ABSTRACT

Due to recent growth in miniaturized electronic devices, a demand to dissipate the large amount of heat over a small area has increased. Two phase cooling technique is an efficient heat management technique that involves phase change, and a large amount of heat is absorbed by the fluid during this phase change. Various surface modification techniques have also been investigated to further help to improve this performance. This paper focuses on enhancing the pool boiling performance by creating the micro porous structures on plain copper chip using an electrochemical deposition process involving salt solution as an electrolyte. Constant direct current was supplied for the deposition and optimum current density of 70 mA/cm² was supplied based on the performance. Change of anions with using the same cation for the deposition process, along with the combination of carbon, resulted in dramatic improvement in heat transfer performance during pool boiling of water at atmospheric pressure. We demonstrate a Critical Heat Flux (CHF) enhancement of ~68% with CaCl₂ and ~32% with CaSO₄ deposition as compared to a plain copper chip. Similarly, the Heat Transfer Coefficient (HTC) performance was increased by ~72% and ~29% respectively. A Scanning electron microscope (SEM) and Fourier Transform Infrared (FTIR) spectroscopy was employed to understand the surface morphology and chemical composition of the substrates that indicate that the selective anions from the salts used in the study in combined with graphene from graphite block which resulted in diverse, hydrophilic structures that improved the heat transfer performance.

INTRODUCTION

Innovations in miniaturized electronic industry have resulted in multifunctional compact devices with a growing challenge of fire hazards arising from overheating of these devices. There is a growing need of novel systems capable of dissipating the heat in these devices. Traditionally used single-phase air cooling systems are limited by their larger moving parts and single phase cooling systems require larger areas [1]

Two phase cooling technique involves heat transfer through phase change in a fluid and offers more capacity to absorb the heat by reducing the wall superheat i.e. temperature of the surface. [2] In order to improve the pool boiling performance further, several surface enhancement strategies have been investigated, such as micro porous and nanoporous coatings to produce additional nucleation sites, micromachining of the surfaces, nanotubes and also the use of nanofluids. [3,4] The main objective of all these enhanced surfaces is to decrease the wall superheat temperature and increase the critical heat flux (CHF) and heat transfer coefficient (HTC).

LITERATURE REVIEW

Improvement in pool-boiling performance by employing micro porous surfaces is widely implemented technique for enhancing the performance [3]. Porous structure increases the surface area and the number of nucleation sites which helps to keep the surface temperature low. Li et al. [5] reported pool boiling heat transfer of de-ionized water on sintered copper mesh layer with 56 µm wire diameter. CHF enhancement of 350% was reported over the polished plain surface. Their results indicated that that the boiling heat transfer coefficient depended on the extended surface area and was independent of the thickness of the surface, however, the CHF was substantially influenced by the surface thickness increases proportionally with increase in the wick thickness. Mori and Okuyama [6] showed an increased CHF by the attachment of a honeycomb structured porous plate with different thickness (1.2 mm, 5 mm, 10 mm) on a heated surface. As the thickness of the honeycomb porous plate on the heated surface decreased, the CHF increases to 250 W/cm².

Webb [7] conducted a series of boiling experiments to study the effect of geometric parameters of porous coatings like particle size, pore size and coating thickness. The results revealed that a maximum HTC was obtained with a porous coating thickness of roughly four to six times the particle diameter. Furthermore, he suggested that pore size has a more significant role compared to porosity in enhancing the boiling heat transfer performance of porous surfaces.

Cavities on the porous network play an important role by acting as nucleation sites. As the bubbles depart, the smaller nucleation sites underneath the departing bubbles become active. The rapid evolution of bubbles creates a large vapor column and turbulent convective flow, enhancing the heat transfer rate [8].

The surface wettability is another key factor affecting CHF surface that further enhances phase change heat transfer [9]. The wettability of a surface can be tuned by changing its surface morphology. Wang et al. studied the microstructure characterization in which they obtained 55% porosity copper sample made from 5 mm copper powders [10]. Also, the binder plays an important role in conserving the shape of pores and the structure of the porous metals, especially during the
They studied the aluminum plate, the auxiliary cartridge type 96 W/cm²’s results on 10 mm area of the test section was exposed to the boiling and a heater. The test chip was placed in the ceramic chip boiling test. Test setup included a water bath, a test chip, and a heater. Grafoil sheet was placed above the heater to hold the middle garolite plate and the top aluminum plate. In the top aluminum plate, the auxiliary cartridge type heater (60-VDC, 200W) circular in cross section was fitted along with a small circular hole to insert the saturation thermocouple probe.

Four cartridge type heaters 120-VDC, 200W capacity, were inserted into a copper heater block and was placed on the ceramic block below the test chip. The heater block fits exactly into the groove on the bottom of ceramic chip holder. Grafoil sheet was placed above the heater to minimize the air gap between chip and the heater. Aluminum block is placed below the ceramic block, which is supported by four compression springs to establish the correct contact between the heater block and the test chip. This also ensured only 1-D steady state conduction from heat source to the test chip.

In this study, the salts were deposited and distilled water was used for pool boiling test. Objective of this work was to demonstrate an improved HTC and CHF by a) creating porous structure by depositing salts and, b) varying the anions of the salt to improve the performance. Since the electrodeposition process is less time consuming, and the porosity of the surface can be controlled by controlling current density and time of deposition, it was used to create all the surfaces. The coatings deposited on the surface are combined with carbon and hence are stable composites of carbon and salts. SEM and FTIR results confirm the formation of composites. [3]

EXPERIMENTAL PROCEDURE

Experimental setup as explained earlier was used for all pool boiling tests [15]. Before starting the experiment, it was confirmed that there is no leakage in the setup. A DC power source was used to supply power to the main heater. All the temperatures of chip and fluid were measured using K type thermocouples. Initially, the voltage of DC power source was increased rapidly to achieve saturation of the fluid. Once the saturation was achieved, voltage was increased by 5 volts and data was recorded once the temperature attained a steady state and temperature fluctuation was ±0.1°C for 10 minutes. [15]

POOL BOILING SETUP

The schematic as shown in fig.1 was used for the pool boiling test. Test setup included a water bath, a test chip, and a heater. The test chip was placed in the ceramic chip holder with slots to insert the thermocouples. Only 10 mm x 10 mm area of the test section was exposed to the boiling and remaining area was covered with Kapton tape which acts as an insulation. Above the test chip, a quartz glass water bath rectangular in cross section, with dimensions 14 mm x 14 mm x 38 mm was placed. To seal the contacting surfaces at the bottom and the top, a rubber gasket was used. [15]

Two stainless steel socket head cap screws were used to hold the middle garolite plate and the top aluminum plate. In the top aluminum plate, the auxiliary cartridge type heater (60-VDC, 200W) circular in cross section was fitted along with a small circular hole to insert the saturation thermocouple probe.

For the development of electrodeposition technique, the chips were cleaned with Iso Propyl Alcohol (IPA) and dried under the pressurized air. Only desired 10 mm X 10 mm area was exposed and the other area was insulated using Kapton tape. This step was important to prevent the electrodeposition happening in other locations apart from the desired area. [16]
For the electrodeposition process, two separate electrolyte solutions were prepared. For the first study of calcium sulphate electrodeposition, 10% w/v anhydride CaSO₄ powder was mixed with 4 mL distilled water and this solute was added to 40 mL distilled water. Once the electrolyte bath was prepared, test copper chip i.e. cathode was connected to cathode section while the graphite block was used as an anode were inserted in the bath. For the deposition of calcium sulphate on the plain copper chip, constant current method called as chrono potentiometric method was used. Constant current density of 70 mA/cm² was supplied for 45 minutes. After the deposition, chips were removed from the bath and were allowed to dry in atmospheric air in a closed container. [12]

For the electrodeposition of CaCl₂, similar electrolytic solution was prepared with 10% w/v calcium chloride powder mixed with 4 mL distilled water and then the solute was added to 40 mL distilled water. Same anode i.e. graphite block was used for the deposition process and the cathode was plain copper chip. Chips were tested using the pool boiling setup as mentioned earlier.

![Copper chip Test section](image)

As shown in Figure 3, the distance between the two successive holes on a rectangular base is 3 mm (Δx) while the distance between the hole near the chip surface and the top of the chip is 1.5 mm. (x₁). To read the three temperatures, T₁, T₂, T₃, thermocouples were inserted into the holes.

Heat flux is calculated using steady state 1D conduction equation

\[ q'' = -k_{Cu} \frac{dT}{dx} \quad (1) \]

Where, the temperature gradient \( \frac{dT}{dx} \) was calculated using the three point backward Taylor’s series approximation

\[ \frac{dT}{dx} = \frac{3T₁ - 4T₂ + T₃}{2\Delta x} \quad (2) \]

The boiling surface temperature was obtained by using eq. (2) and is given by

\[ T_{wall} = T₃ - q'' \left( \frac{x₁}{k_{Cu}} \right) \quad (3) \]

**UNCERTAINTY ANALYSIS**

Uncertainty analysis was performed same as Kandlikar and Patil [16]. Each of the thermocouple was calibrated in the temperature range higher than the testing temperatures. Based on the factors such as distance between thermocouples and the spacing between them, thermal conductivity of copper, and calibration data of thermocouples, total uncertainty in heat flux and heat transfer coefficient was calculated. The error analysis was obtained by the method of partial sums, which showed that the maximum uncertainty was from thermocouple value measurements. At higher heat fluxes, a maximum uncertainty of 8% was obtained for the test surfaces.

**SURFACE CHARACTERIZATION**

**Scanning Electron Microscope (SEM)**

The morphology of the substrates was studied by JSM-6400V scanning electron microscope (SEM), JEOL, Ltd., Tokyo, Japan at an accelerating voltage of 15 kV. The energy dispersive X-ray spectroscopy (EDS) measurements were done on Bruker Quantax EDS with XFLASH 5010 detector attached to a field emission scanning electron microscope MIRA II LMH. [18]
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From Fig.4, SEM images show the structure which look like the clouds. Images a and b are at the magnification of 2882x, image a shows the top view while image b shows the Energy Dispersive Spectroscopy mapping of image a. EDS shows the spatial distribution of elements in a sample. As seen from image b, the distribution of carbon, calcium and chlorine is observed which shows the formation of cloudy structure. Pores size of 2-4 µm are present on the surface. Image b shows the distribution at higher magnification of 3.6kX at 70° tilt angle. The tilted images enable to detect the thickness of the coating. The image c is at the scale of 10 µm. The height of the coating is observed to be 4-5 µm. Finally, image d shows the EDS mapping of image c, i.e. the distribution of carbon, chlorine and calcium.

Image a) in Fig.5 shows the top view of the coating at 565 X while image b) shows the 70° tilt at 3.6 kX. All the other images from c to f, show the EDS mapping of image b. The coating of CaSO₄ has formed marble type structure as seen on Fig 5b. From image b, the diameter of these marbles is observed to be ~5µm. All the other images show the distribution of carbon, calcium, oxygen and Sulphur.

**Fourier Transform Infrared (FTIR)**

Fourier Transform Infrared (FTIR) spectroscopy (Shimadzu IR Prestige 21) was used to confirm the presence of copper and graphene oxide. Absorption peaks are observed at different energy levels indicating the bonds as characteristics of molecules. Absorption of IR by molecules is plotted against the wavelength. The different peaks signify the different bonds. [17]

FTIR of both electrodeposited chips is shown in Fig.5a and Fig.5b. The different peaks signify different bonds. FTIR analyzes the samples at molecular level and the peaks signify the bonds that are characteristics of molecules. For chip 2, as shown in Fig.5, a sharp peak at 1150 cm⁻¹ shows the C-O stretch. The peak at 1500 cm⁻¹ signifies the C=C aromatic stretch. Both these peaks are due to the counter electrode which is graphite block. The fusion of calcium and sulfate ions is seen at 1630cm⁻¹. However, the fusion of calcium with carbon is not observed from the FTIR.

For chip 3, as shown in Fig.6, again peaks at 1150 and 1500 cm⁻¹ signify the C-O and C=C bonds. Since the same counter electrode was used for electrodeposition of both chips 2 and 3, the peaks are similar. But, for chip 3, carbon and calcium form carbon halide bond, which is seen from the peaks between 600-800 cm⁻¹.

**Contact Angle**

Static, advancing and receding contact angles were measured for the surfaces. Contact angle measurements were done using VCA Optima. For measuring the advancing contact angle, syringe was used to drop a certain volume of water (4 µL) on the surface. The angle was measured whenever the size of the droplet was increased and the droplet of water slide outward on surface compared to the previous position. The maximum value of the contact angle was recorded as the advancing contact angle.

Similarly, for measuring receding contact angle, water from the chip surface was drawn into the syringe, and the point at which the water droplet changed its shape from convex...
toconcave shape, contact angle was measured. This was recorded as the receding contact angle. The difference between advancing and receding contact angle is called as the hysteresis of contact angle.

Table 1: Contact angle data for Plain copper chip (chip 1), and two electrochemically deposited chips (chip 2 – CaSO₄, and chip 3 – CaCl₂)

<table>
<thead>
<tr>
<th>Test chip</th>
<th>Contact Angle</th>
<th>Static</th>
<th>Advancing</th>
<th>Receding</th>
<th>Hysteresis</th>
</tr>
</thead>
<tbody>
<tr>
<td>Plain Cu</td>
<td></td>
<td>82.5</td>
<td>89.5</td>
<td>24.6</td>
<td>64.9</td>
</tr>
<tr>
<td>Chip 1</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>CaSO₄</td>
<td></td>
<td>51.25</td>
<td>56</td>
<td>19</td>
<td>37</td>
</tr>
<tr>
<td>Chip 2</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>CaCl₂</td>
<td></td>
<td>29</td>
<td>36</td>
<td>12.85</td>
<td>23.15</td>
</tr>
<tr>
<td>Chip 3</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

RESULTS AND DISCUSSIONS

All the characterization was done before the pool boiling tests. From the contact angles, it is seen that the chip with CaCl₂ was more hydrophilic as compared to CaSO₄ chip. From it is observed that for CaCl₂ electrodeposited chip, the carbon, calcium and chlorine deposition is present on the chip. This is explained in details in surface characterization section.

All the pool boiling tests were conducted under atmospheric pressure and the distilled water was used for all the tests. Fig.6 shows a comparison of pool boiling curve with plain copper chip. It is observed that both electrodeposited chips performed better than plain copper chip. A maximum CHF of 205 W/cm² with a wall superheat of 22.7°C was achieved by chip 2. This CHF corresponds to ~ 68% improvement in CHF when compared to plain copper chip.

Fig.7 shows the HTC as a function of heat flux. It is seen that HTC increases as the heat flux goes on increasing. Maximum of 91.1 kW/m²°C heat transfer coefficient was achieved for chip 3.

Fig.8 shows the repetitive testing of the chip 3. All the tests were performed at interval of 24 hours. It is observed that in all the three tests on the same chip, heat transfer performance is consistent. This shows that the coating of the chip is stable and performance somewhat remains constant after repetitive pool boiling test.

DISCUSSIONS

During the electrodeposition, when current is supplied across the electrodes, current flows from anode to cathode. Anode loses its electrons and cathode gains electrons and material gets deposited at cathode. Apart from this, an
electrolyte solution also plays important role in electrodeposition process. It forms cations and anions and get deposited at the cathode. [18] For the electrodeposition of CaSO4 and CaCl2, reactions at anode and cathode are:

At cathode: \(2 \text{H}_2\text{O} + 2e^- = \text{H}_2 + 2 \text{OH}^-\)

At anode: Oxidation of graphite (release carbon atoms)

Electrolyte: \(\text{CaSO}_4 = \text{Ca}^{2+} + \text{SO}_4^{2-}\)
\[
\text{Ca}^{2+} + 2e^- = 2\text{Ca} \\
\text{Cl}_2 = \text{Cl}^{2+} + 2e^- \quad [19]
\]

Hence, the calcium ions from electrolytic bath along with carbon from graphite block i.e. anode gets deposited on cathode. The cloud like structure from the SEM as shown in Fig.10 confirms the presence of carbon and calcium on the test chip 3. Also, FTIR confirms the carbon halide i.e. C-Cl bond as explained in the FTIR section above. Due to this combination, CHF is enhanced by marginally as compared to plain copper chip.

Since in the calcium sulphate deposition, anions did not get deposited on the test chip, there is still a scope for the calcium derivative whose anions can get deposited. Hence, an inorganic salt containing both cations and anions that can be used in deposition were used. Calcium chloride is the most common inorganic salt was used for the deposition. When constant current is supplied, calcium chloride forms cations and anions which get deposited on the cathode along with carbon from graphite block, forms a combined C-Cl structure.

It can be seen from Table 1 that CaCl2 deposited chip is more hydrophilic as compared to CaSO4 deposited chip. Hydrophilic surfaces help to keep the cavities filled with liquid all the time and hence there is a continuous supply of fluid for the evaporation. This helps in preventing the dry out and keeps the surface temperature to minimum value. [20]

Hence, the combination of hydrophilic surface and CaCl2 deposition along with carbon halogen bond improved the heat transfer performance drastically and wall superheat temperature was observed to be comparatively lower. CHF enhancement of ~68% was achieved as compared to plain copper chip.

**CONCLUSION**

Electrodeposition of micro porous structures by depositing salts and by varying selective anions of inorganic compounds was successfully tested for pool boiling performance. The novelty of this work lies in implementing different anions while keeping the cation same to enhance the performance. The combination of selective anion and carbon formed a carbon-chloride based structure which improved the morphology of the substrate and improved the bonding.

**NOMENCLATURE**

- \(T_i\): temperature at the top hole of chip, °C
- \(T_m\): temperature at the middle hole of chip, °C
- \(T_b\): temperature at the bottom hole of chip, °C
- \(\alpha\): temperature gradient
- \(q^*\): heat flux, W/m²
- \(h\): heat transfer coefficient, W/m²°C
- \(k_c\): thermal conductivity of copper, W/m°C
- \(T_{wall}\): wall temperature, °C

**ABBREVIATIONS**

- CHF: Critical Heat Flux
- HTC: Heat Transfer Coefficient

**REFERENCES**


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