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Layer-by-layer carbon nanotube coatings for enhanced pool boiling heat transfer on metal surfaces

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ABSTRACT

Micro/nanotextured coatings have enabled the manipulation of thermal characteristics in pool boiling heat transfer such as the heat transfer coefficient (HTC) and critical heat flux (CHF) because of the ability to optimize bubble formations and departures. However, fabricating such coatings on substrates involves high cost-bulky setup, and is limited by the materials and adhesion properties. Herein, we report layer-by-layer (LbL)-assembled polyethylenimine (PEI)-multi-walled carbon nanotube (MWCNTs) coatings on stainless steel (SS316) to enhance HTC and CHF in pool boiling heat transfer. LbL-assembled PEI-MWCNTs coatings (10, 20, 40 bi-layers) on SS316 were fabricated, whereas bare SS316 substrate and vacuum-filtered MWCNTs coating were prepared as controls. Because of the nano-cavities and the inner-nanoporous structures, LbL coatings showed significant enhancement of HTCs and CHFs compared to bare SS316. Furthermore, denser networks of MWCNTs due to electrostatic bonding and hydrophilic nature of PEI in the LbL coatings could enhance the HTCs and CHFs compared to vacuum-filtered MWCNTs coating. LbL-assembled PEI-MWCNTs of 20 bi-layers showed the highest improvement in HTC, whereas the 40 bi-layers coatings achieved the best enhancement ratio of CHF, (~147%). Further development of the LbL-assembled coatings on metal surfaces would enable potential applications for thermal management, from micro/nanoscale platforms to macroscale systems.

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1. Introduction

The development of micro/nanoscale platforms has revolutionized both existing products and new applications such as central processing units, memory devices, circuit systems, micro fuel cells, and batteries. Meanwhile, the rising temperatures due to restricted structures in the micro/nanoscale platforms has emerged as a big obstacle for further enhancement of the overall performance and the operating stability. Furthermore, even in macroscale systems including power plants, thermal energy management is quite critical to improve the efficiency and to maintain the thermal safety margin. For example, the failure of thermal energy dissipation in nuclear power plants may lead to a damage of the fuel rod. Liquid-vapor phase change heat transfer is one of the most significant phenomena to be addressed for managing such thermal energy in multiscale systems [1]. The heat transfer coefficient (HTC) of liquid-vapor phase change, which is much higher than that of the single phase, allows efficient cooling performance to dissipate thermal energy or to maintain operating temperatures in small devices and macroscale systems [2]. As well, achieving higher critical heat flux (CHF) extends the thermal operability of the systems.

The overall characteristics of liquid-vapor phase change heat transfer significantly depend on the solid-surface conditions that are directly exposed to the working fluid. In general, porous structures are known to increase the site density of bubble nucleation and enhance the HTC [3]. In addition, the wetting properties of surfaces are regarded as other crucial factors to manipulate the HTC [4]. Furthermore, the surface conditions can affect another

critical factor of phase change heat transfer, viz. CHF, which determines the maximum applicable power density in the thermal systems [5,6]. The CHF is the upper limit of nucleate boiling heat transfer during liquid-vapor phase heat transfer. When the system reaches the CHF, vapor blankets cover the contact surfaces at the interface with working fluids. Such a circumstance results in a dramatic decrease in the HTC because of the low thermal conductivity of the vapor phase on the surface and a decrease in bubble departures from the surfaces [7]. The sudden drop in the thermal energy dissipation causes a decline in the cooling performance, collapsing the stability of the systems.

Coatings of micro-nanostructured materials are ideal candidates for the manipulation of contact surfaces in liquid-vapor phase change heat transfer. Three desirable physical conditions, porosity [8], roughness [9], and wettability [10] should be optimized at the interface with the working fluids. Various micro-nanostructured materials such as silicon, metal, metal oxide, organic, and inorganic materials have been tested as contact interfaces between working fluids and the heat transfer substrates [11]. For example, copper [12], aluminum [13], CuO [14], ZnO [15], and TiO_2 [16] effectively improved HTC or CHF compared to the bare substrates. Moreover, coatings of specific micro/nanomaterials, which were composed of flower-like structures [17] or vertically aligned nanowires [18], allowed controlling the HTC and CHF in identical materials. In particular, carbon nanotubes (CNTs) have been regarded as potential candidate materials for contact interfaces between the working fluid and the heat transfer substrate. CNTs have ideal fiber-like structures for the formation of porous structures with increased surface roughness, while the wettability of CNTs can be modified from hydrophobic to hydrophilic by a facile chemical method [19]. In addition, the superior thermal conductivity of CNTs can contribute to the minimization of the thermal resistance at the contact interfaces between CNTs and the heat transfer substrates [20]. In fact, the use of CNTs at contact interfaces was effective in changing both the HTC and CHF in pool [21,22] and flow boiling heat transfer [23,24].

However, there are fundamental limitations of contact interfaces using micro/nanostructured materials for boiling heat transfer applications. Micro/nanofabrication processes are inevitable for directly developing micro/nanotextured surfaces on the substrates, despite being high-cost and not applicable to large-scale surface areas [25]. Moreover, the fabricated structures are not strong enough to endure the high pressure or harsh operating conditions of liquid-vapor transfer and bubble formation-departure in boiling heat transfer [26]. Another approach is the indirect coating of pre-synthesized micro-nanostructured materials on the bare substrates by simple solution processing methods [27]. In this case, the adhesion with the substrate or among the micro/nanomaterials is not strong because it is based on only weak physical contacts due to surface energy. Moreover, it is quite difficult to form uniform structures and surfaces along the textures of the original heat transfer substrate. In other studies, the difficulty of fabrication and surface transfer, the weak adhesion and non-uniform surfaces formation were major problems causing high cost, long processing times, and a limit on the applicable sizes [28]. In this respect, the development of a new coating method of micro/nanomaterials on the heat transfer substrate is essential to facilitate further application for the enhanced pool boiling heat transfer.

Layer-by-layer (LbL) deposition is a fabrication method for uniform thin film layers on various material substrates [29,30]. The repetitive alternate immersion of substrates into a positively charged and a negatively charged solution forms stable film layers along the original surfaces of the substrates [31–34]. Compared to conventional coating methods, LbL deposition can be used on large areas and requires simple processing equipment because it is based

on solution-processing methods rather than high temperature processing [35]. The manipulation of properties is implemented by adjusting the bi-layer number [36], by controlling the pH of suspensions [37], or by adopting different techniques such as immersive [38–40], spin [41], spray [42], or microfluidic LbL [43]. The LbL methods can be used to fabricate diverse coatings that are highly porous to form large surface area films of micro/nanostructured materials [44]. In particular, the electrostatic force among bi-layers allows formation of more uniform, adhesive, and substantial coating layers than films depending on physical contacts to surfaces.

Herein, we report LbL-assembled polyethylenimine (PEI)-multi-walled carbon nanotubes (MWCNTs) coatings on stainless steel (SS316) metal surface to significantly enhance HTC and CHF in pool boiling heat transfer (Fig. 1). SS316 is one of the most widely used substrates in various thermal systems because of its low-cost and anti-corrosion property, but it has a relatively low HTC and poor CHF to satisfy the requirements for fast-cooling and high power applications. Nowadays, MWCNTs can be mass produced and are ideal nanomaterials for using on surfaces for controlling boiling heat transfer because they have excellent thermal properties [45,46], good mechanical-chemical stability [47], and ideal fiber-like structures for the formation of porous layers [48]. The hydrophilic property of PEI can control wettability by mixing with the hydrophobic MWCNTs. In this work, we fabricated three LbL-assembled PEI-MWCNTs coatings (10, 20, 40 bi-layers) on SS316, and measured the enhancement in HTC and CHF in pool boiling heat transfer. They were compared with the control performances of bare SS316 substrate and MWCNTs-coated SS316 fabricated by vacuum filtration. These two controls provide a basis to evaluate the effectiveness of LbL-based PEI-MWCNTs coatings.

2. Experimental

2.1. Preparation of SS316 substrates and test heater assembly

The bare substrate, SS316, was polished with sand paper of grade #800, for all heat transfer substrates for pool boiling experiments. This procedure retained the microscale wave-like structures on SS316 surfaces. After the polishing step, the bare SS316 substrates were cleaned with deionized (DI) water, acetone, ethanol, and DI water, sequentially [49]. The dimensions of the heating element were $35 \times 10 \times 2$ mm^3 in length, width, and thickness, respectively. Since thin films of the coating materials were deposited onto the SS316 surface, the heated area (35×10 mm^2) for all the test samples was identical to the bare substrate.

2.2. Fabrication of LbL-assembled PEI-MWCNTs films on SS316

The MWCNTs were purchased from JEIO, Korea (95 wt% purity, 5–20 μm length, 20–40 nm outer diameter). In order to prepare the negatively-charged MWCNTs solution for the LbL immersion process, MWCNTs were refluxed with H_2SO_4/HNO_3 (3/1 v/v, 95–98%/70% concentration) at 70 °C for 2 h [44]. They were filtered from the acid solution and washed using DI water several times. The filtered MWCNTs were dried at room temperature for 24 h to remove the solvent. Because of the previous steps, the surfaces of MWCNTs were functionalized with carboxylic group (−COOH), which made the negatively charged surface. The carboxylic-functionalized MWCNTs (0.1 mg/mL) were sonicated for 1 h and uniformly dispersed in DI water. In order to prepare the positively charged layers for the LbL immersion process, polyethyleneimine (PEI, 50% w/v in water) was purchased from Aldrich. PEI solution (1 mg/mL) was diluted in DI water. The prepared SS316 was