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Experimental investigation of heat transfer of supercritical carbon dioxide flowing in a cooled vertical tube

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ABSTRACT

This work presents an experimental investigation on the heat transfer characteristics of a cooled vertical turbulent flow of supercritical carbon dioxide. The test section consists of two sectors (vertical tube-in-tube) connected in series by means of a U-bend. An integral method is used to treat the experimental data. Experimental results illustrate the influence of buoyancy forces on the heat transfer process. Results are presented in dimensionless form commonly used in the studies of mixed convection in heating. Correlations have been developed for upward and downward flows. The present results complete the literature on turbulent vertical mixed convection under cooling conditions.

1. Introduction

From the design requirements for industrial systems, such as heat pumps, cryogenic engines, nuclear power plants or extraction processes, numerous experimental and theoretical studies on supercritical fluids have been performed over the last 50 years. Most of these were carried out with water (see for example the review by Cheng and Schulenberg [1]), helium (Ito et al. [2], Valyuzhin and Kuznetsov [3]), hydrogen (Dziedzic et al. [4]) or carbon dioxide (Liao and Zhao [5], Pioro and Duffey [6]) and were focused on the thermal-hydraulic behaviour. Despite this intense research, there are significant gaps in the understanding of fundamental phenomena in mass and energy transfer in supercritical fluids. Moreover, almost all these studies were performed in heating conditions. Investigations on cooled flows of supercritical fluids in general, and supercritical carbon dioxide in particular, were performed in past years, when industry required substitutes to the current refrigerants. Nonetheless, the available database remains relatively limited.

Due to international agreements and European legislation, environmentally harmful refrigerants are being gradually phased out, and the use of natural substances has therefore attracted much attention. Among several possible candidates as substitute refrigerants, carbon dioxide (CO_2) may be a good successor for air-conditioning and residential heat pump systems due to its environmental benign properties (zero Ozone Depletion Potential, ODP, and a Global Warming Potential, GWP, equal to unity by definition) and its attractive physical and transport properties (Lorentzen [7]).

Above the critical point, the main observed phenomena are the disappearance of distinct liquid and vapour states and the large variations in the thermodynamic and transport properties at varying temperatures and pressures. Fig. 1 shows the effect of temperature on the specific heat C_p, the thermal conductivity λ, the dynamic viscosity μ and the mass density ρ at the constant pressure of 80 bars for carbon dioxide. All the thermophysical properties were evaluated using the Refprop 7.0 software [8]. The density and specific heat are plotted using the equation of state developed by Span and Wagner [9], while the viscosity and thermal conductivity are determined according to Vesovic and Wakeham [10]. These two studies can be considered as key references in the evaluation of CO_2 properties in the supercritical region (Kim et al. [11]). The uncertainties claimed by the authors are less than 1% and about 5%, respectively.

As the pressure approaches its critical value (73.8 bars), the variations become more severe. For a given pressure, the maximum of C_p is reached at the so-called pseudo-critical temperature T_{pc}. As seen in Fig. 1, large variations are encountered in all properties. Thermal conductivity, density and dynamic viscosity change continuously from a liquid-like state to a gas-like state. Specific heat variations are quantitatively high compared to other properties and take the form of a peak at T_{pc}. As the pressure approaches the critical value, the peak becomes sharper and narrower.

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Nomenclature

$C_p, \bar{C}_p$	specific heat, average specific heat J kg^{-1} K^{-1}
D	diameter, m
$\dot{Q}$	heat flow rate, W
G	mass flux, kg m^{-2} s^{-1}
g	acceleration of gravity, m^2 s^{-1}
Gr	Grashof number
h	specific enthalpy, J kg^{-1}
L	length, m
LMTD	logarithmic mean temperature difference, °C, K
$\dot{m}$	mass flow rate, kg s^{-1}
Nu	Nusselt number
Pr, $\bar{\text{Pr}}$	Prandtl number
Re	Reynolds number
Ri	Richardson number
S, dS	area, m^2
T	temperature, °C, K
$U, \bar{U}$	overall heat transfer coefficient, average overall heat transfer coefficient, W m^{-2} K^{-1}

Greek symbol

α	heat transfer coefficient, W m^{-2} K^{-1}
λ	thermal conductivity, W m^{-1} K^{-1}
μ	dynamic viscosity, Pa s
$\rho, \bar{\rho}$	density, average density, kg m^{-3}

Subscript and superscript

b	bulk
CO$_2$	refers to carbon dioxide
exch	exchange
exp	refers to experimental data
ext	external
h	hydraulic
in	inlet
int	internal
out	outlet
pc	pseudo-critical
w	wall
water	refers to water

2. Selected bibliography

Researches on heat transfer processes with supercritical fluid flows have been outgoing since the 1950s, mainly involving supercritical fluids as coolants in nuclear power plants or fuel in rockets, mainly in heating conditions (Pethukov [12], Jackson and Hall [13], Polyakov [14]). These researches have revealed that the heat transfer coefficient of supercritical fluid flows under heating conditions is strongly affected by heat flux and flow direction. A complete literature review on the heat transfer behaviour of supercritical carbon dioxide in heating condition is presented by Pioro and Duffey [6]. Due to rapid and large variations of thermophysical properties with small temperature changes, supercritical fluid flows are characterized by the development of mixed convection. A complete overview of experimental observations and of related phenomena of mixed convection in vertical tubes is given by Jackson et al. [15]. It appears in particular that in turbulent upward flows (turbulent aiding mixed convection, i.e., free and forced convection flows are in the same direction), a strong degradation of heat transfer is observed with the intensification of buoyancy forces. This is interpreted as a local "laminarization", i.e., an important reduction in turbulence level due to modification of the velocity profile under the effect of Archimedes forces (Aicher and Martin [16], Jackson and Hall [17]). For downward flows (turbulent opposing mixed convection), any development of free convection leads to intensification of heat transfer. These behaviours are commonly represented on a general single graph representing the growing influence of free convection on the heat transfer process. Aicher and Martin [16] announced that the features of mixed convection may be similar in heating and cooling conditions, but that no systematic and complete study was performed to confirm these considerations. Liao and Zhao [18] measured the convection heat transfer coefficient of turbulent flow of a supercritical carbon dioxide in heated horizontal and vertical miniature tubes of 0.7 mm, 1.4 mm and 2.16 mm. It was found that buoyancy effects were significant for all the flow directions and that the trends obtained were inconsistent with the existing literature of turbulent vertical mixed convection.

Contrary to heating, the cooling process has not been extensively investigated to date because of the slow development of practical applications. A complete review of the studies performed in cooling of supercritical CO_2 is given by Bruch [19]. Most of these studies were performed with horizontal flows, with no consideration of buoyancy and they present similar results both in their form and in the chosen approach. All available investigations showed that the heat transfer coefficient reaches a peak at a temperature close to the pseudo-critical temperature T_{pc} and discussed the effect of operating parameters such as mass flux or pressure. Huai et al. [20] experimentally studied the local and average heat

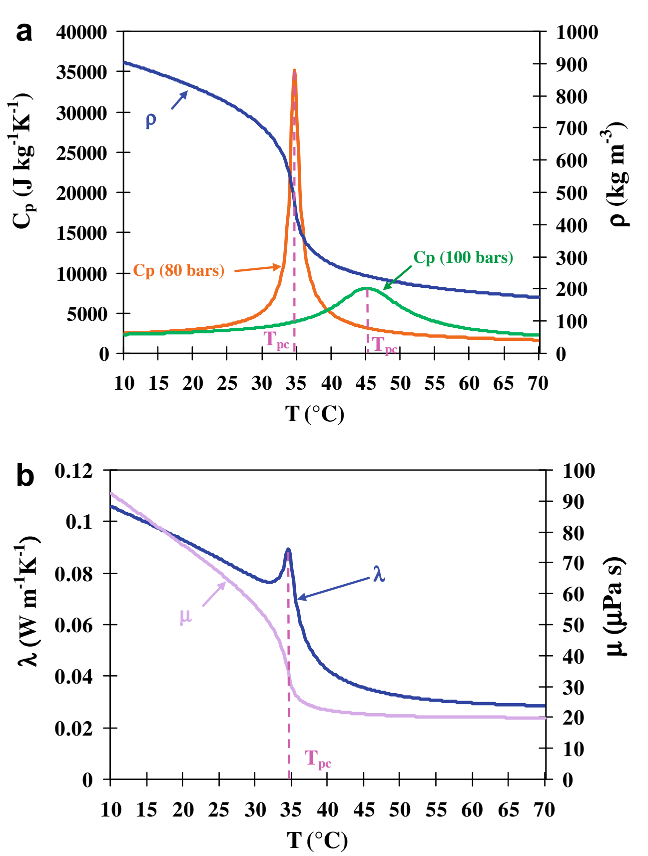

Fig. 1. Evolutions of thermophysical properties of CO_2; (a) specific heat and density; (b) viscosity and thermal conductivity.

transfer coefficient of supercritical carbon dioxide in a flat multi-port mini channel of 1.31 mm hydraulic diameter and discussed the effect of pressure and mass flux on heat transfer. Pitla et al. [21] conducted numerical and experimental investigations on a horizontal tube of 4.7 mm of inner diameter and 1.8 m length and proposed a new prediction model based on their data. Yoon et al. [22] conducted experiments using a 7.73 mm hydraulic diameter and discussed the effect of mass flux and pressure on heat transfer and pressure drop. Liao and Zhao [23] performed numerical simulations of laminar convective heat transfer of supercritical carbon dioxide in miniature vertical tubes and obtained typical velocity, temperature profiles and skin-friction coefficients with significant mixed convection effects. Liao and Zhao [5] measured the heat transfer coefficient for horizontal tubes of inner diameter ranging from 0.5 mm to 2.16 mm cooled at constant wall temperature and observed that the Nusselt number decreases with the tube diameter. The authors (Liao and Zhao [5]) discussed the influence of buoyancy in horizontal flows using a dimensionless parameter of mixed convection, the Richardson number (Ri). However, the deduced criterion does not completely explain the Nusselt variations.

This indicates that the study of the heat transfer process in cooled vertical flows, taking into account the effects of buoyancy forces, is an interesting field of research, not only for applications such as heat pumps or air-conditioning systems, but also to complete the missing data of mixed convection in cooling conditions.

3. Experimental apparatus

The experimental test loop constructed for the present study is schematically introduced in Fig. 2. Only the main CO_2 circuit is represented in this figure. Each heat exchanger represented in Fig. 2 is connected to a specific heating or cooling loop. The main CO_2 circuit is composed of a liquid CO_2 pump, a supercritical pre-heater, the test section, an absolute pressure transducer, two differential transducers, an expansion valve, a condenser, a CO_2 tank, a subcooler and a flow meter. The test loop was filled with CO_2 with purity of 99.5%.

The liquid CO_2 is circulated and compressed by a three head diaphragm pump (model LEWA ECOFLOW LDC3) which allows independent controls of discharge pressure and mass flow rate. The fluid passes through the pre-heater (5 kW hot water heating

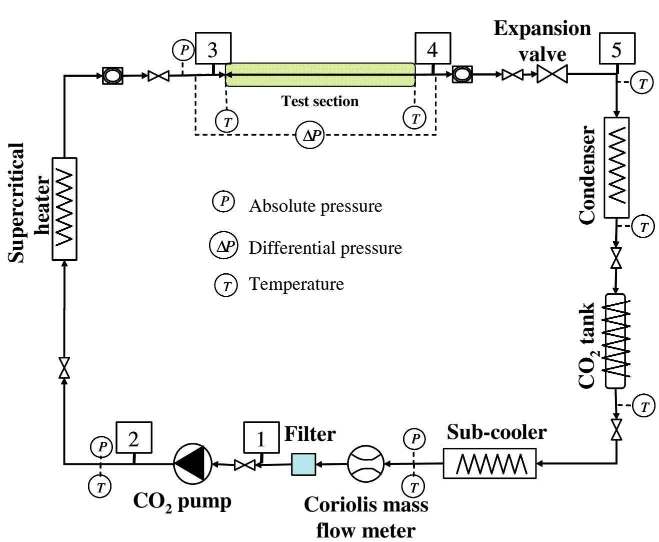

Fig. 2. Schematic diagram of the experimental facility. For an aim of simplicity, the vertical test section is schematically represented in horizontal position.

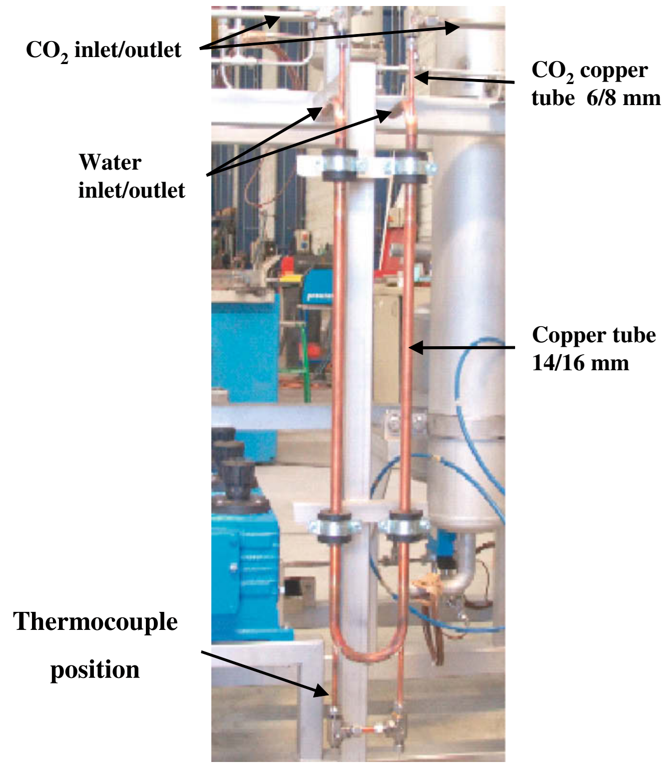

Fig. 3. Tube-in-tube test section without thermal insulation.

Table 1
Operating parameters.

P (bars)	T (°C)	$\dot{m}$ (kg h^{-1})	G (kg m^{-2} s^{-1})	Re	Flow direction
74–120	15–70	5–60	50–590	3600–1.8 × 10^6	Upward downward

exchanger) to adjust the temperature at the inlet of the test section where it is cooled. After entering the expansion valve, the pressure is lower than the critical pressure, and the fluid is condensed (5 kW cold water exchanger), stored in the CO_2 tank (connected to a 2 kW cold water cooling loop) and subcooled (2 kW cold water cooling circuit) to increase its density and its viscosity and to avoid cavitation before circulated by the CO_2 pump.

As illustrated in Fig. 3, the test section contains two subsections connected in series and forming a U-tube. Each subsection is a 0.75 m-long vertical tube-in-tube counter flow heat exchanger. CO_2 flows in the inner cooper tube of 6 mm diameter, cooling water flows in the annulus. The temperature at the inlet and the outlet of the test section (CO_2 side and water side) was measured using T-type thermocouples calibrated with an accuracy of 0.1 °C. Inlet pressure and pressure drop in the test section were measured with an error less than ±0.05%. The mass flow rate of carbon dioxide was measured with an accuracy of 0.1% using a Coriolis mass flow meter. Accuracy of cooling water side mass flow rate measurement was 0.5%.

Tests were conducted to quantify the influence of pressure drop in the test section on the evaluation of physical properties of carbon dioxide. Differences in the evaluation of the properties were less than 1%, assuming that the inlet absolute pressure can be used to calculate the thermophysical properties of CO_2 in the test section.

The ranges of the operating parameters in the test section are listed in the Table 1.

4. Data reduction

In studies of cooling of supercritical CO_2, the overall heat transfer coefficient U over the entire cooled length of each subsection is commonly calculated using a logarithmic method, LMTD method (Log Mean Temperature Difference), as expressed below:

$$\dot{Q} = U S_{exch} LMTD \tag{1}$$

where the logarithmic mean temperature difference $LMTD$ is given for counter current flows by:

$$LMTD = \frac{(T_{CO2,out} - T_{water,in}) - (T_{CO2,in} - T_{water,out})}{\ln\left(\frac{T_{CO2,out} - T_{water,in}}{T_{CO2,in} - T_{water,out}}\right)} \tag{2}$$

Because of the high temperature dependency of the thermophysical properties of CO_2 (and in particular the specific heat), the temperature difference between the cold water and the CO_2 varies non-linearly with heat load and distance from the inlet of the test section.

Thus an integral method proposed by Ngo et al. [24] was preferred to the classical logarithmic mean rate equation. Considering an infinitesimal region dS of the exchange surface of the test section, the heat flow rate between cold water and carbon dioxide $d\dot{Q}$ can be written as:

$$d\dot{Q} = U(T_{water} - T_{CO2})\,dS \tag{3}$$

where U is the overall heat transfer coefficient between water and carbon dioxide. The averaged overall heat transfer coefficient over the entire length of each subsection of the test section is defined by:

$$\bar{U} = \frac{1}{S}\int_0^S U dS = \frac{1}{S}\int_0^{\dot{Q}} \frac{d\dot{Q}}{T_{water}(\dot{Q}) - T_{CO2}(\dot{Q})} \tag{4}$$

For all experimental conditions, temperature variations of water side were less than 1 °C; leading to an approximation of constant water temperature along the test section:

$$T_{water}(\dot{Q}) = T_{water} \tag{5}$$

In the infinitesimal region dS, the exchanged heat can be expressed as:

$$d\dot{Q} = \dot{m}\,(dh)_{CO2} \tag{6}$$

where h is the specific enthalpy of the supercritical carbon dioxide. Eqs. (4)–(6) lead to the final expression of the overall heat transfer coefficient over the entire length of each subsection:

$$\bar{U} = \frac{\dot{m}}{S}\int_{hin}^{hout} \frac{dh}{T_{water} - T_{CO2}(h)} \tag{7}$$

The heat transfer coefficient on the CO_2 side is determined from the overall heat transfer coefficient as expressed below:

$$\frac{1}{\bar{U}} = \frac{1}{\alpha_{CO2}} + \frac{S_{exch,int}}{S_{exch,ext}\alpha_{water}} + \frac{S_{exch,int}}{2\pi\lambda_w L}\ln\left(\frac{D_{ext}}{D_{int}}\right) \tag{8}$$

where α_{water} is calculated using a forced convection correlation developed for heated turbulent flows (Dittus and Boelter [25]) with a correction due to annulus configuration (Pethukov and Roizen [26]):

$$Nu = 0.0243\mathrm{Re}^{0.8}\mathrm{Pr}^{0.4}0.86\left(\frac{D_{int}}{D_{ext}}\right)^{-0.16} \tag{9}$$

As mentioned previously, variations of cooling water temperature were typically less than 1 °C for all test conditions, allowing us to consider the water properties to be constant along the test section.

The CO_2 side heat transfer coefficient based on the integral method was compared with that calculated with the commonly used LMTD method and presented in Fig. 4 for upward flows. Similar behaviour is observed in downward flows. For all pressures, main differences between the integral and the LMTD methods are localized in the vicinity of the pseudo-critical temperature T_{pc} and these differences are damped with increasing pressure. These observations are related to the evolution of the specific heat C_p of supercritical CO_2 with temperature and pressure: the value of C_p peaks at the pseudo-critical temperature T_{pc} and the extent of this ride dampens with increasing pressure. Fig. 5 shows a direct comparison of heat transfer coefficients obtained with the two methods for different pressures and a downward mass flux of $G = 490$ kg m^{-2} s^{-1}. For pressures of 74 bars and 90 bars, differences are noted in the vicinity of T_{pc}, whereas no significant difference is observed in the liquid-like and the gas-like regions. At the high pressure of 120 bars, the choice of the data reduction method does not induce significant differences in the entire temperature range.

In most experimental conditions, the differences between the integral and the LMTD method do not exceed 5% far from the pseudo-critical temperature T_{pc} and 10% in the pseudo-critical region. Small temperature differences of the CO_2 along the test section (less than 2 °C) occurred for bulk temperature close to T_{pc}, while for bulk temperatures far from the pseudo-critical temperature, larger differences were recorded (more than 3–4 °C).

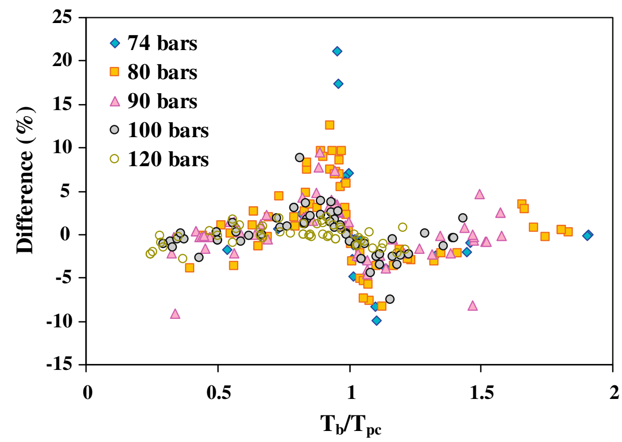

Fig. 4. Difference between integral method and LMTD method for different pressures, upward flows.

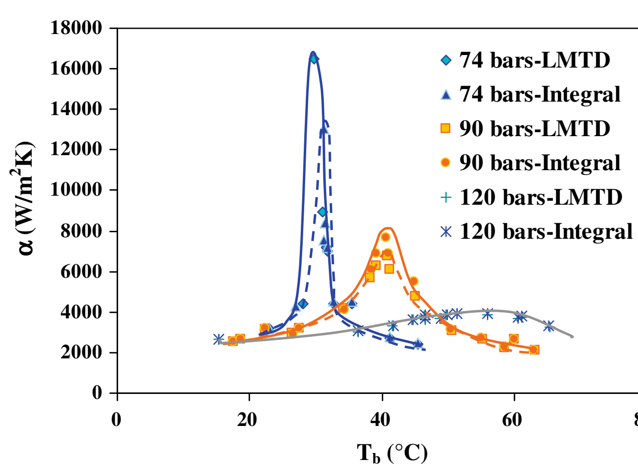

Fig. 5. Comparison of heat transfer coefficient for LMTD and integral methods, downward flows, $G = 490$ kg m^{-2} s^{-1}, LMTD method is represented in solid line, integral method in dash line.

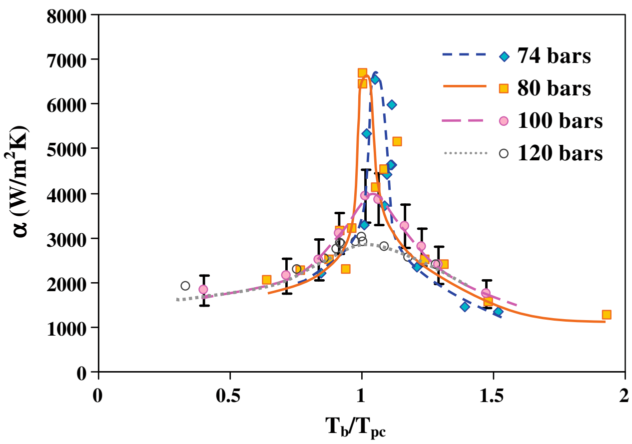

Fig. 6. Effect of pressure on heat transfer coefficient as a function of the dimensionless temperature T_b/T_{pc}. T_b is the bulk fluid temperature, $G = 490$ kg m^{-2} s^{-1}; upward flow.

From an uncertainty analysis, the accuracy of the heat transfer coefficient was generally less than ±20%. Typical uncertainties are illustrated in Fig. 6 at a pressure of 100 bars. With an aim of legibility, uncertainties are not presented in the rest of the figures.

5. Results and discussion

Measurements were performed by systematically studying the influence of the different operating parameters (pressure, inlet temperature, mass flow rate and flow direction) on the heat transfer coefficient.

5.1. Influence of the operating pressure

Figs. 6 and 7 show the local heat transfer coefficient of CO_2 for operating pressures ranging from 74 bars to 120 bars at a given mass flux of $G = 490$ kg m^{-2} s^{-1}, for downward flows. Similar behaviours are observed for upward flows, and are not presented here. The heat transfer coefficient is represented as a function of two different characteristic temperatures, (i) the average bulk fluid temperature T_b (Fig. 6) defined as

$$T_b = \frac{(T_{in} + T_{out})_{CO2}}{2} \tag{10}$$

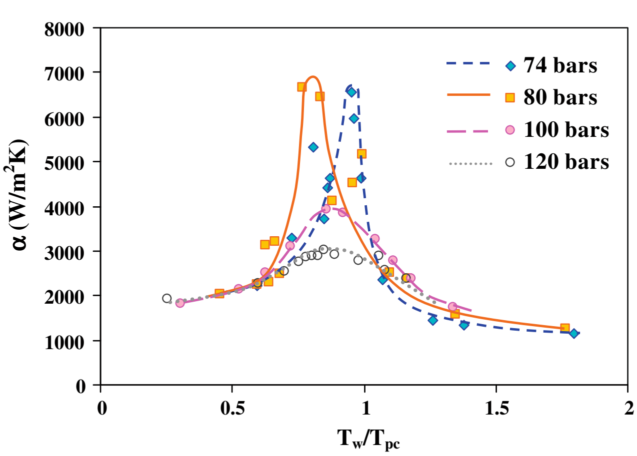

Fig. 7. Effect of pressure on heat transfer coefficient as a function of the dimensionless temperature T_w/T_{pc}. T_w is the wall temperature, $G = 490$ kg m^{-2} s^{-1}; upward flow.

(ii) the mean wall temperature T_w (Fig. 7) calculated from the mean bulk water temperature and the water-side heat transfer coefficient as:

$$T_{w,\mathrm{int}} = T_{b,water} + \dot{Q}\left[\frac{1}{\alpha_{water}S} + \frac{\ln(D_{ext}/D_{\mathrm{int}})}{2\pi\lambda_w L}\right] \tag{11}$$

For all pressures, the heat transfer coefficient is almost constant in the liquid-like region and in the gas-like regions. It reaches a peak near the pseudo-critical temperature, whose value is damped with increasing pressures. These behaviours are related to the corresponding evolutions of specific heat with temperature and pressure, as shown in Fig. 1: in the liquid-like and gas-like regions, the influence of pressure on the specific heat, and thus on the heat transfer coefficient, is weak. Near the pseudo-critical temperature, the higher the pressure the smaller the specific heat, and thus the smaller the heat transfer coefficient.

When the bulk fluid temperature T_b is used, the heat transfer coefficient values are centered at a value close to the pseudo-critical temperature T_{pc}. With other tested characteristic temperatures, a dispersion of the results is observed, indicating that the bulk fluid temperature T_b is the most adequate to represent the evolutions of the heat transfer coefficient.

5.2. Influence of the mass flux

The influence of the mass flux on heat transfer coefficient is illustrated in Figs. 8 and 9, for upward flows and downward flows, respectively, at an operating pressure of 80 bars.

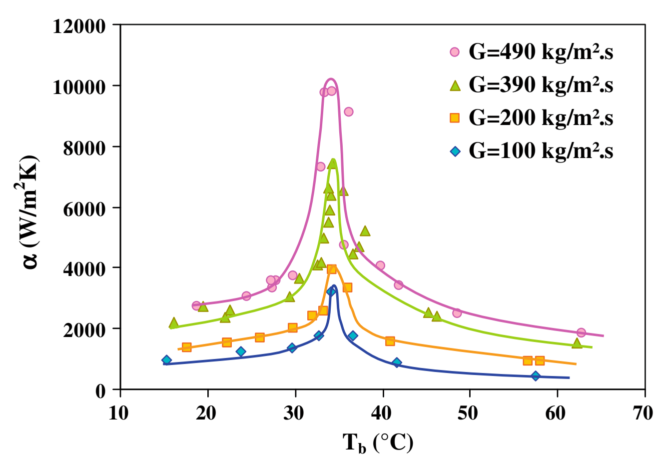

Fig. 8. Influence of mass flux on heat transfer coefficient; upward flow, $P = 80$ bars.

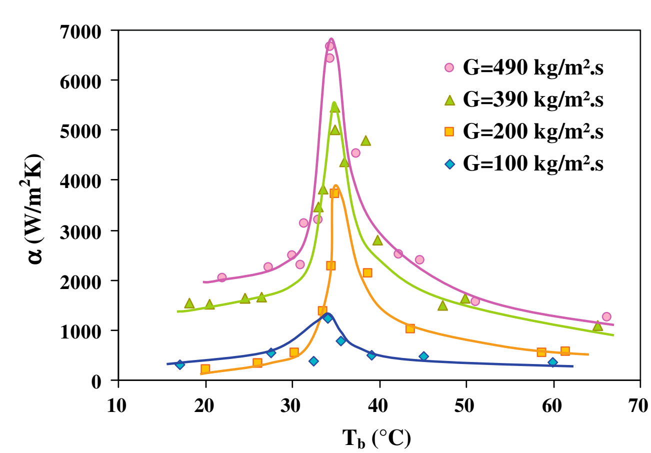

Fig. 9. Influence of mass flux on heat transfer coefficient; downward flow, $P = 80$ bars.

For upward flows, as in the case of a constant-property fluid flow, an increase in G causes an increase in the heat transfer coefficient of carbon dioxide due to an enhancement of turbulent diffusion.

For downward flows, different behaviours are observed, depending on mass flow rate and temperature. For $T_b \geqslant T_{pc}$, classical trend is observed, i.e., the heat transfer coefficient decreases with decreasing mass flux. For $T_b \leqslant T_{pc}$, the heat transfer coefficient decreases when mass flow rate drops from 490 kg m^{-2} s^{-1}to 200 kg m^{-2} s^{-1} Any further reduction of the mass flow rate results in an augmentation of the heat transfer coefficient. This particular behaviour, not observed in upward flows, suggests a different influence of free convection on thermal regime.

5.3. Influence of flow direction/mixed convection effects

The typical form of the local heat transfer coefficient with respect to the variations of bulk fluid temperature T_b and for different operating pressures is presented in Figs. 10 and 11 for constant values of mass fluxes of G = 490 kg m^{-2} s^{-1} and of G = 200 kg m^{-2} s^{-1}, respectively. Upward and downward flows are considered and compared for each value of pressure and mass flux.

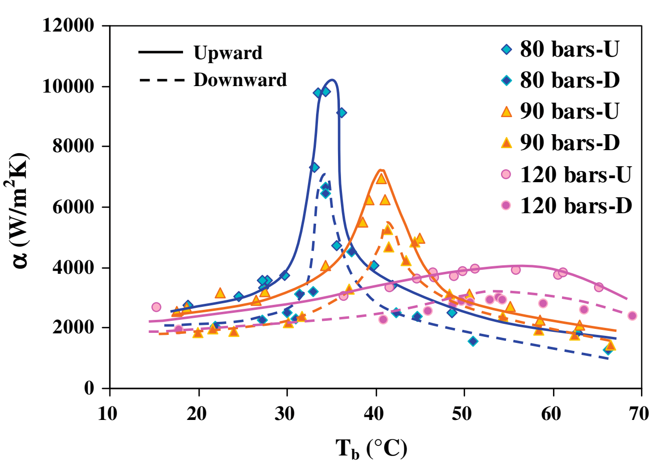

Fig. 10. Comparison of heat transfer coefficient for upward and downward flows; G = 490 kg m^{-2} s^{-1}; the solid line represents the upward flows; the dash line represents the downward flows.

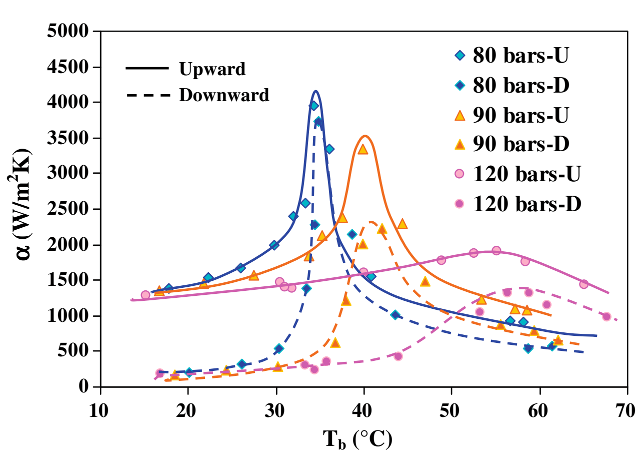

Fig. 11. Comparison of heat transfer coefficient for upward and downward flows; G = 200 kg m^{-2} s^{-1}; the solid line represents the upward flows; the dash line represents the downward flows.

In the gas-like region, the heat transfer coefficient reaches a limit value which only depends on mass flow rate. This indicates that forced convection is the predominant mechanism of heat transfer. However, due to the lack of measurements for temperatures above 65 °C, it is not possible to draw any final conclusion on the influence of flow direction on the heat transfer coefficient.

Strong differences with flow direction are noticed in the pseudo-critical region for all mass fluxes, indicating a strong influence of buoyancy forces on heat transfer. Except for the highest mass flow rate, similar behaviours are also observed in the liquid-like region.

The onset of mixed convection is related to the radial density gradient. When mixed convection occurs, the heat transfer coefficient is enhanced in upward flow and deteriorated in downward flow, in accordance with literature data on, respectively, opposing and aiding turbulent mixed convection.

These considerations are confirmed using a mixed convection parameter. The comparison between the buoyancy forces and the inertial forces can be made using the Richardson number defined by:

$$\mathrm{Ri} = \frac{\mathrm{Gr}}{\mathrm{Re}^2} \tag{12}$$

Jackson and Hall [17] have developed a semi-empirical parameter ($\mathrm{Gr}/\mathrm{Re}^{2.7}$), whose form is similar to that of the Richardson number, to characterize the influence of natural convection on turbulent vertical flows of heated supercritical carbon dioxide. Using this parameter, mixed convection has a significant effect on heat transfer when:

$$\frac{\mathrm{Gr}}{\mathrm{Re}^{2.7}} > 10^{-5} \tag{13}$$

Using the integral method, the Reynolds number Re is calculated as:

$$\mathrm{Re} = \frac{GD_h}{\mu_b} = \frac{GD_h}{h_{out} - h_{in}} \int_{hin}^{hout} \frac{dh}{\mu(h)} \tag{14}$$

The Grashof number Gr used by Jackson and Hall [17] is calculated as:

$$\mathrm{Gr} = \frac{(\rho_b - \bar{\rho})\rho_b g D^3}{\mu_b^2} \tag{15}$$

In Eq. (15), the influence of the radial density gradient (which is the main cause of the ignition of buoyancy flows) is expressed as the difference between the bulk density ρ_b and the average density $\bar{\rho}$ which is calculated according to the approximation proposed by Bae and Yoo [27]:

$$\bar{\rho} \approx \begin{cases} (\rho_w + \rho_b)/2 & \text{if } T_w > T_{pc} \text{ or } T_b < T_{pc} \\ \frac{[\rho_b(T_b - T_{pc}) + \rho_w(T_{pc} - T_w)]}{T_b - T_w} & \text{if } T_w < T_{pc} < T_b \end{cases} \tag{16}$$

Fig. 12 illustrates the evolution of the mixed convection parameter $Gr/\mathrm{Re}^{2.7}$ with the dimensionless temperature T_b/T_{pc} from high to low mass flux, at different pressures. In Fig. 12, the horizontal dotted line represents the limit value of the mixed convection parameter defined by Jackson and Hall [17] for significant effect of buoyancy forces in heating condition. Since similar evolutions are observed in upward and downward flows, only results for downward flows are presented. Conclusions drawn from Figs. 10 and 11 are confirmed: a significant mixed convection influence exists in the pseudo-critical region for all considered mass fluxes; in the liquid-like region, forced convection is predominant at high mass flux and buoyancy forces significantly affect the heat transfer process only for moderate to low mass fluxes; forced convection is the main mechanism of heat transfer for all mass fluxes in gas-like region. It has to be mentioned that at G = 100 kg m^{-2} s^{-1}, the experimental values of the $\mathrm{Gr}/\mathrm{Re}^{2.7}$ parameter in the gas-like re-

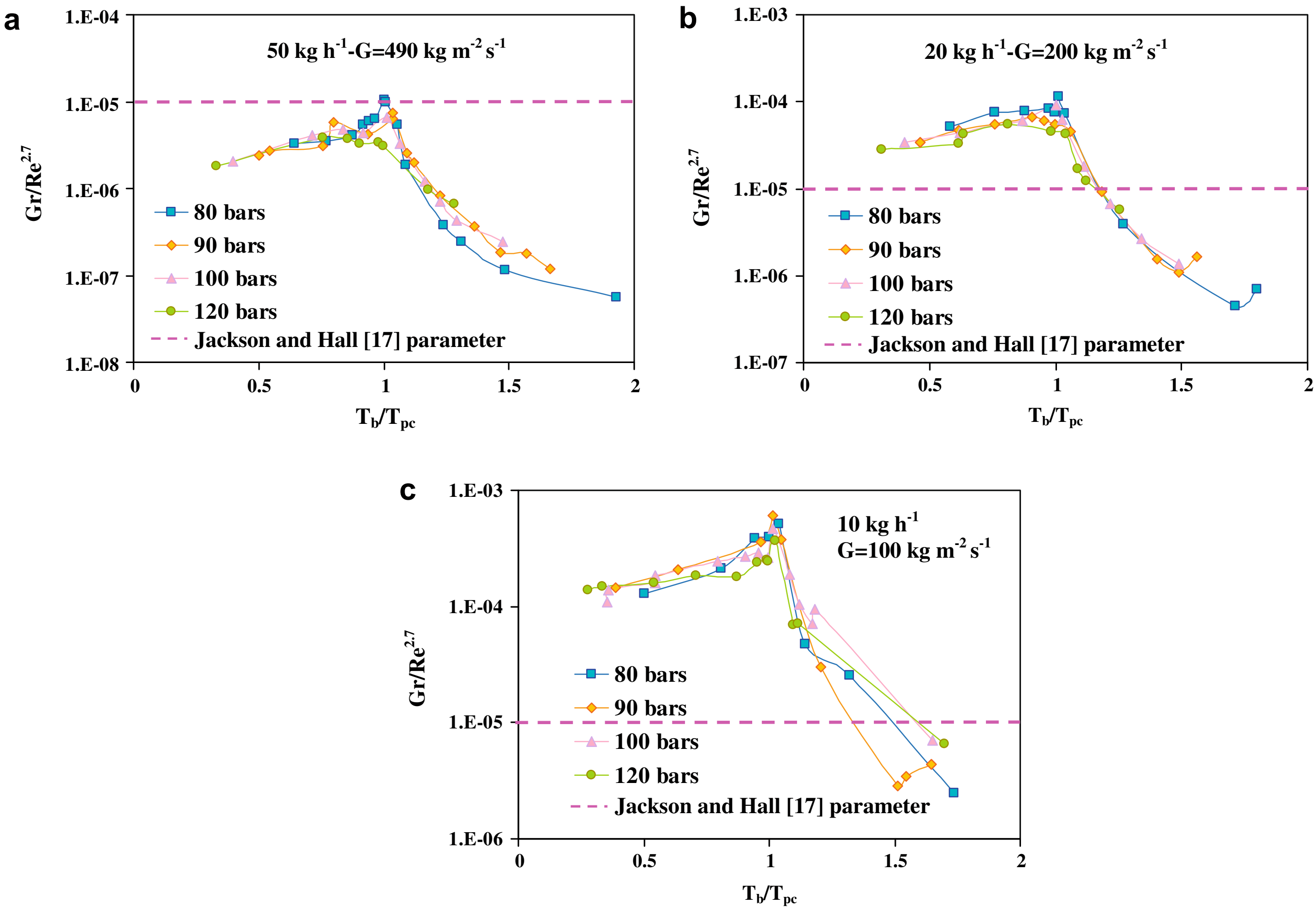

Fig. 12. Evolution of the mixed convection parameter with dimensionless temperature T_b/T_{pc}; (a) $G = 490$ kg m^{-2} s^{-1}; (b) $G = 200$ kg m^{-2} s^{-1}; (c) $G = 100$ kg m^{-2} s^{-1}.

gion are very close to the limit value defined by Jackson and Hall [17], indicating that mixed convection effects are significant in the whole range of temperatures.

In Fig. 13, results are presented in dimensionless form commonly used in studies on mixed convection in heating condition. The experimental Nusselt number is divided by a Nusselt number calculated with a pure forced convection correlation and is plotted as a function of the mixed convection parameter defined above.

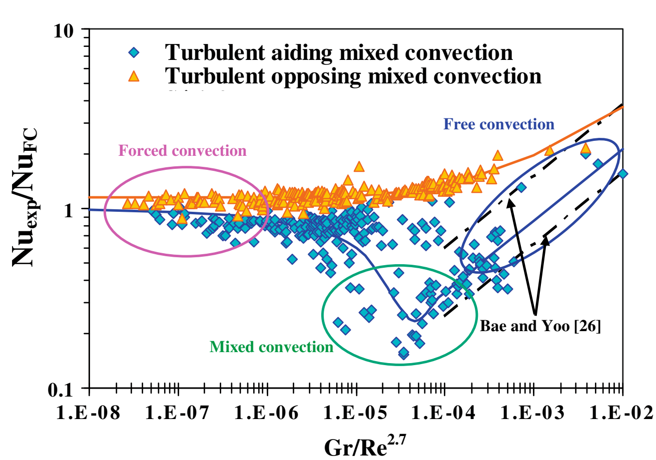

Fig. 13. Evolution of Nusselt number with the mixed convection parameter; FC refers to Forced Convection.

The chosen pure forced convection correlation is the one of Krasnoshchekov modified by Jackson and Hall [13]:

$$Nu_{FC} = 0.0183\mathrm{Re}_b^{0.82}\bar{P}r_b^{0.5}\left(\frac{\rho_b}{\rho_w}\right)^{-0.3} \tag{17}$$

The Prandtl number $\bar{P}r$ is calculated using the average specific heat $\bar{C}_p$ integrated between the bulk fluid temperature T_b and the wall temperature T_w, i.e.:

$$\bar{C}_p = \frac{h_b - h_w}{T_b - T_w} \tag{18}$$

and

$$\bar{P}r = \frac{\bar{C}_p}{h_{out} - h_{in}} \int_{hin}^{hout} \frac{\mu(h)}{\lambda(h)} dh \tag{19}$$

When the Nusselt number is plotted as a function of the parameter Gr/Re$^{2.7}$, single curves are obtained, respectively, for upward and downward flows, whose forms are similar to that obtained in mixed convection in heating conditions. Based on the Fig. 13, three different thermal regimes are observed:

- for low values of the mixed convection parameter Gr/Re$^{2.7}$, forced convection is the predominant mechanism of heat transfer, the influence of free convection is negligible and the ratio $\mathrm{Nu_{exp}/Nu_{FC}}$ is close to unity, for all flow directions. This also indicates a good agreement between the experimental measurements and the Jackson and Hall [13] forced convection correlation;

- as the parameter $Gr/Re^{2.7}$ increases, buoyancy forces are stronger and differences appear with flow direction. This confirms the observed behaviours (Figs. 10 and 11): heat transfer is enhanced in upward flows (turbulent opposing mixed convection) and deteriorated in downward flows (turbulent aiding mixed convection). This can be explained by modifications of velocity profiles and, thus, of turbulence production under the effect of buoyancy forces. In turbulent aiding mixed convection, this phenomenon is called "relaminarization" (Aicher and Martin [16], Jackson and Hall [17]). The lowest values of heat transfer coefficient are observed for $Gr/Re^{2.7}$ ranging from 2.10^{-5} to 4.10^{-5};
- for higher values of $Gr/Re^{2.7}$, free convection becomes predominant and experimental heat transfer is in good agreement with the empirical law developed by Fewster [28] with supercritical carbon dioxide under heating conditions:

$$\frac{Nu_b}{Nu_{FC}} = 15\left(\frac{Gr}{Re^{2.7}}\right)^{0.4} \tag{20}$$

5.4. Correlations for the prediction of the heat transfer coefficient

Basic contributions to the prediction of the heat transfer coefficient of cooled supercritical carbon dioxide only include correlations developed for horizontal flows. In these cases, the influence of mixed convection was considered only by Liao and Zhao [5] through a correlation including the Richardson number Ri. The experimental results are compared with four correlations developed for supercritical CO_2 under cooling condition, i.e., Huai et al. [20], Pitla et al. [21], Yoon et al. [22] and Son and Park [29]. Figs. 14 and 15 show a comparison between these predictive models and our experimental data, for moderate mass flow rate of $G = 200\ \mathrm{kg\ m^{-2}\ s^{-1}}$, where significant effect of buoyancy forces was noted.

For downward flows, large differences between the prediction models and the experimental data are noted, in particular in the liquid-like region and in the pseudo-critical region. These behaviours are related to the development of mixed convection, and as a consequence, to the degradation of heat transfer due to the relaminarization phenomenon. In the gas-like region, a correct agreement is observed with the correlations developed by Huai et al. [20], Pitla et al. [21] and Yoon et al. [22]. This confirms that without a significant effect of buoyancy forces, no influence of flow direction should be noticed. The Son and Park [29] correlation shows a peculiar behaviour, overestimating the limit value at high temperature. For upward flows, differences are still observed at peak values. In the liquid-like and gas-like regions, a correct agreement is noted, except with the Son and Park [29] correlation.

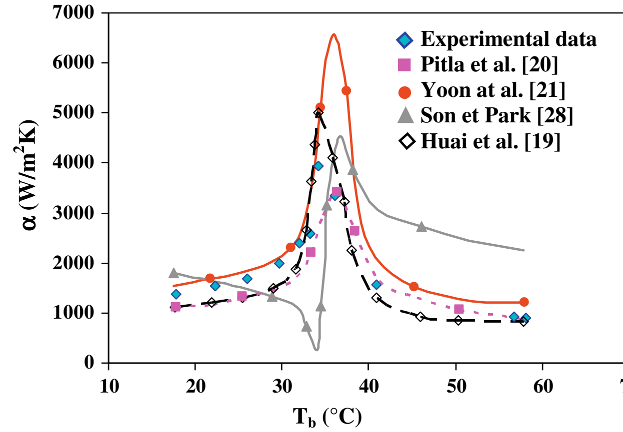

Fig. 15. Comparison of measured heat transfer coefficient with various correlations; $G = 200\ \mathrm{kg\ m^{-2}\ s^{-1}}$, $P = 80$ bars, upward flow.

The above cited correlations were developed for horizontal flows. Thus, new correlations are proposed from the current data on vertical turbulent aiding and opposing mixed convection under cooling conditions.Using the Jackson and Hall [17] parameter, in turbulent aiding mixed convection, the best fit on our experimental results leads to:

$$\begin{aligned} Gr/Re^{2.7} < 4.2\cdot 10^{-5} &: \frac{Nu_b}{Nu_{FC}} = 1 - 75\left(\frac{Gr}{Re^{2.7}}\right)^{0.46} \\ Gr/Re^{2.7} > 4.2\cdot 10^{-5} &: \frac{Nu_b}{Nu_{FC}} = 13.5\left(\frac{Gr}{Re^{2.7}}\right)^{0.40} \end{aligned} \tag{21}$$

In turbulent opposing mixed convection, the developed correlation is analog to those developed by Fewster [28] in heating conditions with supercritical carbon dioxide. The fit on the experimental data was made keeping the same exponents of the Fewster [28] correlation:

$$\frac{Nu_b}{Nu_{FC}} = \left(1.542 + 3243\left(\frac{Gr}{Re^{2.7}}\right)^{0.91}\right)^{1/3} \tag{22}$$

The accuracy of the developed correlations is illustrated in Figs. 16 and 17, where an excellent agreement is observed in opposing

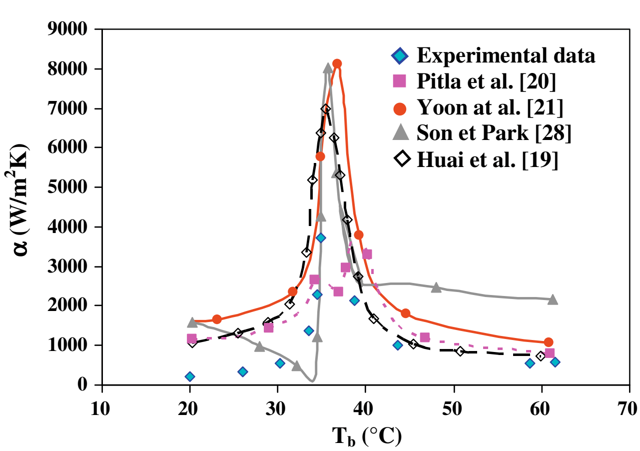

Fig. 14. Comparison of measured heat transfer coefficient with various correlations; $G = 200\ \mathrm{kg\ m^{-2}\ s^{-1}}$, $P = 80$ bars, downward flow.

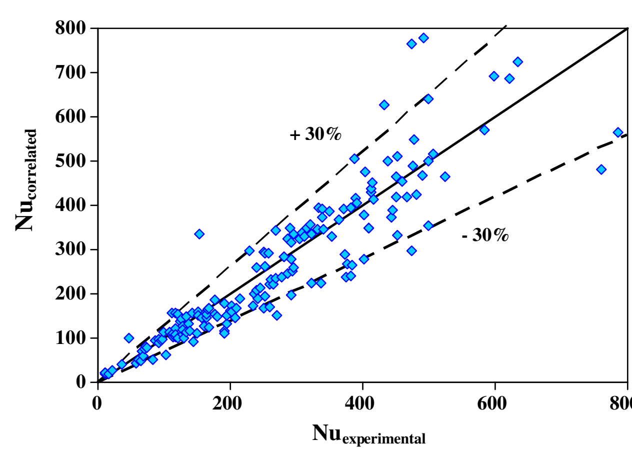

Fig. 16. Comparison of experimental data with the proposed correlation, downward flow.

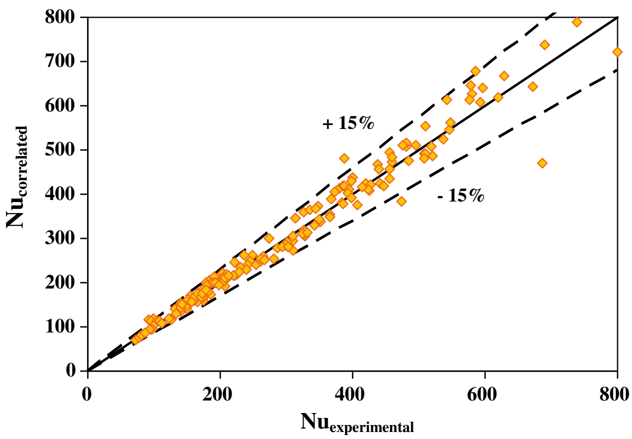

Fig. 17. Comparison of experimental data with the developed correlation, upward flow.

mixed convection (up to 90% of the 302 valid data points where predicted within the ± 15% accuracy limits). Due to dispersion of experimental data in the deteriorated region, the accuracy is about ± 30% in aiding mixed convection. The data dispersion in turbulent aiding mixed convection was commonly observed in both experimental (Fewster [28], Jackson et al. [15]) and numerical (Bae and Yoo [27]) studies of mixed convection in heating and might be explained by instabilities during the relaminarization phenomenon.

6. Conclusion

Heat transfer during cooling of turbulent vertical flows of supercritical carbon dioxide was experimentally investigated. The heat transfer coefficients were measured using specific data reduction and effects of operating parameters such as pressure, mass flux and flow direction on heat transfer process were analyzed. The main conclusions are summarized as follows:

- the heat transfer coefficient reaches a maximum for a bulk temperature close to the pseudo-critical temperature T_{pc}. The peak value decreases as the pressure increases. In the liquid-like and gas-like regions, no significant influence of pressure on the heat transfer coefficient is observed. These behaviours are related to similar evolutions of the specific heat with temperature and pressure;
- in upward flows, an increase of mass flux leads to an increase of heat transfer coefficient. In downward flows, experimental results revealed that there exists a limit value of the mass flow rate below which a reduction leads to an enhancement of the heat transfer coefficient. This may be explained by the development of mixed convection;
- direct comparison of the heat transfer coefficient for upward and downward flows shows that an influence of flow direction, and thus of buoyancy forces, exists, mainly in the liquid-like and the pseudo-critical regions. In the gas-like region a possible effect of free convection may be noticed for lowest values of mass flow rate;
- investigation of mixed convection was conducted according to a similar approach as in the studies under heating condition. In particular, the possible influence of mixed convection was investigated using the mixed convection parameter and criterion developed by Jackson and Hall [17]. A good agreement with experimental data in cooling was obtained. The Nusselt number was plotted as a function of the dimensionless mixed convection parameter. A single curve is obtained in turbulent aiding mixed convection and in turbulent opposing mixed convection. This indicates that the thermal behaviour of mixed convection is comparable in heating and cooling conditions and that the chosen dimensionless group can be used to reduce the data. The present results complete the missing data of the literature on turbulent vertical mixed convection in cooling conditions;
- specific correlations were developed for upward and downward flows, with accuracy of 15% and 30%, respectively.

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