



Effect of a graphene oxide coating layer on critical heat flux enhancement under pool boiling



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ARTICLE INFO

Article history:

Received 6 December 2013

Received in revised form 5 June 2014

Accepted 5 June 2014

Available online 3 July 2014

Keywords:

Critical heat flux

Pool boiling

Graphene oxide

ABSTRACT

We report an investigation of the boiling heat transfer characteristics of graphene-based materials. Pool boiling experiments were carried out to investigate the critical heat flux (CHF) enhancement in a graphene oxide (GO) colloidal suspension with various concentrations as the working fluid with a Nichrome wire heat source. We found that nucleate boiling resulted in deposition of the GO colloids onto the heated wire, whereby the GO flakes formed a smooth laminated film, and that the thickness of this layer was approximately proportional to the observed increase in the CHF. The surface wettability could not explain the enhancement in the CHF. Instead, we focused on thermal activity analysis based on heat dissipation effects of the GO layer with extraordinary thermal properties. The large thermal conductivity of the GO layer inhibited the formation of local hot spots, thus preventing the formation of dryout regions and delaying the CHF. Furthermore, a thermal reduction of the GO colloids led to further increase of its thermal conductivity, also thermal activity, and hence additional enhancement of the CHF.

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1. Introduction

Nucleate boiling is one of most efficient methods of heat transfer because of the latent heat associated with the phase transition of the working fluid. It is possible to transfer a large heat flux with less wall superheat using nucleate boiling than with a single-phase heat transfer process or other boiling regime heat-transfer methods. For this reason, nucleate boiling has been employed in many applications, including power plants and cooling systems for electronic chips.

The critical heat flux (CHF) is the operating limit of the nucleate boiling regime, whereby a transition from nucleate boiling to film boiling occurs. When CHF phenomena occur, a vapor film covers the surface so that the liquid supply for cooling is inhibited [1–3]. This leads to an increased thermal resistance, and causes the surface temperature to increase rapidly, which may result in failure of a system; for example, a severe accident in a power plant.

For this reason, an understanding of CHF phenomena is important, and can lead to improved efficiency and safety margins. There have been a number of reports of predictions of CHF phenomena [4–11] and methods of increasing the CHF (i.e., extend the nucleate boiling regime) [12–15].

Nanofluids are one approach to increasing the CHF. When a nanofluid is used as the working fluid in pool boiling applications, the nanoparticles build up and form a porous medium on the heating surface, which alters the surface wettability, resulting in enhancement of the CHF [12]. Pool boiling experiments using nanoparticle-coated surfaces and distilled water as the working fluid have shown that the nanoparticles induced a surface morphology, which enhanced the CHF [13]. The modified surface showed enhanced wettability and capillary-wicking characteristics, whereby the water supply to the deposited layer increased the CHF by preventing the growth of localized dry patches [14]. The effects of wettability and capillary wicking on CHF phenomena have also been investigated using artificially modulated micro- or nano-structured surfaces [15].

Graphene has received a lot of interest due to its highest thermal conductivity [16] and favorable mechanical properties [17]. It is a two-dimensional (2D) carbon structure and may be coated onto surfaces, and has recently been demonstrated to be a heat-spreader [18]. Moreover, there has been considerable progress in

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developing chemical processes for preparation of graphene as a colloidal suspension in water [19–21]. Via a chemical process employing hydrazine, graphene oxide can be reduced to form reduced graphene oxide (RGO), which has properties similar to those of graphene. Films of RGO synthesized by oxidation and reduction processes from natural graphite have shown a thermal conductivity of 110–1100 kW/m/K at room temperature, for thicknesses of 1–100 nm [22]. This indicates that RGO films may have applications in heat-transfer systems.

Studies of boiling heat transfer using colloidal suspensions of graphene oxide (GO) and RGO colloids have resulted in an increase in the CHF of 200% and 100%, respectively, compared with distilled water as the working fluid [23]. An important point here is that the increase in the CHF could not be explained solely by the wetting characteristics of the GO/RGO layers, which showed large contact angles and limited capillary-wicking performance. Instead of wettability analysis, a phenomenological analysis based on the Rayleigh–Taylor (RT) wavelength [5] was attempted with a morphological observation of RGO-coated wires following CHF experiments [24].

RGO flakes form unique three-dimensional (3D) structures on the heated surface—a so-called self-assembled 3D interconnected foam-like graphene network (SFG) and a base graphene multilayer (BGL), which formed due to the bubble dynamics of the nucleate boiling regime [25,26]. The 3D structure of the RGO flakes allows it to act as a heat spreader and a liquid-saturated porous medium in the nucleate boiling regime, leading to a higher boiling heat transfer coefficient and an increased CHF, while eliminating the rapid temperature jump at the CHF [27].

In this study, we investigated the effects of a surface coating using GO flakes on the CHF during pool boiling. We describe experiments using a wire heater in a saturated GO colloidal suspension under atmospheric conditions. To modify the surface characteristics, various concentrations of the colloids were used and the effects on the CHF were investigated. Furthermore, the surface morphology and wettability of the modified surface were characterized. We explain the mechanism for the enhancement of the CHF in terms of heat dissipation within the deposited GO layer, which can prevent the growth of dry spots due to the high thermal conductivity.

2. Experimental

2.1. Preparation of graphene oxide colloids

GO colloids were synthesized using chemical oxidation of graphite and sonication procedures. Graphite oxide powder was prepared via the modified Hummer's method [19,20]. Two grams of graphite powder (<45- μm particle size, Aldrich) and 1 g of sodium nitrate (Aldrich) were mixed using a stirrer bar in 50 ml of sulfuric acid (ACS 95–98%, Alfa Aesar) at 2–3 °C. Six grams of potassium permanganate were added slowly to oxidize the mixture, while maintaining the temperature below 10 °C. After 30 min, the solution was heated to 35 °C for 2 h and mixed vigorously. Distilled water (100 ml) was then added slowly, and the mixture was heated to 98 °C. Following this, the color changed from dark black to yellow–brown due to the oxidation. The mixture was then cooled and diluted with 300 ml of distilled water at 40 °C to prevent precipitation of residual salts and metallic acids. Hydrogen peroxide (3%; 40 ml; 36 ml of distilled water and 4 ml of hydrogen peroxide (30 wt.%, Aldrich)) was added to neutralize the potassium permanganate oxidizer. The solution was vacuum-filtered twice using a 0.45- μm cellulose membrane filter, leaving the GO. The GO was then freeze-dried for 4–5 days.

The GO powder was dispersed in distilled water by ultrasonication for 1 h (Fisher Scientific Sonic Dismembrator, Model 500,

400 W, 20 kHz, 30% amplitude). The synthesized powder (0.25 g) was suspended in 500 ml of distilled water to create a 0.5 mg/ml GO colloidal suspension. The solution was transparent brown, as shown in Fig. 1-(a). Fig. 1-(b) shows transmission electron microscope (TEM) images of the GO flakes, revealing a 2D-sheet-like structure. Fig. 1-(c) shows selected area electron diffraction (SAED) data at the center of the flake shown inset in Fig. 1-(b). The diffraction pattern includes an amorphous ring, which is attributed to the folding of sheets and defects due to oxidation. The solution was diluted in distilled water into 0.0001, 0.0005, 0.0010, and 0.0050 wt.% samples for use as the working fluid in pool boiling experiments.

2.2. Pool boiling experiment

The pool boiling apparatus consisted of a main pool containing the working fluid, the wire heater, and a data acquisition system, as shown in the schematic diagram in Fig. 2. The main pool was a 200 × 100 × 200-mm Pyrex glass bath containing ~3 L of the working fluid. The bath was placed on a hot plate (DAIHAN, HP-30D, 2 kW) for heating and maintaining the working fluid at the saturated condition. The bath was sealed using a Teflon cover, whereby a reflux condenser was installed to prevent loss of the working fluid due to evaporation, and to maintain atmospheric pressure. The Nichrome wire heater, which was 0.2 mm in diameter and 79.2 mm in length, was submerged in the working fluid and arranged horizontally by suspending it under tension between the two cylindrical 40-mm-diameter stainless steel electrodes. All of the wires used were cleaned to remove any residual dust or contamination on the surface. The electrodes were mounted on the Teflon cover and connected to a direct current (DC) power supply (Sorensen, 60 V/500 A). A data acquisition system (Agilent, 34970A) was employed to measure the voltage applied to the wire heater, and a 1- Ω resistor ($\pm 0.001 \Omega$) in series with the heater was used to measure the current as a reference resistance. This resistor was cooled and maintained at 10 °C using a constant temperature bath (Jeiotech, RW-3040G).

The working fluids were preheated on the hot plate for 2 h for degassing prior to the experiments, and were maintained in the saturated condition under atmospheric pressure during the experiments. The heat flux applied to the wire continuously increased with a rate of 50 kW/m² per min until the CHF was reached. When the CHF was reached, the temperature of the wire increased rapidly, causing it to melt. The CHF was calculated as follows:

$$q''_{CHF} = \frac{V_{max} I_{max}}{\pi DL}, \quad (1)$$

V_{max} and I_{max} are the voltage and current immediately prior to the wire melting, and D and L are the diameter and length of the wire. The experimental uncertainty was determined as follows [29]:

$$\frac{U_{q''_{CHF}}}{q''_{CHF}} = \sqrt{\left(\frac{U_{V_{max}}}{V_{max}}\right)^2 + \left(\frac{U_{I_{max}}}{I_{max}}\right)^2 + \left(\frac{U_D}{D}\right)^2 + \left(\frac{U_L}{L}\right)^2}, \quad (2)$$

where U is the maximum uncertainty. The main sources of uncertainty were the applied voltage and length of the wire. In addition, the contact resistance between the wire and the stainless steel electrodes was not taken into consideration. The uncertainty in the applied voltage was $\pm 4.0\%$ and the uncertainty in the length of wire was $\pm 1.7\%$. Based on Eq. (2), the maximum uncertainty in the CHF was $\pm 4.4\%$.

2.3. Water level during pool boiling

We found that the distance between the heated surface and the free surface of the working fluid during pool boiling was strongly

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