

Slip flow, capillary evaporation, and hydrovoltaic power generation in graphene nanochannels

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論 文 内 容 の 要 旨

The discovery of ultrafast water flow in graphene channels holds significant implications for various applications like electronics cooling, thermally driven energy harvesting, and solar-driven desalination. However, existing slip flow measurements on graphene are confined to room temperature, limiting our understanding of temperature dependent behaviors and constraining potential thermal applications. Furthermore, nanostructured and porous materials can generate electricity upon interaction with liquid, a phenomenon termed hydrovoltaic power generation. This process has the potential to significantly enhance water energy harvesting and enable the creation of self-powered devices. Hydrovoltaic power generation has garnered intense research interest due to its ability to directly harvest green energy from liquid flow and evaporation. However, much remains to be explored regarding hydrovoltaic power generation, including the following issues: (1) how to decouple the contributions of moving electrical double layer and capillary evaporation to the hydrovoltaic power generation, (2) how slip flow and capillary evaporation affect hydrovoltaic power generation, (3) how the channel size and temperature affect the hydrovoltaic power generation. Resolving these issues will lay the foundation for advancing hydrovoltaic power generation, enhancing our understanding of electrokinetic phenomena at the nanoscale and paving the way for new sensors and electrical power sources. This thesis utilizes a single graphene nanochannel to simultaneously measure capillary evaporation, slip flow, and hydrovoltaic power generation and clarify the underlying mechanisms. Here's a summary of each chapter:

Chapter 1 provides the general background on slip flow, capillary evaporation and hydrovoltaic power generation in graphene channels, along with a review of previous reports on nanoscale water slip length, capillary evaporation and hydrovoltaic power generation.

Chapter 2 investigates the fabrication process of graphene nanochannels and silicon nanochannels, outlining the steps from electron-beam lithography (EBL), wet-etching methods and wet transfer method for graphene to create the desired structure. The experiment stage includes a visualization system, a voltage test system, a current test system, and a temperature control system which the heater is to control the temperature to adjust capillary evaporation.

Chapter 3 investigates the simultaneous measurement of water slip length and evaporation flux in graphene nanochannels across a range of temperatures spanning from room temperature to 85 °C, with varying channel depths. A capillary flow model combined with capillary evaporation is employed, a factor that cannot be ignored at high temperatures. The measured water slip length ranged from approximately 20 nm to 80 nm, exhibiting a decrease with rising temperature. This temperature-dependent behavior is attributed to the intensified momentum transfer across the solid-liquid interface at higher temperatures. Additionally, the measured water slip length showed a decrease with the channel depth decreasing from 290 nm to 78 nm. The measured evaporation fluxes in the graphene nanochannels ranged approximately from 0.05 mm/s to 0.25 mm/s, which is one to two orders of magnitude lower than the Hertz-Knudsen limit. The

evaporation fluxes showed an increase with both rising temperature and greater channel depths. This temperature and channel depth dependence can be explained by the capillary evaporation theory. These experimental findings enhance our understanding of the temperature-dependent behaviors of slip flow and capillary evaporation in graphene nanochannels, providing valuable insights applicable to various fields such as electronics cooling, thermally driven energy harvesting, and solar-driven desalination.

Chapter 4 have demonstrated the assembly and characteristics of a hydrovoltaic power generation by the graphene single-nanochannel that can generate capillary potential directly from the movement of liquid. A model for hydrovoltaic power generation by slip flow and capillary evaporation in nanochannels is employed. In silicon nanochannel, the capillary potential is induced by the moving EDL and capillary evaporation. Owing to the large resistance of silicon, the voltage is very small. Furthermore, in graphene nanochannel, the capillary potential is also induced by the moving EDL and capillary evaporation on the graphene surface. The voltage of graphene nanochannel is more than that of silicon nanochannel. Through the examination of the correlation between voltage, water meniscus location, water meniscus area, and water meniscus speed, capillary potential can be affected by slip flow. A larger slip length corresponded to a significantly higher fitting voltage. Through theoretical calculations and experiment there is little influence on the induced potential under different depth. As the capillary evaporation increase with temperatures, the open-circuit voltage and short-circuit current increase. In the ethanol experiment, the mechanism is akin to that of water. Like the water, the capillary evaporation can increase the induced potential. At the nanoscale it was verified that the induced potential between polar molecules and graphene is higher than that of non-polar ethanol. Similar to water in the graphene nanochannel, as the capillary evaporation increase with temperatures, the open-circuit voltage and short-circuit current increase. This work lays the foundation for the macroscale hydrovoltaic power generation for studying electrokinetic phenomena, with possible application in the new sensor and new hydrovoltaic power generation sources.

In Chapter 5, the concluding remarks provide a comprehensive overview of the findings from the preceding chapters, highlighting their significance in advancing our understanding of nanoscale fluid properties and power generation. The potential contributions of this study to the field are also discussed. Overall, through the investigation of liquids interacting with graphene/SiO₂/Si at the nanoscale, this study serves as a platform for advancing nanoscale fluid research. Furthermore, recommendations for future research directions are provided to further deepen our understanding of nanoscale fluid behavior and power generation, paving the way for continued progress in this field.

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