DEVELOPMENT AND USE OF IMAGE SCANNING ELLIPSOMETER
TO STUDY THE DYNAMICS OF HEATED THIN LIQUID FILMS

Progress Report

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ABSTRACT

The long-term objectives of this research are to determine the heat transfer characteristics and stability of evaporating ultra-thin films. Some of these films may form permanent coatings. The current objective is to develop and use a microscopic image-processing system (IPS) to measure the film thickness profile. The IPS has two possible configurations: an image analyzing interferometer (IAI) and/or an image scanning ellipsometer (ISE).

An IAI was designed, constructed and used to measure the thickness profiles of extended menisci in the contact line region of equilibrium and evaporating liquid films under steady state and oscillating conditions. The design and construction of our first ISE was also completed and the system was successfully tested by measuring the thickness profile and the refractive index of a non uniform solid film. The profiles of both wetting and partially wetting draining isothermal films were also measured. The profiles of the wetting films were analyzed to obtain the dynamics of transport processes in thin films. The analysis of the nonwetting profiles and the redesign of the ISE were initiated. The two doctoral students who were responsible for this research completed their theses. During the current year, the development of the second generation ellipsometer is being emphasized along with the further analysis of our previous experimental results. Next year the research will emphasize the use of the new ISE to measure and evaluate the formation of thin coatings. Subsequently, evaporating thin pure liquid films will be studied.

REVIEW OF PROGRESS

The long-term objectives of the research are to determine the heat transfer characteristics and stability of evaporating ultra-thin films. Some of these films may form permanent coatings. The current objective is to develop and use a microscopic image-processing system (IPS) to measure the film thickness profile. These measurements are important because the thickness profile gives information on the stability of the thin films and the transport processes in the liquid film. The results of the research have been published in a broad range of publications because they cover not only the development of a unique experimental technique but also a fundamental study in interfacial transport phenomena. For example, we found that the critical boundary condition in the contact line region was not sufficiently well defined. However, since it was needed to understand fluid flow and change-of-phase heat transfer in our research on thin films, we rectified this deficiency. These results were discussed in the publications coded P5, P6, and P8, and P9, which are listed below.

The IPS has two parts: an image analyzing interferometer (IAI) and an image scanning ellipsometer (ISE). The design and use of the IAI was discussed in numerous publications sent to DOE, which are listed below as P1, P2, P3, P4, P7 and P11. We found that the convenience and efficiency of microcomputer enhanced video microscopy naturally led to a better understanding of the transport processes in the contact line region. We also found that the processes of change-of-phase heat transfer and fluid flow in thin films are intrinsically connected because of their common dependence on the intermolecular force field.

The ISE is a rapid full-field imaging technique which we developed with high spatial resolution that will enable measurements of the changes in the very thin liquid film thickness profiles associated with instabilities, fluid flow, evaporation and drying. The initial ISE (originally called IAE in our initial report DOE/ER/14045-1), which was designed for proof of principle testing, was configured using an available null ellipsometer modified with laboratory optical equipment. We added an autocollimator to obtain accurate alignment, a video camera, and a long working distance microscope to improve resolution and magnification. After many modification the first generation ISE system was described in P10 and P15. The use of this ISE technique to measure the thickness profile and refractive index of a non uniform thin solid film were discussed in P10 and DOE/ER/14045-5. The ISE results were evaluated by comparing the experimental results with those obtained using our interferometer and also with those supplied by the manufacturer. The profiles of both wetting and partially wetting draining isothermal films were also measured. The profiles for the wetting films were analyzed and the results were published. This work is detailed in P14. Two additional papers concerning the ISE are P12 and P13. The analysis of the nonwetting profiles was commenced and the doctoral student who developed the ISE completed the writing of his thesis (P15).

Since the ISE can measure thinner films than the IAI, we have decided to emphasize the further development, evaluation and use of the ISE technique. Therefore, current research emphasizes the development of the second generation ellipsometer which is being built and which should be operational by May, 1995. The further analysis of our previous experimental results continues. Next year the research will emphasize the use of the new ISE to measure and evaluate the formation of thin coatings. Subsequently, evaporating thin pure liquid films will be studied.

LIST OF PUBLICATIONS/PRESENTATIONS/DOE REPORTS

PUBLICATIONS

- P1) "Microcomputer Enhanced Optical Investigation of Transport Processes in Thin Liquid Films", M. Sujanani and P.C. Wayner, Jr., pp. 79-86, DOE CONF-9005183, Proceedings of Eighth Symposium on Energy Engineering Sciences, Argonne National Laboratory, Argonne, IL, May 9-11, 1990.
- P2) "Interfacial Phenomena in an Evaporating Thin Film", M. Sujanani, *Ph.D. Thesis*, Rensselaer Polytechnic Institute, Troy, NY, 1990. Abstract in Appendix A of DOE/ER/14045-2.
- P3) "Microcomputer Enhanced Optical Investigation of Transport Processes with Phase Change in Near-Equilibrium Thin Liquid Films", M. Sujanani and P.C. Wayner, Jr., *J. Colloid and Interface Science*, 143, 472-487, 1991. (DOE/ER/1045-3).
- P4a) "Transport Processes and Interfacial Phenomena in an Evaporating Meniscus", M. Sujanani and P.C. Wayner, Jr., paper preprint AIAA 91-4002 presented at 1991 ASME/AICHE/AIAA National Heat Transfer Conference, Minneapolis, MN. (sent to DOE as a manuscript)
- b) "Transport Processes and Interfacial Phenomena in an Evaporating Meniscus", M. Sujanani and P. C. Wayner, Jr., rewritten version of P4a, *Chemical Engineering Communications*, 118, 89-110, 1992. (sent to DOE as a reprint)
- P5a) "Evaporation and Stress in a Small Constrained System", P.C. Wayner, Jr., Extended Abstract, pp. 269-272, in Paper Summaries, *IS&T's 44th Annual Conference*, May 12-17, 1991, St. Paul, MN, (sent to DOE as DOE/ER/14045-4).
- b) P5a published as part of a book of selected articles *IS&T's Proceedings of the 44th Annual Conference, Symposium on Coating Technologies for Imaging Materials*, Edited by J. Truong, IS&T, Springfield, VA, 22151, 1991, pp. 174-181.
- P6) "Spreading of a Liquid Film on a Substrate by the Evaporation-Adsorption Process", P.C. Wayner, Jr. and J. Schonberg, *J. Colloid and Interface Science*, 152, 507-520, 1992. (sent to DOE as a manuscript and reprint)
- P7) "Interfacial Phenomena, Evaporation and Stress in a Constrained System", S. DasGupta, I.Y. Kim, and P.C. Wayner, Jr., preprint # AIAA 92-4053, Washington, 1992. (sent to DOE as a preprint)
- P8) "Evaporation and Stress in the Contact Line Region", P.C. Wayner, Jr., in "Pool and External Flow Boiling", V. K. Dhir and A.E. Bergles Editors, Engineering Foundation by ASME, New York, New York, 1992, pp 251-256. DOE/ER/14045-6
- P9) "Spreading of a Liquid Film with a Finite Contact Angle by the Evaporation/Condensation Process", P.C. Wayner, Jr., *Langmuir*, 9, 294-299, 1993. (sent to DOE as a manuscript and reprint)

- P10) "Image Scanning Ellipsometry for Measuring Non-Uniform Film Thickness Profiles", A.H. Liu, P.C. Wayner, Jr., and J.L. Plawsky, *Applied Optics*, **33**, 1223-1229, 1994. (Sent to DOE as a manuscript and reprint)
- P11) "Use of the Augmented Young-Laplace Equation to Model Equilibrium and Evaporating Extended Menisci", S. DasGupta, J.A. Schonberg, I.Y. Kim, and P.C. Wayner, Jr., *J. Colloids and Interface Science*, **157**, 332-342, 1993. (sent to DOE as a reprint)
- P12) "Microscopic Image Processing System for Measuring Nonuniform Film Thickness Profiles: Image Scanning, Ellipsometry", Liu, A.H., J. Plawsky, and P.C. Wayner, Jr., Proceedings of 11th Symposium on Energy Engineering Science, Conf.-9305134, pp. 107-204, Department of Energy, Argonne National Laboratories, Argonne, IL, May 1993.
- P13) "Microscopic Image Processing Systems for Measuring Nonuniform Film Thickness Profiles", A.H. Liu, J.L. Plawsky, and P.C. Wayner, Jr., Proceedings of the 10th International Heat Transfer Conference, Brighton, England, **2**, 262-272, 1994, Edited by G. F. Hewitt, Institution of Chemical Engineers, Rugby, Warwickshire, UK. (sent to DOE as a conference paper, DOE/ER/ 14045-8)
- P14) "Image Scanning Ellipsometer for Measuring Transient Non-uniform Very Thin Liquid Film Thickness Profiles", A.H. Liu, P.C. Wayner, Jr., and J.L. Plawsky, *Physics of Fluids A*, **6**, 1963-1971, 1994. (Sent to DOE as a manuscript and reprint)
- P15) "Design, Development, and Applications of Image Scanning Ellipsometer for the Measurement of Thin Film Thickness Profiles", A. H. Liu, *Ph.D. Thesis*, Rensselaer Polytechnic Institute, Troy, NY, 1994. Abstract in Appendix A of DOE/ER/14045-10

PRESENTATIONS

- PR1) "Fluid Flow in Ultra Thin Films", M. Sujanani and P.C. Wayner, Jr., Presentation 167C-b, 1989 Annual Meeting of A.I.Ch.E., Nov. 5-10, San Francisco, CA.
- PR2) P1 above
- PR3) P4a above
- PR4) P5a above
- PR5) P7 above was presented at the ASME/AIChE National Heat Transfer Conference, August 9-12, San Diego, CA, 1992.
- PR6) P8 above was presented at the Engineering Foundation Conference on Pool and External Flow Boiling, Santa Barbara, CA, March 22-27, 1992.

PR7) P9 above was presented at the Research Conference on Capillarity and Wetting, Dynamical Phenomena, Arnhem, Netherlands, September 1 -6,1992 at no expense to DOE.

PR8) P9 above was presented at the AIChE 1992 Annual Meeting, November 1-6, Miami Beach, FL.

PR9) P12 above

PR10)"Image Scanning Ellipsometer for the Measurement of Thin-Film Thickness Profiles", A.H. Liu, P.C. Wayner, Jr., and J.L. Plawsky, Optical Society of America Annual Meeting, Toronto, Canada, Aug. 1993 (P10 above)

PR11)"Image Scanning Ellipsometer for the Measurement of Thin-Film Thickness Profiles", A.H. Liu, P.C. Wayner, Jr., and J.L. Plawsky, 1993 Annual Meeting of AIChE, St. Louis, MO, Nov. 1993 (P14 above).

PR12)"Development and Use of Image Scanning Ellipsometer to Study the Dynamics of Heated Thin Liquid Films", OPA Peer Review Assessment, Gaithersburg, MD, Nov. 16-18, 1993.

PR13) P 13 above.

PR14)"Film Thickness Profiles for a Partially Wetting System, Dodecane on Silicon: An Application of Image Scanning Ellipsometer", A.H. Liu, P.C. Wayner, Jr., and J.L. Plawsky, AIChE 1994 Annual Meeting, November 13-14, San Francisco, CA.

DOE REPORTS

DOE/ER/14045-1) "Microcomputer Enhanced Optical Investigation of Spreading and Evaporative Processes in Ultra Thin Films", P.C. Wayner, Jr., M. Sujanani and A.H. Liu, DOE Progress Report DOE/ER/14045-1, January 1990.

DOE/ER/14045-2) "Microcomputer Enhanced Optical Investigation of Spreading and Evaporative Processes in Ultra Thin Films", P.C. Wayner, Jr., M. Sujanani and A.H. Liu, DOE Progress Report DOE/ER/14045-2, December 1990.

DOE/ER/14045-3) P3 above

DOE/ER/14045-4) P5a & P5b above

DOE/ER/14045-5) "Microcomputer Enhanced Optical Investigation of Spreading and Evaporative Processes in Ultra Thin Films", P.C. Wayner, Jr. and A.H. Liu, DOE Progress Report DOE/ER/14045-5, December 1991.

DOE/ER/14045-6) P8 above

DOE/ER/14045-7 "Microcomputer Enhanced Optical Investigation of Spreading and Evaporative Processes in Ultra Thin Films", P.C. Wayner, Jr., DOE Progress Report DOE/ER/14045-7, January 1993.

DOE/ER/14045-8) P12 above

DOE/ER/14045-9) P.C. Wayner, Jr., "Development and Use of Image Scanning Ellipsometer to Study the Dynamics of Heated Thin Liquid Films", DOE Progress Report DOE/ER/14045-9, January 1994.

DOE/ER/14045-10) P.C. Wayner, Jr., "Development and Use of Image Scanning Ellipsometer to Study the Dynamics of Heated Thin Liquid Films", DOE Progress Report DOE/ER/14045-10, January 1995.

DISCLOSURE OF INVENTION

Image Scanning Ellipsometer (ISE), to Dr. Chris LeMaistre, Head, Patent & Licensing at Rensselaer Polytechnic Institute, September 18, 1992. After a patent search through the Patents and Licensing Department at RPI, it was decided that an application for the above invention would be rejected (in their opinion) on the basis of anticipation and/or obviousness. Nonetheless, the ISE has many uses.

BUDGET INFORMATION

At the end of the current grant, there will be no funds available for the proposed renewal research.

Design, Development, and Applications of Image Scanning Ellipsometry for
the Measurement of Thin-Film Thickness Profiles

by

An-Hong Liu

An Abstract of a Thesis Submitted to the Graduate Faculty of Rensselaer
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Abstract

A novel technique, Image Scanning Ellipsometry, to measure the two dimensional thickness profile of a non-uniform, thin film, from several nm up to several μm , in the transient state as well as in the steady state was developed and tested in this thesis. Image Scanning Ellipsometry (ISE) is a full-field imaging technique which can study every point on the surface at the same time with high spatial resolution and thickness sensitivity; i.e., it can measure and map a liquid or solid film thickness profile in two dimensions. The long-term objective of the development of ISE is to determine the stability and heat transfer characteristics of evaporating thin films. The main purpose of this thesis was to develop the basic concept of ISE and demonstrate its use by measuring the thickness profiles of non-uniform solid films in a steady state as well as the profile of draining liquid films of wetting and partially wetting fluids in a transient state.

In this thesis, ISE has been proven to be as accurate as a null ellipsometer by measuring a known solid wedge profile of ThF_4 on a Si substrate. In addition, the ability of ISE to measure liquid draining films such as FC-5311, FC-77, and FC-70 in a transient state was demonstrated. Moreover, ISE was also used to measure a partially wetting, draining film of dodecane, and to record the details of film rupture. The approximate solutions of a modeling equation for the thickness profile during draining was compared to the experimental profile. The agreement between theory and experiment is quite good. The theoretical profiles agree with the experimental profiles in both the thicker hydrodynamic region and in the thin film region which is under 100 nm.

However, because the current limited magnification of the ISE hinders the exact location of a null point and the allocation of the exact position of the dark fringes is limited by the ability to accurately digitize and analyze the images, discrepancies between the modeling and the experiments are expected. Moreover, since the initial draining film

thickness is not infinite as assumed in the model, there is a difference between the real draining time and the model time. With calibration, this is not a hinderance to model evaluation.

The present ISE design is not optimum but can be easily improved by redesigning and rebuilding the instrument. Suggestions for improvement include replacing the optical components for improved resolution, using better image aquisition hardware, and faster data analysis tools.