Non-trivial capillary hydrodynamics and strong evaporative cooling in multilayer two-dimensional materials: application in thermal management systems of electronic and energy devices

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Abstract: Studies of evaporation from graphene nanostructures with nontrivial capillary hydrodynamics are being carried out. Data were obtained for the rates of evaporation and heat fluxes during cooling of experimental heated cells. Models were built and compared with experimental data. It is shown that the existing models overestimate the evaporation rates and heat fluxes and heat transfer coefficients.

1. Introduction
The problems of thermal stabilization and thermal management of various energy (fuel cells, solar photo panels, solar thermal power engineering, etc.) and electronic (processors, nanotransistors, semiconductor micro- and nanolasers, etc.) cannot be solved (in particular, "thermal wall "of Moore's law) without using more radical cooling methods than are currently used [1-3]. Today it is believed that the most effective cooling method is the liquid-vapor phase transition, when the latent heat of vaporization is used. However, the limiting factor of this process is the rate of fluid supply to the interface [3]. More recently, it was found that the use of layered 2D materials (graphene, molybdenum disulfide, etc.) leads to nontrivial capillary hydrodynamics in nanoscale pores and channels [4, 5], which allows a sharp increase in the rate of supply of a cooling evaporating liquid to the interface, practically removing the limiting factor of the fluid flow rate.

The aim of this study is to compare the calculated and experimental cooling rates of heated silicon substrates using a layer of graphene material, which is a partially oriented graphene nanoflakes 1.5-3.5 nm thick with lateral dimensions 10-30 μm, obtained by supercavitational cleavage of high-purity graphite in distilled water (Graphene Institute Ltd, Russia).

In this paper, modern data on the capillary thermohydrodynamics of nanopores and nanochannels are presented, the mechanisms of strong evaporative cooling using two-dimensional materials are considered, and examples of the use of such approaches to promising thermal management are given. In natural phenomena, strong evaporative cooling occurs very often, therefore, many mechanisms of capillary thermohydrodynamics and strong evaporation can be "spied" from nature, which only in recent years has become an obvious fact.

The nanoporous structure is very difficult to implement in conventional materials, however, the possibility of its creation is directly related to two-dimensional materials (in several atomic layers),
which at present can be implemented, for example, using graphene nanoflakes (thickness 2-4 nm, lateral size - from 20 to 100 μm) in the manufacture of bulk structures. It is important that in such structures the transverse size of nanochannels can range from 2 to 8-10 nm, which naturally leads to an abnormally high velocity of water movement, as shown by direct experiments with graphene nanomembranes [3,4] and our experiments on evaporation from such structures under the action of solar radiation [2,4].

In addition to traditional models for describing capillary thermohydrodynamics with an interphase boundary, new physical effects appear in some problems - taking into account the slip of the liquid phase near the boundary, peculiarities of surface wetting with nano-roughness, electroosmotic, thermoosmotic flows and thermophoresis [4].

2. Materials and method
The experimental cell consisted of a silicon heated substrate, a container inside which an evaporating liquid was supplied (in this work, data are given only for water and aqueous graphene nanofluids), a graphene structure block placed inside steel meshes of copper or stainless steel, and also consisting of a nanoporous structure steam condensation unit. The condenser could force condensed liquid into the cooling unit (or similar to heat pipes) (figure 1).

![Figure 1](image1.png)

**Figure 1.** Schematic of an experimental cell for studying evaporation from nanoporous graphene structures.

Graphene nanoflakes with an average size of 10-30 μm (thickness - 1.5-3.5 nm) were received from Graphene Institute Ltd (Russia) (figure 2). Chemically clean graphite microparticles ~300 μm were used. As a mesh we used cooper M1(Cu>99.6%) or stainless steel mesh with cell side size 0.3 mm and wire diameter 0.1 mm supplied by TDMC, Russia.

![Figure 2](image2.png)

**Figure 2.** Oriented graphene nanoflakes for evaporation.

A nanoporous oriented layer of graphene nanoflakes before being placed inside the experimental cell is shown in figure 3. The geometrical dimensions of the evaporating block were chosen differently, however, a high graphene layer of about 2 mm and a diameter of 10-12 mm was found to
be optimal. For more efficient operation, the graphene layer was saturated with water or other liquids using drop dispensers (figure 3). The temperature of the silicon substrate was varied from 25 to 65 °C. It is clear that the evaporation rates and cooling efficiency increased with an increase in the substrate temperature. The substrate temperature was measured with thermocouples and heat flow sensors. The temperature was measured with an accuracy of about 0.5 °C. The evaporation rate was measured by an accurate electronic balance inside the nanoporous capacitor, as well as by the liquid flow rate through the heating unit.

3. Evaporation rate data
In addition to pure liquids, we were the first to study the evaporation of graphene nanofluids inside nanoporous structures. Preliminary data show that such fluids heat up and evaporate more efficiently, as shown by the empirical data in figure 4.

![Figure 3. Graphene structure (wick) with mesh for placement inside the evaporation unit.](image)

![Figure 4. Evaporation rates of various liquids using graphene nanochannels and nanopores.](image)

The physical mechanisms of the increased evaporation rate are not yet completely clear and require additional study. At the same time, the use of graphene nanofluids together with graphene nanoporous structures makes it possible to create an active system of evaporative cooling of a continuous cycle, which is extremely important for a large number of applications. The obtained experimental data on the evaporation of water and aqueous graphene nanosuspensions with mass concentrations of 1% and 3% are shown in figure 4.
4. Comparison with mathematical models
A feature of evaporation from oriented graphene nanostructures is nontrivial capillary hydrodynamics - a strong influence on the movement of the meniscus of the interface of the boundary conditions on the liquid - graphene plate surface (almost complete water sliding without tangential resistance - (Navier boundary condition and significant slip length) [4]. In this work, we used models close to those developed in the [5].

4.1 Capillary-driven flow
The imbibition of liquid into a nanoporous structure occurs when the solid–vapor specific surface energy ($\sigma_{sv}$) is greater than the solid–liquid specific surface energy ($\sigma_{sl}$). This usually occurs when the contact angle is less than 90°. The driving force for imbibition is characterized by the Young–Laplace equation

$$\Delta p = p_l - p_v = 2\sigma_v \Gamma$$

where $\Delta p$ is the local capillary pressure, $p_l$ is the local liquid pressure at the liquid–vapor interface, $p_v$ is the pressure of the vapor phase, $\sigma_v$ is the liquid–vapor surface tension, and $\Gamma$ is the local mean curvature of the liquid surface.

In contrast to the work [5], where the Brinkman equation (or an analogue, the Darcy equation) was used for the flow of a liquid inside a porous structure, in this study we describe the flow of a liquid inside a graphene structure based on non-slip flow. In this case, the slip length significantly exceeded the characteristic scale of nanopores $L_s$>>$\delta$ ($L_s$~50 nm, $\delta$ ~3-5 nm).

4.2 Heat transfer
Heat in thermal management devices is generated due to Joule heating and is transferred to the liquid vapor interface through the combined solid-liquid matrix by the heat conduction mechanism. Further, heat is dissipated by convection, heat conduction and thermal radiation into the environment. Typically thermal management systems use a heat transfer coefficient $\alpha$=$q(T_s$-$T_m)$, $q$ is the steady state heat flux dissipated by the graphene nanostructures (wick), $T_s$ is the temperature at the solid interface, and $T_m$ is the temperature of the far field ambient.

The effective thermal conductivity of the graphene nanostructures is dependent on the working fluid, the solid material and nanostructures and operating conditions. In the simplest case, the following model can be adopted (the study showed that the complication of the model insignificantly affects the final result)

$$\lambda_{eff} = \lambda_s \frac{2 + \lambda_s \lambda_v^{-1} - 2\phi(1 - \lambda_s \lambda_v^{-1})}{2 + \lambda_s \lambda_v^{-1} + \phi(1 - \lambda_s \lambda_v^{-1})}$$

where $\lambda_s$, $\lambda_v$ - the thermal conductivity of the solid and liquid, $\phi$ - the porosity. When using nanofluids, the quantity is replaced by the effective thermal conductivity of the nanofluid ($\lambda_v$→$\lambda_{v,eff}$).

4.3 Vapor flow
The mass flow of vapor during evaporation can be determined by the following relationship

$$\vec{j}_v = \rho_m \vec{u} - \rho_m D_\nu \nabla \varphi$$

where $j_v$ is the vapor mass flux vector, $\rho_m$ is the mixture (air + vapor) density, $\varphi$ is the vapor mass fraction, and $D_\nu$ is the diffusivity of vapor in the gas (or mixture of gases). The evaporative mass flux is related to the evaporative heat flux by $q_v = j_v \alpha_v$, equivalent thermal resistance is $R_v = (T_s - T_m)(j_v \alpha_v S)^{-1}$.
Using the above model, the rates of evaporation of various liquids (water and water suspensions of graphene) were calculated. These calculated data are presented in figure 4, which also shows experimental data in the study of evaporation from graphene oriented nanostructures.

Unfortunately, within the framework of the used model, there is no good agreement between theoretical and experimental data, which speaks, first of all, of the shortcomings of the existing models of evaporation from nanostructures. Note that the results of the calculations give data exceeding the evaporation rate found experimentally.

It should be especially emphasized that for the case of liquid sliding inside graphene nanochannels and evaporation from the surface of the meniscus inside the channel, an interesting situation can arise - the liquid inside the channels can move much faster (slip boundary condition) than the liquid-vapor interface. As far as we know, such cases have not been previously considered within the framework of mathematical models.

In our opinion, it is precisely in these processes that the nontrivial capillary hydrodynamics of liquids inside the nanochannels manifests itself. Therefore, in addition to the importance of further constructing models of vapor transport in the external environment, as noted in, it is necessary to more consistently consider capillary nanohydrodynamics inside channels, especially when it concerns two-dimensional materials such as graphene and its structure.

4.4 Cooling

The fraction of heat dissipated by evaporation was determined by subtracting the perceptible cooling and parasitic heat loss from the heating source. The heat flux used for cooling was calculated from the ratio of the energy balance

\[ q_c = (j_v + j_L)C_p(T_{out} - T_m)S_{Cool}^{-1} \]

where \( q_c \) - heat flow for cooling, \( j_v \) - vapor mass flux, \( j_L \) - evaporation loss, \( C_p \) - specific heat capacity of the working fluid, \( S_{Cool} \) - cooling surface area. The table shows the calculation results and experimental data on the dependence of heat fluxes during cooling on the type of working fluids and the heat transfer coefficient.

| Working fluids | \( \alpha \), W/cm²K | Maximum heat flux, W/cm² | Publication |
|----------------|----------------------|---------------------------|-------------|
| Water          | 9.6                  | 140                       | [6,7]       |
| Water          | 14.8                 | 180                       | [6,7]       |
| Water          | 38                   | 390                       | [6,7]       |
| R-245fa        | 39.7                 | 550                       | [6,7]       |
| Pentane        | 18.4 (th – 20.2)     | 230 (theory – 244)        | [this work] |
| Water          | 24.7 (th – 20.2)     | 297 (th – 316)            | [this work] |
| Water+GNF1%    | 31.2(th – 34.8)      | 340 (th – 369)            | [this work] |

5. Results and discussions

A preliminary comparison with the rate of evaporative cooling in other systems, as well as using heat pipes, showed that the efficiency of evaporative cooling using oriented graphene nanostructures is several times higher. The use of active cooling systems in thermal management systems (thermal interface materials and efficient capillary hydrodynamics with strong evaporation from nanopores) is
currently the most attractive approach to thermal stabilization of energy-intensive electronic and energy systems.

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