



Review

Studies of boiling chaos: a review

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Abstract

Research into boiling has been carried out over the last several decades and extensive data have accumulated from experimental studies carried out under various conditions and configurations, leading to the development of the currently available empirical and phenomenological correlations. However, most correlations apply to situations under a relatively narrow range of conditions and exhibit a considerable error band, even for the data sets on which they are based. In contrast to this multitude of correlations, the development of mechanistic models based on the underlying physical processes has been sporadic and limited. Thus, it is said that the use of boiling is not yet a “mature” technology, though it is one of our important technologies. This paper summarizes recent developments in research addressing the nonlinear dynamical behaviors of pool nucleate boiling for the purpose of suggesting some potential reasons why we have had limited success in mechanistic modeling, and to provide a promising perspective on future boiling research. In the introductory section, the problems facing current boiling research are described. In the following two sections, recent nonlinear experimental as well as theoretical research achievements are overviewed, while in the last section, the problems that remain unsolved are discussed, along with some concluding remarks.

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Keywords: Pool nucleate boiling; Conjugated boiling problems; Nucleation site interaction; Boiling chaos; Nonlinear dynamics

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1. Introduction

It is believed that humans may have started boiling a few thousands of years ago. Scientific research into boiling and the industrial use of boiling spans several decades. Starting from the pioneering paper of Nukiyama [1], extensive data have accumulated from experimental studies dealing with a diverse array of conditions. The accumulated data have led to the development of both empirical and phenomenological correlations, and many such correlations have been incorporated into design and analysis methods. Most correlations, however, are applicable to only a relatively narrow range of conditions. In contrast to this multitude of correlations, the development of mechanistic models based on the underlying physical processes has been

sporadic and limited. Thus it is said that the use of boiling has yet to develop into a mature technology, despite it being one of our important technologies. In recent years, new boiling applications to systems such as micro-mini scales, highly transient, or reduced-gravity conditions have come to light, so a full understanding of the boiling phenomenon is urgently required. This article summarizes recent developments in research, addressing the nonlinear behaviors of boiling in order to suggest some potential reasons why we have had limited success in the mechanistic modeling of boiling, and to provide a promising perspective on future boiling research.

This article focuses primarily on pool nucleate boiling because it provides a simple configuration from which it is easy to address fundamental issues. Nucle-

ation boiling has been utilized extensively in industry because it is one of the most efficient modes of heat transfer, particularly in high-energy-density systems such as nuclear reactors, power plants, electronics packaging and the like. The reason for this is that nucleate boiling is capable of transferring amounts of energy many times greater than those transferred by convection or conduction. This undoubtedly comes from phase changes (bubbles generation) on the heated surface. Thus, the main purpose of boiling research has been to obtain the correlation between the heat transfer and the surface superheat, including the parameters related to phenomena taking place on the heated surface.

1.1. Conjugate problem of boiling

Nucleate boiling involves many processes and sub-processes, as shown in Fig. 1. This includes three aspects of the liquid, the heated wall, and the liquid–wall in-

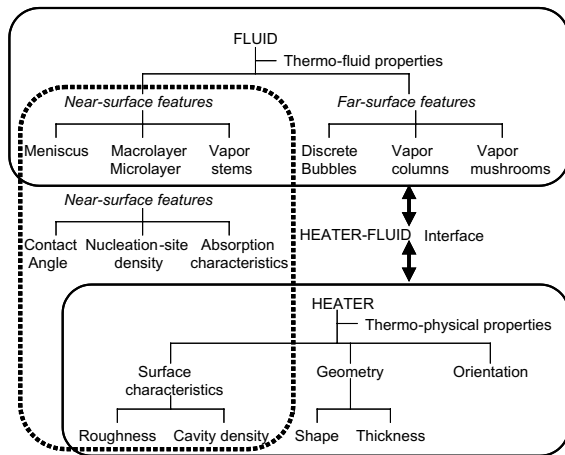
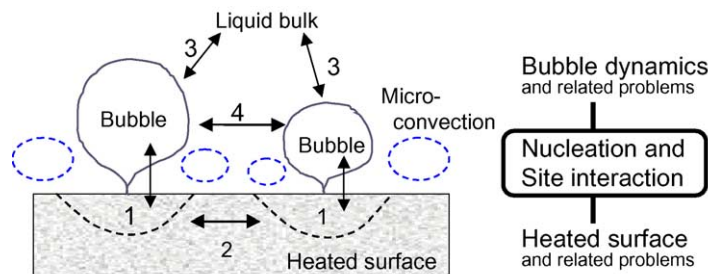


Fig. 1. Conjugate problem of pool boiling.

terface. Most of the processes are nonlinear in nature, interacting in a complex manner. In terms of boiling heat transfer, the most important but difficult process is the nucleation and site interaction that takes place at the liquid–wall interface. The interaction can be divided into four main types, as shown in Fig. 2. This involves problems associated with the liquid side as well as with the wall side. For the heated wall, the thermal interaction determines the temperature distribution. This includes two aspects; namely the thermal interaction between the nucleation site and the heated wall (interaction 1), and the thermal interaction between nucleation sites (interaction 2). On the liquid side, the hydrodynamic interaction dominates the behavior of the bubbles, which can be further divided into the hydrodynamic interaction between the bubble and the liquid bulk (interaction 3) and the hydrodynamic interaction between the bubbles themselves (interaction 4). Furthermore, interactions 1 and 3 can be considered to be auto interactions at a single nucleation site, while interactions 2 and 4 exist between two adjacent nucleation sites. Each interaction includes many hydrodynamic and thermal problems: the wall-related problem includes the nucleus distribution, the activation and deactivation of the nuclei, nucleation, the site interaction, and so on. The bubble-related problem includes micro-layer and macro-layer formation at the base of a bubble, the triple evaporating meniscus at the periphery of the bubble base, bubble departure, bubble coalescence and interference, and bubble-induced micro-convection and its contribution to heat transfer. Yet, if the underlying mechanisms of such interactions were better clarified, it would be possible to predict the rate of boiling heat transfer a priori and to design and manufacture boiling surfaces to specification. More importantly, the ability to predict the boiling condition from the surface characteristics would enable the performance of boiling heat transfer surfaces to be optimized.



Interactions on a Heated Surface.

- 1: Thermal interaction between bubble and heated surface.
- 2: Thermal interaction between nucleation sites.
- 3: Hydrodynamic interaction between bubble and liquid bulk.
- 4: Hydrodynamic interaction between bubbles.

Fig. 2. Classification of nucleation site interaction.

1.2. Problems and perspectives of boiling research

Historically, an averaging procedure has been employed for deriving heat transfer correlations for boiling heat transfers. Most of the correlations have been proposed as relationships between the heat flux and the wall superheat for a steady-state or a quasi-steady-state condition, averaged with respect to both time and space. While the averaging procedure is useful for deriving the correlation, if it is not accompanied by a physical understanding of the underlying processes, it hides the dynamical aspects and restricts the application of the correlation to limited conditions. In the averaging procedure, temporal averaging requires a certain minimum amount of time, δt_{\min} , and a certain minimum area, δA_{\min} , over which the average must be taken to obtain a statistically meaningful result [2]. If either the transient changes are faster than δt_{\min} or a time resolution less the δt_{\min} is required, the quasi-steady assumption is not valid and an instantaneous representation of the heat flux and temperature is required. Similarly, if consideration of either the process where the scale of the phenomenon is influenced by the scale of the apparatus or the area is less δA_{\min} , a spatial-scale sensitivity is produced and a local representation of the heat flux and/or temperature is needed. In many of the systems associated with recent technologies that are encountered, such as electronic chip cooling, micro-mini thermal equipment, bubble jet printers and amorphous material production, the older averaging-based correlations cannot be applied.

In the framework of classical boiling research, the factors or processes affecting boiling phenomena are often considered separately as independent variables. Typically, in terms of the heated wall, it is usual to assume a uniform wall temperature by considering that the density of the active nucleation sites can be obtained directly from a specified size distribution and an activation superheat relationship, and that each active site makes the same contribution to the heat flux. Thus, increasing the wall superheat yields progressive additions to the population of active sites. This sort of model (principally a linear model containing both implicit and imprecisely defined averaging procedures) results in a static picture of the active sites, which conflicts with observations of hysteresis, intermittent activity, the deactivation of sites, and above all, the number of active nucleation sites when the wall superheat (and, as a result, the heat flux) is increased, as found by Judd and his coworkers [3–10] and as well as summarized by Kenning [11]. Thus, in most of the classical models, the conjugate problem in terms of the heater, and how the heater is represented, is separated and ignored by assuming a uniform wall temperature. However, the key issue in boiling research is not only to understand the separate underlying processes but also to consider the interactions among processes. This statement may relate to the

suggestion of Dhir [12] in that although there are numerous subprocesses involved in boiling, one of the key issues is the density of the active nucleation sites and the relationship between active cavities. A similar statement was given more definitively by Kenning [11] who suggested that the mechanistic modeling efforts thus far have overlooked some of the key characteristics of the boiling system, one of which is the issue of spatial and temporal variations on the heated surface. This point of view of Kenning led Sadasiavn et al. [13] to discuss boiling as a locally instantaneous conjugate problem and to suggest that improvements in the mechanistic modeling of nucleate boiling would only result when viewed from this perspective (Fig. 1). The importance of wall temperature fluctuations in mechanistic modeling was exactly the consensus that was reached by the boiling specialists after vigorous discussion and summarized by Bar-Cohen at the boiling conference held in Banff, Canada, in 1994.

For many years, the complexity of boiling phenomena has been attributed to nuisance variables such as the aging of the heated surface or unknown noise imposed from outside the system. However, according to recent advances in nonlinear chaos dynamics, it is known that even in noise-free simple deterministic systems, “noisy” information can be generated as a result of nonlinearity (e.g., Moon [14], Ott et al. [15]). Most of the problems and subprocesses of boiling in Fig. 1 are fundamentally nonlinear in nature. Therefore, nonlinear chaos dynamics has the potential to be a powerful analysis tool for clarifying the underlying physical processes of boiling. Using this new form of dynamics, it is possible to better understand the dynamical structure of the phenomena and to evaluate quantitatively the complexity. In the last few years, study has begun into a new approach to boiling from a promising perspective based on nonlinear chaotic dynamics in the framework such as that shown in Fig. 3. The main purpose of the new approach is to find the nonlinear elements involved in boiling and to clarifying the dynamic mechanism that yields the observed complex behaviors. Some works

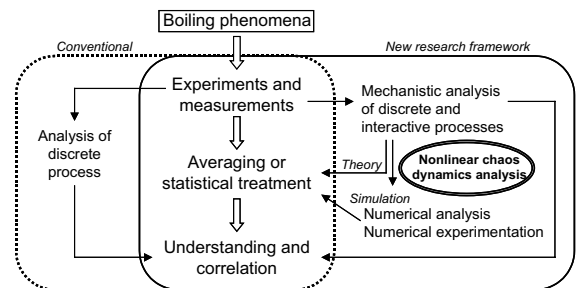


Fig. 3. Perspective on boiling research: classical and new approaches.

show that boiling is one example of nonlinear spatio-temporal systems where even very small systems exhibit very complex behavior. They also suggest that such nonlinear behavior is one of the reasons why the mechanistic predictive capabilities for the boiling process have remained elusive. This review article is concerned with such research. The achievements in western countries in the early stage were reviewed by Nelson et al. [16] and Shoji [17], and for those in eastern countries by Shoji [18]. The present article summarizes such achievements, including some others not directly connected to nonlinear research but suggestive of the nonlinear nature of boiling.

In connection with the present topic, it is noted that nonlinear chaotic theory is well suited to two-phase-flow boiling systems. With regard to the chaotic phenomena and nonlinear methods of analysis in that field, the reader is referred to the review articles of Lahey [19–21]. For the nonlinear dynamics and chaos of heat transfer and fluid flow, the reader is referred to other literature (e.g., Dorning [22,23] and Arpaci et al. [24]).

2. Nonlinear experimental studies

Experiments focusing on the nonlinear chaotic features of pool boiling are rather limited. The serial works of Shoji and his coworkers in Tokyo, Kenning and his coworkers in Oxford, and Golobič and his coworkers in Slovenia are exceptions to this, and will be described in this section. Some topical results found by Fong et al. in Canada on the fractal roughness effects on critical heat flux and by Kuzma-Kichita in Russia on bubble surface nonlinear oscillations will also be described briefly.

2.1. Wall temperature fluctuations: the complexity of boiling

In order to be able to discuss the chaotic nature of boiling, we need temporal or spatio-temporal information. In many nonlinear experiments of boiling, the wall temperature fluctuation has been measured and been found to be a good indicator of the integrated effects of the conjugate processes of boiling.

2.1.1. Temporal spatially averaged characteristics on a wire

Shoji et al. [25] studied experimentally the complexity of boiling phenomena exhibited within a small simple configuration. They boiled saturated water on a platinum wire, 0.05 mm in diameter and 0.8 mm in length, under atmospheric pressure. The wire acted not only as a heater but also as a sensor for temperature measurements. Every mode from nucleate to film boiling was realized, and a high-speed video camera was used to

observe the hydrodynamic features in the various boiling regimes ranging from a few isolated bubbles on the wire at low-heat-flux nucleate boiling to only one coalescence vapor mass over the test wire at high heat-flux of nucleate, transition and film boiling. The wire temperature fluctuations were detected using the electrical-resistance method with a sampling rate of 50 KHz. This method produced an indication of the volume-averaged wire temperature. To address the nonlinear nature of the boiling, Shoji et al. used the temperature time series, Fourier spectrum and attractors (trajectories) reconstructed from the original data using the embedding method and the fractal dimension (correlation dimension) of the attractor calculated using the method of Grassberger and Procaccia [26–28]. It was found that the wall temperature fluctuations differ depending on the boiling mode and that the fluctuation is largest in transition boiling where the maximum fluctuation reached around 70 K. The attractor of transition boiling shows an intermediate structure, indicating that both nucleate and film boiling partly occur. The results of fractal dimensions show that the level of complexity is highest around CHF and decreases somewhat in nucleate and transition boiling. Film boiling is the simplest phenomenon measured in the experiment. These results suggest that chaotic behavior is involved in the boiling system. This conclusion was further supported by analysis of the power spectrum, found to be definitely broad-banded for nucleate as well as transition boiling.

2.1.2. Spatio-temporal characteristics on a flat surface

The experimental results of Shoji et al. [25] in the previous section suggest that significant nonlinear effects occur in boiling. These are important conclusions, but they provide no clarification as to what creates such behavior. Kenning and his coworkers [29–32] and Ellepola [33] provided valuable information along this line which relates primarily to the heater side problem, including the interfacial and hydraulic aspects of the problem (see Figs. 1 and 2).

Kenning and Yan [29] boiled water at atmospheric pressure on a horizontal stainless-steel plate of 28 mm×41 mm in size and 0.125 mm in thickness, and measured the locally instantaneous temperature distributions on the back side of the heater using liquid crystal thermometry combined with high-speed video recording at 200 Hz. In all of the tests, bubble motion on the surface was simultaneously recorded by video. The video frames were digitized, and a detailed analysis was performed on a central area of 11 mm×20 mm. It was found that, even when time-averaged, the wall superheat was far from uniform. The instantaneous distributions of the wall superheat during bubble nucleation and growth were measured, and the changes in wall temperature during bubble growth and departure were found to be confined to a circular region corresponding

to the maximum projected area of the bubble. The heat removal seemed to be consistent with the evaporation of a liquid micro-layer meniscus. The rate of heat removal during bubble departure was much less than that predicted by the model of wall quenching by the bulk liquid; however, the general level of convective heat transfer between bubbles was enhanced. A pattern-recognition program for the cooled spots identified nucleation sites and the size of the bubbles they produced, with confirmation from a direct view of the boiling surface at low heat fluxes. It was then possible to obtain the wall temperature time series at individual sites. These site characteristics showed that nucleation depends on the locally instantaneous wall superheat and the triggering wall superheat. A striking feature was the intermittent nature of bubble production, even at the most active sites. These sites had an identifiable base frequency that depended on the rate of recovery from the local cooling during bubble growth; however, the recovery was often interrupted. In a limited study of a small group of sites, nearly all of the interruptions were identified with the cooling effect of bubble growth at an adjacent site. Some sites produced only an occasional bubble; however, these could have had a significant effect on the more active sites. With an increase in the heat flux, there was an exchange of activity, thus suppressing the sites that were active at the lower flux. The increasing heat flux also caused a general increase in the site-triggering superheating.

2.1.3. Local spot characteristics on a flat surface

Thermocouple measurements have long been a part of boiling experiments. Such measurements have generally involved the time averaging of thermocouples embedded in the heater so that the detailed temporal temperature information necessary for nonlinear analysis has not been available. However, the recent measurements below have provided the necessary data to identify the local chaotic characteristics on a horizontal flat surface in saturated pool boiling.

2.1.3.1. Embedded thermocouples. The experiments of Shoji et al. [34] were devised to provide high-resolution temporal temperature measurements using micro-thermocouples located on a heated surface. The boiling experiment was performed using water as the test fluid at saturated conditions under atmospheric pressure. The boiling surface was a 20 mm diameter copper surface, finished with 600-grade sand paper, with six Chromel–Alumel fine thermocouples located on the surface with the appropriate spacing (mostly 2 mm). The thermocouple junctions were embedded in the heater at a depth of 0.1 mm from the surface. The response of the thermocouples was about several kHz. The recording of temperature fluctuations was made for 30 s with a 1 kHz sampling rate at each of the spots. A PID control

technique was employed to stabilize the transition boiling. This paper of Shoji et al. [34] discussed the nonlinear characteristics of the boiling system by employing a temperature time series, its power spectrum (Fourier spectrum) and the attractor reconstructed from the time series using the embedding method.

It was found that although the spot temperature shows different aspects in terms of fluctuations from spot to spot on the heated surface, it includes two modes of fluctuations with either small amplitudes and high frequencies or large amplitudes and low frequencies. The former fluctuation mode may be attributed primarily to bubble generation and departure, and the latter to vapor mass generation and departure. The former was observed in the wide regime of nucleate boiling while the latter was observed only in the high-heat-flux nucleate boiling regime. For most boiling conditions, the Fourier spectrum was broad-banded and the reconstruction of data showed some structure, probably suggesting chaotic behaviors. Also, the analysis of the spot temperature time series was undertaken in terms of the correlation dimension and the maximum Lyapunov exponent calculated using the algorithm of Wolf et al. [35]. All points on the boiling curve exhibit positive Lyapunov exponents, supporting the above suggestion of the existence of chaotic behavior. In many of the cases, however, the complexity of the behavior appears so great (suggested by a high correlation dimension) that convergence is not obtained. Thus, they concluded from these results that chaotic behavior probably exists in pool boiling on a flat surface but that the behavior at high-heat-flux nucleate boiling is likely to be random with high degrees of freedom.

A similar investigation was performed by Shoji et al. [36] on a smaller heated disk of 2 mm in diameter, where only a few bubbles were generated. The results were almost the same to those found in the previous work of Shoji et al. [34] on the larger heated surface. Another objective of this experiment was to capture the nonlinear features of boiling from the liquid side through bubble generation. These results are described later.

2.1.3.2. Liquid crystal thermography. It is difficult to fabricate arrays of miniature temperature sensors to a sufficient extent to be able to measure the varying wall temperature around a large number of nucleation sites. To provide a two-dimensional instantaneous wall temperature distribution, Ellepola and Kenning [32] performed an experiment, similar to that of Kenning and Yan [29], using thicker copper plates of 0.05 and 0.1 mm in thickness. Video recordings were analyzed to obtain the local cooling caused by individual bubbles and the temperature time series under individual members of a group on nucleation sites. Bubble growth commenced whenever the local temperature reached the activation superheat for a particular site. Cooling, caused by

bubbles produced at adjacent sites overlapping the site, disrupted the regular production of bubbles. This behavior is of the same general nature as for the case on a stainless-steel plate [29], except that the temperature fluctuations occur at shorter intervals. An attempt was made to construct an attractor in two dimensions for a special time series using the method of delays. However, noise in the liquid crystal data prevented the determination of the fractal dimension. So Ellepola [33], supervised by Kenning, employed the singular-value-decomposition method (SVD) to reduce the signal noise generated by the video recording/replay system. Thus, they succeeded in constructing an attractor to determine the fractal dimension and to evolve the Lyapunov exponents for the SVD data. The structure evident in the attractor and the positive Lyapunov exponents for the embedding dimensions thus obtained are indicative of low-dimensional chaos in the system [37]. In addition to such analysis, Ellepola proceeded bravely with a trial for predicting the future values of the filtered time delays one step ahead to within ± 0.1 K (the accuracy of the temperature calibration) using radial basis functions. Recently, by using a nonorthogonal empirical function (NEF), Mcsharry et al. [38–40] identified the nucleation sites and discussed the site interactions.

2.1.3.3. Radiation thermography. A radiation thermometer may be another possible tool for measuring the heated wall temperature. It has, however, been rather difficult to obtain detailed and accurate data because of the limited capacity and quality of the conventional thermometer. Fortunately, recent developments in equipment have made it possible for us to satisfactorily record spot temperatures and line temperature distributions. This was employed by Shoji and his coworkers [41–46] in their serial experiments of boiling on artificial surfaces, which are described later in the section on “Nucleation site interaction”.

Summarizing the works of the Tokyo and Oxford groups mentioned above, it is said that low-dimensional chaotic dynamics seem to exist in the boiling system of small or locally simple configurations. Their results also indicate that even a simple system of boiling exhibits rather complicated features, and that the complexity increases with increasing scale of the boiling system. It is therefore not surprising that modelers have had difficulty in developing mechanistic models for actual boiling systems.

2.2. Nonlinear bubble generation

2.2.1. Bubbling features in boiling

Bubble generation affects boiling heat transfer through micro-layer or macro-layer formation and evaporation at the bubble base, evaporation or con-

densation at the bubble surface, and heat transfer due to micro-convection induced on a heated surface. So many researchers have developed an interest in the bubble dynamics of boiling. Most have concerned themselves with the growth rate, the departure size and the frequency of an isolated or coalesced bubble in relation to heat transfer (e.g., Gorenflo et al. [47]). Only a few recent studies have addressed the nonlinear features.

2.2.1.1. Nonlinear bubbling features. To reveal the nonlinear behavior of bubbling in pool boiling, Shoji et al. [36] carried out a boiling experiment to measure the liquid motion (micro-convection) induced by a growing bubble using a hot wire anemometer located slightly above a heated surface. In order to avoid producing numerous bubbles, a small heater consisting of a disc 2 mm in diameter was employed. The experiment was conducted in a copper–water system for a saturated condition and atmospheric pressure. The strength of the micro-convection was detected as a time series for various heat-flux conditions. From the time serial data, the Fourier spectrum and the Lyapunov exponent (maximum exponent) were calculated for discussing the nonlinearity and complexity. The maximum Lyapunov exponent obtained turned out to always be positive. This result, together with the broad-banded power spectrum, indicated the existence of chaotic behavior in bubble generation. It was difficult to clarify the mechanism associated with such nonlinear bubbling behaviors due to the numerous interactive factors. For this reason, Shoji and his coworkers performed studies of bubble dynamics in nucleation-site-regulated systems [41–43], and in air-water isothermal systems [48–50]. These will be described in the next section.

2.2.1.2. Interaction between bubbles: coalescence characteristics. Bubble coalescence, which is a special bubble interaction in boiling, has evoked the interest of many researchers. Some preliminary conclusions have been drawn from experimental investigations. Williamson and El-Genk [51] and Buyevich and Webbon [52] classified the coalescence into three types: (a) lateral coalescence far away from the heated wall, which had no effect on boiling heat transfer, (b) vertical coalescence between consecutive bubbles near the wall, and (c) lateral coalescence between adjacent bubbles near the wall. Yang et al. [53] also presented similar results obtained by numerical simulation. Recently, Zhang and Shoji [44,45] and Shoji and Zhang [46] discovered the existence of a fourth type of coalescence, (d) lateral coalescence between adjacent declining bubbles near walls. In their experiment of a simplified boiling system employing regulated artificial cavities, they reported that coalescences of types (c) and (d), which are both important in heat transfer indicating an interaction

between adjacent bubbles, take place only when the distance between nucleation sites is less than 1.5 times the bubble departing diameter.

In relation to boiling heat transfer, Bonjour et al. [54] pointed out that adjacent bubble coalescence allowed the vaporization of a larger micro-layer volume. Indeed, the total micro-layer volume was found to be the sum of the volume of the liquid between the stems and the volume of the micro-layer of each bubble. This supplementary micro-layer vaporization increased the latent heat transfer and locally reduced the wall temperature. However, if a large number of adjacent bubbles coalesced, wall dry-out occurred, which implied a heat transfer impairment or a triggering of the critical heat flux.

2.2.1.3. Oscillation of bubble surface: subcooling effects. Kuzma-Kichta [55,56] investigated a special boiling feature of bubble interface oscillations during boiling using acoustic diagnostics and a laser method. Under the subcooled liquid condition, the bubble fluctuates in size before departing from the heating surface because of the simultaneous effects of evaporation and condensation. The bubble surface oscillation causes a complex micro-convection of the liquid around the bubble, affecting heat transfer on a heated surface. They succeeded in obtaining beautiful trajectories of the interface motion, revealing fractal and chaotic characteristics. The bubble fluctuation phenomenon was also observed by Yasui and Shoji [57] in subcooled boiling on an artificial surface with a fabricated cavity. As for the oscillation phenomenon mentioned here, interesting results have been reported by Fuchigami and Kiyono [58], and Kiyono and Fuchigami [59]. They dealt with the density-reversed system of a dripping faucet, showing that the frequency of droplet formation relates directly to oscillations of the droplet surface.

2.2.2. Nonlinear bubbling experiments in isothermal systems

Bubbles are widely used in industry such as in chemical plants and the like, and numerous studies on bubbling features and dynamics have been available in the past. Most studies have dealt with a single isolated bubble (e.g., Cliff et al. [60]). This belongs to the older research category in the sense that the authors do not consider interactions with adjacent or preceding bubbles. In contrast with the multitude of studies that fall into the older category, a few recent studies have observed the nonlinear features of bubble formation from submerged orifices [61–65]. Shoji [49] and Zhang and Shoji [50] successfully produced air bubbles from a single orifice submerged in water for various air flow rates for the purpose of comparing the result with that in boiling on a small heated disc [36]. In the experiment,

the diameter of the nozzle was set to the same as that used in the previous boiling test (2 mm), and the local velocity of liquid motion induced by the successively generated bubble was measured using a hot wire anemometer set close to the orifice, as was carried out in the boiling experiment [36]. The time series of the hot wire signal was analyzed for measures of chaos dynamics, and it was concluded that the trivial system displays very complex behavior as the preceding bubble had an effect on the fluid around the nozzle, and influenced the production of the following bubble. Shoji [49] clarified further the nonlinear characteristics of bubble formation and departure to indicate that the bubble departure frequency shows a bifurcation similar to the period-doubling route to chaos when the air flow rate is increased. Zhang and Shoji [50] proposed a nonlinear model that successfully explains the phenomenon.

The results discussed above are in relation to a single successively generated bubble. The situation becomes more complex for the case in which bubble generation is from multiple orifices. Actually, in the case of twin orifices, very interesting but complex bubbling modes (regular and periodic, irregular and chaotic bubble formation) repeatedly appear with changes in the air flow rate. The mechanism of the peculiar phenomenon has been made clear theoretically by Tange and Shoji [66].

Chaotic bubble behaviors observed in isothermal systems are not directly connected with those in boiling systems (e.g., see the paper of Di-Marco et al. [67] concerning bubble coalescence). However, it is clear that even in a simple system of bubble generation, the behavior is not simple. Hence, the more complex system of boiling could lead to significantly more complex behaviors in bubble generation and the interaction between the liquid and the wall.

2.3. Surface roughness and fractal structure

Surface roughness is the most important factor in boiling heat transfer as it closely relates to the nucleation site for bubble generation (e.g., Luke [68]). However, it is difficult to discuss the effects of surface roughness on nucleation because nucleation does not merely relate to the geometric size of the roughness but also to aging, oxidation, and wettability [69]. Moreover, the actual solid surface has numerous cavities of various shapes and sizes. In contrast, we have no proper physical measures for capturing such a surface property; at least it is impossible to represent the surface property only by the “height” of the roughness. In the older framework of boiling research, the roughness size has been measured by assuming that nucleation takes place depending on its size and the wall superheat required to nucleate [70]. This kind of treatment has serious problems in mechanistic modeling, as mentioned in Section 1. The study of

how to represent the solid surface in boiling is elusive but the study of Fong et al. [71,72] discussed below provides valuable insights.

In relation to CANDU reactors, Fong et al. [73] had an interest in enhancing the CHF (critical heat flux) on zirconium alloy calandria tubes by treating the tube surfaces. They employed glass peening to successfully increase the CHF. In order to correlate the enhancement of CHF as a function of the property of the treated surface, they not only measured the height of the surface roughness but also evaluated the surface characteristics of the micro-topology by the “fractal” roughness using stereo-pair micro-graphs obtained from scanning electron microscopy (SEM) and photogrammetry techniques. Thus, they found that CHF is well correlated with the fractal roughness and not with the roughness height [71,72]. This result is very suggestive in terms of the nonlinear research of boiling because the fractal feature is known to be the geometrical structure of chaotic dynamical systems.

2.4. Nucleation site interactions

2.4.1. Nucleation site interactions in boiling

The interaction between heat transfer and bubble formation is the most fundamental issue in nucleate boiling [47], where the nucleation site interaction plays the essential role. So over the past 30 years, the relationship between bubble behavior and cavity spacing has been studied experimentally by several authors. Chekanov [74] performed the earliest experiments in order to investigate the interaction between two artificial nucleation sites, which were created by two heated copper rods that were placed in contact with a thick Perm-alloy plate covered with water. The experimental results revealed that the elapsed time between the bubble departures at neighboring nucleation sites was random and possessed a Gamma distribution. When the dimensionless cavity spacing, S/D (S : cavity spacing, D : bubble departure diameter) was less than three ($S/D < 3$), the formation of a bubble at one nucleation site inhibited the formation of a vapor bubble at the other nucleation site, while for $S/D > 3$, the growth of a bubble on one nucleation site promoted the growth of a bubble on the other. When $S/D \gg 3$, there were no interactions. Cheknov postulated that the bubbles affect one another by acoustic actions and by hydrodynamic mixing but gave no further explanations of the interaction mechanisms. Judd and his coworkers [3–10] investigated the interaction phenomena occurring at adjacent nucleation sites on a transparent glass surface with a thickness of 3.6 mm. Dichloromethane was caused to boil on the glass surface, which was coated with a 0.3 μm thickness of stannic oxide that conducted electrical current and permitted heat to be generated. In order to decrease the number of active sites and to increase the

size of the bubbles, as well as to enable operation at a lower surface temperature thereby avoiding damage to the heat transfer surface, the pressure in the vessel was maintained at lower than atmospheric pressure and the input heat flux was controlled. The experimental results were analyzed by a similar method to that of Chekanov [74] and concluded that when a nucleation site that is unable to capture vapor nuclei lies within the area influenced by a continually active nucleation site that can deposit nuclei in it, bubbles will form at the adjacent nucleation site more frequently than would otherwise be the case. This type of interaction, which was observed to occur when $S/D < 1$, is said to be “promotive”. When a nucleation site that is unable to capture vapor nuclei lies within the area influenced by an intervening nucleation site capable of depositing/displacing nuclei in it, that is itself under the influence of a continually active nucleation site, bubbles will form at the adjacent nucleation site less frequently than would otherwise be the case. This type of interaction, which was observed to occur when $1 < S/D < 3$, is said to be “inhibitive”. When bubble formation at one nucleation site is in no way influenced by bubble formation at another nucleation site, the events are said to be “independent”. This type of interaction was observed to occur when $S/D > 3$. They applied the assumption of “site seeding” to interpret the experimental results. Kitron et al. [75] presented a stochastic model for describing the boiling site interaction. Baldwin et al. [76] and Bhavnani et al. [77–79] studied the bubble size and latent heat contribution for different cavity spacings and heat fluxes on the 0.6 mm thick silicon wafer, which was immersed in FC-72. They found that the latent heat dissipation is only a minor part (<16%) of the total heat flux being dissipated. From a heat transfer application point of view, the single-spaced test surface was superior to the plain, double, and triple surfaces in terms of maintaining low wall superheats, and hence lower operating temperatures for the heat source. However, it featured a rather low ceiling of operation governed by its low departure from the nucleation boiling value (DNB). This low DNB, brought about by excessive bubble coagulation near the surface, was the result of nucleation sites whose primary areas of influence overlapped.

The above research has provided valuable basic information about nucleation site interactions and their relationship to boiling heat transfer. However, no consideration was paid to the interaction of the nucleation sites with the heated wall. In contrast, as mentioned in Section 2.1, Kenning and Yan [29] investigated the temperature fluctuation beneath the bubble growth region on a 0.13 mm thick, electrically heated stainless-steel plate by recording the color-play of a thermo-chromic liquid crystal layer on the back of the plate. The measurements confirm the importance of variations in wall temperature for the removal of heat by bubbles and

the activity of nucleation sites. It was found that throughout the growth and departure of a bubble, its direct cooling effect was remarkable and was confined to its maximum contact area with the wall. The radius of the “area of influence” was equal to the maximum bubble radius, and the cooling effect decreased near the outer edge of this area. They noted that the nucleation sites located within one bubble radius of each other interacted through the fluctuations in wall temperature caused by bubble growth.

The complexity of nucleation site interaction can be partially attributed to some hydraulic factors primarily related to bubble behaviors on the heated surface. As described above, the frequency of bubble formation at different nucleation sites is not equal [8] and so the bubble formations at different sites must be interconnected. The hydrodynamic interactions due to the non-uniform bubble formation may affect the activation of nucleation [6], together with the thermal interactions between the sites through the heater [29,32,80]. Thus, the irregular activity of nucleation sites may surely be caused by a combination of hydrodynamic as well as thermal interactions. However, no useful information has been revealed with regard to the hydrodynamic interaction. The recent research described below considers these topics by employing simplified nucleation sites regulated using recently advanced laser and micro-machining techniques.

2.4.2. Controlled nucleation sites (1): laser beam splitting method

To study the behavior of nucleation sites and bubble generation, Golobič and his coworkers [81–87] presented a new experimental method in which the nucleation sites were spatially controlled using a laser beam. They used 25 μm thick copper and titanium foil submerged in a saturated water pool and heated the backside of the foil using a laser. The laser was split into four proportional beams of equal diameter, and to study the interactions between the two, three and four simultaneously active nucleation sites in various arrangements, the diameter of each circular heated area, as well as the distance between them, were changed. They observed the bubble formation in columns from artificially activated nucleation sites and naturally activated nucleation sites on their activity and activation–deactivation of the sites. Their measurements indicated that the nucleation sites exert effects on each other and that when the nucleation site spacing is reduced, the individual and total activity of the nucleation site are reduced. At very small spacings, the deactivation of a weaker, less-active nucleation site might occur. Thus, they concluded that the intensity of the interaction depends not only on the distance-to-diameter ratio but also on the diameter of the heating area itself. Based on these results, they proposed a computer model for nucleation site interactions and heat

transfer [82], and recently a more sophisticated simulation model at actual size [87].

2.4.3. Controlled nucleation sites (2): artificial cavities

In recent years, the micro-machining technique has developed expeditiously, and it is now possible to move boiling research through its biggest dilemma concerning the elusive surface roughness and nucleation sites. By applying such advanced surface manufacturing techniques, together with modern measurement techniques, some researchers have made significant attempts to elucidate boiling features, including bubble behaviors and temperature fluctuations, for a single as well as multiple nucleation sites [88–91]. Qiu et al. [92] studied the growth and detachment mechanisms of a single bubble on a 1.0 mm thick silicon surface with an artificial cavity 10 μm in diameter and 100 μm in thickness. The bubble growth time, bubble size and shape from nucleation to departure were measured under subcooled and saturated conditions. It was found that the effect of wall superheating and liquid subcooling on the bubble departure diameter is small, whereas the growth periods are very sensitive to liquid subcooling at a given wall superheat. On the other hand, the heat transfer characteristics of the artificial surfaces with different cavity patterns have been studied by some researchers: Kubo et al. [93] studied experimentally the boiling heat transfer of FC-72 from newly enveloped treated surfaces with a thickness of 0.5 mm and micro-reentrant cavities. Four kinds of treated surfaces with the combinations of two cavity mouth diameters (about 1.6 and 3.1 μm) and two number densities of micro-reentrant cavities (81 and 96×10^3 $1/\text{cm}^2$) were tested along with a smooth surface for the liquid subcoolings of 3 and 25 K with the degassed and gas-dissolved FC-72. It was found that the heat transfer performance of treated surfaces is considerably higher than that of the smooth surfaces. The highest performance was obtained with the treated surface with the larger cavity mouth diameter and the larger cavity number density. Honda et al. [94] conducted experiments to study the effects of the dimensions of fins on the pool boiling of FC-72 on silicon chips with rectangular micro-pin fins. The pin-finned surfaces showed a steep increase in the heat flux with increasing wall superheat. The wall temperature at the critical heat-flux point was always less than 85 $^\circ\text{C}$. The maximum value of the critical heat flux was 3.5 times larger than the heat flux at a wall temperature of 85 $^\circ\text{C}$ obtained using a smooth chip. Baldwin et al. [76] and Bhavnani et al. [77–79], as mentioned in the previous section, studied the heat transfer performance of different cavity sizes and spacings.

All of the above research has been performed in the older research context, without considering the interaction with the heated wall, by assuming a uniform wall temperature. In contrast, Shoji and his coworkers

[41–45,57] performed serial experiments of boiling to investigate the various problems of nucleation sites and interactions, with a special interest into their nonlinear interactive features and mechanisms. They employed a 15 mm×15 mm and 200 μm thick copper or silicon disk with regulated single or twin artificial cavities as the test surface. The vicinity of the manufactured cavities was heated by Nd:YAG laser irradiation from the backside of the test disk. The temperature fluctuations just under the artificial cavity were recorded using a radiation thermometer with sufficient resolutions of space, temperature and time. The corresponding bubbling status was recorded by high-speed video camera. The heat input to the disk surface was controlled using the power of a laser. The working fluid was distilled water at atmospheric pressure under the saturated as well as the subcooled pool boiling condition. The achievements of these experimental studies have been reported elsewhere [41–45], and the main results in the present context are described briefly below.

2.4.3.1. Cavity shape effects. In the first test of serial experiments, Shoji and Takagi [41] investigated the effect of cavity shape on the bubble behaviors and the surface temperature fluctuations on an artificial copper plate with a 0.1 mm thick 10 mm diameter. To reveal the bubbling characteristics depending on the geometry of the cavities, three types of artificial cavities were manufactured on the center of copper disk surfaces, and, in total, five different cavities with two different sizes of 50 and 100 μm and two different depths of 30 and 50 μm were arranged. It was found from this experiment that the conical cavity shows intermittent bubbling with large temperature fluctuations and requires a rather high degree of wall superheat to maintain the bubbling, while the cylindrical as well as the reentrant cavity show continuous and stable bubbling from rather low superheating. From nonlinear analysis for the wall temperature time series, it was found that rather low correlation dimensions and positive maximum Lyapunov exponents are obtained, suggesting the existence of low-dimensional chaos in the systems. The return maps constructed from the bubble departure intervals for cylindrical and reentrant cavities reveal chaotic structures of bubbling in the boiling phenomena.

2.4.3.2. Cavity size effects. Based on the first test results of Shoji and Takagi, Yasui and Shoji [57] employed a silicon test surface with a single cylindrical cavity to investigate the cavity size effects on nucleation and bubbling features. The cavity was produced using DRIE (deep reactive ion etching), and the diameter of the cavity varied from 5 to 100 μm and depths from 20 to 80 μm . They found that the cavity depth has strong effects on bubble generation and the bubbling features, while the diameter has little influence. The results of nonlinear

analysis showed that the wall temperature fluctuation underneath the cavity becomes complex, and that the complexity (correlation dimension) increases with decreasing cavity depth.

2.4.3.3. Complexity around the nucleation site. In the experiment of Yasui and Shoji [57], the spatial and temporal variations of wall temperature were measured along the line crossing the center of the cavity. From the results of the nonlinear analysis for the data set, it was found that the complexity is relatively low at the bubble base but increases with distance from the cavity center in regions larger than the bubble base periphery. This increment in complexity may be due to liquid motion and heat transfer at the meniscus formed at the periphery of the bubble base, and may also be due to the micro-convection induced around the bubble by the bubble growth and departure.

2.4.3.4. Cavity spacing effects: nucleation site interaction. To investigate the nucleation site interactions, a silicon plate with twin cylindrical cavities was employed as the test surface. Based on the results of the second experiment of Yasui and Shoji [57], Zhang and Shoji [44,45], selected a cavity of 10 μm in diameter and 80 μm in depth and kept the size of the cavity constant throughout their experiment but the spacing between the two cavities was changed from 1.0 to 8.0 mm. As the size of the departing bubble without interference was approximately 2.4 mm in diameter, the spacing employed corresponds to a change in the spacing-to-bubble diameter ratio, S/D , from 0.3 to 3.3. It was found that as the spacing decreases, the bubble size becomes small, the bubble departure frequency increases, and that the heat transfer is enhanced with decreasing cavity spacing. According to detailed analysis of the data and photographic observations, it was made clear that the heat transfer enhancement could be attributed to the agitated micro-convection due to strong interference of the neighboring two bubbles. In general, as shown in Fig. 2, nucleation interactions may involve three significant factors: (*H*) the hydrodynamic interaction between bubbles, (*T*) the thermal interaction between nucleation sites, and (*C*) vertical, horizontal and declining bubble coalescences. Based on the comprehensive observations and analysis, the intensity, competition and dominance relationships among these three factors determines the four different interaction regions. This classification of nucleation site interaction is essential and universal, with the validity confirmed by changing the wall material and thickness [45,46]. These results do not conflict with the available classification given by Judd and his coworkers [4–10] for actual boiling surfaces with various roughnesses. Thus, the present study reveals some essential aspects of nucleation site interactions in pool boiling,

significantly improving our understanding of boiling mechanisms.

3. Nonlinear theoretical studies and models

The sophisticated nonlinear modeling of boiling has been attempted in recent years spanning a wide variety of topics. A few of these studies have been purely theoretical and qualitative, focusing on the essential problem of boiling, such as the structure of the boiling system, boiling modes and their transitions. Some of the research has been devoted to finding the sources or elements leading to the chaotic features of boiling using simplified dynamical models. Other studies have proposed sophisticated models and simulations, for the purpose of being able to understand the actual underlying processes and interactions. In addition to such studies, some articles discuss the principles and philosophy when formulating their model of boiling. As seen in the previous section, the hydrodynamic behaviors of bubbles have important roles in boiling so that the nonlinear modeling of bubble dynamics has been attempted in both isothermal as well as in simple boiling systems. In what follows, such currently available studies will be overviewed in a categorized order.

3.1. Theoretical models of boiling

In general, grasping the fundamental nature of the phenomena using simplified minimal models is one strategy for improving our understanding of the complex system. It is believed that the qualitative nature of the phenomena is generally universal, independent of the minute structure of the dynamical processes. Boiling may be a typical example of such a category. From this point of view, Yanagita [95,96] proposed a highly idealized theoretical model of boiling. He employed the CML (coupled map lattice) method to simulate boiling. The CML method is a tool for grasping the qualitative nature of complex spatio-temporal systems and is well suited to many physical systems (e.g., Kaneko [97,98], Yanagita and Kaneko [99]). CML associates a dynamical system with continuous field variables but with discreet space and time, in which the local dynamics are advanced in time by mapping and propagation in space in the form of “diffusion” or “flow”. The CPU time required for the calculations is much shorter than those of other simulation methods. The CML method can be applied to nonlinear analysis as well as to statistical treatments. Details of the CML method are available elsewhere [97,98]. Using this CML method, Yanagita succeeded in explaining the appearance of boiling modes and transition. He assumed that boiling phenomena are decomposed into the following three dynamical pro-

cesses: (1) thermal convection, (2) bubble generation and floating motion, and (3) phase change. Then, he expressed each dynamical process in a set of parallel mappings and applied these to the lattices of the boiling region. In this model, a periodic boundary condition is assumed in the horizontal direction and both the top and the bottom temperatures in the calculation domain are prescribed. The temperature of the fluid lattices is initialized using a small uniform random number, and a set of mappings was successively carried out for the time advancement. From the results, Yanagita showed the boiling curve, together with snapshots of stationary boiling patterns. He also plotted the maximum Lyapunov exponent for the data on the boiling curve. Thus, he summarized his study by concluding that his model explains well the boiling modes and their transition and that boiling is a dynamical system of spatio-temporal chaos.

Yanagita’s study is surely epochal in boiling research as it was the first trial of boiling simulation covering the full range. However, his model conflicts in some aspects with actual boiling phenomena. Firstly, the model permits liquids to vaporize in bulk. This phenomenon is known as a homogeneous nucleation phenomenon, not boiling in its usual sense. Secondly, boiling takes place when the temperature of a heated surface is less than the phase change temperature of the liquid. Thirdly, film boiling was not strictly realized. This is the most serious drawback of Yanagita’s model. To solve such problems, Shoji and Tajima [100] revised Yanagita’s model by adding two additional dynamical processes related to nucleation on a heated surface, and Taylor instability at high wall superheat. With this revision, all of the boiling modes are well realized and the influence of parameters such as subcooling and surface roughness are at least qualitatively well explained [101].

3.2. Idealized models of nucleation site interactions

In nucleate boiling, the wall superheat is not uniform. Nucleation sites can interact as a result of a variety of processes that change the local wall superheat and other parameters that determine the stability of nuclei. The interaction affects the number of active sites and causes intermittence in their production of bubbles. Thus, chaotic features can typically be observed in wall temperature fluctuations during boiling. These ideas were made clear experimentally by Kenning and his co-workers, as was seen in the previous section on “non-linear experiments”. They provided instantaneous spatial information on wall temperatures. However, the amount of information available in experimental data is so large that comparisons between the model output and the experiment observations were a very difficult process. So they addressed a much smaller scale problem by considering only the temperature time series at a small

group of nucleation sites and the insights that they might provide into the physical processes associated with site interactions. Then, they applied some of the general tools available for the analysis of nonlinear systems, first to the model and next to the real experimental data from pool boiling on a copper plate. They suggested that this method of analysis would be very valuable for temperature signal analysis at near-critical heat fluxes, such that the direct observation of bubble behavior close to the wall is impractical. The application of the results to experimental data has already been described in the previous section. The highly idealized model is explained below.

3.2.1. *Ellepola and Kenning model*

Ellepola and Kenning [32] constructed a model for considering the interaction of just two nucleation sites on a thin uniformly heated strip. The strip was represented by three lumped thermal capacities, one around each site and one for the intervening wall which was cooled by convective heat transfer. Heat is exchanged between the three capacities by conduction. A site was assumed to become active at a high activation superheating and then remain active, producing bubbles in a near-continuous stream, removing heat until the site's superheat fell to a lower deactivation value; then the superheat had to rise back to the activation value before bubble production was resumed. The fluctuations in the heat removal due to individual bubbles were not modeled, nor were the details of the temperature distribution around a site. For this reason, the site superheating was not necessarily the value right at the site itself. This model may be appropriate for high flux boiling.

The model equations were solved numerically for conditions of zero initial wall superheat and for a uniform electrical heating supply. The parameter values were chosen to approximate water boiling on a thin and thick copper plate, and asymmetry was introduced by specifying different deactivation and activation wall superheats at the two sites. The switching behavior of the site is strongly nonlinear, as indicated in the article of Moon [14], and an additional weak nonlinearity was introduced in the convective cooling. The boiling cooling rates were chosen to satisfy the inequality that causes the sites to cycle on and off. The model system had three independent variables. Ellepola and Kenning [32] demonstrated the application of several tools that have been developed for detecting the chaotic behavior of nonlinear, multiple-degree-of-freedom systems by examining just one variable at a time. The attractor was reconstructed using the method of delays and embedding [102], indicating that the increase in heat flux causes the system to become chaotic. The attractor trajectories at the low heat flux quickly converge from the starting

point onto a limited cycle, repeating for hundreds of cycles. It was found that the combined effects of the two sites cause a stronger chaotic behavior. This qualitative interpretation can be quantified by correlation dimensions, derived by applying the Grassberger and Procaccia algorithm [26–28]. It was also indicated that another route to chaotic behavior appears when the plate thickness is reduced.

These examples using simulated, noise-free data have illustrated ways by which a boiling system may develop chaotic behavior, which can be diagnosed by examining temperature time data from a single point. Ellepola and Kenning [32] did not consider combined spatio-temporal data, nor whether the chaotic behavior is of practical significance. In the example, however, the superheating of the central capacity is the most chaotic but the range of variation is small. Such behavior might explain the experimental observations of very occasional bubble nucleation at some sites between the more active sites shown by Kenning and Yan [29]. These results were well summarized by Ellepola [33].

3.2.2. *Mosdorf model*

Simplified models similar to that of Ellepola and Kenning [32] have been extensively proposed by Mosdorf [103–108]. He proposed the simplest one-dimensional mathematical model by considering the slow and fast processes of heat transfer in the heating surface [103]. He extended his model to two and three dimensions by considering further single and multiple nucleation sites, active and nonactive nucleation depending on the wall temperature, and also the two types of heat transfer depending on the wall temperature [106]. Recently, to investigate how the change in thermal boundary condition influences the change in wall temperature, he proposed a simple but more sophisticated two-dimensional model, in which he involved a single nucleation site, activation and deactivation of the nucleation site, bubble waiting and growing times, and two separated regions with different surface heat transfers. With this model, he concluded that the chaotic change in the heating surface temperature can take place when the heat transfer to the thin layer of the heated surface is less than the heat absorption by the boiling liquid from the heating surface with the vapor bubble.

Mosdorf has thus considered boiling chaos by presenting simple theoretical models to show the chaotic nature of boiling, and also the influence of bubble growth, departure and interactions [109–111] which will be described later. Although some of the assumptions in his model do not agree with actual boiling databases, his models hit the essential nature of boiling phenomena, based on his definite philosophy [112]. This review paper was published independently of the article of Nelson et al. [16] but the opinions expressed coincide surprisingly well.

3.3. Nonlinear simulation models of nucleate boiling

3.3.1. Numerical simulation of an isolated single bubble: older mechanistic modeling

It is really difficult, even at present, to carry out comprehensive simulation work on boiling phenomena, due to the unknown underlying interaction mechanisms of the system. Historically, the modeling of active sites has assumed that the sites are constantly active and arranged in a square geometrical pattern (e.g., Mikic and Rohsenow [113], Lay and Dhir [114]). Numerous models of individual discrete bubbles have been documented over the years. A numerical treatment for single bubble generation and growth was made by Hatamiya et al. [115] and Murata and Hatamiya [116]. Lee and Nydhal [117] found from a numerical solution of the Navier–Stokes and energy equations during bubble growth and departure, that the thermal layer was affected by bubble departure only in the region immediately beneath the bubble, with a maximum effect at the center. This result suggested that the transient heat conduction in the thermal layer could not be integrated over a region larger than the area occupied by the bubble. Mei et al. [118,119] numerically analyzed bubble growth in saturated boiling without taking into account the hydrodynamic effects induced by the growing bubble. They concluded that the thermal influence field depended primarily on four parameters: the Jacob number, the Fourier number, the thermal conductivity, and the thermal diffusivity, with quite complicated relationships among them. Guo and El-genk [120] developed a transient model for studying the evaporation of a liquid micro-layer under a growing vapor bubble during nucleate boiling on the surface of a flat composite wall. It was found that as either the thickness or the thermal conductivity of the heated wall was increased, the evaporation rate increased due to improved lateral heat conduction, approaching that for an isothermal wall. Recently, Fujita and Bai [121] performed a numerical simulation of a single bubble by combining hydrodynamics and heat transfer behavior. It was assumed that the evaporation heat was supplied from the superheated layer around the bubble by convection, and from the meniscus part of the micro-layer. The two-dimensional transient equations of mass, momentum and energy were solved by an arbitrary Lagrangian-Eulerian finite element method. The growth process of an isolated bubble was simulated continuously. Son and Dhir [122] and Dhir [123] also carried out similar work associated with a single bubble by solving the conservation equations of mass, momentum, and energy for the liquid and vapor, phased simultaneously with the continuously evolving interface at and near a heated surface. The simulation results of the bubble behaviors showed good agreement with the experimental data under subcooled as well as saturated condi-

tions, and also under reduced-gravity conditions [92].

These simulations have provided some useful insights into boiling phenomena. However, there is still limited understanding of the mechanisms because of the inevitable simplifications and assumptions made in the simulation process. They attempted to simulate the hydrodynamic interaction and the thermal interaction separately. Namely, they neglected the interactive processes (Fig. 2) by calculating only the interaction of the liquid side with the assumption of an isothermal boiling surface, and only computing the interaction of the heated side with the assumption of bubble behaviors. More significantly, the heated surface temperature was assumed to always be constant. In other words, they have been performed within the framework of the older modeling.

3.3.2. Sophisticated numerical simulation: new mechanistic modeling

In 1990, liquid crystal thermography was applied to the back of a thin heater and combined with high-speed video to study the dynamical behavior of surfaces by Kenning [124]. Kenning and Yan [29] extended the study to reveal an on–off (active and inactive) site behavior and interactions between nucleation sites, as described in the previous section. They made it clear that the site distribution was not uniform. Until recently, the detailed mechanistic modeling of boiling has been severely handicapped by limited computing capabilities. However, the recent works described below were performed in the new framework of Fig. 3, and provide valuable insights into the basic mechanisms that may be involved in nucleate boiling.

3.3.2.1. Discrete bubble region. The work of Pasamehmetoglu and Nelson [125] represented the first effort to model multiple nucleation sites and bubbles for studying the boiling process on a locally instantaneous basis, that is, at spatial and temporal scales of less than the minimum δt_{\min} and δA_{\min} (see Section 1). In this model, the hydrodynamic side problem was dealt with using models associated with single bubbles. So the work applies only to the low-heat-flux isolated-bubble regions. Their model revealed that the primary nonlinearity within the system is associated with the on–off (activation and deactivation) behavior of the sites that is driven by the thermal distribution within the heater. The model also revealed that thermal distributions must occur on the heater surface and that intermittent bubble behavior is possible. See the more recent work of Unal and Pasamehmetoglu [126].

3.3.2.2. Mushroom bubble region. Under a single vapor mass (mushroom bubble) at high-heat-flux nucleate boiling, the problems associated with the modeling of

discrete bubble behavior and possible hydrodynamic interactions could be minimized, and only the consideration of thermal site interactions and macro-layer (liquid rich layer) consumption may be possible. Maruyama et al. [127] and He et al. [128] simulated two-dimensional transient macro-layer consumption and heat transfer for a wide region of nucleate to transition boiling, showing that the simulation results agree well with the available information obtained from past experiments. Both studies assumed a uniform wall temperature (He et al. recently considered thermal diffusion inside the heater yet assumed a uniform surface temperature). However, in the recent study of He et al. [129] and He [130], temporal as well as spatial fluctuations in wall temperatures were considered in the model. Sadasivan et al. [13,131,132] presented a novel model capable of representing hundreds of potential nucleation sites on a heated surface where all of the sites are located under a single mushroom bubble and exposed to the same hydrodynamic boundary conditions. The model allows for the study of the temporal as well as spatial nonlinear effects associated with on-off site behavior, macro-layer thinning and unequal site spacing that occur naturally on real heated surfaces. They employed a configuration where a single mushroom bubble had grown above the surface just before its departure, under which mushroom a small segment of heater is located. The segment was 5 mm on each side (25 mm² in area), and the transient heat conduction equation was solved in both the heated and liquid macro-layer left behind at the birth of the bubble. The effects of macro-layer thinning caused by evaporation from the various interfaces of the macro-layer into the bubble were included. Enhancements to the evaporation process caused by meniscus effects at the base of the vapor stem associated with each active cavity were also included. The heater was 50 μm thick copper. The saturated water at atmospheric pressure was assumed to be the boiling fluid. 180 potential nucleation sites ranging from 0.5 to 5 μm in diameter were located on the heated surface, and the site spatial distribution was assigned randomly in a manner that satisfied a Poisson distribution. All sites were assumed to have geometries conducive to activation such that the activation and deactivation of the sites are controlled solely by the local wall superheat values. The potential wetting effects and cavity shape were hidden within this criterion. In the calculation, based on the heat flux, the bubble's lifetime (hovering period) was determined and the bubble departure was assumed to be periodic. At the end of each bubble's lifetime, the bubble departed, thus returning the liquid macro-layer to its initial state. Sites were allowed to activate any time their cavity temperature exceeded the activation temperature, but were deactivated only from liquid re-supply at the time of bubble departure if their temperature was less than their deactivation temperature. Because the system

was relatively small in size, the locally instantaneous surface temperatures were area-averaged for the analysis. The transient variation of the temperature averaged over the full heated surface exhibited markedly different characteristics depending on the heat flux. The time series showed a periodic behavior at low heat fluxes but showed bifurcation to a period-six at high heat flux.

The details of the site behavior and thermal response are not mentioned here because of limited space, but their results revealed that the active-site density, a characteristic of the heater–fluid interface, is a dynamical quantity and a result of the mechanisms involved, not of a correlated parameter that the older models require. The active cavity sets do not repeat, and thus produce the aperiodic thermal response at high heat fluxes. Some site activities could be characterized as being extremely intermittent in showing “period-doubling” features. Period doubling is a hallmark characteristic of many nonlinear chaotic processes found in nature. The period doubling and subsequent breakdown of periodicity that we see in the current problem would suggest the presence of deterministic chaos in the system. To investigate further the potential chaotic behavior, Sadasivan et al. [132] used the correlation-integral technique to determine the fractal dimension as approximately 4.8 for the time series at 1.04 MW/m². Coupled with period doubling, this suggests that significant nonlinear dynamical behavior is present in the system.

Golobič and his coworkers [81–86] performed an experiment on nucleate site interactions employing the novel laser heating and splitting technique and by controlling spatially the nucleation sites, as mentioned in the section on “nonlinear experiments”. Based on their experimental results, they proposed a computer model for nucleation site interactions [82]. More recently, in collaboration with Kenning and Nelson, they proposed a sophisticated model and simulation method of nucleate boiling [87], similar to that of Sadasivan et al. [13,131, 132], but for a more realistic size, and compared their simulation results with the data on which their model was based.

3.4. First principle in modeling: self-organization and self-similarity

The self-organizing principle within the boiling process has been proposed by Nelson and Bejan [133]. The approach employed is based on the view that a naturally occurring flow (geometric structure) is the end result of the process of internal geometric optimization in constructing an assembly of paths of minimal resistance for the “current” flow through the system. The process is related to the general principle of entropy generation minimization. Nelson and Bejan applied this principle successfully to various thermal and fluid problems,

including boiling. The self-organization for boiling is restricted to the available cavities and their distribution on the heater surface. Thus, the process becomes one of constrained minimization [134–136]. This method of self-organization does not seem to connect directly to nonlinear dynamics, but it provides an analysis method for solving systems where a number of competing mechanisms is involved, and should be one objective of nonlinear dynamics [16]. For this reason, it is described briefly below.

Boiling self-organization performed by Nelson and Bejan [133] begins with the determination of the system's organizational preferences within the mechanisms available to it. For high-heat-flux nucleate boiling (e.g., the boiling of water on copper), the preferences are found to be in the following order: (1) if a cavity of the proper size and shape is available in a locally hot region, it should be activated; (2) if a cavity is not available, time-dependent conduction penetrates the macro-layer; (3) when conduction has penetrated the macro-layer, Marangoni–Benard convection completes the scenario. This third mechanism requires a sufficiently thin macro-layer and a sufficiently long bubble lifetime. The active-site distributions resulting from the self-organization mentioned here vary both in time and in space and are driven by both the thermal distribution within the heater and the ability of the liquid to accept the heat flow via one of the mechanisms just noted. A simple picture of the spatio-temporal behavior in the simulation was provided by showing the distribution of active cavities at the bubble departure, from which spatial information might be compared to that obtained experimentally by Wang and Dhir [70]. Temporal information associated with the site activation is not available, so the figure represents a time-lapsed picture of the process and should be compared to the end-of-bubble-life state in the simulation. Regarding the results, Nelson and Bejan mentioned that the visual similarity between the estimated and the experimental results may be fortuitous, but it is also expected.

Chai, together with his collaborators, has also discussed extensively the complexity, nonequilibrium and random nature of boiling by analyzing the nonlinear processes of boiling. After discussing the dissipative structure of boiling systems from a thermodynamical point of view, they explained the critical as well as the minimum heat flux by considering nucleate site interactions [137,138]. They also showed that the randomness of boiling comes not only from the nonlinear interaction between bubbles [139] but also the complicated nucleation site distribution [140]. They discussed the self-organization and self-similarity of the boiling systems [141], made clear the bifurcation and catastrophe of boiling curves [142,143], and revealed a new possible understanding of transition boiling [144] and boiling mode transitions [145].

3.5. Modeling of nonlinear chaotic bubbling in isothermal systems

As mentioned in the previous section on “nonlinear experiments”, even in a simple system where a chain of bubbles is generated from a submerged orifice and a nozzle, bubble behavior is rather complex, showing chaotic features under certain conditions of gas flow rate [48–50,146]. Several models of bubble formation have been presented in the past but most of them have dealt only with isolated single bubbles, and include no interactions between preceding or adjacent bubbles. In other words, all of the past models are “linear models” and it is impossible to explain using these models the nonlinear features of bubbling that are actually observed.

Mosdorf [147] presented a very simple mass-spring model for a single bubble-formation system to explain the nonlinear interaction between a single bubble and the surrounding liquid, and recently extended his model to include the interaction between two neighboring bubbles [148]. In the model, he showed that the chaotic movement of bubbles (mass) may appear when the spring force (distance between bubbles decreases) increases, the dumping force decreases, and the amplitude of the external force (the diameter of the bubbles) increases. His model is quite unique in considering the bubble–bubble as well as bubble–liquid interactions but involves some problems in terms of how we determine the values of mass, spring and dumping to fit the actual systems. On the other hand, Zhang and Shoji [50] proposed a nonlinear dynamical model and succeeded in explaining the nonlinear bubbling features. They concluded that the nonlinear feature comes mainly from the vertical interaction between the bubble and the preceding bubble.

Bubbling features becomes more complex in multiple-bubble-generating systems. In a couple-of-bubbles-generating system, Shoji and his coworkers [149,150] found curious phenomena such that regular periodic and irregular chaotic bubble generation occurred repeatedly when the gas flow rate was decreased in a monotonous manner, which was later theoretically explained by Tange and Shoji [151].

Thus the bubble dynamics are rather complex, even in isothermal systems isolated from phase changes, so it may be said that the situation becomes more complex in boiling systems.

4. Discussions

4.1. Research achievements and remaining problems

As described in the previous sections, much research has been carried out in recent years into nonlinear boiling, and, as a result, considerable advances have

been made. However, in spite of this, the physical mechanisms of boiling phenomena are still not yet fully understood, and continuous efforts in research are still required. In terms of the nonlinear features of boiling, it needs to be emphasized that the available theoretical models reveal the existence of low-dimensional dynamics in boiling systems. However, there is little clear experimental evidence to show that this is the case. This means that actual boiling phenomena, even in simple systems, are complex, far beyond the capabilities of our detection techniques and analysis tools.

Boiling is a conjugate phenomenon, the subprocesses of which interact with each other (Figs. 1 and 2). It is generally very hard to investigate the underlying mechanism of each process separately in isolated conditions, even when we separately change the effect factor of the boiling, such as one of the properties of the liquid, such as the material or subcooling, of a wall property such as the material, thickness, or thermal capacity, or the surface roughness or system pressure. For example, if we change the test liquid, the wettability of the liquid to the heated surface also changes, which accordingly changes the nucleation site and bubble generation. So we have to unravel the processes from the integrated effects of all of the factors. However, such parametric experiments have yet to be satisfactorily performed.

As suggested by Kenning [11] and Dhir [12], the most important issue in boiling processes is the nucleation site and interactions, and their dependence on the nonuniform wall temperature. This issue relates to the properties of the heated surface. How to treat the surface is one of the most difficult and elusive problems of boiling. This problem includes various problems such as surface roughness, nucleation site distribution, effects of aging and oxidation, and wettability, all of which connect directly to the activation and deactivation of the nucleation site. In most of the research that has been carried out, the surface roughness and/or contact angle have been used to describe the surface property. However, it is almost impossible to define the complicated solid surface using only such limited quantities. In other words, the actual solid surface has numerous cavities or stretches with various kinds of shapes and sizes. It is obvious that the height of the roughness does not represent the geometrical properties of the actual surface. A better and more plausible measure would be the “fractal” roughness, as suggested by Fong et al. [71,72], but the usefulness and generality of it has yet to be validated. When the surface is heated and subjected to a liquid, the situation becomes more complicated. The wettability of the wall surface to the liquid is an important effect in nucleation from the site. The wettability is usually represented by the contact angle, but we need to differentiate between “receding” and “preceeding” contact angles, depending on the movement of the liquid–solid–vapor triple interface. Moreover, the contact angle itself

is a “vague” physical quantity [152,153] in that it is not clear whether such a property exists on a such high temperature solid surface as that heated above the saturation temperature of the liquid. In other words, we have no physical measure at present to rigorously capture this property of the actual heated surface subject to boiling. This is undoubtedly one of the reasons why we have had limited success in the mechanistic modeling of boiling. A possible method for solving this dilemma of the heated surface may be to simplify the surface as far as possible. In actual fact, in recent years, quite a few experiments have been carried out on such “artificial” surfaces using modern micro-machining techniques, and some valuable information has been obtained as a result. However, their mechanistic achievements are limited to small scales, and the problems of scaling up to real sizes remains unsolved.

In most of the currently available models dealing with nucleation site interactions, the effects of the liquid-side interactions are excluded from rigorous treatment by assuming convective heat transfer, a micro- or macro-layer thickness, a bubble departure frequency, and the like. This may indicate a lack of mechanistic studies on bubble-related phenomena. Exceptions to this are the studies of Shoji and his coworkers described in the previous section on “nonlinear experiments”, where bubbling features were shown to be complex and strongly nonlinear in nature. It is particularly noticeable that even bubble generation is periodic, the micro-convection induced by generating bubbles is not periodic because of the nonlinearity involved in liquid motion.

4.2. Measurement methods and tools

At the present time we are fortunate in that we have various modern tools available for analysis and measurement. Nonlinear chaos dynamics is one of them. The others are the recently advanced micro-machining technique for surface treatments, computers, and various kinds of measuring equipment. However, each has limitations and problems in their application to boiling research. In terms of nonlinear chaos dynamics, we have many physical measures such as strange attractors, fractal dimensions and Lyapunov exponents, but it is rather hard to evaluate the value of such measures from the experimental “noisy” data. In addition, the current methods of evaluation can only be applied well to temporal series. Boiling is a more complex spatio-temporal multi-chaos system, where the complexity changes from position to position and a huge amount of data are needed for the analysis. At present, unfortunately, we are limited in the methods of analysis available and the equipment capacity of the data acquisition systems to solve these problems.

In terms of measurement tools, the thermocouple has been used to measure temperature as well as heat flux. Actually, in some nonlinear experiments [34,36], the wall temperature fluctuations were measured by embedding the thermocouple inside the heated wall. However, it is actually impossible to obtain the special information that is indispensable to investigating the essential processes of boiling, the nucleation and site interactions. So, in some recent experiments, liquid crystal thermography or a radiation thermometer has been employed to measure the local as well as the spatial information of wall temperature. However, both have some disadvantages in terms of time and temperature resolution. We need some technique for filtering, just as was done by Ellepola [33], and Kenning and his coworkers [38–40]. More significantly, both can be applied only from the backside of the heated wall and so the thickness of the heated wall is limited to thin walls. To solve these problems, new sensing devices such as MEMS (method of electrical and mechanical system) or μ -TAS (micro-totally analytical system) could be applied, but no attempts have yet been made.

In terms of computing tools, at present, we can use a high-quality and huge-capacity computer. For dispersed gas–liquid systems, Ohashi [154], and Hashimoto and Ohashi [155] have attempted to perform large-scale simulations using the method of lattice-gas cellular automata, and suggested a new direction for thermo-hydrodynamical numerical analysis. In order to rigorously solve the governing system of equations of boiling [156], however, we need to prescribe the hydrodynamic as well as the thermal boundary conditions of the solid–liquid interface and the liquid–vapor interface, all of which remain elusive. In other words, unless the physical mechanisms of the boiling process are made clear, it is almost impossible to carry out rigorous numerical simulation that is fully deserving of the name.

4.3. Modeling efforts: philosophy and perspective of boiling research

In general, concerning nonlinear modeling, we should differentiate between studies aimed at achieving a better understanding and those aimed at better predictions, as emphasized by Sadasivan et al. [13]. We know that in some complicated systems, we can reach a better understanding much more easily by grasping the fundamental qualitative nature rather than by providing too much quantitative information. Thus, some of the current theoretical nonlinear models, especially the pure model of Yanagita [95,96], are not aimed at quantitatively predicting the behavior of the phenomena. They are aimed at achieving a better understanding, with the hope that they will eventually lead to better predictions. What is attempted in these models is to mimic the behavior of real phenomena. Mimicking the behavior

merely presents us with a means of exploring the possible characteristics of the system to better understand the effects of particular processes and their potential interactions. Sadasivan et al. [13] called this theoretical approach “numerical experimentation”. Future efforts in such modeling (or numerical experimentation) are necessary and it is hoped that the results of such studies might either identify future experiments to clarify dominant phenomena or focus future mechanistic modeling efforts.

As described in Section 1, the method of temporal and spatial averaging has previously been used to develop our current correlation-based boiling technology. This technology has provided the means with which to design and analyze, but has required either experimental re-testing or the inclusion of safety factors for obtaining believable results. In the historical experience, the older mechanistic models have proven to be of little value because of their inability to predict a priori the results we desire since they are not based on the underlying physics.

Studies associated with the possible nonlinear behavior of boiling systems have recently emerged. The experiments carried out in Tokyo, Oxford, and Slovenia reveal, as presented in the previous section, that significant nonlinear chaotic effects are possible in boiling systems. These studies suggest some fundamental reasons for the difficulties experienced by previous researchers studying boiling and two-phase phenomena in providing mechanistic models. Over the last couple of decades, as work with nonlinear systems has progressed, it has become clear that linear approximations to nonlinear systems are of limited use. In regard to nucleate boiling heat transfer, the previous paradigms have been built around average nucleation site behavior. That is, single nucleation site behavior was investigated, the number of active nucleation sites was determined, and the overall average heat transfer was estimated using a linear extrapolation of heat transfer from a single average site assuming uniformly spaced constantly active sites. If any of the nonlinear effects noted above do exhibit significant influence over the behavior of the boiling system, such as chaotic behavior, these previous paradigms must be downgraded. As already suggested in this article, a possible way of solving the dilemma regarding surface roughness is to simplify the surface by employing a smooth surface with only fabricated cavities. In such a system, it is much easier to investigate the underlying processes and mechanisms. However, the surfaces used in industry are not like these, so research into actual surfaces and simplified surfaces need to be performed simultaneously and the results should be compared with each other.

Computer simulations are strongly recommended for future studies into boiling. They may yield information that would otherwise not be found using measurement

tools alone. In order to perform rigorous sophisticated simulations of boiling, we need to make clear beforehand the underlying physical mechanisms of the boiling process. When we succeed in achieving such simulations, the averaging technique will then become a more suitable tool for deriving useful correlations for applications. Judging from the present situation of boiling research, we will need some time to reach the stage where we can achieve satisfactory simulations of boiling. In the meantime, computer simulations of simplified surface systems are recommended, just as has been done by Dhir and his coworkers [92,122] who showed that it is possible to validate the calculation algorithm and the assumptions for the dynamics of boiling, hopefully leading to final success. Of course, at the same time, attempts to simulate the actual system in a large scale, such as the efforts of Sadavisan et al. [13,131,132] and Golobič et al. [87] should also be continued.

This article focuses on the research and achievements based on nonlinear dynamics. It is mentioned here that nonlinear dynamics is no more than an analysis tool. The most important aspect of boiling research is not the tool employed but the definite perspective and philosophy of the individual researcher. In this article, the terminology “older” and “classical” are used in terms of the research framework. They are not used to mean “invaluable”, “useless”, or “inferior”. They are used only to classify or categorize the approach methods. It should be noted that some studies belonging to the “older” mechanistic framework are more suggestive and informative than the “new” ones. The articles and the papers of Dhir [12,122,123] are some such typical examples.

5. Concluding remarks

In the present article, the available nonlinear research into boiling chaos has been critically reviewed, focusing on the interactive and conjugated processes of boiling. The results of these studies surely improve our understanding of the dynamical mechanisms of boiling. However, many problems remain unsolved. In terms of experiments, we have yet to discover how the local heat transfer from the boiling surface is determined in space and time and how it relates to the wall temperature, bubble growth and departure, and liquid micro-convection. In terms of theoretical studies, there remains a need to formulate a sophisticated model that includes both the subprocesses of the liquid side and the wall side.

Although the present article also suggests a new avenue for boiling research, the complexity of the problem is tremendous, and our current correlation-based technology for design and analysis will continue for years to come.

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