SUPERCRITICAL HEAT TRANSFER TO REFRIGERANTS: ADVANCES ON A NEW EXPERIMENTAL TEST RIG

Jera Van Nieuwenhuyse[1]*, Steven Lecompte[1] and Michel De Paepe[1]

[1]Ghent University, Department of Electromechanical, Systems and Metal Engineering, Ghent, Belgium

*Corresponding Author: jera.vannieuwenhuyse@ugent.be

ABSTRACT

In a transcritical Organic Rankine cycle, the working fluid is pressurized above its critical pressure before heat is added in the vapor generator. Insight into supercritical heat transfer is therefore crucial to make an accurate design of the vapor generator. Supercritical heat transfer has already been widely investigated for water and CO_2. For these working fluids, the majority of the investigated geometries are vertical tubes. These results are however not generally applicable to the refrigerants used in Organic Rankine cycles due to the differences in operating conditions and fluid properties. So far, a limited number of refrigerants have been examined, nonetheless, there remain areas where further research is required. Therefore, this work provides an overview of the literature on experimental and numerical investigations on supercritical heat transfer to refrigerants. The remaining gaps in literature are highlighted and discussed. Based on this overview, it can be concluded that the main lack of knowledge lies in the large diameter sized geometries and in the low variety of tested refrigerants. To close this gap, a new experimental test rig is designed based on the experience from a previously built test rig. The design and development of this new test rig as well as the selection of the refrigerants that will be tested on the test rig are discussed in the current work. The test rig will be able to examine different low Global Warming Potential refrigerants with a wide range of critical temperatures. The unique data generated will contribute to the understanding of supercritical heat transfer and enable more accurate sizing of vapor generators for their application in transcritical Organic Rankine cycles.

1 INTRODUCTION

Transcritical Organic Rankine cycles (TORCs) have been put forward as novel heat-to-power conversion cycles for low-grade heat sources such as geothermal, solar and waste heat. A conventional technology such as the steam/water Rankine cycle cannot be applied efficiently under these operating conditions, as low-grade heat sources consist of low-to-medium temperature heat. Therefore, alternative heat-to-power cycle systems with alternative working fluids are required, such as the Organic Rankine cycle system. This system is already commercially available and mature, however, in order to improve the rather limited efficiency of this cycle, different adjustments to the basic cycle configuration are currently under investigation as well, research ranging from theoretical assessments to practical implementations. These adjustments include alteration of cycle design (e.g. by implementing internal regeneration, by adding different evaporation pressure levels, etc.), alteration of the working fluid (e.g. by using zeotropic mixtures) or optimization of cycle components (e.g. of the pump, expander or heat exchangers) (Lecompte *et al.*, 2015). In a transcritical ORC, the cycle design is altered as heat is added by the heat source to the working fluid which is under supercritical conditions. The working fluid is thus first pressurized above its critical pressure before it enters the heat exchanger. This reduces the temperature differences in the heat exchanger and enables increased cooling of the heat source. By bypassing the isothermal two-phase region during heat addition a better thermal match exists between the heat source and the working fluid (Lecompte *et al.*, 2015, Schuster *et al.*, 2010). Applications with supercritical heat transfer are not new. Many mature applications exist such as water-cooling of nuclear reactors or supercritical steam generators (Pioro *et al.*, 2004). In recent years, more novel applications such as supercritical CO_2 power and cooling cycles and supercritical heat pumps have also come forward.

In order to design the heat exchanger in TORC systems, which is called a vapor generator instead of an evaporator, accurate knowledge about supercritical heat transfer to the working fluid is required. However, correlations which hold at subcritical conditions cannot be applied in this region as the working fluid undergoes large and drastic thermophysical property variations when being heated in its near supercritical region. This is illustrated for R1234yf in Figure 1, which has a critical temperature T_c of 368.85 K and a critical pressure p_c of 3.38 MPa. The properties depicted in the figure are calculated with CoolProp v6.3.0 (Bell *et al.*, 2014). In the figure, the variations of specific heat capacity c_p, density ρ, thermal conductivity k and dynamic viscosity μ are depicted in function of temperature, for different supercritical pressures (expressed relative to p_c). When passing the pseudo-critical temperature T_{pc}, which is the temperature at a certain supercritical pressure where the isobaric specific heat capacity reaches a maximum, the fluid changes from a liquid-like to a vapor-like fluid (Yoo, 2013). These large property variations over a small temperature range will heavily influence heat transfer (Pioro *et al.*, 2004). For example, when a fluid flowing through a tube is heated from the outside at a supercritical pressure, fluid closer to the tube wall will become significantly less dense than fluid located in the bulk of the tube when the pseudo-critical temperature is reached. This will induce a secondary flow as less dense fluid located near the tube wall will flow upwards and denser fluid located near the bulk will flow downwards due to gravity (buoyancy effect). Therefore, constant property correlations such as the Dittus-Boelter or Gnielinski correlation cannot be applied to predict the heat transfer on the working fluid side, and specific correlations for fluids flowing under supercritical conditions need to be developed.

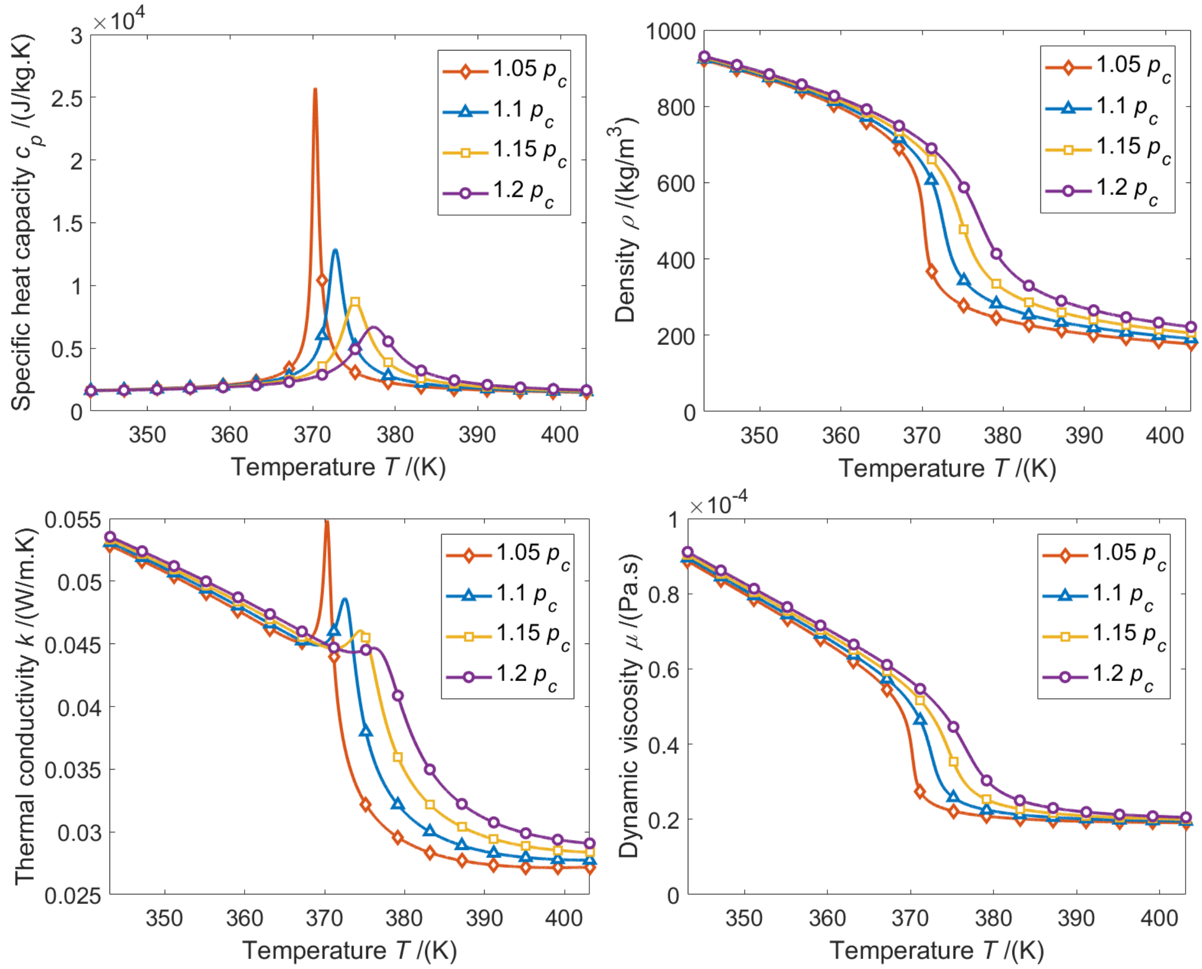

Figure 1: Thermophysical property variations of R1234yf

2 SUPERCRITICAL HEAT TRANSFER TO REFRIGERANTS

As already mentioned, supercritical heat transfer applications are not new. Consequently, the heat transfer under these conditions has already been studied, both numerically and experimentally. The large majority of these studies have focused on water (for its application in e.g. steam generators, water reactors), helium (for cooling of magnets) or CO_2 (as a simulant fluid for water) and experimental data available is mainly for vertical flows (Pioro *et al.*, 2004, Yoo, 2013, Duffey and Pioro, 2005). In addition, hydrocarbon fuels have also been investigated for their use as rocket engine coolant (Huang *et al.*, 2016). In these studies, several heat transfer correlations and buoyancy criteria (which predict when the buoyancy effect starts influencing the convective heat transfer as it transitions from forced to mixed convection) have been developed. However, due to differences in experimental conditions and fluid properties, these results are not generally applicable to refrigerants. Certain buoyancy criteria were found to be able to qualitatively predict the onset of buoyancy, but the threshold values differed significantly, and depending on operating conditions, heat transfer correlations only had low prediction capability (Tian *et al.,* 2018, 2019a, Cui and Wang, 2018). Research on refrigerants is much more scarce compared to the other fluids mentioned above. For vertical flow under heating conditions, several studies exist on supercritical heat transfer to R12, R22 and R134a, where the refrigerant served as a simulant fluid for water. Cui and Wang (2018) investigated vertical flow of R134a applied to TORCs. Other TORC working fluids investigated in vertical flow configuration are R245fa (He *et al.,* 2018a), R1233zd(E) (He *et al.*, 2018b) and hexamethyldisiloxane (MM) (Fu *et al.*, 2018, Xu *et al.*, 2020). R134a is the most widely investigated refrigerant under horizontal flow conditions. Experimental measurements were performed by Tian *et al.* (2019a, 2020), Wang *et al.* (2019a ,2019b, 2019c) and Cui *et al.* (2019) and numerical simulations were performed by Tian *et al.* (2019b) and Cui *et al.* (2019). In addition, Tian *et al.* (2020) did the same experimental and numerical campaign with R1234yf as well and compared the results of the two refrigerants to each other. Other refrigerants that have been investigated for horizontal flow are R125, which was experimentally investigated by Lazova *et al.* (2017), and NOVEC 649 which was investigated by Fu and Lin (2020). In Table 1 an overview is provided of the mentioned studies for horizontal flow, together with the investigated refrigerant, hydraulic diameters (d_h), and whether the work is numerical (N) or experimental (E). Hydraulic diameters are mentioned for the studies of Wang *et al.* (2019a, 2019b, 2019c) as the investigated geometries here also included a micro-finned and a ribbed tube.

Table 1: Overview of literature on supercritical heat transfer to refrigerants flowing horizontally, E = experimental, N = numerical.

Author	Refrigerant	d_h	Type of study
Tian *et al.* (2018)	R134a	d_i = 10.3; 16 mm	E
Cui *et al.* (2019)	R134a	d_i = 8 mm	N + E
Wang *et al.* (2019a)	R134a	d_h = 7.85 mm; d_i = 10.3 mm	E
Wang *et al.* (2019b)	R134a	d_h = 7.85 mm	E
Tian *et al.* (2019a)	R134a	d_i = 10.3 mm	E
Tian *et al.* (2019b)	R134a	d_i = 10.3 mm	N
Wang *et al.* (2019c)	R134a	d_h = 15.5 mm	E
Tian *et al.* (2020)	R134a, R1234yf	d_i = 10.3 mm	N + E
Lazova *et al.* (2017)	R125	d_i = 24.77 mm	E
Fu and Lin (2020)	NOVEC 649	d_i = 2, 4, 6 mm	E

From this table and the literature overview provided above, it can be concluded that refrigerants have already been investigated, mainly under vertical flow configuration. Flow direction, however, plays a major role in supercritical heat transfer. For vertical flows, the buoyancy force acts in the same direction as the flow direction, in horizontal flows it acts perpendicular. Therefore results on vertical tubes are not directly applicable to other orientations. In addition, the number of investigated refrigerants is limited. To the authors' knowledge, for horizontal flow under heating conditions, only four different refrigerants have been investigated, with most of the studies focusing on R134a. Nevertheless, a wide database of tested refrigerants is required in order to create general heat transfer correlations which can be applied to a large range of refrigerants. Finally, all authors, except one, performed measurements or simulations on tubes with a hydraulic diameter equal or smaller than 16 mm. As diameter size also influences heat transfer under supercritical conditions (buoyancy effects increase in larger diameter tubes), additional measurement campaigns on larger diameter tubes are necessary to achieve a full understanding of supercritical heat transfer to horizontal flowing refrigerants. Moreover, for TORC applications, a horizontal shell-and-tube configuration is considered more suitable for the vapor generator design and larger tube diameters correspond to practical (industrial) applications (Lazova *et al.*, 2016, Lazova, 2020).

In this work, the design and development of a new experimental test rig will be discussed. The test rig will be able to measure the heat transfer to different refrigerants in their near supercritical region, flowing horizontally in a large diameter tube. The data generated by this test rig will contribute to the understanding of supercritical heat transfer to refrigerants under TORCs conditions, and will enable more general correlation development in order to accurately size vapor generators.

3 DESIGN OF EXPERIMENTAL TEST RIG

3.1 Selection of refrigerants and measurement range

The choice of an appropriate working fluid in ORC systems is not straightforward and can be based on multiple criteria, such as cost, performance and safety requirements. In addition, the environmental impact of a refrigerant is becoming an increasingly important factor (Chen *et al.*, 2010). Therefore, the refrigerants that will be investigated with the new test rig were chosen based on their environmental impact, meaning zero (or low) Ozone Depletion Potential (ODP) and low Global Warming Potential (GWP). Furthermore, the conversion efficiency in low-grade heat conversion systems is taken into account (Molés *et al.*, 2017, Yang *et al.*, 2019). Actually, the energy use required to manufacture the refrigerants should also be an important environmental aspect to take into account when selecting a working fluid. However, available data on this is not easily accessible and this aspect is often not considered in literature when selecting a suitable working fluid. Therefore, refrigerant manufacturers should make this information publicly available in order to be able to make a fairer comparison between different refrigerants. Table 2 gives an overview of the refrigerants under consideration. The critical properties in the table as well as the properties used in calculations in order to design the test rig are calculated with REFPROP v10.0 (Lemmon *et al.*, 2018).

Table 2: Overview of refrigerants under consideration.

Refrigerant	T_c [K]	P_c [MPa]	ODP [-]	GWP_{100} [-]
R1234yf	367.85	3.38	0	4
R1234ze(E)	382.51	3.63	0	6
R1336mzz(E)	410.85	3.15	0	18
R1224yd(Z)	428.69	3.34	0[*]	1
R1233zd(E)	439.6	3.62	0[*]	1

As measurements on large tube diameters will be performed, a tube with an inner diameter of 24.77 mm was used as design diameter. With a test section length L of 4 m (large enough to attach a high amount of thermocouples to the tube), and heat fluxes ranging from 10 - 100 kW/m^2 (covering a large

[*] R1224yd(Z) and R1233zd(E) are hydrochlorofluoroolefins, indicating they contain a chlorine atom. This means their ODP is not completely zero. However, due to their very short atmospheric lifetime they have a minimal effect on the stratospheric ozone and are not considered as Ozone Depleting Substances (EPEE, 2018).

part of the application range in TORCs (Cui and Wang, 2018)), this results in a maximum required heating power in the test section of around 32 kW. The maximum temperature that will be reached is set by R1233zd(E). At a supercritical pressure of 1.25 p_c and a minimum mass flux of 320 kg/m^{2}s, the maximum temperature at the end of the test section is around 500 K (assuming that the pseudo-critical temperature is reached in the middle of the test section). The maximum design mass flux is 1000 kg/m^{2}s and the design pressure range of the refrigerants lies between 1.05 and 1.25 p_c. Higher pressures are less desirable as this would require even more pumping power and would complicate component selection. In addition, as can be seen in Figure 1, the variation in thermophysical properties reduces when pressure increases. Therefore, it is assumed that the interesting pressure range is covered by the proposed design pressure range. The refrigerant R1234ze(E) is the most critical one for the pressure limitation, resulting in a maximum pressure of 4.54 MPa.

3.2 Component selection and temperature and pressure control

Based on the maximum temperature and pressure level derived in Section 3.1, the overall system will make use of steel tubing. More specifically, stainless steel is chosen in order to avoid corrosion issues that could occur during construction or operation of the test rig. The actual inner diameter of the stainless steel tube will depend on what tube sizes are commercially available, however, an inner diameter in the range of 24 – 25 mm is a requirement. In addition, a thick walled tube is preferred for the installation of the thermocouples, explained further in Section 3.3. In the previously built test rig (by Lazova *et al*, 2018), a thermal oil unit was used to preheat the refrigerant and the desired test section inlet temperature was regulated by a PID temperature controlled electrical preheater. However, during measurements, the electrical preheater acted unstable and impeded the control of the test rig and complicated steady-state conditions. Therefore, in the current design, the electrical preheater will be omitted, and only the thermal oil unit will be used for preheating and temperature control. After the test section, the supercritical fluid will be cooled down in a plate heat exchanger. The intermediate cooling fluid is a water-glycol mixture which in its turn is cooled down by a chiller.

Pressure control of the test rig will be realized by a gas charged accumulator which is connected to a nitrogen cylinder. By lowering or raising the amount of nitrogen in the accumulator, pressure will, respectively, decrease or increase. Use of an expansion valve, as implemented in the previously built test rig, was considered as well. However, due to the high temperature requirements, no suitable expansion valve could be found, and eventually, the nitrogen vessel option was chosen. By omitting the expansion valve, the entire test rig will be pressurized to supercritical pressures. Therefore, a suitable pump needs to be chosen that can handle these high inlet pressures. This pump will not have to pressurize the refrigerant from a sub- to supercritical pressure and will only have to compensate for the limited pressure losses of the system. As a safety measure, a safety relief valve will also be installed close to the pump.

Mass flux through the test section will be regulated through a combination of bypassing and frequency control of the pump. A bypass will redirect part of the flow (by using a three-way diverting valve) to the inlet of the pump. Finetuning is then done by regulating the frequency of the pump. Finally, a filter drier will be added as well to protect the pump and the rest of the installation from unwanted particles. A schematic representation of the test rig is illustrated in Figure 2.

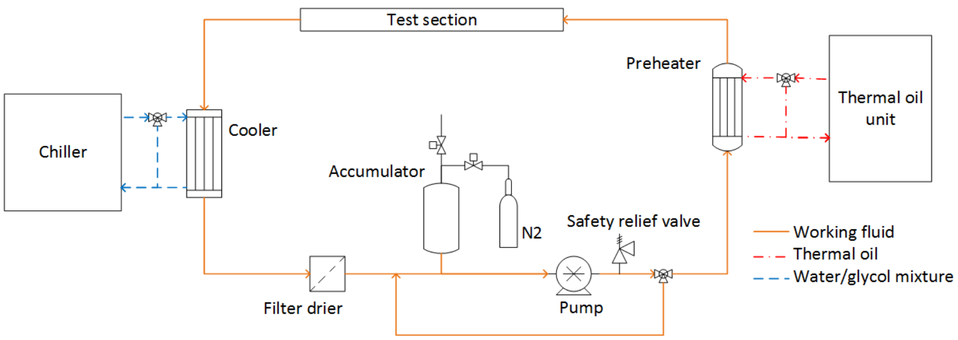

Figure 2: Schematic of test rig

3.3 Test section layout

In the test rig built by Lazova *et al.* (2018), the heating power in the test section was applied by thermal oil in a tube-in-tube heat exchanger configuration, with oil flowing in the annulus and refrigerant in the inner tube. However, this design has some drawbacks. First of all, by working with a tube-in-tube layout, construction of the test section is more complicated. Specifically the attachment of the thermocouples to the outer wall of the inner tube is challenging due to the thermal oil flowing over it. Secondly, the capacity of the current thermal oil unit is limited to 20kW, with a maximum allowable temperature of 453.15K. The heating capacity and maximum temperature are not high enough to reach the desired measurement conditions as explained in Section 3.1. Finally, the heat flux to the refrigerant cannot be independently regulated with this system, as the heat flux depends on numerous variables such as oil mass flow rate, refrigerant mass flow rate and test section inlet temperature. Based on the reasons mentioned above, heat flux in the current design will be applied electrically by means of Joule heating (also called resistance heating) of the steel tube. By Joule heating is meant that a certain current will be send through the test section (which is electrically and thermally insulated from the remainder of the test rig) by using a DC power source. As the stainless steel tube has a certain electrical resistance, heat will be generated in the tube.

The heat transfer to the refrigerant will be expressed by the local heat transfer coefficients of the refrigerant (htc_r), which will be determined from the measurements according to Equation (1):

$$htc_r = \frac{\dot{q}}{T_{w,i} - T_{r,b}} \tag{1}$$

In this equation, $\dot{q}$ is the heat flux to the refrigerant, $T_{w,i}$ the temperature of the tube at the inner wall and $T_{r,b}$ the bulk temperature of the refrigerant. The heat flux will be determined by measuring the applied electrical power to the tube through voltage and current measurements. In addition, the bulk temperature in Equation (1) and the outer wall temperature, from which the inner wall temperature can be deduced, will be directly measured by thermocouples. The inner wall temperature is calculated according to Equation (2) (one dimensional heat conduction problem with internal uniform heat source), with d_o the outer diameter of the test tube, k_{ss} the thermal conductivity of stainless steel and $\dot{q}_v$ the volumetric heating rate (which is the power supply of the DC power source divided by the volume of the heated stainless steel tube which is $\frac{\pi(d_o^2 - d_i^2)L}{4}$):

$$T_{w,i} = T_{w,o} + \frac{\dot{q}_v}{16k_{ss}}\left(d_o^2 - d_i^2\right) - \frac{\dot{q}_v}{8k_{ss}} d_o^2 \ln\left(\frac{d_o}{d_i}\right) \tag{2}$$

As already mentioned in the introduction, buoyancy plays an important role in supercritical heat transfer. Consequently, the radial temperature distribution of the wall is not uniform, and heat transfer differs in function of radial position. Therefore, three wall thermocouples will be attached at one axial position in order to be able to quantify and gain insight into the effect of buoyancy on the heat transfer. The resulting thermocouple layout at a given axial position in the test section, placed every 200 mm, is illustrated in Figure 3. The bulk temperature will be measured by T-type thermocouples, as these have a higher inherent accuracy. However, K-type thermocouples will be used for the wall temperature measurements in the test section as they are capable of dealing with higher temperatures, which is required in order to be able to weld them to the tube.

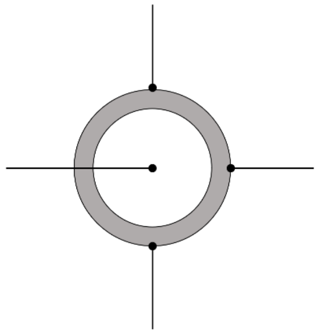

Figure 3: Thermocouple layout

3.4 Expected measurement uncertainty

Compared to the previously built test rig, uncertainty on the heat transfer coefficient calculations can be significantly reduced, which is a requirement in order to construct reliable heat transfer correlations based on experimental data. This reduction in uncertainty is due to the direct measurement of the outside wall temperatures (instead of deducing them from bulk oil temperature measurements) and the different application of the heat flux. For the new test rig, the absolute uncertainty (U_{htcr}) on htc_r is a function of the uncertainty on the heat flux measurement ($U_{\dot{q}}$) and on the thermocouple measurements (U_T) and is calculated according to:

$$U_{htc_r} = \sqrt{\left(\frac{1}{T_{w,i}-T_{b,r}}\right)^2 U_{\dot{q}}^2 + \left(\frac{\dot{q}}{\left(T_{w,i}-T_{b,r}\right)^2}\right)^2 U_{T_{b,r}}^2 + \left(\frac{\dot{q}}{\left(T_{w,i}-T_{b,r}\right)^2}\right)^2 U_{T_{w,i}}^2} \tag{3}$$

After calibration of the thermocouples, an uncertainty of maximum 0.1K will be achieved. For the heat flux measurement, a relative uncertainty of 5% is taken as a reference. For heat fluxes ranging from 10 - 100 kW/m^2 and heat transfer coefficients ranging from 500 – 5000 W/m^{2}K, the expected relative uncertainties on the heat transfer coefficients will lie in between 5 – 9 %.

Future work includes construction and commissioning of the test rig and performing the extensive measurement campaign to determine the supercritical heat transfer to different low GWP refrigerants. Based on the measurements, insight into supercritical heat transfer will be extended, and a larger database will be available in order to create general heat transfer correlations.

4 CONCLUSIONS

In this work, the design and development of a new experimental test rig is discussed. The test rig is designed to measure the heat transfer to low GWP refrigerants flowing horizontally in a large diameter tube under supercritical conditions. The refrigerants under consideration are R1234yf, R1234ze(E), R1336mzz(E), R1224ydZ and R1233zd(E), which cover a wide range of critical temperatures. The test rig will be able to test supercritical pressures up to 1.25 p_c, heat fluxes from 10-100 kW/m^2 and mass fluxes from 320-1000 kg/m^{2}s. Advancements compared to the former test rig include pressure control through the use of a gas charged accumulator, use of steel tubing to extend the allowable temperature range and electrical application of the heat flux for a wider range and more independent control. Furthermore, in the test section thermocouples will be attached to the tube at three radial positions in order to quantify and gain insight into the effect of buoyancy on heat transfer in larger diameter tubes. The unique data generated will contribute to the understanding of supercritical heat transfer to refrigerants and development of a general correlation which will enable more accurate sizing of vapor generators in transcritical Organic Rankine cycles.

NOMENCLATURE

c_p	specific heat capacity	(J/kgK)
d	diameter	(m)
E	experimental	
GWP	Global Warming Potential	(-)
htc	heat transfer coefficient	(W/m^{2}K)
k	thermal conductivity	(W/mK)
L	test section length	(m)
MM	hexamethyldisiloxane	
N	numerical	
ODP	Ozone Depletion Potential	(-)
ORC	Organic Rankine cycle	
p	pressure	(Pa)

$\dot{q}$	heat flux	(W/m^2)
$\dot{q}_v$	volumetric heating rate	(W/m^3)
T	temperature	(K)
TORC	transcritical Organic Rankine cycle	
U	absolute uncertainty	
μ	dynamic viscosity	(Pa.s)
ρ	density	(kg/m^3)

Subscript

c	critical
h	hydraulic
i	inner
o	outer
pc	pseudo-critical
r	refrigerant
ss	stainless steel
w	wall

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ACKNOWLEDGEMENT

The work presented in this paper was funded by a FWO – Flanders grant for strategic basic research (1SA3720N). This financial support is gratefully acknowledged.