Physical features of Leidenfrost effect on the surface of a graphene nanocomposite for the problems of thermal and nuclear energy

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Abstract. We investigate the possibility of overcoming the Leidenfrost point to achieve the stable boiling of a pool (not a film) at temperatures much higher than the Leidenfrost temperature for water on smooth surfaces. We have studied the Leidenfrost effect (droplets on a surface overheated above the film boiling temperature). Surfaces made of hybrid graphene nanocomposites are used as substrates. We investigated two types of nanocomposites based on graphene flakes (the size of the latter being about 3-5 nm in thickness, longitudinal dimensions of about 10-20 µm), sintered at a pressure of 200-300 bar, and hybrid nanocomposites consisting of graphene flakes, metal microspheres (a diameter of about 100-200 microns) and a small amount of polymer (epoxy resin).

1. Introduction
The film boiling of liquid droplets on highly superheated surfaces (the Leidenfrost effect) has been an area of extensive studies in recent years, both in connection with new problems of thermal physics, and with applications, in particular, for jet cooling of high-temperature surfaces in power engineering and electronics [1-3]. Heat dissipation from hot surfaces through cooling droplets is limited to the Leidenfrost point (LFP), in which an insulating vapor film prevents direct contact between the cooling droplet and the hot surface. Evaporation and boiling are the most important processes in a wide range of applications in various industries, such as power generation (thermal and nuclear), refrigeration and cooling, water purification, electronics etc. [1-5]. For boiling heat transfer, the latent heat of the liquid-vapor phase transformation allows one to achieve a large dissipated heat flux. At low superheat values, the direct contact of the liquid with the hot surface ensures an efficient heat transfer. However, at high superheat values, a vapor film is formed between the liquid medium and the hot surface and minimizes the rate of heat transfer. The onset temperature of this phenomenon is indicated by the Leidenfrost point (LFP). Investigation of the possibility of increasing the temperature of Leidenfrost or, in general, eliminating the film boiling (overcoming the Leidenfrost limit) is one of the most important problems of heat and mass transfer on new mesoscopic and nanostructured surfaces. Recently, a surface concept of decoupled hierarchical structures [6,7] was developed which could eliminate the LFP.

2. Material and methods
Graphene substrates consisting of pure graphene flakes or graphene flakes, metallic monodisperse pellets and a small amount of polymer as an adhesive filler were used in the work. The method of obtaining composite materials is as follows. Graphene flakes produced by the NanoGraphen Group were used to prepare a nanocolloid solution (nanofluid) for which graphene flakes of various volumes were added to pure distilled water and stirring was carried out for 20-30 minutes in a rotating centrifuge. Graphene flakes 5-10 microns in size with 3-10 layers of graphene (few-layer graphene, FLG) were obtained (figure 1a). In addition, hybrid nanocomposites were fabricated using the technology presented in [4] (figure 1b). The production of the tablets was carried out by means of an automatic machine for hot pressing the Buehler SimpliMet 1000.
Then the press-fitting parameters are selected on the control panel of the press and the process is started. At the end of the process, the cap is opened and the mold with the obtained graphene tablet is lifted using the control buttons. For this work, samples of graphene tablets were made with various press parameters and powder volumes.

As a result, samples of different thicknesses were obtained. In production, pressures of 200 and 300 bar were used. At each pressure value, four samples of different thicknesses were made. The thickness depended on the amount of graphene powder used. The quantity was measured in volumetric units with a syringe, which allowed the measurement of a certain amount and was convenient for loading the metered powder into a mold. Since this press is highly specialized equipment, mainly intended for metallographic research, it constantly heats the material loaded in the mold. Therefore, the lowest possible temperature of 150 °C was used for manufacturing. As mentioned above, four samples with powder volumes equal to 2, 5 and 10 ml were prepared for each pressure value, respectively. Different values of pressure and volume gave different thicknesses of the obtained samples (figure 2, left). Figure 2 (on right) shows the optical image of the surface (sample $h = 2$ mm).

### 3. Contact angles and evaporation rate on graphene tablet surfaces

We investigated two types of nanocomposites based on graphene flakes (the size of the latter being about 3-5 nm in thickness, longitudinal dimensions of about 10-20 µm), sintered at a pressure of 200-300 bar, and hybrid nanocomposites consisting of graphene flakes, metal microspheres (a diameter of about 100-200 microns) and a small amount of polymer (epoxy resin). The Kruss EasyDrop measuring system was used with a special heater and temperature control system. First, contact angles were studied as a function of temperature for different samples of graphene substrates.

We also investigated the evaporation rates of droplets of distilled water on the surface of graphene tablets at different temperatures. To heat the substrates, the ITO (Indium Tin Oxide substrate) was used, the measurements were made by a non-contact infrared thermometer and thermocouples.
Various properties of hybrid nanocomposites based on microspheres and graphene were studied. In some cases, the microspheres coated with graphene were placed in a polymer matrix. The temperature dependences of the contact angles of water droplets on hybrid graphene substrates over a wide temperature range are studied. We also investigated the evaporation rates of droplets of distilled water and nanocolloids of water + silver nanoparticles (AgNP - 0.5% by volume; the average nanoparticle size is about 20 nm), as well as water + graphene flakes (GNF - 1% by volume). As an example, figure 3 shows the dependencies of the evaporation rate of distilled water and nanocolloid (water + silver nanoparticles) droplets on the graphene tablet surface (pressure of 300 bar). It is important to note that water droplets with silver nanoparticles evaporate more strongly, which, apparently, is associated with an increased effective thermal conductivity of such nanofluids. According to estimates of the evaporation rates for nanocolloid droplets, such an increase can be about 11-14%, which roughly corresponds to our experimental data.

Figure 3. Evaporation rate of distilled water and nanocolloid (water + silver nanoparticles) droplets on the graphene tablet surface

4. Leidenfrost effect on graphene tablet surfaces

After experiments on the measurement of contact angles and evaporation rate on graphene tablet surfaces, studies of the boiling processes on graphene substrates at various temperatures were conducted. In this paper, we investigate the possibility of overcoming the Leidenfrost point for organizing a stable boiling of the pool (not the film) at temperatures much higher than the Leidenfrost temperature for water on smooth surfaces. We studied the Leidenfrost effect (droplets on a surface overheated above the film boiling temperature). Surfaces made of hybrid graphene nanocomposites, which were first developed and created in [4-6], are used as substrates.

A special feature of hybrid graphene nanocomposites is a noticeable increase in the Leidenfrost temperature $T_L$. If on metal substrates for water $T_L \sim 175-190$ °C, then the minimum value for graphene substrates is $T_L \sim 210-230$ °C. At temperatures below $T_L \leq 120$ °C normal evaporation and pool boiling droplets on graphene substrates are observed (figure 4a). The next fact that has been clarified is the partial destruction of graphene substrates starting from a temperature $T_L \geq 120$ °C with the removal of graphene flakes from the surface (figure 4b). This, apparently, can be explained by the presence in the material of air caverns, which when heated leave the surface and destroy graphene substrates. It is interesting to note that for hybrid composites containing metallic microspheres, flocculation of smaller-sized graphene flakes was observed at temperatures above $T_L \geq 200$ °C, in the case of pure graphene samples at temperatures $T_L \geq 120$ °C.
Figure 4. Pool boiling droplets (a) and destruction of substrates (b) on graphene tablet surfaces

A study of the evaporation of droplets with a transition of the surface temperature above the film boiling point (Leidenfrost temperature) is performed. In particular, figure 5 shows a drop of water with a diameter of 2 mm on the surface of a graphene nanocomposite in the Leidenfrost regime (figure 5a) and a violation of the stability of levitation at times of about 5-6 seconds (figure 5b). When the temperature is above $T_L \sim 230 \, ^\circ C$, in the first seconds on graphene nanocomposites a very stable Leidenfrost effect is observed (figure 5a). At the same time, after the regime of stable film evaporation, the stability of levitation is disturbed at times about 5-6 sec (figure 5b). The figure shows that the drop is in contact with the hot surface, without a vapor film, quickly evaporates and boils. The mechanism of this phenomenon is not very clear, but perhaps the reason for this is the melting of the polymer that is part of the hybrid nanocomposite and the observation of the breakdown of the Leidenfrost regime on mesoscopic surfaces, as was first noted in [7].

Figure 5. Water drop on the surface of a graphene nanocomposite in the Leidenfrost regime (a) and violation of the stability of levitation (b)

5. Conclusions
Due to high-efficiency the boiling heat transfer is frequently used in the heat transfer devices, such as a heat exchanger, boiler, and so on. The evaporation rate of the drops, which is important for the cooling systems of power plants in the cooling systems of jets and droplets, showed a very large amount on the graphene nanocomposite surfaces. This allows us to recommend such surfaces for efficient cooling systems for power and electronic equipment. We investigated the critical heat fluxes for the transition from the pool boiling to the film boiling (Leidenfrost effect). Unfortunately, the accuracy of such measurements is not more than 15-20%. At the same time, it was possible to establish that there is a noticeable difference in the critical heat fluxes for the four types of surfaces studied: the mesoscopic surface (carbon microspheres, 5-7 µm in diameter), the graphene tablet 200 bar and 300 bar, and the graphene nanocomposite.

We conducted studies of critical heat fluxes for various types of nanocomposites, which may be important for energy applications. Figure 6 shows a comparison of the critical heat fluxes for these surfaces. Mesoscopic surfaces based on carbon microspheres (diameter 5-7 µm), graphene tablets...
made at 200 bar and 300 bar, graphene nanocomposites were studied. It is clearly seen that the highest critical heat fluxes are observed in graphene nanocomposites - up to 140 W/m², which makes them very promising for many applications in thermal and nuclear energy. In addition, the latter materials are significantly more stable at higher temperatures than graphene tablets. This allows us to recommend hybrid graphene nanocomposites for use as materials in thermal and nuclear power engineering.

![Graphene nanocomposite critical heat fluxes](image)

**Figure 6.** Critical heat fluxes for various surfaces

The features described above play an important role in the development and creation of new functional surfaces for thermal and nuclear power engineering, in the problems of thermal stabilization of electronic and optoelectronic devices [8]. New functional surfaces, as a rule, should have a high Leidenfrost temperature, a stable levitation of droplets or, conversely, a rapid transition when the droplets interact with superheated surfaces, into the pool boiling mode and efficient evaporation.

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