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FINAL REPORT NO. 1

**RESEARCH ON THE
WETTED FIRST WALL CONCEPT
FOR FUTURE LASER FUSION REACTORS**

from October 1, 1974 to January 31, 1976

Co-principal Investigators:

Professor Myron A. Hoffman

Professor Zuhair A. Munir

Intramural Order 6774105

MASTER

UNIVERSITY OF CALIFORNIA
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Abstract

Research is in progress to determine the feasibility of the wetted first wall concept for a future laser fusion reactor. The basic idea involves the use of a thin coating of lithium on the inner wall of the laser fusion containment vessel to protect it from the micro-explosion blast debris. This report contains a review of the available information on contact angles and wettability of alkali metals on various metal substrates as well as a review of literature on thin falling liquid films. A proposed experiment to measure the contact angles of lithium on stainless steel and niobium is described. The requirements for a second experiment to measure certain key characteristics of thin falling films are also included.

CONTENTS

Abstract

List of Symbols

1. Introduction
2. Review of the Literature on Liquid-Solid Interfacial Phenomena
3. Review of the Literature on Thin Falling Films
4. Design of Contact Angle Experiments
5. Design Considerations for the Initial Falling Film Experiments
6. Summary

Appendix A - Cleaning and Operating Procedures

Appendix B - Laboratory Handling of Lithium

Tables

Figures

References

LIST OF SYMBOLS

A	Dimensionless wave amplitude (defined on page 3.15)
a	Wave amplitude
b	Film thickness
c	Wave speed
C_r	Dimensionless wave speed
g	Acceleration of gravity
N_{FR}	Froude no. (defined on page 3.1)
N_{RE}	Reynolds no. (defined on page 3.1)
N_{WE}	Weber no. (defined on page 3.1)
N_G	Surface tension no. (defined on page 3.1)
N_T	Film thickness parameter (defined on page 3.13)
u	Velocity
α	Dimensionless wave number (defined on page 3.10)
β	Plate inclination angle from the horizontal
Γ	Mass flow rate per unit film width
θ	Contact angle between liquid and solid substrate
λ	Wavelength
μ	Dynamic viscosity
ν	Kinematic viscosity
ρ	Density
σ, γ	Surface tension coefficient
ω	Wave frequency (radians/sec)

RESEARCH ON THE SUPPRESSED-ABLATION
WETTED-FIRST-WALL CONCEPT
FOR LASER FUSION REACTORS

1. INTRODUCTION

The objective of this research is to provide some basic design data on the coating of solid metallic walls with thin, uniform films of liquid lithium. The specific application is to the laser-induced fusion reactor concept of the Lawrence Livermore Laboratory described in References 1.1 and 1.2. In order to appreciate the unique requirements of this application, a brief description of the salient features of this so-called suppressed-ablation LCTR (laser controlled thermonuclear reactor) concept will be given first. Then the lithium coating requirements for the protection of the blast-chamber first wall which faces the plasma will be discussed.

a. Suppressed-Ablation LCTR Concept

The LCTR concept of Reference 1.1 is an attempt to create a laser-induced micro-explosion which produces relatively low pressure pulses on the blast-chamber first wall and structure. The overall reactor layout is shown in Figure 1.1 (Ref. 1.3) and the key parameters of the LLL reference design of Reference 1.2 are shown in Table 1.1. This design has some general features in common with several other proposed laser-fusion conceptual designs. Provision is made for multiple laser beams to simultaneously irradiate a small pellet containing deuterium and tritium. If adequate spherical symmetry of the radiation flux is achieved the pellet will ablate uniformly enough to cause a thousand to ten-thousand fold density compression of the core. As described in Reference 1.4,

thermonuclear conditions can thereby be reached, followed by a micro-fusion explosion of the pellet core. Energy gains, defined as the ratio of the TN (thermonuclear) energy released to laser light energy incident on the pellet, of between 10 and 100 appear possible with bare D-T spherical pellets.

The combination of design features unique to the "suppressed-ablation" concept which result in relatively low pressure pulses on the first wall are described by Hovingh et al. in Reference 1.2:

- 1) Reduced energy yield per shot
- 2) Improved pellet design
- 3) Increased first-wall area

The first item implies that the laser light energy input per shot is greatly reduced compared to other designs. For example, the LLL reference design of Reference 1.2 uses 10^5 J per shot compared to 10^6 J per shot for an early LASL reference wet-wall design. This would imply about an order of magnitude lower fusion energy yield per shot for the LLL design, all other things being equal. However, as indicated by the typical gain curves of Figure 1.2, the yield for bare D-T spheres may be reduced by an additional factor of .5 to .8 due to the order of magnitude lower laser light energy input per shot of the LLL design. Consequently, the fusion energy yield per shot for an assumed gain of 70 is 7MJ for the LLL reference design (compared to about 100MJ for the LASL wet-wall reference design referred to above).

Clearly, this feature alone can result in over an order of magnitude reduction in the amount of lithium blow-off from the wet first wall. Since it is this lithium blow-off or ablation which can cause a strong reaction pressure pulse on the first wall and any structure connected to it, this also implies over an order of magnitude reduction in the pressure pulse.

The improved pellet design of Item (2) above refers to the use of a low mass (10^{-4} grams) hollow D-T pellet without any layers of high Z material on the outside. This results in much less pellet debris impinging on the first wall, and hence much less lithium ablation. (The hollow pellet allows some reduction in peak laser power required compared to a solid D-T pellet but does not appear to improve the gain significantly according to Reference 1.4.)

Further reductions in the energy flux impinging on the first wall are achieved by using a blast chamber of larger radius to increase the first-wall area, as mentioned in Item (3) above. In addition the LLL reference design employs a pyramidal-shaped surface on the first wall to increase the effective surface area as shown in Figure 1.3.

The combination of all these features results in a low energy flux of only about 1.1 J/cm^2 normal to the first-wall surfaces (compared to about 70 J/cm^2 in the early wet-wall reference design of LASL). The resulting pressure pulse in this suppressed-ablation design is reduced to a peak value of less than 0.1 bar (Ref. 1.2).

This impressively low pressure pulse is not achieved without some penalty, however. Clearly, the power output of each laser cavity would be over an order of magnitude lower than other wet-wall designs for the same pulse rate. However, the very much lower lithium vapor load on the vacuum pumps may permit use of a pulse rate of 10 Hertz (rather than the 1 Hertz pulse rate used in some other wet-wall designs). This higher pulse rate can result in an economically reasonable power output per cavity while still retaining the advantages of the suppressed ablation concept.

While the LLL reference design appears to result in relatively low energy fluxes to the blast-chamber first wall, protection of this wall from thermal

shock damage, sputtering and surface ablation is still essential. The LLL reference design employs a very thin "first-wall liner" or shield of niobium (0.4 mm thick) coated with a 0.3 mm layer of liquid lithium. These are made up of triangular plates 6 cm on a side, forming the pyramidal first-wall liner shown in Figure 1.3.

The various contributions to the energy flux on the first wall of this LCTR arrive on roughly the time scales shown in Table 1.2. The X-rays can be seen to arrive first at about 6 to 30 nanoseconds after the thermonuclear (TN) burn. The neutrons arrive next (with some overlap) at about 20 to 100 ns. Finally the pellet debris including unburned deuterium and tritium ions as well as some of the alpha particles arrive after roughly 1 microsecond.

The corresponding penetration depths of the various components are shown in Figures 1.4 and 1.5. The unburned deuterium and tritium ions (about 75 to 80% of the original pellet mass) can be seen to be able to penetrate up to about 10 microns in liquid lithium (Fig. 1.5). The 3.5 MeV alpha particles which lose most of their energy in the plasma have about the same order of magnitude penetration distance. However, the alpha particles which do not lose a significant portion of their initial energy can penetrate up to 50 microns. Also from Figure 1.4 we see that the 1-10 MeV knock-on deuterium and tritium ions can also be seen to penetrate several hundred microns.

As a result of preliminary calculations such as these, the Laser Systems Studies Group at LLL has requested a study of the feasibility of coating large areas of the first wall of the blast chamber with liquid lithium. The liquid lithium must be thick enough to protect a first-wall liner or shield from serious erosion and sputtering in order to obtain a reasonable lifetime, perhaps on the order of a year or more.

1. Possible Wetted-Wall Concepts

The original LLL preliminary design study of Reference 1.2 suggested the possibility of coating a 400 micron thick niobium first-wall liner or shield with a 300 micron thick lithium layer utilizing capillary pumping. The multiple lithium feed ports envisioned are shown in Figure 1.3. Many basic questions regarding the feasibility of this scheme as well as other possible schemes remain to be answered. One basic question regards simply the feasibility of coating such a complex surface with a relatively uniform film of liquid lithium. This is the subject of the present research program.

It must be recognized that this question is only part of a much broader design problem. The complexity of the overall first-wall design problem for an LCTR is illustrated by Figure 1.6 (prepared by J. Hovingh of LLL). It will take many research programs and many years of effort and experimentation to fill in all the details of the various interactions which determine the ultimate feasibility and lifetime of a first-wall design concept.

Common to many of the lithium coating schemes are some basic questions regarding the behavior of thin films of lithium on metal walls at various inclination angles. As a result, one of the fundamental objectives of this research is to provide basic data on lithium contact angles and wettability which can be used in a wide variety of design concepts employing thin lithium films. The factors affecting the contact angle and its measurement are discussed in Section 2 and the initial contact angle experiment design is described in Section 4.

The whole question of the wettability of a metal surface by liquid lithium is obviously very closely related to the contact angle. However, it is clear that many additional factors influence whether a stable liquid film of relatively uniform thickness can be produced and maintained on the complex first-wall geometry

of the blast chamber. Some of these factors which ultimately must be considered are the following:

- 1) Surface inclination angle
- 2) Dimensions and shape of the surface segment to be coated
- 3) Liquid feed mechanism
- 4) Change in surface condition with time (e.g., due to X-ray, ion and/or neutron bombardment)
- 5) Temperature and temperature gradient time dependence during a cycle
- 6) Nature of other disturbances suffered by the film during and after a shot.

For the first wall of a closed, roughly spherical blast chamber, all inclination angles from 0° (horizontal with the film on the top of the surface) to 180° (horizontal with the film on the bottom of the surface) are theoretically possible. In practice, it may be possible to limit the range of inclination angles somewhat to avoid the region near 180° .

The dimensions and shape of the individual surface segments to be coated are clearly strongly coupled to Item (3), the liquid-feed mechanism proposed. Some of the possible conventional and unconventional feed mechanisms which can be considered are:

- a) Capillary pumping of films (often involving rising films)
- b) Gravity pumping of films (i.e., so-called falling films)
- c) Porous-plate liquid-feed systems
- d) Centrifugal force coating (e.g., in rotating chambers)
- e) Solid extrusion from holes or other feed ports (e.g., lithium hydride as proposed by R. W. Werner of LLL)
- f) Flash evaporation followed by vacuum deposition on the walls (e.g., employing a low-power laser to evaporate a lithium pellet in between D-T pellet shots as suggested by M. A. Hoffman)
- g) Combinations of the above.

Which of the above feed mechanisms are considered "conventional" and which "unconventional" depends entirely on one's prejudices and past experience, of course.

For this particular application, we began our study and literature search on what we consider to be the conventional feed mechanism (b), the so-called falling film approach, for the following reasons. First of all, the falling-liquid films utilize the relatively large gravity forces rather than capillary forces. This offers the possibility of employing rather simple, widely-spaced feed ports, or slots, and of coating relatively large surface areas from each feed port. Secondly there is a wealth of theoretical and experimental data on falling-film behavior, primarily from the field of chemical engineering. However, almost all of these data are for water; in fact, we have not yet found a single experiment specifically on liquid metal falling films.

In Section 3 we describe the falling-film data for water and show some predictions for liquid lithium based on the available theoretical scaling laws. These laws have been verified for a few other liquids than water but not for any liquid metals as far as we know.

In Section 5 we discuss the first experiments we plan to run on thin falling water films. These experiments are primarily to calibrate our measuring instruments and to refine our measurement techniques for conditions of variable plate inclination angle. We plan to continue to study and compare the other possible liquid-film coating mechanisms to the falling-film approach while performing these initial calibration experiments. The research program status and plans are summarized in Section 6.

2. REVIEW OF THE LITERATURE ON LIQUID-SOLID INTERFACIAL PHENOMENA

A. Review of Fundamentals

While experimental data of contact angles between liquids and solids are relatively abundant, they tend to be highly restricted in terms of the investigated materials and the chosen parameters. In this section we shall first review the fundamentals of the interaction between liquids and solids and then examine recent results demonstrating the influence of various parameters on the wettability of solids with special emphasis on liquid lithium data.

The original contributions of Laplace have been modified by Young so as to describe the interaction between a liquid drop and a solid substrate (Refs. 2.1-2.4). The contact angle, θ , shown in Figure 2.1 defines the following equilibrium relationship between the three interfacial energies:

$$\gamma_{SV} = \gamma_{SL} + \gamma_{LV} \cos \theta \quad [2.1]$$

where γ_{SV} , γ_{SL} and γ_{LV} represent, respectively, the energies of the solid-vapor, solid-liquid and liquid-vapor interfaces. Complete wetting is attained when the value of θ is zero degrees and an arbitrary value of 90° has been selected as the boundary between partial wetting and nonwetting. The early work of Dupre has introduced the concept of a reversible work of adhesion between liquids and solids (Ref. 2.5). In terms of the interfacial energies defined in Eq. [2.1] this work term is defined as:

$$W_A = \gamma_{SV} + \gamma_{LV} - \gamma_{SL} \quad [2.2]$$

A similar and somewhat more useful term is that defining spreading as:

$$S = \gamma_{SV} - (\gamma_{LV} + \gamma_{SL}) \quad [2.3]$$

For spreading to occur the spreading coefficient, S must be positive. Furthermore, a corresponding but not sufficient condition for spreading is that $\gamma_{LV} < \gamma_{SV}$.

This latter criterion is often used as a screening device in the selection process of wetting liquids. As defined by Harkins (Refs. 2.6-2.9), the spreading coefficient is the difference between the work of adhesion and the work of cohesion, or

$$S = W_A - W_C \quad [2.4]$$

Relating this equation to Eqs. [2.2] and [2.3], it can be seen that the reversible work of cohesion, W_C , is twice the vapor-liquid interface energy.

The importance of adsorbed gases on the contact angle between a liquid drop and a solid substrate has been demonstrated by Bangham and Razouk (Refs. 2.10 and 2.11). The relationships obtained by these authors are a generalized form of the Young-Dupre equation. The relation between the contact angle and the non-modified interfacial energies are

$$\gamma_{SV}^o - \gamma_{SL} = \gamma_{LV}^o \cos \theta \quad [2.5]$$

and

$$W_A = \gamma_{SV}^o + \gamma_{LV}^o - \gamma_{SL} \quad [2.6]$$

where γ_{SV}^o is the solid-vacuum interfacial energy and γ_{SV}^o and γ_{LV}^o are the energies of the interfaces when the condensed phase is saturated with the adsorbed vapor. Thus by substituting the value of γ_{SL} (Eq. [2.1]) into Eq. [2.6] the work of adhesion for this case becomes

$$W_A = (\gamma_{SV}^o - \gamma_{SV}^o) + \gamma_{LV}^o (1 + \cos \theta) \quad [2.7]$$

The concept of a contact angle hysteresis is associated with the difference between angles at advancing and receding edges of a liquid drop. The difference between these two contact angles arises from the fact that the advancing angle is that between the liquid and the dry substrate while the receding angle represents the interfacial equilibrium between the liquid and a wetted solid surface. For a tilted substrate, the balance between gravitational and surface forces gives the following approximate relationship:

$$mg \sin \alpha \approx w \gamma_{LV} (\cos \theta_R' - \cos \theta_A') \quad [2.8]$$

where α is the angle of tilt of the substrate, w is the average width of the drop and θ_A' and θ_R' represent the angles of the advancing and receding edges of the drop. At a critical value of α a balance between the surface and gravitational forces can be maintained. Beyond this value, the drop will no longer be able to adjust itself and will roll off the substrate. In most cases, hysteresis does not necessarily result from the difference between "wetted" and clean surfaces, but rather from possible chemical changes at the solid-liquid interface. Such changes can come about as a result of the diffusion of the liquid into the solid substrate. An experimental verification of such a process will be given in a later portion of this discussion.

The above formalism, while being useful in relating the γ quantities to each other is not without serious shortcomings. For example, the derivation of Eq. [2.1] ignores the contribution of the vertical component of γ_{LV} . Since we normally deal with rigid substrates, this simplification is not unacceptable. A more serious shortcoming of this formalism, however, is that it cannot be used to calculate γ values from observed contact angles. Surface energies, at least two of the three shown in Figure 2.1 must be determined independently. Attempts aimed at calculating these quantities theoretically are necessarily based on idealized surfaces. For instance, calculations of the solid-vapor surface free energy can be made on the basis of bond considerations between atoms on a well-defined crystalline plane. As an example of such calculations, the high density plane (111) of a face-centered cubic (fcc) metal will be considered. The coordination number (N_c) of surface atoms is 9 (compared to 12 in the bulk). Thus, by cleaving an fcc metal along the (111) plane a total of three bonds per atom are broken. Since two planes are created in this process, this number must be divided by two. To calculate the energy of each bond we take advantage of the

enthalpy of sublimation ΔH_s which, of course, is equal to the product of the total number of bonds broken and the energy per bond, i.e.,

$$\Delta H_s = \phi \left[\frac{1}{2} N_c \right] N_0 \quad [2.9]$$

where ϕ is the energy per bond, N_0 is the number of atoms in the crystal, and N_c is the coordination number which is equal to 12 for fcc structures. The enthalpy of surface formation ΔH_{sf} , as stated earlier, is $\frac{3}{2} \phi$ per surface atom. Therefore,

$$\Delta H_{sf} \text{ (per atom)} = \frac{3\Delta H_s}{N_c N_0} \quad [2.10]$$

or, for an fcc metal,

$$\Delta H_{sf} \text{ (per atom)} = \frac{\Delta H_s}{4N_0} \quad [2.11]$$

Since it is convenient to consider surface energy per unit area, we introduce the quantity N as the number of surface atoms per unit area. For a large number of metals, an atomic diameter of $\sim 3 \text{ \AA}$ is typical. This gives an atomic density of $1.1 \times 10^{15} \text{ atoms/cm}^2$ on the (111) plane. Thus, the enthalpy of surface formation per unit area is

$$\Delta H_{sf} \approx \frac{10^{15}}{4} \cdot \frac{\Delta H_s}{N_0} \quad [2.12]$$

Since heats of sublimation are usually given on a molar basis, N_0 is then Avogadro's number.

When these calculated surface enthalpies are compared with measured surface free energies, a reasonable agreement is obtained for many face-centered cubic metals. In accordance with the method outlined above, the calculated enthalpies of surface formation for Cu, Ag and Au are 1409, 1185 and 1580 ergs/cm², respectively. These compare reasonably well with the experimentally determined surface free energies of 1650, 1140 and 1460 ergs/cm², respectively (Refs. 2.12-2.14). It should be emphasized, however, that the calculated values

are enthalpies and that agreement with the surface free energies is only anticipated at low absolute temperatures since the entropy of surface formation is expected to have a finite positive value. Aside from its failure to account for the entropy change, this approach suffers from the assumption of first-neighbor interaction only. In other words we have neglected the influence of second and subsequent neighbors in our calculation of the bond energy. Indeed it has been suggested that the discrepancy between calculated and measured values is in part the result of this simplification. For crystal structures with lower coordination numbers, body-centered cubic solids with $N_c = 8$, the theoretically calculated surface enthalpies are significantly different from measured surface free energies. For iron we calculate a ΔH_{sf} of 964 ergs/cm² while the reported experimental value is about 1800 ergs/cm² (Ref. 2.15). Similarly, the calculated value for Nb is 1742 ergs/cm² and the experimentally determined quantity is about 2200 ergs/cm² (Ref. 2.15).

Calculations of the liquid-vapor surface energies are more complicated and involve additional simplifying assumptions. The coordination number in liquid metals ranges from about 6 to 11 with most metals having an N_c value between 9 and 11. Lacking long-range order, liquids are more difficult to deal with. Calculations of the number of bonds broken in the process of forming a surface can only be based on a statistical estimate of $\frac{1}{2} N_c$. Furthermore, the density of surface atoms must be estimated. In spite of these difficulties, in principle at least, surface energies of liquid-vapor interfaces can be calculated.

The picture concerning the solid-liquid interface is much less clear and simplifying assumptions cannot be justifiably made in this case. Unlike solid-vapor and liquid-vapor interfaces which are created by breaking bonds, the solid-liquid interface results from breaking solid-solid and liquid-liquid bonds and then creating new solid-liquid bonds. As shown above, the energies associated

with the solid-solid and liquid-liquid bonds can be calculated, albeit with varying degrees of assumptions. This is not the case for the solid-liquid bond which in the absence of diffusional processes across the interface, can not be assumed to be the same as that of an intermetallic compound or a solid solution between the metals of liquid and solid phases. In the case of Li/Nb, information is not available for the thermodynamic properties of this binary. Thus, even crude assumptions cannot be made. What complicate the problem further, are interdiffusional processes across the liquid-solid interface. The composition, and hence energetics of the phases near the interface change with time in accordance with Fickian's laws of diffusion. In a subsequent section the influence of diffusional process will be demonstrated.

B. Review of Recent Findings

In reviewing published results on contact angle and wettability measurements, only those investigations with a critical analysis of the parameters were carefully examined. Of these parameters, temperature has been the most widely investigated. A generalized behavior for contact angles between liquids and ceramic systems has been proposed by Rhee (Ref. 2.16):

$$\cos \theta = 1 + B(T - T_C) \quad [2.13]$$

In this equation T_C is the temperature at which complete wetting is attained and B is a constant. The obvious conclusion from this relationship is that the contact angle is a decreasing function of temperature. Experimental verifications for this conclusion are at hand (Ref. 2.17). However, their interpretation is made complicated by phase changes occurring at higher temperatures. Thus it is difficult to assess unambiguously the influence of temperature on the γ values (and hence θ) of Eq. [2.1]. In other words observed changes in θ

may result from changes in γ caused by compositional variations rather than by temperature changes. The recent work of Zee and Murir (Ref. 2.18) on the system $\text{Au-Ge}(l)/\text{GaAs}(s)$, shown in Figure 2.2, is an example of this point. Contact angles fall into three distinct temperature regions. In the first, (low temperature) region a decreasing contact angle is interpreted as an indication of equilibration of the liquid drop with the solid substrate. No significant dependence of the contact angle on temperature is observed in the second stage. However, the decrease of contact angle values with temperature in the last (high temperature) region has been shown to be the result of diffusional processes altering the original interface. Electron microprobe analysis showed extensive phase changes at the high temperatures. Figure 2.3 shows an electron microprobe scan of the interface demonstrating considerable compositional changes. Electron microprobe scans of the interface between liquid Au-Ge alloys and oriented gallium arsenide established at temperatures corresponding to the flat portion of the curves of Figure 2.2 are demonstrated by Figure 2.4. No diffusional process across the interface appears to have taken place in this sample and all others corresponding to the flat portion of Figure 2.2.

The influence of substrate characteristics on the wettability by a liquid has been examined (Ref. 2.19). Figure 2.5 demonstrates the influence of surface roughness on the contact angle. The ratio between the real (θ) and apparent (θ') contact angles has been utilized by Wenzel to define a surface roughness factor:

$$r = \frac{\cos \theta'}{\cos \theta} \quad [2.14]$$

In accordance with the basis of derivation of such a factor, apparent contact angles should increase with r for $\theta' > 90^\circ$ (Refs. 2.20 and 2.21). The influence of the crystallographic orientation of the substrate on the contact angle is

demonstrated by the curves of Figure 2.6. Since our plans call for the utilization of polycrystalline substrates, such information is of no direct use.

However, the dependence of contact angles on the degree of preferred orientation of the grains may very well be a factor. Preferred grain orientation can result from the anisotropic rolling of the material in the fabrication process.

The wettability of solids by liquid lithium has not been extensively investigated. Of the few reported investigations, those of Achner (Ref. 2.22), Jordon and Lane (Ref. 2.23) and Taylor (Ref. 2.24) are useful sources of information. The surface tension and contact angle of pure lithium and lithium containing nitrogen or oxygen impurities were determined by Achner. The maximum bubble pressure method was used for surface tension measurements and contact angles were determined by the dip tube method. From the capillary rise in the 304 stainless steel tube, the contact angle is calculated in accordance with:

$$\cos \theta = \frac{\rho g h r}{2\gamma} \quad [2.15]$$

where g is the gravitational acceleration, h is the height of the capillary rise, r is the radius of the dip tube and γ is the liquid-vapor surface tension (in this analysis γ_{SV} and γ_{SL} are ignored). Temperature ranges within which contact angle measurements were made were 210-617°C for pure Li, 240-514°C for lithium containing two concentration levels of nitrogen and 220-545°C for lithium containing 3000ppm of oxygen. Achner observed that both surface tension and contact angles varied with temperature and N and O content. In the case of pure lithium complete wetting (contact angle of 0°) occurred at about 200°C above its melting point, when a clean dip tube was used. However, if the dip tube was exposed to the atmosphere after cleaning a contact angle of zero was never attained. Figure 2.7 demonstrates these results. The presence of nitrogen had a significant effect on the wettability of lithium. Figure 2.8 shows that even at the highest

experimental temperature the lowest contact angle measured was about 45°. Furthermore, changing the nitrogen concentration from 1040 to 2000 ppm had no discernible influence on the wetting characteristics between lithium and niobium.

Taylor (Ref. 2.24) determined the surface energy and its temperature coefficient for pure lithium in the temperature range ~200-500°C. Surface energies were determined by adopting the maximum bubble pressure technique. Subsequently Bohdanský and Schins (Ref. 2.25) used the above-mentioned technique to determine surface energies of lithium at significantly higher temperatures, 1200°-1600°K.

Jordan and Lane (Ref. 2.23), in their investigations on the wetting of various metals by liquid alkali metals, considered the copper-lithium system. They used the vertical plate balance method to measure surface tension and then calculate contact angles. Complete wetting between liquid lithium and copper was attained at 250°C. In contrast, copper substrates could not be completely wetted by liquid sodium or potassium. The role played by the relative stabilities of the substrate and the liquid metal oxides is an interesting consideration at this point. Relative to the oxide of copper, Li_2O is more stable than the oxides of sodium and potassium. In other words, the reaction

is energetically more favorable than either

or

Calculated standard Gibbs free energies at 473°K for the reaction represented by Eqs. [2.16] through [2.18] (-91.8, -47.2 and -51.8 kcal/mole, respectively)

are in agreement with this conclusion. Using such an approach, the presence of oxides on niobium is judged as detrimental in terms of its wettability by lithium. If we consider the reaction:

a $\Delta G^\circ_{500^\circ\text{K}}$ of approximately -247 kcal/mole Nb_2O_5 is calculated. However, to make a meaningful comparison with the values calculated for Eqs. [2.12] through [2.18], this number must be divided by five and hence a resulting value of -49.4 kcal is not significantly different from that given for the $\text{Cu}_2\text{O/K}$ system. Since the oxidation behavior and the resulting phase for niobium is not clearly understood, it is instructive to examine NbO as an alternative for a calculation similar to that presented above. Unfortunately, high temperature thermodynamic data for NbO are not available. Based on the reaction:

a ΔG°_{298} of -37 kcal/mole NbO was calculated, with a more negative value anticipated at higher temperatures.

The concept of misfit energy arising from atomic differences between the liquid metal and the solid substrate at the liquid-solid interface, has been shown to give a correlation with wetting for numerous metals with liquid alkali metals. Jordon and Lane (Ref. 2.23) showed that as the ratio between the atomic radii of the liquid to solid metals exceeds an approximate value of 1.4, complete wetting is not attained. A corresponding ratio for Li/Nb was calculated to be approximately 1.1. Thus, presumably if clean Nb surfaces can be prepared, they will be wetted by liquid lithium. It is our feeling, however, that while such a correlation may be useful in a general way, it lacks the fundamental approach necessary to make understandable predictions. In other words, the interaction between metallic atoms must be viewed and analyzed from an electronic energies point of view. The Engel-Brewer theory and the proposals of Samsonov

(Refs. 2.26 and 2.15) are examples of the needed approach. Unfortunately, however, the Engel-Brewer analysis has been limited to interaction between transition metals only.

The influence of surface strain, resulting from the preparation and polishing of the substrate has been cited by others (Ref. 2.23) as the reason for the high degree of scatter in contact angle determinations. In the work of Jordan and Lane (Ref. 2.23), no attempt was made to anneal the specimens prior to their use as substrates. The work on Ga(L)/GaAs(s) by Kashkooli and Munir (Ref. 2.19) had also demonstrated the significant influence of the surface strain on contact angles and hence wettability. It was necessary to dissolve the strained surface layer by chemical etching. Such a technique may also be necessary in the planned work on Li(L)/Nb(s).

The solubility of niobium in lithium is another parameter which enters the present considerations. The resulting changes in the surface energy of lithium are a function of concentration and the nature of the binary diagram between these metals. Unfortunately no phase diagram data are available and reported solubility data are at higher temperatures than those planned for this study (Refs. 2.27 and 2.28). The other side of this Li/Nb interaction is the susceptibility of Nb to liquid lithium attack. The key factor according to experimental observations (Refs. 2.29-2.31) is the oxygen content of the niobium. If the oxygen content is below 50 ppm, lithium and niobium are compatible up to 1200°C.

3. REVIEW OF THE LITERATURE ON THIN FALLING FILMS

3.1 Nondimensional Groups

There is a great deal of information on falling films, but primarily for water. In this report we shall only highlight the historical background of falling film studies and the key ideas presented that relate to our specific application. A detailed survey of falling film research was presented in 1964 by Fulford (F-1).

As in any field of fluid mechanics there has been a development of dimensionless groups that can be used to characterize the flow. A dimensional analysis has shown that three of the key dimensionless parameters are the Reynolds, Weber and Froude numbers. A fourth dimensionless parameter, the surface tension number N_σ , occurs often in recent literature and is a combination of the three previously mentioned groups. In this report the following definitions of these groups are used:

$$\text{Reynolds Number} \quad \equiv \hat{N}_{RE} \triangleq \frac{4 \bar{u} \bar{b}}{\nu} \quad 3.1A$$

$$\text{Weber Number} \quad \equiv N_{WE} \triangleq \frac{\rho \bar{b} \bar{u}^2}{\sigma} \quad 3.1B$$

$$\text{Froude Number} \quad \equiv N_{FR} = \frac{\bar{u}^2}{g \bar{b}} \quad 3.1C$$

$$\text{Surface Tension Number} \equiv N_\sigma \triangleq \sigma \left(\frac{2}{\rho^3 \nu^4 g} \right)^{1/3} = \left[\frac{0.20 \hat{N}_{RE}^{4/3} N_{FR}^{1/3}}{N_{WE}} \right] \quad 3.1D$$

The factor of 4 appearing in the Reynolds number corresponds to a Reynolds number based on the hydraulic diameter of an infinitely wide film. In many papers on falling films, one encounters the Reynolds number defined without the 4, but we adopt the use of the 4 since this form seems to pre-

dominate in the literature. However, to alert the reader and ourselves to the potential problem in comparing our values with others, we will adopt the symbol $\hat{N}_{RE}$ for our Reynolds number in this work.

One also encounters in the literature the Weber and Froude numbers defined as the square root of Equations 3.1B and 3.1C, respectively. However, the definitions used here are consistent with the forms that appear in the nondimensionalized equations of motion. The surface tension number represents the nondimensionalized fluid properties group and is very useful for the purpose of data presentation. An alternative fluid properties group used by Fulford (F-1) and others is $K_F = 2 N_G^{-3}$.

In addition to the above parameters, the surface inclination angle from the horizontal, β , is a fourth independent parameter. If waves are present, additional dimensionless parameters such as the wave number and amplitude ratio appear in the problem. To the theoretician these are often independent parameters, while to the experimentalist they are functions of the four normally independent parameters $\hat{N}_{RE}$, N_{WE} (or N_G), N_{FR} and β .

3.2 Qualitative Description of Falling Film Flow

The flow in thin liquid films is a special case of flow in which gravity, surface tension and viscous effects are all significant. In order to have a feeling for the nature of the flow, the case of the fully-developed flow of water on a vertical surface will be described qualitatively in terms of the flow rate (or equivalently the Reynolds number) in the next section. Then a brief description of the spatial development of the thin film flow down the plate will be given.

3.2.1 Fully-Developed Thin Film Flow on a Vertical Plate

After the thin film has had a chance to fully-develop, usually after many centimeters or meters of flow from the entrance region, its character is completely determined by the flow rate for a given fluid. The mass flow rate per unit film width, Γ , is related to the Reynolds number by:

$$\hat{N}_{RE} = \frac{4 \Gamma}{\mu}$$

At very low flow rates (up to a Reynolds number of perhaps 15 for water), the film surface appears completely smooth to the eye. There may be an occasional small random disturbance, usually in the form of a dimple, but it is damped out in the flow direction. This regime is called the smooth laminar flow regime.

At slightly higher flow rates small, symmetrical regular waves appear. The wave fronts are fairly straight and perpendicular to the direction of flow. The appearance of periodic waves on the surface has been called the wavy laminar regime (F-1). At larger flow rates the regular, symmetrical waves tend to become less regular, and the cross section of the waves assumes the nonsymmetrical shape with a steep front and a long gentle sloping tail. Fulford refers to these waves as roll waves. These roll waves are frequently preceded by a number of small waves, called push waves, which move as a group with the mean wave.

There is really no sharp division between these two wavy flow regimes and they are sometimes grouped together and referred to as the transitional flow regime between smooth laminar and fully turbulent film flow. Shown in Fig. 3.1 are some time traces of water waves recorded by Dukler (D-1)

at four Reynolds numbers at a distance of 2.135 m from the entrance. In Fig. 3.1a, steep-fronted, nearly-periodic, symmetrical waves characteristic of the fully-developed wavy laminar flow regime can be seen ($\hat{N}_{RE} = 213$). There are often regular small waves between the large waves even in this flow regime. As the Reynolds number is increased the wave amplitude tends to increase. In Fig. 3.1b, the appearance of the steep-fronted roll waves preceded by the small amplitude push waves is evident. (These two wave types can be seen even more clearly in Fig. 3.2, to be discussed later). This push wave phenomenon is due to the fact that the radius of curvature at the base of the steep leading edge of the roll waves is small enough to lead to the formation of surface-tension-dominated ripples. When there is a complex mixture of large roll waves and smaller waves this is often referred to as the wavy turbulent flow regime. However, it is usually very difficult to distinguish the transition between wavy laminar and turbulent flow regimes.

As the Reynolds number is increased further, the wave fronts are no longer always straight but show a tendency to form bulges and to split and overtake each other; this is beginning to be evident in Fig. 3.1c.

As the liquid flow rate is increased a stage is reached where the roll waves and their accompanying push waves coalesce and become randomly mixed, as depicted in 3.1d. This random mixing is to such an extent that the individual wave fronts can scarcely be distinguished, and the surface appears to be covered with a mass of small and large jagged waves. The waves in this region, due to their randomness, are called turbulent waves. This final regime is called the fully turbulent film flow regime.

There is a lack of agreement among experimentalists regarding the values of the transition Reynolds numbers between the various film flow regimes (and even the names of the regimes). This is due to at least two factors. First, the transitions in film flow are not expected to be as sharp as in pipe flow due to the fact that the laminar sublayer of the film will occupy a rather large fraction of the total film thickness even at large flow rates (F-1). Secondly, the recorded character of the waves is quite sensitive to the laboratory setup, measurement techniques, recording instruments and to the extraneous disturbances and vibrations (see, e.g., P-1). (This latter effect may account for some of the differences in the wave profiles of Figs. 3.1a and 3.2b which are quite similar Reynolds numbers).

In addition, as we stated in Section 3.1, the Reynolds number is only one of at least four independent parameters required to specify the behavior of a thin falling film. If we restrict ourselves to water films on a vertical surface ($N_G = 4280$ and $\beta = 90^\circ$), we can provisionally define the various flow regimes as indicated on Fig. 3.3, based on the Reynolds numbers where there appear to be important changes in the slopes of various film flow parameters such as the mean film thickness, the mean wave speed and so forth.

3.2.2 Development Lengths in Wavy Film Flow

In nearly all of the observations of falling film flow, a smooth region near the liquid inlet has been noted at all flow rates. The length of the smooth entrance condition varies with the flow rate and with the

manner in which the liquid is introduced. In general, the length of the smooth zone is usually only a few centimeters long for water, and increases as the flow rate is increased, but the increase is not in direct proportion to the flow rate ($F-1$).

Fulford proposes that this entrance regime is a potential-like flow with the boundary layer growing in thickness. Once the boundary layer reaches the free surface its vorticity is sufficient to trigger the wave disturbances, which propagate or not, depending on whether the flow is unstable or stable. Thus, one observes the dimples dissipating in the smooth laminar flow regime while the onset of waves is observed at this point at higher Reynolds numbers.

At the onset of observed waviness a small disturbance region exists where the waves are small in amplitude and fairly periodic. In Figs. 3.4a, b and c, Pierson has photographed waves characteristic of this region ($P-1$). There is a little randomness, but one can visualize a periodic solution to the equations of motion using small disturbance theory which would fit the observations. The linear growth region in water is typically of the order of a few centimeters long at moderate flow rates.

The next region is characterized by a nonlinear growth of the waves. The length of this region is highly sensitive to external disturbances due to its unstable nature. Fig. 3.2 gives an indication of the changes which can take place in this region. The shape of the waves has changed drastically from 3.2a to 3.2b. Furthermore, there are important stochastic features that are now evident. The wave amplitude, while not as regular as before, is still about the same for all waves. But note that the separa-

tion distance or time between waves, the slope of the front and the slope of the back, all vary significantly for different waves. The nonlinear growth region can extend for many centimeters up to a few meters.

Following the nonlinear growth region, the fully-developed flow region occurs, where changes of the statistics of the wave profiles with length are small. This is the region where the measurements of Fig. 3.1 were taken.

The complicated flow patterns described above for falling thin films indicate the existence of potentially undesirable characteristics as far as our laser-fusion application is concerned. Ideally, we would want to produce and maintain a thin film of liquid lithium without any waves on the surface. In this way we could insure that the entire surface would be covered by a known thickness of lithium, which would enable us to make accurate predictions on the energy transfer characteristics of the lithium during the pulse cycle.

Unfortunately the description of falling film flows indicates that some waves at the surface will almost certainly occur. Thus, we need to develop scaling laws, based on the nondimensional groups, to predict what the surface characteristics of liquid lithium films might be at various flow rates and surface inclination angles. We can then decide if there are regions where the film thickness might be sufficiently uniform to be attractive for the laser-fusion application.

In order to develop scaling laws for thin falling films, we will examine the theoretical results published in the literature first. A comparison with experimental results will be made wherever possible. Then using these

results, estimates of falling lithium film characteristics will be made. The emphasis in the following sections will be on the lower Reynolds number flow regimes, principally the smooth and wavy laminar regimes, because of the greater attractiveness of these more uniform films to the laser-fusion reactor application.

3.3 Smooth Laminar Flow Regime

Nusselt ($N = 2$) originally developed the equations for the fluid mechanics and heat transfer of falling thin films by applying a force balance on an element of fluid. He assumed viscous flow with no shear or wave motion at the surface (i.e., smooth laminar flow). Thus, the only parameter needed to describe the geometry of this laminar falling film is the film thickness, which is uniquely determined by the flow rate for constant film properties and angles of inclination. His equation for the film thickness, b_N , with a modification to include the angle of inclination from the horizontal, β , is:

$$b_N = \left(\frac{3 \mu \Gamma}{(g \sin \beta) \rho^2} \right)^{1/3} \quad 3.2A$$

$$= \left(\frac{3 \nu^2 \hat{N}_{RE}}{4 g \sin \beta} \right)^{1/3} \quad 3.2B$$

The experimentalists attempted to obtain verification of Equation 3.2. However, the inception of wavy motion of the falling water films made these measurements very difficult, except for creeping or near-horizontal flows. Ultimately, excellent agreement between the measured and the predicted Nusselt film thicknesses has been obtained in the smooth laminar flow regime ($\hat{N}_{RE} \lesssim 25$).

Using the Nusselt film thickness in place of b in Equations 3.1A to 3.1C, it is possible to uniquely relate the Froude and Weber numbers to the Reynolds number for a given liquid:

$$N_{FR}(b_N) = \frac{\hat{N}_{RE}}{12} \sin \beta$$

$$N_{WE}(b_N) = \frac{(\hat{N}_{RE})^{5/3} \sin \beta^{1/3}}{11.5 N_G}$$

These relationships are strictly valid only for the smooth laminar flow regime. However, it will be shown in the next section that the average film thickness in the wavy laminar and wavy turbulent flow regimes up to Reynolds numbers of as high as 800 to 1000 are only slightly smaller than the Nusselt film thickness. As a result, the relationships among the nondimensional parameters retain their significance up to these Reynolds numbers. This will be shown to have great importance with regard to the interpretation of the observed characteristics of the waves in the transition regime.

The Froude and Weber numbers are plotted as a function of the Reynolds number for water and lithium in Fig. 3.5a. The corresponding Nusselt film thicknesses for vertical film flow are plotted in Fig. 3.5b.

3.4 Onset of Wavy Flow

In 1948 Kapitza (K-1) developed the pioneering work in stability analysis of thin films. By introducing a small perturbation to the equations of motion that included surface tension, Kapitza showed that in the small disturbance region the mean film thickness for waves at the surface was slightly smaller than the case of no waves. His equation for the mean film

thickness, $\bar{b}_K$, is

$$\bar{b}_K = \left(\frac{2.4 \mu \Gamma}{\rho^2 g} \right)^{1/3} \approx 0.93 b_N \quad 3.3$$

His analysis yielded the result that if the mean thickness with waves were smaller than without waves, then the wave state would be the more stable state. A necessary condition for wave inception developed by Kapitza was that the Reynolds number must be larger than a critical Reynolds number calculated from:

$$\hat{N}_{RE1} = 2.44 \left[\left(\frac{\mu^4 g}{\rho \sigma^3} \right) \sin \beta \right]^{-1/11} \quad 3.4A$$

$$= 2.29 [N_\sigma^{-3} \sin \beta]^{-1/11} \quad 3.4B$$

For a vertical falling water film this relation predicts the onset of waves at a Reynolds number of about 23. Kapitza's analysis was valid only for long wave-length waves where $\lambda/\bar{b} \geq 13.7$.

It should be noted that for such long wave-length waves, the gravity forces would tend to be dominant. It can be seen from Fig. 3.5a that for $\hat{N}_{RE}$ of about 23, the Froude number is almost unity. However, the surface tension effects evidently still exert a crucial influence even though the Weber number is only about 10^{-3} . This influence of the surface tension forces may be explained by the approximate method of solution of the equations of motion developed by Javdani and Goren (J-1). This method obtains solutions for the stream function, Ψ , expanded as a power series in wave number, α , where:

$$\alpha \triangleq \frac{2 \pi \bar{b}}{\lambda}$$

This method of solution generates terms in the equations with coefficients proportional to α^2/N_{WE} . Thus the importance of surface tension terms is related to α^2/N_{WE} , and for wave numbers as small as 0.03, this ratio is on the order of unity for N_{WE} only 0.001.

The variation of Kapitza's predicted wave-inception Reynolds number with water surface inclination angle, β , is shown as curve 3 on Fig. 3.6 (taken from Fulford's review paper, F-1). Two other theoretical predictions by Benjamin (curve 1) and Ishihara (curve 4) are also shown. Benjamin predicts the onset of waves at:

$$\hat{N}_{RE_1} = \frac{10}{3} \cot \beta$$

Thus, Benjamin's simplified theory as well as Ishihara's more refined version lead to the conclusion that the film is unstable to waves at all Reynolds numbers for vertical film flow ($\beta = 90^\circ$).

The last theory, curve 2, is based on the simple idea that if the waves are gravity-dominated at the onset of waviness, they should first appear at Froude numbers on the order of unity for a vertical surface; from Fig. 3.5a this implies a wave-inception Reynolds number of about 12. The equation for curve 2 for arbitrary surface inclination is simply:

$$\hat{N}_{RE_1} = \frac{12}{\sin \beta}$$

The data for water show fair agreement with both Kapitza's curve 3 and with curve 2.

The more recent theoretical work (e.g., L-1, J-1 and F-1) tends to agree with the predictions of Benjamin (B-1) that the flow is unstable to waves at all Reynolds numbers. However, the amplification ratio of the

unstable waves initially is so small that they are not observed until a Reynolds number of 25 or so. Thus, in recent literature the Reynolds number for the inception of waves as measured visually or by ordinary sensors has been termed the "visual" wave inception Reynolds number. More refined measurement techniques such as laser interferometry on longer plates could possibly detect waves at even lower Reynolds numbers if it were possible to create an absolutely disturbance-free environment for the falling film measurements.

Based on Kapitza's results (Equation 3.3), we have estimated the visual wave inception Reynolds number for lithium at 500°C. This is shown as the dashed curve 5 on Fig. 3.6.

Even though the concept of a critical Reynolds number for wave inception does not appear to be correct, we believe the work by Kapitza does show an appropriate trend. For example, a higher surface tension number should cause the growth rate of the instability to be less (L-1). Thus, we would expect the observed waves in lithium with $N_G \approx 67,840$ to be produced at a higher Reynolds number than for water, where $N_G = 4280$.

A lower limit on the visual wave inception for lithium may be the Froude number criterion, curve 2. These estimates are the best we can do at present in trying to scale the water experimental observations to lithium for the onset of wave flow.

3.5 Average Film Thickness

Early experimental results did not appear to agree with Kapitza's predictions for the mean film thickness, $\bar{b}_K$, near the onset of waviness. However, according to Fulford (F-1), more recent results are in reasonably

good agreement with Equation 3.3 over a limited range of Reynolds numbers near $\hat{N}_{RE_1}$.

In order to estimate the mean film thickness at higher Reynolds numbers, Dukler and Bergelin (D-2) proposed in 1952 that the thin film mean velocity profile could be approximated by "universal" velocity profiles for the laminar sublayer, a buffer zone and a turbulent zone. Using the Von Karman universal velocity profile for this latter zone, in analogy with pipe flow, they integrated these velocity profiles to obtain mean film thicknesses. An improved version of this model was published by Dukler in 1960 (D-3). The mean film thickness parameter, N_T , obtained from this improved model is shown plotted in Fig. 3.7a (F-1). N_T is defined as follows:

$$N_T \triangleq \frac{\Delta \bar{b}}{h_N} \times \left(\frac{3 \hat{N}_{RE}}{4} \right)^{1/3} \quad 3.5$$

This parameter is also useful for comparing various liquids.

Also shown in the figure are the Nusselt and Kapitza film thickness lines. The water data can be seen to agree reasonably well with Dukler's "smooth" turbulent film flow model over a Reynolds number range from about 200 to several thousand. At very high Reynolds numbers, the data systematically fell above the Dukler curve and were matched better by a semi-empirical equation of Brotz (F-1):

$$N_T \approx (3.2 \times 10^{-4} \hat{N}_{RE}^2)^{1/3}$$

At lower Reynolds numbers, Fulford states that modern data approaches the Kapitza line and then at still lower Reynolds numbers tends back toward the Nusselt line. It can be seen that the Dukler curve goes smoothly into the Nusselt line at low Reynolds numbers.

In Fig. 3.7b, Fulford has compiled all the experimental data he could obtain in 1964, and several empirical and theoretical curves to predict the film thickness. As one can see from this figure, many of the early film thickness data which were obtained mainly by the micrometer or simple drainage techniques were not of a high degree of accuracy. However, the figure indicates that the bulk of the data fell between the Nusselt and the improved Duklar curves over the Reynolds number range shown.

3.6 Wave Characteristics in the Initial Regions

The smoothest regions of a falling film are generally the entrance region and the small disturbance region immediately following it (e.g., see Figs. 3.4a, b and c). As indicated previously, these two regions are on the order of several centimeters long for water. We believe, as does Whitaker (W-2), that due to the increased surface tension of lithium, the characteristic length of these two regions will be greater than for water. Thus, the combined length may be as much as 5-10 cm for lithium. This region is of particular interest for the laser fusion application, as mentioned previously, because the lithium film would have the greatest uniformity of thickness.

A collection of experimental results for the visual wave inception line for water is shown in Fig. 3.8. Some attempts have been made using linear stability theory to predict the length of this smooth entrance zone for fluids other than water (e.g., C-1, C-2). However, as Atherton and Homsy (A-2) have indicated, the visual wave inception line is a function of the amplitude of the waves detectable by a given device, and is to that extent an ambiguously defined parameter. The authors compared the attempts made to predict this

length with experimental data which showed fairly poor agreement. They further concluded that the wave onset is basically a nonlinear phenomenon whose description needed a nonlinear hydrodynamical treatment (even though linear theory gives good results for the wave speed and wave length).

Thus, there are no scaling laws or equations to enable us to predict the characteristic lengths of the smooth acceleration and small disturbance regions. However, by examining the wave characteristics (wave speed, wave number, growth rate, and equilibrium amplitude) it is possible to indicate trends observed in water and hopefully extendable to lithium by analogy. All these wave parameters except for the wave amplitude and the development lengths are, in principle, obtainable from linearized stability theory. The method of solution until recently has been to allow for a small perturbation to the equations of motion and then to assume a periodic solution to the resulting perturbation equation. Thus, the solutions are valid only in the small disturbance (periodic) region. This method was initially developed by Kapitza (K-1). He also employed a physical argument related to minimum energy dissipation to estimate the equilibrium wave amplitude.

Lee (L-1) extended Kapitza's work and added mathematical rigor. In the small disturbance region he assumed a periodic solution of the form:

$$\phi = \left(\frac{a_{\max}}{b} \right) \sin(\omega x) = A_{eq} \sin(\omega x) \quad 3.8$$

$$\text{and found that: } A_{eq} = (3 N_{WE})^{1/2} \quad 3.9$$

where A_{eq} is the nondimensional wave amplitude. Fig. 3.9 shows a comparison of theory with experiment; the solid curve line is Equation 3.9. Thus, it appears that Lee's work represents the lower limit of experimental data. Kapitza's original solution yielded a constant value of $A_{eq} = 0.46$.

For the low flow rates associated with reasonably long small-disturbance regions, the Nusselt equation (3.2A) for the film thickness is fairly accurate. Using this equation and the definition of the Reynolds number, 3.1A, we can transform the Weber number (for $\beta = 90^\circ$) as follows:

$$N_{WE} = \frac{\hat{N}_{RE}^{5/3}}{14.5} \left(\frac{v^4 \rho^3 R}{\sigma^3} \right)^{1/3} = \frac{\hat{N}_{RE}^{5/3}}{11.5 N_G} \quad 3.10$$

thus:

$$A_{eq} = 0.5 \frac{\hat{N}_{RE}^{5/6}}{N_G^{1/2}} \quad 3.11$$

Again, this result is valid only in the small disturbance region. If we include the dependence of the film thickness on the angle of inclination from the horizontal, β , we get:

$$A_{eq} = 0.5 \frac{\hat{N}_{RE}^{5/6}}{N_G^{1/2}} (\sin \beta)^{1/6} \quad 3.12$$

In Figure 3.10a we have calculated values using Equation 3.11 for water at 20°C and lithium at 500°C . We note that the increased surface tension of lithium produces a decrease in the predicted equilibrium amplitude by more than a factor of 2 for a given Reynolds number.

In order to show the effect of the angle of inclination, β , on the wave amplitude, we have calculated two curves for water from Equation 3.12 as shown in Figure 3.10b. Unfortunately, the $(\sin \beta)^{1/6}$ term does not give us much help until we get to angles less than 30° . However, the trend does indicate that a small decrease in the wave amplitude is possible if the plate inclination angles can be reduced to small values.

The range of validity for this method of predicting the wave amplitudes in the small disturbance region is probably limited to Reynolds numbers less than 100 to 200. In order to obtain estimates of the wave amplitudes at higher Reynolds numbers, we must look to the experiments. Dukler (D-1) has obtained experimental results up to a Reynolds number of 6000 as shown in Fig. 3.11. Looking only at the upper curve for a gas Reynolds number of zero (i.e., no gas flow above the liquid film), we see that the dimensionless wave amplitudes seem to "saturate" at an R.M.S. value of about 0.7.

We have now extended Kapitza's small disturbance theory about as far as possible. To obtain theoretical results for higher Reynolds numbers requires the solution of the Orr-Sommerfeld equation, in general (e.g., Whitaker, W-3). Even this powerful method cannot yield any information on equilibrium wave amplitudes or the length of the linear wave growth region, since these wave characteristics must come out of a nonlinear analyses.

Pierson (P-1) has recently solved this equation numerically for surface tension numbers from 0 to 7000. In Fig. 3.12, we present Pierson's curves for the dimensionless wave number for the maximum growth rate, α_m , versus Reynolds number at different surface tension numbers, where

$$\alpha_m = \frac{\Delta}{\lambda} \frac{2 \pi \bar{b}}{\lambda}$$

We have included our extrapolation of his results to lithium, where

$$N_G = 67,840 \text{ at } 500^\circ\text{C} \quad .$$

In Fig. 3.13 are shown the predictions of two other theories reported in Pierson (P-1). There is remarkable agreement between Pierson and Anshus and Goren (A-1). This agreement is startling at first as Anshus and Goren

developed approximate solutions to the Orr-Sommerfeld equation by neglecting the parabolic velocity profile in the fluid. The authors' a priori assumption was that surface effects were governing so that the velocity profile could be replaced by the surface velocity. The good agreement achieved with the more rigorous solutions of Pierson is clearly evident in Fig. 3.13 and seems to confirm that surface effects are controlling. Thus, it is reasonable to expect that the higher surface tension number fluids would be stabler than the ones with lower N_Q .

Figure 3.14 from Pierson shows the excellent agreement between his solution (and that of Anshus and Goren) with the available data on water up to the unexpectedly high Reynolds number of almost 4000. This good agreement is further verified in Fig. 3.15 where the nondimensional wave speed (relative to the mean surface velocity), C_r , is plotted versus nondimensional flow rate.

Pierson also presents the nondimensional spatial growth rate parameter, α_1 , as a function of the nondimensional wave number α_r , for water, where:

$$\alpha_r = \frac{\Delta}{\lambda_r} \frac{2 \pi \bar{b}}{\lambda_r}$$

This is reproduced in Fig. 3.16. The wave number of maximum growth rate should be a good indication of the wave which is observed and this is corroborated for water by the data of Fig. 3.14. It also gives some qualitative indication of the length from the entrance where waves will first be observed. The visual measurements of the wave inception line shown in Fig. 3.8 seem to verify that the lower growth rates for Reynolds numbers above and below about 200 do result in a longer inception distance.

The data in these figures represent the type of results which have been published to date on numerical solutions of the Orr-Sommerfeld equation for thin falling films. Unfortunately, it is not sufficient to develop a complete set of analytical scaling laws applicable to lithium.

Before we discuss the requirements for complete surface wetting we will discuss very briefly the experimental approaches to obtaining statistical data on wavy films, particularly on the higher Reynolds numbers.

3.7 Recent Experimental Wave Data

In the recent work of Dukler and a few others, the older experimental approaches aimed primarily at measuring the mean film thickness, average wave length and average wave speed, have been superseded by more detailed statistical analysis of the wavy surface. Due to the random waviness of falling film surfaces at Reynolds numbers above a few hundred, these statistical data are felt to be essential for modern heat and mass transfer studies. This is particularly true in the wavy turbulent flow regimes ($N_{RE}^* > 300$) where many practical applications are found. This statistical analysis can only be accomplished using high quality experimental data on the instantaneous film thickness.

A relatively simple but powerful experimental technique developed by Wicks (W-4) using conductance probes, has enabled Dukler to obtain the wave profile traces as shown in Figures 3.1 and 3.2. If one considers the wave amplitude as a stochastic process, it can be characterized by its moments, calculated from the measured probability density.

Chu and Dukler (C-3) have recently presented a statistical analysis of the wavy films. They have shown that waves on falling films display certain

stochastic features. The authors found that at least two classes of such random waves exist; large waves which carry the bulk of the flow and smaller waves which appear on a sublayer film that exists between large waves. These two classes of random waves are illustrated in Fig. 3.17 and can be seen fairly clearly in Figs. 3.1b, c and d.

The wave speed data of Fulford (F-1) for water, shown in Fig. 3.18 seem to indicate a related phenomenon occurring at a specific Reynolds number. It can be seen that the wave speed slope seems to change abruptly at a certain Reynolds number. This Reynolds number corresponds to a Weber number of about 0.8, indicating that small capillary waves are becoming important. Thus the existence of two classes of waves, one class composed of the longer-wave-length, gravity-dominated waves, and the other class composed of short-wave-length, surface tension-dominated waves, appears to be reasonable above Weber numbers near unity.

The R.M.S. wave amplitude data shown in Fig. 3.11 are typical of the statistical results obtained by Dukler (D-1). They indicate that the wave amplitudes for water increase very rapidly between a Reynolds number of 100 and about 1000, and then tend to saturate. Thus, for our laser-fusion application, it appears that we may have to restrict ourselves to the low Reynolds number range of at most a few hundred in order to limit the liquid film nonuniformity.

3.8 Effects of Wavy Flow on Transport Phenomena

There have been many efforts made to examine the effect the wavy surface of a falling film has on transport phenomena (e.g., see D-1). Unfortunately, the concepts used in these works are for ordinary transport

mechanisms. The energy transport mechanisms involved in a pulsed fusion reactor cannot be compared to these works, due to the difference in time scales. For the most part the film will be standing still during the transfer process. One obvious effect of any waves on the surface would be to increase the effective surface area of the first wall coating. Since this present study is focused on the establishment and maintenance of the thin coatings of lithium before the fusion shot, much additional research will have to be done on the behavior of thin lithium films during and after the laser-fusion micro-explosion.

3.9 Minimum Wetting Rates

It is well known (H-1) that when a thin film flows over a solid surface under the action of a driving force, dry patches can form and spread. If the flow rate is increased, the dry patch can be rewetted. The flow rate necessary to cover a surface completely with a stable film is called the *minimum wetting rate*, $\Gamma_{\min}$. Experimentalists (N-1) attempted to develop empirical relationships to predict the minimum wetting rate. Unfortunately, these were valid only for the particular fluid or flow condition used, due to the dependence on contact angle.

Hartley and Murgatroyd (H-1) developed one of the first theoretical criteria for fluid breakup based on a force balance at the upstream stagnation point of a dry patch. The flow condition was a vertical isothermal film flowing under the influence of gravity on a solid surface. This force balance led to the following equation for the minimum wetting rate, $\Gamma_{\min}$:

$$\frac{\Gamma_{\min}}{\mu} = 1.69 \left[\frac{\rho g^3}{4\mu^8} (1 - \cos \theta)^3 \right]^{1/5} \quad 3.13A$$

$$= 1.47 [N_G (1 - \cos \theta)]^{3/5} \quad 3.13B$$

where θ is the contact angle. We note that $\hat{N}_{RE} = 4 \frac{\Gamma}{\mu}$ so that this equation specifies the minimum Reynolds number for a stable vertical film. Here stable refers to the absence of a dry patch.

Equation 3.13 is plotted for lithium at 500°C in Figure 3.19. Also plotted is an equation developed by Ponter et al. (P-2) for the minimum wetting rate where the only difference is that the constant is 1.116 instead of 1.69 in Equation 3.13A. It is noted that if these equations are correct and if the contact angle is large (e.g., 25° or larger), the Reynolds number required to produce a stable film on a vertical surface can be as high as 1000 or more. One consequence of this is that a continuous film might not be attainable in the smooth, low Reynolds number film flow regimes. However, there is some reason to believe that by preconditioning the substrate surface to cause diffusion of lithium into the metal substrate, that the contact angle can be reduced to low values.

Another consequence of Equation 3.13 is that there is a minimum film thickness, since it is specified uniquely by the flow rate for constant fluid properties and angle of inclination. Figure 3.20 shows a curve that represents the Nusselt film thickness for lithium calculated for the minimum wetting rates using Equation 3.2. We note that, unless the contact angle is smaller than about 25°, a film thickness less than 0.4 mm (400 microns) is unattainable (according to Equation 3.13) for a vertical plate.

Hobler and Czajka (H-2) measured minimum wetting rates for fluids flowing over a series of inclined planes. The functional dependence of the minimum wetting rate on the angle of inclination from the horizontal, β , was found to be (based on plots of their data as reported in W-1):

$$\frac{\Gamma_{\min}}{\mu} \propto (\sin \beta)^{2/3} \quad 3.14$$

In Fig. 3.21 we have included this angular dependence using Eq. 3.14. These curves give us the hope that it may be possible to produce a thin film at a relatively low flow rate for the surfaces at small angles of inclination.

Recently, Watanabe, et al., (W-1) have examined the predictions of the breakdown of falling films in the absence of mass or heat transfer. The authors showed that the experimental results often disagreed significantly in magnitude with Eq. 3.13, even though the trend was correct. The authors showed that differences of the top shape of the wetted surface at the entrance region could produce a significant difference in the minimum wetting rate. Thus, it seems reasonable to use a scaling law for the minimum wetting rate of the form:

$$\frac{\Gamma_{\min}}{\mu} = C_F [N_G (1 - \cos \theta)]^{3/5} (\sin \beta)^{2/3} \quad 3.15$$

where C_F is a constant determined experimentally for the flow conditions.

We hope to evaluate this constant experimentally for lithium. However to be able to use this scaling law, we will need the contact angle measurements provided by the other members of our research team.

3.10 Summary

As mentioned in the Introduction, we believed initially that falling films were preferable to climbing (i.e., capillary) films because of the

opportunity to use the rather large gravity forces. The literature search has indicated that it may be possible to work with falling films in spite of the complicated variation of the film surface with flow length, flow rate, surface inclination angle and so forth.

This literature search has indicated most of all the need for experimental data on the basic characteristics of falling liquid lithium films, since the literature has failed to produce a complete set of scaling laws that would enable us to make a decision on the use of falling lithium films. Thus, we intend to do some basic first experiments on falling lithium films with the goal of determining the conditions under which it may be feasible to use such films in future laser-fusion reactors.

4. DESIGN OF CONTACT ANGLE EXPERIMENTS

After examining literature accounts of the various methods reported for contact angle measurements, it was decided to develop two approaches for short- and long-range plans. A general outline of the various experimental considerations in contact angle determinations and a description of the two adopted plans will be given below.

Basically there are two general methods for determining the contact angles between liquids and solids (Ref. 4.1). In the first general method, knowledge of the surface tension of the liquid is essential. Eq. [2.15] in the second section of this report ($\cos \theta = g\rho h r / 2\gamma$) describes the relationship of θ to the height of the fluid's capillary rise, h . In addition to this approach, that of capillary pull can also be utilized in contact angle measurements. The difference in weight of a flat plate when in air, W_0 , and when partially immersed in a fluid can be related to the contact angle by

$$W - W_0 = 2(l + \delta)\gamma \cos \theta - l\delta z\rho g \quad [4.1]$$

where l and δ are the width and thickness of the plate, z is the depth to which the plate has been immersed, ρ is the density of the liquid, γ is its surface tension and g is the gravitational acceleration.

A more commonly employed general approach is the direct measurement of the contact angles. This can be accomplished through a variety of techniques. Perhaps the most popular is the measurement of θ from the contact periphery of a sessile drop. Use of telescopes for direct reading of θ , or the use of a camera to record the drop's image then measure the angle are two examples of the direct techniques. Calculations of the contact angles from known drop dimensions are also possible. Referring to Figure 4.1, the contact angle θ can be calculated from the height, h , and width, w , of the drop in accordance

with the geometric relationship:

$$h = R(1 - \cos \theta) \quad [4.2]$$

and since

$$R = \frac{w/2}{\sin \theta} \quad [4.3]$$

the ratio

$$h/w = \frac{\tan(\theta/2)}{2} \quad [4.4]$$

A similar relationship can be derived for obtuse contact angles.

Another less common technique is termed the level surface method. A partially immersed plate is moved around an axis parallel to the surface of the liquid and the angle is adjusted until the liquid surface remains horizontal all the way to the plate. At that point the angle between the plate and the surface of the liquid is the desired contact angle.

A variety of sessile drop techniques have been reported in the literature. Each reflects the differences in the desired conditions needed to accommodate the specific properties of the materials being studied. Furnace configuration, the capability of measuring trailing and advancing contact angles, and direct or photographic measurement of the contact angle are some of the characteristics of the different systems. The furnace designs vary considerably, depending on the particular requirements of the experiment. An important distinction is whether the entire furnace is in an evacuated chamber or whether the furnace surrounds the evacuated chamber.

The systems comprised of a furnace external to the evacuated system generally consist of a fused-silica tube containing the sample and substrate. The section of the tube to be heated is placed in the furnace and the sample can be observed from the ends. In this system, only the small glass tube requires evacuating, and any manipulation of the system can be accomplished directly. In other systems, the entire furnace and sample assembly is enclosed in an

evacuated chamber. When the whole system is enclosed, any manipulation or observation of the sample is more difficult since viewing ports and remote control devices are required.

Most furnaces examined had cylindrical shapes. This provides for symmetric heating and simple design. The tubes are long enough to provide a sizable region of constant temperature in the center and allow viewing through the ends. An assortment of other furnace designs comprised the remaining techniques examined. Several types of designs are shown in Figure 4.2 (Refs. 4.2-4.5).

If successive sessile drop experiments are to be performed, the ability to make several determinations without breaking the vacuum or inert atmosphere seal is highly desirable. Only one reported system had the provision for measurement of several drops sequentially (Ref. 4.6, Figure 4.3). The entire system was enclosed in an evacuation chamber containing a circular conveyor that accommodates 20-30 specimens.

After considering all of the criteria pertaining to the lithium-niobium couple, a bell jar type vacuum system was designed to perform the sessile drop experiment. This preliminary design was reviewed and a simpler one suggested for the initial set of experiments.

Our initial, short-range design will be directed towards individual sessile drop measurements. A stainless steel tube, housing the substrate, will be heated externally by a horizontal tube furnace. Provisions will be made for a viewing port on one end of the stainless steel tube and a vacuum coupling valve at the other. The tube will be first evacuated and back-filled with argon. It will then be taken into a glove box which in turn will be evacuated and purged with argon gas. Small pieces of lithium will be cut and shaped, then placed onto niobium substrates and finally inserted into the stainless steel tube. After closing the tube, it can then be removed from the glove box and inserted into the furnace. Following evacuation of the tube, contact angle measurements can

commence. Alternatively, various atmospheres (e.g., N_2 , O_2 , etc.) can be introduced at previously chosen partial pressures and the contact angles determined under these conditions.

Measurements of the angles of contact between liquid lithium and the niobium substrates will be made directly by means of a cathetometer equipped with movable cross hairs and a goniometer. For low value contact angles, photographic techniques may be utilized to ensure a higher degree of accuracy. The vacuum system which will be coupled with the tube in which contact angles will be measured is a modified bell jar apparatus. This apparatus, which utilizes an oil diffusion pump backed by a mechanical pump, is capable of providing a vacuum in the range of $2-5 \times 10^{-7}$ torr.

For the long-range design, we anticipate developing the capability of measuring sequentially a number of contact angles without changing the atmosphere of the experiment. The capability of rotating the substrate will also have to be developed. To accomplish these capabilities, a design has been made.

The final long-range design is a system that incorporates all of the required features and is entirely enclosed in an 18" bell jar. The furnace is a Ta cylinder (Figure 4.4) 10 inches long. The lithium samples can be added from the dispenser shown in Figure 4.5 to the substrate which is a 4" long niobium strip. For successive tests, an unused portion of the strip can be positioned under the loading tube with the mechanism shown on the left side of Figure 4.4. The substrate can be tilted with the mechanism on the right side of this figure. By tilting the substrate beyond the critical angle, the old drop can be forced to roll to the side. Because of the sophisticated mechanical linkages needed to perform the required functions from the outside of the bell jar, the simpler interim design described has been adopted.

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5. REQUIREMENTS FOR THE INITIAL FALLING FILM EXPERIMENTS

5.1 Liquid Metal Alternatives

The literature review on falling films discussed in Section 3, has indicated the need for experimentation to adequately verify and extend the scaling laws presented. In order to verify these scaling laws, the basic experiment must be designed to give us data on the following parameters for a fluid with a surface tension number significantly higher than that of water:

1. Film thickness as a function of flow rate and angle of inclination.
2. Characteristic Reynolds and Weber numbers at which waves appear and grow.
3. Characteristic lengths of the smooth acceleration region and the small disturbance region.
4. Minimum flow rate required to insure the existence of a stable film (minimum wetting rate).

The minimum wetting rate depends crucially on the contact angle, and consequently the interpretation of these experimental results requires prior knowledge of the contact angle for the particular liquid/metal-substrate combination.

Since reliable contact angle data for lithium are not yet available, we have given some consideration to the possible use in the initial experiments of another liquid where contact angle data are available.

In Table 5.1, we have listed the pertinent thermophysical data for some liquid metals. Examination of the calculated values of the surface tension number indicates that all of these liquid metals probably have values of N_G significantly higher than room-temperature water, where

$$N_G = 4280 - 4660 \text{ at } 20^\circ\text{C}.$$

This indicates that any of these liquid metals would be suitable for testing the scaling of the falling film equation with N_G . However, only limited contact angle data have been uncovered so far, mostly for sodium.

We have found some interesting data from two experimental programs which evaluated the contact angle of sodium on stainless steel (A-1, H-1). The experimental conditions were basically the same with close to identical results, even though the measurement techniques employed were quite different. The results are discouraging as can be seen from Figure 5.1. It appears that very high sodium temperatures are required to obtain the desired small contact angles. It is not clear from these reports whether preconditioning the substrate surface by allowing it to soak for a period of time in a pool of liquid sodium at high temperature would improve the low-temperature contact angles.

Although the use of sodium does not look promising, we are still exploring the possibility of using some other lower-melting-point liquid than lithium for the first liquid metal experiments. This would have several potential advantages. First of all, operation at lower temperature, preferably near room temperature, would simplify the experimental procedures required inside a glove box. The establishment of high purity conditions inside the glove box will be complex enough without the added complexity of the several heaters required to maintain the desired temperature distributions. Lower temperature operation should facilitate achieving either isothermal conditions or producing a controlled temperature gradient; once again, near room-temperature operation would probably be the best from this point of view.

Operation at the lower temperature would also enable us to use a relatively inexpensive Bently-Nevada proximito probe for film thickness measurements. This could provide us with a check on an optical reflection technique for measuring film surface smoothness which we plan to investigate. This optical technique is extendable to higher temperatures than the Bently-Nevada probes, and is relatively inexpensive but potentially of high accuracy. This technique is described further in Section 5.2.

We are continuing our investigation of various possible liquids to see if any one of them seems preferable to lithium for the high surface tension number experiments. In the meantime, we are designing a simple falling water film apparatus for use as an instrumentation test bed. This is described in the next section. It is even conceivable that we may be able to use hot water in place of a liquid metal for our high surface tension number experiments since

$$N_g = 16,250 \text{ at } 90^\circ\text{C} \quad (\text{See Table 5.1.})$$

5.2 Instrumentation Development Apparatus

In order to perform the liquid-metal thin-film experiments, it will be necessary to make measurements of the instantaneous film thickness, flow rate and fluid temperature distribution. Most of the fluid properties may be calculated with sufficient accuracy if the fluid temperature is known. Since it is known that heat transfer affects the minimum wetting rate, the temperature of the plate must also be controlled and measured accurately. As mentioned previously, we intend to design the system for both isothermal operation and for operation with a controlled temperature gradient.

The most difficult aspect of the diagnostics is the instantaneous film thickness measurement. We would like to use the simple but very effective conductance probe technique developed by Wicks (Sec. 3, Ref. W-4) and employed successfully by Dukler to obtain the water film profiles shown in Figures 3.1 and 3.2. However, the probes do not appear to be able to produce large enough output signals in a highly electrically conducting liquid such as lithium.

An alternate technique which is elegant but very expensive is laser interferometry. A typical laser system will give better than $1 \mu\text{m}$ accuracy but can cost \$25,000 which is well beyond our limited budget.

A survey of other available techniques suggested that we try to employ some form of light reflection from the wavy film surface rather than the more common

light refraction techniques, since lithium is an opaque liquid. We have tentatively chosen to investigate the use of a helium-neon laser light source reflected off the flowing liquid surface. The reflected light will be detected by an array of phototransistors or photodiodes. There seems to be no obstacle to using this light reflection technique at the relatively high temperatures encountered with liquid lithium.

In order to develop and calibrate this optical diagnostic technique, we are designing a simple but versatile falling water film apparatus. The basic reason for this choice is that the characteristics of falling water films are well known and thoroughly documented (as described in Section 3), particularly at the lower Reynolds numbers of greatest interest to the laser-fusion application. In addition, the apparatus can be outside of a glove box and at room temperature. One potential problem with the use of water films is that we will require a higher laser intensity than for lithium due to the relatively small percentage of light reflected off a water surface at angles of incidence less than about 70° .

The optical measurements and the theoretical predictions for water films will be backed up by the use of Bently-Navada proximitors; however, these probes are limited to temperatures less than about 90°C . These probes radiate an RF signal in the neighborhood of the probe tip and measure the displacement of any conducting medium in that neighborhood by the field attenuation.

There are several additional important problem areas which can be investigated by building a suitably flexible apparatus, including:

1. Techniques for isolating the apparatus from external disturbances.
2. Minimum usable plate widths and lengths for interpretable data.
3. Minimum run durations for interpretable data.

4. Behavior of the films for various plate inclination angles including angles greater than 90° (i.e., for films on the underside of inclined plates).
5. Variable N_G effects (e.g., by heating the water close to 100°C).
6. Effect of small temperature gradients and small departures from isothermal conditions.

All of this information is essential for the design of a minimum-size liquid metal experiment which can fit conveniently into a glove box. In fact, we are fairly certain that we will modify the falling water film apparatus to fit inside the glove box upon satisfactory achievement of the initial experimental objectives described above.

In the modified apparatus, the fluid system will consist of two reservoirs, one constant head tank to introduce the fluid onto the plate and one to catch the fluid after it flows over the plate. These tanks will be under pressure from an inert gas. To replenish the source tank, we will either pump the liquid from the receiving tank, or we will interchange the two tanks. The reservoirs and plate will have heaters in order to check out higher temperature operation. (Fig. 5.2).

To insure fairly isothermal conditions (when we operate above room temperature), additional guard heaters will be used. One will be placed below the plate to keep it at the desired temperature, and the other will be placed above the fluid to keep it at the proper temperature. Thermocouples will be attached to the plate to measure the plate temperature, and the temperature of the fluid will be measured in the two reservoir tanks.

The flow rates will be measured by flowmeters and/or by the simple measurement of fluid height in the dump tank as a function of time. It may be necessary to separate the flow in the middle of the plate from the edge dominated flow before measurements are taken.

The thermocouples and other instrumentation will be sheathed and put through electrical feed-throughs in the glove box and connected to the appropriate electronics outside the glove box.

Since it will take some time to build the water film apparatus, we plan to test the laser reflection technique using a wavy film analog in the meantime. We plan to construct solid wavy surfaces of amplitudes and wavelengths typical of the liquid films and then evaluate the accuracy possible with the measurement scheme. We will also search for the optimum angle of incidence and the optimum location of the photosensitive detectors using this analog.

Once the concept is tested and shown to be sufficiently accurate on the wavy film analog, we will test it on our water film apparatus. The procedure will be to first statically calibrate both the laser reflection measurement system and the proximator probes. Then we will make several experimental runs with water measuring the film thickness as a function of flow rate and the characteristic lengths from the entrance. These runs will hopefully verify the accuracy of our measurement systems when used on falling films.

After the initial water runs are shown to be reproducible and to compare favorably with other data, we will make several runs with the modified apparatus in the glove box to show that we can achieve the same results within the constraints of the glove box. Thus, when we begin to experiment with the liquid metals, we will have our procedures perfected as best we can.

6. SUMMARY

The present status and future plans for the research on the wetted first wall concept for future laser fusion reactors can be summarized as follows:

- (a) In order to ascertain the feasibility of the wetted wall concept, it is crucially important to generate reliable data on the contact angle between lithium and various solid substrates including niobium and stainless steel.
- (b) The few available contact angle data for lithium are not useable quantitatively because of their large scatter.
- (c) However, the qualitative trends shown by these data indicate a significant sensitivity of the contact angle to surface condition and impurity level in the lithium.
- (d) A simplified initial experiment to measure the contact angle of lithium on various solid substrates employing the sessile drop technique has been designed and is partially built.
- (e) The principal objective of these initial experiments is to establish a technique through which reproducible contact angle data can be obtained.
- (f) A second objective is to determine the substrate characteristics and lithium purity conditions which provide for maximum wettability of liquid lithium.
- (g) The ability of the lithium to wet the materials of the reactor first wall is only the first basic requirement which must be fulfilled. Another basic requirement is the ability to uniformly

cover the relatively complex geometry of the entire first wall using reasonably simple lithium feed port and collection port configurations.

- (h) The initial research on the coating of large areas of the first wall with liquid lithium has focussed on the use of thin falling liquid films.
- (i) This research has revealed many potential problems associated with the use of falling liquid films including:
 - 1) the probable occurrence of surface waves on the film after only a few centimeters of flow from the feed ports
 - 2) the uncertainty regarding the ability of falling films to coat the undersides of the surfaces near the top of the reactor
- (j) In spite of these potential problem areas, the use of falling films is still sufficiently attractive for large portions of the reactor first wall that it has been decided to design a falling film experiment.
- (k) The principal objective of the initial experiment will be to verify and extend the existing scaling laws for falling films to the higher surface tension numbers characteristic of liquid metals such as lithium.
- (l) In order to accomplish this objective the development of a relatively simple and inexpensive laser light reflection diagnostic system for measuring liquid film thickness and smoothness will be undertaken.

- (m) In parallel with the above experimental efforts, research will continue on the other possible methods for coating all or certain portions of the first wall with liquid lithium.

Appendix A

Cleaning and Operating Procedures

Bell Jar Vacuum System

Components and Assembly

The vacuum system referred to in the text was assembled from two incomplete facilities, donated by Fairchild Semiconductor Corporation. They are typical high vacuum bell jar systems with a diffusion pump, copper piping and pneumatic valves. By cannibalizing the two units, a complete vacuum was built with the exception of a mechanical pump and a bell jar. The main platform required some hand grinding around the bell jar seal and the high-vacuum valve seal. The high-vacuum valve and roughing valve required no repair. A new solenoid was installed in the vent valve and a new activation shaft was manufactured for the foreline valve.

The mechanical pump, obtained on a loan basis from LLL required cleaning and new oil. The new bell jar and seal was purchased from Van Waters and Rogers Corporation. The O-rings were obtained from Porter Seal Company and are made of Viton. An ionization gauge was purchased from Veeco Corporation.

During assembly, Dow's silicon vacuum grease was used exclusively in the system.

Cleaning

The basic parts of the present system had been in storage for some time, and needed a thorough cleaning. This was accomplished by first cleaning with soap and water. Next, a cleaning apparatus was set up consisting of three plastic trays filled with water, acetone and trichloroethylene, respectively. The acetone removed any remaining grease or oil which might have been left from the soap and water treatment. The trichloroethylene removed additional grease and also the film left by the acetone. Freon R-11 was used to remove the residue from the trichloroethylene. Varnish and scale from soldered parts were removed with concentrated HCl and, more successfully, with a nitric acid solution consisting of 40% nitric acid, 30% sulfuric acid, .5% HCl,

29.5% H_2O ; or 50% nitric acid and 50% H_2O . The acid bath was followed by rinsing with distilled water. Rubber gloves were used to handle the parts during the cleaning to protect the hands from the harsh chemicals and to keep skin oils from contaminating the cleaned parts. The ion gauge tube was cleaned by rolling a small amount of trichloroethylene, acetone and HCl around on the inside of the glass. Care must be taken not to allow the fluid to come in contact with the filament.

Operation Procedure

1. Before starting any part of the vacuum system, a check should be made to ensure that cooling water is flowing through the system. Checking the drain outlets of the cooling water lines is desirable since it is possible to turn on the water at the tap and have a blockage somewhere in the lines. In addition, for operations involving the bell jar, the high vacuum valve must not be opened until the pressure has been substantially equalized on both sides of this valve to within about 10μ at about the 20μ level or better.
2. After preliminary checks, the main breaker may be turned on. If the panel key is off the switches will display red. If the switches are in the on mode they will not be activated until the key is turned on. At this point, the protect button may be set after the high vacuum (Hi-Vac) valve is closed. (Caution: the protect button may be used to close the Hi-Vac valve, but will also open if the Hi-Vac button is not turned off,) Close all valves. (Note: turning the main breaker on with the panel key left in the on position causes a loss of any existing vacuum in the system, as valves will open if their switch has been left on.)

3. The mechanical pump may be turned on to evacuate the primary system between the roughing and foreline valves. Thermocouple gauge No. 1 (TC 1) should read below 50μ (usually about 10μ) before the foreline valve is opened and the diffusion pump is evacuated up to the high vacuum valve. Once the pressure in the entire system up to the high vacuum valve has stabilized at about 10 to 50μ , the bell jar may be evacuated if it is to be used. Check to be sure that the vent valve is closed. Close the foreline valve then open the roughing valve. When the pressure comes close to the pressure under the Hi-Vac valve, the foreline valve may be reopened to pump on the entire system.
4. The diffusion pump has very poor efficiency at pressures above 50μ . When the pressure in the entire system has stabilized below 50μ , the diffusion pump can be started. The lower the pressure in the system at which the diffusion pump is started, the faster the ultimate vacuum required can be attained. After turning on the diffusion pump, a warm-up time of about 45 minutes is required before it is fully effective. The Hi-Vac valve may be opened. It may take several hours to reach a vacuum of about 5×10^{-7} torr.
5. Before the system may be left unattended, recheck the cooling water drain to be sure that it is flowing properly.

Shut Down

1. Allow any heating facilities used in the bell jar to cool to room temperature.
2. Close the Hi-Vac valve.
3. Turn off the diffusion pump but continue pumping with the mechanical pump for at least 45 minutes.
4. After the diffusion pump has cooled, close all valves and turn off the mechanical pump.
5. If access is required to the bell jar, open the vent valve first.

Opening the Bell Jar

1. Close the Hi-Vac valve.
2. Close the roughing valve.
3. Open the vent valve.
4. Raise the bell jar - close the bell jar.
5. Close the foreline valve.
6. Open the roughing valve.
7. After at least 20 μ pressure has been reached in the bell jar, close the roughing valve and open the foreline valve.
8. Open the Hi-Vac valve.

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Appendix B

Laboratory Handling of Lithium and Safety Considerations

To prevent contamination of lithium by oxygen and nitrogen, lithium is usually contained in an inert atmosphere or in a vacuum. The recommended maximum impurity levels in inert gases for glove boxes in which high purity lithium is handled are as follows: water 5ppm, oxygen 10ppm, nitrogen 10ppm and carbon dioxide 10ppm. In vacuum boxes samples of lithium are found to remain clean for long periods of time at vacuum pressures of 10^{-3} to 10^{-5} torr.

Moisture diffusion through gloves in glove boxes is found to be a problem and this could be eliminated by the use of two layers of butyl rubber for the gloves. Continuous removal of nitrogen, oxygen and carbon dioxide from the inert gas in the glove box is necessary to maintain the purity of the lithium.

Glasses, plastics and ceramics are generally attacked by molten lithium near its melting point. Low carbon steels and iron can be used to contain lithium up to about 500°C and for temperatures above 1000°C refractory metals could be used.

The surfaces of apparatus in which lithium is to be contained should be thoroughly cleaned. Degreasing with trichloroethylene followed by several acetone rinses is recommended.

Since lithium is a highly reactive material, it can be dangerous if proper precautions are not taken. Lithium is generally stored in hydrocarbons, mineral oils, or in containers sealed with purified argon. Lithium is lighter than most mineral oils and floats on the surface when stored in this manner. Hence these storage vessels should be opened with caution. The major hazard encountered in the use of lithium are explosions or fires that could occur when lithium is in contact with water, air, chlorinated hydrocarbons or other reactive materials.

Those handling pound quantities of lithium in glove boxes should protect themselves by wearing fire-resistant aprons and safety glasses. Since molten lithium may burn with an intense flame when in contact with moist skin, containers

should be leak proof. Personnel handling containers with liquid lithium should wear safety gloves and chrome leather protective clothing.

Powdered graphite, zirconium silicate, potassium chloride and lithium chloride are effective lithium fire extinguishing agents. The powdered carbon preparation called Met-L-X is pressurized and stored in cylinders and is commonly used for extinguishing lithium fires. It is important that normal fire extinguishing agents are not used, as these compounds (water, carbon dioxide, sand, sodium bicarbonate and carbon tetrachloride) react highly with burning lithium.

Any lithium that spills should be immediately transferred to buckets containing 100 mesh powdered graphite. Lithium-contaminated areas should be wiped with flame-proof rags. All unopened packages containing lithium should be clearly marked, "Hazardous material - keep away from water".

REFERENCE

J. O. Cowels and A. D. Pasternak, "Lithium Properties Related to Use as a Nuclear Reactor Coolant", UCRL-50647 (1967).

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TABLE 1.1-- NOMINAL REFERENCE DESIGN PARAMETER (LL, REF. 1.2)

Net Electrical Power per Reactor, MW	27.5
Number of Reactors	10
Net Plant Efficiency, percent	36
Pulse Rate per Reactor, Hertz	10
Pellet (hollow sphere)	
Pellet mass, grams	10^{-4}
Pellet composition, atom percent	50/50 D/T
Pellet Diameter, mm	2.
Pellet wall thickness, mm	.07
Reactor	
Cavity shape	Icosahedron
Cavity dimension (radius of inscribed sphere), m	2.2
First wall liner thickness, mm	.7
Maximum pressure before shot, torr	.1
First Wall Liner Parameter, per pulse	
Energy deposition, charged particle, J/m ²	10^4
Energy deposition, x-rays, J/m ²	10^3
First Wall Liner Material and Thickness	
Lithium, mm	.3
Nb, mm	.4
First Wall	
Neutron loading, MW/m ² (time averaged)	1.
Vacuum pumping area, steradians	.5
Laser beam area, steradians	1.2
Blanket	
Thickness, m	1.
Breeding ratios (tritons/14 MeV neutron)	1.45
Materials	Lithium, Niobium
Laser System Parameters	
Energy output, Joules/shot	10^5
Wave length, micron	$.3 \leq \lambda \leq .6$
Efficiency, percent	10.
Power supply (plant)	
Energy, Joules	10^6
Pulse frequency, Hertz	10^2
Lifetime, pulses	10^{11}
Amplifier systems per reactor	1.
Number of beams to reactor	12.
Optics rating, J/m ² per pulse	2×10^4
Peak power into pellet, watts	5×10^{13}

TABLE 1.2
TIMESCALE OF EVENTS FOR LCTR PELLET FUSION PULSE

<u>TIME</u>	<u>PRIMARY EVENTS</u>	<u>SECONDARY EVENTS</u>
-20 TO -5MS	PELLET ENTERS CAVITY	
-150NS	LASER PULSE FIRED	
-10NS	LASER PULSE ARRIVES AT PELLET SURFACE	
0	THERMONUCLEAR BURN BEGINS	
+10PS	TH BURN COMPLETE	
+6NS	X RAYS STRIKE FIRST WALL	
		ABLATIVE MATERIAL BEGINS EXPANSION FROM FIRST WALL
+30NS	X RAYS STRIKE LAST OPTICAL SURFACE	
+20-100NS	NEUTRONS DEPOSITED IN REACTOR VESSEL	SHOCK WAVE INDUCED IN LITHIUM
+60NS	NEUTRONS STRIKE LAST OPTICAL SURFACE	
		ABLATIVE MATERIAL AND PELLET DEBRIS INTERACT
0.3 TO 1.2 μ S	PELLET DEBRIS STRIKES FIRST WALL	
		CAVITY ATMOSPHERE EQUILIBRATED
+1MS	CAVITY BLOWDOWN BEGINS	
3.01 TO 10 SEC	RESTORATION OF ORIGINAL CAVITY CONDITIONS COMPLETE	WEITED WALL BLOWDOWN COMPLETE, LITHIUM VORTEX RESTORED, TURBULENCE IN RARIFIED DRY WALL CAVITY DISSIPATED.

TABLE 3.1. Fluid Properties of Water and Lithium

Fluid	ρ , g/cm ³	μ , g/cm-s	σ , dynes/cm	N_σ	$\bar{b}_N = f(\hat{N}_{RE}, \beta)$ (cm)	$N_{FR}^* = f(\hat{N}_{RE})$	$N_{WE}^* = f(\hat{N}_{RE}, \beta)$
Water at 20°C	1.00	.00936	72.5	4280- 4660	$4.06 \times 10^{-3} \left[\frac{\hat{N}_{RE}}{\sin \beta} \right]^{1/3}$	$\hat{N}_{RE}/16$	$1.9 \times 10^{-5} \hat{N}_{RE}^{5/3} \sin \beta^{1/3}$
Lithium at 200°C	.515	.00575	400	39,480	$4.57 \times 10^{-3} \left[\frac{\hat{N}_{RE}}{\sin \beta} \right]^{1/3}$	$\hat{N}_{RE}/16$	$2.2 \times 10^{-6} \hat{N}_{RE}^{5/3} \sin \beta^{1/3}$
Lithium at 500°C	.481	.00340	349	67,842	$3.37 \times 10^{-3} \left[\frac{\hat{N}_{RE}}{\sin \beta} \right]^{1/3}$	$\hat{N}_{RE}/16$	$1.28 \times 10^{-6} \hat{N}_{RE}^{5/3} \sin \beta^{1/3}$

* N_{FR} , N_{WE} , and $\hat{N}_{RE}$ based on Nusselt film thickness, $\bar{b}_N$. It is noted that in this case N_{FR} is the same as the Reynolds number divided by a constant for all fluids.

TABLE 5.1. Thermophysical Data for Several Liquid Metals from the
Handbook of Heat Transfer (P-1)

ELEMENT	MELTING POINT (°C)	TEMPERATURE (°C)	DENSITY (g/cm ³)	VISCOSITY (g/cm-s)	SURFACE TENSION (dynes/cm)	N _G
Cesium	28.5	28.5	1.84	.0065	-	-
Gallium	30.0	50	6.09	.0189	735	33,720
Lithium	179	500	.484	.0034	349	67,840
Mercury	-39.0	20	13.5	.0155	465	36,340
Potassium	63.7	100	.819	.00458	86	13,419
Rubidium	39.0	39	1.475	.00673	-	-
Sodium	97.8	100	.928	.00686	206	19,590
Sodium-Potassium Alloy (N _G % wt 22)	-11.0	100	.847	.00468	120	18,400
Hot Water	0	90	.97	.0032	60.7	16,250

Figure 1.1 Schematic diagram of a laser fusion reactor (Ref. 1.3).

Figure 1.2 Gain of tamped pellets compared to bare pellets (Ref. 1.4).

Fig. 1-3. A conceptual suppressed-ablation laser-fusion reactor blanket element. (Ref. 1-2)

Figure 1.4 Typical penetration depths of the various particles produced in a laser-fusion micro-explosion into the containment vessel structure (Ref. 1.3).

Figure 1.5. Charged Particle Pellet Debris Energy Deposition in the Lithium Portion of the First Wall Liner with a Fluence of 24.3 kJ/m^2 at an Angle of Incidence of 70.5° from the Normal to the Surface. (Ref. 1.2)

Figure 1.6 Schematic diagram of some of the complex interactions which must be considered in design of the first wall of a laser-fusion reactor (J. Hovingh, LLNL).

Figure 2.1 Schematic Representation of the Contact Angle
Between a Liquid Drop and a Solid Substrate

Figure 2.2 The Dependence of the Observed Contact Angle on Temperature for the System Au-Ge(l)/GaAs(s) (Ref. 2.18)

Figure 2.3 Electron Microprobe Analysis of the Solid-Liquid Interface
Showing Significant Compositional Changes
in the System Au-Ge(l)/GaAs(s) (Ref. 2.18)

Figure 2.4

Electron Microprobe Scan Demonstrating the Absence of Diffusional Processes Across the Liquid-Solid Interface Between Au-Ge(L) and GaAs(s) (Ref. 2.18)

Figure 2.5

The Dependence of the Contact Angle
on Surface Roughness for the System Ga(l)/GaAs(s) (Ref. 2.19)

Figure 2.6 The Influence of the Substrate Crystallographic Misorientation on the Contact Angle for the System Ga(ℓ)/GaAs(s) (Ref. 2.19)

Figure 2.7 - EFFECT OF AIR EXPOSURE OF STAINLESS STEEL SAMPLES ON THE CONTACT ANGLE WITH LIQUID LITHIUM.

Figure 2.8

EFFECT OF NITROGEN IMPURITY ON THE CONTACT ANGLE OF LITHIUM ON STAINLESS STEEL

$$Re \equiv \hat{N}_{RE}$$

Figure 3.1(a,b,c,d) Time traces of water surface-wave amplitudes at increasing flow rates in the fully-developed region for thin film flow on a vertical plate (D-1).

(a) $Re = 180$ 2" BELOW LINE OF WAVE INCEPTION

$$Re \equiv \hat{N}_{RE}$$

(b) $Re = 180$ 2" BELOW LINE OF WAVE INCEPTION

Figure 3.2(a,b) Time traces of wave amplitude at low water flow rates for thin film flow on a vertical plate (D-1).

Figure 3.3 Definition of approximate flow regimes in terms of the Reynolds, Weber, and Froude numbers for a falling water film on a vertical plate.

Figure 3.4a Photograph of water waves on a thin vertical falling film
at a Reynolds number of 80 (P-1).

Figure 3.5b Photograph of water waves on a thin vertical falling film at a Reynolds number of 200 ($P=1$).

Figure 3.4c Photograph of water waves on a thin vertical falling film at a Reynolds number of 400 ($P=1$).

Figure 3.5a Calculated values for Froude and Weber numbers as functions of the Reynolds number of thin water and lithium films on a vertical plate.

Figure 3.5b Calculated values for the Nusselt film thickness, b_N , as a function of Reynolds number for thin water and lithium films on a vertical plate.

Figure 3.6 Reynolds number for wave inception as a function of angle of inclination from the horizontal (F-1).

Figure 3.7a Comparison of various correlations for the film thickness parameter, N_T , in the turbulent flow regime with some water data on a vertical surface (F-1).

Figure 3.7b Comparison of film thickness data with various correlations of the film thickness parameter as a function of Reynolds number (F-1).

Fig. 3.8 - VISUAL INCEPTION LINE VS. N_{Re} FOR WATER (P-I).

Figure 3.9 Comparison of theoretical prediction of nondimensional equilibrium wave amplitude, A_{eq} , by Lee with experimental data for water films on a vertical surface (L-1).

Figure 3.11 Experimental data for the mean wave amplitude ratio as a function of Reynolds numbers (D-1).

Figure 3.10a Calculated values of the equilibrium wave amplitude ratio, A_{eq} , as a function of Reynolds number for water and lithium films on a vertical surface.

Figure 3.10b Calculated values of the equilibrium wave amplitude ratio for water films as a function of Reynolds number at differing angles of inclination from the horizontal.

Figure 3.12 Theoretical prediction of the dimensionless wave number for the maximum growth rate, α_m , versus Reynolds numbers for pure liquids with differing surface tension numbers (P-1).

Figure 3.13 Comparison of various theories to predict the wave number for maximum growth rate versus Reynolds number for water films on a vertical surface (P-1).

Figure 3.14 Comparison of theory with experimental data for the wave number for maximum growth rate versus Reynolds number for water films on a vertical surface (P-1).

Figure 3.15 Comparison of theory with experimental data for the non-dimensional wave speed relative to the mean surface velocity, C_r , versus Reynolds number for water films on a vertical surface (P-1).

Figure 3.16 Theoretical prediction for the spatial growth rate, α_i , versus nondimensional wave number, α_r , for water films on a vertical surface (P-1).

Figure 3.17 Time trace of typical surface wave forms on a thin water film showing two distinct wave forms (C-3).

Figure 3.18 Comparison of various theories with smoothed experimental data for the ratio of wave velocity to mean film velocity, c/u , as a function of Reynolds number (F-1).

Figure 3.19 Estimated values for the dimensionless minimum wetting rate, $\Gamma_{\min}/\mu$, as a function of contact angle, θ , for lithium films.

Figure 3.20 Estimated Nusselt film thickness, $\bar{b}_N$, as a function of contact angle, θ , for the minimum wetting rate condition for lithium films.

Figure 3.21 Estimated effect of angle of inclination, β , on the minimum wetting rate at various contact angles, θ , for lithium films.

Representation of the Relationship Between Drop Geometry and the Contact Angle

(a) Ref. 4.2

(b) Ref. 4.3

(c) Ref. 4.4

(d) Ref. 4.5

Figure 4.2 Various Furnace Designs Utilized in Contact Angle Measurements (Refs. 4.2-4.5)

Figure 4.3 An Apparatus with Multidrop Measurement Capabilities
(Ref. 4.6)

Figure 4.4

Schematic Diagram of a Cylindrical Heating Element with a Drop Dispenser

Figure 4.5 Detailed Schematic of Sessile Drop Dispenser

Fig. 3.1 Effect of temperature on wetting of 316 and 316L coupons by sodium (H-I).

Figure 5.2 Initial Experimental Apparatus for Verifying the Falling-Thin-Film Scaling Laws for Opaque or Transparent Liquids.

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