BASIC DEVELOPMENT OF A NOVEL HIGH TEMPERATURE HEAT PUMP SYSTEM USING LOW GWP WORKING FLUIDS

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Abstract: A large application potential of industrial heat pumps is still not utilized due to limited supply temperatures of about 100 °C. With increased supply temperatures, more industrial processes could be improved in their energy efficiency. The main reason for the limited temperatures is the absence of adequate working fluids. An ideal working fluid should be non-flammable, non-toxic and should have a low GWP, no ODP and a high critical temperature. Four ideal working fluids are identified: LG6, MF2, R1233zd and R1336mzz. This paper presents the basic development of a novel high temperature heat pump using ideal working fluids. At first, the particular thermodynamic properties of the working fluids are presented. They show different extents of an overhanging behavior caused by the complex molecular structure. As a consequence, alternative cycle designs are necessary with deviating operation characteristics. It is shown, with newly defined terms, which cycle design is suitable for which thermodynamic property. In doing so, the phenomenon is ascertained, that the necessary cycle design can be concluded only from knowing the molar mass of the working fluid. Secondly, experimental performance investigations are presented for two ideal working fluids at different combinations of evaporation and condensation temperatures. The experimental results indicate a very promising performance of this novel heat pump.

Key Words: Performance, high temperature, low GWP, overhanging, molar mass

1 INTRODUCTION

“Heat pump technologies are widely used for upgrading low-temperature free heat from renewable sources, such as air, water, ground and waste heat, to useful temperatures. They are used for residential and commercial space and water heating, cooling, refrigeration and in industrial processes. […] 1.8 billion tones of CO2 per year could be saved by heat pumps, corresponding to nearly 8% of total global CO_2 emissions.” (IEA 2013)
Commercially available heat pumps can supply heat only up to 100 °C. However, high temperature heat pumps (HTHP, >100°C) are of great interest because the higher temperature range leads to a higher application potential. The absence of commercially available products in this higher temperature range is mainly due to the lack of suitable working fluids (Lambauer et al. 2012).
The choice of working fluids is narrowed down because of environment and safety issues. At the moment, large research efforts are expended for the development of alternative fluids and corresponding heat pump systems.
Newly considered working fluids for the use in HTHP are more complex in their molecular structure and the molar mass is higher. Along with this trend, the shape of the phase

boundary of new fluids is overhanging. This means, that the saturated vapor line has a low positive slope. The operation of such fluids in heat pumps has to be investigated. Condensation and thus liquid droplets forming during the compression process could occur. This could damage the compressor by liquid hammer and has to be prevented by alternative cycle designs.

2 WORKING FLUIDS FOR HIGH TEMPERATURE HEAT PUMPS

This section describes important properties of working fluids for the use in high temperature heat pumps. In addition, this section gives an overview of currently existing projects for the development of working fluids and HTHPs. In the end of the section, considerations about the shape of the phase boundary, which turns out to be an important working fluid property for the heat pump cycle design, are shown.

2.1 PROPERTIES AND CURRENT SITUATION

An important thermodynamic property is the critical temperature. The higher the critical temperature, the higher is the heat of condensation at a distinct temperature. Due to performance reasons, the critical temperature should be far above the condensation temperature (Cube et al. 1997). This is a criterion for exclusion for the most common working fluids for the use in HTHPs.
Besides the applicable temperature range there are important requirements about the safety and environmental properties of the working fluids. Ideally, working fluids should be non-flammable and non-toxic. Environmental properties are the ozone depletion potential (ODP) and the global warming potential (GWP). Fluids with an ODP value higher than zero are banned by the Montreal protocol from 1987. Restrictions for fluids with a high GWP are currently in discussion. There is already a restriction by law valid in the European Union (EU) for working fluids in motor vehicles. The EU Directive 70/156/EEC (2006) forbids the use of working fluids for motor vehicles air conditioning with a GWP higher than 150. The EU commission further decided to reduce the fluorinated gas emissions to 20% compared to now until 2030. The decisions of the EU institutions emphasize the political will to reduce the use of high-GWP working fluids by law restrictions. This creates research necessity for alternative environmentally friendly working fluids.
At the moment, there is no commercially available HTHP with heat supply temperatures higher than 100 °C. Earlier, R114 was used as working fluid for HTHPs, but it is forbidden now because of its ODP being higher than zero.
Currently there are several working fluids in discussion for the use in HTHPs like R245fa (Bobelin 2011) or ECO3 (Bobelin et al. 2012). However, these working fluids show a high GWP and the usage will probably be limited in the future.
The aim of this paper is to develop a HTHP using only working fluids with ideal environment and safety properties (non-flammable, non-toxic, low GWP, no ODP). Four working fluids with these ideal properties are identified, which also show a high critical temperature (see Table 1 and following list).

R1233zd (DeBernadi 2013):
is a recent development of Honeywell especially for the high temperature range. Like HCFCs it contains a chlorine atom, nevertheless the ODP is extremely low (0.0003), so that it is not forbidden by the Montreal protocol. Data on thermodynamic properties are freely available since mid 2013.

R1336mzz (Kontomaris 2011 and 2013):
is a recent development of DuPont especially for the high temperature range. It was introduced since 2011 under the code name DR-2. Since October 2013 the chemical formula is revealed as R1336mzz(Z) and some data on thermodynamic properties are available.

LG6:
is a working fluid with ideal properties. Reissner et al. (2013a, 2013b) showed theoretical and experimental performance investigations. The denomination "LG6" is used throughout this paper as code name.

MF2:
is presented for the first time in this paper and also shows ideal properties. The denomination "MF2" must be used throughout this paper as code name.

Table 1: Properties of ideal working fluids for high temperature use; Sources from working fluid list above

Working fluid	T_{crit} [°C]	Flammable or toxic	ODP	GWP
R1233zd	166	no	0.0003	6
R1336mzz	171	no	0	9
LG6	>165	no	0	1
MF2	>145	no	0	<10

2.2 SHAPE OF PHASE BOUNDARY

The shape of the phase boundary (saturated liquid line and saturated vapor line) of a working fluid in the temperature-entropy diagram varies significantly for different fluids. Fluids can be distinguished by their saturated vapor line slope (see Figure 1) in negative (type-A) and partly positive (type-B).
These two different behaviors of the fluids are named with various terms in the literature. Type-A fluids are named "dry" and "bell-shaped" (Lai and Fischer, 2012), whereas type-B fluids are named "wet", "re-entrant" (Itard, 1995), "skewed" (Morrison, 1994) or "overhanging" (Lai and Fischer, 2012). The terms "dry" and "wet" are process depending and not clearly defined, so that when other processes like Organic Rankine Cycles are considered the terminology is vice versa (Qiu, 2012). In this paper the terms "bell-shaped" and "overhanging" are used.

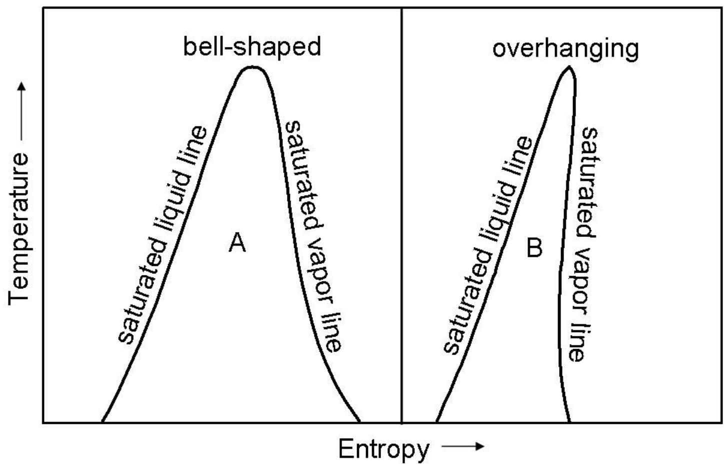
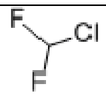
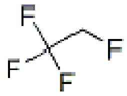
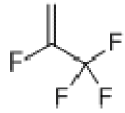


Figure 1: Scheme in a temperature-entropy diagram A: bell-shaped; B: overhanging

2.2.1 MOLECULAR STRUCTURE AS REASON FOR THE PHASE BOUNDARY SHAPE

In the history of refrigeration and heat pump technology, many working fluids have been investigated. As a result, all derivatives of methane (hydrogen substitution by fluorine or chlorine) are well known. However, they were mostly ozone depleting fluids (e.g. R22) and are forbidden by the Montreal protocol. Later, derivatives of ethane (e.g. R134a) were developed and are used until today. But they have a high global warming potential, so that they must be replaced by propane (or propene) derivatives (e.g. R1234yf). As the search for fluid alternatives continues, fluids with ether (HFE) and olefin (HFO) groups are investigated as well. Consequently, the molecular structure of alternative fluids becomes more and more complex. This is illustrated by the historical development in Table 2.

Table 2: Historical view on fluid replacement and increased complexity of molecular structure; Sources: (Buchwald et al. 2010), (Bitzer2010), (Calm and Domanski2004), (Franklin 1993), (Honeywell 2011)

Working fluid	**R22**	**R134a**	**R1234yf**	**e.g. LG6, MF2, R1336mzz**
Molecular structure				-
C-atoms [count]	1	2	3	≥4
Molar mass [g mol^{-1}]	86	102	114	>150
Fluid type	bell-shaped	bell-shaped	overhanging	overhanging
ODP	0.05	0	0	0
GWP	1700	1300	4	<10
Year of commercialization	1936	1990	2010	>2014

Fluids tend to be overhanging with the increased molecular complexity. This can be a problem for the compression process of the heat pump cycle. When the slope of the saturated vapor line is very low, a typical amount of superheat after the evaporation might not be enough to avoid liquid compression.

The slope of the saturated vapor line is a good indicator to quantify the overhanging behavior. Fluids, which are very near at the boundary from overhanging to bell-shaped, have extreme high slopes. This is a problem when fluids are compared. Consequently, the inverse slope is more useful, as suggested by Lai and Fischer (2012). They also suggest defining a mean slope for each fluid by taking the minimum and maximum entropy of the positive slope range. However, if a specified temperature is considered, there is a deviation from their definition to the actual slope of the tangent on the saturated vapor line at the specified temperature. Consequently, the inverse slope (IS) at a specified temperature (80 °C) is used in this paper to describe the degree of the overhanging behavior, see equation (1).

$$IS = \frac{s_{sat.vap.,T+1\,K} - s_{sat.vap.,T-1\,K}}{2\,K} \quad (1)$$

Morrison (1994) showed that the IS is directly correlated to the molar mass. Figure 5 shows this correlation. For all following working fluid properties and calculations Refprop 9.1 (Lemmon et al., 2013) is used.

2.2.2 CONSEQUENCES FOR OPERATION

Fluids with a high IS have to be further investigated regarding the consequences for operation in heat pumps. Because of the high IS the amount of superheat provided by the evaporator might not be enough to avoid liquid compression. Therefore the term “minimum required superheat” (minSH) is defined in this paper.

The minSH is calculated for a specified evaporation temperature (exemplary: 80 °C) and condensation temperature (exemplary: 130 °C). The isentropic compressor efficiency for this calculation is assumed to be 0.8. Additionally a minimum safety distance of the compression end temperature to the saturated vapor line (at the same pressure) of 5 K is considered as prerequisite, see Figure 2 and Equations (2)-(6).

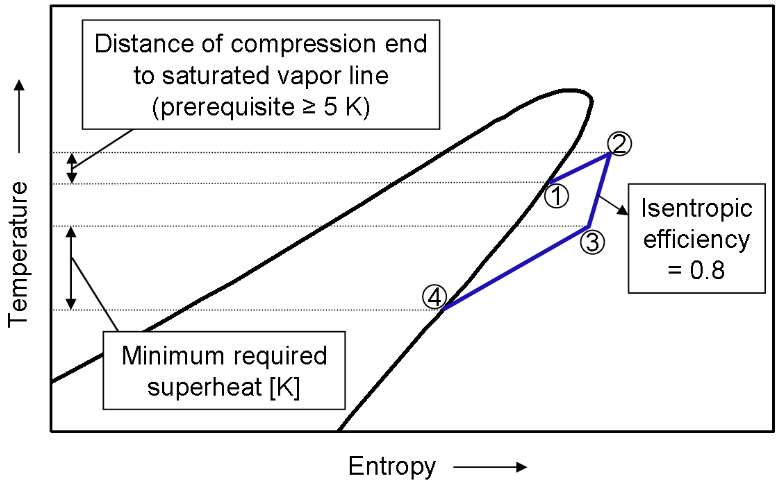


Figure2: Definition of minimum required superheat in a scheme of a temperature-entropy diagram

$$\min SH = T_3 - T_{4,evap} \quad (2)$$

$$T_3 = f\left(p_{evap}; h_3\right) \quad (3)$$

$$h_3 = h_2 - \frac{h_2 - h_{3s}}{\eta_s} \quad (4)$$

$$h_2 = f\left(T_{cond} + 5\ \text{K}; p_{cond}\right) \quad (5)$$

$$h_{3s} = f\left(p_{evap}; s_2\right) \quad (6)$$

The minSH is calculated for the considered working fluids and can be seen in Figure 6.

3 OPERATION OF LOW SLOPE FLUIDS WITH AN INTERNAL HEAT EXCHANGER

To meet the demands of a large superheat for fluids with a high IS as explained in section 2.2.2, an additional heat source is required. An internal heat exchanger (IHX), which subcools the condensate, can be used as an additional heat source (see Figure 3B).
As described by Domanski et al. (1994) this is done to improve the COP by increasing the evaporator pressure and superheat partly in the IHX. Another purpose of the IHX is to increase the temperature at the suction line of the compressor, to prevent water of the surrounding air from condensing.
In contrast to these purposes, where the IHX is an optional apparatus, the IHX for cycle designs with fluids with a high IS is indispensible for the operation of such fluids. This can be seen in Figure 3, where the compression end in a basic heat pump cycle without an IHX is partly liquid.

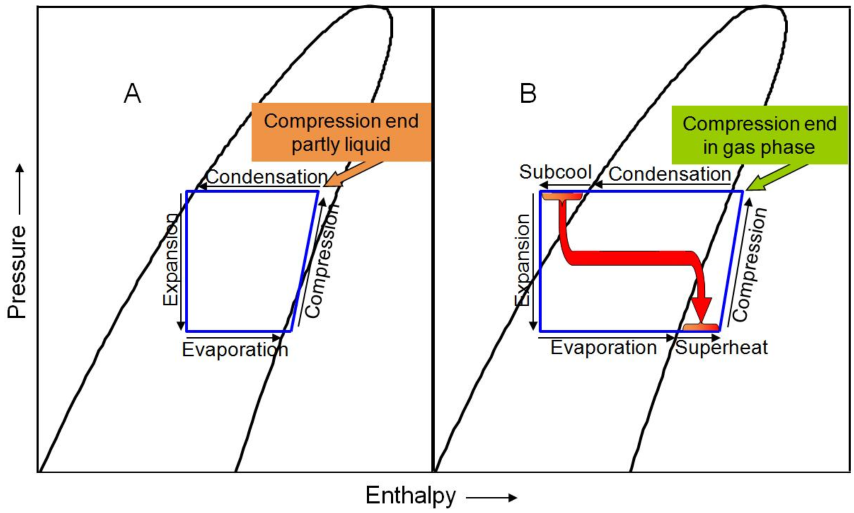


Figure3: Scheme of vapor compression heat pump process without (A) and with (B) internal heat exchanger in a pressure-enthalpy diagram for fluids with a high IS

For the following calculations a minimum temperature difference inside the IHX between the subcooled condensate and the superheated suction gas is 5 K. Subcool in condenser and superheat in evaporator are 5 K. The expansion is isenthalpic.
The maximum transferable heat amount through the IHX (Δh,IHX) depends on the temperature lift. This dependency is shown in Figure 7A for the fluid LG6 as an example. For a compression operation in gas phase only, the IHX has to deliver enough heat to fulfill the minSH requirement.
The minSH has a corresponding required minimum Δh (Δh,required). To reach the minSH the Δh,IHX must be larger than the Δh,required. If Δh,IHX is smaller than Δh,required an external additional heat source (Δh,external) is needed. These three terms are shown in Figure 7A at different temperature lifts and at a constant evaporation temperature of 80 °C for the working fluid LG6 as an example.
The temperature lift at the intersection point of Δh,IHX and Δh,required is here defined as the limiting temperature lift (LTL). At temperature lifts lower than LTL an additional external heat source is needed. At temperature lifts higher than LTL the IHX delivers enough heat to reach the minSH.

4 EXPERIMENTAL PERFORMANCE INVESTIGATION

A novel high temperature heat pump cycle has been developed. The performance of the working fluid MF2 is for the first time experimentally investigated in this paper and compared to LG6.

Setup

The mentioned novel high temperature heat pump cycle is a lab-scale single-stage vapor-compression heat pump. It has a maximum condenser (plate-type) heat load of 12 kW and evaporator (coaxial-type) heat load of 10 kW. The compressor is of a piston type with a maximum motor power of 6 kW.
The IHX is a counter-current gas-liquid heat exchanger, designed to transfer heat from the subcooled condensate to the evaporated fluid for further superheat after the evaporator.

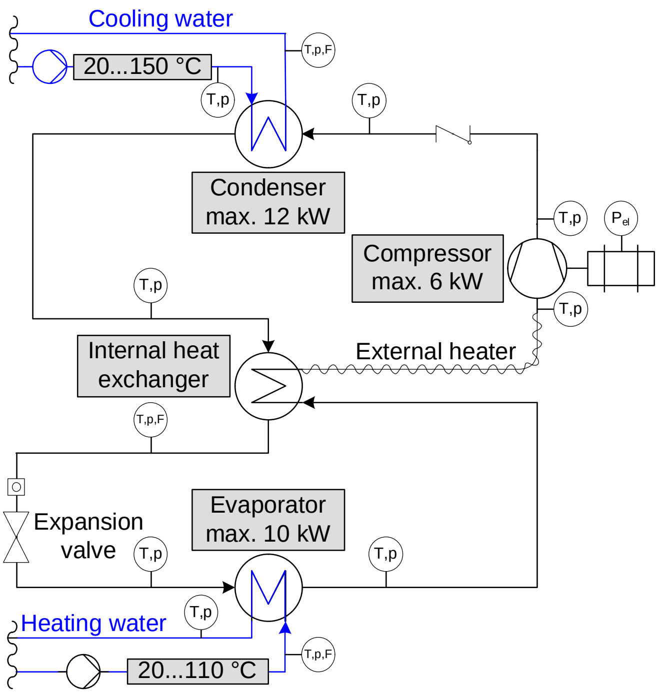


Figure4: Setup of developed HTHP functional model

The working fluid can be evaporated with hot water at up to 110 °C and condensed with cooling water at up to 150 °C. Besides the temperature, the flow of the two heating and cooling water cycles can be adjusted as well. Temperature, pressure and flow measuring points can be seen in Figure 4. Temperatures are measured with thermocouples type J (accuracy ± 1 K). Pressures are measured with transmitters (accuracy ± 0.004 MPa). Flows are measured with turbine flow meters and magnetic flow meters (accuracy ± 2%).
From temperature and pressure the enthalpy, entropy, density and heat capacity is calculated and displayed (log p,h diagram) online at all state points via Refprop 9.1 which is implemented in the data acquisition system LabVIEW™.
An IHX is integrated into the cycle, which shall provide the necessary superheat to reach the minSH. If the Δh,IHX is not enough to reach the minSH, an external heater is turned on. The heater is integrated at the pipe connecting the IHX vapor outlet and the compressor inlet. The expansion valve, the temperatures and the flow of the heating and cooling water cycles are adjusted to set the evaporation and condensation temperatures.

Procedure

The experimental COP is measured at 15 different operation points with temperature lifts of 30 to 60 K. The evaporation temperature ranges from 40 to 70 °C. The condensation temperature ranges from 70 to 130 °C.
The COP is always measured at both condenser sides (MF2: inner COP and water: outer COP) and the power of the compressor and is shown in Figure 9 and Figure 10. The deviation of the inner and outer COP for all operation points is below 5%.

5 RESULTS AND DISCUSSION

5.1 Working fluid properties and operation characteristics

Figure 5shows that the reason for the IS of the saturated vapor line is also valid for working fluids for high temperature heat pumps. The IS increases linear with molar mass by approximation. Consequently, only by knowing the molar mass it is possible to determine the extent of the overhanging behavior in terms of IS.

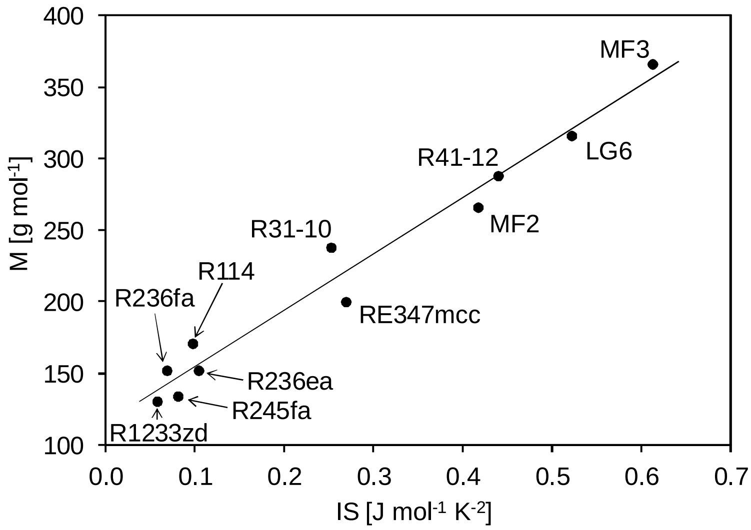


Figure5: Correlation of the inverse slope and the molar mass with trend line; LG6, MF2 and MF3 are code names

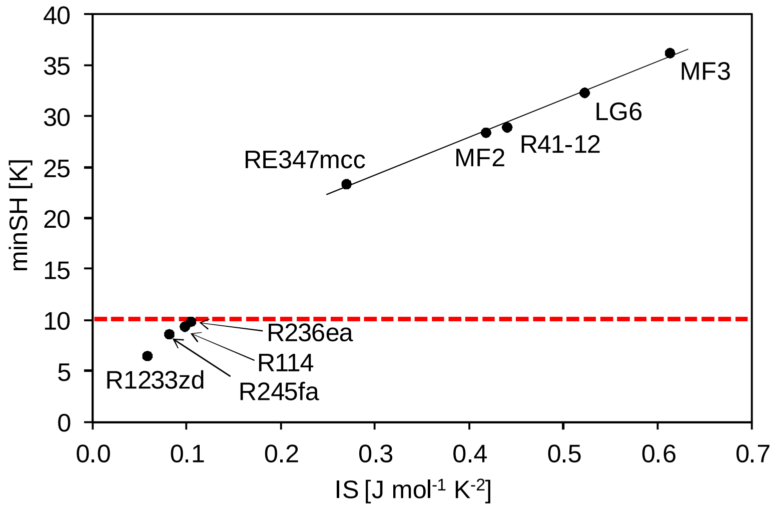


Figure6: Correlation of the inverse slope and the minimum required superheat with trend line; LG6, MF2 and MF3 are code names

The new defined term minSH is also linear increasing with IS by approximation (see Figure 6). For high IS the minSH is so large, that it cannot be provided by the same heat source, which is used for the evaporation. The border situates around 10 K depending on the particular heat source and heat pump application. The working fluids R1233zd, R245fa, R114, R236ea show a minSH value below 10 K. Here the evaporator heat source in the standard cycle is enough to superheat for avoidance of liquid compression. However, all other working fluids need an alternative cycle with additional heat source to provide large superheat. The border to distinguish working fluids in standard cycle operable and alternative cycle operable is approximately around IS values of 0.15 J mol^{-1} K^{-2}.

The additional heat source of an alternative cycle can be an IHX. The Δh,IHX is depending significantly on the temperature lift as shown in Figure 7A for the fluid LG6.The reason for the significant increase is the increasing temperature difference of the condenser outlet and evaporator outlet, which allows more heat amount to be transferred.

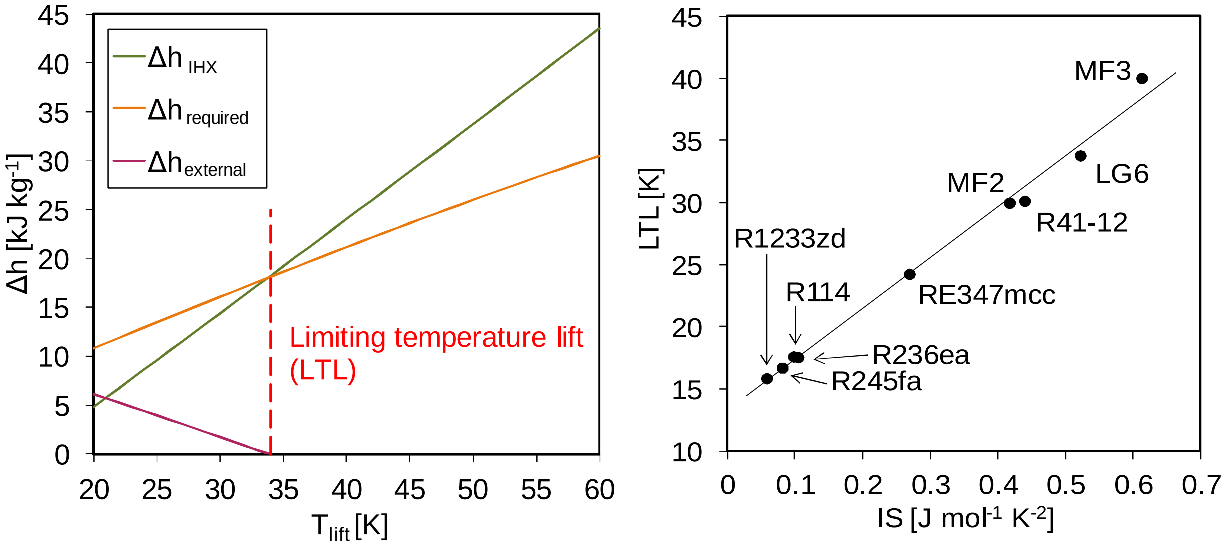


Figure7A: Limiting temperature lift as intersection point of Δh,IHXand Δh,required; Working fluid LG6; 7B: Correlation of the inverse slope and the limiting temperature lift with trend line; LG6, MF2 and MF3 are code names

The temperature lift of the intersection point of Δh,IHX and Δh,required is the LTL. The LTL is fluid-specific (see Figure 7B) and indicates if an intended operation point is feasible with the IHX alone as additional heat source. If the temperature lift of an operation point is below the LTL, an external heat source (Δh,external) is needed for superheat.
The LTL increases linear with the IS by approximation (see Figure 7B). Consequently, only by knowing the molar mass it is possible to predict if a cycle with an IHX is enough for operation or if a external heat source is needed. This is valid for the investigated working fluids. Other working fluids might deviate from these correlations.
Figure 8 summarizes the linear correlations and the consequential conclusions for cycle design.

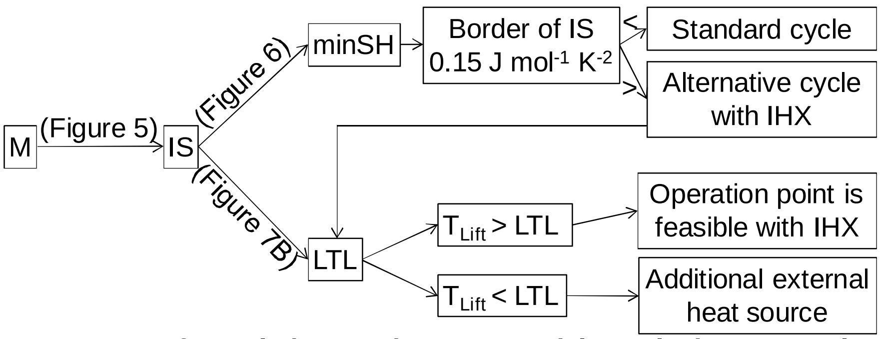


Figure8: Summary of correlations and consequential conclusions on cycle design

R1336mzz is also an ideal working fluid and is taken as example to verify these correlations. Data for calculations with Refprop are not freely available at this time. However, it is possible to estimate the minSH from Kontomaris (2013) with a value of 18 K ± 2 K (inaccuracy in graphical readout). R1336mzz has a molar mass of 164 g mol^{-1}. The linear correlations of Figure 5 and Figure 6 lead to a minSH of 19 K. This emphasizes the validity of the established correlations for further working fluids.

5.2 EXPERIMENTAL PERFORMANCE

The experimental setup from section 4 was used to investigate the performance of the working fluid MF2. MF2 has an IS of 0.42 J mol^{-1} K^{-2} and a minSH of 28 K. Thus, an IHX is needed for operation and included in the setup. The LTL is 30 K. Therefore, operation points with temperature lifts above 30 K are measured with the external heater to avoid liquid compression.
Figure 9 shows the results of the performance measurements of the working fluid MF2.

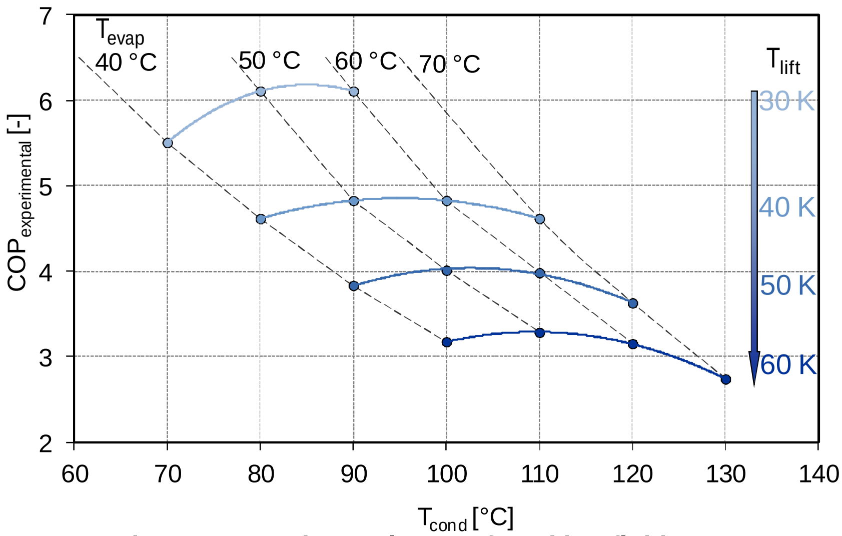


Figure9: Experimental COP of working fluid MF2

Naturally, the COP is highest at the lowest temperature lift, but still at a temperature lift of 60 K the COP is well above 3. The operation point T_{evap} = 70 °C; T_{cond} = 130 °C is an exception. Here the condensation occurs very near the critical point and thus the condensation enthalpy is lower. The best performances of each temperature lift are around condensation temperatures of 90 to 110 °C. Consequently, it is advisable to use MF2 at these temperatures.
For even higher condensation temperatures up to 140 °C, it is advisable to use LG6 instead of MF2. Reissner et al. (2013a) published experimental performance results of LG6 earlier. Figure 10 shows the experimental results for both working fluids LG6 and MF2 at a temperature lift of 50 K.

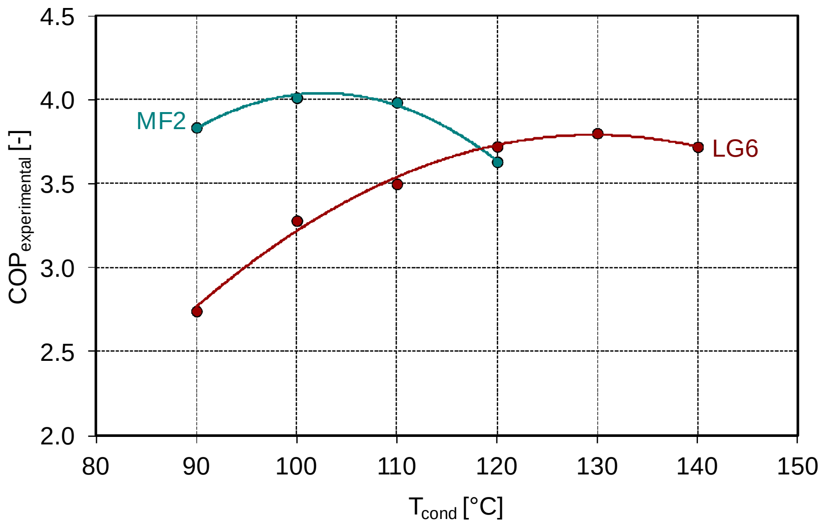


Figure9: Experimental COP of working fluid MF2 and LG6 at T_{lift} = 50 K

At condensation temperatures higher than 120 °C LG6 shows a better performance. The large gap between the COPs of MF2 and LG6 at condensation temperature from 90 °C to 110 °C bases on the low volumetric heating capacity (VHC) of LG6. At these temperatures the VHC ranges from 700 to 1300 kJ m^{-3}. In contrast to that, MF2 shows a VHC of 2000 to 3000 kJ m^{-3}. VHC values of LG6 above 2000 kJ m^{-3} are only at temperatures 120 °C and higher.
Finally, two working fluids with ideal environment and safety issues can be used for high temperature heat pumps in alternative cycles. Each shows a very promising performance for a certain condensation temperature range.

6 CONCLUSION

It is shown, that HTHPs need new future-proof working fluids for industrial dissemination of this technology. Ideally, a working fluid should be non-flammable, non-toxic and show a low GWP, no ODP and high critical temperature. Currently investigated working fluids like R245fa show no ideal properties because of the high GWP.
Four new ideal working fluids are identified for high temperature use: MF2, LG6, R1233zd and R1336mzz. These working fluids show high molar masses and complex molecular structures and thus the saturated vapor line is overhanging. An approximately linear correlation of the molar mass to the slope of the saturated vapor line is proved.
Overhanging working fluids show problems with liquid compression. The extent of the overhanging behavior and the consequences for operation are quantified and classified with two newly defined terms: “minimum required superheat” and “limiting temperature lift”. It is possible to establish alternative cycle designs with these new terms. Moreover it is possible to conclude to the necessary cycle design (e.g. internal heat exchanger, additional external heat source) only by knowing the molar mass of the investigated working fluids. The coverage of this correlations and conclusions on different molecule classes and further working fluids is verified.
In the second part of the paper, the experimental performance of the working fluids LG6 and MF2 are presented. These working fluids are measured at condensation temperatures up to 140 °C. The measured experimental COP is very promising for future applications. MF2 and LG6 complement in a positive way. MF2 shows best performance at T_{cond} = 90 to 110 °C and LG6 at T_{cond} = 120 to 140 °C. Consequently, for each desired condensation temperature, ideal working fluids with a very promising performance established within this paper.

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