Measurements of spray water cooling heat transfer coefficients under oxide scale formation conditions

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INTRODUCTION

Spray water cooling is an important technique used for cooling in steel materials production processes, e.g. as part of the casting and rolling procedure [5,14]. Due to the high temperatures, especially after a furnace treatment, the steel surface is oxidized. A lack of knowledge about the influence of this oxide layer (scale) on heat transfer conditions may prevent a quantitative prediction of the heat transfer coefficients during water spray cooling. For predictable homogenous cooling procedures, especially the dependence of the heat transfer coefficient (HTC) on its principal parameters, the surface temperature and the water mass flow density, is required. For instance, a lower and particularly non-uniform cooling can lead to structure inhomogeneities, tensions and finally cracks. Also the plant layout and the installation of optimal spraying plans aggravate themselves by insufficient knowledge about the heat influence of the oxide scale. An optimal temperature control can be hindered at heat treatments, at hardening and tempering as well as in the outlet area after warm-rolling, depending on character of the formed oxide layer.

It is very important to know the interrelationship between oxide scale formation and heat transfer and after that to be able to predict them for different process conditions. The temperature of the metal surface is one of the decisive factors for the growth of the oxide layer, which again is determined by the heat transfer. The layer of the oxide scale forms a resistance for the heat transfer. Additionally due to the cooling process mechanical tensions arise between the oxide layer and the bulk material which lead to spallation and thus again to changes of the heat transfer conditions. Spray water cooling as an important technology used in steel industry for the cooling of materials from temperatures up to 1800K and it will be investigated in this paper.

For the description of boundary conditions for heat transport problems, the concept of the Heat Transfer Coefficient (HTC) $\alpha$ is used, which is defined through its relation to the heat flow density to the surface $q$ [W/m²]:

$$q = \alpha \cdot (T_s - T_w) .$$

For the case of spray water cooling, $T_w$ is the water temperature, while $T_s$ is the (local) surface temperature. This approach is most valuable for situations where $\alpha$ is constant, i.e. not depending on $T_s$. It can also be used for any general heat flow $q$ by using a function for $\alpha$ depending on the same parameters as $q$. In the so-called steady film boiling regime, $\alpha$ is known to be a function of the water mass flux density [3-10]. Below a specific surface temperature $T_{Le}$, the heat transfer coefficient shows a strong dependence on temperature (Leidenfrost effect [1-2,13]). In this paper, the heat transfer coefficient was measured by an automated cooling test setup (stationary method, see Figure 1 and the discussion in [4]) under oxidizing and non-oxidizing sample surface conditions. Compared to the previous state-of-the-art [3-10,12-14], an additional temperature dependency in the high temperature regime was investigated and more emphasis on the determination of the measurement precision was undertaken (which is known to be a significant problem, e.g. [4]).

Spray water cooling of steel materials introduces additional effects due to the formation of oxide layers (scale) [10,12]. The growth of the oxide scale on the investigated steel surface will be described. Heat transfer experiments under scale formation conditions will be presented and discussed.
EXPERIMENTAL PROCEDURE

The thin sheet specimens of cold rolled metals with a thickness from 1-3 mm were machined to discs with 70mm in diameter. Three thermocouple pairs, one in the middle and the others with 10 mm of distance, were spot welded to the lower side of the sample discs. The temperature measurement was carried out with Ni-CrNi thermocouples with a wire diameter of 0.5mm. The thermocouple wires were isolated by thin ceramic tubes. The experimental set-up sketched in Figure 1 was used for heating, oxidizing the samples and measurement of the temperature during spray cooling.

After installation in the furnace sample holder, the disks prepared as described above were heated up e.g. to 1000 °C under protective atmosphere. For the measurements with scale, the sample was oxidized in the furnace by supplying Air instead of Argon for a specific time and temperature. Reaching the test condition (temperature, oxidation time) the sample was moved automatically from the furnace under the full cone nozzle which takes approximately 4 seconds and subsequently cooled with spray water. The spray water mass-flux density \( V_s \) was determined experimentally and varied in the range of about 3.8±1 to 25±1 kg m\(^{-2}\)s\(^{-1}\). The water temperature \( T_w \) was approximately 18 °C. The compositions of two materials investigated, obtained by chemical analysis, are presented in Table I.

<table>
<thead>
<tr>
<th>Specimen</th>
<th>Material</th>
<th>C</th>
<th>Mn</th>
<th>P</th>
<th>S</th>
<th>Si</th>
<th>Cr</th>
<th>Ni</th>
</tr>
</thead>
<tbody>
<tr>
<td>Thermax</td>
<td>AISI 314</td>
<td>0.15</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>2.0</td>
<td>25</td>
<td>20</td>
</tr>
<tr>
<td>DC04</td>
<td>AISI 1008</td>
<td>0.03</td>
<td>0.21</td>
<td>0.009</td>
<td>0.007</td>
<td>0.02</td>
<td>0.02</td>
<td>-</td>
</tr>
</tbody>
</table>

Table I: Chemical analysis of the specimens (in wt. %).
MEASUREMENT OF THE HTC TO A NON-OXIDIZED SURFACE

For the investigation of the HTC from a non oxidized surface, heat resistant steel (Thermax, AISI 314) was used. It was chosen as the test material because surface oxidation is minimised, and the physical properties are known within the temperature range being studied. The materials were annealed at 1000 °C under protective atmosphere and consequently cooled with spray water. The cooling curves were measured at three points for each sample as described above. Figure 2 shows the cooling curves of the Thermax specimens for different water mass flux densities. In the diagram only one curve for every sample is demonstrated. The sample reaches the spray water cooling position after about 4.2 seconds. The cooling starts in the range of stable film boiling. The primary effect of the water mass flux density $V_s$ can be seen by comparing the different cooling curves. When increasing $V_s$ from 3.8 up to 25 kg m$^{-2}$ s$^{-1}$ the sample is cooled more quickly. At approximately 400-500 °C, dependent on $V_s$, the cooling curves bend. At this point the Leidenfrost temperature is reached and more rapid cooling begins (unstable film boiling).

![Cooling curve for different water mass flux densities $V_s$ (spray water cooling of the Thermax steel).](image)

Figure 2: Cooling curve for different water mass flux densities $V_s$ (spray water cooling of the Thermax steel).

Figure 3 shows heat transfer coefficient $\alpha$ in dependence on the temperature difference $\Delta T= T_s- T_w$ between surface temperature and the water temperature for different water mass flux densities $V_s$. The heat transfer coefficient was calculated analytically from the measurement data. The following equation was used:

$$\alpha \approx \frac{\rho \cdot c_p (T) \cdot d}{T - T_w} \left. \frac{\partial T}{\partial t} \right|_{z=0}$$

with:
- $\alpha$ heat transfer coefficient [W m$^{-2}$ K$^{-1}$]
- $c_p$ specific heat capacity of the sample [J kg$^{-1}$ K$^{-1}$]
- $d$ thickness of the sample [m]
- $z$ vertical coordinate (bottom: $z=0$, top: $z=d$) [m]
- $t$ time [s]
- $\rho$ mass density of the sample [kg m$^{-3}$]
- $T$ temperature measure at the lower side (U, Fig.5) [K]
- $T_w$ temperature of spray water [K]
Equation (1) is valid for thin samples such as in this case (d=1 mm) because of the small temperature difference between the cooled top and the measurement point at the bottom. The Specific heat capacity $c_p$ of the AISI 314 depending on temperature was calculated with the Thermocalc® Software based on the chemical analysis and using the TCFE3 database.

In the open literature, the HTC in the range of stable film boiling (i.e. above $\Delta T=600$K) is assumed to be independent of the surface temperature. As shown in Figure 3, for the higher spray water densities, $V_S = 18$ and $25$ kg m$^{-2}$s$^{-1}$ there is a decrease in the measured HTC even in the stable film boiling regime.

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\[ \text{Figure 3: Heat transfer coefficient towards a non-oxidised sample in dependence on temperature difference for different water mass flux densities.} \]

**MEASUREMENT OF THE HTC TO AN OXIDIZED SURFACE**

Following the initial considerations (see below and [10]) and the basic knowledge on scale properties (e.g. [11]), the determination of the influence of scale on the heat transfer conditions introduces some difficulties due to the following reasons:

- Thin scale layers (<10µm) may show enough adhesion for measurement but imply a very small heat resistance (“no effect”).
- Small gas layers between the scale and the substrate (blistering) imply a big effect on heat resistance (“random effect”).
- The adhesion of medium thickness scale layers (10..200µm) may be not sufficient to withstand spray water – spallation and descaling occurs (“instationary effect”).
- Thicker scale layers (>200µm) are removed rapidly by the spray water – the thermal effect becomes inhomogeneous.

Regardless these arguments, the practical importance remains and thus the influence of an oxide layer on heat transfer was investigated in a second set of experiments in which the low alloy material AISI 1008 was used. In order to get well defined scale formation, the oxidation kinetics was determined in the first step.

**Measurement of the oxidation kinetics**

In the first part of the study the high temperature oxidation kinetics of different steel materials was examined. The specimens were oxidized in a horizontal furnace at 650, 750, 850 and 1000 °C. The furnace was connected through a two-way valve to gas lines supplying protective gas (Ar 97% + H$_2$) or air. The samples were kept under protective atmosphere until the desired temperature was reached. Subsequently, the atmosphere was changed over to air for oxidizing of the steel for a certain time. After the oxidation, the sample was cooled down to room temperature again under protective gas atmosphere. The pressure was 1 bar. The thickness of the
oxide layers was measured by means of an optical microscope on polished cross sections of the samples. All experiments in air show, in principle, the well known a parabolic growth law. The time dependent oxidation process can be approximated by the function:

\[ s = \sqrt{2 \cdot k \cdot t} \]  

(2)

- \( s \) thickness of the oxide scale \([\text{m}]\)
- \( k \) growth constant \([\text{m}^2 \text{s}^{-1}]\)
- \( t \) time \([\text{s}]\)

The parabolic growth “constant” shows an Arrhenius type temperature dependence:

\[ k = k_0 \cdot e^{\frac{E_A}{RT}} \]  

(3)

- \( k_0 \) oxidation constant \([\text{m}^2 \text{s}^{-1}]\)
- \( E_A \) activation energy \([\text{J mol}^{-1}]\)
- \( R \) molar gas constant \([\text{Jmol}^{-1}\text{K}^{-1}]\)

Figure 4 shows the scale growth on low carbon steel (AISI 1008). For calculation of the thickness of oxide scale depending on oxidation temperature and time, the values \( E_A = 200 \text{ kJ mol}^{-1} \) and \( k_0 = 5 \cdot 10^{-3} \text{ m}^2 \text{s}^{-1} \) were used. As can be seen in this diagram, the growth of the oxide scale layer enhance parabolic with increasing time and exponentially with increasing temperature. It should be noted, that, depending on thickness and cooling conditions, the spallation of the oxide scale begins. Scale layers are thus not stable against thermal cycling. This investigation of the growth of oxide scale was determined in order to predict the scaling of the specimens for the heat transfer experiments, where these results were used. The samples were heated up to 1000 °C as described in the first set of experiments. After reaching of the temperature the samples were oxidized for a specific time to obtain a certain thickness of the oxide scale and subsequently cooled by spray water to ambient temperature.
Modelling of HTC for an oxidized surface

For understanding the influence of oxide layer on heat transfer during spray water cooling, the mechanism of heat transfer at an oxidized surface is theoretically described first. Since design calculations often can not include thin layer effects, an effective heat transfer coefficient containing all scale layer effects is introduced. The heat flow is thus described by an effective heat transfer coefficient which is using the temperature difference between the steel surface and the temperature of the water (see Figure 5):

\[
q = \alpha_{\text{eff}} \cdot (T_B - T_w)
\]

\( q \)  
heat flow density  
\([\text{W m}^{-2}]\)

\( \alpha_{\text{eff}} \)  
effective heat transfer coefficient  
\([\text{W m}^{-2} \text{K}^{-1}]\)

\( T_B \)  
temperature at the steel-scale interface  
\([\text{K}]\)

The heat transfer from the surface to the spray water is given by

\[
q = \alpha \cdot (T_S - T_w),
\]

and the heat transfer through the oxide layer can be approximated by

\[
q = \frac{\lambda_{sc}}{\delta_{sc}} \cdot (T_B - T_S)
\]

\( \lambda_{sc} \)  
heat conductivity of the scale layer  
\([\text{W m}^{-1} \text{K}^{-1}]\)

\( \delta_{sc} \)  
thickness of the scale layer  
\([\text{m}]\)

\( T_S \)  
temperature at the surface of the oxide scale  
\([\text{K}]\)

Under quasi-stationary conditions and for uniform cooling, the heat flow (4) from the surface to the cooling water (5) is equal to the heat flow through the scale layer (6) and we get the effective HTC defined by (7):

\[
\alpha_{\text{eff}}(T_B) = \frac{1}{\frac{1}{\alpha(T_S)} + \frac{\delta_{sc}}{\lambda_{sc}}}
\]

\( \alpha_{\text{eff}} \) contains the influence of the oxide layer (thermal insulation, temperature drop). It can be calculated by (7) using the HTC \( \alpha(T_S) \) at the surface, which does not depend on bulk material properties. Accurate measurements of \( \alpha \) without scale thus can be applied for surfaces with well defined oxide according to equation (7). Additionally, a measurement using an oxidized surface and equation (1) for analysis will yield to a "measured" \( \alpha_{\text{eff}} \).
RESULTS

The “expected” (calculated from equation (7)) and the corresponding “measured” temperature dependence of the effective HTC for an oxidized sample is shown in Figure 6. As material AISI 1008 and for the water impact density \( V_S = 3.8 \text{ kg m}^{-2}\text{s}^{-1} \) was used. The solid lines show the dependence of the HTC on temperature as calculated according to (1) from the experimental data for un-oxidized (0 min, i.e. \( \alpha_{\text{eff}} = \alpha \)) and oxidized samples (3 min and 5 min air at 1000°C). Assuming a scale layer thickness of 100 and 200 µm and using a heat conductivity of the oxide layer of \( \lambda_{\text{sc}} = 3 \text{ W m}^{-1}\text{K}^{-1} \) [11], the effective HTC calculated by (7) is also plotted (dashed lines). These investigations indicate, that with increasing oxide layer thickness at constant water mass flux density the \( \alpha_{\text{eff}} \) values decrease only in the burnout regime (partial film boiling) and the temperature corresponding to the Leidenfrost point move to higher values. In the area of stable film boiling the \( \alpha_{\text{eff}} \) values are approximately equal to \( \alpha(T) \), as long as \( \lambda_{\text{sc}}/\delta_{\text{sc}} \) is much greater than \( \alpha \). As can be seen in the diagram, the experimental findings show qualitative agreement with the calculations. The “0 min” curve corresponds to the sample without scaling. Increasing oxide scale to ~100 µm in thickness (3 min air at 1000°C, second line) respectively to ~140 µm (third line) the predicted behaviour was observed. The Leidenfrost point enhances from \( \Delta T \sim 420 \text{ K} \) to \( \sim 450 \text{ K} \) at 3 min and further more to 540 K for 5 min oxidation time. This can be explained by a lower temperature at the scale layer top surface leading to an earlier onset of unstable film boiling.

It should be noted that for these experiments, the oxide scale was adhesive in the region of the sample where on the back side the thermocouples for the measuring of the cooling curves were located. It must be mentioned that during a cooling process the state of the oxidized steel surface can change rapidly due to the separation or partial spallation of the oxide scale. This process is purely stochastic. Formation of a gas gap between the steel surface and the oxide layer can also happen. In this case, the gap establishes a larger heat transfer resistance and prevents the heat from flowing towards the scale layer. If, for instance, a 100 µm air gap is formed this will imply an \( \alpha_{\text{eff}} \) decrease from approximately 600-700 W m\(^{-2}\text{K}^{-1}\) down to 200 W m\(^{-2}\text{K}^{-1}\) in the temperature range of stable film boiling. As a consequence, spatial inhomogeneous heat removal from the surface will cause heat flows by conduction within the surface plane. These \( \alpha_{\text{eff}} \) “measurements” are thus not as accurate as the measurements without surface oxidation.

![Figure 6: Effective heat transfer coefficient in dependence on temperature difference for different oxidation times.](image-url)
For discussion, the HTC in the stable film boiling regime is plotted against its parameter, the water impact density. Figure 7 exhibits the heat transfer coefficient with and without scaling, depending on the water mass flux density under variation of material and temperature difference $\Delta T = T_s - T_w$. The grey area demonstrates the open literature values for $\Delta T = 700$ K, which proves that the heat transfer coefficient is a strong function of water mass flux density in the range of stable film boiling. Additionally, the results indicate an additional temperature dependency even in the so called stable film boiling regime. A further discussion of the measurement precision seems to be very important. The current developments approach a 25% accuracy for the determination of the HTC while the scatter in the literature data is about 40% in the stable film boiling regime. Below $\Delta T=500$K, approaching the Leidenfrost point, the measurements are somewhat less accurate.

For oxidized surfaces showing descaling during cooling, the local HTC varies spatial and with time. The mean value and its statistics needs to be investigated in greater detail. Without blistering, stable adhesive scale surface layers need to grow thicker than 250µm in order to influence the local heat transfer coefficient (see equation (7)).

Figure 7: Heat transfer coefficient without oxidation under variation of the material and temperature.
SUMMARY AND CONCLUSIONS

In this study, spray water cooling of steel materials from initial temperatures up to 1000°C was investigated. The (low alloy) AISI-1008 steel and - for comparison - the (stainless) AISI-314 was used for the experiments. The heat transfer coefficient was measured by an automated cooling test (instationary method). This allows for a determination of the HTC $\alpha$ depending on the surface temperature $T$, within a single experiment (Figure 6). The second parameter, the water mass flux density $V_w$ was varied from 3.8 to 25 kg m$^{-2}$s$^{-1}$ (Figure 3 and Figure 7).

Furthermore, spray water cooling of steel materials introduces additional effects due to the formation of oxide layers (scale). In order to investigate the influence of an oxide layer on heat transfer the high temperature oxidation of steel was examined first. For these experiments the AISI-1008 steel was used. The samples were oxidized in atmospheric air at different temperatures and for specific times. Subsequently the thickness of the oxide scale was determined. The temperature-dependent parabolic growth “constant” $k$ was determined from the experimental data for the scale layer thickness versus oxidation time at a certain temperature. The temperature is the main parameter for the thickness of the oxide layer, because the oxidation reaction rate increases exponentially with temperature, according to activation energy $Q_0$. Using $k_0$ and $Q_0$ it was possible to describe the oxide layer growth on the surface of AISI-1008 steel materials (Figure 4). These results are used to calculate the scale layer thickness in the HTC experiments.

Finally, the heat transfer coefficient was measured for samples with (initial) scale layers (Figure 7). For the assessment of the scale layer effect and application purposes, an effective heat transfer coefficient $\alpha_{eff}$ was defined. Heat transfer experiments for oxidized samples were presented and discussed. Summarizing, we observed:

- An additional temperature dependency in the high temperature (stable film boiling) regime was found, thus the HTC is also temperature-dependent above the Leidenfrost temperature – simple $\alpha(V_w)$ relations are very inaccurate.
- For the quantitative description of the oxide scale formation, spallation has to be taken into account.
- In the lower $\Delta T$ (unstable film boiling) regime, scale layers can dramatically influence the apparent HTC.
- The oxide layers mainly impede heat transfer by random formation of an isolating gas gap between the oxide and the bulk material (blistering).
- Spallation of the oxide scale and thus scale plates moving around additionally influence local and global cooling conditions.

As a conclusion, the heat transfer coefficient (HTC, $\alpha$) introduced by

$$q = \alpha \cdot (T_S - T_w)$$

fulfils numerical stability requirements for spray water cooling calculations. Nevertheless, $\alpha$ is a complex function of the surface temperature, the water mass flow density and the surface condition. The indirect measurement of $\alpha$ was demonstrated by thin sheet cooling experiments. Future work should be devoted to further enhancements with respect to the measurement precision and the statistics caused by the blistering and spallation of scale layers. While the effect of a stable adhesive scale layer on heat transfer was predicted and measured in this paper, further investigations of the mean HTC and its statistics under scale formation, blistering and spallation conditions are subject to current research. This paper and future work provides the data required for more defined and predictable spray water cooling and e.g. thus avoid material inhomogeneity due to inhomogeneous cooling procedures.

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