Explosive Vaporization of Water and Isopropyl Alcohol on a Flat Microheater

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Abstract. The MEMS control systems, such as ink jet printers, optical switches and valves, use the explosive vaporization of metastable liquid for the rapid phase change. The initial stage of explosive vaporization of water, isopropyl alcohol and ethanol has been studied experimentally on the surface of a thin-film microheater, covered with the submicron silicon-carbide layer. In experiments, the multilayer thin-film resistor of the Think Jet printhead with $100 \times 110 \ \mu m^2$ surface area was used. Applying the optical method, we investigated the patterns of the liquid–vapor phase transition under pulse heating and obtained the characteristics of nucleation at the rate of temperature growth of up to 800 MK/s. Pulsed laser illumination was applied for the high-speed photography of liquid vaporization. Data for the vapor-covered surface area vs. time depending on the supplied heat flux and the heating time are reported. The theoretical model of explosive vaporization on a flat heater has been developed and numerical simulations have been performed for water and isopropyl alcohol nucleation on the multi-layer microheater. Comparison of calculation results with experimental data is discussed in this paper.

Keywords: Explosive Vaporization, Microheater, Pulse heating, MEMS

1. Introduction

The MEMS control systems, such as ink jet printers, optical switches and valves, use the explosive vaporization of metastable liquid for the rapid phase change. In recent years, experimental studies on the explosive vaporization under liquid pulsed heating on thin wires and rectangular microheaters were performed. Homogeneous and heterogeneous nucleation becomes dominant for explosive vaporization as distinct from conventional boiling incipience. The kinetic limit of liquid superheat in the metastable state is defined by spontaneous nucleation. Volmer and Weber (1926), Zeldovitch (1942), Skripov (1974) gave physical and mathematical completeness to the classical theory of homogeneous nucleation.
One of the most significant recent applications of explosive vaporization is the ink-jet or bubble-jet printing technology (Allen et al., 1985). An impressive progress in this area during the last decades was accompanied by an increased interest to physics of vaporization near the superheat limit. Detailed experimental data have been obtained for explosive vaporization on the thin wires by Pavlov et al. (1965), Skripov et al. (1970). A series of experiments on the flat microheaters were reported by Okuyama et al. (1988), Asai et al. (1992). The pressure pulse measurements during explosive vaporization were performed by Meyer (1986), Zhao et al. (2000). A few theoretical considerations of explosive vaporization available in literature are founded by the classical theory of homogeneous nucleation (e.g. Skripov, 1974). Meanwhile, none of them accounts correctly the effect of pressure rise near the heated surface due to collective vapor nuclei growth. Although these studies were focused on different aspects of explosive vaporization, the patterns of rapid liquid–vapor phase transition and characteristics of nucleation in liquid at high heat flux remain insufficiently understood.
This article presents the results of experimental study of the initial stage of explosive vaporization of water, isopropyl alcohol and ethanol on the surface of a flat microheater coated with a submicron silicon-

carbide layer. Applying the optical method, the patterns of the liquid–vapor phase transition under pulse heating and characteristics of nucleation at the rate of temperature growth of up to 800 MK/s were obtained. Numerical simulations have been performed for explosive vaporization of water and isopropyl alcohol on the multi-layer microheater. The comparison of calculation results and experimental data is discussed in this paper.

2. Experimental Setup

The experimental setup is shown in Fig.1. Resistors array of a printhead for the Hewlett Packard ThinkJet printer was used as a heaters board. The set of multilayer resistors of 100×110 μm^2 heater surface was deposited layer-by-layer by the PECVD on a glass substrate including the electrical pads. The first layer is silica with thickness of 1.1 μm, the second layer is heat-emitting TaAl film whose conduction is weakly dependent on the temperature, the third layer is 0.5 μm N_2O_4 film, and last layer is 0.25-μm SiC film (Bhaskar and Aden, 1985). This sandwich-like structure of the heater ensures the high growth rates of the surface temperature under pulse heating.

One resistor was selected for the test and its electrical pads were connected to the power supply using the coaxial cable. The microchip (1) was immersed into working liquid (2) open to the atmosphere. The single current pulse
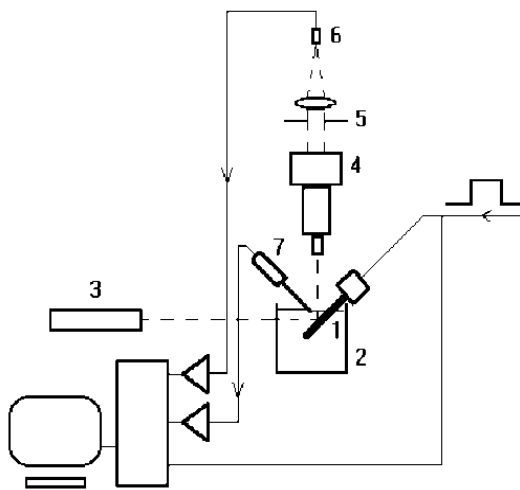

Fig. 1. Schematic diagram of experimental setup.

from HP 214 pulse generator was applied to the microheater for liquid heating up. The pulse had duration and amplitude, sufficient to ensure explosive vaporization. The heat flux into fluid reached the values of 400 MW/m^2. The corresponding temperature rate of the heater surface was about $8 \cdot 10^8$ K/s.

An optical method for recoding nucleation and dynamics of the vapor cavity was developed. It is based on measuring the intensity of He–Ne laser beam reflected regularly from the heater surface. The microscope with photo camera adapter (4) created the image of the heater under reflected light of the laser (3). Rectangular aperture (5) in the image plane cuts out the necessary area of the resistor image, which was focused further on photoreceiver (6). The 75-fold heater's image was created in the diaphragm plane. Then the light beam was focused onto the face of the light fiber with a diameter of 400 microns. The light optical signal modulated with the process of firing was received by high-frequency (up to 40 MHz) optic-electronic converter and was registered by the current signal with help of a fast (50 ns/count) ADC.

When vapor bubbles appear on the heater surface and grow up to the sizes comparable with the laser wavelength (0.6 μm), the integrated coefficient of specular reflection diminishes and an inverted signal from the photodetector yields the dynamics of filling the heater surface with microbubbles. It eventually reaches almost zero value, when bubbles occupy all surface of the heater. A high-frequency storage oscilloscope and fast (50 ns/reading) digital recording system assembled by PC was applied to record the signals. The experiments involved simultaneous recording of the current and voltage pulses, optical reflection and pressure probe signals. The numerical solution to the Fourier equation for multilayer heater was performed to define the resistor surface temperature and heat flux corresponding to the time of boiling incipience.

A pulsed solid-state laser with 20 ns flash duration was used also to make image of very small growing bubbles on the heater surface.

3. Experimental Results

Typical patterns of ethanol decay under pulse heating are shown in Fig. 2. Here, time t is measured from the start of heating and effective heat flux q_e is determined as the total amount of heat, divided by heater surface area. Nucleation occurs, when liquid temperature achieved the homogeneous nucleation limit (Skripov, 1974) in the surface layer on the heater. It can be identified with the phase explosion, whose properties are determined by the physical properties of liquid. At the initial stage of bubble nucleation the bubbles are unevenly distributed across the surface of the heater, and their number is small. The emission of pressure waves was observed at the time of bubbles origination. In the final
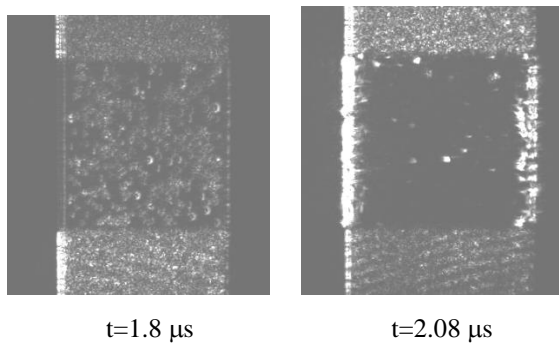

t=1.8 μs   t=2.08 μs

Fig. 2. The patterns of ethanol decay for $q_e = 521.5$ MW/m^2, $dT_w/dt = 85.7$ MK/s.

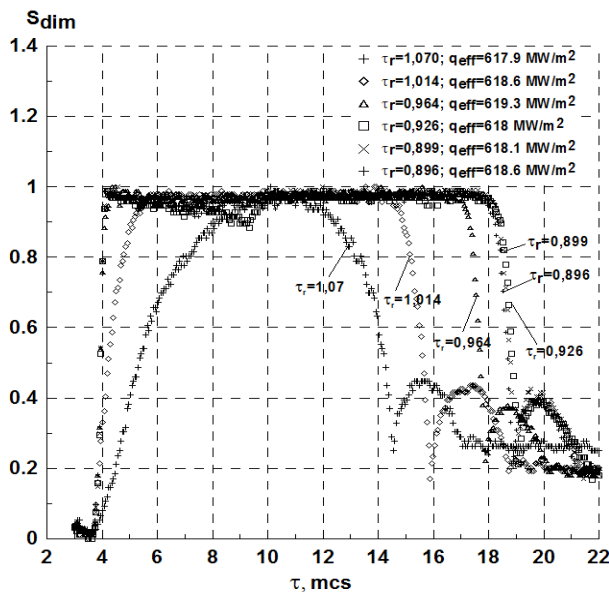**Fig. 3.** Dependence of reflected light intensity on time at effective heat flux is about 618–619 MW/m^2.
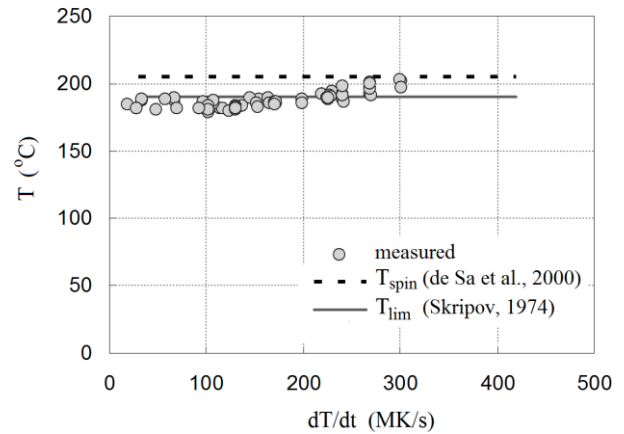

Fig. 4. Nucleation temperature of isopropyl alcohol depending on liquid temperature growth rate.

stage of liquid decay the heater surface is covered with a cloud of bubbles, which combine then to form a continuous vapor film (see Fig. 2 at t=2.08 μs).

The experiments show that water phase explosion occurs at higher heat fluxes and the number of bubbles is much less than that for the alcohol. It shows a significant influence of liquid physical properties on the decay patterns.

Figure 3 shows dynamics of coverage of the heater surface by vapor bubbles during phase explosion of water for different relative times of decay initiation. The relative time of decay initiation τ_r is defined as the ratio of heated pulse duration to nucleation time. As can be seen, water decay is explosive in nature and occurs in the period less than 300 ns.

Figure 4 shows the dependence of nucleation temperature on liquid temperature growth rate for isopropyl alcohol. When the temperature growth rate is less than 200 MK/s, the nucleation temperature is close to the temperature of homogeneous nucleation limit (Skripov, 1974). For the higher growth rate it begins increasing with an increase in dT_w/dt and approaches the spinodal temperature. The calculation of spinodal according to de Sa et al. (2000) is shown in Fig. 4 by the dotted line. Experiments showed that the nucleation temperature for water remains below the temperature of homogeneous nucleation limit, if the rate of temperature growth is less than 400 MK/s. It is caused by reduction of the work of critical nucleus appearance on the

poorly wetted surface (Skripov, 1974).

Measuring amplitude of the emitted pressure wave shows that bubble appearance during nucleation is suppressed by the pressure growth. It proves the interconnection of hydrodynamic, thermal and acoustic processes during intensive phase transition at phase explosion near the temperature of homogeneous nucleation limit. Such approach is particularly important for selection of the mathematic model of explosive evaporation, where the pressure field development near the heater surface should be taken into account.

4. Explosive Vaporization Model

The theoretical model of decay of metastable liquid is based on the classical theory of homogeneous nucleation of Volmer and Weber (1926), Skripov (1974). In the nucleation model, frequency of nucleation is given as follows:

$$J_{\text{hom}} = N_1 B \exp(-G), \quad G = W_*/kT, \quad (1)$$

here $W_* = 16\pi\sigma^3/3(p_{sat} - p_f)^2(1 - \rho_g/\rho_f)$ is the work of critical nucleus appearance, N_1 is the number of molecules per a unit volume, B is the kinetic coefficient that takes into account dynamics of the vapor bubble growth (Kagan, 1960). The reduction of wettability of solid surface decreases the work of critical nucleus formation (Skripov, 1974):

$$W_*^{'} = W_*\psi(\theta), \quad (2)$$

and frequency of nucleation determined by heterogeneous nucleation mechanism on a flat surface is determined as follows:

$$J_{het} = N^{2/3}\frac{1+\cos\theta}{2}\sqrt{\frac{6\sigma}{(3-b)\pi m\psi(\theta)}}\exp[-G\psi(\theta)]. \quad (3)$$

The total number of bubbles nucleated in the volume of liquid with non-uniform temperature in time τ is determined as the integral, where it is recorded as heterogeneous and homogeneous nucleation mechanisms:

$$n(\tau) = S_0\int_0^\tau\left\{J_{\text{het}}(T_w,q_w,p_f)+\int_0^\infty J_{\text{hom}}[T_f(z,t),p_f]dz\right\}\cdot \\ (1-\tilde{S}_g(t))dt. \quad (4)$$

Here S_o is surface area of a heater, $\tilde{S}_g = S_g/S_0$ is normalized area of a heater surface occupied by vapor bubbles and S_g is the area of a heater surface occupied by vapor bubbles. Frequency of heterogeneous nucleation accounts nucleation on low energy spots according to Eq. (3).

The important feature of explosive vaporization is radiation of the pressure wave (Meyer, 1986; Kuznetsov and Vasserman, 2004). One supposes that nucleation takes place in a vicinity of the heater surface. For this case, the bubbles are localized on the surface and the pressure increase is due to inertia of liquid, surrounding the heater, see Fig. 5. The pressure in a vicinity of the heater is set as p_f and the pressure in the surrounding fluid away from the heater is set as p_∞. The pressure in a vicinity of the heater, averaged over its surface, can be determined using calculation of acoustic radiation in accelerated motion of a flat piston with an effective diameter equivalent to the surface area of the heater. Let $z_g(\tau)$ is the displacement of fluid on the outer boundary of the bubble layer caused by emergence and growth of bubbles $z_g(\tau) = V_g(\tau)\cdot(1-\rho_g/\rho_f)/S_0$ where $V_g(\tau)$ is the total volume of vapor, accumulated at time τ. Then the pressure in a vicinity of the heater, averaged over its surface, can be determined as follows (Rayleigh, 1945):

$$m_p\,d^2z_v/d\tau^2 = S_0(p_f - p_\infty), \quad m_p = 8\rho_f l^3/3\pi^{3/2}. \quad (5)$$

The total volume of vapor can be represented as the integral over time from superposition of homogeneous and heterogeneous nucleation:

$$V_g(\tau) = S_0\int_0^\tau\left\{J_{\text{het}}(T_w,q_w,p_f)+\int_0^\infty J_{\text{hom}}[T_f(z,t),p_f]dz\right\}\cdot \\ V_b(\tau-t)(1-\tilde{S}_g(t))dt. \quad (6)$$

Here $V_b(\tau-t)$ is the volume of gas bubble nucleated at time t and S_g is implicitly defined as follows:

$$S_g(\tau) = S_0\int_0^\tau\left\{J_{\text{het}}(T_w,q_w,p_f)+\int_0^\infty J_{\text{hom}}[T_f(z,t),p_f]dz\right\}\cdot \\ S_b(\tau-t)(1-\tilde{S}_g(t))dt. \quad (7)$$
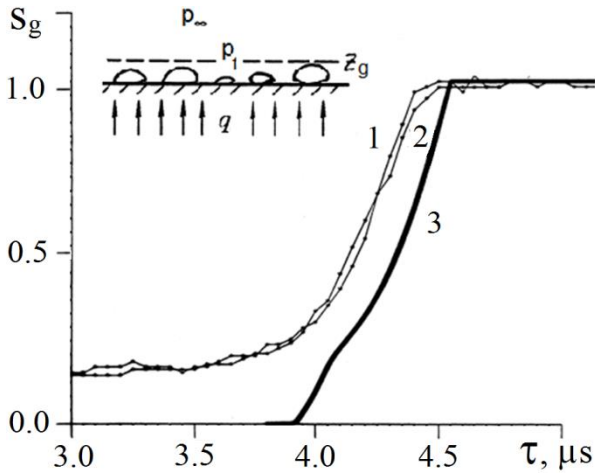
Fig. 5. Variation of the normalized area of a heater surface occupied by vapor bubbles with time.

where $S_b(\tau, t)$ is the area of bubble projection nucleated at time t on the surface of the heater. Together with the equations of hemispherical bubble growth, these equations allow us to calculate the decay of metastable liquid.

The closed self-consistent model of spontaneous nucleation and growth of nuclei in a flat microheater at decomposition of bubbles of deeply metastable liquid can be constructed from the following equations: Eqs. (1) − (7), bubble growth equation and Fourier equations for the heater, liquid around the bubble, microlayer under the bubble and vapor phase. Figure 5 shows the comparison of calculation results of heater surface coverage by vapor bubble in time (line 3) with the experimental data for water (lines 1 and 2). The experimental data were obtained for two heater firing followed one after the other. In calculations, the reduction of critical nucleation work on the silicon-carbide surface was taken into account. The pressure increase during explosive vaporization limits the number of emerging bubbles and allows us to predict correctly the initial stage of liquid-vapor phase transition, bubbles growth and collapse at the high rate of thermal energy supply.

5. Conclusions

Experimental study of formation of the metastable state and its decay under pulsed heating of liquid showed that duration of the

phase of explosion is less than 300 ns. The decrease in activation energy for nucleation on the poorly wetted surface was determined for water. For isopropyl alcohols, the nucleation temperature is close to the temperature of homogeneous nucleation limit and tends to spinodal at the rate of temperature growth of more than 200 MK/s.

A new model of explosive vaporization on the heater surface is proposed. It accounts for the pressure increase near the heater surface during bubble nucleation. The calculation of explosive evaporation for water shows a good agreement with experimental results, if the reduction of critical nucleation work is taken into account.

Acknowledgments

Nomenclature

B kinetic coefficient [s^{-1}],
J homogeneous nucleation rate [m^{-3} s^{-1}],
J_{het} heterogeneous nucleation rate [m^{-2} s^{-1}],
k Boltzmann constant [J/K],
N_1 molecules number density [m^{-3}],
n total number of bubbles [dimensionless],
P pressure [N/m^2],
q heat flux [W/m^2],
S_o resistor surface area [m^2],
S_g surface area occupied by vapor [dimensionless],
T temperature [K],
t nucleation time [s],
V_g total vapor volume [m^3]
W_* critical nucleation work [J],
z coordinate, normal to resistor surface [m]
ρ density [kg/m^3]
τ time [s]
σ surface tension [N/m]

Subscripts
g gas
f liquid
w wall

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