Effect of material properties on heat transfer during cooling of high-temperature cylindrical bodies in liquids

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Abstract. The aim of the present paper is an experimental study of the cooling processes of high-temperature cylindrical samples with different thermal conductivity of the metal in liquids. Studying the influence of material properties on heat transfer during cooling and the possibility of accurately prediction of the transition surface temperature to the intensive cooling regime will allow substantiating the choice of a new type of tolerant fuel from the standpoint of thermophysics. The experiments were conducted for cooling in water and ethanol with different subcoolings under atmospheric pressure. For metals with low thermal effusivity, an increase in the transition temperature with a significant increase of the removed heat fluxes from the surface was experimentally discovered.

1. Introduction
The process of cooling high-temperature bodies in liquids is an important physical process which occurs in metallurgy, nuclear industry and cryogenic. During quenching it usually occurs film, transient and nucleate boiling on solid surface, and each of the boiling regimes is accompanied by its own heat transfer characteristic. In the list of abovementioned boiling regimes, it should be included so called “microbubble boiling” regime, which was first studied by Hewitt and Kenning [1]. Their study showed that microbubble boiling begins at high surface temperature and featured by high heat transfer rates during quenching in subcooled water. This combination causes large temperature gradients near the heat exchange surface, which cause stresses in the metal and cause the sample to crack [2]. The study of this regime is important not only from a fundamental point of view (there is still no strict physical model), but also for calculating the strength of materials.

In relation to nuclear power plants (NPP), it is a core cooling of nuclear reactor using an emergency cooling system when the fuel cladding overheating during a loss-of-coolant-accident (LOCA). After the accident at the Fukushima-1 NPP in 2011, large-scale research was launched to find a new accident tolerant fuel. The zirconium cladding of the fuel rods has different advantages under normal operating conditions in water-cooled nuclear reactors, however, there are serious drawbacks in the event of severe accidents, such as the occurrence of a steam-zirconium reaction, cracking and embrittlement of the fuel cladding. New materials for use as coatings or as a replacement for zirconium fuel elements must be investigated thoroughly under reactor conditions, as well as in the event of emergency situations.

There are several potential claddings materials among which the most developed are chromium coating on zircaloy, FeCrAl, NiCr and SiC. The analysis of influence of different fuel and cladding materials on performance during nominal operation regime and accidents is presented in [3]. The authors
concluded that the use of the ATF fuel allows for increasing the time margin for the actions preventing possible NPP core damage. The accident tolerance of the SiC cladding was studied in [4]. Tiubeheaters of SiC and zircaloy-4 was placed in the pool of deionized water, which was heated to boiling at atmospheric pressure. An increase in critical heat flux (CHF) of 63% was observed for SiC compared to zircaloy, which the authors attribute to a higher thermal conductivity and roughness of SiC. Also, the physical degradation did not occur during heating even after the CHF with transition to film boiling for SiC cladding. The chromium is considered as a coating of zirconium fuel elements, which demonstrates excellent properties. In the work of Korean authors [5], when cooling samples with a chromium coating after oxidation at 1200 °C in water, the temperature of the oxidized tube dropped from 1200 to 60 °C in just 5 s. This experiments and studies conducted at CEA (France) [6] showed that less hydrogen is formed after oxidation in coated than in uncoated sample.

So, studies of cooling processes and the influence of the materials properties are of great importance. The intensification of film boiling is also interesting for cryogenic technology. Different studies show that the cooling intensity can be increased by changing the condition of the heat transfer surface. In the paper [7] discuss the results obtained during the transient cryogenic quenching by liquid nitrogen jet impingement. The quenching samples were circular plates made of rock and aluminum alloy. The experimental results showed that after jet impingement, the surface with low thermal conductivity and microstructural porosity was directly wetted by liquid nitrogen jet without film boiling, the rewetting temperature of rock plate is almost 100 °C higher than of aluminum surface. For a quantitative interpretation of the high rewetting temperature on the surface, the authors use the theory of the possibility of the local direct liquid/solid contact, which may explain the absence of film boiling at stagnation point and enhancing the transition temperature on the rock surface. The paper [8] consists experimental results on quenching of stainless steel spheres (polished, hoarfrost and iced surfaces) in liquid argon and nitrogen. The experiments on hoarfrost surface (high porosity and low thermal effusivity) demonstrated higher cooling rates. The same effect of low thermal conductivity coating was observed during cooling of copper plate by falling nitrogen film [9]. In [10] present the results of an experimental study of heat transfer regimes during cooling in water of high-temperature heated steel balls with a technically smooth and modified surface. The modification consisted of the application of a finely dispersed carbon coating to the surface followed by electron beam treatment. The carbon coating leads to a decrease in the surface temperature which corresponds to the transition to the intensive cooling regime.

The study of the cooling processes of high-temperature bodies is important in metallurgy, where it is required to obtain metal with desired properties as a result of quenching. In [11] was analyzed quenching processes of the heated to 850 °C Inconel 600 probe in a mineral oil, the authors estimated the heat flux transients along axial and radial locations during immersion quenching. For this, time–temperature data were obtained at different axial and radial locations for cooling of the Inconel probe. Thanks to this, it was possible to fix that the wetting front starts at the bottom and ascends to the top of the quench surface, and the variation in peak heat fluxes estimated at the radial location indicates the non-uniform nature of the wetting front. The study [12] also noted that the quench front propagates upwards from the bottom for both the untreated and superhydrophilic rodlets. It was shown that surface superhydrophilicity and increase in subcooling of the liquid can significantly accelerate the quenching process. The stainless steel rodlets with a superhydrophilic surface were cooled in saturated and subcooled water. Superhydrophilicity was created by applying silica nanoparticles to the samples, which increased the surface roughness. The cooling of the superhydrophilic surface was faster than for the untreated surface in saturated water, and the cooling time was further reduced with an increase in subcooling. Another paper [13] studied experiments on cooling of the heated metal balls made of carbon and stainless steel in brines and water-based suspensions of graphite and clay, temperature was measured in the center of the balls. The experiments showed that a change in the mass fraction of the solid admixtures in a coolant weakly affects the cooling process. Also, weak fluctuations in temperature near the liquid/vapor interface were found during the pool film boiling of the subcooled deionized water, the authors attribute this to phenomenon of the “step” change of the vapor film thickness, associated with the short-term multiple liquid/solid contacts.
The present paper is devoted to study the influence of surface properties and thermophysical properties of cooling liquids on heat transfer during cooling of high-temperature cylindrical bodies in liquids, and determination of transition temperature to an intensive heat transfer regime.

2. Experimental facility and test samples
The experiments were conducted at the experimental facility shown in Figure 1, is the same that was depicted in [14]. The sealed housing of the experimental volume (6), made of stainless steel pipe with an external diameter of 219 mm, is the base of the stand. The copper coil (2) of a high-frequency (HF) induction heater (3) is placed in the upper part, and the cooling liquid is poured into the lower part. A metal diaphragm (5) separates the heating zone and the volume of liquid. The test sample is a cylinder (1) which is located inside the coil (2) using a lever system (4). Thermocouples are installed inside the sample, by which heating is controlled. The sample is heated to a set temperature (400–700 ºC for the experiments discussed below) in less than 2 minutes and then moved to the cooling liquid at the level of viewing windows. The temperature of the liquid is maintained at a predetermined level by means of a thermostat (8) and a connected coil immersed in the liquid (7). During the cooling process, the signal from the thermocouples (11) passes through the Ni SCXI-1303 connector to the Ni SCXI-1102 (12) measuring module, which is a part of the Ni SCXI-1001-based assembly. The signal from each thermocouple is registered at a frequency of 100 Hz. The measurement results are transmitted via a USB interface to a personal computer (13), where the temperature-time dependence is constructed in the Lab View program. The advantage of this stand is the ability to conduct experiments in a wide range of pressures (0.1 – 1.0 MPa) and temperatures (-200 ºC – 200 ºC). For performing experiments under pressure, the working sample is heated in the inert gas (argon or nitrogen) environment coming from a high-pressure reservoir (9). The gas pressure is regulated by a reduction gear (10).

![Figure 1. Schematic of experimental facility: 1 – tested sample, 2 – HF-inductor coil, 3 – HF-inductor, 4 – displacement system, 5 – metallic diaphragm, 6 – experimental camera, 7 – coil pipe, 8 – thermostat, 9 – vessel with inert gas, 10 – manometer, 11 – thermocouples, 12 – measuring module, 13 – personal computer, 14 – video camera, 15 – halogen lamp.](image-url)
At this stage of research video was not taken which is due to the difficulties of obtaining reliable information when shooting the boiling process [15].

The working samples are cylinders with the same size: 10 mm in diameter and 50 mm in length, made of different metals with and without coating. The bottom part had a spherical shape to reduce the end effects. Note that the experimental results in the study [16] showed an increase in the heat transfer performance of about 20% for cooling of a high-temperature cylinder with a hemispherical bottom compared to a flat bottom surface, the transition temperature also increased for this sample.

Two samples were prepared - nickel (thermal effusivity is approximately 18900 Ws\(^1/2m\)^2K\(^{-1}\)) and copper cylinder with a gold coating (thermal effusivity of copper and gold are 37000 and 28000 Ws\(^1/2m\)^2K\(^{-1}\), respectively). Copper has low resistance to oxidation, especially at high temperatures. Therefore, the gold coating performed a protective function, while this metal has a similar high thermal conductivity as copper. The gold coating was deposited by the galvanic method, after that the sample was polished. Arithmetical mean deviation of the assessed profile (Ra) is 1 μm. The nickel cylinder also was polished using polishing ceramic (Al\(_2\)O\(_3\)) and porcelain particles, the profilometry results showed that Ra is 0.2 μm. Polishing was carried out using vibratory finishing equipment by Walter Trowal in Finishing Technology Center (Moscow).

A single central thermocouple was installed in the copper cylinder with gold galvanic coating, since copper has a high thermal conductivity. The technology for mounting the thermocouple is shown in Figure 2a. The K-type cable thermocouple (4) is inserted at the center of the sample (1). The hot junction is firmly fixed due to the graphite sealing (2) after force screwing the metal tube holder (3) in the sample. The technology for mounting thermocouples in a nickel cylinder has differences, since nickel has a low thermal conductivity. It was decided to measure the temperature in two places - on the surface (at the bottom of the cylinder) and in the center, which necessitated the mounting of two thermocouples. The Figure 2b shows the mounting scheme for this method. The thermocouples were mounted using a clamping collet (4), and a mixture of liquid indium and gallium metals was used for better contact of the hot junction with the surface.

![Figure 2. Mounting of a thermocouple. a - in the copper sample: 1 – sample, 2 – graphite sealing, 3 – tube holder, 4 – thermocouple cable (TC); b – in the nickel sample: 1 – sample, 2 – holder tube (d=3 mm), 3 – TC passing inside the holder tube (type K) d=1 mm, 4 – clamping collet, 5 – release of the thermocouple cable from the holder tube, 6 – heat shrink, 7 – extension (compensation) wire, 8 – plug.](image)

3. Results and discussion

The Figure 3 shows cooling thermograms obtained by cooling the cylinders in subcooled ethanol \(AT_{sub} = 48\) K. For the nickel cylinder, cooling thermograms were obtained by the surface temperature, for the copper sample - by the center temperature. However, the thermal conductivity of copper is high, so the temperature of the center and surface differ slightly.

As can be seen, the cooling proceeds with low intensity, and the surface temperature at which intensive boiling regime starts (transition temperature further) are lower than critical temperature for ethanol in the case of copper and nickel cylinders. It is predictable result because the local direct liquid/solid
contact becomes possible when temperature of metal body is lower than the attainable limiting temperature of a liquid [17].

Figure 3. Experimental quenching curves obtained during cooling in ethanol +30 °C under atmospheric pressure:
1 – nickel cylinder (surface temperature), 2 – copper cylinder with gold coating (center temperature).

The picture of cooling in water is different. Note that the cooling thermograms for nickel and copper cylinders in water have a different temperature scale: the nickel sample was heated to higher temperatures of about 700 °C (Figure 4a).

For cooling in water +80 °C (ΔT_{sub} = 20 K), part of the cooling process also proceeds in the regime of stable film boiling (curves 1 in Figure 4a, b). However, the transition to intensive cooling was observed at higher temperatures than during cooling in ethanol. For the nickel cylinder it exceeds the attainable limiting temperature of water. An increase in subcooling of water by 20 K leads to an increase in the transition temperature, and a microbubble boiling regime is observed for both cylinders (curves 2 in Figure 4a, b). When subcooling is 40 K, the transition temperature is higher than 400 °C for cooling of the copper cylinder with a gold coating and higher than 600 °C for cooling of the nickel cylinder. At such temperatures, the direct liquid-solid contacts are impossible.

Figure 4. Experimental quenching curves obtained during cooling in water at different temperatures under atmospheric pressure: a – nickel cylinder (the averaged surface temperature); b – copper cylinder with gold coating. 1 – ΔT_{sub}=20 K, 2 – ΔT_{sub}=40 K.
Dependence of heat flux from temperature head were obtained for comparing during cooling in ethanol \( \Delta T_{\text{sub}} = 48 \text{ K} \) and water \( \Delta T_{\text{sub}} = 40 \text{ K} \) for nickel and copper cylinders (Figure 5). For this, the inverse heat conduction problem was solved using the ANSYS software package in a two-dimensional axisymmetric formulation. Using ANSYS Fluent, at each time step the temperature field was determined within the computational domain. The temperature field obtained at the current time step (for heat transfer coefficient satisfying the optimization condition) was used as the initial condition for calculation at the next step.

During cooling in ethanol, stable film boiling, which is characterized by low values of heat fluxes removed from the surface (not more than 500 kW/m\(^2\)), lasts for a long time for both copper and nickel cylinders. Figure 5 clearly demonstrates a significant difference in the values of the removed heat fluxes for cooling in water. Heat fluxes reach enormous values up to 5 MW/m\(^2\), which is not characteristic for film boiling. Film boiling during cooling in water with a large subcooling takes only a few seconds, after which a sharp increase in the heat flux density is noticed. Moreover, for cooling a sample with lower thermal effusivity (the nickel cylinder), the maximum of the heat fluxes is almost 2 times higher than for cooling a copper sample with high thermal effusivity. Also, this maximum is observed at higher temperature head for cooling the nickel cylinder.

![Graph showing heat flux vs. temperature head](image-url)

Figure 5. Dependence of heat flux from temperature head for: 1 – nickel cylinder in ethanol +30 \(^{\circ}\)C, 2 – copper cylinder with gold coating in ethanol +30 \(^{\circ}\)C, 3 – copper cylinder with gold coating in water 60 \(^{\circ}\)C, 4 – nickel cylinder in water 60 \(^{\circ}\)C (according to Figure 3 and Figure 4a, b curves 2).

The temperature distributions during cooling of the samples with different thermal effusivity were obtained using the ANSYS software package. Moments when the maximum heat fluxes are removed from the cooled surface are of interest, since at this time a large difference in the temperature values of the center and surface is possible. Figure 6 shows the temperature fields in the central section of the cylinders at times of the maximum heat flux removing. For clarity, parts of cooling thermograms for those temperature fields are presented on the left in Figure 6.

For the copper cylinder, the temperature field is more uniform, the difference between the center and surface temperature is about 40 \(^{\circ}\)C (Figure 6a). For the nickel cylinder, this difference is more than 150 \(^{\circ}\)C (Figure 6b), a strongly warmed center and a sharp decrease in temperature near the surface are observed.
Figure 6. The temperature field obtained at the time of intensive cooling during cooling in water +60 °C of: a – copper cylinder with gold coating; b – nickel cylinder. t₀ – time of the maximum heat flux removing, corresponding to the temperature field on the right.

In addition, temperature gradients are more pronounced along the axial location than at the radial, which was also noticed by Indian researchers for quenching samples in a mineral oil [11]. These results confirm the need for temperature measurements in low thermal conductivity samples not only in the center, but also on the surface.

4. Conclusion
The experiments confirm an existence of an extremely high intensity heat transfer process in water with large subcoolings \( \Delta T_{\text{sub}} \geq 20 \) \( K \) for cooling low and high thermal conductivity cylinders made of copper and nickel, while the transition temperature was higher than the critical temperature for water. For water subcooling \( \Delta T_{\text{sub}} = 20 \) \( K \), the intensive heat transfer regime was observed at temperature higher than critical only for cooling the sample with rather low thermal effusivity (Ni). Thermograms demonstrate that cooling processes are more intense for cooling in water, a microbubble boiling regime in ethanol was not observed at all. Although, as evidenced by the results of [18], in ethanol it is possible to achieve a transition to the intensive regime, but with very high subcoolings, when the ethanol has an extremely high viscosity. The emergence of microbubble boiling is influenced by many factors, such as the properties of the cooling liquids (heat of vaporization, surface tension, viscosity) and its subcooling [19], as well as the thermal effusivity of the cooled samples. An approximate physical model proposed in 2018 [20] provides explanations for mechanisms of intensive heat transfer and allows predicting the transition temperature with acceptable accuracy. To correct and verify the model, there is a need for further experiments for cooling high-temperature samples with various properties and roughness.

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