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Experimental study of water droplets on over-heated nano/microstructured zirconium surfaces

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HIGHLIGHTS

- Heat transfer performance of a droplet on a modified zirconium surface is evaluated.
- Modified (nano/micro-) surfaces enhanced heat transfer rate and Leidenfrost point.
- A highly wettable condition of the modified surface contributes the enhancement.
- Nano-scaled modification indicates the higher performance of droplet cooling.
- Investigation via visualization of the droplet support the heat transfer experimental data.

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ABSTRACT

In this study, we observed the behavior of water droplets near the Leidenfrost point (LFP) on zirconium alloy surfaces with anodizing treatment and investigated the droplet cooling performance. The anodized zirconium surface, which consists of bundles of nanotubes (~10–100 nm) or micro-mountain-like structures, improved the wetting characteristics of the surface. A deionized water droplet (6 μL) was dropped onto test surfaces heated to temperatures ranging from 250 °C to the LFP. The droplet dynamics were investigated through high-speed visualization, and the cooling performance was discussed in terms of the droplet evaporation time. The modified surface provided vigorous, intensive nucleate boiling in comparison with a clean, bare surface. Additionally, we observed that the structured surface had a delayed LFP due to the high wetting condition induced by strong capillary wicking forces on the structured surface.

1. Introduction

When a liquid droplet is placed on a heating block whose temperature is significantly higher than the boiling point of the liquid, the droplet can hover over the block surface without contact. This is due to the existence of a vapor cushion layer produced by strong vaporization beneath the droplet, otherwise known as the Leidenfrost (LF) phenomenon. At or above the Leidenfrost Point (LFP), thermal energy is transferred from the heated surface to the droplet mainly via conduction and radiation through the vapor layer. This layer acts as thermal insulation, reducing the heat transfer performance compared with the case of liquid–solid contact below the LFP. Therefore, the evaporation time for a droplet at or above the LFP can be much longer than that observed in the lower-temperature nucleate boiling regime. This long evaporation process and reduced heat transfer are undesirable for cooling purposes in many thermal applications. For a cooling system that has a surface temperature below the LFP, a spray-cooling method is appropriate due to the small droplet formation that occurs with vigorous nucleate boiling (Duffey and Porthouse, 1973). For a cooling system that has overheated components (above the LFP) in a high-power-density thermal system, such as nuclear power plants during accidents, a higher LFP is desirable for rapid cooling of the system.

Over the last several decades, the LFP mechanism has been investigated by analyzing film boiling and observing droplet behavior on overheated surfaces. Because the LFP is associated with the onset of film boiling, the LFP has been studied by examining the vapor layer that forms during the film boiling process. The LFP has also been studied based on Rayleigh–Taylor instability analysis of film boiling (Zuber, 1958) because film boiling is triggered by a stable liquid–vapor interface due to sufficient heat flux and phase change. Numerous film boiling studies related to the LFP

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have been performed using various experimental and numerical approaches (Kunugi, 2012; Kim et al., 2003; Berenson, 1961; Hoseler and Westwater, 1962; Yao and Cai, 1988). Gottfried et al. (Gottfride et al., 1996; Gottfried and Bell, 1966) proposed an analytical solution for a hovering droplet, which involved solving the heat and mass transfer relationships for a spherical droplet on a heated surface. Recently, Biance et al. (2003) derived the state of a gradually evaporating droplet on a surface; their analytical results were supported by droplet LFP experiments. A hovering droplet experiences heat transfer and a phase change via conduction through the vapor layer; an evaporated vapor layer provides supporting pressure to the droplet (Chung and Lee, 1982).

Recently, the LFP of a droplet with initial inertia (ρvD) on a heated surface has been investigated (Bernardin and Mudawar, 2002). As the droplet approaches and contacts the heating surface, the subcooled droplet experiences nucleate boiling or rebound from the surface. Studies on the droplet transient processes have revealed the mechanism behind the spray-cooling condition, which involves transient phenomena from impact to the point of nucleate boiling or rebounding. Bernardin et al. (Bernardin and Mudawar, 1999, 2002) reported that rapid nucleate boiling after droplet impact on a heated surface contributes to droplet rebounding and continuous vapor-layer formation. Kandlikar and Steinke (2002) focused on the effect of strong evaporation at the droplet edge on droplet rebounding. The intersection between the edge of the droplet and the surface is composed of three phases (liquid–vapor–solid), and is referred to as the triple line. The fast evaporation of the triple line under overheated surface conditions brings about a recoiling force on the droplet, which pushes the droplet up; this rebounding behavior is called “cutback”. Recently, Kim et al. (2011) focused on the liquid–solid (L–S) contact of the cutback phenomenon; they observed that the L–S ‘liquid filament’ contacts induced nucleate boiling that destabilized the droplet behavior via the Kelvin–Helmholtz instability.

Based on the previous research efforts described above, many researchers have studied various methods to enhance the LFP. Takata et al. (2005) reported that the hydrophilic condition of a surface provides a higher LFP than the hydrophobic condition. Kandlikar and Steinke (2002) also focused on the effect of wettability on droplet rebounding. The triple line behavior of the droplet on the heated surface is affected by the wetting characteristics, and the hydrophilic condition makes droplet rebounding more difficult due to the enlarged surface tension acting on the droplet. Recently, Kim et al. (2011) evaluated the effect of porosity and roughness on the LFP. Many cavities on a surface enhance nucleate boiling during droplet bounding and hovering, which, in turn, delays the LFP due to unstable droplet dynamics. Avedisian and Koplik (1987) reported that a porous layer on the heated surface can affect the vapor flow via evaporation at the liquid–vapor interface; the porous layer absorbs the vapor flow and decreases the vapor pressure supporting the droplet. Thus, in this case, a higher LFP is required to compensate for the vapor pressure loss. Fatehi and Kaviany (1990) also described the vapor flow into a porous layer during droplet hovering after LFP.

In this study, we prepared a nano- and micro-scaled-structured zirconium surface to investigate the LFP mechanism by observing droplet behavior via a high-speed visualization technique in an attempt to enhance rapid quenching for overheated surfaces (e.g., nuclear power plant accidents). For this reason, zirconium alloy, a nuclear fuel cladding material, was used as the base material for the test surface. The modified test surface has already been evaluated in terms of nucleate boiling heat transfer and critical heat flux (CHF); significant enhancement of the CHF has been observed due to the superhydrophilic surface characteristics (Ahn et al., 2010, 2011; Ahn and Kim, 2011; Kim et al., 2006). The actual condition (prototypicality) of the nuclear cladding material is rough, with a porous oxide layer as compared with a bare zirconium surface. According to the literature (Byers and Deshon, 2004; Cinosi et al., 2011; Leistikow and Schanz, 1987) on the zirconium surface structure with respect to the surface morphology, an oxide layer, a few microns to several hundred microns thick, forms during normal or abnormal operation. The modified test surface used in this research closely resembled the rough, porous features of actual nuclear cladding. Recently, Buongiorno (2014) reported that corrosion and crud affect the safety margins of nuclear reactors via thermal–hydraulic effects.

In this study, we focused on the droplet cooling performance and the LFP of the modified zirconium surface as an extension of the previous phase-change heat transfer research. The results from this study may provide valuable information for future nuclear reactor design and the safety of these systems.

2. Experiments

2.1. Preparation and characterization of test surfaces

In this study, rectangular zirconium alloy plates (20 mm × 25 mm × 0.7 mm) were used as test samples. The plates were mechanically polished with #1200 silicon carbide abrasive to remove impurities and produce a uniform surface. The polished samples were then cleaned with acetone and methanol in an ultrasonic bath. After rinsing in deionized water (DI), the samples were completely dried (Ahn et al., 2010). The samples underwent anodic oxidation with 0.5-wt% hydrofluoric acid solution as the electrolyte. A constant electric potential (20 V) was applied between the anode (test surface) and cathode (carbon bar) using a DC power supply (N5771A; Agilent). The solution temperature was fixed at 10 °C to maintain a constant reaction speed. The exposure times of the oxidation process were 0, 300, and 600 s to obtain zirconium oxide surfaces with different morphologies. After the anodic oxidation process, the treated samples were heated for 6 h at 300 °C in an electric muffle furnace to eliminate fluoride residue.

The characteristics of the modified zirconium were controlled by the exposure time in the solution. To characterize the modified zirconium alloy, we evaluated the static contact angle and imaged the modified surface using scanning electron microscopy (SEM). The contact angles of the prepared test surface were measured by dosing with 5-μL DI water for a few seconds. The contact angle decreased from 49.3° for the bare surface (at time $t = 0$ s) to 0° for the modified zirconium alloy surface. When the anodic oxidation time was 300 s, nanostructures, specifically nanotubes (diameter: ∼10–100 μm) were observed by SEM (Fig. 1); the contact angle was less than 10°, and in some cases, even 0°. After 600 s, however, the nanoscale structures had almost completely disappeared, and microstructures resembling a valley–mountain structure were observed. The contact angle indicated complete surface wetting (for a contact angle of 0°). In previous studies (Ahn et al., 2010, 2011; Ahn and Kim, 2011), we defined this complete wetting as “liquid spreading,” observed on both nano- and microstructured surfaces. The surface roughness of the zirconium alloy was measured by a surface profiler (Alpha-step IQ; ZKLA Tencor). The average roughness (Ra) of the bare zirconium alloy was 0.15 μm, and the peak Ra of the modified zirconium alloy (at 600 s for the microstructure) was 0.32 μm.

2.2. Experimental setup of droplet impingement

Fig. 2 shows a detailed schematic diagram of the experimental setup of the droplet impingement test. The setup consisted of an assembly of rectangular zirconium alloy test samples and an