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Effect of oxide layers on spray water cooling heat transfer at high surface temperatures

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Abstract

The influence of surface oxidation phenomena on spray water cooling heat transfer is an important issue in steel industry. In practical applications, spray cooling is regularly used for the cooling of steel surfaces from high temperatures with an (initial) oxide (scale) layer. This paper investigates the changes in the heat transfer due to the oxide layer and its removal during cooling. After a theoretical treatment of the principal effects of thin and stable layers, the heat transfer coefficient was measured by an automated cooling test stand (instationary method) for oxidized steel samples. The heat transfer is described by the concept of an effective heat transfer coefficient. Compared to the clean surface state, scale layers show a different effective heat transfer coefficient in the transition boiling regime. Additionally, the local delamination of the oxide, the formation of vapor gaps at the oxide–metal interface and the movement of scale plates makes the heat transfer more erratic. The heat transfer is therefore inhomogeneous and has to be described statistically. For water temperatures around 291 K, surface temperatures between 473 and 1173 K, i.e. $\Delta T > 180$ K and water impact densities between $V_S = 3$ and $30\ \mathrm{kg/(m^2\,s)}$ the heat transfer coefficient α was measured. As sample material, different steel grades, oxidized in air for a specific time ($0, \ldots, 80\ \mu\mathrm{m}$ scale layer) were used. The results are compared with the clean surface state.

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Keywords: Spray water cooling; Heat transfer coefficient; Full cone nozzle; Subcooled liquid; Film boiling; Surface state; Oxidation; Scale formation; Continuous casting; Hot rolling

1. Introduction

Heat transfer from oxidized surfaces is an important issue for the practical application of spray water cooling technology in steel production and processing industry. Spray water cooling is used as secondary cooling procedure in strip casting and for the final microstructure optimization after hot rolling. In both cases, the initial surface is not clean, it is oxidized. The oxides form a scale layers of up to several 100 μm (thicker layers require special descaling practices). Based on the quantitative understanding of the heat transfer for clean surfaces [11], the special phenomena induced by the oxide layer needs to be investigated experimentally and theoretically.

Earlier investigations give some fundamental ideas on the effects of scale layers on heat transfer [3] and provide the properties of the scale [8]. Fundamental research on the droplet impingement process [2] provide basic knowledge applicable to mist cooling procedures at lower water impact densities. For immersion cooling, the oxidation level is known to influence the onset of transition boiling [7], similar to the effect of surface roughness [1]. For low temperature differences, the roughness effects were investigated in greater detail [5]. Technological aspects where investigated theoretically [4]. Due to the scale heat conductivity value of 0.2 W/(m K) used by Lin et al., the scale

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Nomenclature

α	heat transfer coefficient (HTC) (W/(m^2 K))
α_{eff}	effective HTC as a function of $f(\Delta T_{\text{B}} := T_{\text{B}} - T_{\text{W}})$ (W/(m^2 K))
c_p	specific heat (J/(kg K))
$\dot{V}_{\text{S}}$	spray water mass flux density (kg/(m^2 s))
v	velocity (m/s)
d	thickness/diameter (m)
h	specific enthalpy (J/kg)
λ	heat conductivity (W/(m K))
ρ_{s}	sample density (kg/m^3)
q	heat flux density (W/m^2)
t	time (s)
T	temperature (K)
ΔT	temperature difference (K)

Subscripts

air	air
B	metal surface, scale metal interface
d	droplet
f	fluid (water)
g	gas/vapor
ox	oxidation
s	sample
sc	scale
S	(scale) surface
U	lower surface (thermocouple position)
W	water
min, LF	minimum (Leidenfrost point)
max, CHF	maximum (critical heat flux)

effects are overestimated. As measured earlier, a more realistic value of the scale heat conductivity is 3 W/(m K) [8]. Nevertheless, the qualitative technological importance of the issue was clearly demonstrated. The initial investigations of [3] are, therefore, continued on the basis of the measurement results currently available for clean surfaces (see [11]).

For this paper, water ($T_{\text{W}} = 18\,^\circ\text{C}$) impact densities from $\dot{V}_{\text{S}} = 3\text{–}30\ \text{kg/(m}^2\ \text{s)}$ and surface temperatures between 200 and 900 °C are investigated. The spray was produced with full cone nozzles as described in [11].

Before describing the measurements in Section 3, the expected results are calculated in the next section.

2. Analysis of the scale layer effect

2.1. The effective heat transfer coefficient

For a simple application and a comparison of the results with those obtained for clean surfaces, the concept of the effective heat transfer coefficient is introduced. The situation is sketched in Fig. 1 and the effective heat transfer coefficient α_{eff} is defined by

$$q = \alpha_{\text{eff}} \cdot (T_{\text{B}} - T_{\text{W}}) \tag{1}$$

For the calculation of cooling processes involving oxidized surfaces, a simple usage of α_{eff} as a function of $(T_{\text{B}} - T_{\text{W}})$ instead of α as a function of $\Delta T_{\text{S}} := (T_{\text{S}} - T_{\text{W}})$ is required. All effects of the surface scale layer have to be included into the unknown function α_{eff}. For thin oxide layers, the heat transfer is quasi-stationary, i.e. the heat content of the scale layer is much lower than that of the metal. The heat flux density q_{S} from the surface is again described by a heat transfer coefficient $\alpha_{\Delta T_{\text{S}}}$:

$$q_{\text{S}} = \alpha_{\Delta T_{\text{S}}} \cdot (T_{\text{S}} - T_{\text{W}}) \tag{2}$$

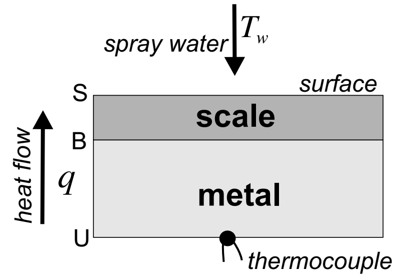

Fig. 1. Cooling scheme situation for an oxidized surface.

In the thin layer limit, it is equal to the heat flux density through the layer q_{BS}, calculated from the scale heat conductivity λ_{sc} and layer thickness d_{sc}:

$$q_{\text{BS}} = \lambda_{\text{sc}} \cdot \frac{T_{\text{B}} - T_{\text{S}}}{d_{\text{sc}}} \tag{3}$$

For the case of a thin layer and a spatially homogeneous cooling, α_{eff} can be calculated from the HTC α acting at the surface of the layer and the layer properties:

$$\alpha_{\text{eff}} = \left(\frac{1}{\alpha_{\Delta T_{\text{S}}}} + \frac{d_{\text{sc}}}{\lambda_{\text{sc}}}\right)^{-1} \tag{4}$$

The thermal isolation and temperature drop in the layer is thus included in α_{eff}. This concept has the following advantages:

- The modelling of the cooling of the sample itself requires only an exchange of the boundary conditions $\alpha \rightarrow \alpha_{\text{eff}}$, but no extra consideration of the scale layer.
- A measurement of α_{eff} and its statistical fluctuations includes all layer effects, even those currently not known in detail.
- The calculation of the cooling boundary condition requires only the knowledge of the temperature of the bulk metal surface, as it is for clean surfaces.

2.2. Scale layer effects for spray cooling

For a known clean surface HTC, α_{eff} can be estimated in advance under the assumption of a stable layer with known thickness d_{sc} and heat conductivity λ_{sc}. The given parameters are the metal surface temperature T_{B}, the scale layer thickness d_{sc} and its heat conductivity of $\lambda_{\mathrm{sc}} \approx 3\ \mathrm{W/(m\,K)}$ [8]. From the equal heat flux density condition in the quasi-stationary state, the (scale) surface temperature T_{S} can be calculated for a given T_{B}:

$$q = \alpha_{\Delta T_{\mathrm{S}}} \cdot (T_{\mathrm{S}} - T_{\mathrm{W}}) = \frac{\lambda_{\mathrm{sc}}}{d_{\mathrm{sc}}} \cdot (T_{\mathrm{B}} - T_{\mathrm{S}}) \tag{5}$$

The correlation from [11], valid also for the scale surface (ΔT_{S}),

$$\alpha(\Delta T_{\mathrm{S}}, V_{\mathrm{S}}) = 190 + \tanh\left(\frac{V_{\mathrm{S}}}{8}\right) \times \left(140 \cdot V_{\mathrm{S}}\left[1 - \frac{V_{\mathrm{S}} \cdot \Delta T_{\mathrm{S}}}{72,000}\right] + 3.26 \cdot \Delta T_{\mathrm{S}}^2 \left\{1 - \tanh\left(\frac{\Delta T_{\mathrm{S}}}{128}\right)\right\}\right) \tag{6}$$

is a non-linear function of ΔT_{S}. Thus (5) is a non-linear equation to be solved for T_{S}. From the resulting T_{S} and $\alpha_{\Delta T_{\mathrm{S}}}$ (Eq. (6)), the effective heat transfer coefficient (4) is calculated.

The results are shown as a function of ΔT_{B} in Figs. 2 and 3. While the effect of the scale layer is negligible in the stable film boiling regime, it is significant in the partial film boiling regime. The appearing maximum heat flux (CHF) decreases with increasing layer thickness, while the appearing Leidenfrost temperature increases.

These findings are from the viewpoint of an observer comparing an oxidized surface with a clean one. They are explained by the formation of a temperature drop in the scale layer. Thus the apparent earlier onset of partial film boiling is due to the viewpoint. While the sample surface temperature T_{B} is above T_{LF}, the (scale) surface temperature T_{S} has already reached T_{LF}.

Estimating the effect for different values of the layer thickness d_{sc} and its heat conductivity λ_{sc} by Eq. (4) shows no significant HTC changes in the stable film boiling regime, while $d_{\mathrm{sc}} < 1$ mm. Since real scale layers tend to be unstable (see the experiments below), the additional formation of a heat isolating gas or vapor gap between the scale and the metal surface has to be taken into account. Due to the very low heat conductivity of e.g. water vapor ($\lambda_{\mathrm{g}} \approx 0.08\ \mathrm{W/(m\,K)}$), this phenomenon is important – at least locally – in all boiling regimes. Such a gap formation and the scale layer instability, finally, results in statistical fluctuations of α_{eff} which have to be determined experimentally.

3. Experimental methods

For the experimental investigation of the HTC for oxidized steel samples during spray cooling, the instationary test stand as described in [11] was used. Additional video recording (IR and VIS) of the experiments allowed the identification of changes in the surface structure during cooling (see Fig. 6).

Sheet specimens of the different steel grades (Table 1) were milled to discs with 70 mm in diameter. Up to five thermocouple pairs, one in the center and the others perpendicular at a radius of 10 mm, 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 0.5 mm. The thermocouple wires were isolated by thin ceramic tubes. After installation in the furnace sample holder, the disks were heated under

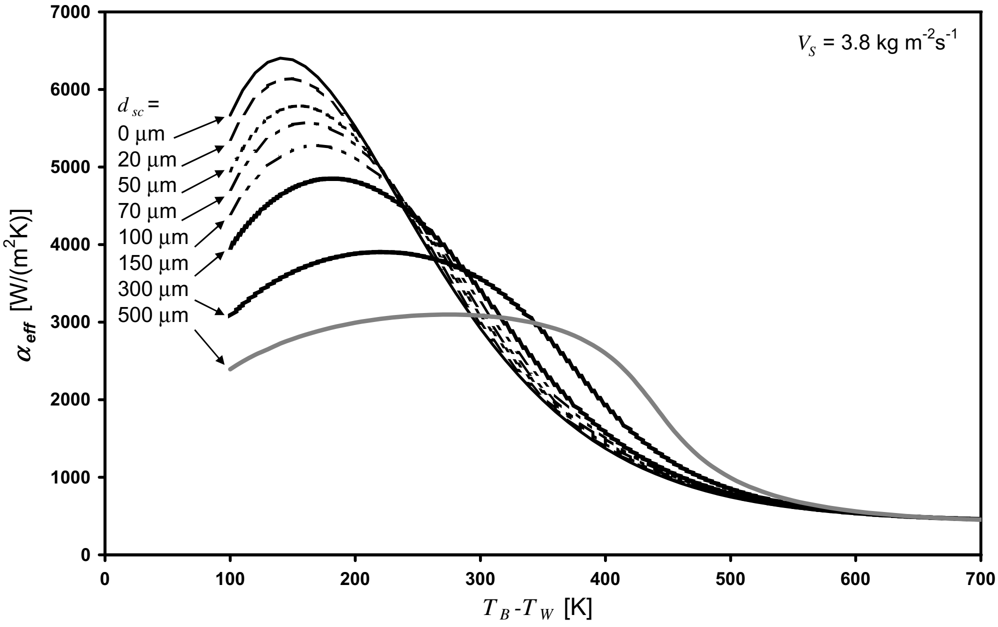

Fig. 2. Effective HTC α_{eff} for different values of the scale layer thickness and $V_{\mathrm{S}} = 3.8\ \mathrm{kg/(m^2\,s)}$.

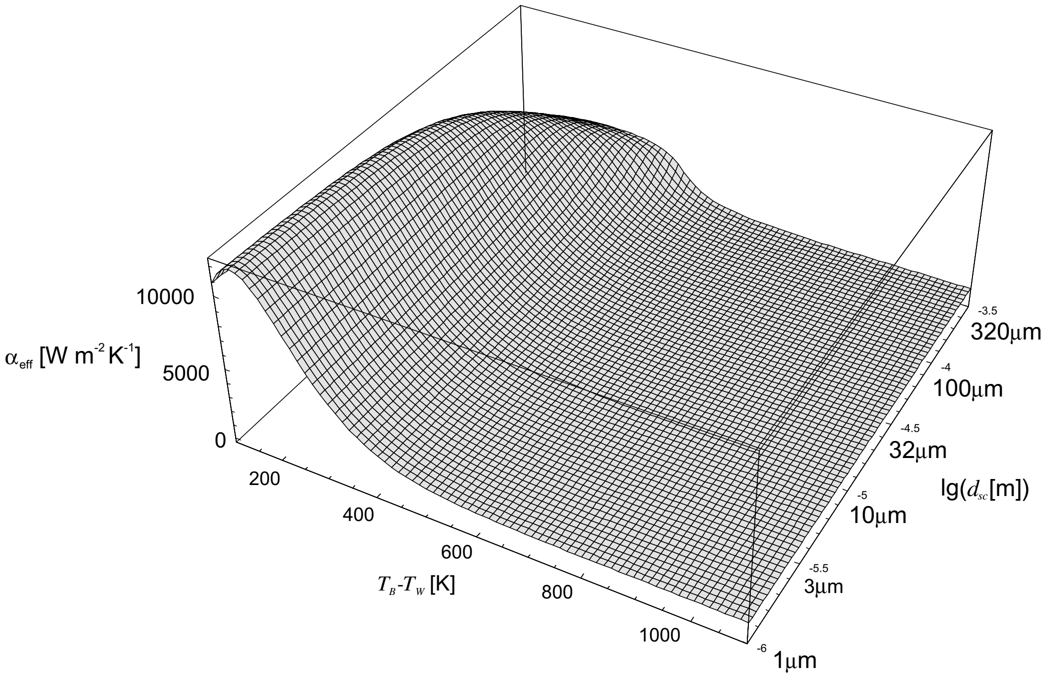

Fig. 3. Calculated effective HTC as a function of ΔT_B and scale layer thickness (logarithmic scaling).

Table 1
Analysis of the steel sample materials (wt%, balance: Fe)

Name	EN100[2,9]5/AISI No.	C	Si	Mn	Cr	Ni	Al	V
Thermax	1.4841/314	<0.2	2	–	25	20	–	–
DC04	1.0338/1008	0.03	0.01	0.20	0.03	0.04	0.04	0.001
SZMS1200	–	0.16	0.12	1.80	0.40	0.04	0.04	0.007
15,385	1.0570/573	0.13	0.01	1.51	0.03	0.03	0.03	0.003
51CrV4	1.8159/6150	0.52	0.23	0.84	1.04	0.04	0.03	0.12

protective atmosphere up to the desired starting or oxidation temperature. After oxidation in air for a specific time, the samples were moved automatically from the furnace under the full cone nozzle, which takes approximately 4 s. In this position, it is subsequently cooled with spray water (mass flux density V_S). The water temperature T_W was approximately 18 °C.

3.1. Oxidation kinetics

Since the scale layer thicknesses and properties cannot be determined experimentally in 4 s after oxidation and before spray cooling, the oxidation kinetics were determined by independent methods [9]. The layer thickness d_{sc} can be calculated from

$$d_{sc} = \sqrt{2 \cdot k \cdot t} \tag{7}$$

with

$$k = k_0 \cdot e^{-\frac{Q_0}{R \cdot T}} \tag{8}$$

with the oxidation constant k_0 ($m^2\,s^{-1}$), the activation energy Q_0 ($J\,mol^{-1}$) and the oxidation temperature T (K) ($R = 8.314472\,J\,mol^{-1}\,K^{-1}$ is the molar gas constant).

The values for k_0 and Q_0 are summarized in Table 2 allowing a calculation of the layer thickness from the oxidation time t and the oxidation temperature T. Control measurements of the layer thickness after cooling of the samples showed good agreement with the calculated values – if there is some portion of the surface where the layer was not removed during cooling.

3.2. Measurement accuracy

For homogeneous cooling, the HTC α_{eff} can be determined from the experimental cooling data by the thin sheet approximation (see [11]):

$$\alpha_{eff} \approx -\frac{c_p(T_U) \cdot \rho_s \cdot d_s}{T_U - T_W} \cdot \left.\frac{\partial T_U}{\partial t}\right|_U \tag{9}$$

Compared to the nickel samples used in [11], the specific heat of the sample materials is not accurately known in temperature intervals where phase changes occur. As demonstrated in Fig. 4, the $\gamma \to \alpha$ phase transformation occurring below 900 °C introduces potential errors due to unknown values of the specific heat in the temperature re-

Table 2
Oxidation law parameters k_0 and Q_0 for different steel grades (after [9])

Steel	Unit	DC06	DC04	MS1200	15,385	51CrV4
k_0	$m^2\,s^{-1}$	7.5×10^{-4}	5.2×10^{-3}	0.076	9×10^{-6}	2.7×10^{-4}
Q_0	$kJ\,mol^{-1}$	187	200	239	147	194

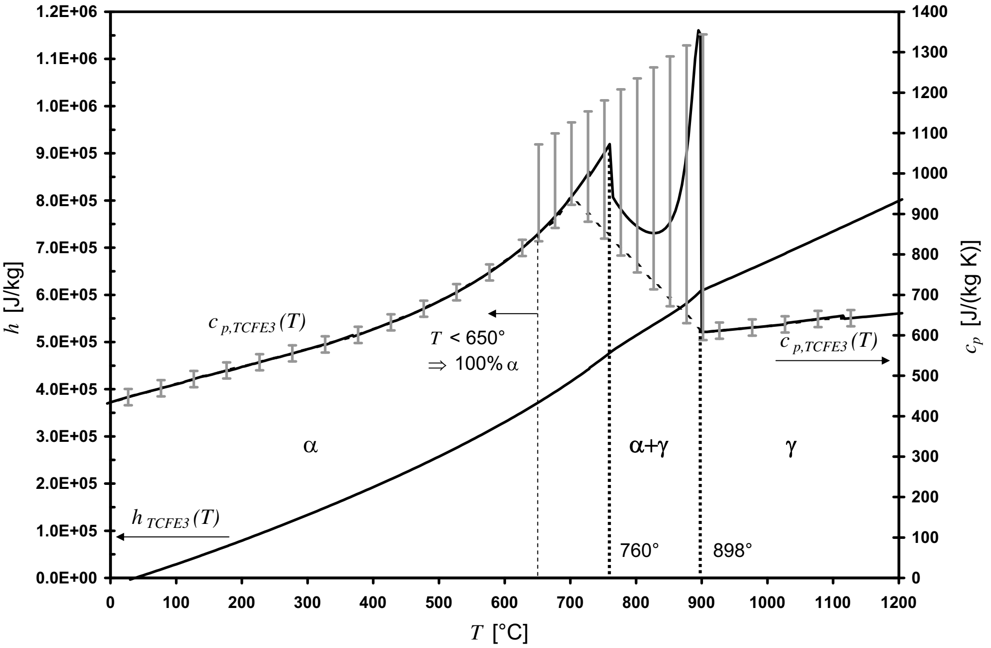

Fig. 4. Specific enthalpy and heat capacity of the low alloy steel DC04.

gion were the phase transition takes place and the real specific heat thus depends on the kinetics of the transformation. The error bars indicate the possible c_p-values in this temperature range.

The spray cooling parameters under investigation ($V_S = 3, \ldots, 30\ \mathrm{kg/(m^2\,s)}$) imply the occurence of descaling phenomena during cooling. These phenomena introduce spatially inhomogeneous cooling of the surface. Since there are no realistic alternative measurement methods for this special situation, the HTC measurement errors introduced from these phenomena using this method has to be discussed in greater detail.

As sketched in Fig. 5, the different thermocouples led to differing cooling curves and local HTC's. Visual and infrared observation showed the partially descaling of the sample during spray cooling.

Between 9 and 12 s, the individual T–t curves differ, e.g. at $t = 11$ s, $T_1 = 350$ °C and $T_2 = 451$ °C. By heat conduction in the sample plane, additional heat flows from T_2 to T_1, slowing down the cooling at T_2, i.e. $-\partial T_1/\partial t$ is decreased. The HTC calculated by (9) appears, therefore, smaller than in reality. The initial scale layer thickness was 78 ± 8 μm.

These phenomena allow accurate measurements and comparisons with the theory (Fig. 2) only for adherent scale layers. The onset of descaling induces both, measurement uncertainties and real spatial and temporal fluctuations of the heat transfer.

The HTC determination in the following section is thus limited by the following restrictions:

- Thin layers (<10 μm) show good adherence, but due to their small heat resistance there is no significant effect on α_{eff} – as calculated above (Fig. 3).
- Gas gaps between the steel surface and the scale layer introduce a significant, but stochastically fluctuating effect on the heat resistance.
- The adherence of scale layers between $10, \ldots, 200$ μm is not sufficient to withstand spray cooling (spallation and partial descaling), the cooling comes inhomogeneous.
- The movement of scale plates additionally influences the local heat transfer.
- Thick layers (>200 μm) rapidly spallate during spray cooling – the heat transfer becomes inhomogeneous until the scale is removed.
- The measurement accuracy is lower due to the specific heat problem (see above) and inhomogeneous cooling.

Nevertheless, it is possible to get a quantitative picture on the HTC values and their fluctuations by this measurement method – especially for comparison with the detailed predictions from Section 2.

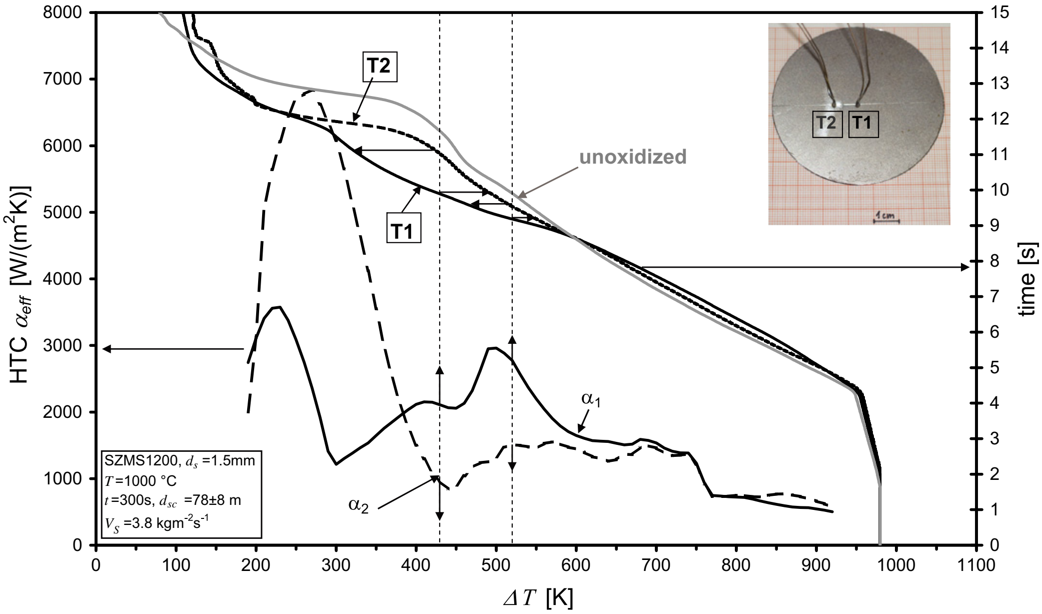

Fig. 5. Cooling curves and corresponding HTC's; SZMS1200, $d_s = 1.5$ mm; $t = 300$ s, $T = 1000$ °C, $V_S = 3.8$ kg/(m^2 s).

3.3. Descaling and statistics

For the oxidized samples the measurements of the different thermocouples spot welded to a single probe as well as the results of different measurements using the same parameters are analysed regarding their statistical mean HTC $\alpha_{\text{eff}}(\Delta T_B)$ and its standard deviation. Additionally, the descaling during spray cooling was recorded as shown in Fig. 6. The videos clearly demonstrate the inhomogeneous surface state and temperature distribution of the oxidized samples during spray cooling.

4. Experimental results

For the different steel materials (see Table 1) the parameters water impact density V_S and oxide layer thickness d_{sc} were varied and the cooling curves and video recordings were analysed with respect to qualitative systematic statements on the observations and quantitative statements on the statistics of the scale induced HTC $\alpha_{\text{eff}}(\Delta T_B)$ variations.

4.1. Variation of the water impact density

The water impact density increases heat removal also for oxidized samples (Fig. 7). In the stable film boiling regime ($\Delta T > 600$ K), accurate measurements are difficult due to $\gamma \rightarrow \alpha$ phase transitions in the material. The fluctuations in this regime seem to be below the measurement accuracy. Especially for higher values of V_S the cooling becomes inhomogeneous below the Leidenfrost point, where statistical fluctuations of 30–50% can be observed for initial scale layers of $d_{sc} \approx 55$ μm. The fluctuations can be explained by local descaling and the formation of gas gaps at the metal–oxide interface due to the rapid quenching of the thin scale layer. Both phenomena result in spatially and temporarily inhomogeneous cooling.

4.2. Different materials

The alloying elements in the materials imply scales with differing properties, especially different mechanical stabil-

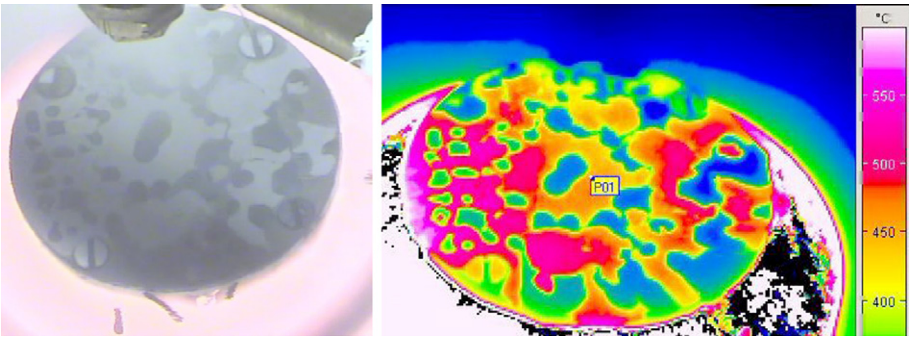

Fig. 6. VIS (left) and IR (right) recording of the cooling of steel MS1200 (5 min oxidation at 1000 °C, $\Delta T_B = 600$ K, $V_S = 4.7$ kg/(m^2 s)).

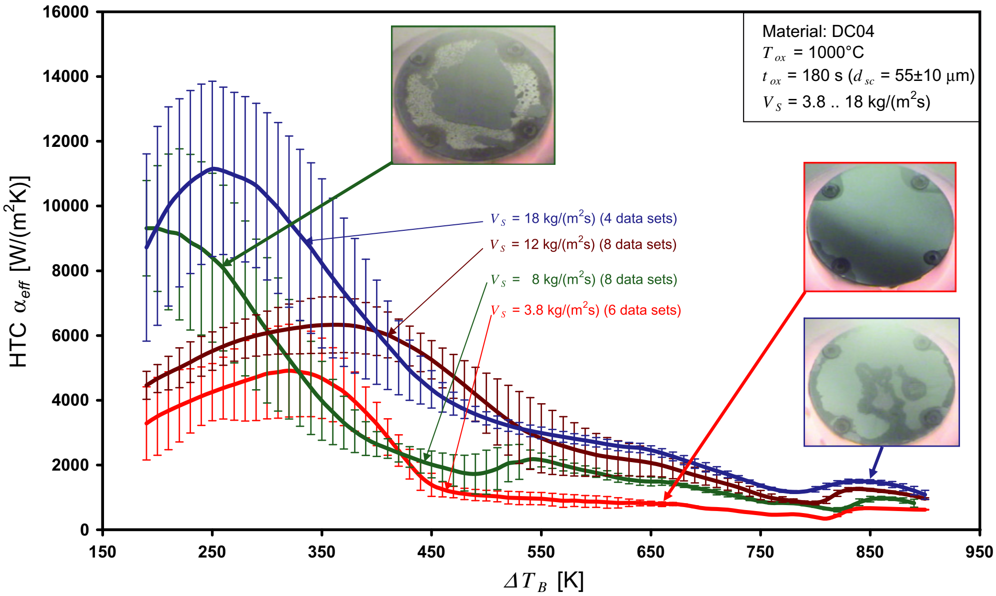

Fig. 7. Temperature dependence of the measured effective HTC α_{eff} for different water impact densities V_S (DC04 steel oxidized at 1000 °C for 3 min ($d_{sc} \approx 55$ µm)).

ity. The differences in the descaling kinetics led to different mean HTC values and fluctuation amplitudes (Fig. 8). The quantitative measurement of these phenomena is limited by the complex and material specific phase transformation kinetics during cooling of the different steel grades. The resulting accurate $c_p(T)$ variation in the transformation temperature range is unknown and reliable α measurements are thus difficult.

The general impression is as follows. For materials with instable oxide layers the HTC fluctuations are small because of the rapid descaling (see 51CrV4 in Fig. 8). For materials forming scale layers of intermediate stability (see DC04 and 15,385 in Fig. 8), the local HTC can be influenced dramatically. Compared to the very instable scales, the detachment occurs at lower temperatures and the scale plates moving around are larger, therefore, the spatial and temporal fluctuations are significant. Due to the smoothing role of heat conduction, the fluctuations are damped out inside the material.

For the high alloy steel Thermax, the results are comparable to those using Ni samples (see [11,10]). Due to the protective and adhesive nature of the oxides, the scale layer on the Thermax surface is very thin (<1 µm) and stable. Differences in the apparent HTC can be explained by the

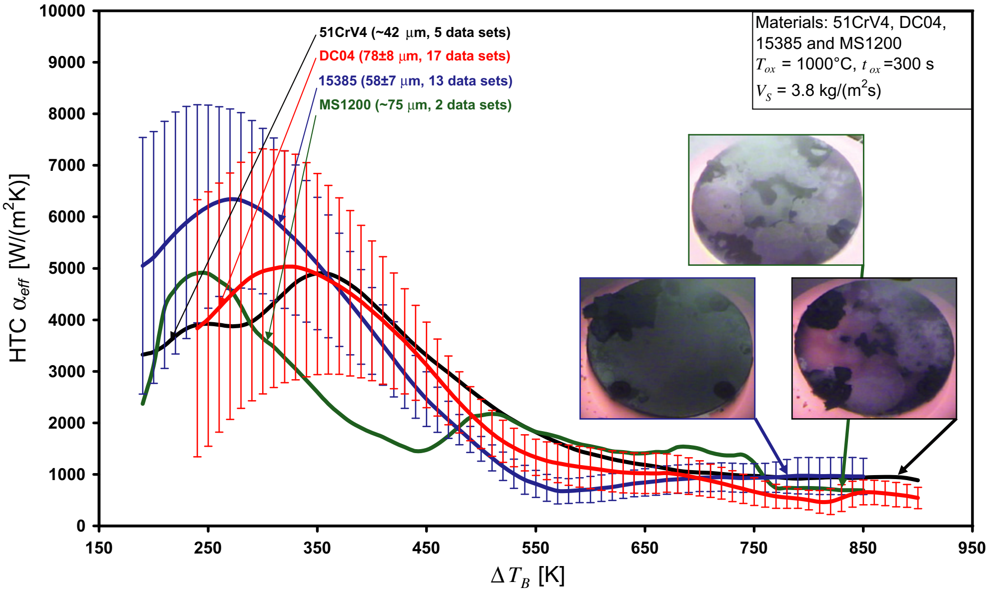

Fig. 8. Temperature dependence of the measured effective HTC α_{eff} for different materials (5 min oxidation at 1000 °C, $V_S = 3.8$ kg/(m² s)).

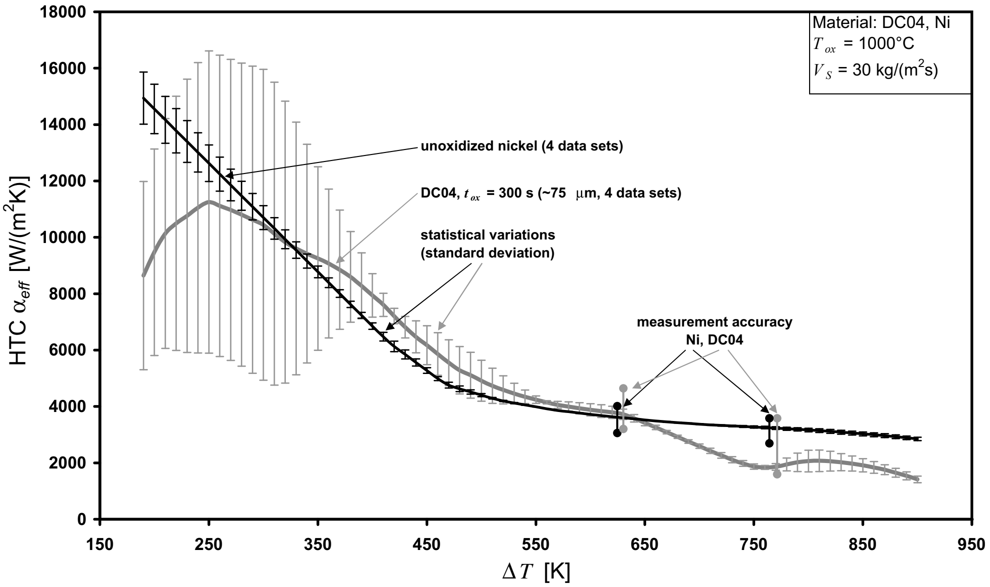

Fig. 9. Temperature dependence of the measured effective HTC α_{eff} and its statistical fluctuations for oxidized steel DC04 and for Ni cooled at $V_{\mathrm{S}} = 30\ \mathrm{kg/(m^2\,s)}$.

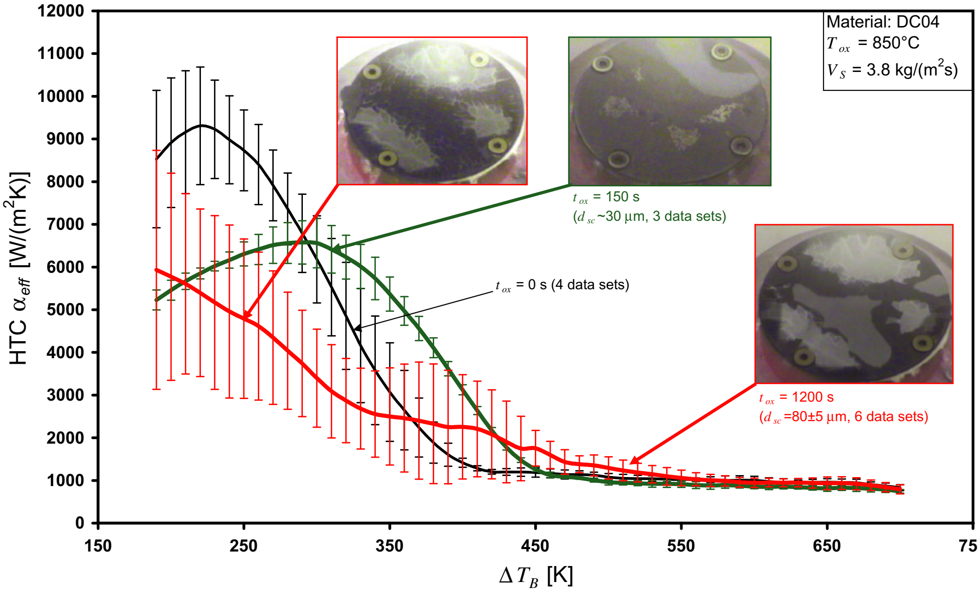

Fig. 10. Temperature dependence of the measured effective HTC α_{eff} under variation of the initial oxidation level.

complex phase transformations during cooling (unknown $c_p(T)$).

4.3. Oxide layer thickness

Comparing the observed HTC fluctuations for the low alloy steel DC04 with an initial 75 µm oxide layer with those from the nickel measurements (Fig. 9), significant fluctuations or differences are found in the transition boiling regime below $\Delta T_{\mathrm{B}} \approx 400$ K. These fluctuations tend to require a minimum layer thickness of ≈ 50 µm (Fig. 10). For scale layers above this value, there is a tendency of decreased mean HTC below $\Delta T_{\mathrm{B}} \approx 400$ K with fluctuations of ±50% at $\Delta T_{\mathrm{B}} \approx 300$ K.

5. Summary and conclusions

As sketched in Fig. 11, the theoretical effect of thin homogeneous and adhesive oxide layers on heat transfer is significant only for layers of 100 µm thickness and above. The measurements support this finding, but only for adhesive scale layers (e.g. those on stainless steels). Real scale layers on low alloy steels are normally not stable enough to withstand spray cooling. The heat transfer from oxidized

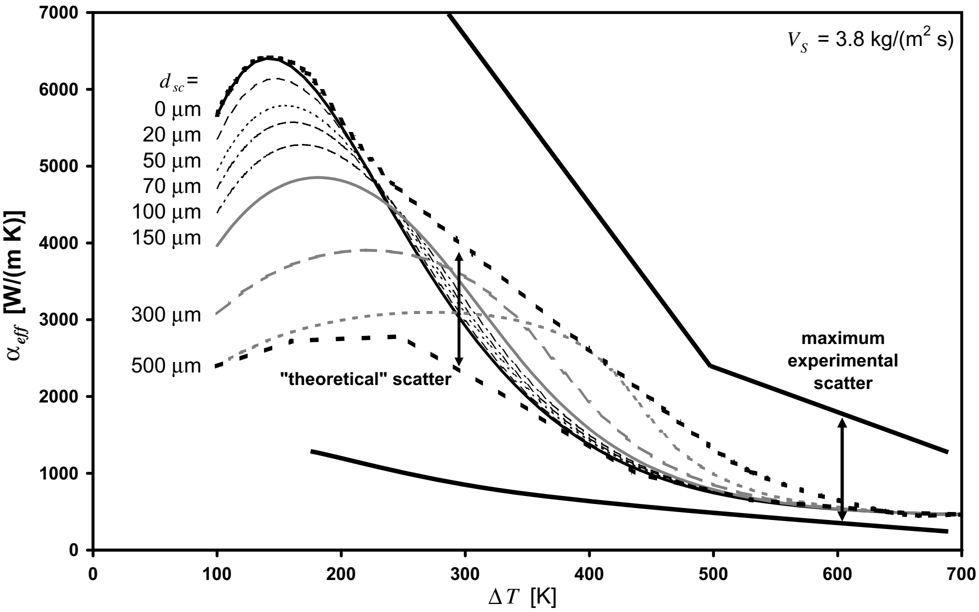

Fig. 11. Effective HTC α_{eff} as a function of ΔT for different oxide layers (theory) and minimum and maximum local values from all experiments.

(steel) surfaces is thus influenced by the instability of the scale layer – especially in the temperature region of transition boiling. Regardless the difficulties of an accurate measurement of the HTC from such surfaces, several general trends were observed:

- Above the Leidenfrost point, the oxide layers normally do not influence the mean HTC but its statistical spatial and temporal fluctuations can reach practically significant values.
- The Leidenfrost point itself can move to higher temperatures thus the heat transfer is significantly enlarged for oxidized surfaces in this regime.
- The oxide layer effect on the HTC decreases with increasing stability and with decreasing thickness of the layer.
- The (local) formation of gas gaps between the scale and the hot metal surface can drastically reduce the (local) HTC.
- As long as the clean surface HTC should be measured, steel samples should not be used in temperature ranges where accurate values of the specific heat $c_p(T)$ are not available due to phase transformations in the material.

The big scatter of all experimental data – as shown in Fig. 11 – is partially originated by the measurement inaccuracies induced by the oxide layer and the inaccurate c_p-values for the steel samples. The *theoretical scatter* is due to the assumption of a removal of the layer during cooling.

For the application in steel industry, the inhomogeneity amplification by the boiling curve (increasing HTC with decreasing ΔT) implies a strict control of oxidation and cooling procedures in hot rolling, while the scale seems not to be very crucial as long as high surface temperatures are maintained.

For a further enhancement of the scientific understanding of the interaction between heat transfer, surface oxidation and descaling, stationary experiments (as proposed by [6]) in the film boiling regime as well as further parametric studies using instationary methods may be helpful. The discussion of measurement errors vs. process specific fluctuations seems to be also a critical issue for future investigations. Especially in the transition boiling regime, the measurement accuracy has to be enhanced with special emphasis on transient and hysteresis phenomena.

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