EXPERIMENTAL STUDY OF THE GAS ENTRAPMENT PROCESS IN CLOSED-END MICROCHANNELS

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ABSTRACT
Earlier studies have shown that for cavities present on any heater surface to become active nucleation sites during boiling, they should entrap gas. The liquid penetrates the cavity due to the capillary and surface forces, but the exact physical mechanisms have not been fully quantified. The physical mechanisms of the gas entrapment process in closed-end microchannels, representing nucleation sites, are investigated in this study. Aside from the fluid properties, the width, length and depth of the cavities, as well as the static contact angle of the test liquid with the solid are considered as main parameters that influence the gas entrapment process. Test pieces consisted of micromachined silicon dices with glass bonded on top. Widths of 50, 30, 15 and 5 μm were chosen based on size distribution probability. The mouth angle was 90° in all cases. Test pieces were held horizontally under a microscope equipped with a CCD camera. A drop of liquid was placed at the entrance of the microchannel and capillary and surface forces drive the liquid into the microchannel. Experiments show two main filling behaviors: (1) A uniform meniscus forms at the entrance and moves inwards, (2) Two menisci: one at the entrance and the other at the closed end of the microchannel. In some cases droplet formation at the walls was observed. A single meniscus typically forms for higher contact angles, while two menisci form for lower contact angles. In all cases, after a sufficient time interval (hours to days) the microchannel was completely flooded. In general, for a given depth, wider microchannels take longer to fill. Surface cleanliness and fabrication process also play a role in modifying the contact angle and hence the time taken to fill the microchannel. A comparison of the interface advancement in the microchannel with a simple mass diffusion model shows reasonable agreement.

INTRODUCTION
Earlier studies have shown that for cavities present on any heater surface to become active nucleation sites during boiling the cavity should entrap gas inside. The liquid penetrates the cavity due to the capillary and surface forces, but the exact physical mechanisms have not been fully quantified.

Gas Entrapment for Boiling Nucleation
During boiling, vapor bubble formation starts from nucleation sites on the heated surface, where the liquid has trapped gas or vapor inside. These nucleation sites are typically a few microns in size. The ability of a liquid to penetrate into the microscopic cavities depends on factors such as the contact angle of the liquid and the size of the cavity. However, once the liquid penetrates into the cavity, it can either entrap the gas/vapor present in the cavity or flood the cavity entirely.

Bankoff (1958) proposed a quantitative criterion for gas entrapment in a wedge by an advancing liquid front. The critical parameters affecting the gas/vapor entrapment in such a cavity were the liquid contact angle and the wedge angle. Conical cavities were approximated as wedge shaped grooves. According to his criterion, a wedge-shaped cavity on a surface will trap vapor/gas when the contact angle is greater than the wedge angle.
Wang and Dhir (1993a) studied the effect of surface wettability on active nucleation site density during pool boiling of water. The objective of their work was to relate the cavities that are present on a surface to those that become active. They used a small copper surface (2 cm diameter) and were able to determine the number density, diameter, and side angle of cavities present on the surface. To vary the surface wettability, the surface was oxidized and static contact angles were used as a measure of the degree of wettability.

The cavity mouth diameter was determined with an optical microscope. Pictures were taken for areas with cavities ranging in size from 3.3 to 56 μm. The shape of the cavities was determined by removing material to a certain depth and remeasuring the cavity diameter. Thus a probability density function for cavity diameters was determined.

The number density of active sites was determined by taking still pictures of a 1 cm² area of the heated surface during pool boiling. Once the active sites were identified their diameter and side angle were then measured.

Based on the detailed measurements, they found that for cavity diameters > 6 μm, the cumulative number density of all cavities present on the surface varied inversely to the square of the cavity diameter. On the other hand, for cavity diameters < 6 μm, the cumulative number density of cavities showed a much stronger dependence on the cavity diameter. Based on the measured cavity sizes and shapes they found that most of the deep cavities present on the surface were spherical in nature. Although a large number of conical cavities were present in the surface, these cavities were very shallow, which according to Bankoff’s criterion were not expected to nucleate. It was found that as the wettability of the surface improves (i.e., contact angle decreases), the number density of cavities that actually nucleate decreases.

Wang and Dhir (1993b) evaluated the change in the Helmholtz free energy of a liquid droplet placed at the mouth of a cavity, with the free surface of the droplet exposed to gas or vapor. The equilibrium position of the interface (which corresponds to the minimum Helmholtz free energy) was calculated by varying the location of the liquid-gas interface. The change in the dimensionless Helmholtz free energy between any two interface positions was written as,

\[
\Delta F^* = \Delta F / (\sigma \sigma^{ab}) = [(A_2^{lb} - A_1^{lb})] + [(A_1^{sl} - A_2^{sl}) \cos \phi] / r_c^2
\]  

(1)

where \(\Delta F^*\) is the change in the Helmholtz free energy and \(\Delta F\) is the dimensionless Helmholtz free energy, \(A^{lb}\) and \(A^{sl}\) represent the liquid-gas and solid-liquid interfacial areas, respectively, \(r_c\) is the cavity mouth radius and \(\phi\) is the contact angle. Subscripts 1 and 2 refer to the interface positions. Figure 1 shows a sketch of the control volume for a liquid droplet placed on a spherical cavity.

For a spherical cavity with side angle (\(\psi_m\)) of 30° they showed that for smaller contact angles (\(\phi < \psi_m\)), the relative free energy decreases as the interface goes to the bottom of the cavity. For these contact angles no gas will be trapped. However, for higher contact angles (from 60° to 150°) the relative free energy reaches a minimum at \(\alpha_s = 180° - \phi\) and then increases to a maximum value at \(\theta_s = 90° - \phi\). Beyond \(\theta_s = 90° - \phi\), the free energy decreases. This suggests that the interface will move from its position above the cavity to the location where \(\Delta F^*\) is minimum and will hence entrap gas in the cavity. From the above analysis, it was concluded that the condition for entrapment of gas could be stated as,

\[
\phi > \psi_{min}
\]  

(2)

where \(\psi_{min}\) is the minimum cavity side angle. A similar analysis was done by Warrier (2002) for a cylindrical cavity. For spherical, conical, and cylindrical cavities, \(\psi_{min} = \psi_m\). Figure 2 shows the experimental data obtained by Wang and Dhir (1993b) relating the contact angle and cavity mouth angle for active nucleation sites on a copper heater surface with water as the test fluid (\(\phi = 90°\)). From Fig. 2 it is clear that all the cavities that entrapped gas and were hence active nucleation sites satisfied the condition that \(\phi > \psi_{min}\). It should be noted that the model of Wang and Dhir was a quasi-static model, which did not consider diffusion of gas into the liquid.

Once a cavity entraps gas, increasing the temperature of the cavity or the liquid may result in an increase in the vaporization at the liquid-gas interface. According to Carey (1992), the degree to which the entrapped gas helps vaporization depends on the rate at which the entrapped gas diffuses into the liquid and is carried away from the interface. Thus, once all the entrapped gas has diffused into the liquid, the cavity will be flooded.

![Figure 1 Positions of the liquid front in the control volume (from Wang & Dhir (1993b)).](Image)
Washburn (1921) derived an equation to describe the spontaneous motion of a liquid in a capillary. His equation expresses the balance of the capillary force, the gravity force and the viscous force given by Poiseuille (1846). Since the inertial forces neglected, the Washburn equation predicts an infinitely high initial velocity for the liquid during the early stages of interface advancement. It was Szekely et al. (1971) who removed this discontinuity by applying the correct energy balance for the entry flow. However since these studies where for open-end capillaries, they do not consider the effect of the gas entrapped within the capillary. L.J. Yang et al. (2002) measured the marching velocity of capillary menisci in microchannels (2mm long). They also developed an analytical model to calculate the marching velocity of the meniscus. In this model the velocity profile was assumed to be parabolic and the pressure difference across the liquid-gas interface was calculated using the “Laplace pressure drop”. Neglecting the acceleration term, they found that the position of the interfaces was proportional to the square root of time. In general their experiments showed that filling time increases with increasing cross sectional area of the microchannels. However, there is no mention as to whether the microchannels had an open or closed end. Also, no information is given regarding the filling mechanisms involved or the role of mass diffusion. Akselrud and Altshuler (1983) proposed that the degree to which the entrapped gas diffuses into the liquid significantly affects the nature of filling of a closed-end capillary. Dovgyallo et al. (1989) observed the phenomenon of bilateral filling of conical capillaries with a closed end. They experimentally studied the filling of closed end capillaries, with lengths varying from 30 to 1000 µm and radii from 0.4 to 15 µm with various liquids (water, ethyl alcohol and acetone), establishing that in a number of cases the capillary is filled not only from the open end, but also from the closed end as well. However, for experiments with cylindrical capillaries, they found that the capillaries are filled only from the open end. In the conical capillaries, the closed end had the smaller cross sectional area. This difference between cylindrical and conical capillaries was associated with the presence of two menisci of different curvatures in the conical case. They theorized that the second meniscus forms at the closed end of the capillary due to condensation from the liquid vaporized from the surface of the meniscus with lower curvature. They also found that, in the case of cylindrical capillaries, the rate at which air dissolves into the liquid was considerably lower than for the conical capillaries. Migun and Azuni (1996) carried out experiments for larger conical capillaries than those considered by Dovgyallo et al. They observed the same double side filling phenomena. In capillaries of radius, \( R \geq 50 \mu m \) they also observed individual drops of water on the channel walls.

From the above discussion it is clear that for a given cavity size (and shape) liquids with certain contact angles will entrap gas in the cavity. Though the important parameters that affect the dynamics of the liquid-gas interface and subsequent entrapment of gas appear to be the liquid contact angle, cavity size, and liquid properties such as surface tension and viscosity, no quantitative information is available regarding the effects of each of these parameters. Additionally, no quantitative information is available regarding the effects of gas diffusion on the dynamics of the interface and the gas entrapment process.

The objective of this study is to develop a basic understanding of the dynamics of liquid-gas interfaces in cavities and the process by which gas can be entrapped in these cavities. This experimental work will focus on the effects of the following parameters: cavity geometry (depth, mouth width, and length), as well as the static contact angle of the test liquid.

**NOMENCLATURE**

- \( A \) interfacial area
- \( c \) molar concentration
- \( d \) microchannel (etched depth) length
- \( D_{12} \) binary diffusion coefficient for chemical species 1 & 2
- \( He \) Henry Number
- \( J \) diffusive mass flux
- \( L \) position of the liquid-air interface inside the cavity
- \( L_0 \) depth of microchannel
- \( P \) pressure
- \( R \) radius of meniscus
- \( r_c \) cavity mouth radius
- \( w \) cavity mouth width
- \( x_i \) mole fraction of species i

**Greek**

- \( \alpha_s \) slope of solid surface
- \( \Delta F \) Helmholtz free energy
- \( \Delta F^* \) dimensionless Helmholtz free energy
- \( \theta_s \) angular coordinate in a spherical cavity
- \( \sigma \) surface tension
- \( \phi \) contact angle of the liquid with solid surface
- \( \psi_m \) cavity mouth angle

**Superscripts**

- \( La \) liquid/gas interface
sl  solid/liquid interface
Subscripts
1  liquid front state 1, or species 1
2  liquid front state 2, or species 2
i  species i
u  liquid phase
s  gas phase

EXPERIMENTAL APPARATUS
Since it is impossible to conduct these experiments with actual cavities, test samples were designed to simulate cavities. In these test samples, the cross section of a cavity is simulated. Hence cylindrical cavities will be represented as rectangular closed end microchannels. Samples were fabricated with four entrance widths (5, 15, 30 and 50 µm) and three depths (50, 150 and 500 µm). The length (dimension normal the plane of the pictures, etched length) was about the same as the width and varied from 7 to 42 µm.

Test samples were built with four inch silicon wafers, using standard micromachining techniques. The samples were etched to the required depth and then diced into 2 x 2 cm pieces. Borofloat glass was anodically bonded to these samples. Figure 3 shows the schematic of the test samples. Each test sample had 3 to 4 microchannels on it.

The criteria for choosing the mouth entrance widths for the microchannels was taken from Wang and Dhir (1993a), where they experimentally measured the cavity mouth diameters on a copper surface and found cavity sizes from 55 µm to 2.2 µm. With their results in mind, cavity mouth widths in the range of 50 to 5 µm were chosen for the present study.

EXPERIMENTAL PROCEDURE AND DATA REDUCTION
The first experiment run for each test piece was conducted without any further surface cleaning, assuming that the microchannel remained clean during the anodic bonding. The anodic bonding process subjects the test pieces to temperatures of around 300°C for two hours.

Before each experiment, the contact angle between the liquid droplet and the silicon surface was measured, so as to quantify the condition of the silicon surface inside the cavity. Since it was not possible to place a droplet in the microchannel, a small drop of the test fluid was placed in the reservoirs using a syringe and an instantaneous photograph was taken. The contact angle was measured from this photograph.

Once the contact angle was recorded, the test piece was placed under the microscope equipped with a CCD camera attached to a framegrabber and computer. The test liquid was then placed in the reservoir and the entire filling process was recorded. Both the liquid and test sample were at room temperature (about 23°C).

After the first use of a test sample, it was thoroughly cleaned using a combination of the following processes:
1) Piranha (4 : 1 mixture of sulfuric acid (H₂SO₄) with hydrogen peroxide (H₂O₂)) bath at room temperature for 50 minutes, thereafter rinse with DI water for five minutes, blow dried with nitrogen and to dehydrate, bake at 110°C for 40 minutes.
2) Supercritical Dryer¹, with CO₂ and pure methanol for one hour.
3) Bake at 160°C for 90 minutes by placing the sample on a hot plate.

The purpose of the surface cleaning was to modify the contact angle of the test fluid in contact with silicon. The contact angles corresponding to the cleaning procedures explained before, are listed as follows:

a) After Anodic Bonding (before first use of new piece): ~ 60°
b) Used piece after Piranha Bath (including rinsing and dehydration bake): ≤ 15°

¹ Supercritical CO₂ drying process allows samples to be dried without any surface tension. This process is typically used to dry samples that have been rinsed in de-ionized water. The de-ionized water is replaced by methanol prior to the drying process, and then the methanol is displaced by liquid CO₂ as part of the drying process. This process may also be used to clean samples.
c) Used piece after Piranha Bath followed by Supercritical Dryer: $\sim 0^\circ$
d) Same as (b) followed by hot bake at 160°C for 40 minutes: $\leq 25^\circ$

The contact angles corresponding to the cleaning procedures (b), (c), and (d), were obtained from the shape of the meniscus inside the microchannel.

The distance from the entrance of the microchannel to the liquid-air interface was measured from the images captured. At the beginning (first 20 images) of the experiments an image was captured every 10 seconds and afterwards every 30, 60 and 120 seconds depending on the test liquid, contact angle and microchannel geometry.

Once the liquid had penetrated the microchannel, the contact angle inside the microchannel was measured from the shape of the meniscus. This was considered to be the actual contact angle. Since two or more surfaces were in contact with the liquid, a mean value was used for the contact angle.

The error associated with the measured penetration depth was about ±4 pixels. Hence the calculated error for the penetration depth was about ±5 µm. The uncertainty of temporal measurements is 10 seconds, which is given by the frame rate of the camera software. The uncertainty for the contact angles measured on the reservoir of the test samples was about ±5°, whereas the contact angles measured inside the microchannels had an uncertainty of ±8°.

RESULTS AND DISCUSSION

Experiments to study the filling of one-side-closed silicon microchannels were carried out with DI water as test fluid. For the rectangular microchannels the mouth widths (w) varied from 9 to 57 µm and the length (d) varied from 7 to 42 µm while their depths (Lo) varied from 50 to 500 µm.

Figures 4 through Figure 8 show the shape of water-air interface and the location of the interface in the microchannel with time. The liquid is entering from the left and is the darker shade. The lighter gray shade is the part of the microchannel that is still filled with air. Two different types of filling behavior were observed; (i) Only one interface is present which progresses from the left to the right. (ii) Two interfaces are seen, one moving from the left to right and the other moving from right to left. In Fig. 4(a) only one uniform water-air interface can be seen. From the measured penetration depth it can be seen that the motion of the liquid-air interface is not smooth. The interface moves intermittently; the interface moves, remains stationary and then moves again. However close to the closed end of the microchannel, the interface moves quite rapidly.

A variation of this filling mechanism (i.e. only one interface) occurs when in addition of the single interface, liquid droplets are also present on the channel walls as shown in Fig. 5(a). It can be seen that the initial droplets merge to form larger droplets and sometimes even merge with the advancing interface, which causes sudden jumps in the interface location. These sudden jumps are clearly seen in Fig. 5(b).

![Figure 4 Front advancing interface.](image_url)

![Figure 5 Front advancing interface, with droplets.](image_url)
A second mechanism by which the channels can be filled has also been observed. In this mechanism, which occurs at low contact angles, at least two interfaces are present; one at the entrance of the microchannel and the other at the end of it (Fig. 6(a)). Referring to the sequence of photographs shown in Fig. 6(a), initially only one interface is present but after 52390 s a second interface appears close to the closed end of the microchannel and starts moving towards the entrance, finally meeting up with the front interface and thus filling the microchannel. In most of the cases, when two interfaces were present, a thin film of liquid on the walls of the channel connected the two menisci. Figure 6(b) shows the corresponding interface locations as a function of time.

Figure 6  Front and back advancing interfaces, with presence of thin film.

A variation of this second mechanism was also observed. In this case, in addition to the two menisci, liquid droplets were also present. This can be seen in Fig. 7(a). The film connecting the two menisci can be clearly seen. The droplets on the walls of the channel grew and merged with each other and with menisci.

Figure 7  Front and back interfaces, thin film and droplets.

In some instances more than two interfaces were seen, usually for longer channels. This generally happened as a result of droplets growing and merging to fill up the channel and forming an interface. This is the case shown in Fig. 8(a), which starts with only one meniscus but after 23385 s the liquid flowing along the side walls feeds the droplets present in the middle of the channel with enough liquid to grow suddenly and merge forming two new interfaces, one moving towards the closed end of the microchannel while the other advances towards the first interface located closer to the entrance of the channel. Later, at 28425 s, the middle meniscus that was moving towards the entrance merges with the original front meniscus. The described behavior can be seen clearly in Fig. 8(b), which shows the position of the interfaces as a function of time. There are three distinct stages: one corresponding to the original front interface (until 23385 s), noted in the figure as diamonds, the second is the creation of the second and third interfaces, seen as crosses and triangles, respectively, and finally the merging of two interfaces (at 28425 s crosses meet diamonds symbols) leaving only one interface (triangles) closer to the end of the channel.

All the microchannels with rectangular cross section tested in this study were completely flooded given sufficient time. The time taken to completely flood the microchannel varied from seconds, to minutes, to hours to even days.
As expected, for a given cross section, deeper microchannels take longer to be flooded. This can be seen in Figs. 9(a) through 9(e) where the time taken to flood a microchannel is plotted as a function of its depth for microchannel mouth widths of 9, 19, 24, 38 and 57 µm, respectively. The contact angles for each data point are indicated in Fig. 9. Also, for most of the cases, the time to flood the microchannel showed to increase as the mouth width increased as shown in Fig. 10(a) through 10(c) for a given depth. This is due to the fact that the effect of the bounding walls diminishes. From Fig. 11(a) through 11(e) it can be seen that, in general, as the contact angle increases, the time taken to flood the microchannel also increases, while depth, mouth width and length are kept constant. Again pointing to the role of solid-liquid force, which increases as the surface wettability improves. This force resulting from disjoining pressure pulls the liquid along the wall.

The experiments conducted, indicate that for $\phi \leq 40^\circ$, two interfaces connected by a thin liquid film were always observed. On the contrary, for $\phi > 40^\circ$, only one interface was observed.

**Figure 8** Front and middle advancing interfaces.

**Figure 9** Flooding time as function of depth for different microchannel mouth widths.
MODEL DEVELOPMENT

Both the surface forces and mass diffusion determine the advancement of the liquid-gas interface in the cavity. A one-dimensional transient mass diffusion model was used to assess the role of diffusion of gas into liquid.

In this model the liquid is assumed to be a semi-infinite medium. The air is assumed to diffuse into the liquid, while the diffusion of the liquid into air is assumed to be negligible. Figure 12 shows the control volume for the model.

The molar flux of air diffusing into water is given by,

\[ J_{1,\text{a}} = c_{1,\text{a}} \sqrt{\frac{D_{12}}{\pi \cdot t}} (x_{1,\text{a}} - x_{1,0}) \]  

\( (3) \)
where $J_{1,u} \text{[mol/m}^2\text{s]}$ is the molar flux of air diffusing into water, $c_{\text{liq}} = \rho / M$ in mol/m$^3$ ($\rho$ is density and $M$ is the molecular weight) is the molar concentration of water, $D_{12}$ is the diffusion coefficient, $x_{1,u}$ and $x_{1,0}$ are the mole fractions of air at the interface and the initial mole fraction of air in the fluid, respectively, and $t$ is time. The gas pressure is taken as the liquid pressure plus the capillary pressure, i.e.,

$$P_{\text{gas}} = P_{\text{liq}} + \frac{\sigma \cos \phi}{R},$$

$$\frac{1}{R} = \left( \frac{1}{R_1} + \frac{1}{R_2} \right) = \frac{2 \cos \phi (w + d)}{w \cdot d}$$

The initial concentration of air ($x_{1,0}$) in water is assumed to be zero, i.e., ($x_{1,0} = 0$) and that there is no liquid present in the air. Hence, $x_{1,s} = 1$.

The mole fraction of air at the interface ($x_{1,u}$) can be calculated as $x_{1,u} = \frac{x_{1,s}}{H_e}$, where $H_e$ is the Henry number calculated as $H_e = \frac{C_{He}}{P_{\text{gas}}}$, where $C_{He}$ is the Henry constant for $P_{\text{gas}}$ in Pa. In the present work a value of 0.689 was used for the Henry number.

Using the numerical values for the fluid and air properties given by Mills (1999), the diffusion coefficient was calculated to be $D_{12} = 2.77 \times 10^{-9}$ m$^2$/s.

The distance of the interface from the entrance of the channel, as a function of time, can be calculated by applying the mass conservation equation for the volume of air in the channel, which is given as,

$$J_{1,u}(t) \cdot A = -\frac{d}{dt} (c_{\text{air}} \cdot Vol_{\text{air}})$$

(4)

where

$$Vol_{\text{air}} = (L_0 - L(t)) \cdot A$$

(5)

and $c_{\text{air}} = \rho / M$ in mol/m$^3$ is the molar concentration of air. Substituting for $J_{1,u}$ and $Vol_{\text{air}}$ in Eqn. (4) and integrating yields the position of the interface as,

$$L(t) = \frac{2c_{\text{liq}}}{c_{\text{air}}} \sqrt{\frac{D_{12} t}{\pi}} \cdot x_{1,u}$$

(6)

Equation (6) was used to predict the position of the interface as a function of time. Figure 13 shows a comparison of the model prediction with the experimental data for various cases. In general, the mass diffusion model gives higher values for the position of the interface inside the microchannel compared with the ones obtained from experiments, but the general behavior is similar. The first experimental data point was used as the initial position (at $t = 0$) for $L(t)$ for the model. For the filling mechanism with only one meniscus (Figs. 13(a) and 13(b)) the model generally over predicts the penetration depth. The over prediction can be due to a number of factors such as specification of gas concentration at the interface, initial position and movement of the interface and non uniformity in the channel dimensions. When two menisci were present, each one was treated independently. For example, for the case shown in Fig. 13(c), when a second meniscus appears after some given time, its position (as measured from the experiment) at the time of creation was set as the initial interface position in the model. It can be seen in Fig. 13(c) that the microchannel has been completely filled when front and back interfaces meet. It is also clear from the graph that the time necessary to flood the microchannel predicted by the diffusion model matches closely the experimental time taken to flood the microchannel.

The model is based on simple mass diffusion and does not account for the diffusion of air into the thin film and into the droplets. A totally predictive model for flooding behavior of the microchannel would require simultaneous modeling of role of surface forces and diffusion of non-condensibles into the liquid.

SUMMARY

Experiments were conducted to investigate the gas/vapor entrapment process in closed-end microchannels. In all tests performed, the microchannels were completely filled. The cavity mouth angles were about 90° while the contact angles measured were less than 90°. Hence the condition for gas entrapment given by Eq. (2) was not met. As a result, all the microchannels were flooded given sufficient time.

The presence of two or more interfaces connected by a thin liquid film was observed for $\phi \leq 40^\circ$. For $\phi > 40^\circ$, only one interface was observed. The dimensions of the microchannels only seem to affect the flooding time, not the physical mechanisms that cause it. One or more interfaces were present independent of the microchannel depth or mouth width.
One dimensional Mass diffusion model shows trends similar to those observed in the experiment with respect to penetration of the interface, but generally over predicts the penetration depth and the flooding time.

**Figure 13** Comparison of experimental data with mass diffusion model.

### ACKNOWLEDGMENTS

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