On the dynamic effects of evaporation and boiling of drops on inhomogeneous graphene substrates for effective thermal management of energy devices

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Abstract: This research examines thermal performance of metal-graphene nanocomposites for thermal management. Graphene nanoflakes and graphite microparticles were compressed with cooper meshes under different pressures. The prepared samples were characterized using optical microscopy. The fabricated samples of cooper-graphene composites were studied to determine the droplet evaporation rates and contact angles at different temperatures including film boiling.

1. Introduction
Due to constant growth of thermal design, power of modern electronic devices there is growing interest in new thermal interfaces materials.[1-4] One of the main goals of the work is to study the properties of thermal interface materials for intensifying heat transfer and increasing the efficiency of cooling systems in microelectronics and high-current electronics. The physicochemical and thermohydrodynamic properties of such surfaces are important for more efficient heat dissipation in cooling systems using the drop cooling method. In such systems, the intensity of evaporation of the working liquid from the surface, as well as the conditions for the transition from the bubble mode of boiling to the film mode, are extremely important. The structure of the tablet has a strong effect on the evaporation process. Moreover there was investigated that pressing pressure on graphene composite heavily impacts on evaporation processes and on the destruction of the surface at high temperatures. The structure with a grid on the surface was chosen to check an impact on evaporation time and contact angles at wide range of temperatures.

Previously, surface modification using various polymer networks to study wettability and evaporation rates has already been used [5]. However, we are using metal meshes with graphene flakes here for the first time to study contact angles and evaporation rates of droplets.

In this work, we investigated the processes of evaporation and boiling, up to the Leidenfrost temperature, of liquid droplets (mainly water) on the surface of inhomogeneous graphene and graphite substrates with metal meshes. The effect of an inhomogeneous surface on the evaporation rate of droplets, as well as the behavior of contact angles depending on the temperature and parameters of the substrate surface, is shown.
2. Materials and method
Graphene nanoflakes with an average size of 10-30 μm (thickness - 1.5-3.5 nm) were received from Graphene Institute Ltd (Russia). Chemically clean graphite microparticles ~300 μm were used. As a mesh we used cooper M1(Cu>99.6%) mesh with cell side size 0.3 mm and wire diameter 0.1 mm supplied by TDMC, Russia. Samples were pressed in hydraulic press under 180-300 bar pressure in inch diameter tablet. We prepared 4 samples in total: a) graphene nanoflakes under 300 bar; b) graphene nanoflakes with cooper mesh under 300 bar; c) graphene nanoflakes with cooper mesh under 180 bar; d) graphite microparticles with cooper mesh under 300 bar (figure 1).

The technique for measuring the contact angle and the evaporation rate is similar to that presented in our works [6,7].

![Figure 1. Photos and microscopy of samples. a) graphene nanoflakes p=300 bar; b) graphene nanoflakes with cooper mesh p=300 bar; c) graphene nanoflakes with cooper mesh p=180 bar; d) graphite microparticles with cooper mesh p=300 bar; e) sample B optical microscopy x4 magnification; f) sample B optical microscopy x10 magnification; g) sample of cooper mesh 03x01.](image)

As we see from microscopy metal wires are not cylindric so we consider the surface as flat and somewhere graphene covers metal wires as we see in 10x magnification. For all types of samples, contact angles and evaporation rates were studied. Kruss EasyDrop was used as a system for measurement the contact angle and evaporation rates of drops on the composites. Also to measuring system was added a special heater with temperature control realized by PID controller for making a research in wide range of temperatures. Evaporation rate and contact angles were measure for all 4 samples. For each sample and each temperature was made from minimum 3 measurements at temperatures 30-40 °C and minimum 5 measurements for 50 °C and more. Distilled water was used as a working liquid.

3. Contact angles for samples nanocomposites and evaporation rates
Wettability makes a heavy impact on heat exchange between operating fluid and working surface. Various materials can be used for this purpose and more various methods can be used for surface
modification [4-5]. In our work we made metal-graphene composite with regular structure made by webbed wires of cooper mesh (figure 1 e,f).

As a result, we can see that adding cooper mesh in graphene tablet leads to increase of contact angle from several degrees till 20 degrees under different surface temperatures (figure 2). But from another hand we see that on the sample C contact angle is dropping relatively to sample A. For proper description we need further researches, but preliminarily we explain it with porosity of graphene samples made with relatively low pressure. Also pressure heavily impacts on surface durability on high temperatures which described in more details in next section.

Figure 2. Contact angles at the start of the evaporation of water droplets for nanocomposites.

Relating to evaporation rates we see relatively the same times on 40-90 °C. But closer to boiling point of water we see great difference between samples. As we see from figure 3 adding a mesh to the surfaces decreasing the evaporation time on both ranges from 100 °C to Leidenfrost point and over Leidenfrost point. On lower pressure sample we see even more rapid evaporation in 100 °C to Leidenfrost point but over it while film boiling evaporation rate is lower than on sample B. Even more Leidenfrost point is higher approximately on 10 °C. On graphite sample we see medium situation relatively to sample B & C on 100 °C to Leidenfrost point range, but after it there is very rapid difference in evaporation rate.

Figure 3. Evaporation time of water droplets for nanocomposites at different surface temperatures.
4. Surface destruction

Previously we described surface destruction of surface of graphene tablets [6]. In this work we made 3 samples under 300 bar pressure. For this samples there is no visual destruction even in optical microscope with 40x magnification. But we investigated that no matter how you clean the sample before the droplet evaporation, on temperatures higher than Leidenfrost point, levitating droplet finds some particles and we see an evaporation of colloid solution (water + graphene nanoflakes).

![Figure 4. Levitating droplet of water with graphene nanoflakes from surface.](image)

Cleaning the surface after each droplet didn’t helped as well. This particles are not visible on the surface and we observed them only closer to the end of the evaporation process because color of water changes from transparent to black during the process and leaves batch of graphene nanoflakes at the end (figure 4).

![Figure 5. Photos of samples exposed to destruction sample C (left), sample D (right).](image)
For the sample C we observed nearly the same situation that in [6]. The only difference that size of partial destruction is limited by the metal cell dimensions (figure 5). Also graphene detachment from surface of the wire remains possible. So in case of durable using we can get a rough surface that radically differs from initial.

For the sample D we observed a bit different situation. If for the graphene samples 300 bar is enough pressure to obtain stable surface, graphite has lower adhesion force between particles, and after all measurements there was significant surface destruction at the edge of the sample (figure 5). Wires of the mesh was separated from the graphite at the edge and inner layers of graphite was more subjected to destruction, so when the droplet moved on the edge of the sample character of evaporation radically changed and early on we limited surface of the sample, and later made the 2nd same sample for measurements on high temperatures. To confirm accuracy of results we compared evaporation rate on both samples and didn’t observe significant differences. That’s why we didn’t make any other samples containing graphite as surface destruction is too heavy for using. To prevent this destruction there is a possibility of sintering processes while manufacturing or adding some polymers to keep the sample intact.

5. Results and discussions
In this work, we were the first to use metal meshes with graphene flakes to study the contact angles and evaporation rates of droplets. In our opinion, such inhomogeneous structures are very promising for active thermal interface materials with drop cooling of surfaces. This is especially important when the surface temperature is close to or higher than the Leidenfrost temperature. In this situation, thermal stability of the surface is required as a thermal interface material.

The structure with a grid on the surface reduces the evaporation time in the temperature range between the boiling point of the working fluid and the Leidenfrost temperature, moreover, for a sample with a lower pressing pressure, as a consequence, with a higher porosity, by more than 4 times. On a sample with graphite and a grid, it was also possible to achieve improvements in the rate of evaporation, as well as the values of contact angles are smaller than on samples with graphene, but graphite samples are also less resistant to water, as well as sample C (see Fig. 5) that they become nearly impossible to use in real arrangements. Moreover very high evaporation times while film boiling could be imperfect on the working temperatures close to Leidenfrost point. Using structures with high porosity allows us to increase Leidenfrost point and also adding cooper mesh to the surface leads to the same result, but we should admit that evaporation time after Leidenfrost point on the sample with high porosity is higher than on the same surface pressed under higher pressure with less pores. Destruction of samples with pressed graphene nanoflakes is need to be investigated, but as we can see 300 bar is enough pressure to evade much surface destruction.

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