Available online at www.sciencedirect.com
SCIENCE DIRECT®
International Journal of Heat and Fluid Flow 25 (2004) 298–304
International Journal of HEAT AND FLUID FLOW
www.elsevier.com/locate/ijhff

Identification of nucleation site interactions

Jost von Hardenberg [a], David B.R. Kenning [b,*], Huijuan Xing [b], L.A. Smith [b]

[a] CIMA, University of Genoa, Savona, Italy
[b] Department of Engineering Science, University of Oxford, Parks Road, Oxford OX1 3PJ, UK

Abstract

Simple models of nucleate boiling consider nucleation sites in isolation. In practice, they interact in ways that depend on the distance between them. In this paper, statistical evidence of interactions is obtained for a large number of non-uniformly distributed sites with different activation superheats during pool boiling of water on a thin plate at a heat flux of 51 kW/m^2. By analysis of spatio-temporal data for wall temperature obtained by liquid crystal thermography over a period of 30 s, the timing, position, activation superheat and radius of cooled region are obtained for every nucleation event, without need for direct observations of the bubbles. For each event, the number of subsequent events at all other sites during different delayed time intervals is obtained as a function of distance from the original site. The number is compared with a null hypothesis obtained by assigning random times to all events: a higher number indicates promotion, a lower number inhibition. It is found that there is promotion during very short time delays of the order of the bubble growth time at distances less than the bubble radius and inhibition for slightly longer delays and shorter distances. There is no statistically significant effect at distances greater than the bubble radius. This finding may be influenced by the low bubble frequencies characteristic of these particular experiments.

Keywords: Nucleate boiling; Nucleation site interactions

1. Previous studies of interactions

Interactions between bubble nucleation sites may influence the contribution of potential sites to the heat transfer process. In flow boiling of water at low subcooling on a thin, electrically heated plate with its surface in as-manufactured condition, Eddington and Kenning (1978) identified active sites by cine recording at 64 Hz over 5 s and showed that there was a cut-off in the distribution of nearest neighbour distances S below a distance approximately equal to the bubble departure diameter D. This was attributed to thermal interference. Sites could be deactivated by an increase in heat flux if they were situated close to a newly active site. Some of these new sites were, on the basis of gas nucleation experiments, expected to be deactivated by the initial subcooling of the system. It was found that a high proportion of these sites had been covered by vapour from active sites at lower heat fluxes, suggesting that their potential for activity depended on a vapour seeding mechanism. The camera speed was too low to investigate seeding during steady-state conditions. Del Valle and Kenning (1985) extended this study of nearest neighbour distances to high subcooling and heat high flux, analysing 40 cine frames recorded at 10^4 Hz. They confirmed that there was an inhibiting effect over a distance of about one bubble diameter and that sites deactivated by an increase in heat flux could be reactivated at an even higher flux, depending on the fate of neighbours. It was stated that conditions in the immediate vicinity of an active site favoured the initiation of nucleation at other sites but the simultaneous activity of sites with $0.75 \leqslant S/D \leqslant 1$ could not be sustained. In the light of experience, the period of observation may have been too short. Examples of gradual exchange of activity between adjacent sites have been observed in nominally steady pool nucleate boiling, Kenning et al. (2001). Studies of the spatio-temporal aspects of interaction have been mostly confined to pool boiling. Chekanov (1977) activated two sites at variable spacing and unknown superheats by pressing two heated copper rods against a thin plate in water and measured the shape of the Gamma distribution of the times between bubble departures at the sites, deducing an inhibiting effect at

* Corresponding author. Tel.: +44-1865-273000; fax: +44-1865-273010.
E-mail address: david.kenning@eng.ox.ac.uk (D.B.R. Kenning).

0142-727X/$ - see front matter © 2003 Elsevier Inc. All rights reserved.
doi:10.1016/j.ijheatfluidflow.2003.11.015

separations $S/D \leqslant 3$ and promotion at slightly larger separations. At about the same time, Judd and co-workers initiated a long series of studies of spatial non-uniformities in pool nucleate boiling. Calka and Judd (1985) performed a similar analysis to Chekanov for the "natural" sites for dichloromethane boiling on a uniformly heated transparent layer of tin oxide on a thick glass substrate, through which they were able to observe the bubbles and detect nucleation events by processing optical signals. They deduced promotion for $S/D \leqslant 1$, inhibition for $1 \leqslant S/D \leqslant 3$ and no interaction for larger separations. The methods of analysis and measurement for this fluid–surface combination were further developed by Judd and Chopra (1993), who analysed (again by the shape of fitted Gamma functions) the distributions of delay time after nucleation at one highly active 'dominant' site and nucleation at four other sites within a group of 21 active sites in a region of 10 mm diameter. The dominant site lay on the edge of this group, with the four other sites at distances of 1.6–6.3 mm inside the group. The average bubble size at the dominant site and the number of active sites were controlled by combined changes in pressure and heat flux. The analysis confirmed the previous finding of promotion for $S/D \leqslant 1$, inhibition for $1 \leqslant S/D \leqslant 3$ but showed that the inhibitory effect was much stronger at a higher site density. The authors' interpretation was that the dominant physical processes were the (promotive) seeding of relatively unstable sites when covered by bubbles growing at adjacent sites, and (inhibitory) scouring by fluid mechanical disturbances, with interactions extended to larger distances by a chain reaction involving "third party" sites that introduced a dependence on site density. The seeding process was extended beyond $S/D = 0.5$ based on the mean departure diameter for a site by fluctuations that produced larger bubbles. Encouraging probabilistic simulations of this mechanism were performed by Mallozza et al. (2000). The experiments and their physical interpretation imply asymmetry in the nature of the sites and their interactions.

Gjerkeš and Golobič (2003) performed experiments that excluded any possible "third party" effect. They activated a single "natural" site on a very thin plate in water heated uniformly either electrically or by a laser beam of large diameter. They then activated a second "artificial" site at a controlled distance by localised heating by a much smaller laser beam. (This implies that the second site required a higher superheat for activation.) They used as a measure of site activity the latent heat flux deduced from video recordings of the bubbles. They found that the activity of the natural site decreased and eventually ceased as the second site was moved closer (inhibition). The activity of the second site, which was always greater than that of the natural site, decreased until it occupied the position of the natural site, when there was a recovery. The combined activity of the two sites decreased as they were brought together. The inhibiting effect was asymmetrical: the more active site was less affected than its less active neighbour with a lower activation superheat.

Zhang and Shoji (2003) measured the symmetrical interaction between two similar artificial cavities at different separations for water boiling on a silicon plate 0.2 mm thick heated by a laser beam, with simultaneous measurement of the changing temperatures on the back of the plate by a line-scanning radiation thermometer. They measured the correlation function for temperature fluctuations at the cavities and along the line joining them. For $S/D > 2$, the correlation function became zero at the mid point, indicating no thermal interaction. Site activity was measured by the bubble frequency and a heat transfer coefficient ratio. Three competing mechanisms of interaction were identified: (A) hydrodynamic (promoting), (B) thermal (inhibiting), (C) various modes of coalescence for $S/D \leqslant 1.5$ (promoting). At $S/D > 3$, there was negligible interaction. At $S/D = 2.5$, there was a considerable increase in activity attributed to the hydrodynamic effect A, then a decline back to the original level at $S/D \approx 1.7$ due to a competition between A and thermal interaction B. Further reduction in S/D produced a sharp increase in activity where all three mechanisms competed. Note the contrast with the observations of Gjerkeš and Golobič (2003) for a pair of asymmetrical sites, that activity decreased continuously with decreasing separation.

Kenning and Yan (1996) obtained simultaneous video recordings at 200 Hz of bubble formation for the special conditions of pool boiling of water on a 0.125 mm thick, electrically heated stainless steel plate and the spatio-temporal variations in temperature on the back of the plate, visualised by liquid crystal thermography. The behaviour depended on the method of cleaning the surface and the resulting contact angle. Following cleaning by solvent, small bubbles were produced at a large number of sites at high frequencies beyond the response rate of the liquid crystal. Following cleaning with detergent, rigorous rinsing and protection of the surface from atmospheric contamination up to the instant of immersion, larger bubbles were produced at lower frequencies, within the measuring range of the liquid crystal, at an apparently smaller number of sites. For these conditions, it was shown that the growth of each bubble caused local cooling that extended only to the maximum contact radius of the bubble. This was used to identify nucleation events over a period of 12 s from the liquid crystal measurements, each event being verified by laborious reference to the bubble-side observations. The temperature–time variations at four adjoining sites were analysed at three heat fluxes. It was shown that interruptions in bubble production occurred when a bubble from one site cooled another site. The interactions were asymmetrical, because of the differences in the sizes of bubbles produced at the sites, and the relative activity of the sites changed with increasing heat flux. McSharry et al. (2000) developed a method of computer analysis of the temperature–time sequences at known sites by which sharp falls in temperature at one site were attributed either to nucleation at that site or thermal interference from an adjacent site, without having to consult the bubble-side images. The analysis could then be extended to 30 s and the number of interaction events was shown to be statistically significant. Next, McSharry et al. (2002) developed a new method of reconstructing each temperature field using non-orthogonal empirical functions (NEFs) of a form suggested by the nature of the local cooling processes that efficiently identified nucleation events, again without using bubble-side images. von Hardenberg et al. (2002) showed that the method could be applied to an area containing many sites, obtaining close agreement with the earlier manual identification of sites by Kenning and Yan (1996). The new method identified the position, timing, local wall superheat and radius of cooled area for every nucleation event in an area 21 mm × 11 mm over a period of 30 s at a heat flux of 51 kW/m^2. These data are used in this paper for a statistical analysis of nucleation site interactions.

2. Summary of NEF analysis

The shape chosen for the NEF basis functions is shown in Fig. 1. The analysis is performed on the difference between successive temperature fields, after the application of a 3 × 3 spatial average filter to reduce noise. This field is represented by a linear combination of NEFs of different positive or negative amplitude and radius, so as to minimise the reconstruction error between the original and reconstructed fields. In principle, any desired degree of accuracy can be achieved by using sufficient NEFs; minimization of a cost function which includes both the reconstruction error and a measure of the 'economy' of the reconstruction, is used to balance the reconstruction error against the number of NEFs employed. The advantage of using NEFs with a shape based on a particular physical event is that only a small number is needed to obtain a reconstruction in which those events are identified with reasonable accuracy. The analysis is applied to 6000 fields recorded in 30 s.

Not every near-circular cooled region is necessarily caused by bubble nucleation and growth: e.g. it could be caused by a local eddy. The identification of nucleation events is improved by post-processing, based on the expected time-related characteristics of cooling following nucleation, Fig. 2. Bubbles are known to remain in contact with the wall for 2–5 time steps, causing rapid local cooling followed by a slower recovery. NEFs

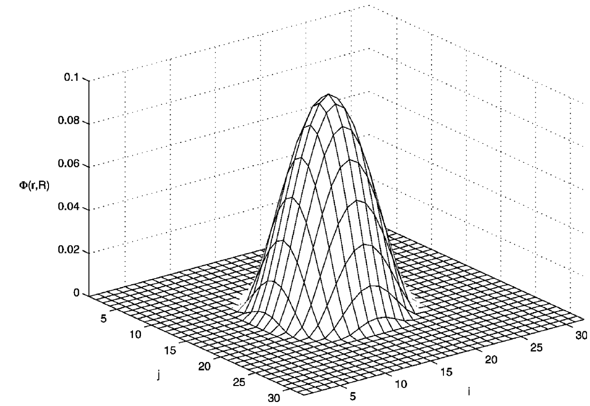

Fig. 1. Typical NEF basis function.

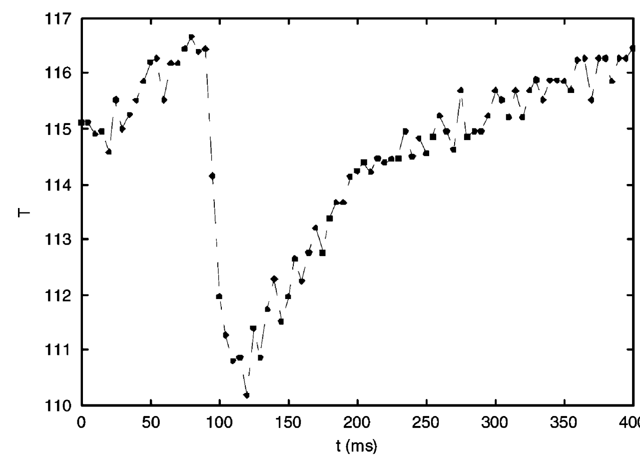

Fig. 2. Temperature evolution at a nucleation site.

centred within a certain radius Δr in at least two successive time steps are labelled as representing the same nucleation event. For the analysis presented in this paper, $\Delta r = 1$ mm. When all NEFs have been assigned to a particular nucleation event, the time and position of the first NEF of each sequence are recorded to represent the nucleation site and time. Each event is tracked in time to obtain the local maximum in wall temperature immediately before nucleation, the first minimum in temperature (coinciding approximately with bubble departure) and the corresponding radius of the NEF (approximately equal to the contact radius at bubble departure). All the nucleation events identified in this way in an area 21 mm × 1 mm over a period of 30 s are plotted in Fig. 3, with an indication of the number of events at each location. The distribution of maximum NEF radii for all nucleation events is plotted in Fig. 4.

There are about 16 concentrated clusters of activity that may, within the accuracy of location, be individual nucleation sites. There are 2 or 3 more diffuse regions of activity and many sites that produce only 1 or 2 bubbles in 30 s. These sites of low activity may be very unstable sites, since it was observed that many more sites were

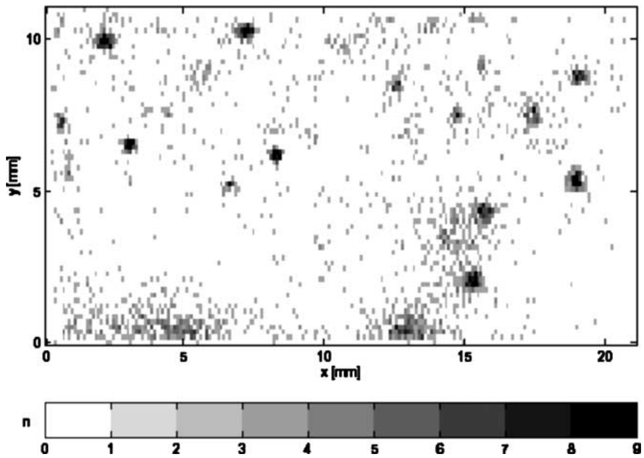

Fig. 3. Position and number of nucleation events.

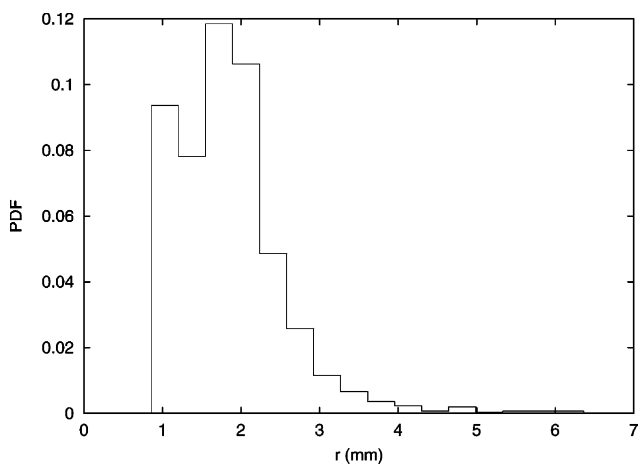

Fig. 4. Distribution of NEF radii for all events.

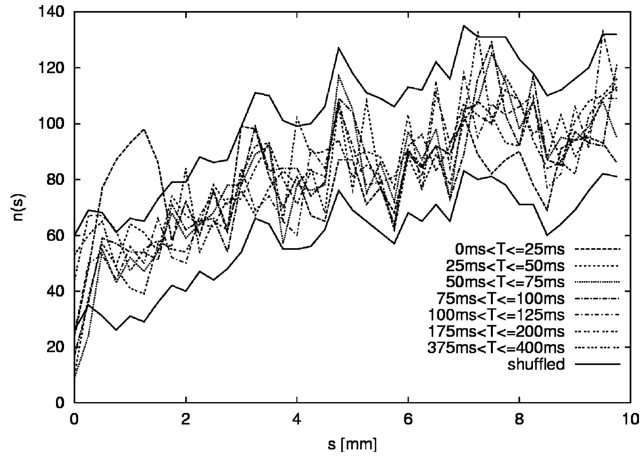

Fig. 5. Number of events at fixed delay time as a function of distance.

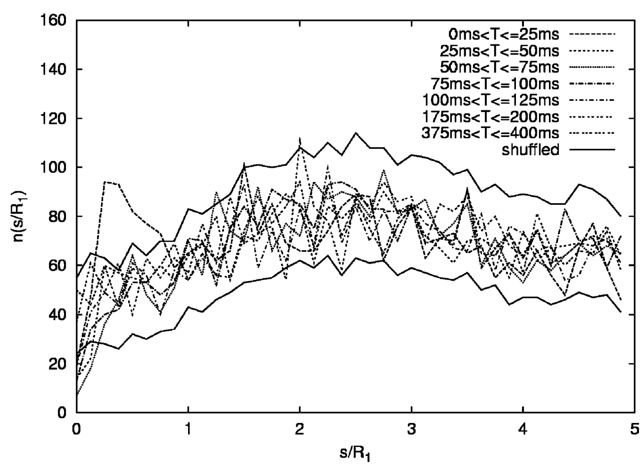

Fig. 6. Number of events at fixed delay time as a function of distance/radius of first bubble.

active on this same surface in a poorly wetted state (Kenning and Yan, 1996), they may be stable sites for which the local temperature is close to their activation temperature or, in the case of single events, they may be associated with gas-stabilised nuclei entrained in the bulk liquid close to the wall. The experimental uncertainty and the good agreement between the computerised NEF analysis and the manual identification of nucleation events from the bubble-side video recordings are discussed by von Hardenberg et al. (2002).

3. Statistical analysis of interactions

The analysis described in the preceding section provides the timing and position of 2499 nucleation events in an area 20 mm$\times$11 mm over a period of 30 s. Treating each event in turn as a 'primary' event, the delay time and relative distance S of all succeeding events are calculated. The number of events in different delay periods at increments of 25 ms are plotted against S at increments of 0.25 mm in Fig. 5, and against non-dimensional distance S/R_1 at increments of 0.125 in Fig. 6. R_1 is the radius of the NEF at the primary site at the time of bubble departure for each event, which usually occurs in the first period 0–25 ms. It is not the average value at the primary site.

Promoting or inhibiting interactions are detected by comparing the measured number of events in each spatial class and delay interval with the average number obtained by randomly shuffling the events in time 1000 times, to destroy any temporal correlation. This procedure allows for the geometrical effect of the distribution of sites so the number of events peaks at a distance approximately equal to the semi-width of the region of observation. The bold lines superimposed on Figs. 5 and 6 represent the 99% confidence limits for these values.

The only statistically significant deviations from the null hypothesis of uncorrelated events occur at small separations and short delay times. For delay times below 25 ms and $S < 1.75$ mm, $S/R_1 < 0.75$, the number of events is up to twice the expected number, indicating promotional interactions. For delay times between 25 ms and 100 ms, the number of events is lower than expected, indicating inhibition, for $S < 0.5$ mm, $S/R_1 < 0.25$.

4. Discussion

4.1. Comparison of methods with other studies

In all the studies of interactions described in Section 1, the conditions differ in some respect (method of heating, surface properties or thermal capacity of the wall) from those of industrial applications and special constraints are applied.

Gjerkeš and Golobič (2003) and Zhang and Shoji (2003) restricted activity to a pair of sites that were asymmetrical in properties in the first case, symmetrical in the second case, and used measures of activity that were averaged over time. Their different findings for the effect of distance between the sites may well be influenced by the different wall conditions and methods of heating but they suggest that asymmetry in respect of activation superheat may have an important effect on interactions. Judd and Chopra (1993) and the present study both use statistical analyses of the time delay between nucleation events to measure interaction in a group of "natural" sites. Judd and Chopra use a 'dominant' site as the primary site, implying asymmetry in some respect, which, according to their interpretation, may be stability in retaining a potentially active vapour nucleus. A similar bias may apply in the manual analysis of a small group of sites by Kenning and Yan (1996) and in the statistical analysis of the same sites by McSharry et al. (2000), which indicate activation superheat as an important property. The present study considers every nucleation event in turn as a primary event so the results should be statistically representative of the general population of nucleation sites. The test for significant deviation from randomness is applied separately to intervals of time delay, rather than fitting a single function to the entire distribution.

4.2. Absence of interactions at $S/R_1 > 0.75$

This finding is at variance with nearly all the other studies citied in Section 1, if R_1 is taken to be nearly equal to the bubble departure radius, as in the examples studied by Kenning and Yan (1996), for which the thermal disturbance appeared to be associated with evaporation from the microlayer and/or the triple interface under the bubble. Although the fluid conditions in this study, i.e. water boiling at atmospheric pressure, are similar to those in Zhang and Shoji's (2003) study, the bubble frequencies are much lower, perhaps because the low thermal diffusivity of stainless steel compared to silicon slows the recovery of local superheat following the departure of a bubble. Streams of bubbles rising in rapid succession may induce longer-range thermal disturbances than bubbles that are effectively isolated in time. Zhang and Shoji measured correlated temperature fluctuations up to a distance of $S/R_1 = 2.5$ and they attributed longer-range promotive effects to hydrodynamic disturbances. There is no statistically significant evidence for either of these effects in the present study.

According to Judd and Chopra (1993), long distance inhibition depends on intervening sites and therefore on the site density, N. The effect should therefore disappear when the ratio of average site separation to bubble radius is sufficiently large, proportional to $(N^{1/2}R_1)^{-1}$. Examples given by Judd and Chopra correspond to values of this parameter of 2.2 for strong inhibition and 2.8 for negligible inhibition. Taking the most conservative view that all the nucleation in the present study is concentrated at 16 clusters in an area 21 mm × 11 mm and that the average bubble radius is approximately 1.75 mm, the site separation parameter is 2.2, which should correspond to strong inhibition. The Judd and Chopra mechanism does not appear to apply in this study but the surface conditions are very different and the mechanism may be sensitive to the proportions of stable and unstable sites, which is difficult to determine.

4.3. Inhibition at $S/R_1 < 0.25$, delay 25–100 ms

This finding is consistent with the earlier findings of Kenning and Yan (1996) and McSharry et al. (2000) that cooling under a bubble that overlaps other sites interrupts the recovery of local superheat at those sites that must precede nucleation. The cooling effect persists for some time after departure of the bubble. The cooling effect declines towards the edge of the bubble contact area but the range deduced here statistically is less than that observed for particular examples in the earlier study. The inhibiting mechanism may be in competition with promotional mechanisms. It is likely to be affected by the thickness and thermal diffusivity of the wall. Judd and Chopra (1993) discount the effect of local variations in wall temperature on short-range interactions in their experiments, even though boiling on a glass wall might be expected to accentuate the variations.

4.4. Promotion at $S/R_1 < 0.75$, delay $\leqslant$25 ms

The short timescale suggests a hydrodynamic mechanism. Seeding of flooded unstable sites would require coverage by a dry spot under the growing bubble and nucleation at a seeded site could not occur before departure of the first bubble. There may be some other asymmetry in the interaction. The primary bubble is

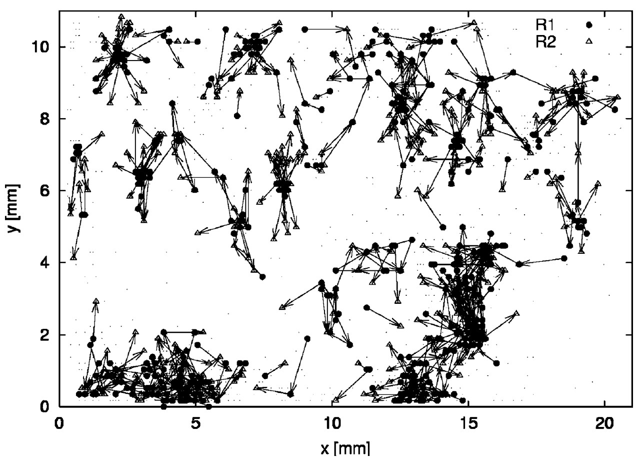

Fig. 7. Interacting sites for $S/R_1 < 0.75$, delay $\leqslant$ 25 ms Circles = primary events, triangles = secondary events, dots = all other nucleation sites.

often formed at a site producing many bubbles, interacting with several surrounding sites that only produce a few bubbles each during the period of observation, Fig. 7.

The conditions of near-simultaneous growth from two sites in close proximity are likely to lead to coalescence, identified by Zhang and Shoji (2003) as an important promoter of vapour production. Coalescence may cause cooling patterns, such as elongated patches, for which the present shape of NEF is not designed so that the identification of new nucleation events may be affected. The bubble-side record has been examined for 10 examples, which was time-consuming, and in half of them the NEF analysis correctly identifies individual nucleation events. Examples of one correct and one incorrect diagnosis are given in Fig. 8. There is an evident deviation from the expected number of random nucleation events in Figs. 5 and 6 in this range but further investigation of the identification of nucleation events from wall temperature patterns influenced by coalescence processes is required. This may require some modification of the post-processing algorithm in the NEF analysis but any technique is ultimately limited by the requirement to interpret complex dynamics on the bubble side *entirely* through the thermal footprint on the rear of the plate. Even a skilled human analyst, trained by combined observations of the bubble-side and thermal records, may have insufficient information to distinguish between a new nucleation event and cooling consequent on the coalescence of two previously nucleated bubbles. Because of the low bubble frequency in the present study, some of the modes of coalescence described by Zhang and Shoji (2003) were not observed. This suggests that the mechanisms of interaction between sites less than one bubble radius apart, like those at larger separations, may be sensitive to the surface

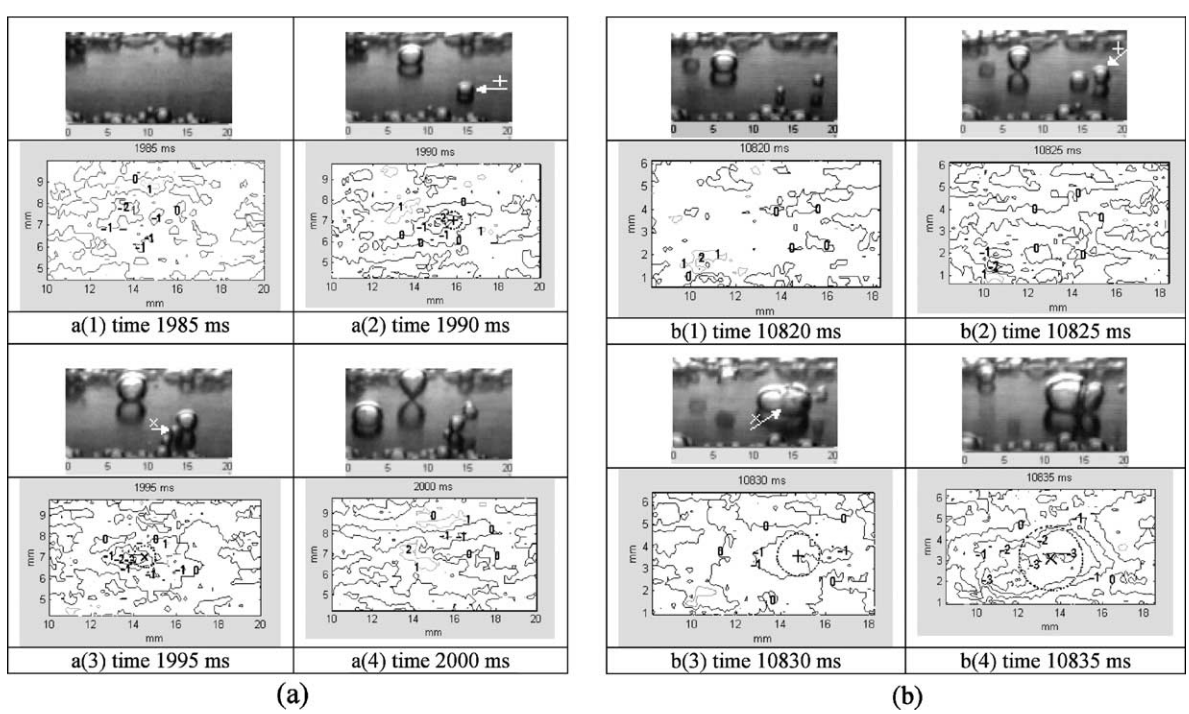

Fig. 8. Diagnosis by NEFs. Bubble images and contours of temperature change between frames: (a) correct identification of events at 1990 and 1995 ms and (b) incorrect interpretation of elongated cooled patch due to rapid coalescence 10,825–10,835 ms.

condition and properties of the wall. Judd and Chopra (1993) do not comment on coalescence in their studies of boiling on a glass wall with a thin-film heater.

5. Conclusion

This paper has described a new statistical method of testing for interactions between all members of a group of nucleation sites, using only data for spatio-temporal variations in temperature measured by liquid crystal thermography on the rear of a thin, electrically heated plate. Promoting and inhibiting interactions between sites less than one bubble radius apart at different times after the primary nucleation event were found to be statistically significant, although the influence of bubble coalescence on the interpretation of promotion requires further investigation. There was no evidence of significant interactions at larger distances.

These findings may well depend on the particular conditions of the experiment, i.e. boiling on a thin, well-wetted plate, which caused low frequencies of bubble production. The differences between the findings of this study and three other studies discussed in the paper, all for different special conditions, confirm that interactions cannot be defined only in terms of the ratio S/D of the distance between a pair of interacting sites and the bubble diameter. Further understanding is required of the influence of factors such as asymmetry in the characteristics of the sites, including their activation superheat and stability, the presence of other active sites and the properties of the wall.

Acknowledgements

This research was supported by EPSRC Grant GR/M 89034.

References

Calka, A., Judd, R.L., 1985. Some aspects of the interaction among nucleation sites during saturated nucleate boiling. Int. J. Heat Mass Transfer 28, 2331–2342.

Chekanov, V.V., 1977. Interaction of centers in boiling. Teplof. Vysok. Temp. 15, 121–128.

Del Valle, M.V.H., Kenning, D.B.R., 1985. Subcooled flow boiling at high heat flux. Int. J. Heat Mass Transfer 28, 1907–1920.

Eddington, R.I., Kenning, D.B.R., 1978. The prediction of flow boiling populations from gas bubble nucleation experiments. In: Proceedings of the 6th International Heat Transfer Conference, Toronto, vol. 1, 275–280.

Gjerkeš, H., Golobič, I., 2003. Measurement of certain parameters influencing activity of nucleation sites in pool boiling. Exper. Therm. Fluid Sci. 25, 487–493.

Judd, R.L., Chopra, A., 1993. Interaction of the nucleation processes occurring at adjacent nucleation sites. J. Heat Transfer 115, 955–962.

Kenning, D.B.R., Yan, Y., 1996. Pool boiling heat transfer on a thin plate: features revealed by liquid crystal thermography. Int. J. Heat Mass Transfer 39, 3117–3137.

Kenning, D.B.R., Kono, T., Wienecke, M., 2001. Investigation of boiling heat transfer by liquid crystal thermography. Exper. Therm. Fluid Sci. 25, 219–229.

Mallozza, R., Judd, R.L., Balakrishnan, N., 2000. Investigation of randomness, overlap and the interaction of bubbles forming at adjacent nucleation sites in pool boiling. Int. J. Heat Mass Transfer 43, 3317–3330.

McSharry, P.E., Smith, L.A., Kono, T., Kenning, D.B.R., 2000. Nonlinear analysis of site interactions in pool nucleate boiling. In: Proceedings of the 3rd European Thermal Sciences Conference, Heidelberg 10–13 September 2000, vol. 2, pp. 725–730.

McSharry, P.E., Ellepola, J.H., von Hardenberg, J., Smith, L.A., Kenning, D.B.R., Judd, K., 2002. Spatio-temporal analysis of nucleate pool boiling: identification of nucleation sites using non-orthogonal empirical functions. Int. J. Heat Mass Transfer 45, 237–253.

von Hardenberg, J., Kono, T., Kenning, D.B.R., McSharry, P.E., Smith, L.A., 2002. Identification of boiling nucleation sites by non-orthogonal empirical functions (NEF) analysis of thermographic data. In: Proceedings of the 12th International Heat Transfer Conference, Grenoble, 18–23 August 2002, vol. 3, pp. 377–382.

Zhang, L., Shoji, M., 2003. Nucleation site interaction in pool boiling on the artificial surface. Int. J. Heat Mass Transfer 46, 513–522.