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Wärme- und Stoffübertragung

Heat transfer in the supercritical region with vertical upflow *

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Abstract. Thermodynamic properties like the specific heat capacity and the volumetric expansion coefficient show a maximum in the critical region under supercritical pressures. Others, like viscosity, undergo a strong change. Depending on the level and distribution of temperature over the cross section of the channel and of the ratio between heat flux and mass flow rate, an enhancement or an impairment of the heat transfer coefficient can occur.

Based on measurements with the refrigerant R12 a criterium is presented for predicting the onset of heat transfer impairment. The criterium is based on 2 dimensionless numbers. In addition correlations are reported which allow to calculate the heat transfer coefficient in R12 and water under supercritical pressures and which cover the region of impairment as well as that of enhancement.

Wärmeübertragung im überkritischen Bereich bei vertikaler Aufwärtsströmung

Zusammenfassung. Stoffwerte, wie die spezifische Wärmekapazität und der volumetrische Ausdehnungskoeffizient, zeigen ein Maximum im kritischen Gebiet unter überkritischem Druck. Andere, wie z.B. die Viskosität, erfahren eine starke Änderung. Abhängig vom Niveau und von der Verteilung der Temperatur über den Querschnitt des durchströmten Kanals und vom Verhältnis zwischen Wärmefluß und Mengenstrom kann durch das Verhalten der Stoffwerte eine merkliche Verbesserung oder auch eine empfindliche Verschlechterung des Wärmeübergangskoeffizienten verursacht werden.

Gestützt auf Messungen mit dem Kältemittel R12 werden Kriterien für die Vorhersage der Wärmeübergangs-Verschlechterung präsentiert. Diese Kriterien sind in Form von 2 dimensionslosen Zahlen formuliert. Zusätzlich werden Gleichungen mitgeteilt, welche den Wärmeübergangskoeffizienten in R12 und in Wasser für überkritische Drücke sowohl unter der Bedingung der Wärmeübergangs-Verschlechterung als auch der -Verbesserung vorhersagen lassen.

1 Thermodynamic properties in the critical and supercritical region

It is well known from the literature [1–12] that there can be an enhancement as well as an impairment of heat transfer if the heat-absorbing fluid is near its critical thermodynamic status under slightly supercritical pressure. The enhancement of heat transfer is used in several apparatus for chemical processing and for conversion of energy. Especially fossil-fired boilers of the so-called Benson-type use the favourable heat transport conditions at supercritical pressures of water. However, with an inadequate layout of the boiler tubes also an inverse situation can occur with a considerable impairment of heat transfer, which leads to unallowable high temperatures of the tube wall and by this to a failure of the boiler tubes.

At the critical point the specific heat c_p and the isobaric expansion coefficient β extend to infinity, as well known. Other properties like the viscosity η, the thermal conductivity λ, the enthalpy h and the density ϱ show a rapid change in their values when passing the critical point. Qualitatively this behaviour can be also observed at supercritical pressures, however then with slightly higher temperatures than the critical temperature. Instead of going to infinity the specific heat and the expansion coefficient show a maximum and the slopes of the other properties are less steep. As an example the behaviour of the thermodynamic properties of water at a supercritical pressure of 245 bar is presented in Fig. 1.

Enhancement and impairment of heat transfer are certainly depending on the gradients of certain thermodynamic properties in the boundary layer near the heat emitting wall. In a first rough and simple deliberation one, therefore, could conclude that enhancement can be expected if the specific heat is increasing with approaching the wall, i.e. if the thermodynamic status of the fluid is over the whole cross section of the tube on the left side of the maximum, in Fig. 2 marked as situation "A". The temperature at which the specific heat has its maximum under supercritical pressures is usually called pseudocritical temperature T_{pkr}. Consequently, if the situation in the boundary layer exceeds the pseudocritical temperature – in Fig. 2 marked as situation "B" – an impairment of heat transfer should be expected. This deliberation is

* Dedicated to Professor E. R. G. Eckert's 80th birthday

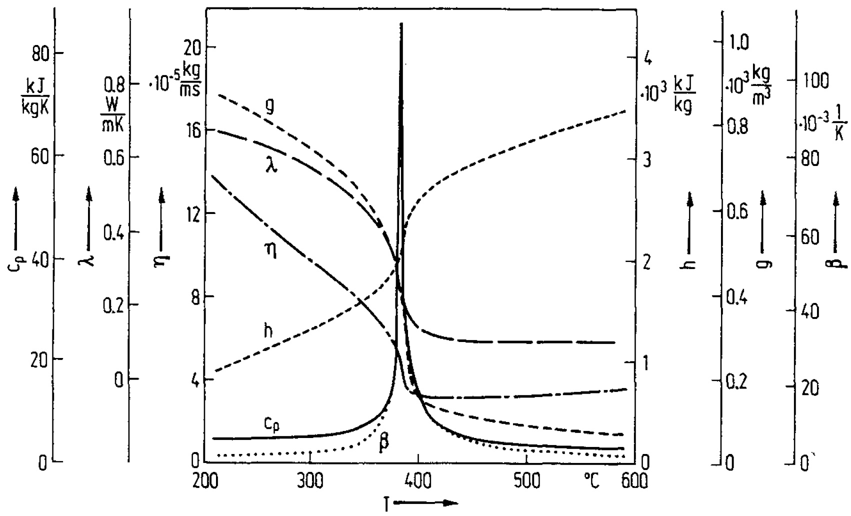

Fig. 1. Thermodynamic properties near the critical point under supercritical pressures (H_2O at 245 bar)

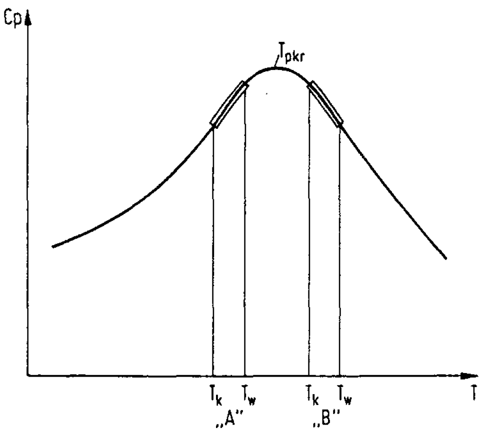

Fig. 2. Course of the specific heat in the boundary layer depending on the temperature level in the channel

simply based on the assumption that specific heat and expansion coefficient are the dominant properties for transporting the heat from the wall.

However, there are also other properties, namely the so-called transport properties – thermal conductivity and viscosity – which are at least as important as the specific heat and the thermal expansion coefficient for the heat transfer behaviour. These properties show no maximum but only a steep decrease in their values which, however, starts well before reaching the pseudocritical temperature.

2 Criteria for impairment of heat transfer

Studying the literature one finds several criteria for predicting impairment of heat transfer in the supercritical region. Some of them make buoyancy phenomena responsible, others regard the density gradient in the boundary layer as main influencing parameter and a third one applies a pseudo-film-boiling theory.

Jackson [13] assumed that due to the rapid change of the density with the temperature the fluid may undergo a buoyancy effect in its laminar sublayer and by this the velocity gradient at the border between this sublayer and an intermediate layer to the turbulent zone decreases, which finally results in a smaller heat transport contribution by turbulent effects. By using the Grashof-number and the Reynolds-number he formulated the criterium

$$\frac{Gr_K}{Re^{2.7}} < 10^{-5} \tag{1}$$

for impairment of heat transfer in the supercritical region with vertical upflow. Brassington [14] checked this criterium and found that it may be a necessary condition, however, that it is certainly not an adequate one. It has to be mentioned here that Jackson's idea of impairing the heat transfer by reducing the turbulent transport in the intermediate layer is not in full agreement with the common theories about the energy transport through boundary layers, because usually it is assumed that the largest resistance is in the laminar sublayer.

Based on stability criteria which Prandtl defined for horizontal layers, Polyakov [16, 17] developed a theory for heat transfer impairment in vertical supercritical flow, which takes in account buoyancy effects. For the turbulent motion he defines the dimensionless number

$$\Phi_g = 1.3 \cdot 10^{-4}\, \overline{Pr} \cdot Re_K^{2.75} \frac{Re_K^{1/8} + 2.4\,(\overline{Pr}^{\,2/3} - 1)}{\log Re_K + 1.15 \cdot \log(1 + 5\,\overline{Pr}) + 0.5\,\overline{Pr} - 1.8} \tag{2}$$

which must be smaller than the Grashof-number

$$Gr > \Phi_g \tag{3}$$

if impairment occurs. The Grashof-number

$$Gr = \frac{g\,\bar{\beta}\,\dot{q}\,D^4}{\lambda_K\, \nu_K^2} \tag{4}$$

is defined with the mean expansion coefficient $\bar{\beta}$

$$\bar{\beta} = \frac{1}{\varrho\,(T_F)} \cdot \frac{\varrho_K - \varrho_W}{T_W - T_K}. \tag{5}$$

The mean Prandtl-number $\overline{Pr}$ in Eq. (2) reads

$$\overline{Pr} = \frac{h_W - h_K}{T_W - T_K} \cdot \frac{\eta_K}{\lambda_K}. \tag{6}$$

Besides buoyancy effects Jackson also makes pseudo-film-boiling phenomena responsible for the impairment of heat transfer. In [1] he states that the heat transfer remains uninfluenced as long as only the laminar sublayer is heated up to the pseudocritical temperature. The thermal resistance in this layer remains approximately constant because the increasing of the specific thermal resistance is compensated by the decreasing of the laminar boundary layer thickness. Lee [7], however, assumes that this laminar layer may behave like a vapour film in subcritical boiling. Wood [18] found in experiments that the velocity profile near the wall is different from the usual form of turbulent velocity distribution if the wall temperature was above and the bulk temperature below the pseudocritical value. He found an accelerated laminar boundary layer and a retarded bulk flow. Petukhov [15] found in his measurements that impairment only occurred if the bulk temperature in the tube was below and the wall temperature was above the pseudocritical temperature. However, in addition certain hydrodynamic conditions of the bulk flow had to be fulfilled. He defined a so-called relative density gradient

$$\Pi_1 = \frac{\frac{1}{\varrho} \cdot \frac{\partial \varrho}{\partial \left(\frac{r}{D_i}\right)}}{Re \sqrt{\frac{\xi}{8}}} \qquad \zeta = (1.82 \log_{10} Re - 1.64)^{-2} \tag{7}$$

for fully developed turbulent transport. Following Petukhov's idea one can modify Eq. (7) by formulating:

$$\Pi_1 = \frac{\frac{1}{\varrho} \cdot \frac{\partial \varrho}{\partial r^+}}{Re_K \sqrt{\frac{\xi}{8}}} = $$

$$= \frac{1}{\varrho} \cdot \frac{\partial \varrho}{\partial r} \cdot \frac{\eta_K}{\dot{m}} \cdot \frac{1}{\sqrt{\frac{\xi}{8}}} \tag{8}$$

In Eq. (8) the derivative for the density with respect to the radius $r^+ = r/D_i$ can be reorganized in the following way:

$$\frac{\partial \varrho}{\partial r} = \frac{\partial \varrho}{\partial T} \cdot \frac{\partial T}{\partial r} = -\frac{\partial \varrho}{\partial T} \cdot \frac{\dot{q}}{\lambda_{\text{eff}}}. \tag{9}$$

The effective thermal conductivity λ_{eff} can be regarded as the sum of the molecular and the turbulent heat transport

$$\lambda_{\text{eff}} = \lambda_l + \lambda_t = \left(1 + \frac{\varepsilon}{\nu} \cdot Pr\right) \cdot \lambda. \tag{10}$$

Assuming that the contribution of the molecular transport is small one can write

$$\lambda_{\text{eff}} \approx \frac{\varepsilon}{\nu} \cdot Pr \cdot \lambda. \tag{11}$$

By substituting Eq. (11) into Eq. (9) and with the definition for the thermal expansion coefficient

$$\beta = -\frac{1}{\varrho} \cdot \frac{\partial \varrho}{\partial T}. \tag{12}$$

Eq. (8) reads

$$\Pi_1 = \text{const} \cdot \frac{\beta}{c} \cdot \frac{\dot{q}}{\dot{m}} \cdot \frac{\nu}{\varepsilon} \cdot \frac{1}{\sqrt{\frac{\xi}{8}}} \tag{8a}$$

which was already proposed in similar form by Petukhov.

Differently to the dynamic viscosity the kinematic viscosity ν near the critical point is not a strong function of the temperature. If we, in addition, make the simplified assumption that the eddy diffusivity and the friction factor ξ are only functions of the flow conditions but not of the temperature, we can incorporate ν/ε and ξ into the constant of Eq. (8a) because at given flow conditions both would approach a certain constant value when impairment of heat transfer occurs. By using a mean expansion coefficient $\bar{\beta}$ as given by Polyakov in Eq. (5) and by refering the specific heat of the fluid to the bulk temperature, we get the dimensionless number

$$\Pi_2 = \frac{\bar{\beta}}{c_{p_K}} \cdot \frac{\dot{q}}{\dot{m}}. \tag{13}$$

For describing the property effects unto the heat transfer, sometimes also a temperature relation is used, which in the special form

$$Ec = \frac{T_{pkr} - T_K}{T_W - T_K} \tag{14}$$

is sometimes called the Eckert-number. In Eq. (14) T_K stands for the bulk temperature and T_W for the wall temperature. With $Ec > 1$ or $Ec < 0$ the course of the specific heat is purely monotonic over the cross section of the tube or the channel, whereas with $0 \leq Ec \leq 1$ the specific heat has a maximum within the flow area. So according to the very first deliberations at the beginning of this chapter, the Eckert-number could also be used as simple criterium for determining the onset of heat transfer impairment. Comparing different models in the literature with measurements, it was found [19] that useful criteria for describing impairment of heat transfer at supercritical pressures could be the Eckert-number (Eq. (14)) and the modified Petukhov-number (Eq. (13)).

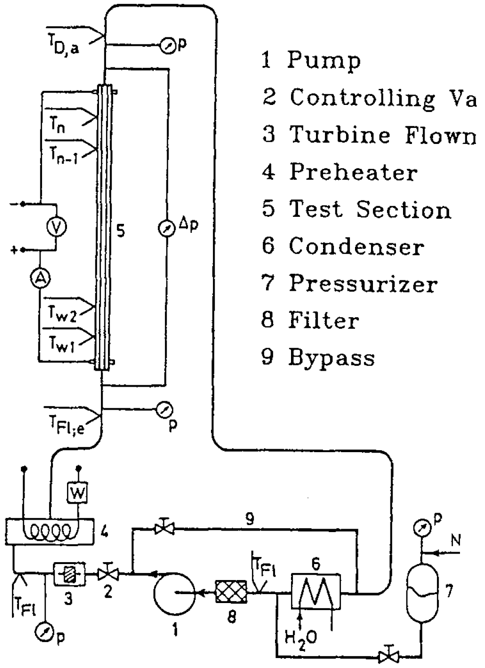

Fig. 3. Schematic view of the test facility

The dimensionless number in Eq. (13) is a function of the heat flux $\dot{q}$ and the mass flow rate $\dot{m}$, and the two thermodynamic properties which show a maximum at the pseudocritical temperature. This means, in addition to the course of the thermodynamic properties via the cross section of the tube or the channel, the onset of the impairment depends also on the ratio of the heat flux versus mass flow rate. This seems to be plausible, because with this increasing heat flux and decreasing mass flow rate an earlier heat transfer crisis can be expected.

3 Heat transfer measurements in the supercritical region

To check the validity of the criteria for the onset of heat transfer impairment and to get limiting values for the dimensionless groups in Eqs. (13) and (14), measurements with the refrigerant R12 were performed. For the measurements a loop was used, as sketched in Fig. 3. The loop was equipped with a vertical tube of 6 m length as test section. A total number of 93 thermocouples were distributed over the length of the tube wall to measure the wall temperature. The tube was electrically heated by using the tube wall as an ohmic resistance. In the experiments tubes with 2 different diameters were used, namely with 17.5 mm and 33.4 mm inside diameter.

Careful calibration tests were performed for determining the mass flow rate and for correcting heat losses. Experimental results are presented in [19] in detail. Here only these results shall be discussed, which demonstrate the behaviour of heat transfer coefficient and wall temperature in the impairment region mainly. With constant heat flux, as it is the case with ohmic resistance heating, the impairment is manifested by a sudden local increase of the wall temperature.

In Fig. 4 the course of the wall temperature (4a) and the heat transfer coefficient (4b) is plotted versus the tube-length. Instead of using length units like meters for the tube-length the enthalpy-increase of the fluid along the tube-length is plotted as abscissa. The wall temperature shows a local maximum between 100 and 150 kJ/kg for the higher heat fluxes applied in these experiments. The pressure was only slightly above the critical pressure of R12. In the graph of the heat transfer coefficient (4b) the impairment shows itself as local minima. In this graph the enhancement of heat transfer following the impairment is more impressive, however, as a comparison with Fig. 4a shows, this enhancement is influencing the wall temperature much less than the impairment. Therefore, the boiler constructor is more concerned about areas of impairment which can endanger the integrity of the tube walls.

In Fig. 5 the influence of mass flow rate density on the impairment is demonstrated. Very high and very low mass flow rates are less susceptible to impairment of heat transfer than the medium ones.

Finally, in Fig. 6 experiments with combinations of heat flux and mass flow rate parameters are presented, in which the ratio between heat flux density and mass flow

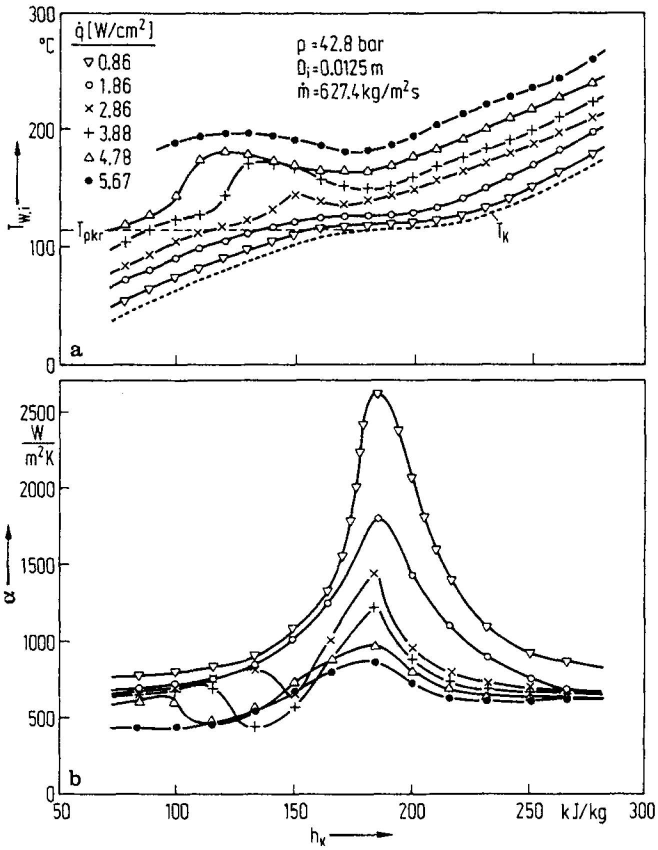

Fig. 4a and b. Heat transfer impairment under different heat flux densities. **a** wall temperature, **b** heat transfer coefficient

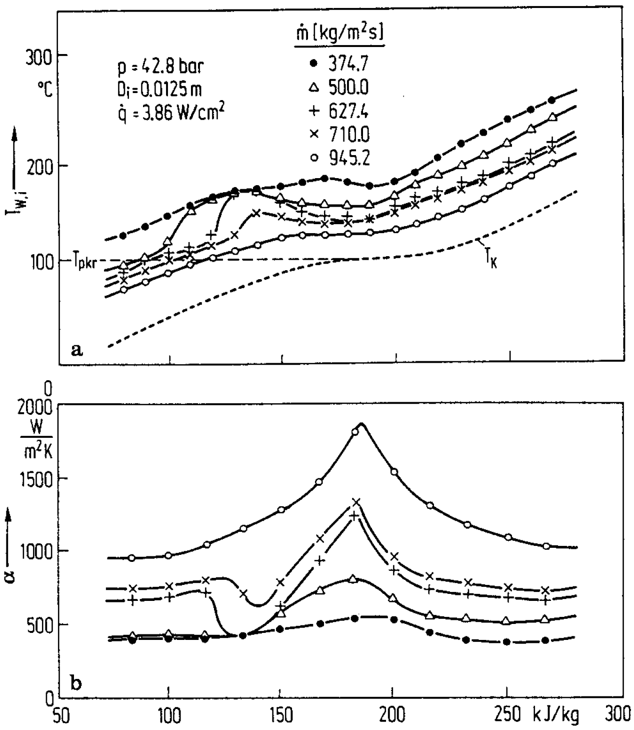

Fig. 5a and b. Heat transfer impairment with different mass flow rates. **a** wall temperature, **b** heat transfer coefficient

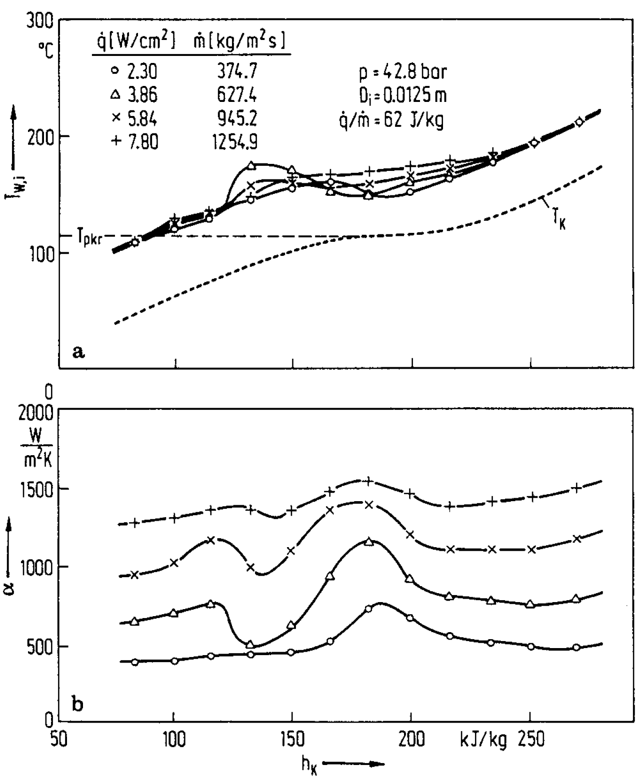

Fig. 6a and b. Heat transfer impairment under a critical ratio of heat flux versus mass flow rate. **a** wall temperature, **b** heat transfer coefficient

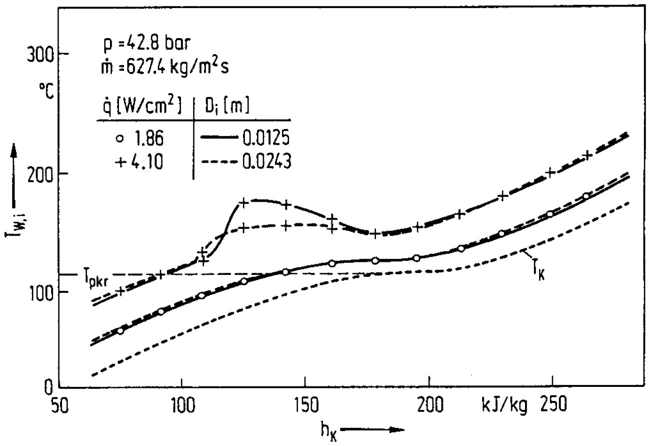

Fig. 7. Influence of tube diameter on heat transfer impairment

rate $\dot{q}/\dot{m}$ has a constant value and which show a strongly pronounced impairment of heat transfer with almost no or only a weak enhancement following the zone of impairment.

The tube diameter has also a certain influence on the development of impairment, even if it is small. Figure 7 gives an example. There is no difference in wall temperature and by this also in the heat transfer coefficient if no impairment occurs, however, with impairment the tube with the larger diameter shows a smaller increase than the more narrow tube.

4 Prediction of the impairment and correlations for heat transfer

From the experiments we saw that heat flux and mass flow rate have a strong influence on heat transfer deterioration and Fig. 7 mediated the impression that the impairing behaviour is especially pronounced at a certain ratio of heat flux versus mass flow rate. This ratio is included in the dimensionless number defined in Eq. (13). Certainly also the thermodynamic properties play an important role, which are a function of the temperature and the pressure, because with increasing pressure the pseudocritical temperature T_{pkr} moves to higher values. The thermodynamic properties mainly influencing the heat transfer in the critical region, namely the expansion coefficient and the specific heat, are contained in the dimensionless number of Eq. (13), too. The temperature course versus the cross section of the channel or the tube is taken in account in the Eckert-number (Eq. (14)).

So based on own measurements [19] and also on measurements of the literature [20, 21, 22], it was found that impairment occurs in R12 if

$$\Pi_{2_{R12}} > 9 \cdot 10^{-4} \qquad (15)$$

$$Ec_{R12} > 0.46 \qquad (16)$$

and in water if

$$\Pi_{2_{H_2O}} > 3 \cdot 10^{-3} \qquad (17)$$

$$Ec_{H_2O} > 0.70\,. \qquad (18)$$

The fact that the dimensionless numbers Ec and Π_2 are not identical for R12 and water gives the hint that there should be taken in account an additional dimensionless number, like the Prandtl-number. To investigate this, however, experiments have to be done with more different liquids and not only with R12 and water.

The experimental results allow also to produce correlations for predicting the heat transfer, not only under conditions of impairment but also in the enhanced area and in areas which are affected neither by impairment nor by enhancement. A screening of the data showed that one has to subdivide in three regions, which can be defined by the Eckert-number.

Region 1: $Ec > 1$; $T_K < T_W < T_{pkr}$

$$Nu_F = C_1 \cdot Re_F^{0.85} \cdot Pr_F^{0.33} \cdot \left(\frac{\varrho_W}{\varrho_K}\right)^{0.20} . \tag{19}$$

Region 2: $1 \geqq Ec \geqq 0$; $T_K \leqq T_{pkr} \leqq T_W$

$$Nu_K = C_2 Re_K^{0.87} \cdot Pr_K^{0.56} \cdot \left(\frac{\varrho_W}{\varrho_K}\right)^{0.21} \cdot \left(\frac{\overline{c_p}}{c_{p_K}}\right)^{0.57} \cdot Ra_K^{0.06} . \tag{20}$$

Region 3: $Ec < 0$; $T_{pkr} < T_K < T_W$

$$Nu_F = C_3 \cdot Re_F^{0.85} \cdot Pr_F^{0.62} \cdot \left(\frac{\varrho_W}{\varrho_K}\right)^{0.155} . \tag{21}$$

In these equations the mean specific heat $\overline{c_p}$ can be expressed by the equation

$$\overline{c_p} := [c_p]_{T_K}^{T_W} = \frac{1}{(T_W - T_K)} \int_{T_K}^{T_W} c_p\ (T, p)\, dT = \frac{h\,(T_W) - h\,(T_K)}{T_W - T_K} \tag{22}$$

and the Grashof-number and the volumetric expansion coefficient in the bulk of the flow are expressed as follows:

$$Gr_K = \frac{g \cdot \beta_K \cdot (T_W - T_K)\, D_i^3}{\nu_K^2} \tag{23}$$

$$\beta_K = \frac{\varrho_K}{(T_2 - T_1)} \left(\frac{1}{\varrho(T_2)} - \frac{1}{\varrho(T_1)}\right) = T_{1,2} = T_K \pm 1 . \tag{24}$$

The constants C_1, C_2 and C_3 in the Eqs. (19)–(21) are presented in Table 1 for R12 and for water. These equations do not yet allow to describe the heat transfer under deteriorated conditions, too. Under these conditions the following equation is recommended:

$$Nu_K = C_4 \cdot Re_K^{0.87} \cdot Pr_K^{0.61} \cdot \left(\frac{\varrho_W}{\varrho_K}\right)^{0.18} \cdot \left(\frac{\overline{c_p}}{c_{p_K}}\right)^{0.28} \cdot Ra_K^{0.12} . \tag{25}$$

The constant C_4 for R12 and water can also be taken from Table 1.

Comparisons between data predicted by these equations and measured values are presented in the Figs. 8–11. The Figs. 8–10 represent the situation with R12 and Fig. 11 gives a recalculation of the wall temperature of boiler tubes cooled by supercritical water. In this last figure the measured data used for comparison are taken from the literature [21, 28].

Table 1. Constants in the Eqs. (19–21) and (25)

	R12	H_2O
C_1	0.0102	0.0128
C_2	0.00166	0.00207
C_3	0.0094	0.0115
C_4	0.00038	0.00024

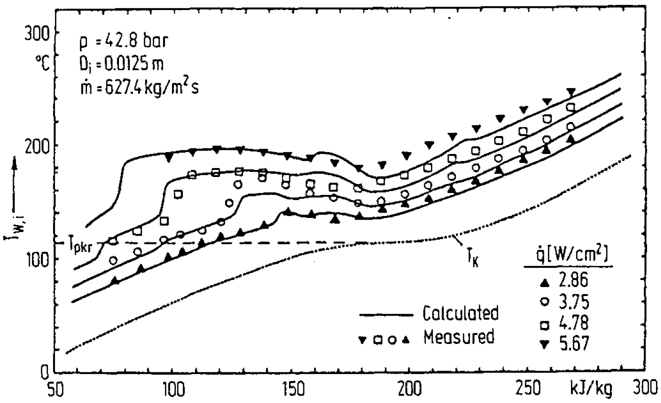

Fig. 8. Comparison of measured and calculated data in R12 for different heat flux densities ($p/p_{krit} = 1.2$)

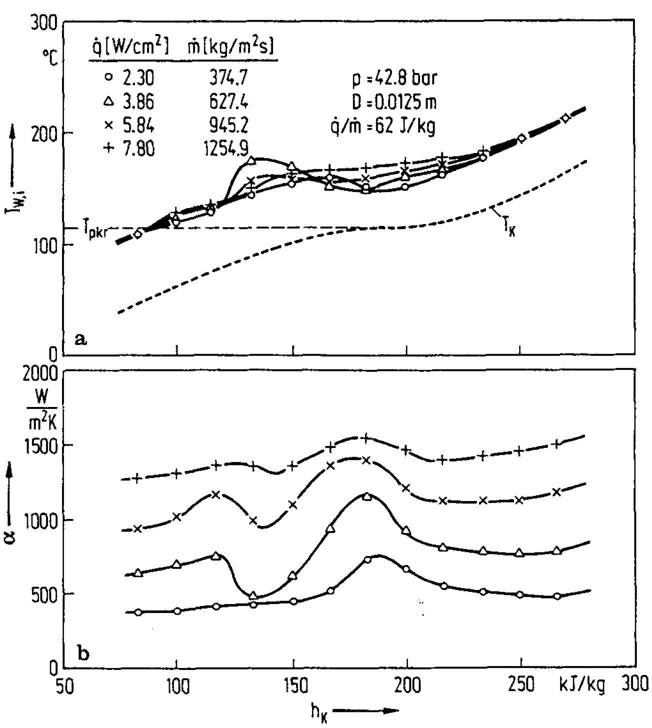

Fig. 9. Comparison of measured and calculated data for R12 ($p/p_{krit} = 1.1$)

The Figs. 8 and 9 show the situation with R12 for 2 different supercritical pressures. One can see that the heat transfer deterioration – and by this the increase in the wall temperature – is less expressed with increasing system pressure. The equations take also in account the influence of the tube diameter, as one can see from Fig. 10, which works via the Grashof-number and – to a smaller extent – also via the Reynolds-number. From Fig. 11 one can see that with water as heat transporting fluid the wall superheating under deteriorated conditions can reach quite unfavourable values, which can endanger the wall integrity.

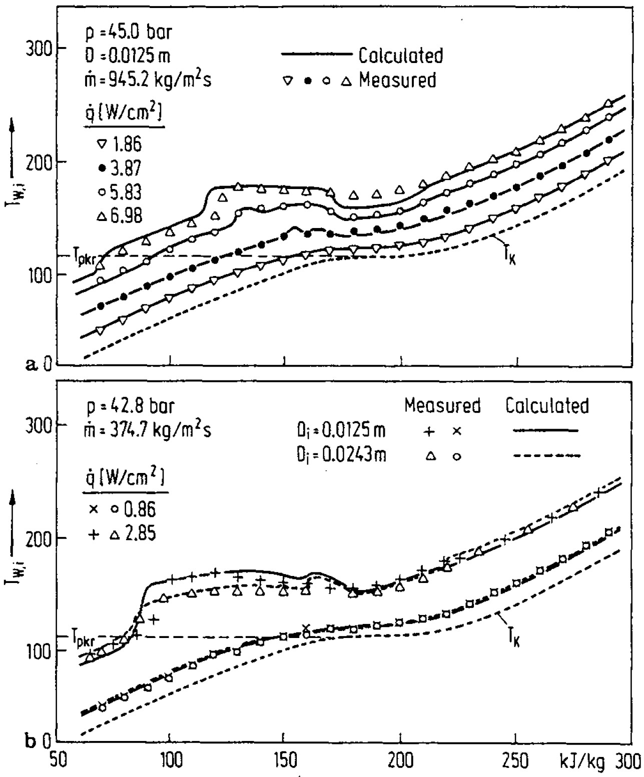

Fig. 10a and b. Comparison of measured and calculated data for R12 influence of tube diameter

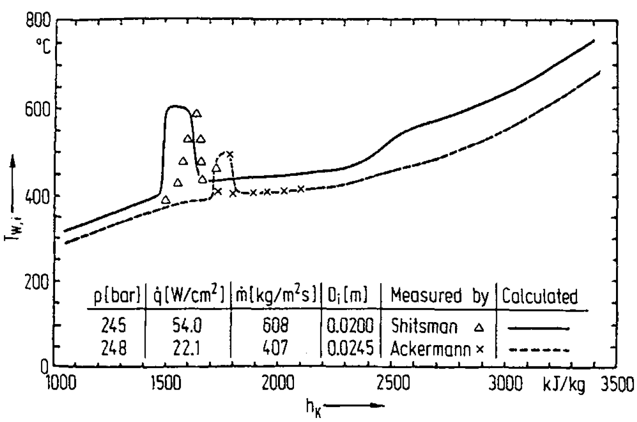

Fig. 11. Comparison of measured and calculated data for water

5 Conclusions

Heat transfer in the supercritical region can show a deteriorated behaviour under certain ratios of heat flux versus mass flow rate and under certain temperature distributions over the cross section of the heated channel or tube. The situation of the temperature distribution can be defined by an Eckert-number and the relation between heat added to the fluid and transported as sensible heat can be expressed by the heat flux density and the product of the mass flow rate density and the specific heat. Together with the volumetric expansion coefficient a dimensionless number can be defined from these variables.

Equations for predicting the heat transfer can be formulated in the well-known Dittus-Boelter-form. In the areas where impairment or enhancement may occur, in addition to the Reynolds- and the Prandtl-number also the Grashof-number has to be incorporated into the formulation.

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