing a non-conserved moment ($m \xi_y^2$) into the Boltzmann Eq. and combining with local conservation eqs. of the type Eq. (6), a bilinear form results:

- $$\frac{\partial a_{\infty}^{-}}{\partial x} = -\frac{P(S_{\infty})}{\lambda_e} (a_{\infty}^{-} - 1) [a_{\infty}^{-} - r(S_{\infty})] \quad (11)$$

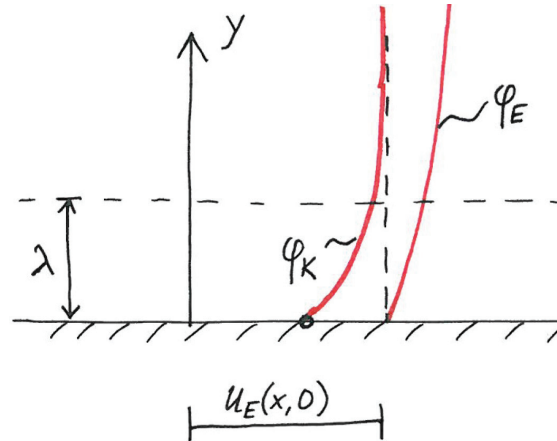
Relation to external equilibrium $a_{\infty}=1$ only if

$$r \leq 1, \text{ implying } \begin{aligned} S_{\infty} &\leq 0.907... \\ M_{\infty} &\leq 0.994... \end{aligned}$$

- Evaporation Knudsen layer restricted to subsonic flow normal to interphase surface.

Matching to External Continuum Flow

Matching principle:



φ_K : Euler
 φ_E : Knudsen layer

$$\lim_{y_K \rightarrow \infty} \varphi_K = \lim_{y_E \rightarrow 0} \varphi_E$$

(12)

Outer limit of
Inner solution

Inner limit of
outer solution

Aoki *et al.* (1991), Abramov & Kogan (1984):

$U_E(x, y = 0) = 0$ Evaporation

$U_E(x, y = 0) \neq 0$ Condensation

Why not Navier-Stokes?

- Navier-Stokes component normal to interphase surface:

$$\underbrace{u \frac{\partial v}{\partial x} + v \frac{\partial v}{\partial y}}_{v \Delta v / l} = - \frac{1}{\rho} \frac{\partial p}{\partial y} + \underbrace{\nu \left(\frac{\partial^2 v}{\partial x^2} + \frac{\partial^2 v}{\partial y^2} \right)}_{\nu \Delta v / l^2}$$

- $\Rightarrow l \sim \frac{\nu}{v} \sim \frac{c\lambda}{v} \sim \frac{\lambda}{M_y}$

that is: Similar scale as Knudsen layer and already accounted for.
Also: $v(y=0) = ?$

- Shear layer "blown off" in evaporation



$$v_{\infty} \simeq \sqrt{RT} \frac{\Delta p}{p} \gg v_{BL}$$

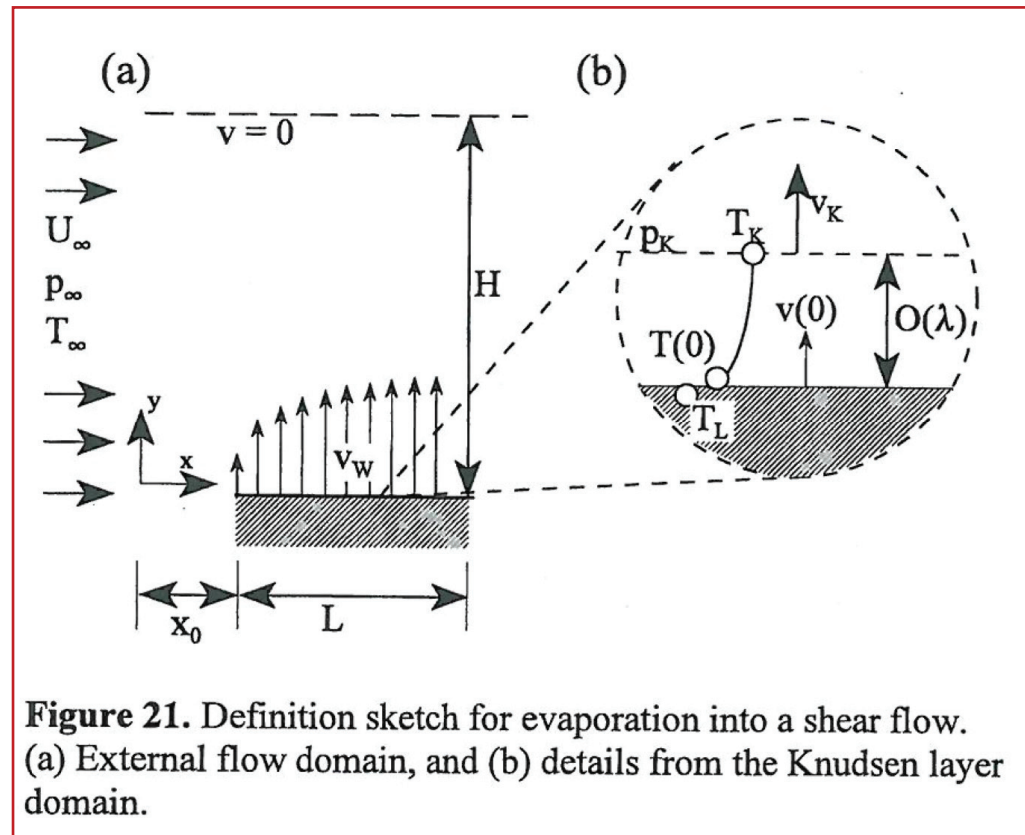
Lateral diffusion: $u / \sqrt{Re}$

Case 1: Evaporation into shear flow

- System: Water/water vapor at conditions such that

- $$\left| \frac{p_\infty - p_e}{p_e} \right| = \mathcal{O}(M_y) \ll 1$$

- $$\frac{U_\infty}{\sqrt{2RT_\infty}} \ll 1$$



Governing equations in external flow

- Description on Navier-Stokes level, mainly to detect eventual influence of turbulence

$$U \frac{\partial U}{\partial x} + V \frac{\partial U}{\partial y} = -\frac{1}{\rho} \frac{\partial p}{\partial x} + \frac{\partial}{\partial x} \left(\nu_{eff} \frac{\partial U}{\partial x} \right) + \frac{\partial}{\partial y} \left(\nu_{eff} \frac{\partial U}{\partial y} \right) \quad (13a)$$

$$U \frac{\partial V}{\partial x} + V \frac{\partial V}{\partial y} = -\frac{1}{\rho} \frac{\partial p}{\partial y} + \frac{\partial}{\partial x} \left(\nu_{eff} \frac{\partial V}{\partial x} \right) + \frac{\partial}{\partial y} \left(\nu_{eff} \frac{\partial V}{\partial y} \right) \quad (13b)$$

$$U \frac{\partial T}{\partial x} + V \frac{\partial T}{\partial y} = \frac{\partial}{\partial x} \left(\gamma_{eff} \frac{\partial T}{\partial x} \right) + \frac{\partial}{\partial y} \left(\gamma_{eff} \frac{\partial T}{\partial y} \right) \quad (13c)$$

Here $\nu_{eff} = \nu + \nu_t$, $\gamma_{eff} = \gamma + \gamma_t = \nu/P_r + \nu_t/P_{r_t}$ where P_{r_t} is turbulent Prandtl number

$$\nu_t = C \frac{k^2}{\epsilon}, \quad C=0.09$$

in standard k- ϵ model, Patankar (1980)

Boundary conditions

- On reactive boundary; $y=0$, $x \in [x_0, x_0+L]$:

$$V = v_w = \sqrt{2RT_L} \frac{4}{\sqrt{\pi}\beta_c} \frac{p_e - p_w}{p_e}, \quad \beta_c = \frac{32 + 9\pi}{4\pi} \quad (14a)$$

$$T = T_w = T_L - \frac{T_L}{\beta_c} \frac{p_e - p_w}{p_e}, \quad (14b)$$

$U = 0$, Evaporation

$U \neq 0$, Condensation

- p_w : Inner limit of external solution, to be determined.
- Iteration on the parameter $(p_e - p_w)/p_e$ after initial guess $p_w = p_\infty$.
- Extent of domain:
 $x_0 = 0.5\text{m}$, $L = 0.55\text{m}$, $T_L = 373.16\text{ K}$, $p_e(T_L) = 1\text{ atm}$, $R = 461.96\text{ J/K/kg}$,
 $p_\infty \in [0.98, 1.03]\text{atm}$, $U_\infty \in [20, 50]\text{m/s}$.

Computed results

- Obtained by TEACH-T code using SIMPLE algorithm, Patankar (1980)

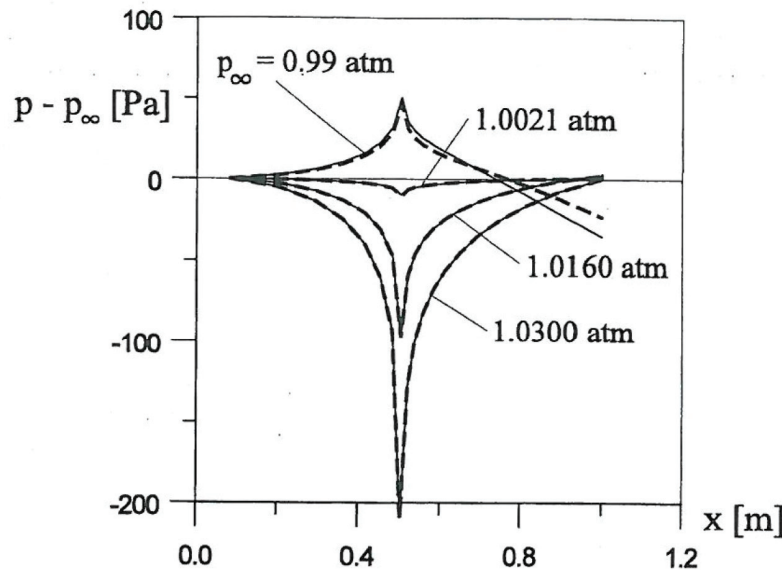


Figure 22. Differential pressure $p - p_\infty$ upstream of and along interphase surface in shear flow, $U_\infty = 20$ m/s. Dashed line: laminar flow, solid line: turbulent flow.

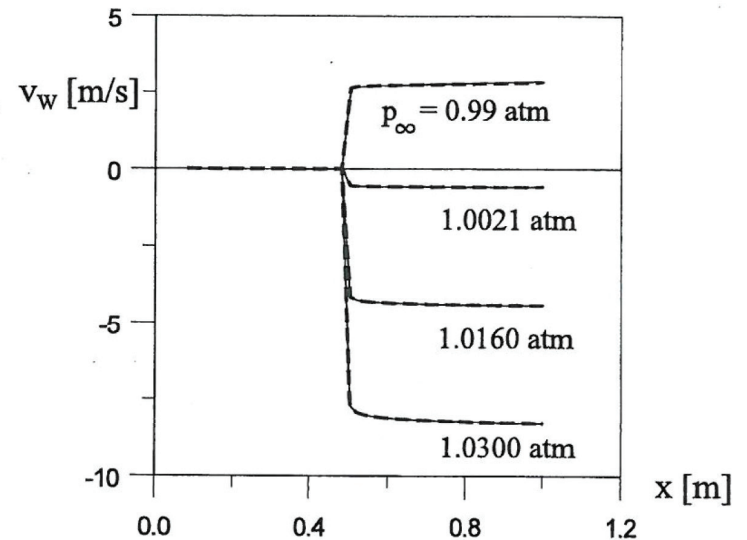
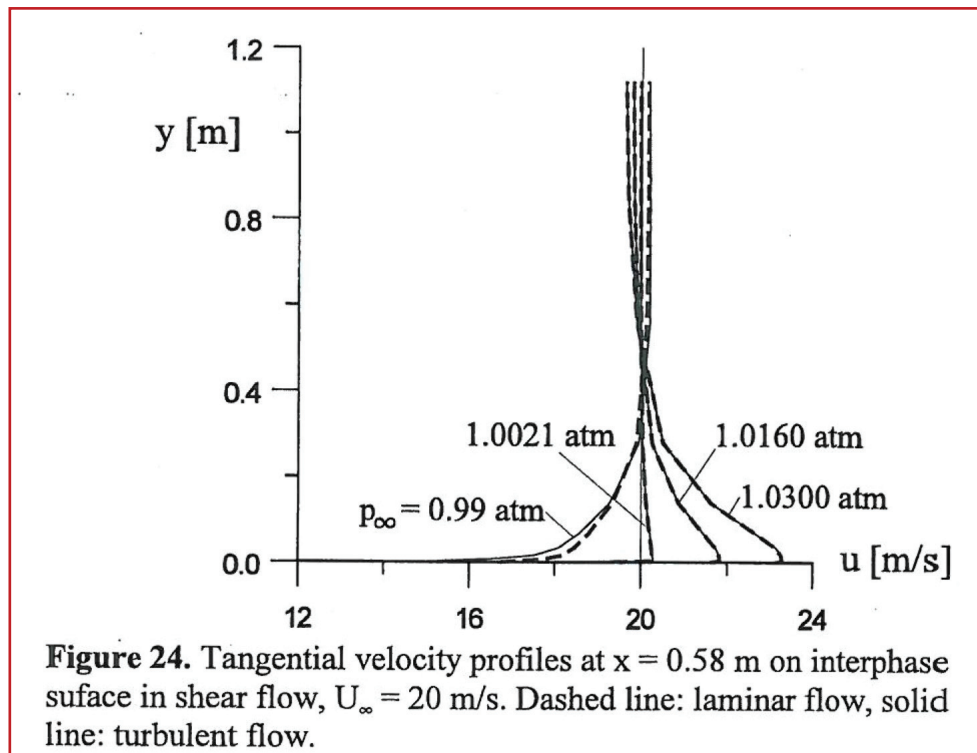


Figure 23. Normal velocity upstream of and along interphase surface in shear flow, $U_\infty = 20$ m/s. Dashed line: laminar flow, solid line: turbulent flow.

Computed results (contd.)



- Thickness of "shear profile", $\delta = 0.35$ m, of same order as geometric scale, $L = 0.55$ m; EULER result.
- Also EULER slip flow at wall $U_E(x, 0) > U_\infty$ for condensation.

Case 2: Explosive boiling of liquid droplet

- Rapid phase transition from highly superheated liquid to vapor.
- Typical situations:
 - Droplet immersed in hot medium
 - High intensity laser radiation
 - Passage through shock wave
 - Sudden decompression of liquid
- Background references and current understanding of problem in Shusser & Weihs (1999), Shusser *et al.* (2000).

Explosive boiling (contd. I)

- Governing eqs. by Shusser & Weihs (1999)
 - Extension of classical theories by Rayleigh (1917), Prosperetti & Plesset (1978).

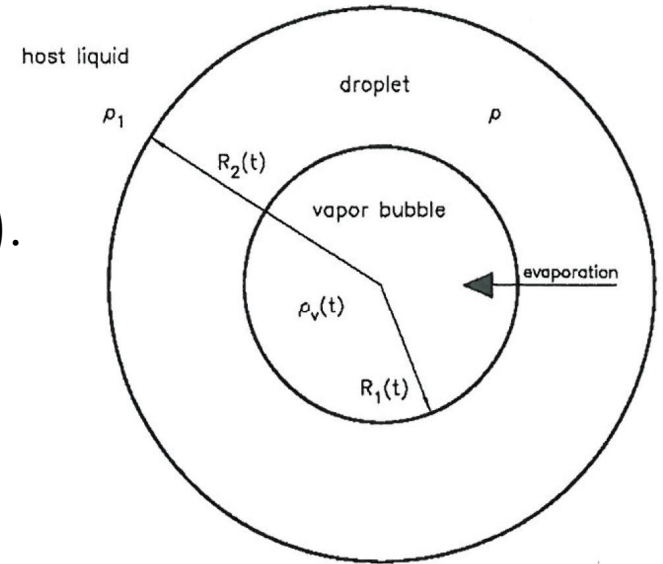


Fig. 1. Schematic illustration of explosive boiling of a liquid droplet.

$$\left(R_1 \ddot{R}_1 + 2\dot{R}_1^2 - 2\dot{R}_1 \frac{J}{\rho} - R_1 \frac{\dot{J}}{\rho} \right) \left[1 - \frac{R_1}{R_2} \left(1 - \frac{\rho_\ell}{\rho} \right) \right] + \frac{1}{2} \left(\dot{R}_1^2 - 2\dot{R}_1 \frac{J}{\rho} + \frac{J^2}{\rho^2} \right) \left[\frac{R_1^4}{R_2^4} \left(1 - \frac{\rho_\ell}{\rho} \right) - 1 \right] - \frac{J^2}{\rho_v \rho} + \frac{J^2}{\rho^2} = \frac{1}{\rho} \left[p_i - p_0 - \frac{2\sigma_\ell}{R_1} \right], \quad (15)$$

$$\dot{R}_2 = \left(\dot{R}_1 - \frac{J}{\rho} \right) \frac{R_1^2}{R_2^2}. \quad (16)$$

Explosive boiling (contd. II)

- Evolution of vapor pressure p_i

$$\frac{1}{p_i} \frac{\partial p_i}{\partial t} = -\frac{3\gamma}{R_1} \left(\dot{R}_1 - \frac{J}{\rho_\nu} \right) \quad (17)$$

- J is evaporated mass flux:

$$J = \sigma \rho_e \sqrt{\frac{RT_L}{2\pi}} - \sigma \rho_\infty \sqrt{\frac{RT_\infty}{2\pi}} \beta F^-(S_\infty) \quad (18)$$

$$\sqrt{\pi} S_\infty (1 + \operatorname{erf} S_\infty) + e^{-S_\infty^2}$$

modified for non-unity evaporation coefficient σ .

- Invokes the complete non-linear gas kinetic connection problem generalized to $\sigma \neq 1$, Ytrehus (1997).

Computed results

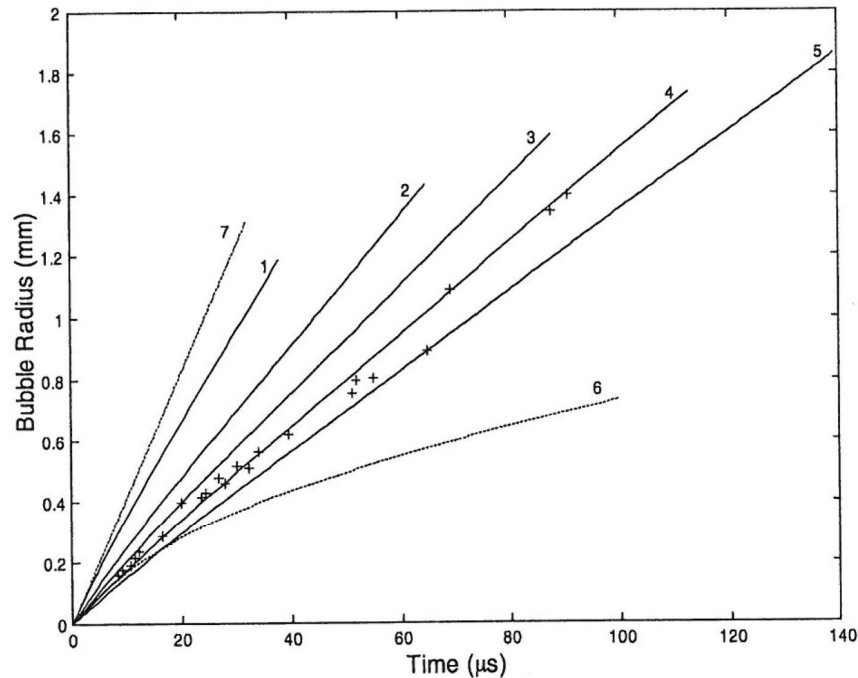
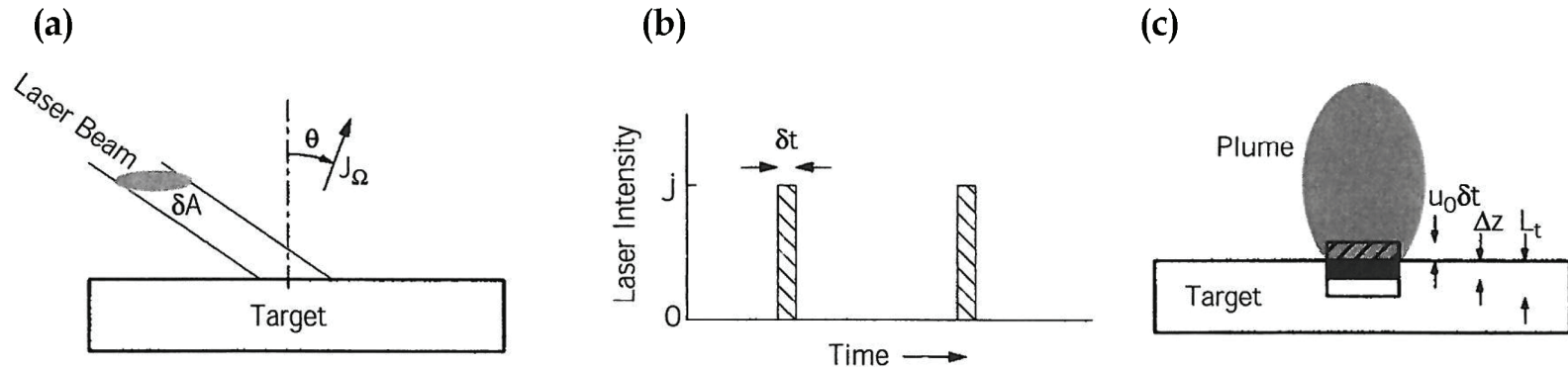


Fig. 2. Bubble radius as a function of time during explosive boiling for several values of the evaporation coefficient: (1) $a_w = 1$; (2) $a_w = 0.2$; (3) $a_w = 0.1$; (4) $a_w = 0.06$; (5) $a_w = 0.04$; (6) classical theory (Shepherd and Sturtevant 1982); (7) classical inertial growth rate (Shepherd and Sturtevant 1982); +, experiment (Shepherd and Sturtevant 1982).

- Butane droplet, 1mm diameter, immersed in ethylene glycol.
- Best fit with experiments of Shepherd & Sturtevant (1982) for $\sigma = 0.06$.
 - Comparable to molecular-dynamics simulations by Matsumoto (1998).

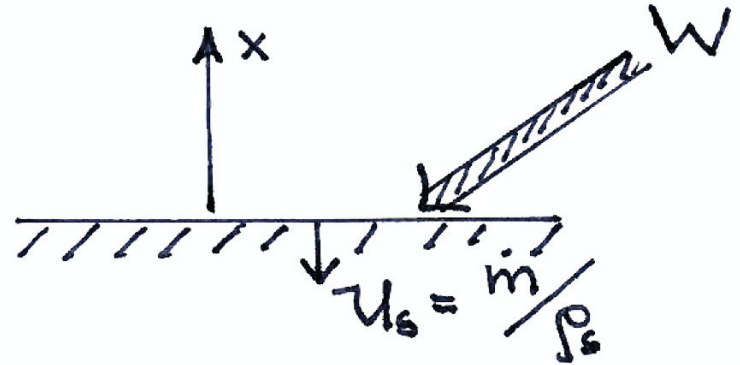
Case 3: Laser ablation of carbon



Objective: To understand and control the physics of laser ablation and mechanism of carbon nanotube growth, M. Shusser (2007), (2010).

Heat transfer in the solid

$$\frac{\partial T}{\partial t} - U_s \frac{\partial T}{\partial x} = \alpha \frac{\partial^2 T}{\partial x^2}$$



Initial and boundary conditions:

$$t = 0 : \quad T = T_0$$

$$x = 0 : \quad k \frac{\partial T}{\partial x} = W - \dot{m}L - \epsilon \sigma T^4$$

$$x \rightarrow -\infty : \quad T \rightarrow T_0$$

U_s : Interface recession velocity

W : Laser energy flux

L : Latent heat of sublimation

External gas dynamic problem

$$\frac{\partial \rho}{\partial t} + \frac{\partial}{\partial x}(\rho u) = 0 \quad (20a)$$

- $$\frac{\partial \rho u}{\partial t} + \frac{\partial}{\partial x}(\rho u^2 + p) = 0 \quad (20b)$$

$$\frac{\partial \rho E}{\partial t} + \frac{\partial}{\partial x}(\rho u E + up) = 0 \quad (20c)$$

- Boundary conditions at $x=0$ supplied from "outer limit" of Knudsen layer solution for evaporation, i.e. Eq. 9(a-c).

- For
$$\frac{p_{\text{sat}}(T_w)}{p_{\infty}} \geq 4.81, \quad M_{\infty} = 1$$

i.e., sonic outflow from Knudsen layer.

Numerical Method

- Heat transfer equation approximated by

$$\frac{T_i(t + \Delta t) - T_i(t)}{\Delta t} = U_s(t) \frac{T_{i+1}(t) - T_{i-1}(t)}{2h} + \alpha \frac{T_{i+1}(t) - 2T_i(t) + T_{i-1}(t)}{h^2}$$

- Gas dynamics eqs. solved by first-order Gudonov scheme:

$$\frac{\rho(t + \Delta t) - \rho(t)}{\Delta t} + \frac{(\rho u)_{i+\frac{1}{2}}(t) - (\rho u)_{i-\frac{1}{2}}(t)}{h} = 0.$$

- Flux at cell boundaries from solution of corresponding Riemann problem, Gottlieb & Groth (1988).

Results

- Calculations performed by Shusser (2007) for conditions corresponding to benchmark experiments by Greendyke *et al.* (2002):
 - Pulse length: 10ns
 - Beam diameter: 5mm
 - Laser intensity: $1.53 \cdot 10^{12} \text{ W/m}^2$

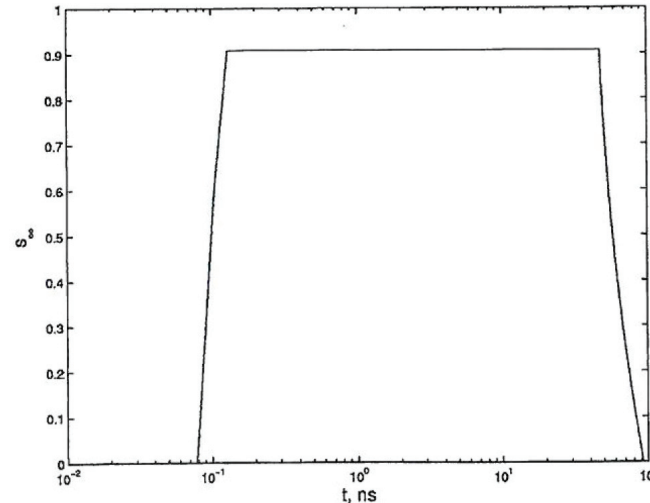


FIG. 1. Time dependence of dimensionless evaporation rate.

- Ablation rate most of the time restricted by kinetic theory limitation $M_\infty \leq 1$, ($S_\infty \leq 0.907$)
- Considerable postpulse evaporation, $\tau \simeq 80\text{ns}$.

Results (contd. I)

- Time dependence of surface temperature

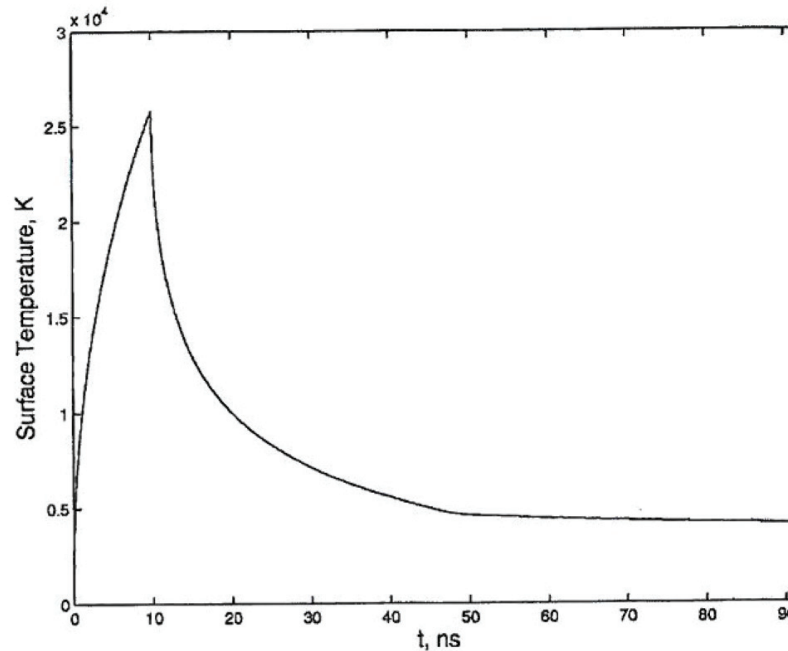


FIG. 2. Time dependence of surface temperature.

- Superheated surface up to 2500°K due to kinetic limit $M_\infty \leq 1$ of mass transfer rate.
 - Also the reason for postpulse evaporation.

Results (contd. II)

- Time dependence of ablated mass

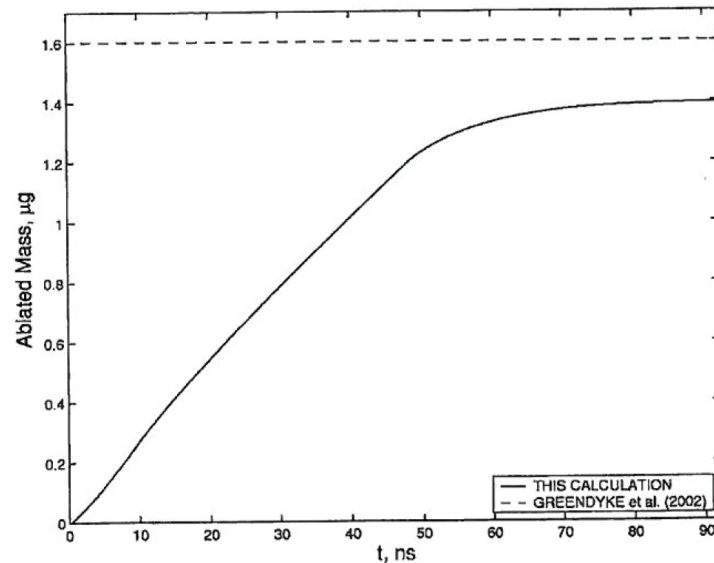


FIG. 5. Time dependence of ablated mass.

- Total ablated mass after postpulse period is within 15% of measured value.
- Accuracy limited by 1D modelling of external gas dynamics, and of heat transfer in solid.

Concluding remarks

- Criteria for validity of continuum macroscopic flow description are generally violated for evaporation/condensation in single-component systems
- Current understanding of such phenomena is based on kinetic theory and the Boltzmann equation.
- A kinetic boundary layer (Knudsen layer) adjacent to the interphase surface should be matched to the external flow.
- Composite solutions are demonstrated for several cases, including nanotechnology applications.

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