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Study on plasma enhanced CVD coated material to promote dropwise condensation of steam

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Abstract—The promoting properties of hard coatings with an amorphous hydrogenated carbon basis to attain dropwise condensation (DWC) of steam on coated copper surfaces were investigated. Using differently produced coatings, equilibrium contact angles of θ_{eq} of 65, 74 and 90° could be reached for water. Stable and well reproducible heat transfer measurements could be performed. For a subcooling temperature of the condensor surface of 5 K, the DWC heat transfer coefficient at the vertical wall is 11 times higher for the surface with $\theta_{eq} = 90°$ than that measured for filmwise condensation (FWC), seven times higher for the surface with $\theta_{eq} = 74°$ and 3.5 times higher for the surface with $\theta_{eq} = 65°$. In comparison to the heat transfer coefficient measured for a contact angle of 90° for the heat flux ranging from 0.4–0.9 MW m^{-2} only 53–45% (for $\theta_{eq} = 74°$) and 1–7.5% (for $\theta_{eq} = 65°$) of the 90°-values were determined. For $\theta_{eq} = 90°$ the observed DWC keeps very well stable up to a technically achievable maximum heat flux of 1.54 MW m^{-2}. For $\theta_{eq} = 74°$ and for $\theta_{eq} = 65°$, however, expanded condensation streams (mixed condensation) appeared on the surface at heat fluxes of 1.03 MW m^{-2} and 0.7 MW m^{-2}. In these situations the performance characteristic is less developed in comparison to pure DWC, but still better than for pure FWC.

INTRODUCTION

Dropwise condensation (DWC) has been very well known for more than 60 years [1]. Compared to filmwise condensation (FWC), the obtainable heat transfer coefficients are pronouncly enhanced. This increase strongly depends on the wettability of the condensation surface which usually can be measured by the size of the contact angle between the condensing fluid and the surface. Incomplete wettability of the surface is the basic condition for forming DWC which usually is not given for metallic surfaces with large surface energies (and small contact angles). Such surfaces, however, are mostly used for industrial condensors, in which therefore FWC appears. In order to reach the surface energy conditions for DWC, appropriate coatings must be used as promoters which have to meet several requirements. These conditions are described in more detail elsewhere [2, 3]. Here, a new type of promoter has been used which is based on amorphous hydrogenated carbon films (a-C:H) with diamond-like mechanical properties [4]. The mechanical properties of these coatings are presented in combination with investigations on its suitability to act as a promoter for DWC.

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AMORPHOUS HYDROGENATED CARBON COATINGS

For promoting DWC, a new type of promoter deposit was used. The basis of these promoter coatings is formed by amorphous layers of hydrogenated carbon (a-C:H or DLC, Diamond-Like Carbon), with its own particular special properties for an industrial application [8–12]: excellent mechanical/tribological characteristics, chemical inertness, high resistance to acids, alkalis and solvents, as well as incomplete wettability with a contact angle of equilibrium of approximately 70° when water is used. Larger contact angles for water can be generated for reducing the surface energy of the pure DLC layer by adding fluorine, silicon or silicon in combination with oxygen during the PECVD (Plasma Enhanced Chemical Vapour

NOMENCLATURE

A	area [m^2]	FWC	filmwise condensation
c	specific heat capacity [J (g K)]	HC	heat conduction
D	diameter [m]	in	inlet conditions
g	acceleration due to gravity [m s^{-2}]	out	outlet conditions
h	heat transfer coefficient [W (m^2 K)$^{-1}$]	P	promoter
$\dot{m}$	mass flow [kg s^{-1}]	p	constant pressure
p	pressure [Pa]	S	saturation condition
$\dot{Q}$	total heat flow [W]	St	steam
$\dot{q}$	heat flux [W m^{-2}]	Surf	surface
Θ	surface radius [m]	W	wall
s	distance [m]	1	position 1
T	temperature [K]	2	position 2
$\dot{V}$	volume flow [m^{-3} h^{-1}]	∞	ambient conditions.
w	[m s^{-1}].		

Greek symbols

Δ	difference
Δh	specific heat of vaporization [J g^{-1}]
η	viscosity [kg (m^{-1} s)]
θ	contact angle [°]
λ	thermal conductivity [W (m K)]
ρ	[kg m^{-3}].

Subscripts

C	condensation
CW	coolant (water)
DWC	dropwise condensation
eq	equilibrium

Abbreviations

a-C : H	amorphous hydrogenated carbon
Al_2O_3	aluminium oxide (suspension, grain size 5 μm)
CVD	chemical vapour deposition
DLC	diamond-like carbon
DWC	dropwise condensation
F	fluorine
FWC	filmwise condensation
PECVD	plasma enhanced chemical vapour deposition
PTFE	polytetrafluoroethylene
REM	screen electron microscope
Si	silicon
Si/O	silicon/oxygen.

Deposition) coating process [4, 5, 7]. The reduction of the surface energy is dependent on the used modifying elements (F, Si, Si/O), and on the operation parameters of the production process [6]. Figure 1 shows the surface energy and the contact angle at equilibrium which can be obtained for such systems. The mechanical/tribological properties of the coatings are listed in Table 1. It can be seen that the pure DLC coating has the most favourable mechanical/tribological characteristics followed by the DLC coating with silicon. Long standing time effects of the promoter coatings which are related to the quality of their tribological properties have not been investigated so far. It may, however, be expected by the results of many applications of these coatings, that the favourable mechanical/tribological properties in general increase their suitability [12] and in particular also for heat transfer measurements.

EXPERIMENTAL SET-UP AND MEASUREMENT PROCEDURE

The used test facility has been described in detail elsewhere [2, 3]. Figure 2 shows the schematic of the condensor cell; the heat flows measured in order to determine the condensation heat transfer coefficients are represented by arrows. The total heat flow for the condensation process is given by

$$\dot{Q}_c = \dot{q}_c A = \dot{m}_c \Delta h. \tag{1}$$

This must be equal to the total heat flow measured on the cooling side

$$\dot{Q}_{CW} = \dot{q}_{CW} A = \dot{m}_{CW} c_{p,CW} (T_{CW,out} - T_{CW,in}). \tag{2}$$

A third heat flux balance can be achieved by two temperature measurements in the condensor wall at a distance of $\Delta s_{1/2}$ in case the thermal conductivity λ_W of the wall is known and one-dimensional heat conduction can be assumed.

$$\frac{\dot{Q}_{HC}}{A} = \dot{q}_{HC} = \lambda_W \frac{T_1 - T_2}{\Delta s_{1/2}}. \tag{3}$$

This balance in particular is advantageous, as the mean surface temperature can be determined by extrapolating the temperature gradient. The precision

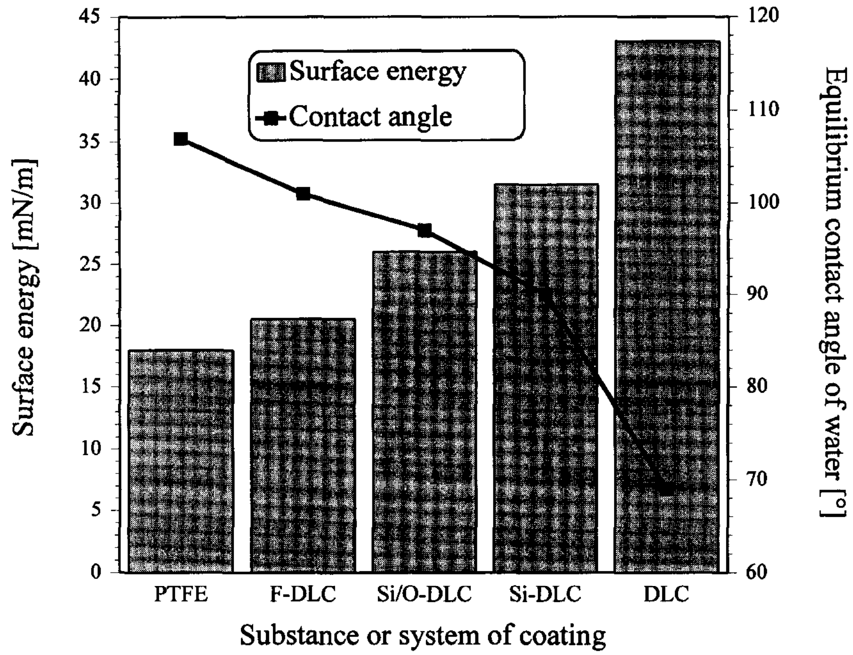


Fig. 1. Surface energy and equilibrium contact angle of water for different coating systems (Table 1) in comparison with PTFE with favourable incomplete wettability.

Table 1. Mechanical/tribological properties of the amorphous hydrogenated carbon (a-C : H) film and three network modification forms (in comparison with PTFE as substance of incomplete wettability)

Substance/ promoter	Modification	Temperature stability [°C]	Micro-hardness [GPa]	Friction coefficient (vs steel)	Abrasive wear (vs Al_2O_3) $[m^{-3}\,Nm]\cdot 10^{-15}$	Young's modulus [GPa]
DLC	Without	350	20–30	0.20	1.0	250
DLC-F	Fluorine	Without statement	2	0.20	150	20–80
DLC-Si	Silicon	400	11	0.12	6–8	120
DLC-Si/O	Silicon/ oxygen	400	7	0.40	8–10	80
PTFE	—	260	0.3	0.12	$\gg 300$	0.5

of this method has already been examined by Wilcox and Rohsenow [13].

The efficiency of the condensation process is given by the heat flux $\dot{q}$ for an adjusted surface subcooling temperature $\Delta T = T_s(p_c) - T_{Surf,C}$. The required heat transfer coefficient follows from

$$h_c = \frac{\dot{q}}{\Delta T}. \quad (4)$$

For FWC the mean temperature of the condensation surface $T_{Surf,C}$ can be determined by extrapolating the temperature gradient measured in the disc body (here, based on T_1; Fig. 2)

$$\text{FWC:} \quad [T_{Surf,C}]_{FWC} = T_1 + \frac{\dot{q}}{\lambda_W} s_1 \quad (5)$$

while for DWC additionally the thermal resistance of the promoter coating must be taken into account

$$\text{DWC:} \quad [T_{Surf,C}]_{DWC} = T_1 + \frac{\dot{q}}{\lambda_W} s_1 + \frac{\dot{q}}{\lambda_P} s_P. \quad (6)$$

For a stable operation point, all three total heat flows, equations (1)–(3), must be equal within a given range of inaccuracy.

FWC MEASUREMENTS

For laminar FWC the heat transfer coefficient can be calculated from Nusselt's theory [14], which has been modified and proved by O'Neill and Westwater [15] for condensation on vertically oriented surfaces.

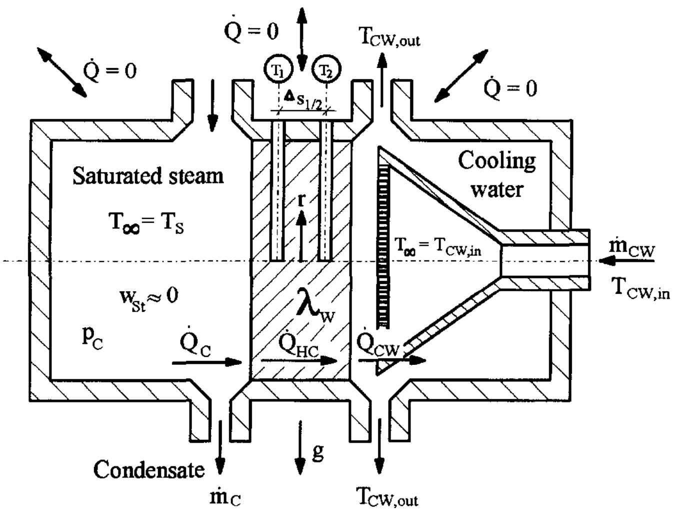


Fig. 2. Schematic of the condensor cell indicating the quantities of interest for setting up the different heat balances (condensor disc ∅ 60 mm, disc thickness about 10 mm).

By comparing these theoretical coefficients as function of ΔT

$$h_{\mathrm{FWC}} = 0.943 \cdot 4 \sqrt{\frac{\lambda^3 \rho^2 \Delta h g}{\eta 8.017 D \Delta T}} \tag{7}$$

with the actually measured values, it was possible to prove the quality of the test facility and the purity of the condensing steam. Laminar FWC was achieved on the surface of a copper disc of diameter 25 mm (Fig. 3).

All data points indicate the calculated average of 10 individual measurements of 4 s integration time. In these measurements, the differences in the different heat flows, equations (1)–(3), did not differ by more than 3%. From this it can be assumed that only stable operation points have been taken into the evaluation and that one dimensional heat conduction in the disc body has been reached.

DWC MEASUREMENTS

The thermal conductivity λ_p of the promoter coating must be known to calculate the heat transfer coefficients, equation (6). The thickness of the coating was determined with an inaccuracy of $\pm 0.05\ \mu$m using REM (Raster-Electron-Microscope) photographs [2]. The thermal conductivity has been determined by performing two series of condensation measurements

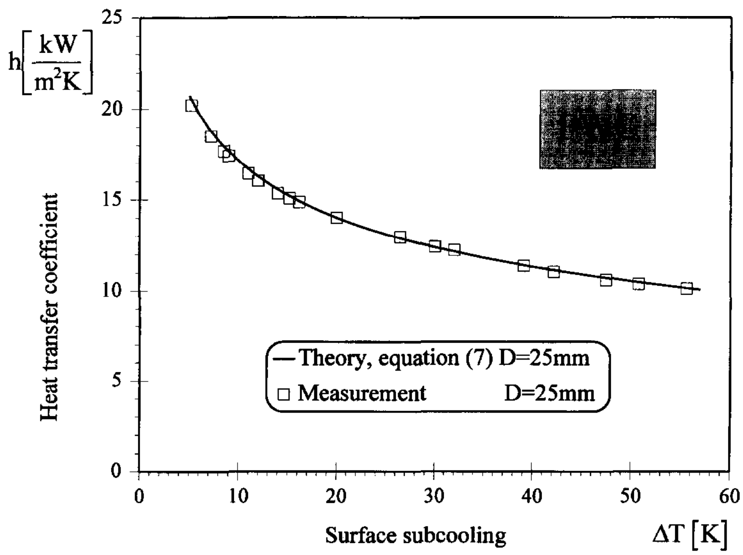


Fig. 3. Experimental and theoretical results for atmospheric FWC of steam on the surface of a fine processed circular copper disc of ∅ 25 mm.

Table 2. Promoter thicknesses (S_p) and equilibrium contact angles of the amorphous hydrogenated carbon (a-C : H) film and three network modification forms (roughness grade number N2, ISO 1302) which were measured immediately after finishing the surface preparation $\theta_{eq}(1)$, and just before starting the condensation measurements, $\theta_{eq}(2)$

Promoter	S_p [μm]	$\theta_{eq}(1)$	$\theta_{eq}(2)$
DLC	3.1	$73 \pm 2^\circ$	$74^\circ \pm 2^\circ$
DLC-F	3.1	$99 \pm 2^\circ$	$65^\circ \pm 2^\circ$
DLC-Si	2.0	$90 \pm 2^\circ$	$90^\circ \pm 2^\circ$
DLC-Si/O	4.0	$98 \pm 2^\circ$	$90^\circ \pm 2^\circ$

using the same promoter with different thicknesses and, as a second method, by two series of measurements with coated and uncoated surfaces. From both methods, a value $\lambda_p = 0.2 + 0.01$ W (m K) has been found [2].

The size of the contact angles have been measured with an adjustable vernier microscope with rotatable magnifying optics, angle vernier and recticle. For the fluorine modified DLC coating, measurements of the contact angle performed immediately after finishing the surface preparation and just before the condensation experiments yielded a contact angle difference of 34° (see Table 2). A difference of 8° was found for the coating modified with Si/O. These differences are far beyond the estimated error of $\pm 2^\circ$ for the contact angle measurements. These alterations in the contact angle, which have not been found for the other coatings, are assumed to be caused by a too large concentration of the added elements resulting in a less dense coating structure. Newer results indicate that with appropriate coating parameters an alteration can be avoided. After the condensation measurements, the contact angles were measured again indicating the same result and by this the stability of the wetting conditions during the experiments (respectively, no further alteration process during the experiments).

As for FWC, also for DWC every measurement point corresponds to the mean value of ten measurements taken with an integration time of 4 s, for which the different heat flux balances agreed within an inaccuracy range of 3%. The measured DWC heat transfer coefficients are shown in Fig. 4.

Stable operation conditions were established on all condensor discs, the performance values were easily reproducible. Increasing the integration time did not alter any results.

The DWC heat transfer coefficient is dependent on the surface subcooling temperature and on the size of the contact angle. The larger the surface subcooling temperature, the lower is the heat transfer coefficient for every DWC surface. This is due to the fact that the amount of condensate on the condensor wall becomes larger when the thermal flow increases. This results in an additional heat conduction resistance, by this approaching for large subcooling temperatures the heat transfer coefficients of laminar FWC.

Figure 5 shows the measured heat flux in dependence on the surface subcooling temperature. While the condensate always appeared in form of drops at contact angles of 90° (Fig. 6a and b), mixed condensation could be observed for smaller contact angles depending on the actual operation point (Fig. 6c and d). Here, besides areas with clear DWC, also areas appear where condensation streams cover a large extent of the surface. The surface energy seems not to be sufficiently low to maintain all the condensate on the wall in form of drops creating a constant condensate motion on the surface, which becomes more intense when the heat flux increases. In spite of this phenomenon, for increasing subcooling a still rising heat flux was observed for the DLC-F coating. This was totally different for the DLC coating alone. With increasing surface subcooling that part of the surface, which was covered with condensation streams, grew continuously and was finally limiting the condensation heat flux. This phenomenon was clearly reproducible when increasing and decreasing the surface sub-cooling. The maximum heat flux for the DLC coating was $\dot{q} = 1.18$ MW m^{-2}.

In order to compare the effect of the size of the contact angle and the resistance of the promoter, in Fig. 7 the obtained heat flux is drawn against the cooling water volume flows. An overall comparison of the efficiency is possible, as the set-up, vapour temperature and inlet coolant temperature were kept constant during these measurements. The cooling water flow can also be considered as a measure of the intensity of the heat sink (this is because the heat transfer coefficient of the heat sink varied only slightly when the volume flow of the cooling water remains constant, in spite of different heat fluxes and varied temperature levels of the wall in contact with the cooling water). At a heat transfer coefficient of the heat sink of approx. 12000 W (m^{-2} K), here at $\dot{V} \approx 0.5$ m^{-3} h, the measured DWC heat flux for the 2.0 μm DLC-Si coating (contact angle of 90°) is 1.5 times larger than for FWC and even for the 3.1 μm DLC-F coating (contact angle of 65°) still larger with a factor of 1.3. For $\dot{V} \approx 0.5$ m^{-3} h DWC could be observed for all different coating systems. The DWC heat transfer coefficients are substantially larger than those of normal convective cooling water streams. An efficient utilisation of DWC can best be realized with the application of extended cooling surfaces (e.g. inner side finned tubes).

CONCLUSIONS

Heat transfer measurements were performed using FWC and DWC of water vapour at ambient pressure. The FWC measurements were carried out to test the cleanliness of the system and the functionality of the measuring techniques. The results found are in good agreement with Nusselt's theory. Different DLC coat-

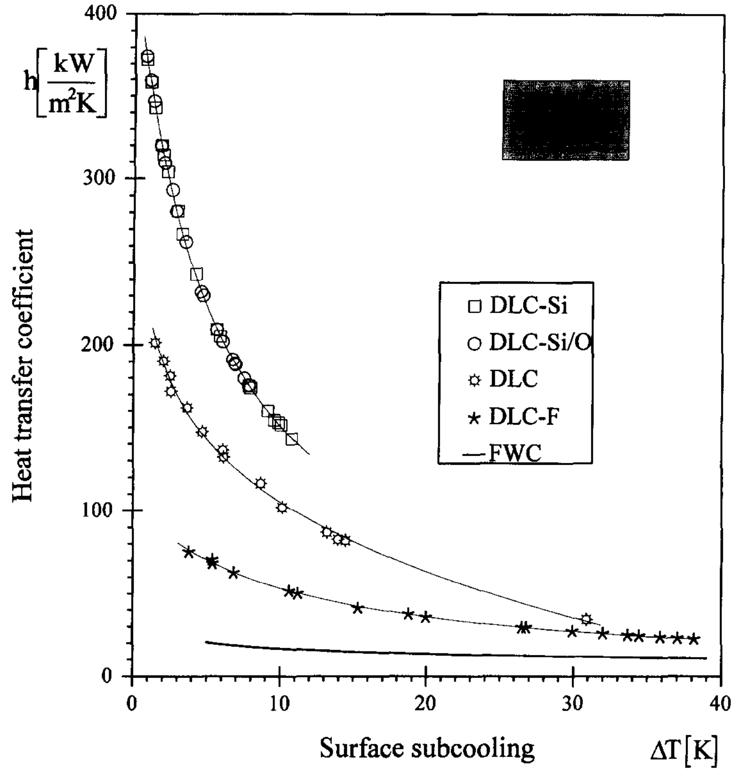


Fig. 4. Measured heat transfer coefficient for different coating systems (FWC measured for comparison).

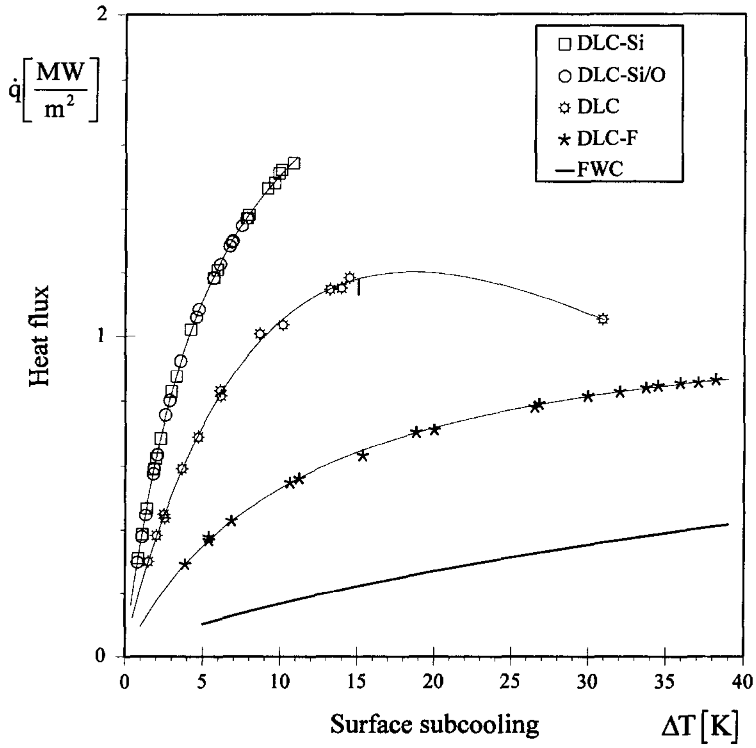
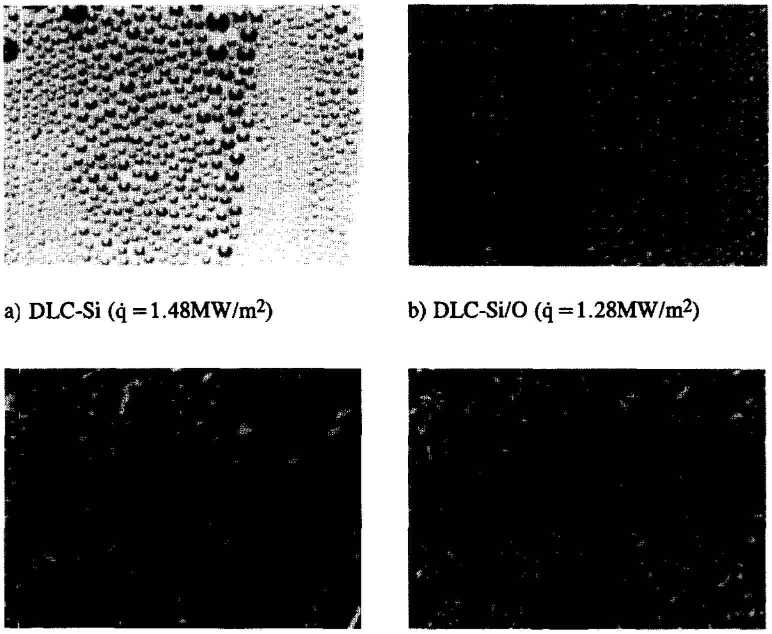


Fig. 5. Heat flux in dependence on surface subcooling temperature (FWC measured for comparison).



a) DLC-Si ($\dot{q}$ = 1.48MW/m²) b) DLC-Si/O ($\dot{q}$ = 1.28MW/m²)

c) DLC ($\dot{q}$ = 1.18MW/m²) d) DLC-F ($\dot{q}$ = 0.84MW/m²)

Fig. 6. Visual observation of the condensation process on coated copper discs for the different coating systems investigated.

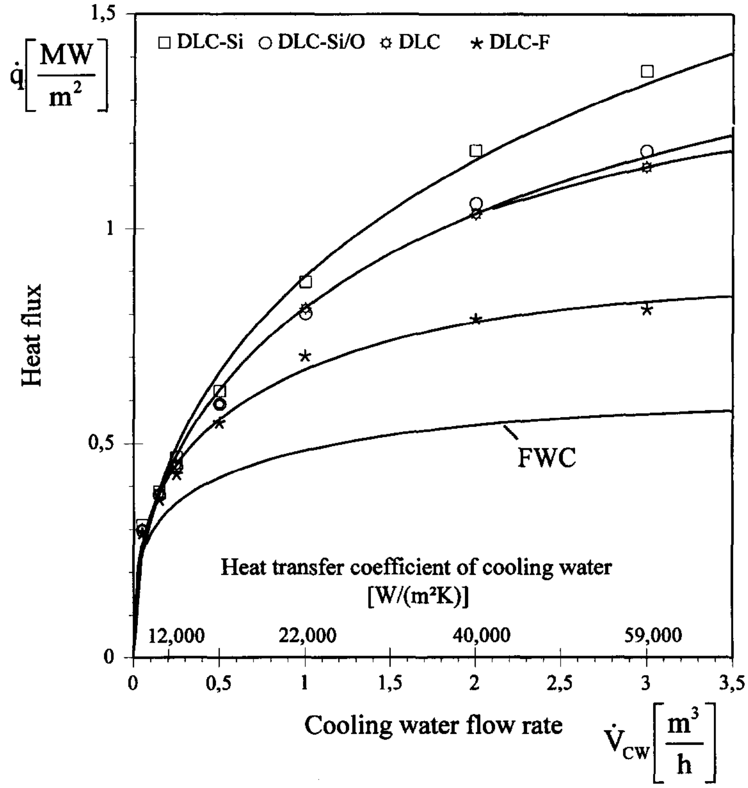


Fig. 7. Measured heat flux in dependence on the cooling water flow rate for coated copper discs with different coating systems (coating systems see Table 2, FWC measured for comparison).

ing systems with different wetting characteristics were applied as DWC promoters. The very well reproducible DWC measurements indicated the expected dependence of the heat transfer performance on the wettability of the surface which is expressed by the contact angle. For an angle of 90° stable DWC could be reached up to 1.54 MW m^{-2}. On surfaces with contact angles of 74 and 65°, the beginning of mixed condensation was observed at different flow densities of the condensate mass, thereby altering the slope of the dependence $\dot{q} = f(\Delta T)$ to negative values. No instabilities of the promoters could be determined during an operation time of approximately 500 h so far.

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