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Thermodynamics bifurcations of boiling structure

L.H. Chai¹, M. Shoji^{*}

Department of Mechanical Engineering, The University of Tokyo, Hongo 7-3-1, Bunkyo-ku, Tokyo 113-8656, Japan Received 14 March 2001; received in revised form 6 March 2002

Abstract

Boiling system is a typical open dissipative structure. The criterion for the transition of boiling modes is derived based on non-equilibrium thermodynamics. It is found that the transitions between different boiling regimes correspond the first thermodynamics bifurcation and second thermodynamics bifurcation: system develops tending to minimize the entropy generation. The self-organized transition processes among different boiling modes caused by the interactions among active sites or dry patches that result in the formation of boiling dissipative structure are then revealed from dynamic perspective. The present studies not only theoretically explain the experimental observations from other researchers, but also give deeper insight on essences of the transitions between different boiling modes. © 2002 Elsevier Science Ltd. All rights reserved.

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1. Introduction

Boiling process is encountered in a number of applications in traditional industry such as metallurgical quenching processes, flooded tube and shell evaporator, immersion cooling of industrial components, to name only a few. A substantial number of such efforts have been devoted to understanding and modeling of the boiling heat transfer since the beginning of last century. The importance of boiling in a wide variety of applications in modern heat transfer technologies related to space, electronics component and nuclear reactors, etc., has provided the even increasing incentive for numerous investigations of its basic mechanisms over the past several decades.

Among all industrial applications related to boiling heat transfer, the understanding of the different boiling regimes is one of the most important topics. Nukiyama [1] first experimentally gave the consideration of the heat flux versus wall superheat boiling curve and the boiling

¹ Tel.: +81-3-5841-6408; fax: +81-3-5841-6408.

transition. Over the past decades, many investigations have been conducted to examine the heat flux versus wall superheat boiling curve. However, because of the complexities of boiling phenomena, the essence of the boiling curve is not yet well understood. There is as yet no consensus as to catastrophe on boiling curves, especially in transition regimes from natural convection to nucleate boiling or from nucleate boiling to film boiling.

It is necessary to look at the whole picture of boiling process from new perspectives [2,3]. Considering that the boiling system is characterized by non-linearity, nonequilibrium, complexity and stochastic performance, it is necessary to deal with potential non-linear effects in boiling systems. Non-equilibrium thermodynamics, as a universal science, may be introduced to resolve complex boiling problems to some degrees. The present paper first introduces dissipative structure concept of boiling systems. The criterion for the transition of boiling modes is then derived based on non-equilibrium thermodynamics. It is found that the transitions between different boiling regimes virtually correspond the first thermodynamics bifurcation and second thermodynamics bifurcation: the boiling system develops tending to minimize the entropy generation. The interactions between active sites, bubbles or dry patches are investigated by use of basis viewpoints in dynamics. Self-organized bifurcation transition processes among different boiling modes are revealed. The

^{*}Corresponding author. Tel.: +81-3-5841-6406; fax: +81-3-5800-6987.

E-mail addresses: lhchai@photon.t.u-tokyo.ac.jp (L.H. Chai), shoji@photon.t.u-tokyo.ac.jp (M. Shoji).

| Nomenciature | | | |
|---|--|--|--|
| с | damping coefficient in Eq. (26) | Greek | symbols |
| G | Gibbs function | ξ | vector defined in Eq. (37) |
| $h_{ m fg}$ | latent heat | ρ | density |
| J^{-} | flux | β | variable contained in matrix L |
| L | matrix defined in Eq. (33) | σ | surface tension |
| q | amount of heat transfer | λ | variable defined in Eq. (35) |
| р | pressure | δ | unit matrix |
| Р | entropy generation | μ, μ' | integer $1, 2, \ldots, n$ |
| r S t T u, v V x X | bubble radius entropy time temperature vectors defined in Eqs. (31) and (38), re- spectively volume element coordinate driving force | Subscr c e fg i l s v | <i>ipts</i> critical, conduction equilibrium, energy, evaporation liquid vapor phase change integer 1, 2,, <i>n</i> , irreversible liquid saturated state vapor |

present studies not only theoretically explain the experimental observations from other researchers, but also give deeper insight on essences of transitions between different boiling modes.

2. Thermodynamics bifurcations—transition of boiling structures

Prigogine [4,5] and Haken [6,7] established dissipative structure theory and synergetic theory, respectively, which have both been appreciated in international academy fields. Starting from mechanistic description of chemical interactions, the Brussels School led by Prigogine investigated the formation of the ordered structure in system far from equilibrium state under the action of non-linear interactions. These kinds of structures are named by dissipative structure for their maintenance by the exchange of mass and energy with the environment. In a similar way, Haken summarized the general characteristics for the formation of various ordered structure, that is, by the non-linear interactions or cooperation under a certain condition, the system composed by lots of subsystems will macroscopically establish temporal ordered, spatial ordered or temporalspatial ordered structures with a certain function. Synergetic theory proposed by Haken can provide a vivid picture for the formation of the dissipative structure in system far from equilibrium state.

For open boiling system, as shown in Fig. 1, dS is total change of entropy, d_iS is entropy generation and d_eS is entropy exchange with environment. The total change of entropy can be written as



Fig. 1. Open boiling system.

$$dS = d_i S + d_e S \tag{1}$$

The total change of entropy is the sum of entropy generation and entropy exchange with environment. According to thermodynamic second law, since system entropy always increases, closed systems always tend to be uniform and disorder. While dissipative structure theory proposed by Prigogine [4,5] states that entropy increase and entropy reduction are both possible for open systems. When $|d_e S| > |d_i S|$, which means the entropy will reduce, the open system will tend to be more order. Boiling system is a typical open system. On one hand, heat and mass transport is the basic process in boiling system. Apparent, this kind of irreversible process will result in entropy increase. On the other hand, boiling system constantly exchanges energy with the environment, which will result in entropy reduction. When the condition that $|d_e S| > |d_i S|$ is satisfied, more ordered boiling system structure would be possibly established.

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Boiling system is a complicated dissipative structure and every part of comprising it interacts each other, not to be independent. It is the interactions and cooperative effect that produce this kind of dissipative structure in boiling system. The detailed analysis is as followed.

Assuming the entropy and entropy generation differences between any states and stationary state are ΔS and ΔP , respectively. ΔS and ΔP can be extended as

$$\Delta S = \delta S + \frac{1}{2} \delta^2 S + \cdots$$
 (2)

$$\Delta P = \delta P + \frac{1}{2}\delta^2 P + \cdots \tag{3}$$

 $\delta^2 S$ in Eq. (2) is called two-order excess entropy.

Providing that the detailed equilibrium is satisfied, the excess entropy $\delta^2 S$ is always a minus, i.e.,

$$\delta^2 S < 0 \tag{4}$$

 $\delta^2 S$ can be considered as a Lyapunov function.

It is known that the derivation of $\frac{1}{2}(\delta^2 S)$ to time equals to one-order excess entropy generation δP [4], i.e.,

$$\frac{\mathrm{d}}{\mathrm{d}t}\left(\frac{1}{2}\delta^2 S\right) = \delta P \tag{5}$$

For the state that is far from the equilibrium-state, depending on the specific conditions, the sign of δP can be positive, minus or zero. Therefore, the sign of $(d/dt)(\frac{1}{2}\delta^2 S)$ can also be positive, minus or zero.

If for any states, the following equation is satisfied,

$$\delta P = \frac{\mathrm{d}}{\mathrm{d}t} \left(\frac{1}{2} \delta^2 S \right) > 0 \tag{6}$$

Then this state is asymptotically stable.

If for any states

$$\delta P = \frac{\mathrm{d}}{\mathrm{d}t} \left(\frac{1}{2} \delta^2 S \right) < 0 \tag{7}$$

The state will be unstable, which physically means that the system will develop tending to minimize the entropy generation.

And if

$$\delta P = \frac{\mathrm{d}}{\mathrm{d}t} \left(\frac{1}{2}\delta^2 S\right) = 0 \tag{8}$$

The state will be critically stable. Three cases are shown in Fig. 2.

For boiling process, the excess entropy generation is

$$\frac{\mathrm{d}}{\mathrm{d}t}\left(\frac{1}{2}\delta^2 S\right) = \delta P = \int \mathrm{d}V \sum_k \delta X_k \,\delta J_k \tag{9}$$

Heat transfer in the zone adjacent to wall is the main transportation mechanism for boiling process, consequently



Fig. 2. The variation of excess entropy with time.

$$\delta P = \int \delta V \delta q \cdot \delta \left[\nabla \left(\frac{1}{T} \right) \right] \tag{10}$$

Equation can be changed to

$$\delta P = \int \frac{\delta q}{\delta T} \frac{\delta T}{\delta x} \left(\frac{\delta x}{T}\right)^2 \left[\frac{2}{T} \left(\frac{\delta T}{\delta x}\right)^2 - \frac{\delta^2 T}{\delta x^2}\right] \delta V \tag{11}$$

Considering the temperature distribution in thermal boundary layer, we have

$$\frac{\delta T}{\delta x} \left(\frac{\delta x}{T}\right)^2 \left[\frac{2}{T} \left(\frac{\delta T}{\delta x}\right)^2 - \frac{\delta^2 T}{\delta x^2}\right] > 0 \tag{12}$$

So the sign of δP depends on $\delta q/\delta T$. If $\delta q/\delta T < 0$, δP will be less than zero and bifurcation will occur. In other word, at case of boiling heat transfer, the well-known Lyapunov criterion for destabilization (bifurcation), i.e., Eq. (7) can be alternately written as

$$\delta q / \delta T < 0 \tag{13}$$

2.1. First thermodynamics bifurcation of boiling structure—transition from natural convection to nucleate boiling

At relatively low superheat, boiling system is in the natural convection states. The system will prevail over stochastic fluctuation resulted from spatial-temporal non-uniform wall temperature and keep in steady state. However, when superheat reaches a certain value, a stable bubble forms on heating wall, as shown in Fig. 3. The condition (13) will be then satisfied for strong evaporation latent heat transfer. Therefore, the formation of stable bubble marks the incipience of nucleate boiling—a new boiling dissipative structure, which is the first thermodynamics bifurcation on boiling curves, as shown in Fig. 4. More mechanistically, when superheat



Fig. 3. Latent heat transfer by micro-layer evaporation.



Fig. 4. Bifurcations of boiling modes.

reaches a relatively high value, boiling system will be in the non-linearity and non-equilibrium states. Non-linear phenomenological relationship exists between the thermodynamic force (temperature gradient in boundary layer) and thermodynamic flux (heat flux) [8]. By nonlinear interactions in systems, the system will develop tending to minimize the entropy generation. This will prompt the formation of a new dissipative structure. In the process of non-equilibrium boiling, outer energy must be constantly provided to maintain the production and growth of bubbles, simultaneously the irreversible process will dissipate fractions of energy. The structure is a kind of dissipative structure for its maintenance by the exchange of mass and energy with the environment.

2.2. Second thermodynamics bifurcation of boiling structure—transition from nucleate boiling to film boiling

When superheat reaches a higher value, a stable vapor film forms on heating wall, as shown in Fig. 5. The condition (13) will be then again satisfied for poor conductive heat transfer by vapor film. Therefore, the formation of stable vapor film marks the incipience of partial film boiling—a new boiling dissipative structure, which is the second thermodynamics bifurcation on boiling curves, as shown in Fig. 4, which physically



Fig. 5. Conductive heat transfer by vapor film.

means that the system transit to another boiling mode tending to minimize the entropy generation.

3. Dynamics process for thermodynamics bifurcations of boiling structure

Above analysis gives the transition of boiling modes from view of thermodynamics. We now turn to the dynamics analysis, which can reveal the vivid process of boiling modes transitions or bifurcations [9].

As shown in Fig. 3, after the formation of an embryo bubble, the change in the Gibbs free energy is equal to [10]

$$\Delta G = \frac{4}{3}\pi r^3 \rho_{\rm v} (G_{\rm v} - G_{\rm l}) + 4\pi r^2 \sigma \tag{14}$$

where G_v and G_l are the Gibbs free energy for the vapor and the liquid respectively, σ the surface tension, ρ_v the vapor density, r the bubble radius.

Considering the bubble is in equilibrium with the surrounding liquid, the pressure difference across the interface satisfies the Young–Laplace equation

$$p_{\rm v} - p_{\rm l} = \frac{2\sigma}{r} \tag{15}$$

The pressure increase in bubble is obtained by use of Clausius–Clapeyron equation

$$\frac{\Delta T_{\rm s}}{\Delta p} = \frac{T_{\rm s}}{h_{\rm fg}\rho_{\rm v}} \tag{16}$$

Substituting Eq. (15) into Eq. (16) yields

$$\Delta T_{\rm s} = \frac{2\sigma T_{\rm s}}{h_{\rm fg}\rho_{\rm v}r} \tag{17}$$

So bubble radius in equilibrium can be written as

$$r_{\rm e} = \frac{2\sigma T_{\rm s}}{\Delta T_{\rm s} h_{\rm fg} \rho_{\rm v}} \tag{18}$$

Derivative of Eq. (15) with respect to r and setting to zero yields

$$r_{\rm c} = -\frac{2\sigma}{(G_{\rm v} - G_{\rm l})\rho_{\rm v}} \tag{19}$$

 $r_{\rm c}$ is critical radius, apparently $r_{\rm c}$ and $r_{\rm e}$ are equivalent, and comparison of Eqs. (18) and (19) produces

$$r_{\rm c} = -\frac{2\sigma}{(G_{\rm v} - G_{\rm I})\rho_{\rm v}} \tag{20}$$

Substituting Eq. (20) into Eq. (14) yields

$$\Delta G = -\frac{4\pi\rho_v h_{\rm fg}\Delta T_{\rm s}}{3T_{\rm s}}r^3 + 4\pi\sigma r^2 \tag{21}$$

For given system, ρ_{v} , $h_{\rm fg}$, $\Delta T_{\rm s}$, $T_{\rm s}$, σ are constants, ΔG is the function of single variable *r*, the following relationship is satisfied

$$\frac{\mathrm{d}\mathbf{r}}{\mathrm{d}t} = -\frac{\partial\Delta G}{\partial r} = \frac{4\pi\rho_{\mathrm{v}}h_{\mathrm{fg}}\Delta T_{\mathrm{s}}}{T_{\mathrm{s}}}r^{2} - 8\pi\sigma r \tag{22}$$

For the convenience of discussion, Eq. (22) is written in a simplified form as

$$\frac{\mathrm{d}r}{\mathrm{d}t} = -ar + br^2 \tag{23}$$

where $a = 8\pi\sigma$, $(b = 4\pi\rho_v h_{fg}\Delta T_s)/T_s$. Eq. (23) is rearranged as

$$\frac{1}{b}\frac{\mathrm{d}r}{\mathrm{d}t} = -\frac{a}{b}r + r^2 \tag{24}$$

Let t' = bt, Eq. (24) is written as

$$\frac{\mathrm{d}r}{\mathrm{d}t'} = -cr + r^2 \tag{25}$$

where

$$c = \frac{a}{b} = \frac{2\sigma T_{\rm s}}{\rho_{\rm v} h_{\rm fg} \Delta T_{\rm s}} \tag{26}$$

For the convenience of discussion, t' is directly written t in later analyses. So

$$\frac{\mathrm{d}r}{\mathrm{d}t} = -cr + r^2 \tag{27}$$

Eq. (27) is basic dynamics equation of analyzing nucleation processes in boiling system. Eq. (27) provides vivid physical picture: only embryos with certain size that satisfies the condition, i.e. $\dot{r} = dr/dt > 0$ can grow, which marks the incipience of first thermodynamics bi-furcation.

As shown in Fig. 6, because of interactions among active sites or bubbles when considering the existence of multiple bubbles, in term of Eq. (27), equations that describe the growth rate of all bubbles are in the following form

$$\dot{r}_{1} = -c_{1}r_{1} + f_{1}(r_{1}, r_{2}, \dots, r_{n})$$

$$\dot{r}_{2} = -c_{2}r_{2} + f_{2}(r_{1}, r_{2}, \dots, r_{n})$$

$$\vdots$$

$$\dot{r}_{n} = -c_{n}r_{n} + f_{n}(r_{1}, r_{2}, \dots, r_{n})$$
(28)



Fig. 6. Illustration of site or bubble interaction (first thermodynamics bifurcation).

where first term on right side is fluctuation effect, second term is evaporation effect that is the function of multiple parameters when taking effects of interactions among active sites or bubbles into account.

According to well-known adiabatic elimination principle [6], for the interacting multi-elements system described by the Eq. (28), the self-organized evolving process prevails, providing that the damping coefficients c_i are non-uniform. More specifically, the development of elements with small damping coefficients (mean small resistances) will dominate the development of elements with relatively large damping coefficients (mean large resistance).

For the existence of all kinds of stochastic factors in boiling system, the wall temperatures are always nonuniform. According to the definition of c in Eq. (26), the damping coefficients c_i in Eq. (28) are correspondingly always different. In this connection, Eq. (28), which reflects the evolving processes of multi-sites/bubbles on wall surface, as shown in Fig. 6, can be generally classified into two groups: one is i = 1, 2, ..., m, which stands for small-damp modes, that is, small c_i (meaning large $\Delta T_{\rm s}$), even changes into unstable modes ($c_i \approx 0$, which means ΔT_s is large enough). Another is $s = m + 1, m + 2, \dots, n$, which stands for stable modes (meaning relatively small ΔT_s). When wall temperatures reach certain values, first thermodynamics bifurcation will be induced. Then, the development of sites/bubbles (i = 1, 2, ..., m) with small damping coefficients will dominate the development of sites/bubbles (s = m + 1, $m + 2, \ldots, n$) with relatively large damping coefficients.

Analysis above supposes that Eq. (28) can be classified into two groups by damp magnitude. Analyses of self-organization process in boiling system without such limits are as follows. Assuming arbitrary boiling system, in which various parameters can not be explicitly grouped, therefore can be applied to any boiling system

$$\dot{\boldsymbol{r}}_j = \boldsymbol{h}_j(\boldsymbol{r}_1, \boldsymbol{r}_2, \dots, \boldsymbol{r}_n) \tag{29}$$

where h_j is non-linear function of $r_1, r_2, r_3, \ldots, r_n$. Assuming stationary solution of Eq. (29) is r_j^0 and right side of Eq. (29) is dependent on a group of parameters $\beta_1, \beta_2, \beta_3, \ldots, \beta_n$, which are chose in a way so that r_j^0 stands for stable value and satisfies the condition when moving *r* coordinate

$$r_i^0 = 0 \tag{30}$$

It stands for stationary state without any motion, let

$$r_i(t) = r_i^0 + u_i(t)$$
(31)

$$\vec{r}(t) = \vec{r}^0 + \vec{u}(t) \tag{32}$$

which mean r can be decomposed into base and perturbed components. It is useful to do linear stability analysis of Eq. (29). Considering the system is stable and thus u_j is small enough, Eq. (29) is linearized by substituting Eq. (31) into Eq. (29)

$$\dot{\boldsymbol{u}}_{j} = \sum_{j'} L_{jj'} \boldsymbol{u}_{j'} \tag{33}$$

Matrix L_{jj} depends on \vec{r}^0 and $\beta_1, \beta_2, \beta_3, \dots, \beta_n$. Eq. (33) is substituted by

$$\vec{u} = L\vec{u} \tag{34}$$

Eq. (33) or (34) is one order ordinal differential equation and the solution is the form

$$\overline{u} = \overline{u}^{(\mu)}(0) \exp(\lambda_{\mu} t) \tag{35}$$

where λ_{μ} is eigenvalue of the following equation

$$\lambda_{\mu}\vec{u}^{(\mu)}(0) = L\vec{u}^{(\mu)}(0) \tag{36}$$

 $\overline{u}^{(\mu)}(0)$ is right eigenvector, generalized solution of Eq. (33) or (34) is the superimposition of Eq. (35)

$$\vec{u} = \sum_{\mu} \xi_{\mu} \exp(\lambda_{\mu} t) \vec{u}_{\mu}(0)$$
(37)

 ξ_{μ} is an arbitrary constant. Introducing left eigenvector $v^{(\mu)}$ and requiring

$$\lambda_{\mu}\vec{v}^{(\mu)} = \vec{v}^{(\mu)}L \tag{38}$$

For the system is stable, the real parts of λ_{μ} are all negative. If we require that Eq. (31) satisfy the non-linearity, Eq. (29) will have the form

$$\vec{u} = L\vec{u} + N(\vec{u}) \tag{39}$$

 $N(\overline{u})$ results from non-linear contribution. If we still express u in a form as Eq. (36), it is necessary to find

appropriate expression $\xi(t)$ so that the following relation is satisfied when left multiplying Eq. (39)

$$\langle \vec{v}^{(\mu)}, \vec{u}^{(\mu')} \rangle = \delta_{\mu\mu'} \tag{40}$$

Eq. (39) simply becomes as

$$\dot{\xi}_{\mu} = \lambda_{\mu}\xi_{\mu} + g_{\mu}(\xi_1, \xi_2, \dots, \xi_n) \tag{41}$$

where $g_{\mu} = \left\langle \vec{v}^{(\mu)}, \vec{N}\left(\sum_{\mu} \xi_{\mu} \vec{u}^{(\mu)}\right) \right\rangle$. Eq. (41) is thoroughly same as Eq. (28) in form, which implies that analysis in similar way as Eq. (28) can be conducted. We can change parameters and thus destabilize Eq. (41), that is to say, let one or a few λ_{μ} equal to or greater than zero with others having negative real part and thus being relating to damped modes. Considering that mode ξ_r for Re $\lambda_{\mu} \ge 0$ acts as order parameter and dominates other modes, adiabatic elimination principle is effective.

3.1. Dynamics processes for first thermodynamics bifurcation

In real nucleating process in boiling systems, as described above, only one or a few sites in a unit will become unstable while most other sites still remain damped, as shown in Fig. 6. This gives the results that all damped sites follow the order parameters and even complicated systems exhibit ordered behaviors: disordered natural convection system will change into nucleate boiling system. We propose that the superheat affects damped modes. The less the superheat, the larger the damped modes will be. Order parameter with large superheat and thus less damped modes often dominates the whole system behaviors.

Self-organization transition process from natural convection to nucleate boiling is analyzed above, which is a transition from a state characterized disordered natural convection to another ordered state characterized by nucleating bubble [11]. Thermodynamically, the transition is a process minimizing entropy generation. Obviously the system becomes more ordered and nucleate boiling system is a typical dissipative structure. Above analysis provides a vivid dynamics picture of formation of this kind of ordered dissipative structure.

3.2. Dynamics processes for second thermodynamics bifurcation

Similarly, during the transition boiling process, wall temperature is extremely non-uniform and dry patches exist at random as shown in Fig. 7. However, most dry patches will disappear rapidly for continuously being rewetted by liquid. They can be eliminated by adiabatic principle for their greater damped coefficients (which means ΔT_s is smaller) and thus be dominated by one or a few dry patches with smaller damped coefficients (which



Fig. 7. Illustration of dry patch interaction (second thermodynamics bifurcation).

means ΔT_s is larger). These predominant dry patches can grow steadily and eventually promise the transition from nucleate boiling to stable film boiling. It is a transition from a state characterized by nucleate boiling to another state characterized by film boiling [12]. Thermodynamically, the transition is again a process minimizing entropy generation. Obviously the system becomes more ordered and stable film boiling system is a typical dissipative structure.

With the development of modern measurement techniques, studies on view of boiling process are available in references. Keening and Yan [13] stated that active sites can co-exist within one bubble radius of each other, then the sites interact through the variation in wall temperature that each produces, introducing intermittency in the regular production of bubbles. Detailed experimental results and descriptions may be found in reference [13]. Judd and Chopra [14] investigated another different mechanism of interactions. We may also interpret them as self-organized phenomena [15,16]. Here, from thermodynamics and dynamics perspectives, we put forward the mathematical theory of boiling process and present mathematical description.

In a word, the development of boiling modes, as Darwin's picture of a flexible, continuously changing and adapting biosphere under the selection pressure of competition and changing environment conditions, is an evolution kinetic process. As anything in nature, nonlinear characteristic existing in boiling system promises the synergetic or cooperative effects and guarantees a certain self-organized behaviors and functions.

4. Concluding remarks

New ideas of analyzing boiling modes transition are introduced from view of thermodynamics theory and dynamics theory, on basis of which dissipative structure concepts of boiling modes are proposed. The self-organized transition processes both from natural convection to nucleate boiling and from nucleate boiling to stable film boiling are revealed. The present investigations not only provide clear physical picture, but also are based on firm theoretical foundation. Considering that the boiling system is characterized by the existence of non-linear, non-equilibrium, and evolution, the traditional theories are congenitally inefficient. The non-equilibrium thermodynamics theory of boiling system significantly wins the advantage over the traditional theory. Where to lead the developing the theories of boiling heat transfer is a serious problem. Currently many problems, such as site interaction, mode transitions etc., still remain elusive. The present investigations provide a renewed effort in understanding these problems.

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