COOLING CHARACTERISTICS OF THE WATER BASED NANOFLUIDS IN QUENCHING

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Izvorni znanstveni rad/ Original scientific paper

ABSTRACT:
Nanofluids are the new quenching media with colloidal suspensions of nanoparticles in base fluids, offering new possibilities for enhancement of the heat transfer characteristics. The recent investigations show that the addition of the metals oxides particles, such as CuO, Al₂O₃ and TiO₂, or graphite particles result in better cooling abilities and higher impact toughness and smaller dimension changes of steels, compared with the traditionally used quenching media.
This paper presents the preliminary results of the investigation of cooling characteristics of water and 30 % Polyalkylene Glycol (PAG) particle water solution based fluids. The investigated fluids were prepared by the addition of Al₂O₃ and TiO₂ powders with different particle sizes - from less than 50 nm to 100 μm and concentrations between 0.1 and 0.4 %wt.
The cooling curve measurements have been conducted by IVF smart quench system using stainless steel probe with 12.5 mm diameter. Derived from these data, the cooling rate vs. temperature and the heat transfer coefficients have been determined using the IVF’s SQintegra software. Alumina particles suspended in water based polymer solution caused shortening of the full film phase. Smaller size particles exhibit greater effect on the start of nucleate boiling phase than solutions with larger particles or without any. This results in shorter quenching process even though the maximum cooling rate is similar for all the media. At lower concentrations of TiO₂ nanoparticles dispersed in water there is no significant difference between cooling curves for deionized water and TiO₂ nanofluid.

Keywords: nanofluid, quenching, heat transfer coefficient
1. Introduction

Quenching still remains one of the most widely used industrial heat treatment processes. As the work piece is heated to austenitizing temperature it has to be rapidly cooled during which phase transformations, heat transfer and stress strain occur. Physical properties of the quench hardened work piece depend on the cooling rate. In order to achieve specific properties different cooling rates are applied for different materials. For that reason various quenching media are used, such as water and water solutions, polymer solutions (ethylene and glycol), oils, molten-salt baths, fluidized beds and compressed gases [1].

New type of quenching media has recently been developed and today world’s leading scientific institutions are conducting research to find out more about the possibilities of this new fluid. It is called nanofluids. Nanofluids are colloidal suspensions of stably dispersed nanoparticles in base fluids. The term was first introduced by Choi [2] in 1995. Nanoparticles dispersed in base fluid can be metals oxides, such as CuO, Al₂O₃ and TiO₂, different carbides or nitrides, but pure metal particles as well [2, 8]. Also, there is a wide use of graphite particles and carbon nanotubes due to their exceptional thermal conductivity. The average size of particles should be less than 100 nm. For base fluid, most commonly, deionized or fresh water are used. As part of the research at QRC polymer solutions and quenching oils have been used as base fluid. Today there is still no confirmed theoretical background that would explain all of the nanofluid properties. That is why several impact factors will be investigated regarding the thermal conductivity and heat transfer coefficient of nanofluids. First the preparation of fluid, since the time of sonification with ultrasonic vibrator or bath effects the fluid properties. Nanoparticles of various sizes will be investigated, as well as mass concentration of particles in the fluid. As nanofluid temperature influences heat transfer coefficient, experiments with nanofluid of same particle size and concentration but different temperature will be conducted. Driving force of this research is higher heat transfer coefficient and thermal conductivity of nanofluids compared to the one of base fluid. There are couple of theoretical reasons for such enhancements: i) high specific surface area, ii) Brownian motion, iii) thermophoresis and iv) diffusiophoresis.

The nanofluids technology is of interest to a variety of industry branches due to a wide range of possible applications such as heat transfer applications, automotive, electronics and biomedical applications. Some of the specific areas include: industrial cooling that could result in energy savings and emission reduction, nuclear energy production by improving the performance of reactor cooling system, nanofluids as coolants in the process of geothermal power extraction and in radiators for automotive industry, resulting in less drag and fuel consumption. Also, the possibility of microchip cooling and drug delivery using nanofluids makes them an interesting research field with findings valuable to various scientific fields.

Several articles concerning behaviour of nanofluids during quenching have been published so far. Most of them were encouraged by the investigations of thermal conductivity that showed extremely high thermal properties compared to base fluid. The research on nanofluids began at Argonne National Laboratory as a part of Advanced Fluids Programme whose main objective was to develop advance energy transmission fluids. Application of nanofluids as quenching media in quenching process was first experimented at 2004 [5]. High temperature copper spheres were quenched in alumina nanofluids in order to investigate the effect suspended nanoparticles have on film boiling heat transfer characteristic. Even though high particle concentrations were used, from 5 to 20 vol%, nanofluids showed lower boiling heat transfer rate than pure water. More important, the experiments conducted on a previously quenched sphere resulted in a much more rapid process compared to quenching of a clean sphere. The researchers’ hypothesis was that the nanoparticles deposited on the probe surface increase the wettability area, thus greatly shortening film boiling phase. This phenomena is in correlation with previous research of nanofluid boiling properties,
which indicated the deposition of particles causing rougher surface and consequently, higher critical heat flux. Next round of experiments was conducted in water based carbon nanotube fluids. Quenching of nickel-plated copper sphere was performed due to the fact that copper spheres can be assumed thermally lumped. Carbon nanotube's (CNT) superior thermal properties caused higher critical heat flux, boiling heat transfer rate and Leidenfrost point (minimum heat flux - MHF) compared to water. Particle deposition on the surface was also experienced. Research at MIT [6] observed quenching of steel and zircaloy spheres in nanofluids with 0.1 %Al2O3, silica and diamond nanoparticles. The results showed that film boiling heat transfer in nanofluids and water is almost identical. Deposited nanoparticles had far greater impact on CHF and MHF, while diamond, unlike alumina and silica particles, had no significant effect. In the next step quench front speed during rodlet quenching was compared after each test, which proved that deposited nanoparticles increase cooling rate and quench front speed. Due to large variations in thermal conductivity measurements of nanofluids, a benchmark study was proposed [8]. A round-robin test among 34 institutions worldwide showed that nanofluids do not experience abnormal thermal conductivity enhancement, but the results of experiments are within Maxwell's theory. Higher particle loading and aspect ratio caused increase in nanofluid conductivity.

2. Heat transfer coefficient

Quenching process is a complex thermodynamic and fluid dynamic problem consisting of several different phases. If the quenching media is vaporizable liquid, four modes of cooling can be observed: i) shock film boiling, ii) full film boiling, iii) nucleate boiling and iv) convection cooling mode, Figure 1. Each of these modes is characterized by different surface heat transfer coefficient (HTC) and cooling rates.

![Figure 1 Four modes of cooling at quenching process](image)

Because of that temperature dependant HTC is determined using some simplifications: the probe is a radially symmetric body and heat transfer problem is considered to be one-dimensional (1-D), dependant only on the probe radius and not length. Calculated HTC is valid for a cross-section at thermocouple position, Figure 2.
There are two main approaches to calculate the surface HTC. First one discussed is lumped heat capacity method which presumes uniform probe temperature with the heat loss equal to the decrease in probe internal energy (1).

\[ Q = hA(T_p - T_q) = -c_p \rho V \frac{dT_p}{dt} \]  

where \( h \) is the surface heat transfer coefficient, \( A \) is the surface area of the probe, \( T_p \) and \( T_q \) are probe and quenchant temperature, \( c_p \) is the specific heat and \( \rho \) is the specific density of the probe material, \( V \) the probe volume and \( dT_p/dt \) the cooling rate of the probe. Since probe temperature and surrounding quenchant temperature are considered uniform, equation (2) for heat flux on the probe surface, \( q \) can be derived:

\[ q = h(T_p - T_q) = \left( c_p \rho \frac{V}{A} \right) \frac{dT_p}{dt} \]  

As lumped heat capacity method directly depends on cooling rate calculation, accuracy and sampling frequency of the data acquisition system are crucial for correct HTC calculation. Also, smaller size probe made of higher thermal conductivity material would give more reliable results of time and temperature dependant HTC. From equation (2), derived is HTC \( h \) (3)

\[ h = -c_p \rho \frac{V}{A} \left( \frac{dT_p}{dt} - T_p - T_q \right) \]  

Second method used is the inverse heat conduction problem. In this case Fourier's partial differential equation (4) has to be solved

\[ \left( k \cdot \frac{\partial^2 T(r,t)}{\partial r^2} \right) + \left( \frac{k}{r} \cdot \frac{\partial T(r,t)}{\partial r} \right) + Q = -c_p \rho \frac{\partial T(r,t)}{\partial t} \]  

where \( r \) is local radius coordinate, \( t \) is time, \( k \), \( c_p \) and \( \rho \) are probe material heat conductivity, specific heat and density, while \( Q \) is latent heat. To solve the equation initial and boundary conditions have to be set. The initial condition for probe temperature is a uniform distribution through entire probe, and the temperature is set to be austenitizing at initial time step (5). Boundary condition for heat transfer (6) is dependent on probe conductivity and radius temperature distribution as well as HTC at probe surface

\[ T(r,t = 0) = T_a \]  

\[-k \frac{\partial T}{\partial r} = h(T)[T(R,t) - T_q] \]
3. Experimental method

To evaluate heat transfer characteristics of different quenching media, a number of experiments in accordance with ISO 9950 standard for oils and ASTM D 6482 standard for polymer solutions were conducted. The media of interest for quenching process are fresh water, water based polymer solutions and quenching oil. Small amounts of added nanoparticles influences heat transfer characteristics of base fluids so the intention of this research is to observe the effects micro and nanoparticles on quenching process. Particle mass content, quenchant temperature and sonification time are modified.

3.1 Probe

Standard test probe used in this set of experiments, Figure 3, is in compliance with international standard ISO 9950. Probe material is Inconel 600 and thermocouple used is standard Type K (NiCr/NiAl), 1.5 mm diameter. Probe is a part of IVF smart quench system consisting of data acquisition unit, certified standard test probe, furnace and software, Figure 4. Data acquisition unit and software have the capacity of gathering 100 samples per second. This frequency is considered to be high enough to register all of the rapid changes that occur during quenching process.

Since particle deposition on probe surface during quenching was previously experienced and it was known to change the surface wettabillity [2], probe was cleaned after each test. This way it was ensured that the condition of the probe surface has minimum effect on the results of the experiment. These particle depositions are thought to be the main reason for heat flux enhancement and heat transfer characteristic modification.

3.2 Quenching media

First of all deionized water was used as a quenchant for comparison with newly developed media. Several nanofluids were suspended for the purpose of this research. All of the nanofluids were produced in 2 step method, meaning pure nanoparticles were added to the base fluid. To homogenize the nanofluid ultrasonic bath, type BRANSONIC 220, frequency 50 Hz and power 120 W, was used. Led by results from other researchers, influence of sonification time will be investigated due to the fact that there is an optimum duration of sonification at which maximum thermal conductivity of nanofluid is reached. Titanium oxide nanopowder was added to deionized water and sonified for 15 min at maximum power and frequency, long enough to obtain stable homogenous suspension. Nanoparticles used for preparation of this nanofluid were 50 nm in diameter average and particle loading of 1 g/l was selected for this set of tests. For another set of experiments Al₂O₃ nanoparticles manufactured by RioTintoAlcan, Canada with average particle size 0.9 µm and 100 µm were used. Reason for such a huge particle size difference is the fact that previous research has
shown strong size dependence of nanofluid’s cooling characteristics. To investigate the effect of particle concentration on cooling rates and quenching process, nanofluids of various particle concentrations were suspended: 1 g/l, 2 g/l and 4 g/l which roughly corresponds to 0.1 %wt, 0.2 %wt and 0.4 %wt. For comparison, water based polymer solution was also tested. Polyalkylene glycol produced by Ucon Quenchant E was added to water. Water based polymer solutions show specific behaviour during quenching process. At a certain temperature solid polymer film is formed around the probe. Since nanoparticles are uniformly suspended in the solution, effect of finely dispersed oxide particles inside the polymer film is observed. Since enhanced thermal properties of metal oxides are the main reason these particles are added to the solution, it will be interesting to see how they effect polymer film conductivity, period of film formation and total quenching time.

4. Results

As many researchers have found, particle deposition has a great impact on cooling characteristics. To observe this phenomena four quenching test were conducted. Using standard IVF probe, cooling curve for deionized water at 20°C was first recorded. This curve would use us as a reference guide to compare the effects of different impact factors. Second, nanofluid with TiO₂ (1 g/l) was prepared using ultrasonic bath and its cooling curve was recorded. When comparing these two experiments no significant differences can be derived, Figure 5. There is a shift to the right for nanofluid quenching curve caused by an extra second it was necessary to bring the probe from the furnace to the cooling tank. Maximum cooling rate and total process time are almost identical.

After the quench test in nanofluid, the probe was cleaned and prepared for a next set of experiments. Again quenching in deionized water was conducted first and after that in nanofluid of same particle loading. In both of those cases maximum cooling rates are higher than in the first set of curves and they reach the maximum value much faster, Figure 6.
Figure 6 Cooling curves and rates for 1st (black line) and 2nd (blue line) set of tests in a) water and b) TiO$_2$ nanofluid

From these observations the effect of particle deposition on the probe surface can be seen, even though the probe was cleaned after the first set of tests.

For the next quenching media, water based polymer solution with and without micro particles was used. This time Al$_2$O$_3$ powder was suspended in fresh water with 30 \%PAG. Two different size powders were used with average size of particles 0.9 and 100 \( \mu \)m. Particles of this dimensions fall within the micron size range, but the main reason for their comparison was to show the effect of particle size on the cooling characteristics of fluids. The fluids were prepared by mechanical stirring and the data were recorded with UT71C/D/E  Interface Program Ver: 1.10 software.
When comparing cooling curves of probe quenched in 30 %PAG water based solution with and without addition of particles, several conclusions can be made. Quenching in fluid solutions results in shorter cooling time, as seen in Figure 7 diagrams a) and b), comparing how long it takes for the probe centre to reach 200 °C.
The biggest difference between polymer solution with and without particles is in the early stage of the process, when full film boiling mode occurs. The cooling rate ascends much faster in fluids with particles in the first couple of seconds, but the maximum rate reached in the initial phase is very similar for all the cases, Figure 7 e) and f). As the probe temperature drops to around 500 °C, nucleate boiling phase begins. This phase is characterised by much higher cooling rates than the full film phase, reaching the maximum just under 60 °C/s, three times the maximum rate of film phase. When particles are dispersed in polymer solution this phase start sooner, Figure 7 c) and d). Even though some differences in cooling curves between various particle concentrations can be seen, they are too small to make any significant impact on the process. The maximum cooling rate was achieved when quenching in polymer solution without particles.

As previously stated particle size is a very important impact factor. Specific surface area increases as the average particle size decreases, causing better heat transfer characteristics. This can be observed through the comparison of cooling curves for fluids with same particle loading, but different size of particles. At smaller loadings, 1 g/l, there is very little variation between quenching in fluids with particles 150 µm and 0.9 µm, Figure 8 a), c) and e). At 4 g/l loading the initial phase of cooling is almost identical for both fluids, Figure 8 b), while fluids with smaller size particles show faster
occurrence of nucleate boiling phase, Figure 8 d). Also, the maximum cooling rate is reached sooner, Figure 8 f), resulting in shorter cooling period to 200°C.

5. Conclusions

- Suspended nano- and microparticles effect the thermal properties of base fluid;
- Deposition of titanium oxide particles on the probe surface increases the maximum cooling rate and shortens the full film boiling phase by means of larger wettability area and disruption of fully developed vapour film;
- Alumina particles suspended in water based polymer solution shorten the full film phase and start the nucleate boiling phase sooner than polymer solution without oxide particles;
- The difference between 1 g/l and 4 g/l particle loading in polymer solution is almost neglectable;
- At higher particle contents the fluids with smaller size particles exhibit better cooling properties than suspensions with larger particles and without particles.

The future work will be focused to effect of nanoparticle size in much more detail since oxides in 10–100 nm diameter. For each of particle sizes nanofluids with various concentrations will be prepared. As one of the most important factors, the effect of quenching fluid temperature on maximum cooling rate and heat transfer coefficient will be of interest. As previous research suggests every nanofluid has optimum sonification time so this variable will be taken under account.

References