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TEMPERATURE EFFECTS ON THE BEHAVIOR OF LIQUID
HYDROGEN ISOTOPES INSIDE A SPHERICAL-SHELL,
DIRECTLY DRIVEN INERTIAL CONFINEMENT FUSION TARGET

Kyekyoon Kim
Lawrence C. Mok

Fusion Technology Laboratory
Department of Electrical and Computer Engineering
University of Illinois
Urbana, Illinois 61801

May 1984

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by

Kyekyoon Kim and Lawrence S. Mok

Fusion Technology Laboratory
Department of Electrical and Computer Engineering
University of Illinois
Urbana, Illinois 61801

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ABSTRACT

An important task in the inertial confinement fusion (ICF) research is to fabricate directly driven reactor targets which will result in high fusion yield. A spherical shell containing a uniform liquid (or solid) deuterium-tritium (D-T) layer on the inner surface represents one of the most popular high-gain directly driven ICF target configurations. The uniformity of the D-T layer is a must to achieve the desired high gain. The present work studies the temperature effects on the formation of a uniform liquid hydrogen layer inside a spherical glass shell (SGS). The profile of the liquid layer is first investigated for an isothermal case. An equation suitable for describing the profile is derived by including the London-van der Waals attractive forces between the liquid and substrate molecules. Two theoretical models are then established to explain the changes in the liquid layer profile under the influence of a vertically applied temperature gradient. It is found that a uniform liquid layer of single-component hydrogen can be created and maintained by an appropriate negative temperature gradient, namely, a temperature gradient which will keep the top of the SGS cooler than the bottom. The liquid layer is kept uniform dynamically: there are continuous fluid flows in the regions of the vapor and liquid. The characteristics of the fluid flows are obtained by solving the fluid equations under the low-Reynolds-number approximations. The effect of the component separation both in the liquid layer and the vapor region, which is induced by the temperature gradient, is studied when the enclosure inside the SGS is a mixture of hydrogen isotopes. A uniform layer can also be formed for the mixture liquid except that the required temperature

gradient is now positive in direction, unlike the case of the single-component liquid. The heating effect due to the radioactive decay of tritium is also evaluated. An experimental apparatus capable of generating a desired temperature gradient across the SGS at liquid hydrogen temperatures is described. The profiles of the liquid layer are observed for different temperature gradients and the results are in qualitative agreement with the theoretical predictions.

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TABLE OF CONTENTS

CHAPTER	Page
1 INTRODUCTION	1
2 LIQUID HYDROGEN PROFILE INSIDE AN ISOTHERMAL SPHERICAL-GLASS-SHELL	6
2.1 Introduction	6
2.2 Derivation of the Young-Laplace Equation	8
2.3 Proof that no Continuous Liquid Layer Solution is Possible with the Young-Laplace Equation	15
2.4 Young-Laplace Equation Containing Attractive Molecular Forces	19
2.5 The Retardation Effect of the London-van der Waals Forces	23
2.6 Summary	30
3 TEMPERATURE EFFECT ON LIQUID HYDROGEN LAYER CONFIGURATION: MODEL I - INTERFACIAL TENSION GRADIENT EFFECT	32
3.1 Introduction	32
3.2 Model I : Effect of Interfacial Tension Gradient	35
3.2.1 Velocity Fields	35
3.2.2 Temperature Fields	42
3.2.3 Condition for a Uniform Liquid Hydrogen Layer	45
3.3 Numerical Examples for Model I	49
3.4 Motion of a Nonconcentric Bubble	54
3.5 Summary	65
4 TEMPERATURE EFFECT ON LIQUID HYDROGEN LAYER CONFIGURATION: MODEL II - COMBINED EFFECT OF THE INTERFACIAL TENSION GRADIENT AND MASS TRANSFER	67
4.1 Introduction	67
4.2 Model II - Combined Effect of Mass Transfer and Interfacial Tension Gradient	69
4.3 Numerical Examples of Model II	75
4.4 Summary	80

5	TEMPERATURE EFFECT ON LIQUID LAYER CONFIGURATION OF MIXTURES OF HYDROGEN ISOTOPES	82
5.1	Introduction	82
5.2	Velocity and Temperature Fields	84
5.3	Concentration Profiles	87
5.4	Numerical Examples for SGS Filled with D-T	90
5.5	Summary	100
6	EFFECTS OF TRITIUM RADIOACTIVITY	101
7	EXPERIMENTAL APPARATUS AND RESULTS	106
7.1	Introduction	106
7.2	Experimental Apparatus	106
7.3	Experimental Results	114
8	CONCLUSIONS	119
APPENDIX		
A	SOLUTIONS FOR THE NONCONCENTRIC BUBBLE	123
	REFERENCES	128

NOMENCLATURE

A_n	expansion coefficients for stream function of liquid flow in concentric bubble case
$\bar{A}_n$	expansion coefficients for stream function of liquid flow in nonconcentric bubble case
$\hat{A}_n$	expansion coefficients for function of wall temperature in concentric bubble case
$\overset{\wedge}{\bar{A}}_n$	expansion coefficients for function of liquid temperature in nonconcentric bubble case
$\bar{a}$	solid-liquid interface in nonconcentric bubble case
B_{hs}	London constant of hydrogen and solid molecules
B_n	expansion coefficients for stream function of liquid flow in concentric bubble case
$\bar{B}_n$	expansion coefficients for stream function of liquid flow in nonconcentric bubble case
$\hat{B}_n$	expansion coefficients for function of wall temperature in concentric bubble case
$\overset{\wedge}{\bar{B}}_n$	expansion coefficients for function of liquid temperature in nonconcentric bubble case
Br	Brinkman number
b	ratio of bubble radius to inner radius of spherical shell
$\bar{b}$	liquid-vapor interface in nonconcentric bubble case
C_H	coefficient of London-van der Waals energy term
$\hat{C}_{He,n}$	expansion coefficients for function of temperature in helium gas column
C_n	expansion coefficients for stream function of liquid flow in concentric bubble case
$\bar{C}_n$	expansion coefficients for stream function of liquid flow in nonconcentric bubble case
$\hat{C}_n$	expansion coefficients for function of liquid temperature in concentric bubble case

$\bar{C}_n^A$	expansion coefficients for function of vapor temperature in nonconcentric bubble case
C_p	specific heat capacity
$D(x,y)$	function for total London-van der Waals energy
$D_B(x,y_B)$	total London-van der Waals energy calculated for lower half of the bubble
D_{DT}	deuterium diffusion constant in tritium
$\bar{D}_{He,n}^A$	expansion coefficients for function of temperature in helium gas column
D_n	expansion coefficients for stream function of liquid flow in concentric bubble case
$\bar{D}_n$	expansion coefficients for stream function of liquid flow in nonconcentric bubble case
$\bar{D}_n^A$	expansion coefficients for function of vapor temperature in concentric bubble case
$D_{ret}(x,y)$	function for retarded London-van der Waals energy
$D_T(x,y_T)$	total London-van der Waals energy calculated for upper half of the bubble
d	ratio of bubble radius to outer radius of a spherical shell
d_b	distance between bubble center and the horizontal axis in bipolar sphere coordinate system
d_i	distance between shell center and the horizontal axis in bipolar sphere coordinate system
d_m	distance between a solid molecule in wall region of a shell and a hydrogen molecule at point p_m
E_g	potential energy
E_{LVDW}	total London-van der Waals energy
E_{lv}	liquid-vapor interfacial energy
E_n	expansion coefficients for stream function of vapor flow in concentric bubble case
$\bar{E}_n$	expansion coefficients for stream function of vapor flow in nonconcentric bubble case
$\bar{E}_n^A$	expansion coefficients for function of vapor temperature in concentric bubble case

$E_p(r)$	London-van der Waals energy at point p_m
E_{sl}	solid-liquid interfacial energy
E_{sv}	solid-vapor interfacial energy
$e_{r\theta}$	rates of strain in θ -direction in spherical coordinate system
$e_1, e_2, e_3,$ $e_4, e_5, e_6,$ $e_7, e_8, \text{ and } e_9$	constants used in Model II
F_d	drag force on the bubble
F_n	expansion coefficients for stream function of vapor flow in concentric bubble case
$\bar{F}_n$	expansion coefficients for stream function of vapor flow in nonconcentric bubble case
$\overset{\Delta}{F}_n$	expansion coefficients for function of vapor temperature in concentric bubble case
Fr	Froude number
f_{mn}	function in calculation of temperature field in nonconcentric bubble case
f_l	function in the calculation of drag force in nonconcentric bubble case
G_n	expansion coefficients for stream function of vapor flow in concentric bubble case
$\bar{G}_n$	expansion coefficients for stream function of vapor flow in nonconcentric bubble case
g	gravitational acceleration
H	function used in calculus of variations
h_f	height of fluid layer
H_h	heat flux associated with evaporation and condensation
H_n	expansion coefficients for stream function of vapor flow in concentric bubble case
$\bar{H}_n$	expansion coefficients for stream function of vapor flow in nonconcentric bubble case
$\overset{\Delta}{H}_n$	function for heat of evaporation and condensation

$h_{\ell v}$	heat of vaporization
I_n	function for interfacial tension gradient of concentric bubble
$\bar{I}_{n+1}$	function for interfacial tension gradient of nonconcentric bubble
J_n	function for temperature fields of Model II
$\bar{J}_1, \bar{J}_1$	functions for velocity fields in nonconcentric bubble case
K_n	function for velocity of concentric bubble
$\bar{K}_n$	function for velocity of nonconcentric bubble
k_{He}	thermal conductivity of helium gas
k_ℓ	liquid thermal conductivity
k_v	vapor thermal conductivity
k_w	thermal conductivity of solid wall
$\bar{L}_n$	function for velocity of nonconcentric bubble
L_t	uniform liquid layer thickness
$\bar{L}$	characteristic length in bipolar sphere coordinate system
M_n	function used in the normal pressure calculation
$\bar{M}_n$	function for temperature fields in nonconcentric bubble case
N	number density of liquid molecules
N_n	function for temperature fields in concentric bubble case
N_s	number density of solid molecules
N_v	number density of vapor molecules
n	integer
Pe	Peclet diffusion number
$P_n(\cos \theta)$	Legendre function
Pr	Prandtl number
p_ℓ	pressure in liquid
p_{sat}	saturated vapor pressure

p_v	pressure in vapor
p_1	derivative of saturated vapor pressure with respect to temperature
Q_l	strength of heat sources in liquid
Q_v	strength of heat sources in vapor
Q_w	strength of heat sources in wall region
R, r	radial distance in spherical coordinate system
Ra	Rayleigh number
R_B	bubble radius
Re	Reynolds number
R_i	inner radius of spherical shell
R_o	outer radius of spherical shell
S	interfacial area
$S_{G,n}, S_{I,n}$	functions for normal pressure calculations of Model II
$\bar{S}_n$	function for velocity of nonconcentric bubble
T	tritium
T_{avg}	average temperature of a nonisothermal shell
$T_{ext}(\theta)$	external temperature
T_{He}	temperature in helium gas column
T_l	temperature in liquid
T_n	function for external temperature
T_{ref}	reference temperature
T_v	temperature in vapor
T_w	temperature in wall region
T_0	constant term Legendre expansion of external temperature
t_B	bottom liquid layer thickness
$\bar{t}_{ls,1}$	temperature gradient at liquid-solid interface

$(t_{ls,1})_S$	temperature gradient at liquid-solid interface required for having uniform liquid layer
$(\bar{t}_{ls,1})_S$	temperature gradient at liquid-solid interface required to maintain stationary nonconcentric bubble
t_n	coefficients in Legendre expansion of external temperature
t_T	top liquid layer thickness
$(t_1)_S$	temperature gradient on outer surface of spherical shell required for having uniform liquid layer
U	steady-state migration velocity of concentric bubble
$\bar{U}$	steady-state migration velocity of nonconcentric bubble
$U(d_m)$	London potential
V_l	liquid volume
V_{ref}	reference velocity
V_v	vapor volume
v_l	liquid flow velocity
v_r	r-component of fluid flow
v_v	vapor flow velocity
v_n	n-component of fluid flow
v_θ	θ -component of fluid flow
v_ξ	ξ -component of fluid flow
$x_n(\xi)$	function for liquid flow in nonconcentric bubble case
x	coordinate of isothermal liquid-vapor interface
x_D	deuterium mole fraction in liquid
x_m	coordinate at liquid-vapor interface where $dx/dy = 0$
x_{n-1}	coefficients in Legendre expansion of interfacial tension
x'	first derivative of x with respect to y
x''	second derivative of x with respect to y
x'_D	mole fraction gradient of deuterium in liquid
$y_n(\xi)$	function for vapor flow in nonconcentric bubble case

y	coordinate of isothermal liquid-vapor interface
y_B	coordinates for lower half of the bubble
y_D	deuterium mole fraction in vapor
y_m	coordinate at liquid-vapor interface where $dx/dy = 0$
y_T	coordinates for upper half of the bubble
$\bar{z}_n$	function for velocity of nonconcentric bubble
α	thermal diffusivity
α, β	constants in Young-Laplace equation
β_{th}	thermal expansion coefficient
β^-	beta particle
γ_{lv}	liquid-vapor interfacial tension
γ_{sl}	solid-liquid interfacial tension
γ_{sv}	solid-vapor interfacial tension
γ_0	constant term in Legendre expansion of interfacial tension
γ_1	magnitude of derivative of the liquid-vapor interfacial tension with respect to temperature
$\delta_{2,n}$	delta function
Δd	distance between the centers of bubble and shell
Δp	pressure difference
Δp_{lv}	pressure difference across liquid-vapor interface
ϵ_1	ratio of liquid and vapor thermal conductivities
ϵ_2	ratio of solid and liquid thermal conductivities
ϵ_3	ratio of helium gas and solid thermal conductivities
ζ_1	ratio of liquid and vapor densities
η, ξ	coordinates in bipolar sphere coordinate system
θ	angle
λ_c	characteristic wavelength of electronic oscillations of atoms

λ_l	Lagrange multiplier
μ_l	liquid viscosity
μ_v	vapor viscosity
ν	kinematic viscosity
$\bar{\nu}$	antineutrino
ρ_D	deuterium density in mixture
ρ_l	liquid density
ρ_T	tritium density in mixture
ρ_v	vapor density
σ	ratio of liquid and vapor viscosities
$\tau_{rr,v}, \tau_{rr,l}$	normal stresses in vapor and liquid on bubble surface
$\tau_{10\%}$	liquid sagging time
Φ	viscous dissipation function
ψ_l	Stokes stream function in liquid
ψ_v	Stokes stream function in vapor
Ω	function for velocity of nonconcentric bubble
$P_{n+1}(\cos\theta)$	Gegenbauer function
$f(R,r)$	function for retarded London-van der Waals energy

CHAPTER 1

INTRODUCTION

As human population grows, demands for energy -- particularly for electrical energy -- will increase inevitably. Because reserves of fossil fuel are limited, society cannot continue to rely on this traditional energy source. Even if petroleum and coal existed in abundance, their continued use at the present rate of consumption would severely pollute the environment. While nonfossil fuel energy sources such as water power, solar heat, geothermal energy, and wind power offer alternative sources of electricity, their intrinsic limitations cause them to be secondary power sources only. Reliable and relatively inexpensive, the use of nuclear fission energy to generate electricity does not offer a satisfactory energy source, either. The scarcity of fissile fuel and the high level radioactive waste that fission reactors produce make it a less attractive power source. Therefore, other potential fuel resources must be explored to meet the ever-growing demands for energy.

Nuclear fusion has been regarded as the most promising choice for the future energy source for many years, because its fuel is inexhaustible and gives out much less radioactive waste. This form of energy is released when two light nuclei are brought together to form a heavier nucleus. Although nuclear fusion occurs extraterrestrially in stars and is successfully produced on earth in an uncontrolled manner, that is, in the explosion of a fusion bomb [1], using it to generate electricity is still far from reality. Numerous research programs are now underway to find an appropriate method to harness the nuclear fusion.

One of the major problems in controlling nuclear fusion is how to handle

the super hot fusion fuel. The temperature of the fuel must be high because the reaction rate of fusion fuel, in general, will not be significant until its temperature reaches 10^7 K (about 1 keV). For instance, the deuterium-tritium (D-T) reaction, which has the highest reaction rate among all the fusion fuel, will not be at the peak level until its temperature reaches 7×10^8 K (about 60 keV) [2]. Two schemes have been proposed and implemented in the current fusion experiments to confine such high temperature fuel. One is magnetic confinement in which highly ionized fuel is confined by a magnetic field. The other is inertial confinement in which fuel is compressed and confined by the inertia of matter. The goal of these experiments is to demonstrate the scientific feasibility of the controlled nuclear fusion, that is, to reach a point where the released fusion energy is equal to the energy needed to start the reaction. This critical point is proportional to the product of the density and confinement time of the fuel at a given temperature, and was first derived by Lawson [3]. For a fuel containing D-T, the product is about 10^{14} ions/cm³ at a temperature of 10 keV [4].

In the inertial confinement fusion (ICF), the fusion fuel is put inside a spherical shell (called a target), which is illuminated by energetic laser or ion beams from all directions. The outer portion of the target is then heated up and blasted off, and the fuel in the middle of the target is subsequently compressed according to Newton's laws. While the fuel at the center is compressed and raised to high temperatures, a series of fusion reactions take place. A sudden release of a large amount of energy at the center of the target then forces the target to explode. After the target is disintegrated, fusion reaction is also stopped. The confinement time in this case is therefore very short; its typical value is about 10^{-10} second. Because of

this limitation in the confinement time, the fuel has to be compressed to high density in order to have a high fusion yield, and to reach or exceed the critical point derived by Lawson. For instance, a D-T fuel must be compressed to 100 times its solid density in order to reach the Lawson criterion.

Because of these characteristics, one of the main tasks in ICF research is to develop targets of optimum design that can maximize the fuel compression. There are two basic target designs: one is directly driven, in which the targets absorb the beam energy in an outer layer, which acts directly to compress the fuel; the other is Hohlraum targets which convert the beam energy into x-rays, which are then used to compress the fuel [5].

Among the direct targets, a spherical glass shell (SGS) containing D-T gas is the most commonly used in the experiments [6-9]. Recently, Mason [10] has theoretically predicted that fusion yield will be enhanced if the glass shell contains a uniform layer of solid D-T on its inner surface instead of gaseous D-T. These kinds of targets are sometimes called cryogenic targets because they have to be kept at solid D-T temperatures prior to being illuminated by the high energy beams. Some researchers have found that fusion yields are indeed enhanced when cryogenic targets are used [11-13].

The use of solid D-T layer instead of gaseous D-T is now widely adopted in the designs of new high gain directly driven targets, where the gain is defined as the fusion energy output divided by the beam energy input [14]. The remaining question is how to fabricate the high gain targets. Several methods have been developed to produce the targets, in particular, the simple cryogenic targets which are spherical glass shells (SGS) containing a uniform layer of solid D-T [15-28]. One of the difficulties encountered in the fabrication is that the thickness of the solid D-T layer is difficult to keep uniform. A

uniform layer, however, is a must because of the stringent symmetry requirements demanded of the target implosion [13,14,29].

A technique known as fast isothermal refreezing is capable of overcoming the difficulty. A target (or an SGS) is first cooled down to around 5K isothermally resulting in frozen D-T inside the target; the frozen D-T is then evaporated by a moderate-power laser beam. After D-T is completely vaporized, the laser is turned off and the gaseous D-T refreezes on the inner surface of the SGS, forming a uniform solid layer. The uniformity of the solid layer depends heavily on the freezing time, which is the time needed for the gaseous D-T inside the SGS to freeze into solid D-T. The thinner the solid layer, the shorter is the freezing time and hence the shorter is the time that the D-T condensate is in the liquid state. Consequently, the effect of the gravity induced liquid sagging on the uniformity of the solid layer is less. A uniform layer of about 2 μm thickness has been produced successfully by this technique [16]. However, for a thicker layer, the result is less satisfactory because of the foregoing described liquid sagging problem.

Based on the considerations of optimum fusion mass [10], requirements of the implosion symmetry [13,14], and the hydrodynamic instabilities during target implosion [10,14,30,31], the solid D-T layer inside the directly driven future ICF target is expected to be much thicker; therefore, a new technique other than the fast isothermal refreezing needs to be developed. Because of the finite freezing time, the key issue here is to find a method with which a uniform liquid layer can be created and maintained inside an SGS. Once a uniform liquid layer is produced, changing it to a solid layer becomes a relatively straightforward task.

The objective of the present work is to study the general behavior of a liquid hydrogen layer inside an SGS. The configuration of a liquid hydrogen

layer inside an isothermal SGS is first investigated in Chapter 2. A continuous liquid hydrogen layer is found inside an SGS, and the liquid layer at the top of the SGS is always thinner than that at the bottom due to gravity. During the target cooling experiments performed respectively by Grilly [17], Henderson et al. [18], Kim and Rieger [28], Fanning et al. [32], and Darling et al. [24], it was found that a vertical temperature gradient across the SGS is useful in creating a uniform liquid layer inside of it. However, no detailed explanation of the phenomenon was given, other than the simple qualitative one presented by Henderson, Simms, and Jacobs [34]. Detailed mathematical models are developed in the present work and are described in Chapters 3, 4, and 5. The models are capable of predicting the temperature gradient on the outer surface of an SGS which is required for the formation of a uniform liquid layer, both for single-component hydrogen and a mixture of hydrogen isotopes. The model described in Chapter 5 explains the reasons why the behavior of a liquid layer of hydrogen mixture is different from that of single-component hydrogen in a nonuniform temperature field, as observed in the early target cooling experiments. If an SGS is filled with deuterium-tritium mixture, heat will be generated internally by the tritium radioactive decay. The effects of tritium radioactivity are studied in Chapter 6. Experiments intended to verify the theoretical predictions have been performed and the results are presented in Chapter 7. Chapter 8 is for the conclusions and some suggestions for future investigation.

CHAPTER 2

LIQUID HYDROGEN PROFILE INSIDE AN ISOTHERMAL SPHERICAL-GLASS-SHELL

2.1 Introduction

The static profile of a liquid inside an isothermal container is generally governed by the fundamental equation of capillarity derived in 1805 by Young [35] and Laplace [36]. This Young-Laplace (YL) equation is

$$\frac{1}{R_1} + \frac{1}{R_2} = \frac{\Delta p}{\gamma_{lv}} \quad , \quad (2.1)$$

where R_1 and R_2 are the two principal radii of curvature of the liquid-vapor interface, Δp is the pressure difference across the interface, and γ_{lv} is the liquid-vapor interfacial tension. In essence, the YL equation is an equation for the macroscopic force balance at the liquid-vapor interface. It was also obtained by Gauss [37] and later proved by Gibbs [38] by means of the minimum energy principle, which says that a system is in static equilibrium if it is in the minimum energy configuration.

Bashforth and Adams [39] were the first to attempt to directly solve the YL equation to find the profile of a sessile drop, i.e., a liquid drop sitting on a horizontal plane. Since then, numerous researchers have derived the YL equation in various forms to describe the liquid profiles in containers of different shapes, for instance, Concus [40] for a meniscus in a vertical right circular cylinder, Benedikt [41] for a liquid inside a rectangular tank, and Li [42] for a liquid in a spherical vessel. In these studies, the contact angle of the liquid is treated as a boundary condition for the YL equation.

The YL equation applicable to the present case, namely, a liquid with a

nonzero contact angle inside a spherical glass shell (SGS), is derived in Section 2.2. The resulting equation is similar to Li's, except that the mathematical method used is slightly different. The YL equation is then solved numerically with arbitrarily assigned contact angles. When the contact angle is zero, however, one finds that the YL equation possesses no solution. The mathematical proof of this nonexistence of solution is carried out in Section 2.3. Since liquid hydrogen has a zero contact angle with glass [43], the YL equation is not applicable to describe its profile inside an SGS.

Because of the geometry of the spherical shell, zero contact angle means that the solid-liquid-vapor contact line disappears and that the liquid hydrogen will wet the entire surface of the shell. The resulting configuration is a hydrogen vapor bubble completely surrounded by a continuous liquid layer. Due to gravity, the bubble is likely to move upward and making the liquid layer near its top area thinner. It will, however, be stopped eventually by the extra pressure in the thin liquid layer, which is created by the intermolecular attraction forces between liquid and solid molecules. In order to predict the final position of the stationary bubble, or the static profile of the continuous liquid layer, such intermolecular forces between liquid and solid molecules are necessary to be included in the YL equation. This idea has been proven useful by Padday [44], Deryagin et al. [45], Philip [46], Renk et al. [47], and Mohanty et al. [48] in their studies on the profile of the transition region between a wetting film and a capillary meniscus. Since hydrogen and glass molecules are non-polar and neutral, only the London-van der Waals attraction forces are needed to account for their interaction. The re-derivation of the YL equation with inclusion of the London-van der Waals forces between the liquid and solid molecules is presented in Section 2.4. The possible retardation effect of the London-van

der Waals forces is discussed in Section 2.5.

2.2 Derivation of the Young-Laplace Equation

Based on the principle of minimum energy, the method of calculus of variations is employed to find a differential equation describing the profile of the liquid-vapor interface inside a spherical glass shell (SGS). The coordinate system to be used is shown in Fig. 2.1a where the thick solid line represents the inner surface of the SGS. Since the y-axis is placed parallel to the gravitational force, the profile of the liquid-vapor interface can be assumed symmetric about the y-axis. The gravitational force and the interfacial tensions at the liquid-vapor, liquid-solid and solid-vapor interfaces, respectively, are considered to be the major energy contributions to the system which consists of an SGS filled with liquid. The expressions of the gravitational energy (E_g), and the energies from the liquid-vapor (E_{lv}), the solid-liquid (E_{sl}) and the solid-vapor (E_{sv}) interfacial tensions are, respectively, as follows:

$$E_g = (\rho_l - \rho_v) g \int_{\tilde{y}_l}^{\tilde{y}} \tilde{y} dv$$

$$= 2\pi \int_{\tilde{y}_1}^{\tilde{y}_2} \frac{1}{2} (\rho_l - \rho_v) g \tilde{y} [R_i^2 - (\tilde{y} - R_i)^2 - \tilde{x}^2] d\tilde{y}, \quad (2.2)$$

$$E_{lv} = \gamma_{lv} \int_{\tilde{S}_{lv}} ds = 2\pi \int_{\tilde{y}_1}^{\tilde{y}_2} \gamma_{lv} \tilde{x} [1 + (\tilde{x}')^2]^{1/2} d\tilde{y}, \quad (2.3)$$

$$E_{sl} = \gamma_{sl} \int_{\tilde{S}_{sl}} ds = 2\pi \int_{\tilde{y}_1}^{\tilde{y}_2} \gamma_{sl} R_i d\tilde{y}, \quad (2.4)$$

Fig. 2.1 (a) Cylindrical coordinate system for the description of an SGS consisting of a liquid layer having a nonzero contact angle with the substrate.
 (b) The system shown in (a) redefined in dimensionless units.

$$E_{SV} = \gamma_{SV} \int_{S_{SV}} ds = \text{constant} - 2 \pi \int_{\tilde{y}_1}^{\tilde{y}_2} \gamma_{SV} R_i d\tilde{y}, \quad (2.5)$$

where g = gravitational acceleration,

ρ = mass density,

γ = interfacial tension,

R_i = inner radius of the SGS,

V = volume,

S = interfacial area,

x' = first derivative of x with respect to $y = dx/dy$,

and the subscripts, l , v , and s are, respectively, for liquid, vapor, and solid; the notation " $\sim$ " indicates that the variables underneath are dimensional quantities (as will be throughout this work).

The variations are now subject to the constraint of the liquid volume inside the SGS, which remains constant at a given temperature and fill pressure of the SGS. The expression of the liquid volume is

$$\tilde{V}_l = \text{constant} + 2 \pi \int_{\tilde{y}_1}^{\tilde{y}_2} \frac{1}{2} [R_i^2 - (\tilde{y} - R_i)^2 - \tilde{x}^2] d\tilde{y}. \quad (2.6)$$

The corresponding function required for the calculus of variations [49] is obtained by combining Eqs. (2.2) through (2.6), i.e.,

$$H = \frac{1}{2} (\rho_l - \rho_v) g \tilde{y} [R_i^2 - (\tilde{y} - R_i)^2 - \tilde{x}^2] + \gamma_{lv} \tilde{x} [1 + (\tilde{x}')^2]^{1/2} + R_i (\gamma_{sl} - \gamma_{sv}) + \frac{1}{2} \lambda_l [R_i^2 - (\tilde{y} - R_i)^2 - \tilde{x}^2], \quad (2.7)$$

where λ_l is the Lagrange multiplier.

Substituting Eq. (2.7) in the Euler equation [49], i.e.,

$$\frac{d}{dy} \left(\frac{\partial H}{\partial x'} \right) - \frac{\partial H}{\partial x} = 0, \quad (2.8)$$

one has

$$x'' [1 + (x')^2]^{-3/2} - x'^{-1} [1 + (x')^2]^{-1/2} = - \frac{(\rho_L - \rho_V) g y + \lambda_L}{\gamma_{LV}},$$

$$\text{where } x'' = d^2x/dy^2. \quad (2.9)$$

It can be seen from this equation that λ_L actually has a physical meaning. It represents the internal pressure of the liquid when the gravitational force is not present. Let

$$\begin{aligned} x &= \frac{\tilde{x}}{R_i}, & y &= \frac{\tilde{y}}{R_i}, \text{ and} \\ \alpha &= \frac{\lambda_L R_i}{\gamma_{LV}}, & \beta &= \frac{(\rho_L - \rho_V) g R_i^2}{\gamma_{LV}}, \end{aligned} \quad (2.10)$$

then the dimensionless form of Eq. (2.9) becomes

$$x'' [1 + (x')^2]^{-3/2} - x'^{-1} [1 + (x')^2]^{-1/2} = - (\alpha + \beta y). \quad (2.11)$$

Two end-point conditions are needed to close this system. They are

$$x' = \infty \quad \text{at } x = 0, \text{ and} \quad (2.12a)$$

$$x(y) = [1 - (y - 1)^2]^{1/2} \quad \text{at } y = y_2. \quad (2.12b)$$

The first condition simply states that the liquid profile is symmetric about the y-axis. The second condition is the so-called transversality condition [49], which says that at $y = y_2$, the liquid-vapor interface lies on the

inner surface of the SGS. Along this solid-liquid-vapor contact line, the following equation must be satisfied,

$$\left\{ H + \left[- (y - 1) x^{-1} - x' \right] \frac{\partial H}{\partial x'} \right\}_{\substack{x = x_2 \\ y = y_2}} = 0 \quad (2.13)$$

It is easy to verify that this equation is identical to the Young's equation for the contact angle [35] :

$$\cos \theta = \cos (\theta_1 - \theta_2) = \frac{\gamma_{SV} - \gamma_{SL}}{\gamma_{LV}} \quad (2.14)$$

The definitions of the angles θ_1 and θ_2 are given in Fig. 2.1b.

Note that Eq. (2.9) can be reduced to the same equation derived earlier by Bashforth and Adams for a sessile drop [39,50], provided that the origin of the coordinate system is shifted to $x = 0$ and $y = y_1$, and that λ_L is replaced by $2\gamma_{LV}/b_0$, where b_0 is the radius of curvature of the liquid-vapor interface at the origin. Equation (2.9) can further be reduced to Eq. (2.1), namely, the YL equation, since the terms on the left-hand side of Eq. (2.9) actually represent the reciprocal of the two principal radii of curvature and the right-hand side is the hydrostatic pressure difference divided by the interfacial tension.

A solution of Eq. (2.11) subject to a given contact angle will be obtained by using numerical integration methods. Before presenting the results of the numerical calculation, certain points must be clarified:

- (i) The liquid-vapor interfacial tension is assumed constant throughout the calculation regardless of the position and curvature of the interface. This assumption is justified because the radii of curvature in the present case are in the range of 100 μm , which is several orders

of magnitudes larger than the cases studied by Tolman [51], Kirkwood and Buff [52], and Benson and Shuttleworth [53]. These authors found that the surface tension of a very small drop decreases with increasing curvature. Tolman estimated that a 4 per cent drop in the surface tension would occur if the diameter of a drop is in the order of $0.01 \mu\text{m}$;

- (ii) The liquid is assumed to be incompressible, and the effect of the vapor pressure on the liquid-vapor interfacial tension is ignored;
- (iii) The recovery of Young's equation resulting from the application of the transversality condition at the liquid-vapor-solid contact line implies that Young's equation is valid for the spherical system being presently investigated, that is, the solid surface is axially symmetric and the gravitational force is parallel to the axis of symmetry. Also concurrently assumed is that the contact line is ideal. An ideal contact line means that two mathematically defined surfaces meet together. Thus, the line should be infinitesimal and no liquid film exists beyond the line. The validity of Young's equation has long been claimed by Gauss [37], and Gibbs [38], and, more recently, by Johnson [54], Li [42], and Goodrich [55]. For arguments advocating its invalidity, the reader is referred to Bikeman [56], Pethica and Pethica [57], and Jameson and del Cerro [58].

Numerical solutions of Eq. (2.11) satisfying the end-point conditions of Eq. (2.12) are shown in Fig. 2.2. The solutions are for liquid deuterium with different contact angles at a uniform temperature of 24 K. The SGS chosen has an inner radius of $67 \mu\text{m}$ and wall thickness of $3 \mu\text{m}$. The room-temperature D_2 -fill pressure of the SGS is 110 atm, which gives a liquid volume of

Fig. 2.2 A set of solutions for Eq. (2.11) for different contact angles. The SGS used here has an inner radius of $67 \mu\text{m}$, wall thickness of $3 \mu\text{m}$, and is kept at a uniform temperature of 24 K . The liquid enclosed is deuterium and has a volume of $1.4 \times 10^{-7} \text{ cm}^3$.

$1.4 \times 10^{-7} \text{ cm}^3$ at 24 K.

As clearly seen from Fig. 2.2, the location of the contact line between the liquid, solid, and gas rises as the value of the input contact angle (one of the end-point conditions, Eq. (2.12b)) is decreased. This is simply because the smaller the contact angle, the larger is the area wetted by the liquid. For zero contact angle, it is, therefore, believed that the liquid will wet the entire inner surface of the SGS. The resulting configuration is a vapor bubble completely surrounded by a continuous liquid layer.

Although different input values of the contact angle at a given temperature are used to produce the results in Fig. 2.2, one must realize that, in reality, only one particular contact angle corresponds to a given temperature. Good and Ferry measured the contact angle between liquid hydrogen and a few different materials and reported that it was zero for all the materials including glass [43]. However, the temperature of the liquid hydrogen during the measurement was not specified in their report. This makes it difficult to see that the contact angle of liquid hydrogen is zero for all liquid hydrogen temperatures. Nevertheless, a zero contact angle for liquid hydrogen including deuterium and tritium is adopted in the present work. Unfortunately, if zero contact angle is used in the numerical integration of Eq. (2.11), no solution can be found. This conclusion is supported by a mathematical proof which is presented in the next section.

2.3 Proof that No Continuous Liquid Layer Solution is Possible with the Young-Laplace Equation

The coordinate system for the description of a continuous liquid layer inside an SGS is defined in Fig. 2.3. The differential equation governing the

(a)

(b)

Fig. 2.3 (a) Cylindrical coordinate system used for the description of a continuous liquid layer inside an SGS.
 (b) The same system in (a) redefined in dimensionless units.
 At $x = x_m$, $y = y_m$ and $x' = 0$.

profile of the continuous liquid-vapor interface can be obtained by the mathematical procedures outlined in Section 2.2. The only difference is that the energy contributions to the system are now from the gravity and the liquid-vapor interfacial tension. The resulting equation is the same as Eq. (2.11).

The necessary boundary conditions in this case are

$$x' = \infty, \text{ at } x = 0, \quad y = y_1, \text{ and} \quad (2.15a)$$

$$x' = \infty, \text{ at } x = 0, \quad y = y_2. \quad (2.15b)$$

Equation (2.15) is hereby called the bubble boundary conditions because it represents a bubble symmetric about the y -axis. Equation (2.15b) also implies a zero contact angle at $y = y_2$. Integrating Eq. (2.11) once and imposing the above boundary conditions, one has

$$x' = \frac{\pm [x^2 - W^2(x)]^{1/2}}{W(x)}, \quad (2.16)$$

$$\text{where } W(x) = \int_0^x \bar{x} (\alpha + \beta y) d\bar{x}.$$

Since x' has both plus and minus values and the liquid-vapor interface is a continuous curve, there is always a point on the curve where $x' = 0$. This point is (x_m, y_m) , as is shown in Fig. 2.3b. From Eq. (2.16), it is not difficult to see that at this particular point, where $x' = 0$,

$$W(x) \Big|_{x=x_m} = x_m, \quad (2.17a)$$

or

$$\int_0^{x_m} x (\alpha + \beta y_T) dx = \int_0^{x_m} x (\alpha + \beta y_B) dx = x_m, \quad (2.17b)$$

where the subscript T denotes the upper (top) portion of the bubble where $x' < 0$, and the subscript B denotes the lower (bottom) portion of the bubble where $x' > 0$.

Taking the first pair of equality in Eq. (2.17b), and rearranging it, one has

$$\beta \int_0^{x_m} x (y_T - y_B) dx = \beta \frac{V}{2\pi} = 0, \quad (2.18)$$

where V_v is the specific volume of the vapor bubble, namely, the actual volume of the bubble divided by R_i^3 .

For nontrivial cases where $V_v \neq 0$, Eq. (2.18) is satisfied only if the gravitational acceleration g is zero. A conclusion can, therefore, be drawn from this point that, when the gravitational force is present, Eq. (2.11) will possess no solution that can satisfy the bubble boundary conditions, Eq. (2.15), and that can still correspond to a continuous curve throughout the liquid-vapor interface. This completes the proof that the classical Young-Laplace equation cannot be used to describe the profile of the liquid-vapor interface of hydrogen, or the profile of a hydrogen vapor bubble inside a spherical shell under gravity.

The same conclusion can be reached qualitatively from another point of view. When there is gravity, the buoyancy force always pushes the hydrogen vapor bubble upward. Therefore, the bubble will not have a stationary position inside a bulk liquid which has a uniform temperature. When it is near a solid substrate, however, the bubble will encounter an extra pressure resulting from the intermolecular forces between the solid and liquid

molecules. If such pressure is large enough to offset the buoyancy force, the bubble will be brought into a stationary position. This phenomenon is further discussed in the next section.

2.4 Young-Laplace Equation Containing Attractive Molecular Forces

Before deriving a differential equation that can describe the profile of a continuous liquid hydrogen layer inside a spherical shell, let us first examine the long-range attractive forces which exist among atoms and molecules. These forces are generally called van der Waals forces and can be classified into four different groups. They are the Debye induction force between a permanent dipole and a neutral molecule [59], the Keesom orientational force between two freely rotating permanent dipoles [60], the London-dispersion force due to electron fluctuations around nuclei [61], and the Margenau force arising from the dipole-quadrupole interactions [62]. Since hydrogen and glass molecules are non-polar and neutral, only the London-dispersion force will be considered.

Two approaches are widely used to calculate the total London-van der Waals attraction energy between two macroscopic bodies. One is Hamaker's approach [63], in which the additivity of the London-van der Waals forces is assumed. The other is Lifshitz's macroscopic approach [64], in which the attraction energy is directly calculated from the imaginary part of the complex dielectric constants of the media, especially their far-ultraviolet portions [65]. To avoid mathematical difficulties and the scarcity of the data required of the Lifshitz's macroscopic approach, the Hamaker's approach is adopted to estimate the total energy resulting from the London-van der Waals forces between the liquid and solid molecules in the system.

The coordinate system used is shown in Fig. 2.4. The energy due to the London-van der Waals forces between a hydrogen molecule at the point p_m and all the molecules in the solid wall is expressed as

$$E_p(\vec{r}) = N_s \int_{V_s} U(d_m) dv, \quad (2.19)$$

where $U(d_m)$ = London potential = $-B_{hs} d_m^{-6}$,

B_{hs} = London constant between hydrogen and solid molecules,

d_m = distance between a solid molecule and the hydrogen molecule
at the point p_m ,

N = number density of the molecules.

Because of the azimuthal symmetry of the system, Eq. (2.19) can be rewritten as,

$$E_p(\vec{r}) = -2\pi B_{hs} N_s \int_{R_i}^{R_o} R^2 dR \int_{-1}^{+1} [R^2 + r^2 - 2Rr\mu]^{-3} d\mu, \quad (2.20)$$

where $\mu = \cos \theta$.

After straightforward integration, Eq. (2.20) becomes

$$E_p(\vec{r}) = \frac{4}{3}\pi B_{hs} N_s [R_o^3 (R_o^2 - r^2)^{-3} - R_i^3 (R_i^2 - r^2)^{-3}]. \quad (2.21)$$

The total energy in the system due to the London-van der Waals forces between the hydrogen and the solid wall is the summation of E_p over the volumes of the liquid and vapor hydrogen, namely,

$$E_{LVDW} = N_l \int_{V_l} E_p(\vec{r}) dv + N_v \int_{V_v} E_p(\vec{r}) dv. \quad (2.22)$$

Fig. 2.4 Spherical coordinate system used to calculate the London-van der Waals attraction energy between a hydrogen molecule at p_m and the solid molecules in the wall of an SGS.

In terms of the normalized cylindrical coordinate system defined in Fig. 2.3b, Eq. (2.22) becomes

$$E_{\text{LVDW}} = \text{constant} - 2\pi (N_L - N_V) \int_{y_1}^{y_2} dy \int_0^x E_p(r) x dx, \quad (2.23)$$

where $E_p(r) = \frac{4}{3} \pi R_i^{-3} N_s B_{hs} D(x,y)$,

$$D(x,y) = R_e^3 (R_e^2 - r^2)^{-3} - (1 - r^2)^{-3},$$

$$r = [x^2 + (y-1)^2]^{1/2}$$

$$R_e = R_0 R_i^{-1}.$$

Following the procedures of the calculus of variations to minimize the total free energy of the system, namely, the sum of the energies from the London-van der Waals and gravitational forces, and the liquid-vapor interfacial tension, one is able to find the governing equation for the profile of the liquid-vapor interface. The mathematical procedures are quite routine and will, therefore, not be reproduced here. The final equation is

$$x'' [1 + (x')^2]^{-3/2} - x^{-1} [1 + (x')^2]^{-1/2} = - [\alpha + \beta y + C_H D(x,y)], \quad (2.24)$$

where $C_H = \frac{4\pi B_{hs} N_s (N_L - N_V)}{3 \gamma_{LV} R_i^2}$,

and the rest of the symbols have already been defined in Eq. (2.11). Integrating Eq. (2.24) once and applying the bubble boundary conditions, Eq. (2.15), one has

$$\beta V_V + 2\pi C_H \int_0^{x_m} x [D_T(x, y_T) - D_B(x, y_B)] dx = 0. \quad (2.25)$$

From this equation, it is easily seen that as B approaches zero and/or C_H approaches infinity, $[D_T(x, y_T) - D_B(x, y_B)]$ will have to take on a value which is vanishingly small. Or, putting it in another way, at either or both of these two limits, the thickness of the liquid layer is uniform, or the bubble is a complete sphere.

The numerical solutions of Eq. (2.24) that satisfy the bubble boundary conditions, Eq. (2.15), are plotted in Fig. 2.5. The SGS chosen is 67 μm in inner radius, 3 μm in shell thickness, and contains liquid deuterium of volume $1.2 \times 10^{-7} \text{ cm}^3$ at 28 K. The calculations are done for two different values of the London constant B_{hs} : one is $4.277 \times 10^{-59} \text{ erg-cm}^6$ which is estimated for the hydrogen and silicon oxide molecules from the data given by Gregory [66]; the other is thousand times larger than the previous one. As is expected, larger B_{hs} produces a liquid layer more uniform in thickness. This effect is more clearly demonstrated by Fig. 2.6 in which both the top and bottom thicknesses of the liquid layer are plotted as a function of B_{hs} . That a larger B_{hs} is required to support a thicker uniform layer is clearly shown by the figure, which is consistent with the competing nature of the gravity and the London-van der Waals attraction between the liquid and solid molecules.

2.5 The Retardation Effect of the London-van der Waals Forces

Since the London-van der Waals forces are electromagnetic-like forces, they will be subject to retardation, namely, the forces will be reduced at large separation because the finite time required for their propagation causes a phase difference between the electronic oscillations of the interacting molecules. This was first proved by Casimir and Polder [67], and later by

Fig. 2.5 Numerical solutions of Eq. (2.24). The broken line is for $B_{hs} = 4.277 \times 10^{-59} \text{ erg-cm}^6$ and the solid one for $B_{hs} = 4.277 \times 10^{-56} \text{ erg-cm}^6$. The volume of the liquid deuterium is $1.2 \times 10^{-7} \text{ cm}^3$ and the temperature is 28 K. The inner radius of the SGS is $67 \mu\text{m}$ and the wall thickness is $3 \mu\text{m}$.

Fig. 2.6 Equilibrium top and bottom thicknesses of a continuous liquid deuterium layer inside an SGS at a temperature of 28 K. The inner radius of the SGS is 67 μm and the wall thickness is 3 μm . The volume of the liquid deuterium is $1.2 \times 10^{-7} \text{ cm}^3$.

McLachlan [68]. When the distance between two molecules is comparable to the characteristic wavelength of the electronic oscillations of atoms, λ_c (which is generally in the order of 0.1 μm), the retardation effect will be significant [67,69,70].

As suggested by Overbeck [69], the retardation effect can be taken into account by introducing a correction function, $f(p)$, into the London potential, namely,

$$U(d_m) = - B_{hs} d_m^{-6} f(p), \quad (2.26)$$

where $p = 2 \pi d_m \lambda_c^{-1}$,

$$f(p) = \begin{cases} 1.01 - 0.14 p, & \text{for } 0 < p < 3 \\ 2.45 p^{-1} - 2.04 p^{-2}, & \text{for } 3 < p < \infty. \end{cases}$$

Unfortunately, this formula is not applicable to the present case since two separate functions are involved in two different ranges. Schenkel and Kitchener [70] later found an empirical expression for $f(p)$ for the range of $1 < p < \infty$. Their expression is

$$f(p) = 2.45 p^{-1} - 2.17 p^{-2} + 0.59 p^{-3}. \quad (2.27)$$

By replacing the London potential $U(d_m)$ in Eq. (2.19) with Eqs. (2.26) and (2.27) and carrying out the straightforward, yet tedious, integration, one obtains the expression for the retarded London-van der Waals attraction energy between a liquid molecule at the point p_m and the solid molecules in the shell (see Fig. 2.4) as

$$E_{p,ret}(r) = 2 \pi R_i^3 B_{hs} N_s D_{ret}(x,y), \quad (2.28)$$

where

$$D_{\text{ret}}(x,y) = f(R_e, r) - f(l, r),$$

$$f(R, r) = \frac{4}{21} a_3 r^6 (R^2 - r^2)^{-6} + \left(\frac{2 a_3}{5} - \frac{4 a_2 R}{15} \right) r^4 (R^2 - r^2)^{-5} +$$

$$\left(\frac{a_3}{4} - \frac{11 R a_2}{30} + \frac{2 r^2 a_1}{5} \right) r^2 (R^2 - r^2)^{-4} +$$

$$\left(\frac{a_3}{24} - \frac{R a_2}{10} + \frac{2 r^2 a_1}{3} \right) (R^2 - r^2)^{-3} + \frac{a_1}{4} (R^2 - r^2)^{-2},$$

$$a_1 = 2.45 \lambda_c (\pi R_i)^{-1},$$

$$a_2 = 2.17 \lambda_c^2 (\pi R_i)^{-2},$$

$$a_3 = 0.59 \lambda_c^3 (\pi R_i)^{-3},$$

and the rest of the symbols have previously been defined.

Following the similar mathematical procedures outlined in Section 2.4, one obtains a differential equation with the retarded London-van der Waals attraction energy. It is similar to Eq. (2.24) except that $D(x,y)$ is now replaced by $D_{\text{ret}}(x,y)$. The effect of the retardation of London-van der Waals forces between the solid and hydrogen molecules is shown by Fig. 2.7, where the top and bottom thicknesses of a liquid deuterium layer inside an SGS, calculated from Eq. (2.24) with and without the retardation corrections, are compared. For liquid deuterium with a volume of $1.208 \times 10^{-7} \text{ cm}^3$ contained in an SGS of 67- μm inner radius and 3- μm wall thickness, the top layer thickness is seen to be about 0.065 μm at 28 K when the retarded London-van der Waals forces are included in the calculation and about 0.10 μm when the retardation effect is not included. The top liquid layer thicknesses are plotted in Fig. 2.8 as a function of the liquid volume.

Note that the equation with the retarded London-van der Waals attraction energy term, Eq. (2.24), is only valid within the range of $1 < p < \infty$. This range might be extended down to $p \approx 0.5$ with a slightly larger error, as pointed out by Schenkel and Kitchener [70]. In the present case, λ_c for hydrogen is 0.085 μm , which means that $p = 0.5$ corresponds to a distance of

Fig. 2.7 Comparison of the equilibrium top and bottom thicknesses of a continuous liquid deuterium layer calculated with nonretarded (solid lines) and retarded (broken lines) London-van der Waals attraction forces. The parameters used are the same as those specified in Fig. 2.6. λ_C used in the calculation of the retarded-van der Waals forces is $0.1\ \mu\text{m}$.

Fig. 2.8 Equilibrium top liquid layer thickness vs. the enclosed liquid volume. The parameters used are the same as those specified in Figs. 2.6 and 2.7. The solid line is for the nonretarded London-van der Waals' forces and the broken line is for the retarded forces.

0.0068 μm . Therefore, as long as the top liquid layer thickness is larger than 0.0068 μm , the error incurred will be negligible.

Finally, it must be pointed out that the liquid-vapor interfacial tension has been assumed constant throughout the calculations. This assumption is substantiated since the thinnest liquid deuterium layer considered in the present case is still far thicker than the "thick" thin film defined by Mohanty, Davies, and Scriven [48]. They claimed that for a liquid layer thicker than the "thick" thin film, the interfacial tension is independent of thickness.

2.6 Summary

It has been shown that the Young-Laplace equation based on the balance of the gravitational force and interfacial tension alone cannot produce a valid solution for the profile of a continuous liquid hydrogen layer inside a spherical shell. The London-van der Waals forces between the liquid and solid (substrate) molecules must be included in the equation. The retardation effect of the London-van der Waals forces needs to be taken into account for thick liquid layers. It has also shown that, in an isothermal environment, a continuous liquid hydrogen layer with a uniform thickness can only be achieved inside a spherical glass shell (SGS) when the gravity is zero and/or the London constant between the solid and liquid molecules is infinity. Since the London constant is always a finite number, the liquid hydrogen layer inside an SGS, in the presence of gravity, is always thinner at the top and thicker at the bottom. The equilibrium thickness and the profile of the layer are determined by Eq. (2.24).

The pressure inside the liquid layer, which is created by the London-van

der Waals forces between the liquid and solid molecules, exerts an extra force on the vapor bubble in the direction opposite to the gravitational force. This pressure is similar to the disjoining pressure of a thin film [71] and is higher when the distance to the solid-liquid interface is shorter. When the resulting force is equal to the buoyancy force on the vapor bubble, the bubble is in an equilibrium position, or the continuous liquid layer has reached its equilibrium configuration.

The conclusion is that, in an isothermal environment, no uniform liquid hydrogen layer can be formed inside an SGS unless gravity is completely eliminated. Other means for producing a uniform liquid layer in the presence of gravity, such as the temperature gradient method, is pursued in the following chapters.

CHAPTER 3

TEMPERATURE EFFECT ON LIQUID HYDROGEN LAYER CONFIGURATION:

MODEL I - INTERFACIAL TENSION GRADIENT EFFECT

3.1 Introduction

It has been shown in the previous chapter that, due to gravity, the thickness of a continuous liquid hydrogen layer inside an isothermal spherical-glass-shell (SGS) is always thinner at the top and thicker at the bottom. Therefore, to achieve a uniform-thickness liquid layer inside an SGS, new schemes are called upon. In the following three chapters, a method is studied, which is designed to use a vertical temperature gradient to offset the gravitational force and bring about a uniform liquid layer on the inner surface of the SGS. Two mathematical models are developed to study this method.

The model to be developed in this chapter scrutinizes the effect of the temperature-induced interfacial tension gradient on the formation of a uniform liquid hydrogen layer inside an SGS. The hydrogen considered here is restricted to the single-component hydrogen isotopes, namely, H_2 , D_2 , T_2 , and molecular HD, HT, and DT. For simplicity, they are referred to as hydrogen(s) hereafter.

The liquid-vapor interfacial tension of hydrogen is generally a linearly decreasing function of temperature. This temperature dependence is shown in Fig. 3.1. If the temperature at the top of an SGS is kept lower than that at the bottom, the hydrogen liquid-vapor interfacial tension near the top portion of the SGS will be higher than that near the bottom. As a result, the liquid hydrogen near the interface will flow against the gravity and toward the top

Fig. 3.1 Liquid-vapor interfacial tension of normal deuterium (nD_2) and tritium (nT_2). The triple points of nD_2 and nT_2 are, respectively, 18.73 K and 20.62 K [79].

of the SGS if the difference of the interfacial tension is large enough. The fluid flows created by the interfacial tension gradient are usually called Marangoni flows [72,73]. If the amount of liquid moving upward due to the interfacial tension gradient is equal to the amount of liquid flowing downward due to gravity, a uniform liquid layer can be formed. The resulting configuration will be that of a stationary bubble concentric with the spherical shell.

The idea of using a temperature-induced interfacial tension gradient to hold a gas bubble stationary in the midway of a liquid bath has already been demonstrated, both experimentally and theoretically, by Young, Goldstein, and Block [74]. Their theoretical treatment, however, is somewhat oversimplified and is only valid for a bubble inside an unbounded liquid medium. A mathematical model, similar to theirs but more appropriate to the present case, is developed in Section 3.2. The model, henceforth referred to as Model I, describes the motion of a hydrogen vapor bubble at the moment concentric with an SGS under the influences of gravity, the temperature-induced liquid-vapor interfacial tension gradient, and the finite size of the liquid container.

The important result of Model I is an explicit expression for the temperature gradient on the outer surface of an SGS which is required to hold a concentric hydrogen vapor bubble stationary (or to form a uniform liquid hydrogen layer) inside an SGS. Some numerical examples for Model I are shown in Section 3.3. In order to show that the prediction of temperature gradient for Model I is unique, the motion of a bubble not at the center of an SGS is also studied. The details of the proof are presented in Section 3.4.

3.2 Model I : Effect of Interfacial Tension Gradient

3.2.1 Velocity Fields

Assuming that hydrogen liquid and vapor obey the continuum and Newtonian fluid mechanics and that the fluid properties are constant throughout the system, one finds that the fluid flows in the liquid and vapor phases can be described by the Navier-Stokes equation [75],

$$\rho \frac{D \vec{v}}{Dt} = - \vec{\nabla} p + \mu \vec{\nabla}^2 \vec{v} + \rho \vec{g}, \quad (3.1)$$

where ρ , v , p , μ , g , and t , respectively, denote the density, velocity, pressure, viscosity, gravitational acceleration, and time; and the notations " $\sim$ " and " $\rightarrow$ " indicate that the variables and the operators underneath are, respectively, dimensional quantities and vectors.

Since only the steady-state solutions are of interest, the time derivative in Eq. (3.1) can be dropped. Rewritten in dimensionless form, Eq. (3.1) becomes

$$\vec{v} \cdot \vec{\nabla} \vec{v} = - \vec{\nabla} p + \frac{1}{Re} \nabla^2 \vec{v} + \frac{1}{Fr} \frac{\vec{g}}{g}, \quad (3.2)$$

where $v = \tilde{v} / V_{ref}$,

$p = \tilde{p} / \rho V_{ref}^2 + \text{constant}$,

$Re = \rho V_{ref} R_B / \mu = \text{Reynolds number}$,

$Fr = V_{ref}^2 / g R_B = \text{Froude number}$,

$R_B = \text{radius of the hydrogen bubble}$,

$V_{ref} = \text{reference velocity, which usually has the value of the average flow velocity in the system,}$

$\vec{\nabla} = R_B \vec{\tilde{\nabla}}$,

$$\nabla^2 = R_B^2 \nabla^2.$$

Because of the nonlinear term on the left-hand side of Eq. (3.2), there is no analytical solution to this equation. However, if the flow rate in the system is very low, then the inertial effect of the fluid flow, which is represented by the nonlinear term in Eq. (3.2), will be negligible. Equation (3.2) can then be linearized as

$$0 = -\nabla p + \frac{1}{Re} \nabla^2 \vec{v} + \frac{1}{Fr} \frac{\vec{g}}{g}. \quad (3.3)$$

This is the so-called low Reynolds number approximation. This approximation is valid when the Reynolds number of the system is less than unity. In two-dimensional cases, an analytical solution to Eq. (3.3) is available in terms of the Stokes stream function.

If the temperature field outside an SGS is azimuthally symmetric and the temperature gradient is parallel to the gravitational force, then the fluid flows in the vapor and liquid phases can be assumed azimuthally symmetric. Furthermore, the hydrogen bubble is assumed to be spherical and to reach its steady-state migration velocity, U , at the moment it is concentric with the SGS. This is sometimes called quasi-steady state and is valid when the acceleration of the bubble is very low. However, a true steady-state can be reached if the bubble velocity is zero. Based on these assumptions, a spherical coordinate system is chosen such that its origin is at the center of the bubble and moves with the bubble, as is shown in Fig. 3.2. The dimensionless Stokes stream function, ψ , can be defined as

$$v_r = -\frac{1}{r^2 \sin \theta} \frac{\partial \psi}{\partial \theta}, \quad (3.4a)$$

Fig. 3.2 Spherical coordinate system used in describing the velocity and temperature fields inside an SGS. The origin of the coordinate system is at the center of the vapor bubble which moves upward at a constant velocity of $\vec{U}$.

$$v_{\theta} = \frac{1}{r \sin \theta} \frac{\partial \psi}{\partial r}, \quad (3.4b)$$

where v_r = r-component of the fluid flows,

v_{θ} = θ -component of the fluid flows,

$$v_r = \tilde{v}_r / V_{\text{ref}},$$

$$v_{\theta} = \tilde{v}_{\theta} / V_{\text{ref}},$$

$$\psi = \tilde{\psi} / R_B^2 / V_{\text{ref}}, \text{ and}$$

$$r = \tilde{r} / R_B.$$

In terms of ψ , Eq. (3.3) is rewritten as

$$E^2 (E^2 \psi) = 0, \quad (3.5)$$

$$\text{where } E^2 = \frac{\partial^2}{\partial r^2} + \frac{\sin \theta}{r^2} \frac{\partial}{\partial \theta} \left(\frac{1}{\sin \theta} \frac{\partial}{\partial \theta} \right).$$

Note that with the Stokes stream function defined in Eq. (3.4), the equation of continuity for a fluid with constant mass density, i.e., $\vec{\nabla} \cdot \vec{v} = 0$, is automatically satisfied. Equation (3.5) is the governing equation to be solved for the velocity fields both in the liquid and vapor phases. According to Happel and Brenner [76], the stream functions, which satisfy Eq. (3.5), are expressed, respectively, for the liquid and vapor phases as follows:

$$\psi_L(r, \theta) = \sum_{n=2}^{\infty} (A_n r^n + B_n r^{-n+1} + C_n r^{n+2} + D_n r^{-n+3}) \int_n(\cos \theta), \quad (3.6a)$$

$$\psi_V(r, \theta) = \sum_{n=2}^{\infty} (E_n r^n + F_n r^{-n+1} + G_n r^{n+2} + H_n r^{-n+3}) \int_n(\cos \theta), \quad (3.6b)$$

where $\int_n(\cos \theta)$ = Gegenbauer function of order n and degree $(-1/2)$,

$$= \frac{P_{n-2}(\cos \theta) - P_n(\cos \theta)}{2n - 1}, \text{ and}$$

$P_n(\cos \theta)$ = Legendre function of order n .

The eight constants in Eq. (3.6) are determined by the appropriate boundary conditions. The nonslip condition at the liquid-solid interface gives rise to

$$v_{r,l} = -U \cos \theta, \text{ at } r = b^{-1} \quad (3.7a)$$

$$v_{\theta,l} = U \sin \theta, \text{ at } r = b^{-1} \quad (3.7b)$$

where $U = \dot{\gamma} / V_{ref}$, and

$$b = R_B / R_i.$$

On the surface of the bubble, the radial component of the fluid flows will vanish since no mass transfer between liquid and vapor phases is assumed, specifically,

$$v_{r,l} = v_{r,v} = 0, \text{ at } r = 1. \quad (3.8)$$

Also, the continuity of the tangential velocity fields requires

$$v_{\theta,l} = v_{\theta,v}, \text{ at } r = 1. \quad (3.9)$$

The shear stress created by the fluid flows and the gradient of the liquid-vapor interfacial tension must also be balanced, i.e.,

$$\sigma e_{r\theta,l} - e_{r\theta,v} = - \frac{1}{\mu_v V_{ref}} \frac{\partial \gamma_{lv}}{\partial \theta}, \text{ at } r = 1, \quad (3.10)$$

where $e_{r\theta}$ = rates of strain in the θ -direction,

$$= \frac{1}{r} \frac{\partial v_r}{\partial \theta} + \frac{\partial v_\theta}{\partial r} - \frac{v_\theta}{r},$$

$\sigma = \mu_v / \mu_v$, and

γ_{lv} = liquid-vapor interfacial tension.

The remaining two boundary conditions are the boundedness requirements on the r - and θ -components of the velocities inside the vapor bubble.

Upon application of these boundary conditions to Eq. (3.6), the eight constants are determined as follows:

$$A_n = \frac{1}{4(2n-1)} I_n K_n b^{2n-3} [(2n-1) - (2n+1) b^2 + 2 b^{2n+1}] + \delta_{2,n} K_n U [1 + \sigma + \frac{5}{4} b^3 - \frac{3}{4} (3-2\sigma) b^5], \quad (3.11a)$$

$$B_n = \frac{1}{4(2n-1)} I_n K_n [2 - (2n-1) b^{2n-3} + (2n-3) b^{2n-1}] + \frac{1}{2} \delta_{2,n} K_n U [1 - (1-\sigma) b^3], \quad (3.11b)$$

$$C_n = \frac{-1}{4(2n-1)} I_n K_n [(2n-3) - (2n-1) b^2 + 2 b^{2n-1}] b^{2n-1} - \frac{1}{4} \delta_{2,n} K_n U [(3+2\sigma) b^3 - 3 b^5], \quad (3.11c)$$

$$D_n = \frac{-1}{4(2n-1)} I_n K_n [2 - (2n+1) b^{2n-1} + (2n-1) b^{2n+1}] - \frac{1}{2} \delta_{2,n} K_n U [3 + 2\sigma - 3(1-\sigma) b^5], \quad (3.11d)$$

$$E_n = \frac{1}{4(2n-1)} I_n K_n [2 - \frac{1}{2} (2n-1)^2 b^{2n-3} + (2n+1)(2n-3) b^{2n-1} - \frac{1}{2} (2n-1)^2 b^{2n+1} + 2 b^{4n-2}] - \frac{1}{4} \delta_{2,n} K_n U \sigma [2 - 5b^3 + 3b^5], \quad (3.11e)$$

$$F_n = 0, \quad (3.11f)$$

$$G_n = -E_n, \quad (3.11g)$$

$$H_n = 0, \quad (3.11h)$$

where $\delta_{2,n} = \begin{cases} 1 & \text{for } n = 2 \\ 0 & \text{for } n > 2 \end{cases}$,

$$\frac{1}{K_n} = 1 + \sigma - \frac{(2n-1)}{4} (2n-1+2\sigma) b^{2n-3} + \frac{1}{2} (2n+1)(2n-3) b^{2n-1} -$$

$$\frac{(2n-1)}{4} (2n-1-2\sigma) b^{2n+1} + (1-\sigma) b^{4n-2} ,$$

$$I_n = \frac{-n(n-1)(2n-1)}{2 \mu_v V_{ref}} \int_0^\pi \frac{\partial y_{lv}(\theta)}{\partial \theta} \int_n (\cos \theta) d\theta .$$

From Haberman and Sayre [77], the drag force on the hydrogen bubble is given in terms of D_n ,

$$F_d = \left[-4 \pi \mu_l V_{ref} R_B D_n \right]_{n=2} . \quad (3.12)$$

Note that only the P_1 -mode of the velocity fields in the liquid phase is responsible for the drag force. The steady-state migration velocity, U , of the bubble is determined by the macroscopic force balance on the bubble, namely, the balance among the drag, weight, and buoyance forces. In dimensionless form, it is

$$U = \frac{2}{3\mu_l} \left[\frac{R_B^2 (\rho_l - \rho_v) g}{V_{ref} K_2} - \frac{1}{4} \mu_l I_2 (2 - 5b^3 + 3b^5) [3 + 2\sigma - 3(1-\sigma)b^5]^{-1} \right] . \quad (3.13)$$

Equation (3.13) can be reduced to an expression similar to the one derived earlier by Young, Goldstein, and Block [74] when b is set equal to zero. The function I_2 in Eq. (2.13) represents the effect of the interfacial tension gradient on the velocity of the bubble. If $I_2 = 0$, Eq. (3.13) is reduced to the formula derived by Haberman and Sayre [77]. In the present case, the interfacial tension gradient is generated by a nonuniform temperature field, so that I_2 can be evaluated once the temperature field is known.

3.2.2 Temperature Fields

Assuming constant fluid properties, the general steady-state energy conservation equation containing temperature is [75]

$$\rho C_p (\vec{V} \cdot \vec{\nabla} T) = k \nabla^2 T + \mu \Phi, \quad (3.14)$$

where C_p , k , T , and Φ are the specific heat capacity, thermal conductivity, temperature, and viscous dissipation function of the fluid, respectively.

By defining the dimensionless temperature and viscous dissipation function, respectively, as

$$T = \frac{T - T_0}{T_{ref}}, \quad (3.15a)$$

$$\Phi = \frac{\mu}{k} R_B^2 V_{ref}^{-2}, \quad (3.15b)$$

where T_0 = the temperature at the outer equator of the SGS,

T_{ref} = reference temperature; which is usually chosen as the maximum temperature difference in the system,

R_B and V_{ref} are as defined in Eq. (3.2) ,

one is able to rewrite Eq. (3.14) in dimensionless form:

$$Re Pr \vec{V} \cdot \vec{\nabla} T = \nabla^2 T + Br \Phi, \quad (3.16)$$

where $Pr = \frac{C_p \mu}{k}$ = Prandtl number,

$Br = \frac{\mu V_{ref}^2}{k T_{ref}}$ = Brinkman number,

ν and Re are as defined in Eq. (3.2).

Since the flow rates in the liquid and vapor phases are assumed to be in the low Reynolds number region, the Brinkman number for hydrogen is in the order of 1×10^{-8} ; hence, the heat generated by the viscous dissipation can be ignored. Moreover, the Prandtl numbers of the liquid and vapor hydrogen are all in the order of unity; the convection term on the left-hand side of Eq. (3.16) can also be dropped. Consequently, Eq. (3.16) becomes

$$\nabla^2 T = 0. \quad (3.17)$$

This is exactly the Laplacian equation, which will be used to describe the temperature fields both in the liquid and vapor phases. The heat conduction in the solid wall region of the SGS can also be described by the same equation since the thermal properties of the material are assumed constant. For azimuthally symmetric temperature fields, the general solutions of Eq. (3.17) (which are written in terms of the spherical coordinate defined in Fig. 3.2) in the regions of the wall, the liquid, and the vapor are, respectively,

$$T_w(r, \theta) = \sum_{n=2}^{\infty} \left[\hat{A}_n r^{n-1} + \hat{B}_n r^{-n} \right] P_{n-1}(\cos \theta), \quad (3.18a)$$

$$T_l(r, \theta) = \sum_{n=2}^{\infty} \left[\hat{C}_n r^{n-1} + \hat{D}_n r^{-n} \right] P_{n-1}(\cos \theta), \quad (3.18b)$$

$$T_v(r, \theta) = \sum_{n=2}^{\infty} \left[\hat{E}_n r^{n-1} + \hat{F}_n r^{-n} \right] P_{n-1}(\cos \theta), \quad (3.18c)$$

where $P_{n-1}(\cos \theta)$ = Legendre function of order $(n - 1)$.

The boundary conditions needed to find the six constants in Eq. (3.18) are

(i) the boundedness requirement of T_v at $r = 0$, i.e.,

$$rT_v = 0, \text{ at } r = 0, \quad (3.19)$$

(ii) the continuities of the temperature fields and heat fluxes at the liquid-vapor and liquid-solid interfaces, i.e., at $r = 1$,

$$T_\ell = T_v, \text{ and} \quad (3.20a)$$

$$\varepsilon_1 \frac{\partial T_\ell}{\partial r} = \frac{\partial T_v}{\partial r}, \text{ where } \varepsilon_1 = k_\ell / k_v, \quad (3.20b)$$

$$\text{and at } r = b^{-1} = R_i/R_B$$

$$T_w = T_\ell, \text{ and} \quad (3.21a)$$

$$\varepsilon_2 \frac{\partial T_w}{\partial r} = \frac{\partial T_\ell}{\partial r}, \text{ where } \varepsilon_2 = k_w / k_\ell, \quad (3.21b)$$

(iii) the temperature field on the surface of the SGS specified as

$$T_w = T_{\text{ext}}^{(0)}, \text{ at } r = d^{-1} = R_o/R_B, \quad (3.22)$$

where R_o is the outer radius of the SGS.

Upon imposing these boundary conditions on Eq. (3.18), one finds the six constants as

$$\hat{A}_n = T_n N_n d^{n-1} (N_n - d^{2n-1})^{-1}, \quad (3.23a)$$

$$\hat{B}_n = -\hat{A}_n N_n^{-1}, \quad (3.23b)$$

$$\hat{C}_n = -(n-1+n\varepsilon_1)(N_n - b^{2n-1}) \hat{B}_n [n-1+n\varepsilon_1 - (n-1)(1-\varepsilon_1) b^{2n-1}]^{-1}, \quad (3.23c)$$

$$\hat{D}_n = -(n-1)(1-\varepsilon_1) \hat{C}_n (n-1+n\varepsilon_1)^{-1}, \quad (3.23d)$$

$$\hat{E}_n = (2n-1) \epsilon_1 \hat{C}_n (n-1 + n\epsilon_1)^{-1}, \quad (3.23e)$$

$$\hat{F}_n = 0, \quad (3.23f)$$

$$\text{where } N_n = \frac{(n-1+n\epsilon_1)(n-1+n\epsilon_2) + n(n-1)(1-\epsilon_1)(1-\epsilon_2) b^{2n-1}}{(n-1)(1-\epsilon_1)[n+(n-1)\epsilon_2] + (n-1)(1-\epsilon_2)(n-1+n\epsilon_1) b^{-2n+1}},$$

$$T_n = \frac{1}{2} (2n-1) \int_{-1}^{+1} T_{\text{ext}}(\theta) P_{n-1}(\cos \theta) d(\cos \theta).$$

By substituting Eq. (3.23) into Eq. (3.18) and setting $r = 1$, one can evaluate, in terms of the system parameters, the temperature at the bubble surface resulting from the external temperature field, i.e.,

$$T_V(r=1, \theta) = \sum_{n=2}^{\infty} \frac{(2n-1) \epsilon_1}{n-1+n\epsilon_1-(n-1)(1-\epsilon_1)b^{2n-1}} \frac{N_n b^{2n-1}}{N_n d^{2n-1}} d^{n-1} T_n P_{n-1}(\cos \theta). \quad (3.24)$$

3.2.3 Condition for a Uniform Liquid Hydrogen Layer

Let the nonuniform external temperature field, $T_{\text{ext}}(\theta)$, on the outer surface of an SGS be expanded in a series of Legendre polynomials, namely,

$$T_{\text{ext}}(\theta) = T_0 + R_0 \sum_{n=1}^{\infty} t_n P_n(\cos \theta). \quad (3.25)$$

With this, the value of I_2 required in Eq. (3.13) for the calculation of the migration velocity of the hydrogen bubble in an SGS can now be evaluated. By an iteration method, a specific value of $T_{\text{ext}}(\theta)$ can be found such that the migration velocity of the bubble is zero. This is precisely the condition for a uniform liquid hydrogen layer inside an SGS, since the bubble is assumed to be spherical and initially concentric with the SGS. Explicitly, the

condition is

$$I_2 = \frac{4 R_B^2 (\rho_L - \rho_V) g}{\mu_L V_{ref} K_2 (2 - 5b^3 + 3b^5)} \quad (3.26)$$

However, an analytical expression for the temperature gradient required for a uniform liquid hydrogen layer is possible because the hydrogen liquid-vapor interfacial tension is very close to a linear function of temperature (see Fig. 3.1), particularly when the temperature variations are small. The resulting expression is

$$(t_1)_S = -\frac{2}{3} \frac{N_2 - d^3}{N_2 - b^3} \frac{1 + 2\epsilon_1 - (1 - \epsilon_1) b^3}{\epsilon_1 \sigma K_2 (2 - 5b^3 + 3b^5)} \frac{R_B (\rho_L - \rho_V) g}{\gamma_1} \quad (3.27)$$

where $\gamma_1 = \left| \frac{\partial \gamma_{LV}}{\partial T} \right| = \text{constant}$,

and the subscript S denotes the temperature gradient required for a stationary bubble.

Note that only the linear temperature gradient (or the P_1 -mode of the external temperature field) gives rise to a uniform liquid layer. Such a linear temperature gradient is a must to ensure that the bubble be spherical. Remember that the pressure normal to the bubble surface has not been considered as one of the boundary conditions in the derivation of the velocity fields in Section 3.2. To be presented is a proof that as long as the liquid-vapor interfacial tension gradient along the bubble surface remains in the P_1 -mode, or in other words, the temperature gradient is a constant (since the hydrogen liquid-vapor interfacial tension is a linear function of temperature), the pressure normal to the bubble surface is automatically balanced.

The general expressions for the hydrodynamic pressure in the vapor and liquid phases are, respectively,

$$\tilde{p}_v = \text{constant} - \mu_v V_{\text{ref}} R_B^{-1} \sum_{n=2}^{\infty} \frac{2(2n+1)}{n-1} G_n r^{n-1} P_{n-1}(\cos \theta), \quad (3.28a)$$

$$\tilde{p}_l = \text{constant} - \mu_l V_{\text{ref}} R_B^{-1} \sum_{n=2}^{\infty} \left[\frac{2(2n+1)}{n-1} C_n r^{n-1} + \frac{2(2n-3)}{n} D_n r^{-n} \right] P_{n-1}(\cos \theta). \quad (3.28b)$$

The general expressions for the corresponding normal stresses are, respectively,

$$\tilde{\tau}_{rr,v} = \text{constant} + 2 \mu_v V_{\text{ref}} R_B^{-1} \sum_{n=2}^{\infty} [(n-2) E_n r^{n-3} + n G_n r^{n-1}] P_{n-1}(\cos \theta), \quad (3.29a)$$

$$\tilde{\tau}_{rr,l} = \text{constant} + 2 \mu_l V_{\text{ref}} R_B^{-1} \sum_{n=2}^{\infty} [(n-2) A_n r^{n-3} - (n+1) B_n r^{-n-2} + n C_n r^{n-1} - (n-1) D_n r^{-n}] P_{n-1}(\cos \theta), \quad (3.29b)$$

The pressure difference on the surface of the hydrogen bubble is then

$$\Delta \tilde{p}_{lv} = \text{constant} + [\tilde{p}_l + \tilde{\tau}_{rr,l} - \tilde{p}_v - \tilde{\tau}_{rr,v}]_{r=R_B} - (\rho_l - \rho_v) g R_B \cos \theta. \quad (3.30)$$

Substituting Eqs. (3.28) and (3.29) into Eq. (3.30), one has

$$\Delta \tilde{p}_{lv} = \text{constant} - I_2 \mu_v V_{\text{ref}} R_B^{-1} \cos \theta + \frac{1}{2} \mu_v V_{\text{ref}} R_B^{-1} \sum_{n=3}^{\infty} \frac{I_n (\sigma K_n M_n - 6n)}{n(n-1)(2n-1)} P_{n-1}(\cos \theta), \quad (3.31)$$

$$\text{where } M_n = 6 + n(4n^2 - 1)(n-2)b^{2n-3} - (2n+1)(2n-3)(2n^2 - 2n-1)b^{2n-1} + (2n-1)(2n-3)(n^2 - 1)b^{2n+1}.$$

The pressure balance equation on the bubble surface is the well-known Laplace-Young equation, Eq. (2.1). For a spherical bubble of radius R_B , it is

$$-\frac{2}{R_B} \gamma_{lv}(\theta) = \Delta p_{lv} . \quad (3.32)$$

Let the interfacial tension be expanded into a series of Legendre polynomials, namely,

$$\gamma_{lv}(\theta) = \gamma_0 + \sum_{n=2}^{\infty} x_{n-1} P_{n-1}(\cos \theta) . \quad (3.33)$$

Substituting Eqs. (3.31) and (3.33) into Eq. (3.32), one has

$$-\frac{2}{R_B} \gamma_0 - \frac{2}{R_B} \sum_{n=3}^{\infty} x_{n-1} P_{n-1}(\cos \theta) = \text{constant} + \frac{2}{R_B} \sum_{n=3}^{\infty} \frac{x_{n-1} P_{n-1}(\cos \theta)}{4(2n-1)} (\sigma K_n M_n - 6n) . \quad (3.34)$$

From Eq. (3.34), it is clear that, for $n = 2$, namely, when the interfacial tension is in the P_1 -mode, the variation of the capillary pressure is completely counterbalanced by the pressure variation created by the fluid flows inside and outside the bubble. For $n > 2$, however, the capillary pressure variation cannot be compensated for because the following equality will never be satisfied:

$$\frac{\sigma K_n M_n - 6n}{4(2n-1)} = -1 , \quad \text{for } n=3,4,5\dots \quad (3.35)$$

3.3 Numerical Examples for Model I

Figure 3.3 is a family of curves corresponding to the temperature gradients on the outer surface of an SGS, $(t_1)_S$, which are required for the formation of a uniform liquid layer on the inner surface of the SGS. $(t_1)_S$ is calculated from Eq. (3.27) and checked by means of the iteration method described in Section 3.2. The difference in the values of $(t_1)_S$ calculated from the two methods is small. In all cases studied, it is less than 0.01%. The SGS chosen here (and in all figures that follow) has an outer radius of $100\text{ }\mu\text{m}$ and a wall thickness of $5\text{ }\mu\text{m}$, and is filled with pure deuterium. The data on the thermal conductivity of glass at cryogenic temperatures are taken from a compendium edited by V.J. Johnson [78]. The properties of deuterium are from a report written by P.C. Souers [79]. From Fig. 3.3, one can see that the thicker the liquid layer, the larger is the magnitude of the temperature gradient required for a uniform layer. Figure 3.4 shows the changes in the values of $(t_1)_S$ when the aspect ratio of the liquid shell is varied, where the aspect ratio is defined as the inner radius of the SGS divided by the liquid layer thickness.

The effect of the liquid layer thickness on the migration velocity of a deuterium bubble at the moment concentric with an isothermal SGS is shown in Fig. 3.5. The isothermal migration velocity of the bubble, U , is calculated from Eq. (3.13) by setting $I_2 = 0$. Knowing this migration velocity, one can estimate the sagging time of a liquid deuterium layer inside an SGS. The liquid sagging time, $\tau_{10\%}$, is defined as the time needed for a bubble initially concentric with the SGS to move a distance equal to ten per cent of the uniform liquid layer thickness. $\tau_{10\%}$ are plotted in Fig. 3.6 for

Fig. 3.3 Temperature gradient on the outer surface of an SGS, $(t_1)_s$, required for the formation of a uniform liquid deuterium layer. $(t_1)_s$ is calculated from Model I. The outer radius of the SGS used is $100\ \mu\text{m}$ and the wall thickness is $5\ \mu\text{m}$.

Fig. 3.4 Scaled temperature gradient, $(t_1)_S / R_B$, on the outer surface of an SGS vs. the aspect ratio of the liquid shell. The temperature gradient is required to form a uniform liquid layer on the inner surface of the SGS. The aspect ratio is defined as the inner radius of the SGS divided by the liquid layer thickness. The parameter d is equal to R_B / R_0 .

Fig. 3.5 Scaled migration velocity, $\tilde{U} / R_B^2$, of a bubble inside of and concentric with an isothermal SGS for various aspect ratios of the liquid shell. The aspect ratio is defined as the inner radius of the SGS divided by the liquid layer thickness. The liquid enclosed is normal deuterium and is at 24 K.

Fig. 3.6 Product of the sagging time of a uniform liquid layer, $\tau_{10\%}$, inside an isothermal SGS and the radius of the bubble, R_B , for various aspect ratios of the liquid shell. The aspect ratio is defined as the inner radius of the SGS divided by the liquid layer thickness. The liquid enclosed is normal deuterium and is at 24 K.

different aspect ratios of the liquid shell. As expected, the thicker the layer the shorter is the liquid sagging time for a given SGS.

When the liquid deuterium layer is brought to be uniform inside an SGS by an external temperature gradient, the layer is in a dynamic equilibrium state. Circular fluid flows exist both in the liquid and vapor phases. Typical stream lines in the liquid and vapor phases inside an SGS are drawn in Fig. 3.7a. In this figure, the thickness and average temperature of the liquid layer are, respectively, $6 \mu\text{m}$ and 24 K . The flow velocity profile in the vapor and liquid phases at the equator of the SGS is shown in Fig. 3.7b. The maximum flow velocities both in the vapor and liquid phases are 0.03 cm/sec , and the corresponding maximum Reynolds numbers are, respectively, 0.04 in the vapor phase and 0.15 in the liquid phase. The Reynolds numbers are much less than unity; therefore, the assumption about the low Reynolds number approximation is justified. The isotherms in the SGS are plotted in Fig. 3.8.

3.4 Motion of a Nonconcentric Bubble

In the foregoing discussion of Model I, an expression for the external temperature gradient required for a stationary hydrogen bubble concentric with an SGS was derived. Is this prediction unique? Will the bubble remain motionless at points other than the center one under the same temperature gradient calculated from Eq. (3.27)? To answer these questions, let us now look at the motion of a bubble not concentric with an SGS.

The coordinate system suitable for describing such a configuration is a bipolar sphere coordinate system which is shown in Fig. 3.9. The lower plane ($\xi < 0$) is the image of the upper plane ($\xi > 0$), and the distance between the centers of the bubble and SGS, i.e., $d = d_j - d_B$, is determined by the characteristic length of the system, $\bar{\lambda}$, which is equal to $(R_B |\sinh b|)$.

Fig. 3.7 (a) Typical stream lines in the vapor and liquid phases for Model I. $(t_1)_S = -0.251$ K/cm. The liquid layer is $6 \mu\text{m}$ thick and at an average temperature of 24 K. Stream line 1: $\psi_L = 0$ and $\psi_V = 0$; 2: $\psi_L = -2.0 \times 10^{-5}$; 3: $\psi_L = -2.0 \times 10^{-4}$; 4: $\psi_V = 0.0015$; and 5: $\psi_V = 0.003$. (b) Flow velocity profile at the equator for the case shown in (a).

Fig. 3.8 Isotherms for the case specified in Fig. 3.7. The temperature difference between two adjacent isotherms is 0.000627 K.

Fig. 3.9 Bipolar sphere coordinate system used in describing the motion of a nonconcentric bubble inside an SGS. In this coordinate system,

$$z \equiv \bar{x} \sinh \xi (\cosh \xi - \cos \eta)^{-1},$$

$$\omega \equiv \bar{x} \sin \eta (\cosh \xi - \cos \eta)^{-1},$$

$$\bar{x} \equiv R_B |\sinh \bar{b}| \equiv R_S |\sinh \bar{a}|.$$

Stimson and Jeffery [80] have adopted this coordinate system successfully in their derivation of an analytical expression for the fluid flows around two adjacent solid spheres.

With the same assumptions described in Section 3.2, the dimensionless stream function equation in the present coordinate system is

$$E^2 (E^2 \psi) = 0, \quad (3.36)$$

$$\text{where } E^2 = (\cosh \xi - \cos \eta) \left\{ \frac{\partial}{\partial \xi} [(\cosh \xi - \cos \eta) \frac{\partial}{\partial \xi}] + \right. \\ \left. (\sin^2 \eta) \frac{\partial}{\partial \cos \eta} [(\cosh \xi - \cos \eta) \frac{\partial}{\partial \cos \eta}] \right\};$$

and the dimensionless stream function is defined as

$$v_\xi = \frac{(\cosh \xi - \cos \eta)^2}{\sin \eta} \frac{\partial \psi}{\partial \eta}, \quad v_\eta = \frac{(\cosh \xi - \cos \eta)^2}{\sin \eta} \frac{\partial \psi}{\partial \xi}.$$

The general solution of Eq. (3.36) is [80]

$$\psi(\xi, \eta) = (\cosh \xi - \cos \eta)^{-3/2} \sum_{n=0}^{\infty} X_n(\xi) \int_{n+1}(\cos \eta), \quad (3.37)$$

$$\text{where } X_n(\xi) = \bar{A}_n e^{-(n-1/2)\xi} + \bar{B}_n e^{+(n-1/2)\xi} + \bar{C}_n e^{-(n+3/2)\xi} + \bar{D}_n e^{+(n+3/2)\xi},$$

and $\int_{n+1}(\cos \theta)$ = Gegenbauer function of order $(n+1)$ and degree $(-1/2)$.

Applying the same boundary conditions stated in Section 3.2, the eight constants for the stream functions in the vapor and liquid phases are obtained. They are listed in Appendix A. Note that the velocity fields are obtained by assuming that the bubble reaches its steady state when it is at Δd

away from the center of the SGS. They are sometimes called quasi-steady-state solutions. Once the velocity fields are solved, the drag force on the bubble can be found by integrating the hydrodynamic pressure along the bubble surface. After a long and tedious mathematical maneuver, one finds

$$F_d = -4(\rho)^{1/2} \pi \mu_L V_{ref} \bar{\epsilon} f_1, \quad (3.38)$$

$$\begin{aligned} \text{where } f_1 &= \sum_{n=0}^{\infty} (\bar{B}_n + \bar{D}_n), \text{ for } \xi > 0, \\ &= \sum_{n=0}^{\infty} (\bar{A}_n + \bar{C}_n), \text{ for } \xi < 0. \end{aligned}$$

Note that f_1 has one unique value for a given Δd . In other words, f_1 is a function of the distance between the centers of the bubble and SGS. Carrying out the overall force balance on the hydrogen bubble, one finds the migration velocity of the bubble as

$$U = - \frac{2 R_B^3 (\rho_L - \rho_V) g}{3 \mu_L V_{ref} \bar{\epsilon} \Omega} - (2)^{-1/2} \Omega^{-1} \sum_{n=1}^{\infty} \frac{1}{2n+1} \bar{I}_{n+1} \bar{K}_n \bar{Z}_n, \quad (3.39)$$

$$\text{where } \bar{I}_{n+1} = - \frac{n(n+1)(2n+1)}{2 \mu_V V_{ref}} \int_0^{\pi} \frac{\partial \gamma_{LV}}{\partial \eta} (\cosh \xi - \cos \eta)^{-1/2} \int_{n+1}^{\pi} (\cos \eta) d\eta,$$

$$\begin{aligned} \frac{1}{K_n} &= \sigma [2 \sinh(2n+1)(\bar{a}-\bar{b}) - (2n+1) \sinh 2(\bar{a}-\bar{b})] - \\ &\frac{1}{2} [(2n-1)(2n+3) + 4 \cosh(2n+1)(\bar{a}-\bar{b}) - (2n+1)^2 \cosh 2(\bar{a}-\bar{b})], \end{aligned}$$

$$\begin{aligned} \bar{Z}_n &= [(2n-1) - (2n+1) e^{+2\bar{a}} + (2n+1) e^{+2\bar{b}} - (2n+1) e^{-2(\bar{a}-\bar{b})} + \\ &2 e^{-(2n+1)(\bar{a}-\bar{b})} - 2 e^{-(2n+1)(\bar{a}-\bar{b})+2\bar{b}}] e^{-(n+3/2)\bar{b}}, \end{aligned}$$

$$\Omega = \sum_{n=1}^{\infty} \frac{n(n+1)}{(2n-1)(2n+3)} e^{-(2n+1)\bar{a}} \bar{K}_n \bar{S}_n,$$

$$\begin{aligned} \bar{S}_n = & \sigma \{ (2n-1) e^{-2\bar{b}} [(2n+1) e^{+(2n-1)(\bar{a}-\bar{b})} - 2] - \\ & 2(2n-1)(2n+3) e^{+(2n+1)(\bar{a}-\bar{b})} + (2n+3)[(2n+1)e^{+(2n+3)(\bar{a}-\bar{b})} + \\ & 2] e^{+2\bar{b}} \} + (2n+1) [2n-1+(2n+3) e^{+4\bar{b}}] e^{-2\bar{b}} + \\ & 2(2n-1)(2n+3) [e^{+(2n+1)(\bar{a}-\bar{b})} - 1] - \\ & (2n+1) [2n-1+(2n+3) e^{+4\bar{a}}] e^{+(2n+1)(\bar{a}-\bar{b})} - 2\bar{a} . \end{aligned}$$

Equation (3.39) can be reduced to an expression previously derived by Meyyappan, Wilcox, and Subramanian [81] for a bubble moving in a direction normal to a plane if $g = 0$, $a = 0$, and $\mu_v \rightarrow 0$. The ratio of the migration velocity of a deuterium bubble at a distance, Δd , away from the center of the SGS to that at the center of the isothermal SGS is plotted in Fig. 3.10. The amount of liquid deuterium enclosed is equivalent to having a uniform liquid layer of $6 \mu\text{m}$ thickness. The ratios are very weakly dependent on the temperature. When $\Delta d \rightarrow 0$, namely, when the bubble becomes concentric with the SGS, the velocity takes on the same value as that calculated from Eq. (3.13).

To find the temperature effect on the velocity of a hydrogen bubble not concentric with an SGS, the temperature fields in the vapor and liquid phases must be solved. The heat equation is again reduced to Laplacian form as before and is written in the present coordinate system as

$$\begin{aligned} \nabla^2 T = 0 , \quad (3.40) \\ \text{where } \nabla^2 = (\cosh \xi - \cos \eta)^3 \left[\frac{\partial}{\partial \xi} \left(\frac{1}{\cosh \xi - \cos \eta} \frac{\partial}{\partial \xi} \right) + \right. \\ \left. \frac{\partial}{\partial \cos \eta} \left(\frac{\sin^2 \eta}{\cosh \xi - \cos \eta} \frac{\partial}{\partial \cos \eta} \right) \right] . \end{aligned}$$

The general solution of Eq. (3.40) is [82]

Fig. 3.10 Ratio of the migration velocity of a deuterium bubble at a distance, Δd , away from the center of an SGS to that at the center in an isothermal environment. This ratio is very weakly dependent on temperature. The amount of liquid deuterium enclosed is equivalent to having a uniform liquid layer of $6 \mu m$ thickness.

$$T(\xi, \eta) = (\cosh \xi - \cos \eta)^{1/2} \sum_{n=0}^{\infty} (\hat{A}_n e^{(n+1/2)\xi} + \hat{B}_n e^{-(n+1/2)\xi}) P_n(\cos \theta). \quad (3.41)$$

By applying the continuity conditions of the temperature fields and heat fluxes at the surface of the hydrogen bubble, it is possible to find the four constants for the temperature fields in the vapor and liquid phases. However, due to the intrinsic characteristics of the bipolar sphere coordinates, the condition of the continuity of heat fluxes at the surface of the bubble is in nonlinear form. To avoid this problem and to find simple analytical solutions to the temperature fields, further restrictions must be imposed. That is, the temperature fields found here are restricted to the cases where the bubble is not far away from the center of an SGS. Mathematically, this means that the temperature fields are valid only when $\cosh(\bar{b}) \gg 1$. Details of this discussion are presented in Appendix A.

After some mathematical maneuvers, the temperature field on the surface of a nonconcentric bubble is obtained in terms of the known temperature field, T_{gS} , at the liquid-solid interface,

$$T_v \Big|_{\xi=\bar{b}} = \epsilon_1 \cosh(\bar{b}) [\cosh(\bar{b}) - \cos \eta]^{1/2} \sum_{n=0}^{\infty} (2n+1) \bar{T}_n \bar{M}_n P_n(\cos \eta), \quad \text{for } \cosh(\bar{b}) \gg 1, \quad (3.42)$$

$$\text{where } \bar{T}_n = \frac{1}{2} (2n+1) \int_{-1}^{+1} T_{gS}(\eta) [\cosh(\bar{a}) - \cos \eta]^{-1/2} P_n(\cos \eta) d(\cos \eta),$$

$$\frac{1}{\bar{M}_n} = (1-\epsilon_1) \sinh(\bar{b}) \sinh(n+1/2)(\bar{a}-\bar{b}) + (2n+1) \cosh(\bar{b}) [\epsilon_1 \cosh(n+1/2)(\bar{a}-\bar{b}) - \sinh(n+1/2)(\bar{a}-\bar{b})].$$

The temperature field in the region of the uniform thickness glass wall

of an SGS will not be calculated here because it is difficult to do so analytically in the present coordinate system. Let the temperature field at the liquid-solid interface be specified as

$$T_{LS}(n) = T_0 + \bar{t}_{LS,1} R_1 \cos n, \quad (3.43)$$

where the plus sign is for $\xi > 0$ and the minus sign for $\xi < 0$; then the value of $\bar{t}_{LS,1}$ that gives rise to a stationary bubble at points along the vertical center line (but not at the center) of an SGS can be found by combining Eqs. (3.39) and (3.42) and setting $U = 0$. It is

$$(\bar{t}_{LS,1})_S = -\frac{4}{3} R_B^3 (\rho_L - \rho_V) g \sinh(\bar{a}) [\sigma \varepsilon_1 \gamma_1 \bar{t} \cosh(\bar{b}) \sum_{n=1}^{\infty} n(n+1) [L_n R_n Z_n]^{-1},$$

for $\cosh(\bar{b}) \gg 1$, (3.44)

$$\text{where } \bar{L}_n = \int_{-1}^{+1} \bar{W}(\cos \eta) \left[\frac{P_{n-1}(\cos \eta) - P_{n+1}(\cos \eta)}{2(2n+1)(\cosh(\bar{b}) - \cos \eta)} - P_n(\cos \eta) \right] d(\cos \eta),$$

$$\bar{W}(\cos \eta) = \sum_{m=0}^{\infty} (2m+1) e^{-(m-1/2)\bar{a}} \left(\frac{m}{2m-1} + \frac{m+1}{2m+3} e^{-2\bar{a}} \right) P_m P_m(\cos \eta).$$

The ratio of the temperature gradient at the solid-liquid interface required for a stationary bubble not at the center of an SGS to that at the center is plotted in Fig. 3.11. The parameters of the SGS used are the same as before. The amount of liquid deuterium enclosed in the SGS is equivalent to having a uniform liquid layer of $6 \mu\text{m}$ thickness. The value of $(t_{LS,1})_S$ is calculated from Eq. (3.27) by setting $d = b$. When Δd approaches zero, the bubble becomes concentric with the SGS; the value of $(\bar{t}_{LS,1})_S$ will be the same as $(t_{LS,1})_S$. This verifies that the external

Fig. 3.11 Ratio of the temperature gradient at the solid-liquid interface required for a stationary bubble at a distance, Δd , away from the center of an SGS to that at the center. The amount of liquid deuterium enclosed is equivalent to having a uniform liquid layer of $6 \mu\text{m}$ thickness. This ratio is very weakly dependent on the average temperature of the SGS.

temperature gradient predicted by Model I (i.e., Eq. (3.27)) required for having a stationary hydrogen bubble at the center of an SGS is unique.

3.5 Summary

In this chapter, a mathematical model, called Model I, has been developed to study the motion of a spherical bubble inside a nonisothermal SGS. The fluid flows inside and outside the bubble at the center of the SGS are determined by solving the low Reynolds number approximation of the fluid equation, and the corresponding temperature fields are derived for low Prandtl number fluids, such as liquid and vapor hydrogen. The velocity and temperature fields are valid when the bubble reaches its steady-state velocity at the moment that it is concentric with the SGS. The migration velocity of the bubble is found related to the sizes of the bubble itself and the SGS, as well as the external temperature gradient. A particular vertical temperature gradient can be applied to hold the bubble stationary at the center of the SGS and hence make the liquid layer uniform. The model further shows that the temperature gradient must be linear to ensure the uniformity of the liquid layer.

The motion of a bubble, not concentric with the SGS, has also been studied. An analytical solution for the fluid flows inside and outside the bubble not at the center (but along the vertical center line) of the SGS has been derived. However, only an approximate solution for the temperature fields is found, due to the mathematical difficulty. The final result of this calculation shows that the isothermal migration velocity of the bubble is maximum when it is at the center of the SGS and the magnitude of the temperature gradient to hold the bubble stationary at the center is also maximum.

Model I has the capability to model the motion of a bubble in a liquid medium under the influences of gravity, temperature-induced liquid-vapor interfacial tension gradient, and the finite size of the liquid container. Therefore, it has broader applications than the early models [74,77]. However, it may not be the correct model for the motion of a hydrogen vapor bubble in a nonisothermal SGS because liquid hydrogen is quite volatile, and the mass transfer between the liquid and vapor phases in the form of condensation and evaporation at the bubble surface may have significant impact on the bubble motion. A new model to account for this mass transfer effect is developed in the next chapter.

CHAPTER 4

TEMPERATURE EFFECT ON LIQUID HYDROGEN LAYER CONFIGURATION:

MODEL II - COMBINED EFFECT OF INTERFACIAL TENSION GRADIENT AND MASS TRANSFER

4.1 Introduction

In the formation of Model I, which has been discussed in the previous chapter, the temperature-induced interfacial tension gradient is the single force responsible for maintaining a uniform-thickness liquid hydrogen layer inside a spherical glass shell (SGS). The mass transfer between the vapor and liquid phases is ignored entirely in Model I. However, this seems unjustified for pure hydrogen since hydrogen is quite volatile and its saturated vapor pressure is a sensitive function of temperature. A small amount of temperature gradient along the hydrogen liquid-vapor interface will likely create a significant vapor pressure difference.

For instance, in the example shown in the previous chapter, the maximum temperature difference in the vapor phase is 0.0049 K (see Fig. 3.8), and the corresponding saturated vapor pressure difference will be 1554.7 dyne/cm^2 (about 1.17 torr). The saturated vapor pressures for pure normal deuterium and tritium are plotted in Fig. 4.1 for a reference. In the above example, the pressure difference raised by the hydrodynamic pressure from the fluid flow in the vapor phase and the hydrostatic pressure from gravity is only about 0.04 dyne/cm^2 . Therefore, one can see that the pressure near the warm spot of the liquid-vapor interface in the vapor phase is far below the saturated vapor pressure, and vice versa for the cool spot. Because of the requirements of the thermodynamic equilibrium at the liquid-vapor interface, liquid will continuously vaporize at the warm spot and vapor will condense at

Fig. 4.1 Saturated vapor pressure for normal liquid deuterium, nD_2 , and tritium, nT_2 . The triple points of nD_2 and nT_2 are, respectively, 18.73 and 20.62 K.

the cool spot. Mass transfer between liquid and vapor phases hence occurs.

If the bottom of an SGS is warmer than the top, the mass transfer induced by the evaporation and condensation will be parallel to the liquid flow created by the temperature-induced interfacial tension gradient, to move the excess liquid from the bottom of the SGS to the top, resulting in a uniform liquid layer. The combined effect of the mass transfer and the interfacial tension gradient on the formation of a uniform pure liquid hydrogen layer inside an SGS is studied in this chapter. A mathematical model, henceforth called Model II, is developed following the procedures similar to those used in the previous chapter.

4.2 Model II - Combined Effect of Mass Transfer and Interfacial Tension Gradient

The assumptions and the coordinate system used in the description of this model are the same as those in Chapter 3 for Model I. The general expressions of the stream functions for the liquid and vapor phases are also the same as those in Eq. (3.6). However, some boundary conditions need to be changed to take into account the effect of mass transfer. The boundary conditions stated in Eq. (3.8) are now changed to

$$v_{r,v} = \zeta v_{r,l}, \text{ and} \quad (4.1a)$$

$$p_v = p_{sat}, \quad (4.1b)$$

where $\zeta = \rho_l / \rho_v$,

p_{sat} = saturated vapor pressure,

and all other symbols are defined in Chapter 3.

Equation (4.1a) is the requirement of the mass conservation between liquid and vapor phases. Equation (4.1b) says that the pressure at the liquid-vapor interface on the vapor side must equal the saturated vapor pressure at the

same temperature.

The eight unknown constants for the stream functions can be rederived by imposing the boundary conditions stated in Eqs. (3.7), (3.9), (3.10), (4.1), and the boundedness requirements of the stream function at $r = 0$. They are

$$A_n = \frac{1}{2} L_n I_n b^{2n-3} \{ (2n-1)(n-1+n\zeta) - (2n+1)(n-3+n\zeta)b^2 - 2(n+2-n\zeta)b^{2n+1} \} + \\ L_n G_n b^{2n-3} \{ (2n-1)[(2n-1)(n-1)+3n\zeta+2(n^2-1)\sigma] - \\ (2n+1)[3(n-3+n\zeta)+2(n-2)(n-3+n\zeta)]b^2 - 2[(n+2)(2n-1)-3n\zeta-2(n^2-1)\sigma]b^{2n+1} \} - \\ \delta_{2,n}^3 \sigma L_n U [2-4\zeta+5b^3+3(1-2\zeta)b^5], \quad (4.2a)$$

$$B_n = \frac{1}{2} L_n I_n [2(n-3+n\zeta)+n(2n-1)(1-\zeta)b^{2n-3} - (2n-3)(n+2-n\zeta)b^{2n-1}] + \\ L_n G_n \{ 2[3(n-3+n\zeta)+2(n-2)(n-3+n\zeta)] + n(2n-1)[2n-1-3\zeta-2(n-2)\sigma]b^{2n-3} - \\ (2n-3)[(n+2)(2n-1)-3n\zeta-2(n^2-1)\sigma]b^{2n-1} \} - \\ \delta_{2,n}^6 \sigma L_n U (1-\zeta)b^3, \quad (4.2b)$$

$$C_n = \frac{1}{2} L_n I_n b^{2n-1} [-(2n-3)(n-1+n\zeta) + (2n-1)(n-3+n\zeta)b^2 + 2n(1-\zeta)b^{2n-1}] + \\ L_n G_n b^{2n-1} \{ -(2n-3)[(2n-1)(n-1)+3n\zeta+2(n^2-1)\sigma] + \\ (2n-1)[3(n-3+n\zeta)+2(n-2)(n-3+n\zeta)]b^2 + 2n[2n-1-3\zeta-2(n-2)\sigma]b^{2n-1} \} + \\ \delta_{2,n}^6 \sigma L_n U (1-\zeta)b^3, \quad (4.2c)$$

$$D_n = \frac{1}{2} L_n I_n [-2(n-1+n\zeta) - n(2n+1)(1-\zeta)b^{2n-1} + (2n-1)(n+2-n\zeta)b^{2n+1}] + \\ L_n G_n \{ -2[(n-1)(2n-1)+3n\zeta+2(n^2-1)\sigma] - n(2n+1)[2n-1-3\zeta-2(n-2)\sigma]b^{2n-1} + \\ (2n-1)[(2n-1)(n+2)-3n\zeta-2(n^2-1)\sigma]b^{2n+1} \} + \\ \delta_{2,n}^6 \sigma L_n U (1-\zeta)(2+3b^5), \quad (4.2d)$$

$$E_n = \frac{1}{2} L_n I_n \zeta [-4 + (2n-1)^2 b^{2n-3} - 2(2n-3)(2n+1)b^{2n-1} + (2n-1)^2 b^{2n+1} - 4b^{4n-2}] + \\ L_n G_n \{ 6(n-1)\sigma - 4(n^2-1)\zeta - 2(n+2)(2n-1)\sigma\zeta + (2n-1)^2[(n^2-1)\zeta - n(n-1)\sigma + \\ (n+2)\sigma\zeta]b^{2n-3} + (2n+1)(2n-3)[(2n^2-2n-1)\sigma - 2(n^2-1)\zeta]b^{2n-1} + \\ (2n-1)^2[(n^2-1)\zeta - (n+2)\sigma\zeta - (n^2-n-3)\sigma]b^{2n+1} - 2[3n\sigma + 2(n^2-1)\zeta - (2n-1)(n+2)\sigma\zeta]b^{4n-2} \} \\ + \delta_{2,n}^3 \sigma L_n U (2-5b^3+3b^5), \quad (4.2e)$$

$$F_n = 0, \quad (4.2f)$$

$$G_n = - \frac{(n-1)(2n-1)}{4(2n+1)} \frac{\rho_v R_B V_{ref}}{\mu_v} \int_{-1}^{+1} p_{sat} p_{n-1}(\cos \theta) d(\cos \theta) -$$

$$\delta_{2,n} \frac{R_B^2 \rho_v g}{10 \mu_v V_{ref}}, \quad (4.2g)$$

$$H_n = 0, \quad (4.2h)$$

$$\text{where } \frac{1}{L_n} = 2 [2n(n-2)\zeta - 3(n-1)\sigma + n(2n-1)\sigma\zeta] + n(2n-1)^2 [(n-1)\sigma - (n-2)\zeta -$$

$$\sigma\zeta] b^{2n-3} + (2n+1)(2n-3)[2n(n-2)\zeta - (2n^2 - 2n - 1)\sigma] b^{2n-1} +$$

$$(2n-1)^2 [(\zeta^2 - n - 3)\sigma - n(n-2)\zeta + n\sigma\zeta] b^{2n+1} +$$

$$2n[3\sigma + 2(n-2)\zeta - (2n-1)\sigma\zeta] b^{4n-2},$$

and the rest of the symbols are defined in Eq. (3.11).

The migration velocity of a bubble passing the center of an SGS is (in dimensionless form)

$$U = \frac{-1}{18} \frac{R_B^2 (\rho_l - \rho_v) g}{\mu_l V_{ref} L_2 e_1} + \frac{1}{6} I_2 \frac{e_2}{e_1} + G_2 \frac{e_2 + \sigma (2+3b^5)}{e_1} \quad (4.3)$$

$$\text{where } e_1 = \sigma (1-\zeta)(2+3b^5),$$

$$e_2 = 1 + 2\zeta + 5(1-\zeta)b^3 - 3(2-\zeta)b^5.$$

Note that only $n = 2$, or P_1 -mode, of the velocity fields has the effect on the bubble velocity. The functions I_2 and G_2 in Eq. (4.3) are to be determined by the temperature fields in the vapor and liquid phases.

The assumptions used in determining the temperature fields are also the same as those in Chapter 3. Equations (3.18) are the general expressions for the temperature fields in the regions of the wall, the liquid, and the vapor inside an SGS. In the present case, i.e., Model II, because of the presence of condensation and evaporation, the extra heat associated with the processes

must be taken into account. The condition for the continuity of the heat fluxes at the liquid-vapor interface, i.e., Eq. (3.20b), is

$$\epsilon_1 \frac{\partial T}{\partial r} = \frac{\partial T}{\partial r} = H_h, \quad (4.4)$$

$$\text{where } H_h = \frac{R_B \rho_v h_{lv} V_{ref}}{T_{ref} k_v} [v_{r,v}]_{r=1},$$

h_{lv} = specific heat of vaporization,

$[v_{r,v}]_{r=1}$ = r-component of vapor velocity at the liquid-vapor interface.

The six unknown constants in Eq. (3.18) for the temperature fields can now be determined by using Eqs. (3.19), (3.20a), (3.21), (3.22), and (4.4).

They are

$$\hat{A}_n = [T_n N_n d^{n-1} - (2n-1) \hat{H}_n d^{2n-1} J_n^{-1}] (N_n - d^{2n-1})^{-1}, \quad (4.5a)$$

$$\hat{B}_n = [-T_n d^{n-1} + (2n-1) \hat{H}_n J_n^{-1}] (N_n - d^{2n-1})^{-1}, \quad (4.5b)$$

$$\hat{C}_n = \frac{1}{(n-1)(1-\epsilon_1)} [\hat{H}_n - (n-1+n\epsilon_1) \hat{D}_n], \quad (4.5c)$$

$$\hat{D}_n = -[(2n-1)(n-1)(1-\epsilon_1)\epsilon_2 T_n d^{n-1} - \hat{H}_n [(n-1+n\epsilon_2) - (n-1)(1-\epsilon_2)(\frac{d}{b})^{2n-1}]] J_n^{-1} (N_n - d^{2n-1})^{-1}, \quad (4.5d)$$

$$\hat{E}_n = \frac{1}{(n-1)(1-\epsilon_1)} [\hat{H}_n - (2n-1) \epsilon_1 \hat{D}_n], \quad (4.5e)$$

$$\hat{F}_n = 0, \quad (4.5f)$$

where

$$J_n = (n-1)(1-\epsilon_1)[n+(n-1)\epsilon_2] + (n-1)(n-1+n\epsilon_1)(1-\epsilon_2)b^{-2n+1},$$

$$\hat{H}_n = \frac{1}{2} (2n-1) \int_{-1}^{+1} H_h P_{n-1}(\cos \theta) d(\cos \theta),$$

and the rest of the symbols are defined in Eq. (3.23).

By substituting $\hat{E}_n$ and $\hat{F}_n$ into Eq. (3.18c) and setting $r = 1$, one obtains the temperature field at the liquid-vapor interface as

$$T_v(r=1, \theta) = \sum_{n=2}^{\infty} \{ (2n-1)^2 \epsilon_1 \epsilon_2 T_n d^{n-1} + \hat{H}_n [n-1 + n\epsilon_2 + n(1-\epsilon_2)b^{2n-1} - (n+(n-1)\epsilon_2)d^{2n-1} - (n-1)(1-\epsilon_2)(\frac{d}{b})^{2n-1}] \} j_n^{-1} (N_n d^{2n-1})^{-1} \cdot P_{n-1}(\cos \theta) . \quad (4.6)$$

The temperature field now depends not only on the external temperature, $T_{ext}(\theta)$, but also on the rates of condensation and evaporation represented by $\hat{H}_n$.

The condition for the stationary bubble concentric with the SGS is then derived from Eq. (4.3) by setting $U = 0$. It is

$$I_2 = \frac{1}{3} \frac{R_B^2 (\rho_L - \rho_V) g}{\mu_L \gamma_{ref} L_2 e_2} - 6 G_2 e_3 , \quad (4.7)$$

$$\text{where } e_3 = 1 + \frac{\sigma (2 + 3b^5)}{e_2} .$$

The definite integrations in I_2 and G_2 can be evaluated numerically using a trial value for $T_{ext}(\theta)$. And, by iteration, a particular $T_{ext}(\theta)$ that satisfies Eq. (4.7) can be found. However, an explicit expression for $T_{ext}(\theta)$ that gives rise to a stationary bubble is possible, if the temperature gradient in the vapor phase is very small, and hence $p_v(T)$ and $p_{sat}(T)$ can be approximated as a linear function of temperature.

The temperature gradient in the vapor phase, in fact, will be minimized by the processes of condensation and evaporation. Heat will be released when condensation occurs at the cool spot of the liquid-vapor interface, thus, raising the temperature at that spot. The opposite will happen at the warm spot as a result of evaporation. The net result is the reduction in the temperature gradient and hence in the rates of condensation and evaporation. A decrease in the rates of condensation and evaporation will subsequently raise

the temperature gradient which will then cause more hydrogen to evaporate and condense. This cycle will continue until a dynamic equilibrium (i.e., steady state) is reached. Because hydrogen is volatile and has a relatively large heat of vaporization, the temperature gradient in the vapor phase is usually less than 10^{-4} K/cm.

Letting $p_1 = \partial p_{\text{sat}}(T) / \partial T = \text{constant}$, one finds that the temperature gradient on the outer surface of the SGS required to maintain a stationary bubble concentric with the SGS, or in other words, to form a uniform liquid layer is

$$(t_1)_S = \frac{-1}{9} \frac{g}{\epsilon_1 \epsilon_2} J_2 (N_2 - d^3) \left\{ \frac{1}{6} R_B (\rho_L - \rho_V) \frac{e_4}{L_2 \sigma e_2} + \rho_V \left[\frac{\gamma_1}{p_1} (e_4 - e_5) + \frac{3}{10} R_B e_3 e_5 \right] \right\} (\gamma_1 + \frac{3}{10} e_3 R_B p_1)^{-1}, \quad (4.8)$$

$$\text{where } e_4 = 1 + \frac{3}{10} \frac{R_B \rho_V h_{LV}}{\mu_V k_V} \frac{R_B p_1}{J_2 (N_2 - d^3)} e_6 e_9 L_2,$$

$$e_5 = 1 + \frac{R_B \rho_V h_{LV}}{\mu_V k_V} \frac{e_6 e_7}{J_2 (N_2 - d^3)} \epsilon \gamma_1 L_2,$$

$$e_6 = 1 + 2\epsilon_2 - (1 - \epsilon_2) \left(\frac{d}{b} \right)^3 + 2(1 - \epsilon_2) b^3 - (2 + \epsilon_2) d^3,$$

$$e_7 = 4 - 9b + 10b^3 - 9b^5 + 4b^6,$$

$$e_8 = 2 - 6b + 5b^3 + 3b^5 - 4b^6,$$

$$e_9 = \frac{10}{3} \epsilon \gamma_1 e_7 (R_B p_1)^{-1} + \epsilon e_7 + \sigma e_8 + (3L_2)^{-1},$$

and γ_1 is as defined in Eq. (3.27).

Carrying out the normal pressure balance at the bubble surface, one has,

$$\begin{aligned} \Delta \hat{p}_{LV} = \text{constant} - I_2 V_{\text{ref}} \mu_V R_B^{-1} \cos \theta + \mu_V V_{\text{ref}} R_B^{-1} \sum_{n=3}^{\infty} \frac{1}{n(n-1)} L_n I_n S_{I,n} \\ + P_{n-1}(\cos \theta) + 2 \mu_V V_{\text{ref}} R_B^{-1} \sum_{n=3}^{\infty} \frac{1}{n(n-1)} L_n G_n S_{G,n} P_{n-1}(\cos \theta), \quad (4.9) \end{aligned}$$

$$\text{where } S_{1,n} = 2(n-1) [2n(n-2)\zeta + (2n^2-n+3)\sigma - 3n\sigma\zeta] + \\
n(n-1)(2n-1)^2 [-2\sigma - (n-2)\zeta + n\sigma\zeta] b^{2n-3} + \\
n(2n+1)(2n-3) [6(n-1)\sigma - (2n^2-2n-1)\sigma\zeta + 2(n-1)(n-2)\zeta] b^{2n-1} - \\
(2n-1)^2 [2(2n^2-2n-3)\sigma - (n^2-n-3)n\sigma\zeta + n(n-1)(n-2)\zeta] b^{2n+1} + \\
2n [-(2n^2-3n+4)\sigma + 3n\sigma\zeta + 2(n-1)(n-2)\zeta] b^{4n-2} ,$$

$$S_{G,n} = 2(n-1)(4n^3-4n^2-2n-3)\sigma + 2n(4n^3-8n^2+2n+5)\sigma\zeta + \\
4(n^2-1)(2n^2-4n+3)\sigma^2 + 4(n-2)(2n^2+1)\zeta + \\
n(2n-1)^2(n-2)[-(n-1)\sigma+(n+2)\sigma\zeta-(2n^2+1)\zeta+2(n^2-1)\sigma^2]b^{2n-3} + \\
n(2n+1)(2n-3)[3(2n^2-4n+3)\sigma-3(2n^2-2n-1)\sigma\zeta+2(n-2)(2n^2+1)\zeta- \\
4(n^2-1)(n-2)\sigma^2]b^{2n-1} + (2n-1)^2 [-(5n^3-9n^2+n+6)\sigma - \\
n(n-2)(2n^2+1)\zeta + 2(n^4-2n^3-n^2+2n+3)\sigma^2+n(5n^2-6n-5)\sigma\zeta] b^{2n+1} + \\
4n(n-2)(2n^2+1)(\zeta-\sigma)(1-\sigma)b^{4n-2} .$$

From Eq. (4.9), and following the similar arguments stated in Chapter 3, one can see that the bubble will remain spherical as long as the interfacial tension and the saturated vapor pressure are linear functions of temperature, and the temperature gradient on the bubble surface is in P_1 -mode. In general, however, the interfacial tension and the saturated vapor pressure are not linear functions of temperature, so that the condition ensuring that the bubble be spherical is difficult to satisfy unless the temperature gradient on the bubble surface is negligible. Some numerical examples for the present model will be presented in the next section.

4.3 Numerical Examples of Model II

The temperature gradient on the outer surface of an SGS, $(t_1)_S$,

which is required for the formation of a uniform liquid deuterium layer inside the SGS, is plotted in Fig. 4.2. The outer radius and wall thickness of the SGS are, respectively, 100 μm and 5 μm . The temperature gradient, $(t_1)_S$, is calculated directly from Eq. (4.8). It is also checked by the iteration method mentioned in Section 4.2. In all cases studied, the results from these two methods have discrepancies about or less than 0.1%. This shows that the temperature gradient at the deuterium liquid-vapor interface is indeed small and Eq. (4.8) produces little error in predicting $(t_1)_S$. The variation of $(t_1)_S$ with respect to the aspect ratio of the liquid shell is shown in Fig. 4.3. Note that the magnitude of $(t_1)_S$ calculated from Model II is much larger than that from Model I for a given layer thickness, due to the fact that a large amount of heat associated with the evaporation and condensation processes must be compensated for.

In Model II, if the temperature at the top of the SGS is lower than at the bottom, then the excess liquid at the bottom of the SGS (resulting from the gravity-induced liquid sagging) is brought up to the top by the mass transfer between the liquid and vapor phases and by the liquid-vapor interfacial tension gradient. This mass movement is clearly shown in Fig. 4.4a, in which the stream lines in the liquid and vapor phases inside an SGS are drawn. The liquid layer in the SGS has a uniform thickness of 6 μm and is at an average temperature of 24 K. In this case, the dominant mass movement is due to the evaporation and condensation, and the circular fluid flows caused by the interfacial tension gradient are hardly seen here. Figure 4.4b is the profile of the flow velocities in the vapor and liquid phases at the equator of the SGS. The maximum flow velocities in the vapor and liquid phases are, respectively, 1.08 and 0.079 cm/sec; and the corresponding maximum Reynolds

Fig.4.2 Temperature gradient on the outer surface of an SGS, $(t_1)_s$, required to form a uniform liquid deuterium layer. $(t_1)_s$ is calculated from Model II. The outer radius of the SGS is $100\text{ }\mu\text{m}$ and the wall thickness is $5\text{ }\mu\text{m}$.

Fig. 4.3 Scaled temperature gradient, $(t_1)_S/R_B^2$, on the outer surface of an SGS vs. the aspect ratio of a liquid shell. The temperature gradient is required to form a uniform liquid deuterium layer on the interior surface of the SGS. The aspect ratio is defined as the inner radius of the SGS divided by the liquid layer thickness. The parameter d is equal to R_B/R_0 .

Fig. 4.4 (a) Typical stream lines in the vapor and liquid phases for Model II. $(t_1)_S = -25.71$ K/cm. The liquid deuterium layer is $6 \mu\text{m}$ thick and at an average temperature of 24 K. Stream line 1: $\psi_2 = 0$ and $\psi_v = 0$; 2: $\psi_2 = -3.0 \times 10^{-4}$ and $\psi_v = -0.02$; 3: $\psi_2 = -2.0 \times 10^{-3}$ and $\psi_v = -0.13$. (b) Flow velocity profile at the equator for the case shown in (a).

numbers are, respectively, 1.43 in the vapor phase and 0.4 in the liquid phase. Note that the fluid flows are in the low Reynolds number range. Figure 4.5 shows the isotherms of the SGS considered in Fig. 4.4. The temperature gradient at the liquid-vapor interface, which is not seen in the figure, is about -1.14×10^{-5} K/cm. The maximum mass flux across the liquid-vapor interface in this case is 1.22×10^{-3} g/sec/cm².

4.4 Summary

A mathematical model capable of explaining the behavior of liquid hydrogen inside an SGS having a vertical temperature gradient has been developed in this chapter. Two temperature-induced effects, namely, the mass transfer between the liquid and vapor phases and the fluid flows resulting from the liquid-vapor interfacial tension gradient, are included in the model calculation. The net result is an analytical expression (Eq. 4.8) for the temperature gradient on the outside surface of an SGS which is required to create and maintain a uniform liquid hydrogen layer inside the SGS. This model is referred to as Model II throughout the present work.

Model II is believed to be more realistic in modeling the behavior of pure hydrogen inside an SGS, compared with Model I in which the mass transfer effect is completely ignored. This view is supported by the experimental data which are shown in Chapter 7. As previously pointed out, these two models are only valid for pure (or single-component) hydrogen enclosed inside an SGS. In the case where the enclosed liquid is a mixture of hydrogen isotopes, the two models will not be appropriate due to the effect of component separation. The model calculation including this component separation effect is presented in detail in the next chapter.

Fig. 4.5 Isotherms for the case specified in Fig. 4.4.
The temperature difference between two adjacent
isotherms is 0.0643 K.

CHAPTER 5
TEMPERATURE EFFECT ON
LIQUID LAYER CONFIGURATION OF MIXTURES OF HYDROGEN ISOTOPES

5.1 Introduction

As pointed out in Chapter 1, the simplest high-gain targets designed to be used in the ICF experiments consist of a uniform layer of liquid or solid deuterium-tritium (D-T) on the inner surface of the spherical glass shells (SGS). It is, therefore, essential to develop a reliable method for fabricating such targets. For a target, or more precisely, an SGS filled with pure hydrogen, a uniform liquid layer can be formed and maintained by a vertical temperature gradient. This has been theoretically shown to be feasible in the previous chapter. Experimental observations have also confirmed it [17,18,28]. However, whether the same method is equally applicable to a D-T target is still a question mark. In some early target cooling experiments, the liquid D-T layer inside a target was found always thicker at the warmer spot of the target [13,17,18]. No detailed explanation of the phenomenon has been given so far except the simple qualitative one presented by Henderson, Simms, and Jacobs [34]. A theoretical model will be developed in this chapter to study the behavior of the liquid mixtures inside a nonisothermal SGS.

It has been shown in the previous chapter that the two major forces that are responsible for bringing about a uniform liquid layer inside an SGS are the liquid-vapor interfacial tension gradient and the saturated vapor pressure difference. For a single-component, two-phase system, the liquid-vapor interfacial tension and the saturated vapor pressure are functions of

temperature only. However, according to the phase rule [83], all the thermodynamic quantities including these two will no longer be a single function of temperature for a multicomponent, multiphase system. For instance, for a binary system having two phases, the quantities will be a function of temperature and the mixture ratio of the two components in the system.

Because deuterium and tritium will react with each other to form molecular DT, the D-T mixture is actually a ternary mixture. However, for simplicity and for the reason that almost all the transport properties of molecular DT are deduced from those of other hydrogen isotopes [79], the D-T mixture is regarded as a binary mixture throughout this chapter. Let us now imagine that an SGS filled with D-T is brought down to the liquid hydrogen temperatures and kept cooler at the top and warmer at the bottom. Deuterium and tritium will evaporate at the bottom and condense at the top. The rates of evaporation and condensation for deuterium, however, will be larger than those for tritium due to the fact that deuterium is more volatile than tritium, or in other words, liquid deuterium has higher saturated vapor pressure than liquid tritium does at the same temperature. The comparison between these two saturated vapor pressures is shown in Fig. 4.1. As a result, the liquid layer near the bottom of the SGS is enriched with tritium and that near the top with deuterium.

The change in the mixture ratio in the liquid layer will have a drastic effect on the total liquid-vapor interfacial tension. From Fig. 3.1, one sees that the interfacial tensions of pure deuterium and pure tritium are both decreasing functions of temperature. However, the interfacial tension of tritium is, in general, higher than that of deuterium. Because of the phase rule, the overall liquid-vapor interfacial tension is now a function of not

only the temperature but also the mixture ratio. Hence, if the enrichment of tritium near the bottom of the SGS is high enough, the total interfacial tension near the bottom will become higher even when the bottom of the SGS is kept warmer than the top. If this is so, the total interfacial tension gradient will point downward and work together with gravity. The configuration of the liquid layer, therefore, depends on the competition between the mass flow rate brought by the evaporation and condensation and that by the interfacial tension gradient plus the gravity. If the enrichment of tritium near the bottom is low, the total interfacial tension gradient may point upward as in the pure component case. The flip-flop of the direction of the total interfacial tension gradient could also happen in a similar manner when the top of the SGS is kept warmer than the bottom.

A complete mathematical model capable of predicting the direction and magnitude of the total interfacial tension gradient, and hence the condition that gives rise to a uniform liquid layer of binary mixture inside an SGS, will be established in the following sections. In Section 5.2, the fluid flows and temperature fields in the vapor and liquid phases will be derived in a way similar to that for Model II. The amount of tritium (and deuterium) enrichment in the liquid layer or, equivalently, the concentration profiles of deuterium (or tritium) in the vapor and liquid phases, are determined by solving the appropriate diffusion equations, which is presented in Section 5.3. Some numerical examples are presented in Section 5.4.

5.2 Velocity and Temperature Fields

The equation of motion for a nonisothermal mixture fluid is [75]

$$\rho \frac{D \vec{v}}{Dt} = -\vec{\nabla} p + \mu \nabla^2 \vec{v} + \rho \vec{g}, \quad (5.1)$$

where ρ = mass density of the mixture; in the present case, $\rho = \rho_D + \rho_T$,

$\vec{v}$ = mass average velocity = $\frac{1}{\rho} (\rho_D \vec{v}_D + \rho_T \vec{v}_T)$,

$\bar{p}$ = pressure of the mixture,

μ = viscosity of the mixture,

the subscripts D and T, respectively, denote the deuterium and tritium portion in the D-T mixture, and the rest of the symbols are the same as those in Eq. (3.1).

Equation (5.1) is identical to Eq. (3.1) in appearance; however, the definitions of all the variables are different. ρ_D is the mass density of the deuterium portion in the mixture and does not equal the mass density of pure deuterium, ρ_{D_2} . The same is true for ρ_T correspondingly. If the mixture is ideal, then $\rho_D = f_D \rho_{D_2}$ and $\rho_T = f_T \rho_{T_2}$, where f_D and f_T are, respectively, the mole fractions of deuterium and of tritium in the mixture. Note that $f_D + f_T = 1$.

The D-T liquid mixture is actually quite close to being ideal; the deviation of its mass density from the ideal mixture is only about 1% [84,85]. There is almost no such comparison currently available for the D-T vapor mixture. For simplicity, the mixtures in both phases are assumed ideal here. Furthermore, the mass densities of the mixture liquid and vapor are also assumed to be constant throughout the system despite the species-enrichments processes described in the previous section. The error incurred from this assumption will be small if the amount of enrichment is confined to a few percent.

The steady-state equations of continuity for deuterium and tritium are, respectively,

$$\vec{\nabla} \cdot \rho_D \vec{v}_D = 0 , \quad (5.2a)$$

$$\vec{\nabla} \cdot \rho_T \vec{v}_T = 0 . \quad (5.2b)$$

Addition of these two equations gives

$$\vec{\nabla} \cdot \rho \vec{v} = 0 . \quad (5.3)$$

Since the mass density of the mixture, ρ , is assumed to be constant, Eq. (5.3) reduces to

$$\vec{\nabla} \cdot \vec{v} = 0 . \quad (5.4)$$

As in the case of a pure component, if the outside temperature gradient is parallel to the gravitational acceleration, then the fluid flows in the vapor and liquid phases can be assumed azimuthally symmetric. Under this assumption and from Eq. (5.4), one can adopt the Stokes stream function defined in Eq. (3.4) and solve Eq. (3.5), instead of Eq. (5.1), for the velocity fields in the vapor and liquid phases.

The final expressions for the velocity fields are the same as those for Model II derived in Chapter 4. The main differences are that the velocity now represents the mass average velocity of the mixture fluid flows and that all the properties of the fluids are replaced by those for the mixtures. The associated temperature fields will also be the same as those for Model II if the thermal conductivities of the mixtures are assumed to be constant.

Equation (4.3) can now be used to calculate the steady-state migration velocity of a concentric bubble inside an SGS which is filled with D-T.

Equation (4.7) again becomes the condition for maintaining a stationary bubble concentric with the SGS. In these equations, all the properties of the fluids are treated as being constant, except the saturated vapor pressure, p_{sat} , and the liquid-vapor interfacial tension, γ_{lv} . p_{sat} and γ_{lv} are now functions of temperature and the mixture ratio in the liquid phase at the liquid-vapor interface. Without information on the mixture ratio, p_{sat} and γ_{lv} cannot be specified, nor the condition for a stationary bubble.

In order to determine the distribution of the mixture ratio throughout the liquid layer, or specifically, the concentration of deuterium since the mixtures are binary and assumed ideal, the diffusion equations for the liquid and vapor phases must be solved. This part of the calculation is presented in the next section.

5.3 Concentration Profiles

If as is mentioned in Section 5.1, one portion of the liquid layer is enriched with deuterium and the other with tritium by the processes of evaporation and condensation along the liquid-vapor interface, a concentration gradient will build up and, consequently, binary diffusion will start. The appropriate equation that describes this kind of mass transport inside a binary mixture is a binary diffusion equation [75]. This equation written in terms of the deuterium density in the mixture has the form

$$\bar{v} \cdot \rho_D \frac{d\bar{v}}{dz} - \bar{v} \cdot D_{DT} \frac{d\bar{v}}{dz} = 0, \quad (5.5)$$

where D_{DT} is the diffusion coefficient of deuterium in the mixture.

Assuming that D_{DT} is constant throughout the system and combining Eqs. (5.4) and (5.5), one has

$$\vec{v} \cdot \vec{\nabla} \rho_D - D_{DT} \nabla^2 \rho_D = 0. \quad (5.6)$$

Equation (5.6) is now the governing equation for the concentration profiles of the deuterium both in the liquid and vapor phases. If the concentration profiles of interest are for the instant that the bubble is concentric with SGS, namely, that the liquid layer is uniform, then the spherical coordinate system previously defined in Fig. 3.2 can be used as it is.

Equation (5.6) can be rewritten in dimensionless form. For an azimuthally symmetric case, the equation for the liquid phase is

$$(Pe)_l \left[v_{r,l} \frac{\partial \rho_{D,l}}{\partial r} + \frac{v_{\theta,l}}{r} \frac{\partial \rho_{D,l}}{\partial \theta} \right] - \nabla^2 \rho_{D,l} = 0, \quad (5.7a)$$

and for the vapor phase,

$$(Pe)_v \left[v_{r,v} \frac{\partial \rho_{D,v}}{\partial r} + \frac{v_{\theta,v}}{r} \frac{\partial \rho_{D,v}}{\partial \theta} \right] - \nabla^2 \rho_{D,v} = 0, \quad (5.7b)$$

where $Pe = \text{Peclet diffusion number [75]},$

$$= R_B V_{ref} / D_{DT},$$

$$r = \tilde{r} / R_B,$$

$$v = \tilde{v} / V_{ref},$$

and R_B and V_{ref} were previously defined in Chapter 3.

The necessary boundary conditions are

$$(a) \text{ At the origin, i.e., at } r = 0, \quad r \rho_{D,v} = 0; \quad (5.8)$$

$$(b) \text{ At the liquid-vapor interface, i.e., at } r = 1$$

$$(i) \quad y_D = \frac{x_D p_{\text{sat},D}}{x_D p_{\text{sat},D} + (1-x_D) p_{\text{sat},T}} \quad (5.9)$$

where y_D = mole fraction of D_2 in the vapor phase,

x_D = mole fraction of D_2 in the liquid phase,

$p_{\text{sat},D}$ = saturated vapor pressure of pure D_2 ,

$p_{\text{sat},T}$ = saturated vapor pressure of pure T_2 ;

$$(ii) \quad p_{D,v} v_{r,v} - \frac{1}{(Pe)_v} \frac{\partial p_{D,v}}{\partial r} = p_{D,l} v_{r,l} - \frac{1}{(Pe)_l} \frac{\partial p_{D,l}}{\partial r} \quad (5.10)$$

(c) At the substrate inner surface, i.e., at $r = b^{-1} = R_i / R_B$,

$$\frac{\partial p_{D,l}}{\partial r} = 0 \quad (5.11)$$

Equation (5.9) is the familiar Raoult's law. It gives the equilibrium partial vapor pressure of deuterium along the liquid-vapor interface.

Equation (5.10) is the mass conservation of deuterium between the liquid and vapor phases. Equation (5.11) simply says that no deuterium is allowed to diffuse into the solid wall of the SGS.

Because of the nonlinearity of Eq. (5.7), no analytical solution exists. The numerical method is therefore pursued, for which the finite difference scheme is used. The computational method of this kind is quite general and will not be elaborated.

To find the temperature gradient $(t_1)_S$ on the outer surface of the SGS that can bring about a uniform liquid layer comprised of deuterium-tritium mixture, one needs to first assume an initial profile of the deuterium concentration, and then use an iterative method to obtain the value of

$(t_1)_S$ that satisfies Eq. (4.7). Once $(t_1)_S$ is known, the velocity fields both in the liquid and vapor phases can be determined. Equation (5.7) is now ready for numerical integration. If the resulting velocity fields cannot satisfy Eq. (5.7), then a new profile of the deuterium concentration will be assumed. The whole procedure will repeat itself until Eq. (5.7) is satisfied. The final result is the correct temperature gradient that can create a uniform liquid D-T layer inside an SGS. Some examples are considered in the next section.

5.4 Numerical Examples for SGS Filled with D-T

In the following examples, the average mole fractions of deuterium and tritium in the liquid are both 0.5. Following the mathematical procedures outlined in Section 5.3, one is able to find the temperature gradient on the outer surface of an SGS that can bring about a uniform liquid layer of D-T mixture on the inner surface of the SGS. This temperature gradient is related to the liquid layer thickness and is plotted in Fig. 5.1. The parameters of the SGS used in this figure are the same as those in Fig. 3.3, namely, the outer radius and wall thickness of the SGS are, respectively, 100 μm and 5 μm . The properties of the mixture are calculated by the additive law of the ideal mixture. The diffusion coefficients of deuterium inside the vapor and liquid D-T are estimated from P.C. Souers' report [79].

The temperature gradient, $(t_1)_S$, that creates a uniform liquid layer of D-T mixture is positive, which means that the top of the SGS is warmer than the bottom. As mentioned in Section 5.1, evaporation will occur at the warmer portion of the liquid layer where tritium enrichment will occur. This can be seen clearly in Fig. 5.2, which contains the mole fraction gradients of deuterium in the liquid layer at the liquid-vapor interface,

Fig. 5.1 Temperature gradient on the outer surface of an SGS, $(t_1)_s$, required to form a uniform liquid layer, plotted against the liquid layer thicknesses. The SGS is filled with $0.5\text{ nD}_2 - 0.5\text{ nT}_2$, and its outer radius and wall thickness are, respectively, $100\text{ }\mu\text{m}$ and $5\text{ }\mu\text{m}$.

Fig. 5.2 Mole fraction gradient of deuterium in the liquid at the liquid-vapor interface vs. the thickness of a uniform liquid layer on the inner surface of an SGS. The SGS is filled with 0.5 nD_2 - 0.5 nT_2 , and its outer radius and wall thickness are, respectively, 100 μm and 5 μm .

x_D , plotted against the liquid layer thicknesses as shown in Fig. 5.1. The minus sign of x_D shows that there is less deuterium in the liquid layer at the top of the SGS than at the bottom. The concentration profile of this kind makes the total interfacial tension gradient point upward and become the force to pull the liquid upward.

Note that, in all cases, the maximum variations of the deuterium mole fraction are less than 0.25% and, therefore, that the assumption of the constant density of the mixture is valid. The variations of the deuterium mole fraction will be larger, however, when the size of the SGS increases. The outer temperature gradient, required for a uniform liquid layer, and the corresponding mole fraction gradient of deuterium in the liquid are plotted, respectively in scaled form, in Figs. 5.3 and 5.4 for various aspect ratios of the uniform liquid shell.

The typical profiles of deuterium concentration in the vapor and liquid phases are drawn in Fig. 5.5. The SGS used has an outer radius of 100 μm and a wall thickness of 5 μm . A uniform D-T liquid layer of 6 μm thickness is created by imposing a vertical temperature gradient of +0.25 K/cm on the outer surface of the SGS. The average temperature (the temperature at the equator of the SGS) is 24 K. From Fig. 5.5, one sees that the variations of deuterium mole fractions are quite linear in the vertical direction both in the vapor and liquid phases. The mole fraction change in the radial direction is very small except in the vicinity of the center of the vapor bubble. In fact, the variation is so small that it is difficult to distinguish the difference in the drawing. This results from the fact that, while deuterium is relatively mobile in the D-T mixture, the fluid flows in the vapor and liquid are slow.

The stream lines and the velocity profile of the fluid flows at the equator are shown in Fig. 5.6. As in the case of Model I (Fig. 3.7),

Fig. 5.3 Scaled temperature gradient on the outer surface of an SGS, $(t_1)_s / R_B$, required to form a uniform liquid layer on the inner surface of the SGS vs. the aspect ratio of the liquid shell. The aspect ratio is defined as the inner radius of the SGS divided by the liquid layer thickness. The SGS is filled with equal molar deuterium and tritium and is at an average temperature of 24 K. The parameter d is equal to R_B / R_0 .

Fig. 5.4 Scaled mole fraction of deuterium in the liquid at the liquid-vapor interface vs. the aspect ratio of the liquid shell. The aspect ratio is defined as the inner radius of the SGS divided by the liquid layer thickness. The SGS is filled with equal molar deuterium and tritium and is at an average temperature of 24 K. The parameter d is equal to R_B/R_0 .

Fig. 5.5 Typical concentration profiles of deuterium in the vapor and liquid phases inside an SGS. x_D and y_D are, respectively, the mole fraction of deuterium in the liquid and vapor. The liquid layer is 6 μm thick and the average mole fraction of deuterium in the liquid 0.5. The outer temperature gradient required to form such a uniform layer is +0.25 K/cm and the average temperature is 24 K.

Fig. 5.6 (a) Stream lines for the case specified in Fig. 5.5.
 Stream line 1: $\psi_{\ell} = 0$ and $\psi_v = 0$; 2: $\psi_{\ell} = -2.0 \times 10^{-4}$;
 3: $\psi_{\ell} = -3.0 \times 10^{-5}$; 4: $\psi_v = 0.001$; 5: $\psi_v = 0.003$.
 (b) flow velocity profile at the equator for the case shown in (a).

circular flow patterns prevail both for the vapor and liquid phases. This shows that the fluid flows are created dominantly by the liquid-vapor interfacial tension gradient. The rates of evaporation and condensation in this particular case are, in fact, very low resulting in the maximum mass flux across the liquid-vapor interface of only about 1.64×10^{-7} g/sec/cm². The maximum vapor-flow velocity is only about 0.034 cm/sec, far less than the one shown in Fig. 4.4 for Model II. The isotherms shown in Fig. 5.7 also show that the rates of evaporation and condensation are low because there is little distortion on the isotherms caused by the heat associated with the evaporation and condensation.

Based on these observations one concludes that, for binary D-T mixture, the processes of evaporation and condensation are essential in creating component separation in the mixture, which, in turn, gives rise to an interfacial tension gradient which is the overwhelming force that creates a uniform D-T liquid layer inside an SGS. However, the rates of the evaporation and condensation are generally low and have little effect on the velocity and temperature fields. Therefore, one is able to use the formula for Model I, i.e., Eq. (3.26), to estimate the external temperature gradient required for producing a uniform liquid layer. Since the rates of the evaporation and condensation are very low, the concentration profile of deuterium can be calculated by matching (via the Raoult's law) the saturated vapor pressure of the mixture to the sum of the hydrodynamic and hydrostatic pressures in the vapor phase. The outer temperature gradients re-calculated from the formula for Model I are quite close to the previously calculated values for all the foregoing examples. The deviation is less than 1% for all cases.

Fig. 5.7 Isotherms for the case specified in Fig. 5.5. The temperature difference between two adjacent isotherms is 0.000625 K .

5.5 Summary

A theoretical model has been formulated which can predict the configuration of a continuous liquid layer consisting of a binary mixture of hydrogen isotopes, inside an SGS exposed to a nonuniform temperature field. For an SGS filled with D-T mixture, the model predicts that a uniform-thickness liquid layer can be created by a positive vertical temperature gradient, namely, by maintaining the top of an SGS warmer than the bottom. A positive temperature gradient ensures that the liquid layer near the top of the SGS is enriched with tritium and that the direction of the overall interfacial tension gradient points upward and acts against gravity to bring about a uniform liquid layer.

In this model, the fluid flows and the temperature fields in the vapor and liquid phases are calculated from the formula derived for Model II in Chapter 4. The concentration profiles of one of the components in the mixtures in both phases are determined by numerically solving the diffusion equations. The major assumption used in these two calculations is that the mass density of the mixture is constant throughout the system. For an SGS filled with D-T mixture, the variations of the mixture density caused by the nonuniform temperature field and the component separations have been found to be small (less than 1%). Therefore, the assumption of constant density is justified.

CHAPTER 6

EFFECTS OF TRITIUM RADIOACTIVITY

If an SGS contains deuterium-tritium, extra heat will be generated inside the SGS from the tritium decay. This internal heating may affect the formation of a uniform liquid layer by the temperature gradient method discussed in the previous chapters. In order to estimate this heating effect, a simple computer simulation for the tritium decay processes is carried out. The result will be presented in this chapter.

Tritium will undergo radioactive decay according to the reaction

where β^- is the beta particle and $\bar{\nu}$ an antineutrino. The daughter He^3 will not be at the excited nuclear states and hence no gamma rays will be generated. The half-life of the decay is 12.346 years [86]. Since neutrino does not interact with matter, the major effects caused by the radioactive decay are from the beta particles, which have a kinetic energy spectrum from zero to 18.54 keV [79]. The heat that results from stopping these beta particles is 0.324 watts per gram of T_2 [87]. When an SGS contains tritium in it, the tritium beta particle will be attenuated by the tritium itself and the glass wall of the SGS. A local heating effect is hence created. These internal heat sources will likely change the temperature fields inside the SGS.

In order to estimate the strength of the decay heat sources, a simple Monte-Carlo-type computer program is written to simulate the tritium decay

processes. At first, an SGS containing a uniform liquid layer of D-T mixture is specified. The concentration profiles of tritium for the vapor and liquid phases, which are assumed azimuthally symmetric, are then put in. The regions of the vapor, liquid, and glass wall are subdivided into numerous equal volumes. A given amount of tritium beta particles will then be generated in each volume in the liquid layer. The direction of each beta particle emitted is random, and the initial kinetic energy of the particle is determined randomly from the probability density function derived from the energy spectrum. Since the density of vapor D-T is much smaller than that of liquid D-T, the number of beta particles emitted from the vapor is small and is hence ignored here. The history of each emitted particle is then followed by assuming that the beta particles will slow down linearly in energy along a straight line. A beta particle is said to be completely absorbed after it travels a distance determined by the cross-section of the beta particle in the medium. The energy of the particle is then added to the volume in which the particle is stopped. The total energy absorbed in each volume now represents the strength of the local heat sources.

The accuracy of the calculations of this type depends on how small the subdivided volume is and how large the amount of tritium beta particles absorbed in each volume is. The smaller the volume and the larger the amount of tritium beta particles absorbed, the higher is the accuracy. A typical result is shown in Fig. 6.1, in which the strength of the tritium decay heat sources along the equator of an SGS is plotted. In this particular calculation, the numbers of the subdivided volumes in the regions of the vapor, liquid, and wall are, respectively, 4300, 1000, and 1000; and the number of particles emitted from each volume in the liquid layer is 2500. The

Fig. 6.1 The radial variation of the strength of the tritium decay heat sources at the equator of an SGS. The outer radius and wall thickness of the SGS are, respectively, $100 \mu\text{m}$ and $5 \mu\text{m}$. The thickness of the uniform liquid D-T layer is $6 \mu\text{m}$. The tritium concentration profiles in the vapor and liquid phases are as those specified in Fig. 5.5. The parameters b and d are, respectively, equal to R_B/R_j and R_B/R_0 . The horizontal broken line indicates that the strength of the decay heat sources inside a bulk liquid D-T is $5 \times 10^5 \text{ erg/sec/cm}^3$ [88].

SGS employed is the same as that in Fig. 5.5, namely, its outer radius and wall thickness are, respectively, 100 μm and 5 μm . The thickness of the uniform liquid D-T layer on the inner surface of the SGS is 6 μm . The strength of the heat sources has little angular dependence even though the concentration of tritium varies. The errors of the calculated strength of the heat sources in the regions of the vapor, liquid, and wall are, respectively, 10%, 2%, and 5%. These errors are estimated by calculating the variances of the mean energy absorbed in the volumes along the equator of the SGS.

Because of these internal heat sources, the temperature fields inside the SGS might now be different from those previously calculated with no heat source included (see Chapter 3). To see the significance of these heat sources, let us now rewrite the heat equations in the regions of the vapor, liquid, and wall as

$$k_v \nabla^2 T_v = Q_v, \quad (6.2a)$$

$$k_l \nabla^2 T_l = Q_l, \quad (6.2b)$$

$$k_w \nabla^2 T_w = Q_w, \quad (6.2c)$$

where Q is the function of the strength of the heat sources shown in Fig. 6.1; the subscripts v , l , and w denote the vapor, liquid, and wall, respectively; and k is the thermal conductivity. Normalizing the temperature in Eq. (6.2) by the definition specified in Eq. (3.15a) and the Laplacian operator by the liquid layer thickness, i.e., $L_t = R_i - R_o$, one has

$$\nabla^2 T_v = \frac{L_t^2 Q_v}{k_v}, \quad (6.3a)$$

$$\nabla^2 T_l = \frac{L_t^2 Q_l}{k_l}, \quad (6.3b)$$

$$\nabla^2 T_W = \frac{L_t^2 Q_W}{k_W} . \quad (6.3c)$$

The values of the right-hand side of Eq. (6.3) can be estimated by using the average values of Q from Fig. 6.1 for each region. For $L_t = 6 \mu m$, they are

$$\nabla^2 T_V = 6.4 \times 10^{-7} , \quad (6.4a)$$

$$\nabla^2 T_l = 5.8 \times 10^{-6} , \quad (6.4b)$$

$$\nabla^2 T_W = 1.1 \times 10^{-5} . \quad (6.4c)$$

The numbers shown in Eq. (6.4) are several orders of magnitude smaller than unity. Therefore, for a thin liquid D-T layer, there is little effect on the temperature fields inside an SGS caused by the tritium decay heat. The upper bound of the liquid D-T layer thickness in which the tritium decay heat can be ignored can be estimated by setting $L_t^2 Q / k = 0.1$. It is about $500 \mu m$.

The other effect associated with the tritium decay might come from the decay product. He^3 is the daughter and is likely to be in the gas state at the liquid hydrogen temperatures. It, therefore, will not disturb the total liquid-vapor interfacial tension. However, it will impede the processes of evaporation and condensation to some degree since it is noncondensable. Nevertheless, the effect caused by the presence of He^3 in the D-T vapor phase is considered negligible here because the rates of the evaporation and condensation are quite low as pointed out in Chapter 5.

CHAPTER 7

EXPERIMENTAL APPARATUS AND RESULTS

7.1 Introduction

The objective of the experiment is to study the configuration of a liquid hydrogen layer inside a spherical glass shell (SGS) in isothermal and nonisothermal environments. An apparatus capable of cooling down an SGS to liquid hydrogen temperatures and maintaining a desired vertical temperature gradient across the SGS has been designed and constructed. An optical interference microscope is used to observe the profile of the liquid layer. The liquid profiles corresponding to different temperature gradients are recorded and analyzed with a computer. The liquid profile at the zero temperature gradient is then compared with the theoretical prediction in Chapter 2. A particular temperature gradient that brings about a continuous liquid layer of uniform thickness is compared with the theoretical model predictions of Chapters 3, 4, and 5.

7.2 Experimental Apparatus

A schematic diagram of the experimental apparatus is shown in Fig. 7.1. It is an open cycle cooling system. Liquid helium coolant is brought into the system and let out to the open air after it is totally vaporized. Two cryostats, which are placed one over the other and are thermally isolated, share the common liquid helium inlet. The temperature of each cryostat can be adjusted separately by changing the flow rate of the helium exhaust gas and the heat input to the cryostat from the heater wire. The two heaters are also regulated automatically and independently by the feedback signals from the

Fig. 7.1 Schematic diagram of the experimental apparatus.

temperature measurement unit for the purpose of reducing the temperature fluctuations.

A cold chamber, in which an SGS is mounted, is inserted in between the two parallel cryostats. A cross-sectional view of the cold chamber is shown in Fig. 7.2. An SGS is placed near the center of the chamber and horizontally supported by a thin glass fiber which is glued to the tip of a nylon screw. The chamber is filled with ultra-high-purity helium gas. A vertical temperature gradient is created, in the helium gas and hence across the SGS, by changing the temperatures of the copper bars on the top and bottom of the chamber. The two copper rods inserted in the chamber are employed to reduce the height of the helium gas column.

The height of the gas column is chosen according to the following three criteria. The first criterion is the Rayleigh number of the column. A Rayleigh number is the parameter to determine whether or not there is a natural convection in a fluid medium when the top of the fluid medium is cooler than the bottom. It is defined as [89]

$$Ra = \frac{g \beta_{th} \Delta T h_f^3}{\nu \alpha} \quad (7.1)$$

where h_f = height of the fluid medium,

ΔT = maximum temperature difference in the fluid,

β_{th} , ν , and α , = thermal expansion coefficient, kinematic viscosity,

and thermal diffusivity of the fluid, respectively,

and g = gravitational acceleration.

If the Rayleigh number of a fluid medium is less than the critical value, there will be no convection in the medium. For a fluid bounded by two infinite horizontal plates, the critical Rayleigh number is 1707.76 [89]. For

Fig. 7.2 Cross-sectional view of the cold chamber (not to scale).

other geometries, no such critical value has been reported in the literature. However, for a closed vertical cylinder, the critical value can be deduced from the results of the numerical calculations and experiments reported by Liang, Vidal, and Acrivos [90]. It is about 4300.

Since the convection current around the SGS inside the helium gas column is likely to reduce the temperature gradient across the SGS drastically, convection in the gas column must be avoided. From Eq. (7.1), one can see that for a given temperature and pressure of the helium gas in the chamber, the only way to ensure that the Rayleigh number be less than the critical value is to decrease the height of the gas column.

The second criterion is the heat flux limitation. Because the lowest achievable temperature of the present system is the liquid helium boiling temperature (about 4.2 K), the maximum attainable heat flux in the gap between the copper rod and the SGS (and therefore the maximum temperature gradient in the gas column) will be limited by the length of the gap. For instance, assuming that the average temperature of the SGS is 24 K, that the rod temperature is 4.2 K, and that the gap length is 1.0 cm, one finds that the temperature gradient across the gap is about 19.8 K/cm and the maximum heat flux going through the gap is about 3.6×10^4 erg/sec/cm². For this amount of heat flux, the maximum achievable temperature gradient across the SGS is only 2.2 K/cm since the thermal conductivity of glass is much higher than that of helium gas. If a higher temperature gradient across the SGS is desired, the length of the gap has to be shortened to raise the heat flux through the gap.

The third criterion is to ensure a linear temperature gradient across the SGS. Since heat conduction is the sole mechanism to cool the SGS in the helium gas column, the temperature field in the helium gas can be described by

the Laplacian equation (see Chapter 3). Let a spherical coordinate be placed at the center of the SGS. Then, the general expression for the temperature field in the helium gas is

$$T_{\text{He}} = \sum_{n=2}^{\infty} [\hat{C}_{\text{He},n} r^{n-1} + \hat{D}_{\text{He},n} r^{-n}] P_{n-1}(\cos \theta), \quad (7.2)$$

where $T_{\text{He}} = (\hat{T}_{\text{He}} - T_{\text{avg}}) / T_{\text{ref}}$,

$\hat{T}_{\text{He}}$ = temperature in the helium gas column in Kelvin,

T_{avg} = average temperature in the column,

$\hat{C}_{\text{He},n}$, and $\hat{D}_{\text{He},n}$ = constants to be determined by the boundary conditions,

and the rest of the symbols are defined in Chapter 3.

Since a linear temperature gradient is required on the outer surface of an SGS to create a uniform liquid hydrogen layer on the inner surface of the SGS, as concluded in the previous chapters, T_{He} must be in the P_1 -mode, namely,

$$T_{\text{He}} = [\hat{C}_{\text{He},2} r + \hat{D}_{\text{He},2} r^{-2}] \cos \theta. \quad (7.3)$$

This form of temperature field is ensured if and only if the isotherms in the gas column near the surface of the inserted copper rod are parallel to the rod surface. From Eq. (7.3), one sees that as $r \rightarrow \infty$, i.e., at distances many times the size of the SGS,

$$T_{\text{He}} \rightarrow \hat{C}_{\text{He},2} r \cos \theta. \quad (7.4)$$

At such distances away from the SGS, the isotherms in the gas column will be

flat and parallel to the rod surface as indicated by Eq. (7.4). In practice, however, a distance about five times larger than the radius of the SGS is found to be sufficient in satisfying this condition [91].

The two constants in Eq. (7.3) are obtained by imposing the requirements of the continuity of the temperature fields and of the heat fluxes on the surface of the SGS. In terms of the two constants which are already derived with Models I and II for the temperature fields of the glass wall, they are

$$\hat{C}_{\text{He},2} = \frac{1}{3\epsilon_3} \left[\hat{A}_2 (1 + 2\epsilon_3) - 2\hat{B}_2 d^3 (1 - \epsilon_3) \right], \quad (7.5a)$$

$$\hat{D}_{\text{He},2} = \frac{1}{3\epsilon_3} \left[\hat{A}_2 (\epsilon_3 - 1) d^{-3} + \hat{B}_2 (2 + \epsilon_3) \right], \quad (7.5b)$$

where $\epsilon_3 = k_{\text{He}}/k_W$

k = thermal conductivity,

$\hat{A}_2$, $\hat{B}_2$, and d are defined in Chapters 3 and 4,

and the subscripts He and w are, respectively, for the helium gas column and the wall of the SGS.

Keeping these three criteria in mind, one is able to select the appropriate lengths of the two copper rods. The radii of the rods and hence the gas column are also required to be as large as possible to ensure a linear temperature gradient across the SGS inside the gas column. In the present system, the height and the radii of the rods are all 2.5 mm which is about twenty five times larger than the radius of the SGS's to be tested. For the detailed design and construction of the entire system, one is referred to the Master's thesis of Mark Erlenborn [91].

Figure 7.3 is a schematic diagram of the Mach-Zehnder interference microscope used to observe the configuration of the liquid hydrogen layer inside the SGS. A coherent light beam from a low-power He-Ne laser is first

Fig. 7.3 Schematic diagram of the Mach-Zehnder interference microscope.

split into two by a beam splitter. The SGS mounted inside the cold chamber is illuminated by one of the two beams, known as the object beam. Since glass and liquid hydrogen are both transparent, the object beam will transmit through the SGS with little attenuation in intensity. However, its divergence angle and phase will change according to the local thickness of the glass wall and the liquid layer. A microscope objective is then used to enlarge the image of the SGS. When the object beam is recombined with the reference beam (which is the other of the two beams and which passes nothing but a microscope objective identical to the one on the object beam) by another beam splitter, the relative phase shifts of the object beam will be revealed. The intensity of the recombined beam will be zero if the relative phase shift is an odd-number multiple of one-half wavelength of the laser light, and will be doubled if it is a multiple of one wavelength. If the wall of the SGS and the liquid layer inside it are both uniform, the phase shifts of the object beam will be a function of the radial distance from the center of the SGS, and the image of the recombined beam (which is called the interferogram) will look like a target commonly used by marksmen, namely, a series of black and white concentric circles.

The interferogram of the SGS is viewed with a television monitor and recorded on photographic films. The recorded interferogram is then analyzed to determine the thickness and uniformity of the liquid layer inside the SGS. Since only those SGS's that are spherical and that have uniform wall thickness are chosen for the experiment, the analysis is relatively straightforward. Some useful methods previously developed by J. Fanning [92] are adopted for the present analysis. The details of the methods are described in Fanning's Master's thesis.

7.3 Experimental Results

Figure 7.4 shows the shadow and interference micrographs of an SGS in an isothermal environment at room temperature and 22 K. The shadowgram is the real image of the SGS and taken with the reference beam blocked. The SGS shown in Fig. 7.4 has an outer radius of $130\text{ }\mu\text{m}$ and a wall thickness of $15\text{ }\mu\text{m}$. The SGS is filled with pure deuterium. The wall thickness of the SGS is quite uniform because the black and white circles in the interferogram at room temperature (Fig. 7.4b) are almost concentric with the SGS. When the SGS is cooled down to liquid deuterium temperatures, a continuous liquid condensate appears on the inner surface of the SGS. The thickness of the liquid layer then increases as the temperature is brought closer to the triple point of deuterium, namely, 18.73 K. The configuration of the liquid layer at 22 K is shown in Figs. 7.4c and 7.4d. The estimated bottom liquid layer thickness is about $16\text{ }\mu\text{m}$. The nonuniformity of the liquid layer is shown clearly in the interferogram (Fig. 7.4d), in which the centers of the black and white circles are above the center of the SGS. Since the resolution of the present optical interference microscope is about $0.3\text{ }\mu\text{m}$ (half of the wavelength of the He-Ne laser light), it is difficult to determine the top liquid layer thickness which is in the order of $0.1\text{ }\mu\text{m}$ (see Chapter 2). Nevertheless, two conclusions can be made from the pictures shown in Fig. 7.4. The first is that the liquid condensate wets the entire inner surface of the SGS forming a continuous layer which is evidence that liquid deuterium has a vanishingly small contact angle with glass. The second is that the profile of the continuous layer is similar to the one predicted by the theory in Chapter 2.

Figure 7.5 is a series of pictures showing the changes in the configurations of liquid deuterium layer inside an SGS under the influence of different vertical temperature gradients. The outer radius and wall thickness

Fig. 7.4 (a) and (b) are, respectively, the shadowgram and interferogram of an isothermal SGS at room temperature filled with pure deuterium. (c) and (d) are the corresponding pictures at 22 K. The bottom thickness of the continuous liquid layer at 22 K is estimated to be about $16\text{ }\mu\text{m}$. The outer radius and the wall thickness of the SGS are $130\text{ }\mu\text{m}$ and $15\text{ }\mu\text{m}$, respectively.

Fig. 7.5 (a) and (b) are the shadowgram and interferogram of an SGS at room temperature. The corresponding pictures that follow are all at an average temperature of 22 K. (c) and (d), (e) and (f), and (g) and (h) are the corresponding pictures for the temperature gradients of about 0.0 K/cm, -4.1 K/cm, and -4.7 K/cm, respectively. The outer radius and wall thickness of the SGS are, respectively, about 90 μm and 8 μm . The amount of deuterium contained in the SGS is equivalent to having a uniform layer of about 3.3 μm .

of the SGS are, respectively, about $90\text{ }\mu\text{m}$ and $8\text{ }\mu\text{m}$. The amount of deuterium contained in the SGS is equivalent to having a uniform liquid layer of about $3.3\text{ }\mu\text{m}$ thickness. Figures 7.5a and 7.5b are, respectively, the shadowgram and interferogram of the SGS at room temperature. The average temperature of the SGS in the following pictures is about 22 K. Figures 7.5c and 7.5d are the pictures for a zero temperature gradient. As expected, the bottom liquid layer in this case is thicker than the top layer. Figures 7.5e and 7.5f are the pictures recorded when the temperatures at the top and bottom of the gas chamber are 15 K and 30 K. Since copper has a very large thermal conductivity, the temperature drop along the copper rods is negligible (in the order of 0.01 K). Therefore, the temperatures at the top and bottom of the helium gas column can be regarded as the same as those at the top and bottom of the gas chamber. After knowing these two temperatures, one is able to estimate the temperature gradient on the outer surface of the SGS by means of Eq. (7.5). In this case, it is about -4.1 K/cm . It is clear from the pictures that the liquid layer is almost uniform under this temperature gradient. For the same parameters of the SGS, the temperature gradient predicted from Model II is -5.3 K/cm . The discrepancy between the experimental result and theoretical prediction is mainly due to the error (about 10%) in the estimation of the liquid layer thickness. In the present case, a 10% change of the liquid layer thickness will result a 35% change in the calculated temperature gradient. Figures 7.5g and 7.5h show that a continuous liquid layer thicker at the top can be created if the applied temperature gradient has a magnitude larger than that required for a uniform layer.

Because of the unavailability of the SGS's filled with deuterium-tritium mixture, no experimental data for the behavior of liquids containing mixtures of hydrogen isotopes inside an SGS could be obtained.

CHAPTER 8

CONCLUSIONS

Spherical glass shells (SGS) containing a uniform layer of liquid or solid deuterium-tritium (D-T) mixture are the simplest high-gain targets employed for the inertial confinement fusion (ICF) experiment. A technique of using a thermal gradient to form and maintain a uniform liquid hydrogen layer inside an SGS has been investigated. Theoretical models capable of describing the liquid layer configuration inside an SGS both in uniform and nonuniform temperature fields have been formulated. Experiments intended to verify these model predictions have been performed.

When an SGS filled with hydrogen is isothermally cooled to liquid hydrogen temperatures, hydrogen will condense and form a continuous liquid layer covering the entire inner surface of the SGS. The principal reason for the formation of the continuous liquid layer is that hydrogen has a zero contact angle with glass. A rigorous proof has been presented to show that the classical Young-Laplace equation cannot predict the continuous liquid layer and that attraction forces between the hydrogen and glass molecules of the London-van der Waals type must be included in the equation. As a remedy, a new equation containing the attractive forces has been formulated which gives rise to a solution that agrees with the experimentally observed liquid layer profiles. Because of gravity, the liquid layer near the top portion of the SGS is always thinner than that near the bottom. The retardation effect of the London-van der Waals forces, which makes the top portion of the liquid layer even thinner, has also been discussed. Although the London-van der Waals forces are weak compared to the gravitational force and the liquid-vapor

interfacial tension, they are the primary forces that bring about a uniform liquid layer inside an SGS when gravity is absent.

A method for offsetting the gravity effect, which employs a vertically imposed temperature gradient, has been studied. Two theoretical models (Models I and II) have been established. In Model I, only the temperature induced liquid-vapor interfacial tension gradient is considered. In Model II, the combined effects of the interfacial tension gradient and the mass transfer between the liquid and vapor phases are included. In both models, constant fluid properties and fully developed fluid flows are assumed. The fluid flows in the regions of the liquid and vapor are determined by solving the low-Reynolds-number approximation of the fluid equations. The net results of these two models are the analytical expressions for the temperature gradient on the outer surface of an SGS that is required for forming a uniform liquid hydrogen layer on the interior surface of the SGS. Experiments have been performed and preliminary results indicate that the predictions of Model II are more appropriate than those for Model I for an SGS filled with single-component hydrogen. It is evident that mass transfer between the liquid and vapor phases is indeed significant when there is a temperature gradient across the SGS. The formula derived for Model I, on the other hand, is useful in calculating the sagging time of a liquid layer inside an isothermal SGS.

When an SGS is filled with a mixture of hydrogen isotopes, it is found that Model II is not appropriate in describing the behavior of the liquid layer. Additional information concerning the component distribution in the mixture is necessary for more appropriate model calculations. In the case of an SGS containing D-T mixture, the concentration profiles of deuterium in both the liquid and vapor phases are obtained by solving two coupled diffusion

equations under the assumptions that the mixture is binary and obeys the Raoult's law. The fluid flows and the temperature fields are calculated from the formula for Model II under the assumptions that the component separations in the liquid and vapor phases are not severe and cause very little change in the properties of the mixture. The final result is the temperature gradient required for the formation of a uniform liquid layer consisting of a hydrogen mixture inside an SGS. In this case, the required temperature gradient is positive, just the opposite of that needed for an SGS filled with single-component hydrogen. (A positive temperature gradient means that the top of an SGS is warmer than the bottom.) Preliminary results from the early experiments appear to support this theoretical prediction.

If the enclosure inside an SGS contains tritium, a certain amount of heat will be generated internally due to the stopping of beta particles released from the tritium decay. Calculations have been performed, which show that the effect of the decay heat is not important for thin liquid layers, say, for the liquid layer thinner than 0.5 mm.

The present work renders the conclusion that a uniform liquid hydrogen layer inside a small-size ICF target under gravity can be created and maintained by a vertically applied temperature gradient. The size of the target and the liquid layer thickness are limited practically by the range of liquid hydrogen temperatures, that is, the highest and lowest temperatures in the liquid layer must fall in between the critical and triple points of hydrogen. The theoretically predicted temperature gradient agrees with the experimental result for the SGS's filled with pure deuterium. However, verification has yet to be made for the mixture case since sufficient D-T targets are not available at the present time. It is hoped that such tests will be performed in the future and that the accuracy of the theory will be

evaluated.

Although the present theoretical models are capable of explaining the behavior of liquid hydrogen (single-component hydrogen and mixtures of hydrogen isotopes) inside a nonisothermal SGS, the stability of the uniform liquid layer on the inner surface of the SGS created by the thermal gradient has not been considered. The hydrodynamic instability such as the Rayleigh-Taylor type may cause problems especially in large targets. Also, the present models do not include the effects of variations in the fluid properties, particularly the fluid densities, which are caused by the temperature gradient and by the tritium decay heat if tritium is involved. These two points are believed to be worthy of further investigation.

Finally, let us look at some possible research proposals that can be drawn from the findings of this research. First, since the London-van der Waals attractive forces between the liquid and substrate molecules are responsible for a uniform liquid layer inside an SGS when gravity is absent, ICF targets with a uniform fuel layer can be fabricated in the outer space or in a free-fall situation. In fact, ICF target fabrication techniques utilizing free-fall isothermal refreezing are now under active study at the University of Illinois [93]. Second, the magnitude of the London constant characterizing the attractive forces between the liquid and substrate molecules can be found by accurately measuring the top thickness of a continuous liquid layer inside an isothermal shell in the presence of gravity. A high-resolution optical interferometer will, however, be required for such measurements. Last, the phenomenon of component separation in a liquid mixture inside a nonisothermal shell suggests that thermal gradients could be employed to purify or extract desired components.

APPENDIX A

SOLUTIONS FOR THE NONCONCENTRIC BUBBLE

The solutions for the velocity and temperature fields for a nonconcentric bubble inside a spherical shell:

(i) Velocity field

The general expressions for the stream functions in the vapor and liquid phases of a nonconcentric bubble inside a spherical shell are

$$\psi_v(\xi, \eta) = (\cosh \xi - \cos \eta)^{-3/2} \sum_{n=0}^{\infty} Y_n(\xi) \int_{n+1}(\cos \eta), \quad (A-1)$$

$$\psi_l(\xi, \eta) = (\cosh \xi - \cos \eta)^{-3/2} \sum_{n=0}^{\infty} X_n(\xi) \int_{n+1}(\cos \eta), \quad (A-2)$$

$$\text{where } Y_n(\xi) = E_n e^{+n_1 \xi} + F_n e^{+n_4 \xi},$$

$$X_n(\xi) = A_n e^{-n_1 \xi} + B_n e^{+n_1 \xi} + C_n e^{-n_4 \xi} + D_n e^{+n_4 \xi},$$

$$n_1 = n - 1/2,$$

$$n_4 = n + 3/2,$$

$$\int_{n+1}(\cos \eta) = \text{Gegenbauer functions of order } (n+1) \text{ and degree } (-1/2),$$

and the upper alternating sign is for $\xi > 0$ and the lower one for $\xi < 0$.

The six constants in Eqs. (A-1) and (A-2) are determined by imposing the boundary conditions similar to those described in Chapter 3. They are

$$\begin{aligned} \bar{A}_n = \bar{K}_n e^{+n_1 \bar{b}} \{ & \frac{1}{2n_2} \bar{I}_{n+1} [e^{+n_2 \bar{d}} \sinh(n_2 \bar{d}) - n_2 e^{+\bar{d}} \sinh(\bar{d})] + \\ & n_4 \bar{J}_1 [\sigma \cosh(n_4 \bar{d}) - n_2 \sinh(n_4 \bar{d}) + n_1 e^{+\bar{d}} \sinh(n_2 \bar{d})] + \\ & \bar{J}_2 [+ \sigma \sinh(n_4 \bar{d}) + n_2 e^{+n_2 \bar{d}} \sinh(\bar{d}) - e^{+\bar{d}} \sinh(n_2 \bar{d})] \}, \quad (A-3) \end{aligned}$$

$$\begin{aligned} \bar{B}_n = \bar{K}_n e^{+n_1 \bar{b}} \{ & \frac{1}{2n_2} \bar{I}_{n+1} [- n_2 e^{+\bar{d}} \sinh(\bar{d}) + e^{+n_2 \bar{d}} \sinh(n_2 \bar{d})] + \\ & n_4 \bar{J}_1 [\sigma \cosh(n_4 \bar{d}) - n_2 \sinh(n_4 \bar{d}) + n_1 e^{+\bar{d}} \sinh(n_2 \bar{d})] - \\ & \bar{J}_2 [+ \sigma \sinh(n_4 \bar{d}) - n_2 e^{+n_2 \bar{d}} \sinh(\bar{d}) + e^{+\bar{d}} \sinh(n_2 \bar{d})] \}, \quad (A-4) \end{aligned}$$

$$\begin{aligned} \bar{C}_n = \bar{K}_n e^{+n_4 \bar{a}} \{ & \frac{1}{2n_2} \bar{I}_{n+1} [n_1 e^{+\bar{d}} \sinh(n_2 \bar{d}) - n_2 \sinh(n_1 \bar{d})] + \\ & n_1 \bar{J}_1 [\sigma \cosh(n_1 \bar{d}) - e^{+\bar{d}} \sinh(n_2 \bar{d}) - n_2 e^{+n_2 \bar{d}} \sinh(\bar{d})] + \\ & \bar{J}_2 [+ \sigma \sinh(n_1 \bar{d}) + n_2 \sinh(n_1 \bar{d}) - n_1 e^{+\bar{d}} \sinh(n_2 \bar{d})] \}, \quad (A-5) \end{aligned}$$

$$\begin{aligned} \bar{D}_n = \bar{K}_n e^{+n_4 \bar{a}} \{ & \frac{1}{2n_2} \bar{I}_{n+1} [n_1 e^{+n_2 \bar{d}} \sinh(\bar{d}) - \sinh(n_1 \bar{d})] + \\ & n_1 \bar{J}_1 [\sigma \cosh(n_1 \bar{d}) - \sinh(n_1 \bar{d}) - n_4 e^{+n_2 \bar{d}} \sinh(\bar{d})] + \\ & \bar{J}_2 [+ \sigma \sinh(n_1 \bar{d}) + \sinh(n_1 \bar{d}) - n_1 e^{+n_2 \bar{d}} \sinh(\bar{d})] \}, \quad (A-6) \end{aligned}$$

$$\begin{aligned} \bar{E}_n = \bar{K}_n e^{+n_1 \bar{b}} \{ & \frac{1}{2n_2} \bar{I}_{n+1} [n_1 n_4 + \cosh(n_2 \bar{d}) - n_2 n_2 \cosh(2\bar{d})] + \\ & 2 n_1 n_4 \bar{J}_1 \sigma \sinh(n_2 \bar{d}) \sinh(\bar{d}) - \\ & \bar{J}_2 \sigma [n_2 \sinh(n_1 \bar{d}) - n_1 \sinh(n_4 \bar{d})] \}, \quad (A-7) \end{aligned}$$

$$\bar{F}_n = - \bar{E}_n e^{+2\bar{b}}, \quad (A-8)$$

where $\bar{d} = \bar{a} - \bar{b}$,

$$n_2 = n + 1/2,$$

$$\bar{J}_1 = \frac{U n (n+1)}{2^{+3/2}} \left(\frac{1}{n_1} e^{-n_1 \bar{a}} - \frac{1}{n_4} e^{-n_4 \bar{a}} \right),$$

$$\bar{J}_2 = \frac{U n (n+1)}{2^{+3/2}} \left(e^{-n_1 \bar{a}} - e^{-n_4 \bar{a}} \right),$$

$$\frac{1}{K_n} = 2 \sigma [\sinh(2n_2 \bar{d}) - n_2 \sinh(2\bar{d})] - \\ 2 [n_1 n_4 + \cosh(2n_2 \bar{d}) - n_2 n_2 \cosh(2\bar{d})],$$

$$\bar{T}_{n+1} = - \frac{n(n+1)(2n+1)}{2 \mu_v v_{ref}} \int_0^\pi \frac{\partial \gamma_{lv}}{\partial \eta} (\cosh b' - \cos \eta)^{-1/2} \int_{n+1}^{\cos \eta} d\eta.$$

(ii) Temperature fields

The general expressions for the temperature fields in the vapor and liquid phases of a nonconcentric bubble inside a spherical shell are

$$T_v(\xi, \eta) = (\cosh \xi - \cos \eta)^{+1/2} \sum_{n=0}^{\infty} \hat{C}_n e^{+n_2 \xi} P_n(\cos \eta), \quad (A-9)$$

$$T_l(\xi, \eta) = (\cosh \xi - \cos \eta)^{+1/2} \sum_{n=0}^{\infty} [\hat{A}_n e^{-n_2 \xi} + \hat{B}_n e^{+n_2 \xi}] P_n(\cos \eta), \quad (A-10)$$

where $P_n(\cos \eta)$ = Legendre function of order n .

The necessary boundary conditions are

$$\text{at } \xi = \frac{+}{-} \bar{b} : \quad T_l = T_v, \quad \text{and} \quad (A-11)$$

$$\text{and } \frac{\partial T_l}{\partial \xi} = \frac{\partial T_v}{\partial \xi}; \quad (A-12)$$

$$\text{at } \xi = \frac{+}{-} \bar{a} : \quad T_l = T_{ext}(\eta), \quad (A-13)$$

The determining equations for the three constants in Eqs. (A-9) and (A-10) become

$$\hat{C}_n e^{-n_2 \bar{b}} = \hat{A}_n e^{+n_2 \bar{b}} + \hat{B}_n e^{-n_2 \bar{b}}, \quad (A-14)$$

$$+ \frac{1}{n_2} \epsilon_1 \sinh(\bar{b}) \left[\hat{A}_n e^{\mp n_2 \bar{b}} + \hat{B}_n e^{\pm n_2 \bar{b}} \right] -$$

$$\epsilon_1 \sum_{m=0}^{\infty} (2m+1) \left[\hat{A}_m e^{\mp (m+1/2) \bar{b}} - \hat{B}_m e^{\pm (m+1/2) \bar{b}} \right] f_{mn} =$$

$$+ \frac{1}{n_2} \sinh(\bar{b}) \hat{C}_n e^{-n_2 \bar{b}} + \sum_{m=0}^{\infty} (2m+1) \hat{C}_m e^{-(m+1/2) \bar{b}} f_{mn}, \quad (\text{A-15})$$

$$\hat{A}_n e^{\mp n_2 \bar{a}} + \hat{B}_n e^{\pm n_2 \bar{a}} = \bar{T}_n, \quad (\text{A-16})$$

$$\text{where } f_{mn} = \int_{-1}^{+1} (\cosh \bar{b} - \cos \eta) P_m(\cos \eta) P_n(\cos \eta) d(\cos \eta), \text{ and}$$

$$\bar{T}_n = \frac{2n+1}{2} \int_{-1}^{+1} T_{\text{ext}}(\eta) (\cosh \bar{a} - \cos \eta)^{-1/2} P_n(\cos \eta) d(\cos \eta).$$

Since f_{mn} is not zero when $m \neq n$, Eq. (A-15) indicates that there are two series consisting of an infinite number of unknowns. This makes it impossible to find a complete solution to the temperature fields for a nonconcentric bubble inside a spherical shell. A simple solution, however, can be found if the $(\cos \theta)$ term in f_{mn} is dropped. This approximation is only valid when $(\cosh \bar{b}) \gg 1$, that is, when the bubble is not too far away from the center of the spherical shell. Based on this approximation, one is able to simplify Eq. (A-15). It becomes

$$\epsilon_1 \left\{ \hat{A}_n e^{\mp n_2 \bar{b}} \left[\pm \sinh(\bar{b}) - 2 n_2 \cosh(\bar{b}) \right] + \hat{B}_n e^{\pm n_2 \bar{b}} \left[\pm \sinh(\bar{b}) + 2 n_2 \cosh(\bar{b}) \right] \right\} = \hat{C}_n e^{-n_2 \bar{b}} \left[\pm \sinh(\bar{b}) \mp 2 n_2 \cosh(\bar{b}) \right]. \quad (\text{A-17})$$

By combining Eqs. (A-14), (A-16) and (A-17), one finds the three constants in Eqs. (A-9) and (A-10) as

$$\hat{A}_n = \frac{1}{2} \bar{T}_n \bar{M}_n e^{\pm n_2 \bar{b}} [\mp (1 - \epsilon_1) \sinh(\bar{b}) + 2 n_2 (\epsilon_1 \pm 1) \cosh(\bar{b})], \quad (A-18)$$

$$\hat{B}_n = \frac{1}{2} \bar{T}_n \bar{M}_n e^{\mp n_2 \bar{b}} [\pm (1 - \epsilon_1) \sinh(\bar{b}) + 2 n_2 (\epsilon_1 \mp 1) \cosh(\bar{b})], \quad (A-19)$$

$$\hat{C}_n = 2 n_2 \epsilon_1 \bar{T}_n \bar{M}_n e^{\pm n_2 \bar{b}} \cosh(\bar{b}), \quad (A-20)$$

where $\frac{1}{\bar{M}_n} = (1 - \epsilon_1) \sinh(\bar{b}) \sinh(n_2 \bar{d}) + 2 n_2 \cosh(\bar{b}) [\epsilon_1 \cosh(n_2 \bar{d}) - \sinh(n_2 \bar{d})]$.

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