Submicron thermal imaging of a nucleate boiling process using fluorescence microscopy

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A R T I C L E   I N F O

Article history:
Received 23 November 2015
Received in revised form 26 April 2016
Accepted 29 April 2016
Available online 24 May 2016

Keywords:
Temperature-dependent fluorescence
Erbium-doped heavy-metal glass
Nucleate boiling
Submicron spatial resolution
Transient thermal imaging

A B S T R A C T

The submicron characterization of transient heat-transfer processes at solid–liquid interfaces is of great importance in many areas of science and engineering. This paper reports on a technique that allows for the transient thermal imaging of the temperature field underneath a growing bubble during nucleate boiling with submicron spatial resolution. The boiling experiments were performed on a temperature-sensitive, erbium-doped, heavy-metal glass, Er:ZBLALiP, used as a robust sensing material for the non-invasive, transient temperature measurements. These measurements were made by analyzing the intensity variations of the fluorescence emission. The thermal imaging of an active nucleation site was performed by utilizing high-resolution, fluorescence microscopy, which enabled a maximum spatial resolution of 370 nm/pixel. The high-speed acquisition above 400 fps ensured sampling of individual bubble-nucleation events. Our transient measurements clearly revealed temperature variations underneath the growing bubble, as well as a measurable bubble-departure frequency under saturated conditions. These encouraging results suggest the need for a systematic use of the corresponding fluorescence technique on enhanced boiling surfaces in order to define the local heat-transfer characteristics and to gain a better understanding of the underlying physical processes.

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1. Introduction

The effective characterization of dynamic thermal processes on the submicron and nano scales provides new scientific understandings and accelerates the development of innovative devices across a broad range of areas, including microelectronics, biochemistry, and biomedicine. Micro- and nano-scale devices are generally subjected to a variety thermal processes that determine their reliability and performance, whereas some are able to exploit their unique sensory abilities, which can then be used in clinical diagnostics and bio-imaging applications to measure the temperature distribution at the phase interfaces of different types [1]. Owing to the variety of complex interactions between the mechanisms of heat transfer, a high-resolution, spatial characterization of the temperature distribution at solid–liquid–gas interfaces requires the use of novel, non-invasive measurement techniques. There have been substantial efforts made over the past decade to implement non-invasive thermometry to analyze the thermal transport across various phase interfaces on the micron scale. Optical, non-invasive temperature techniques, such as infrared thermography [2], liquid-crystal thermography [3], thermoreflectance [4], transmission electron microscopy [5], Raman thermometry [6] and luminescence thermometry [7], have been successfully adopted in the fields of modern microchips [8,9], microfluidics [10,11] and micro heat transfer [12,13]. For example, the high-spatial-resolution temperature imaging of resistively heated micro-wires was achieved using fluorescence thermometry, as reported by Löw, Kim [14]. Similarly, Ross, Gaitan [15] demonstrated the high-spatial-resolution thermal imaging of a multi-branched, microfluidic circuit, while Benninger, Koç [16] and Wu, Kwok [17] presented the three-dimensional mapping of the thermal distribution inside a microfluidic chip realized through optical sectioning. However, most of these techniques were designed for a very specific subject of analysis and are only effective under specific circumstances [18].
One of the most intricate and highly dynamic interfacial processes is the phenomenon of nucleate boiling. In order to understand this highly effective and complex heat-transfer mechanism, high-spatio-temporal temperature measurements at the single-bubble level are required [19]. Consequently, several different techniques, including thermochromic liquid crystals [20,21] and IR thermography, have been used in order to deduce the wall-temperature distribution under a growing bubble. High-speed IR thermography [22–25], for example, has been used extensively for more than a decade to provide useful information about nucleate pool boiling. However, due to a diffraction-limited spatial resolution of the order of ~10 μm, a detailed insight into the solid–liquid–gas interface has not yet been fully realized. In this context, the need for a comprehensive thermal characterization of phase-change processes on the submicron scale has led to the emergence of novel techniques such as fluorescence thermometry [26]. For the application of the latter, rhodamine B (RhB) and other temperature-sensitive dyes have been used in two-phase boiling experiments to measure the temperature field surrounding a single vapor bubble [27]. An alternative approach was proposed by Quinto-Su, Suzuki [28], who used a fluorophore solution for both spatially and temporally resolved temperature-distribution measurements after the oscillation of a single laser-induced microbubble inside a microfluidic liquid gap. However, despite these promising experimental results, there are some severe limitations that call into question the future feasibility of these measurements: firstly, the dye concentration in the fluid, especially around the edges of the vapor bubble, is not homogenous [27], thus drastically influencing the magnitude of the luminescence intensity and the resulting temperature, and secondly, the temperature measurement may also be affected by diffraction effects caused by the liquid/vapor interface.

In this study we focus on the development of a high-resolution imaging technique for non-invasive measurements of the transient temperature field underneath a growing bubble during nucleate pool boiling. To achieve this aim we have developed a robust measuring technique that relies on the stable, temperature-dependent emission characteristics of a heavy-metal fluoride glass doped with rare-earth ions. A transparent erbium-doped fluoride glass, i.e., Er:ZBLA LiP, was used as the sensing material to perform both the nucleate boiling experiments and the transient temperature measurements. Individual bubble-nucleation events were observed by using an inverted confocal microscopy system with acquisition rates of several hundred frames per second. The thermal images were obtained by measuring the local intensity variations of the fluorescence emission in the temperature range of the saturated water nucleate boiling. The results of the submicron temperature imaging of the phase-change phenomena underneath the growing bubble illustrate the potential of this measuring technique.

2. Experiments

2.1. Experimental setup

Fig. 1 shows the schematics of the experimental setup for the nucleate pool-boiling experiments. It consisted of a boiling vessel, a confocal fluorescence microscopy system, and auxiliary equipment such as a data-acquisition unit, DC power supply and variable transformer. The boiling vessel consisted of the stainless-steel (grade 316) metal flanges and the square-shaped Pyrex glass tubing for the visual observations of the water pool-boiling phenomena. The double-walled glazing functioned as an isothermal bath, as hot air was circulating in the wall in order to minimize the heat losses. Flat wall design allowed direct optical access to the area of interest with almost no optical distortion. The saturated boiling conditions were maintained by the temperature of the circulating heated air within the isothermal bath, and with two partially immersed cartridge heaters powered by a HSN 0202 (Metrel, SI) variable transformer. During the pool-boiling experiments the bulk temperatures of the circulating air and the de-ionized water were carefully monitored using K-type thermocouples connected to an Agilent 34970A (Loveland, CO, USA) data-acquisition unit equipped with an Agilent 34901A multiplexer module. The uncertainty of the temperature measurements was estimated to be ±1 °C. A reflux condenser was mounted on the top of the vessel to prevent water-level fluctuations and to ensure the cartridge heaters remained properly immersed.

A test section for the pool-boiling investigations was mounted on the bottom of the boiling vessel and powered by an EA-PS 3032-10B (Elektro-Automatik, Viersen, GER) DC power supply. The Joule heat generation across the boiling surface was determined by measuring the voltage drop across the heated area and the shunt resistor and acquired by the data-acquisition unit. For an accurate positioning of the boiling vessel, a custom-designed, XY, open-frame microscope stage was fabricated and mounted on top of the inverted microscope.

A transient thermal imaging of wall temperature distribution during bubble growth was performed by utilizing fluorescence-based microspectroscopy. This high-resolution technique relies upon the blue light excitation of a fluorescent material at shorter wavelengths, followed by the temperature-dependent emission of green fluorescence at longer wavelengths. The wall-temperature measurements during nucleate boiling were based on the integrated-intensity method where the fluorescence intensity within a broad spectral band was recorded. The FMS (fluorescence microspectroscopy) system has been described in detail in Sedmak, Urbančič [29]. Here, we describe the key features of the microscopy system and its recent modifications to acquire high-speed thermal images. As can be seen in Fig. 2, the system was based on an inverted Nikon Eclipse TE 2000-E microscope platform and the CARV II (BD Biosciences, USA) fluorescence and spinning disk confocal unit. The excitation, dichroic and emission filters were all housed within the fluorescence unit. A fluorescent sample was excited by a Xe–Hg light source through 10 × and 40 × objective lenses, whereas the emission passed through a LCTF (liquid-crystal tunable filter) and an OPTOMASK (Andor, Belfast, UK) adjustable field mask aperture placed in front of the scientific-grade EMCCD (electron multiplying charge-coupled device) camera. Due to the relatively weak fluorescence signal obtained from the investigated sample, the highly sensitive (Best-in-class) Andor iXon3 897 EMCCD camera with high quantum efficiency and with binning capabilities to increase the SNR (signal-to-noise ratio) was used. To perform spectrally-resolved measurements the light was passed through the LCTF prior to reaching the EMCCD, whereas for high-speed imaging of the nucleate pool-boiling the LCTF was removed and replaced with field mask aperture. The removal of the LCTF during broad-band imaging is necessary to compensate for the poor light transmission, which reduces the maximum acquisition rate of the visualization system. The fluorescence images were afterwards processed and analyzed with a custom MatLab script.

2.2. Test section

The boiling experiments were performed on a specially designed test section mounted in the boiling vessel. The setup was designed to perform both nucleate pool-boiling experiments and the transient submicron temperature imaging of the active nucleation sites. As can be seen in Fig. 3, a fluorescence glass sample with a size of 16 × 10 mm² was inserted into a rectangular borehole
milled in the center of a ceramic insulation plate and placed onto the stainless-steel square flange to enable optical observations. Nickel-plated electrical contacts were installed in the test section to supply power to the heater. All the parts were glued together using a flexible heat-resistant adhesive to avoid any thermal expansion damage.

The erbium-doped transparent fluoride glass Er:ZBLALiP was used as a sensing material because of its unique properties as a temperature sensor. Most important amongst these properties are: high temperature sensitivity, chemical stability and mechanical robustness as well as excellent photo-thermal stability against photobleaching and hysteresis effects [30]. The bulk sample of Er:ZBLALiP was double-side grinded and polished to a thickness of 500 μm, which enabled optical observations with high-magnification objectives that typically had short working distances. The sample was coated via a reactive magnetron-sputtering process to produce a uniform Au thin-film resistive heater (11 × 10 mm²) with an overall thickness of 270 nm, and with the following properties: density (ρ) 19,300 kg/m³, specific heat (c_p) 128 J/kgK, and thermal conductivity (k) 317 W/mK. This deposition process was used to avoid excessive thermal stress to the fluoride glass sample and to assure thin-film thickness uniformity, and also the uniformity of the electrically induced heat flux. The thin-film heater was deposited onto the upper surface of the sample, whereas the bottom surface remained uncoated for the thermal microscopy imaging. Due to the heat transfer across the thickness δ of the heater, the thermal time constant δρc_p/k was calculated to be 0.57 ns, which is well below the fastest possible temporal resolution of the measurement system. The small thickness allowed a rapid response to the temperature changes, which is beneficial, since a thicker wall can significantly affect the heat-transfer process during the nucleate boiling [31]. A top view of the boiling vessel and a closer-up view of the test section are shown in Fig. 4 (a). The surface topography of uncoated and coated glass sample was characterized by the Park XE-100 (Park Systems, Suwon, KOR) atomic-force microscope, as shown in Fig. 4 (b) and (c), respectively. The average surface roughness (Ra) was 2.7 nm for the uncoated sample and 2.3 nm for the Au-coated sample. It was found that both surfaces can be classified as smooth heating surfaces without distinctive nucleation cavities which would promote an earlier transition to the onset of boiling according to the nucleation criterion by Hsu [32].

2.3. Calibration procedure

Prior to the boiling experiments the thermal behavior and microspectroscopic analysis of the Er:ZBLALiP bulk sample were conducted. The heavy-metal fluoride glass had the following chemical composition: 62.2%ZrF₄—19.5%BaF₂—6.1%LaF₃—3.6%AlF₃—2.4%LiF—6.1%PbF₂ + x%ErF₃, where x is the erbium-doped concentration in mol%. In our experiments we used a concentration of 6 mol% of erbium ions, which equates to 12 × 10²⁰ Er³⁺ ions/cm³. The preparation of this kind of erbium-doped, heavy-metal glass has been described in detail in Mortier, Goldner [33].

The microspectroscopic and thermal characterizations of the bulk sample were performed as described in Ref. [29]. In short, the sample was excited by light with wavelengths between 430 and 490 nm. The light from the sample was fed back through the CFI (Chromatic aberration-Free (or Chrome Free) Infinity) Plan Fluor 10 × objective lens and the 550/88 emission filter, through the narrowband LCTF for a spectrally resolved detection. The fluorescence was acquired by the LCTF in the range between 510 and 570 nm in 1-nm steps. The exposure time for each picture in the λ-stack was 0.5 s. The emission spectra of the 6% Er:ZBLALiP versus temperature at intervals of 10–15 K are presented in Fig. 5.
The fluorescence spectra consisted of two distinctive emission peaks, where the second peak at 546 nm was considerably higher than the first one, centered at 525 nm, and decreased rapidly at elevated temperatures. This tendency originates from the electron transitions from the metastable levels $^2H_{11/2}$ and $^4S_{3/2}$ to the ground level $^4I_{15/2}$. The obtained emission spectra correspond well to the results reported previously [34], except for the fine features, which were not resolved with our system due to the 10-nm spectral transmittance band of the LCTF.

The wall-temperature measurements during the nucleate boiling were based on an integrated-intensity method and performed in the conventional-fluorescence-microscopy imaging mode. For this purpose the sample was illuminated through the high-magnification CFI S Plan Fluor ELWD (Extra Long Working Distance) 40 x C objective lens, whereas the LCTF was removed and replaced with an adjustable field-mask aperture in front of the EMCCD camera. The adjustable field mask featured a precise aperture control to prevent light leakage on the defined active sub-area of the sensor, which in turn enabled fast imaging of the boiling surfaces with a submicron spatial resolution. Therefore, with an ability to crop the sensor of the camera and in combination with the integrated-intensity method, acquisition rates of up to 500 fps were achieved.

Prior to each boiling experiment the temperature-intensity calibration curve had to be determined due to the different acquisition parameters and camera pre-settings, such as gain and pixel binning. The binning was applied to ensure a reasonable signal-to-noise ratio at faster acquisition rates. Accordingly, two camera pre-settings modes were used, which yielded high-spatial-resolution images of 370 nm/pixel at 68 fps, and lower-spatial-resolution images of 740 nm/pixel at 413 fps. Both calibration curves shown in Fig. 6 were obtained by averaging the intensity of the entire image at the corresponding temperatures. The surface-temperature distribution was derived by correlating the signal value of each individual pixel with the calibration curve. It should be noted that both the fluorescence intensities of the sample were normalized to their fluorescence intensities at the lowest temperatures.

According to the linear least-mean-square fit over the temperature range from 75 °C to 140 °C at different camera pre-settings, the sensitivities of the fluorescence-intensity change were
determined to be $-0.6\%$/K and $-0.4\%$/K, respectively. The absolute measurement uncertainty of wall temperature measurements was determined to be around 2.7 K for the temperature range between 100 °C and 130 °C.

2.4. Image processing

The original fluorescence images were processed using the custom-developed MatLab script. The data-processing flowchart is shown in Fig. 7. The raw images were stored as files and assigned as a matrix of 16-bit image pixel intensity values. As the first image-processing step, the offset signal from the EMCCD camera was subtracted. The next step was the averaging of the images over 10-by-10 neighboring pixels to remove the noise, followed by the transformation of the image grey-level to the temperature. Due to the uneven brightness of the field-of-view, owing to the uneven illumination profile, reflections from the heater, potentially non-homogeneous material, etc., the temperature variations were calculated with respect to the temporal intensity average during the experiment at each location.

2.5. Experimental procedure

The boiling experiments were performed with de-ionized water that was degassed by vigorously boiling for two hours. The bulk temperature of the water in the boiling vessel was maintained at atmospheric pressure in the saturated state by controlling the power of the cartridge heaters and by the temperature of the heated air in the isothermal bath. Once the boiling vessel reached steady-state conditions, the power to the Au thin-film heater was applied in discrete steps to achieve nucleate boiling. Moreover, the generation of large numbers of bubbles on the heated surface was associated with the onset of nucleate boiling. After some time the applied power to the heater was evenly decreased to the point of sparse bubble generation, whereas it was operated at a constant heat flux. The boiling vessel was precisely maneuvered along both the x and y axes using the microscope stage in order to track the randomly distributed active nucleation sites. The fluorescence-intensity response to the temperature variation during the bubble growth was measured through the high-magnification objective lens that was focused on the bottom side of the thin-film heater plane. According to the depth of field in optical microscopy the fluorescence signal was obtained from the optical section, at a distance of about 2.7 μm from the boiling surface. Throughout the experiments, special care was taken to ensure the rapid evacuation of the working fluid from the boiling vessel due to potential leakage.

3. Results and discussion

The implementation of a fluorescence-based technique for the quantitative thermal characterization of the temperature distributions during nucleate boiling resulted in thermal images with a submicron spatial resolution. The microscopy system allowed the highest possible diffraction-limited spatial resolution of ~370 nm/pixel at an image-acquisition rate of 68 fps. Fig. 8 shows an example of the detected surface-temperature variations under the growing bubble during the saturated nucleate boiling at atmospheric pressure. Due to the low acquisition rate, only three consecutive images were acquired for a particular nucleation event. The heat flux supplied to the thin-film heater was 60 kW/m², whereas the analyzed field was $256 \times 256$ pixels, which corresponded to 94.72 μm × 94.72 μm. In the images shown, the entrapped residual vapor left from the previous nucleation event initiated a rapid
Temperature dependence of the relative intensities of the Er:ZBLALiP sample
Fig. 6. Temperature dependence of the relative intensities of the Er:ZBLALiP sample for different camera pre-settings. Fine focus adjustments were needed to compensate for focus shift due to temperature changes within the sample.

However, studying the individual bubble-nucleation events requires acquisition rates of at least several hundred frames per second.

Having obtained the thermal images of the active nucleation site at the highest-possible spatial resolution of the measuring system, the fastest transient-temperature measurements were performed with a reduced spatial resolution and with a reduced field of view. The basic concept of the isolated bubble growth during saturated boiling of water which has been previously studied by Golobic, Petkovsek [35] has been adopted in the current work for the interpretation of nucleate boiling at the submicron level. The spatial resolution of these measurements remained at the submicron level, while the acquisition rate increased up to 500 fps. In order to perform these local measurements, stable microbubble nucleation sites were deliberately chosen due to the reduced field of view. In this example, the input heat flux was set at 45 kW/m². As presented in Fig. 9, the analyzed field was 50 × 32 pixels (37 µm × 23.68 µm), corresponding to a spatial resolution of ~740 nm/pixel at 413 fps.

The beginning of the bubble-nucleation event at 0 ms was associated with a non-uniform wall temperature distribution left by the previous microbubble activity. Bubble nucleation occurred at 5 K wall superheat while the sounding area remained superheated at 7 K. The temperature distribution under the growing microbubble was changing significantly following a rapid decrease of wall temperature during the initial period of the bubble growth. The contact area of the bubble was expanding rapidly at that time, reaching its maximum shortly after the lowest wall superheat of 1 K was reached at around 9.7 ms. During the fast bubble growth, the bubble was sliding on the surface in all directions and the cooled region under the bubble was greatly affected by the thermal fluctuations of the surrounding liquid. After this stage the contact area started to decrease and temperature of the heated wall was progressively reheated until the detachment event appeared to take place. Due to the bubble growth and departure, wall-temperature variations as large as 6 K occurred for the aforementioned input heat flux. After the detachment, the nucleation of another bubble took place at the same spot. As shown in Fig. 10, for a particular nucleation site the bubble-departure frequency was found to be around 45 Hz, which is in good agreement with previous experimental studies for pure water [25,36]. It should be noted that despite the enhanced spatial resolution, the triple contact line evaporation during the growth of the vapor embryo was not clearly observed. Furthermore, transient thermal imaging of the temperature field underneath a growing bubble is feasible only up to a certain point, beyond which the EMCCD camera is no longer capable of capturing rapid bubble growth. Our approach therefore averages out the ultrafast and highly localized events, but is appropriate to image heat transfer during bubble growth, where a local heat flux creates a persisting variation in the temperature field.

This experimental result indicates the successful implementation of the fluorescence-based technique for the quantitative thermal characterization of temperature distributions during nucleate boiling. However, conducting boiling experiments to measure temperature variations with a submicron spatial resolution proved to be challenging: (i) the boiling vessel had to be mounted on an inverted microscope and therefore in close proximity to sensitive optical devices while operating at high temperatures; (ii) there was a potential risk of fluid spillage from the vessel due to the thermal stresses within the test section; and (iii) it was difficult to track the randomly scattered active nucleation sites. It is important to mention that despite these challenges, the proposed technique makes possible robust transient-temperature measurements at a spatial resolution that is almost two orders of magnitude.
better than what has been available in such studies with infrared thermography.

4. Conclusions

We have successfully demonstrated the applicability of a non-invasive, fluorescence-based technique for the thermal characterization of a transient thermal field underneath a growing bubble with a submicron spatial resolution. This technique exploits the unique thermal properties of a transparent, erbium-doped, heavy-metal fluoride glass that can be used for the thermal characterization of micron-sized, transient-temperature fields. The wall-temperature variations underneath the growing bubble were measured with a diffraction-limited spatial resolution of ~370 nm/pixel at 68 fps. By decreasing the resolution to ~740 nm/pixel, a much more convenient acquisition rate of 413 fps was achieved, which enabled the capture of individual microbubble-nucleation events. The resulting combination of a rapid acquisition rate and data post-processing led to an estimation of the bubble-departure frequency of approximately 45 Hz under saturated conditions, which is comparable to other similar experiments. Moreover, the transient-temperature field under the growing microbubble was found to be substantially influenced by the thermal fluctuations of the surrounding liquid. Nevertheless, wall-temperature variations as large as 5 K occurred at the particular

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Fig. 7. Data-processing flowchart.

Fig. 8. (a) Surface-temperature variations under a growing microbubble during saturated nucleate boiling experiments at atmospheric pressure and the input heat flux of 60 kW/m². (b) Horizontal and vertical temperature profiles across the observed temperature fields.
Fig. 9. Local temperature distribution for the individual bubble-nucleation event. (a) Surface-temperature variations under the growing microbubble during the saturated water pool-boiling experiments at atmospheric pressure and the input heat flux of 45 kW/m². (b) Horizontal temperature profile across the observed temperature fields of a growing microbubble.
nucleation site, as a direct result of the microbubble growth, which implied a suitable thermal sensitivity of the sensing material. During repeated boiling cycles the Er:ZBLALiP glass exhibited a stable fluorescence emission without hysteresis effects. However, there is still plenty of room for improvement in terms of acquiring images at faster acquisition rates as well as enhancements to the boiling experiment itself: (i) the measurements can be substantially improved by choosing an optimal sensing material with an even higher thermal sensitivity [37], and (ii) in order to avoid the troublesome tracking of randomly scattered active nucleation sites, artificial ones should be considered [38–41]. Further improvements would require the synchronization of the high-speed video camera and the FMS camera, with a special emphasis on the maximum obtainable spatial resolution, possibly of the same order of magnitude.

Finally, the further development of the measuring technique, together with a new design of the boiling test section, could provide new clues for a better understanding of the nucleate boiling process and the prevailing surface heat transfer mechanisms. This technique could also have applications in the numerous physical, biological and electrochemical processes at solid-liquid interfaces, which may indicate new clues for a better understanding of the nucleate boiling phenomenon as a whole. This indicates that new developments could be made, possibly of the same order magnitude.

Acknowledgments

This research received financial support from the Ministry of Higher Education, Science and Technology of the Republic of Slovenia (Research Program: P2-0223).

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