Transient Nucleate Boiling and Convection Processes Taking Place in Nanofluids that Contain Silver Nanopartiles

G.V. Kovalenko*, N.I. Kobasko#
*ITTF NANU, Kyiv. Ukraine, # PhD, Fellow of ASM International, ITL, Kyiv, Ukraine

Abstract: *The paper discusses transient nucleate boiling and convective processes taking place during cooling steel probes in nanofluids that contain silver nanoparticles. It is shown that nanoparticles in water suspensions cannot increase radically cooling rate of steel probes due to cooling in water is rather intensive that tends to be ideal in condition when any film boiling is absent. Nanoparticles can be used for increasing critical heat flux densities and convective heat transfer coefficients during quenching. In some cases nanoparticles accelerate formation of surface insulating layers during quenching of steel parts that decreases initial heat flux densities resulting in eliminating any film boiling process. It is recommended to use nanofluids for deep cold treatment of tool steel to increase radically their wear resistance. The paper can be helpful in further investigation of nanofluids and can be used for new technologies development.*

I. INTRODUCTION

Suspensions which contain solid particles of a size less than 100 nm (nanofluids) attracted attention of scientists when it was discovered that some nanofluids have abnormally high thermal conductivity at low volume fractions of solids [1]. This feature, as well as their other properties, has made use of nanofluids promising in chemistry, electronics, medicine, energy, in life while protecting the environment.

In [2] it was shown an increase in the thermal conductivity (for 14 - 18%) when the concentration of copper and iron particles (size 10 nm) was increased by 0.6%. The presence of 2.4% of nanoparticles, consisting of 70% Al and 30% Cu, leads to an increase in thermal conductivity 2.25 times. Thus, adding nanoparticles to one phase liquid increases significantly heat transfer in it [3,4]. The researches found the increase the heat transfer coefficients and increase the critical heat flux densities during boiling in nanoliquids. In [5] it is reported that the addition of nanoparticles TiO_2 and Al_2O_3 to a boiling water can increase the heat transfer coefficient in 4 - 5 times.

However, it should be noted that there are two groups of experimental data concerning the heat transfer exchange. At small concentration of nanoparticles in nanoliquid is observed increase of heat transfer for 20 - 40%, while at higher concentrations is observed decrease of the heat exchange by 10% - 30%. Some experiments with nanoparticles CuO in water [6] showed increase heat transfer during nucleate boiling and increase the critical heat flux densities along with increase of the mass concentration of nanoparticles to 1%. At high concentrations of nanoparticles in a nanoliquid the heat exchange gets worse.

Also, some researchers have not received the expected increase in thermal conductivity after adding nanoparticles to a liquid [6]. Classical models of suspensions (Einstein, Nielsen) poorly predict the viscosity of nanofluids. It should be noted successful attempt [7] to explain certain features of the film boiling in nanofluids, particularly by nanoparticles accumulation on the heated surface, and taking into account the thermophorestic and Brownian diffusion mechanisms.

In the literature there are rare descriptions of experiments with nanoparticles of noble metals. Silver nanoparticles themselves tend to agglomerate. To increase the stability of suspensions with such particles, it has been using a variety of coatings. These coatings (often depending on the methods for preparing nanoparticles) create surface charges at the nanoparticle surface that leads to a difference in properties of such nanofluids [3].

Maybe this is the reason of the difficulties in generalizing the characteristics of nanofluids. Often one can see lacking repeatability of obtained data during experiments with nanofluids.

The subject of research in the field nanofluids is the presence (or absence) of solid particles of their critical size, the impact of the physical characteristics of the solid phase on specific effects. In this regard, technically challenging, but promising is the definition of the properties of suspensions of metal nanoparticles.

In the investigation of the critical heat flux densities, it is shown that the addition of nanoparticles to the liquid results in their maximizing to 300 - 450%. Promising properties of nanofluids raise questions about the prospects of their applications in other fields of heat engineering, for example, in heat pipes technique or metal quenching.

In view of the foregoing, it is interesting to study transient nucleate boiling and convection within the wide temperature range including the temperature of transition from film boiling to nucleate boiling upon cooling of the heated metal probe in a suspension containing silver nanoparticles.

II. PROBE FOR TESTING NANOFLUIDS

For experiments was used a steel cylinder with the following dimensions: cylinder diameter - 9.9 mm, height of the cylinder - 28.0 mm, the radius of curvature between the cylindrical surface and the end - 0.1 mm. Along the cylinder axis in the probe at a half depth of its height 1 mm diameter a channel was drilled. In channel was pressed chromel-alumel thermocouple. The hot junction of the thermocouple was welded to the bottom of the channel by contact welding. Simultaneously, the additional thermocouple was welded to the surface of the probe on its middle. The thickness of the thermocouple with its stainless case was 1 mm. Thickness of stainless tube was 0.1 mm. The cold junctions of both thermocouples were thermo-stabilized. The probe was heated to 500°C in tube electrical furnace to provide mostly transient nucleate boiling process than developed film boiling. After sufficient holding the probe in the electrical furnace, that provided smooth temperature through all length of cylindrical probe, it was immersed into tested nanoliquid. Thermocouple signals, measuring the temperature in the center of the cylinder and its middle surface, were recorded by PC via analog-digital device. The surface thermocouple, due to its low thermal inertia, showed exact start cooling time in nanoliquid. For each experiment, temperature field in the probe was calculated using simplified method of calculation which is described in [8]. In the present study the next nanofluids were used:

-- Nanofluids with silver particles formed in the process of biosynthesis were named as the green (Ku1) and (Ku2). The method of their preparation was proposed at the Faculty of Physics and Biology at the University of Alabama, Huntsville (USA) [9]. Nanofluids Ku1 contain silver particles with a size of 422 nm in an amount of 38 ppm. Nanofluids Ku2 contain silver particles with a size of 439 nm in an amount of 43 ppm

-- Collargol 2% solution.

-- Distilled water (for comparison).

It should be noted here that Tensi (Tensi, 1992) considered the four heat transfer modes taking place during quenching the cylindrical probe in liquid media [10]. They are:

1. The area of nucleate boiling moves up along the probe surface replacing film boiling (see Fig. 1)).

2. First, film boiling takes place throughout the entire probe surface area. At a certain point in time, nucleate boiling replaces simultaneously the film boiling and then convection heat transfer replaces nucleate boiling that is well known as a classical heat transfer mode [10,11].

3. Some local areas of the probe surface are covered by the vapor blanket, while at the same time, other areas experience nucleate boiling. These local areas do not move and are reason for big distortion of steel parts during quenching in liquid media.

4. Film boiling and nucleate boiling change each other periodically in all surface area.

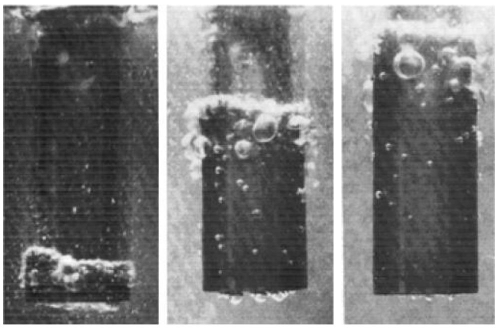

Fig. 1: Wetting process of a cylindrical Cr-Ni steel sample being quenched in water at 60°C without agitation [11].

To already existing four types of heat transfer modes should be added the 5th type of heat transfer that considers cooling without presence of any film boiling process. This type of heat transfer mode was carefully investigated by French (French, 1930) [12]. It was shown by author [13] that 5th type of heat transfer mode is intensive cooling taking place in condition $Bi \to \infty$. It is worth to consider the 5th type of heat transfer mode comparing it every time with the obtained experimental data to see what kind of heat transfer mode was taking place during experiment and to know what was happening on the surface of probe during its quenching.

III. RESULTS OF EXPERIMENTS AND COMMENTS

Figure 1 shows surface and core cooling curves versus time during cooling cylindrical probe 9.9 mm diameter in distilled water.

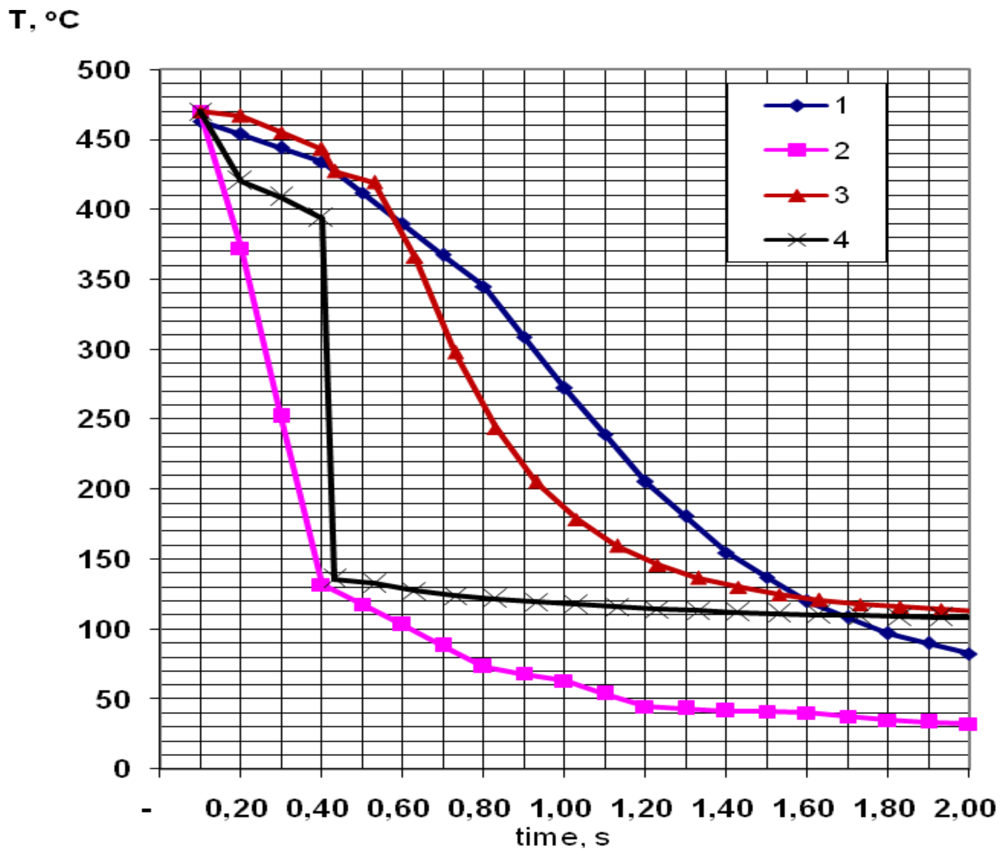

Fig. 2: Cooling the heated steel sample 9.9 mm diameter in distilled water: 1, 2 - experiment; 3, 4 - calculation [8]; 1 - temperature in the center of the cylindrical sample; 2 - temperature measured by the surface thermocouple.

As seen from Fig. 2, experimental cooling curves 1 and 2, according to Tensi’s classification, belong to the first type of heat transfer mode when nucleate boiling and convection move from bottom to upper end of the probe (see Fig. 1). Calculated cooling curves 3 and 4 belong to second type of heat transfer mode when first the film boiling takes place throughout the entire probe surface area and then at a certain point of time the nucleate boiling starts. Duration of film boiling is only 0.3 sec and initial temperature for transient nucleate boiling process is approximately 400°C. So, calculated data can be considered as the 5th type of heat transfer mode needed for comparison when quenching probes in nanofluids. Quenching in nanofluids, in many cases, eliminates film boiling completely. In this case, obtained experimental data should be compared with the mentioned above the 5th type of heat transfer mode to see what is happening on the quenched surface of the probe. No one should compare the first and second heat transfer modes hoping to get useful conclusion. As is said, apple to apple should be compared to get useful conclusions. Taking this approach and consideration into account, let’s see what are cooling curves during quenching in nanofluids (see Fig. 3).

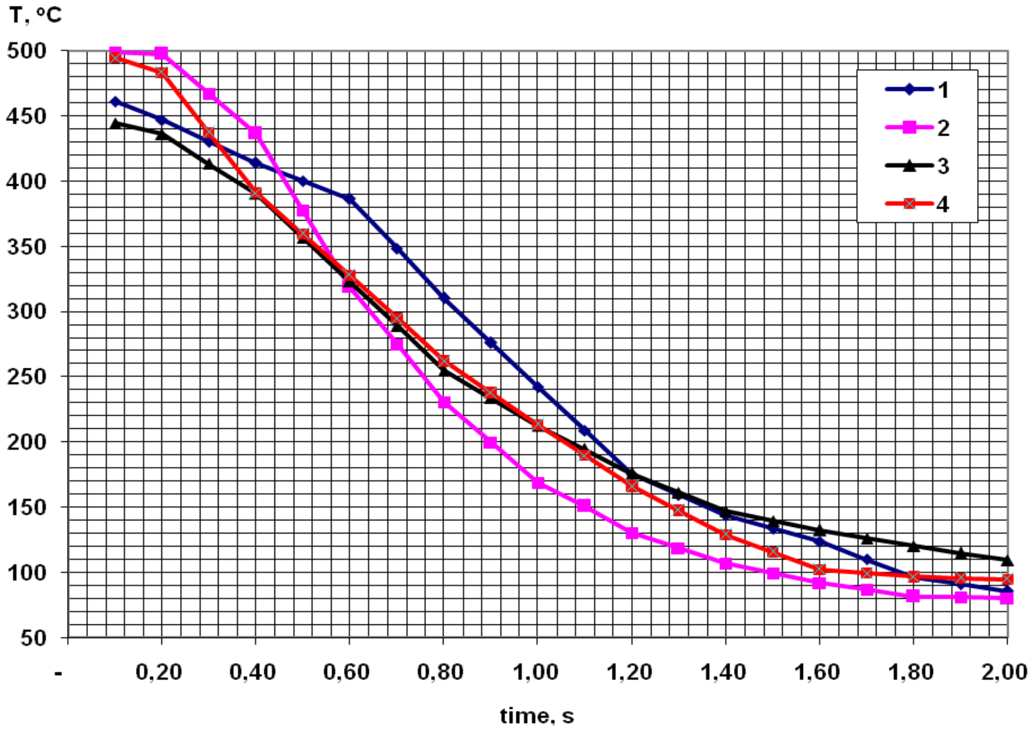

Fig. 3: Core cooling curves during quenching cylindrical probe 9.9 mm diameter in nanoliquids: 1 is distilled water; 2 is 2% water solution of collargol; 3 is nano- suspension of silver (Ku2); 4 is nano- suspension of silver (Ku1).

Let's compare obtained experimental core cooling curves with the second type of heat transfer mode shown in Fig. 2 within the interval of temperatures 150^{o}C – 350^{o}C to exclude film boiling process. The results of experiments concerning the nucleate boiling process measured by the thermocouple located at the center of cylindrical probe are presented in Table I.

TABLE I

Cooling time from 350^{o}C to 150^{o}C and average cooling rate within the given temperatures

Cooling curve name	**Cooling time from 350^{o}C to 150^{o}C in sec**	**Average cooling rate within temperatures 350^{o}C – 150^{o}C**
Core cooling curve from Fig. 1. Calculation	0.55	364
Distilled water	0.66	303
Collargol	0.56	357
Ku2	0.84	238
Ku1	0.73	274

Note: No film boiling within temperature interval 150^{o}C – 350^{o}C.

As one can see from Table I, the average cooling rate for water and collargol is almost the same and is equal to 364^{o}C/s and 357 ^{o}C/s correspondently. Cooling rates for Ku1 and Ku2 nano- suspensions are significantly reduced (see Table I). It can be explained by creation of surface insulating layers during quenching of probe. As known, cooling rate is evaluated as [14]:

$$v = \frac{aKn}{\left(1 + 2\frac{\delta}{R}\cdot\frac{\lambda}{\lambda_{coat}}\right)K}\left(T - T_s\right) \qquad (1)$$

During quenching cylindrical probe in colllargol solution on its surface very thin insulating layer was formed that decreased insignificantly cooling rate of cylindrical probe. Essential decrease the cooling rate during quenching in nano- suspesions of Ku1 and Ku2 is explained by coagulation of suspensions resulting in creation of insulating layer with the larger thickness (see Eq. 1). Note, the average cooling rate for the first type of heat transfer mode during quenching in distilled water is 303^{o}C/s that is larger as compared with cooling probe in Ku1 and Ku2 nano – suspensions and lesser as compared with the second type of heat transfer mode that can take place during cooling in distilled water. This cooling rate is not taken into account because last belongs to completely different heat transfer mode.

Due to above consideration, it is worth to know surface tension and electrical resistance of nanoliquids depending on silver nanoparticles concentration. Such information is provided by Fig. 4 and Fig. 5. Fig. 3 shows the dependence of the surface tension of nanofluids versus silver concentration. Surface tension coefficient was determined by the height of liquid rise *h* in a glass capillary with a cylindrical channel of radius *R* = 0.5 mm using Eq. (2) [15]:

$$\sigma = 0.5\rho_l gRh \qquad (2)$$

where g is gravitational acceleration; ρ_l is density of liquid.

It was noticed reduction in surface tension with increasing nanoparticle content. The relative reduction of σ was from 0.787 to 0.727.

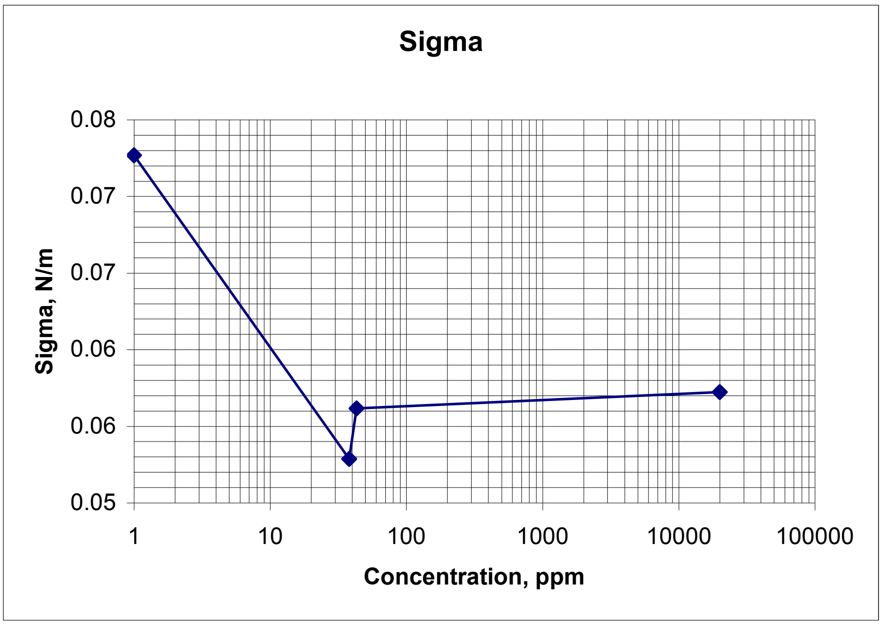

Fig. 4: Surface tension versus concentration of silver in nanofluid: ppm is particles per million.

Similar function of electric resistance versus concentration silver nanoparticles in nanoliquid is shown in Fig. 5

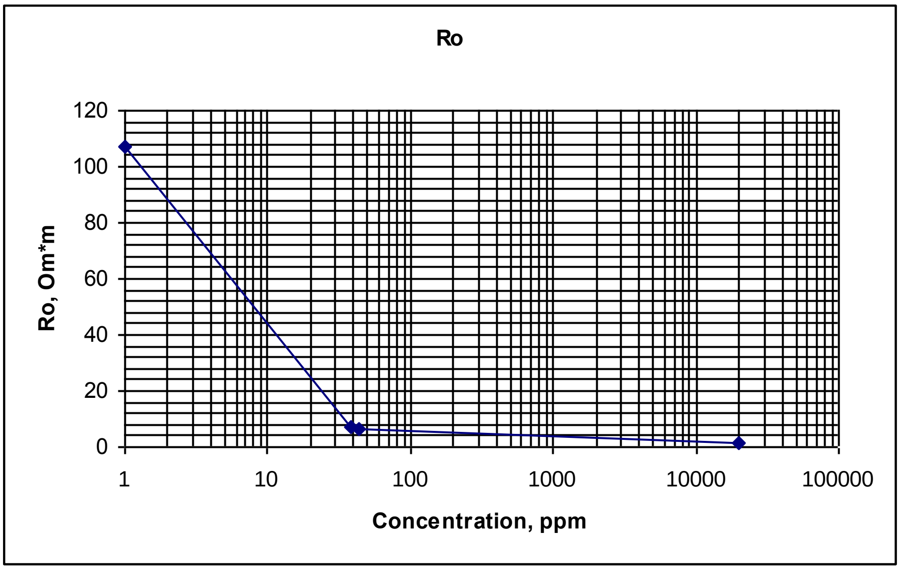

Fig. 5: Electrical resistance versus concentration of silver in nanofluid: ppm is particles per million.

IV. NUCLEATE BOILING AND CONVECTION PROCESSES ANALYSIS

To start nucleate boiling process, the randomly appeared in the overheated liquid tiny bubbles must be larger its critical value R_{cr}. According to [16, 17] the critical radius is evaluated by Eq. (3):

$$R_{cr} = 2\sigma T_s / \left(r^{\bullet}\rho_g\left(T_W - T_s\right)\right) \qquad (3)$$

Here T_s is saturation temperature; T_w is wall temperature; r is latent heat of vaporization; ρ_g is vapor density.

Surface roughness affects the number of possible centers of boiling also. Reduce of surface tension leads to decrease critical size R_{cr}, and consequently, increases numbers of active boiling centers that increases heat transfer.

In Ref. [16] there is an equation establishing relationship between active vapor bubbles n and liquid characteristic parameters:

$$n = 625\cdot 10^{-16}\left(\left(r^{\bullet}\rho_g\left(T_W - T_s\right)/\sigma T_s\right)\right)^3 \qquad (4)$$

With such a strong dependence n on surface tension, its reduction for 21% increases the number of boiling centers for 77%.

Similarity equation for heat transfer during nucleate boiling in accordance with [15] can be expressed as (5):

$$Nu = 0.082 K_z^{0.33} K_q^{0.7} \Pr_l^{-0.45} \qquad (5)$$

Here Nu is Nusselt number; K_z and K_q are dimensionless numbers; $\Pr_l$ is Prandtl number of a liquid; As a geometrical size the capillary constant δ is used:

$$\delta = \left[\sigma / g\left(\rho_l - \rho_g\right)\right]^{0.5} \qquad (6)$$

$$Nu = \frac{\alpha}{\lambda_l}\delta \qquad (7)$$

$$K_z = \frac{c_p\rho_l\left(T_w - T_s\right)R_{cr}}{2\delta r^{\bullet}\rho_g} \qquad (8)$$

$$K_q = \frac{q\delta^2}{\rho_g r^{\bullet} a_l l_x} \qquad (9)$$

$$l_x = \frac{c_p\sigma\rho_l T_s}{r^{\bullet}\rho_g} \qquad (10)$$

Here α is heat transfer coefficient; λ_l is thermal conductivity of a liquid; c_p is specific heat capacity of a liquid; q is heat flux density; a_l is thermal diffusivity of a liquid.

Analysis of considered equations (3), (5) – (10) shows stronger dependence α from thermal conductivity of liquid than rather from σ (see Eq. (11) and Eq. (12)):

$$\alpha \propto \lambda^{1.45} \qquad (11)$$

$$\alpha \propto \sigma^{-0.335} \qquad (12)$$

Proceeding from equations (3), (5) – (12), the decrease of surface tension for 21% – 28% as a result of nanoparticles cannot be a soul factor of increasing heat transfer since it can increase α only for 8% – 11%.

There are some other factors cardinally affecting cooling intensity during quenching.

In Ref. [9] there is an equation establishing relationship between critical heat flux density and physical properties of a liquid during boiling on the horizontal surface

$$q_{cr} = 0.149\, r\, \rho_g^{0.5}\left[\sigma g\left(\rho_l - \rho_g\right)\right]^{0.25} \qquad (13)$$

$$q_{2cr} = 0.05\, q_{cr1} \qquad (14)$$

Based on experimental data published in Ref. [9] and [17], we can see weak effect of silver nanoparticles on increasing intensity of cooling during transient nucleate boiling process. The same conclusion was widely discussed in the recently published paper [13].

Based on published data [8, 18, 19], authors of current paper believe that nanoparticles can affect initial and critical heat flux densities and by this way increase cardinally intensity of cooling during quenching processes. There are two ways to do such things:

- Creation of thin insulating layer on the surface of metals during their quenching [18, 19].
- Multiplying electrical forces in double electrical layer taking place during hardening of metal components [20 -22].

Unfortunately, nobody considered direct effect of electrical forces in the double electrical layer on increasing critical heat flux densities

As for increasing convective heat transfer coefficients and increasing intensity of cooling during hardening of metal components in liquid media via exploring nanoparticles, one should expect a visible progress since metal nanoparticles can significantly increase the thermal conductivity of nanofluids (see Eq. (15)) [22]:

$$\alpha_{conv} = 0.135\lambda_l \left(\frac{g\beta_l \Delta T}{a_l \nu} \right)^{1/3} \qquad (15)$$

Here β_l is volumetric expansion coefficient of a liquid; ΔT is temperature difference; is kinematic viscosity of a liquid.

Metal nanoparticles can be used for increasing thermal conductivity of nanofluids used as a quenchant during cryogenic treatment of tool steels. This area of investigations is very promising because deep cold treatment cardinally increases wear resistance of various tool steels (see Table II) [23, 24].

TABLE II

The improvement in wear resistance of various tool steels after deep cold treatme nt [23, 24].

Steel (AISI)	**Improvement in wear rate (%)**
D2	817
S7	503
52100	420
A10	264
M1	225
H13	209
M2	203

Other improvements of steels after cryogenic treatment are discussed in two books [19, 24].

V. DISCUSSION

It is important to know how intensively the cylindrical steel probe was cooled in distilled water. For this purpose we'll consider the extreme situation. According to [17] the first critical heat flux density is equal to 5.9 MW/m^2 when underheat is 80°C. According to [13] overheat of water at the beginning of transient nucleate boiling process for cylindrical steel probe 9.9 mm diameter is 32°C. Maximal possible heat transfer coefficient during nucleate boiling can be $\alpha = \frac{5.9\,MW/m^2}{32^o} = 184375 W/m^2K$. According to [25, 26] for steel containing 1% carbon thermal conductivity is 43 W/mK and thermal diffusivity at room temperature is 11.72 x 10^{-6}m^2/s and at 250°C it drops below 10.5 x 10^{-6}m^2/s. Kondrat'ev form coefficient for cylindrical probe 9.9 mm diameter is 4.02 x 10^{-6}m^2. Conventional Biot number $Bi = \frac{184375W/m^2K}{43W/mK} \cdot 0.00495m = 21.22$ Generalized Biot number Bi$_V$ = 0.346 x 21.22 = 7.34 that provides Kondrat'ev Kn = 0.907. Let's see what is difference in cooling rates obtained by experiments (see Table I) and calculated data in the extreme condition. Average cooling rate for interval temperatures 150°C – 350°C corresponds to temperature 250°C. Then calculated cooling rate can be evaluated as [27]:

$$v = \frac{aKn}{K}\left(T - T_s\right) = \frac{10.5\times10^{-6}\times0.907}{4.02\times10^{-6}}\times\left(250-100\right) = 355^o C/s$$

According to our experiment and calculations the average cooling rates for temperature interval 150°C – 350°C (second heat transfer mode) are 357°C/s and 364 °C/s. Difference between experimental and calculated data is less than 1%. Due to different types of heat transfer modes, when comparing the second types of heat transfer modes, difference is 14% (see Table 1). It means that nucleate boiling process provides intensive cooling if any film boiling is absent [28]. Nobody can increase cardinally cooling rate during transient nucleate boiling in water by adding nanoparticles to nanofluids. Nanoparticles can affect essentially critical heat flux densities and convective heat transfer taking into account equations Eq. (1) and Eq. (15) [28].

VI. CONCLUSIONS

1. Nanofluids containing metal nanoparticles are promising as the quenchants for heat treating industry since they increase several times convective heat transfer coefficients and increase significantly critical heat flux densities.

2. The main attention should be paid to investigation the effect of metal nanoparticles on increase critical heat flux densities and convective heat transfer coefficients during quenching since nanofluid cannot affect significantly transient nucleate boiling process.

3. In case, when production of nanofluids is stable, their application is possible in the heat pipes, since the intensification of heat transfer due to the presence of nanoparticles in the liquid prevails over the decrease in the surface tension, necessary for the movement of liquid in the wick systems.

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