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Design of a Cascade Refrigeration System for Applications Below -50°C Using CO_2-Sublimation

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ABSTRACT

Within the present paper the design of a CO_2-sublimation refrigeration system is introduced. Different variations of CO_2 cycles were theoretically investigated and compared to a baseline R-23 system under various boundary conditions. It turned out that the overall system efficiency of a modified CO_2-sublimation system could exceed the conventional R-23 cycle performance in many operation points. Based on the simulation results, a cascade design has finally been chosen. The lower stage of the cascade system can be switched between the CO_2-sublimation cycle and a conventional R-23 cycle, so that these two cycles can be compared directly under the same ambient conditions. For the CO_2 sublimation cycle a two-stage compression with intercooling is used as well as an internal heat exchanger. The design of this cascade refrigeration machine is currently used for building-up a demonstrator to prove the concept experimentally.

Keywords: cascade, CO_2, sublimation, R23

1. INTRODUCTION

Environmental simulation and also some specific applications in chemical and pharmaceutical industry, such as freeze-drying processes require a low cooling temperature range of -50 °C to -85 °C. For this temperature range, there are only a limited number of refrigerants available, especially regarding the safety. Trifluoromethane (R-23) is currently the most widely used non-flammable refrigerant for applications down to about -80 °C. However, it has an enormous global warming potential ($GWP_{100,\ R\text{-}23}$=14800). Although it is currently not banned by the new F-gas regulation in Europe due to a lack of alternatives, recent price increases lead to a loss of attractiveness in industry.

In search of a substitute for R-23, some studies in recent years indicated that the application temperature of the natural refrigerant carbon dioxide (CO_2) could be extended beneath its triple point (-56 °C and 0.52 MPa) in a refrigeration cycle in order to use the sublimation heat. Concerning safety issues, conventional CO_2 refrigeration cycles are usually operated above the triple point, in order to avoid the formation of solid CO_2, which could cause clogging in the refrigeration system. Huang *et al.* (2007) experimentally investigated the effect of existing solid CO_2 in a discharge flow from a safety valve. It has been proven that the blockage in the valve or in the downstream line caused by solid CO_2 is quickly spontaneously dissolvable as the pressure rises. A continuously working closed cycle of CO_2-sublimation was firstly established on a laboratory scale (Yamaguchi and Zhang, 2009). Later the system was improved by Iwamoto *et al.* (2015) and a lowest refrigeration temperature of -66.4 °C was achieved. In a previous study at TU Dresden (Langebach *et al.*, 2016), recent publications about CO_2-sublimation were summarized and the possibilities as well as the challenge of using CO_2-sublimation in the closed cycles were discussed. Until now, there is only a limited number of studies regarding sublimation cycles available. Further studies should be carried out, in order to prove CO_2 as a practical replacement for R-23.

This study aims at the development of a practical and efficient CO_2-sublimation system that could potentially replace a conventional system using R-23. By using of a stationary simulation approach different cycle variants were analyzed theoretically and compared to each other for various boundary conditions. Based on these calculations, the most appropriate cycle was selected to build up a demonstrator for future experimental investigations. In the last part of this paper the component design and selection are discussed.

2. FLUID PROPERTIES

Before going into detail of the calculation, a comparison of some chosen fluid properties between CO_2 and R-23 is done, as shown in Table 1. In addition, their vapor pressure curves are shown in Figure 1.

Table 1: A comparison of some fluid properties between CO_2 and R-23

Refrigerant		R-744 (CO_2)	R-23 (CHF3)
Molar mass	[g/mol]	44.01	70.01
Critical temperature	[°C]	31.0	25.6
Critical pressure	[MPa]	7.38	4.83
Triple temperature	[°C]	-56.6	-155.1
Triple pressure	[MPa]	0.52	5.8e-5
Isentropic expansion coef. (0 °C, 0.1 MPa)	[-]	1.30	1.20
Normal boiling point	[°C]	-78.5*	-82.1
Normal evaporation enthalpy	[kJ/kg]	573.0*	239.6

* Sublimation

It can be seen that there is a much steeper increase in vapor pressure of CO_2, especially in the sublimation range, than that of R-23, although these two substances have a similar normal boiling / sublimation point. Therefore a higher compression ratio is required for CO_2 systems, compared to R-23 systems and an even higher discharge temperature can be expected, considering the larger isentropic expansion coefficient of CO_2. These could result in the necessity to build an extra stage of compression in CO_2 sublimation system (see section 3).

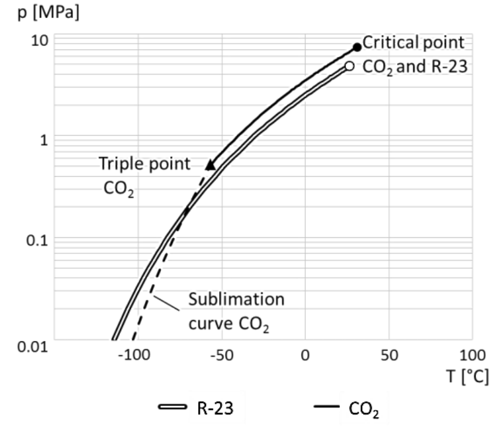


Figure 1: Vapor pressure curves of CO_2 and R-23

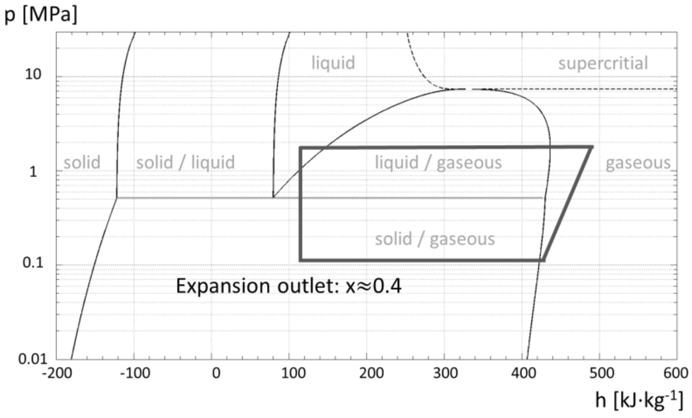


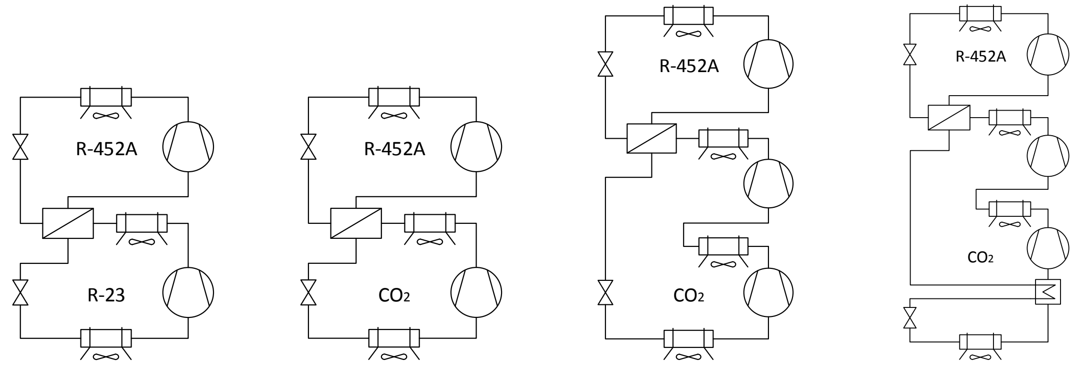
Figure 2: A CO_2-sublimation cycle in log (p)-h-diagram

The larger sublimation enthalpy is an advantage of CO_2. However, it should be pointed out that the usage of the complete sublimation enthalpy is normally not possible. It can be illustrated by a simple CO_2-sublimation cycle in log (p)-h-diagram, as shown in Figure 2. With assumption of isenthalpic expansion and direct sublimation, the vapor quality at the entrance of the sublimation heat exchanger can only reach a maximum of about 0.4 (corresponding to a volume fraction of solid particles of approximate 0.3 %). Using special processes, such as dry ice separation and storage, could theoretically further reduce the vapor quality, however, a minimum fraction still need to be maintained to ensure a stable flow. The highest possible mass / volume fraction of solid particles for a stable flow still remains to be investigated. Within this study, the sublimation cycles are limited to direct sublimation of pure CO_2, which have been experimentally proven (Yamaguchi and Zhang, 2009, Iwamoto *et al.*, 2015).

3. THEORETICAL INVESTIGATION OF CYCLES

3.1 Definition of the circuits

The cascade system is at present most widely used in industry for applications below -50 °C. Therefore, a typical air cooled cascade system R-452A[1] / R-23 (Figure 3a) is set as reference for evaluation of the CO_2-sublimation systems. Although R-404A or R-507A is still currently the most popular refrigerants for the upper stage, both refrigerants will not be permitted for use in new stationary plants in the near future due to their high GWP value[2] according to the new EU F-gas regulation. Therefore R-452A is one of the possible alternatives for both above mentioned refrigerants and the replacement has very little impact on the system performance.



a) Reference: R-452A/R-23 cascade

(R-23)

b) Single-stage CO_2-sublimation cycle as cascade

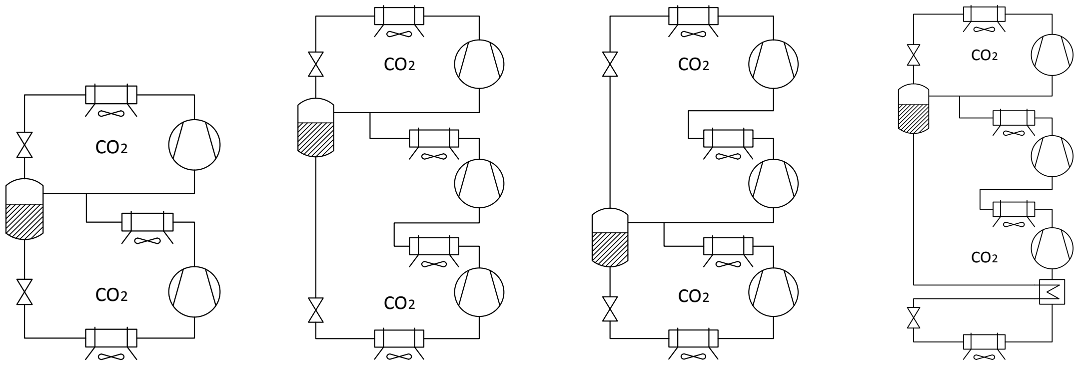
(CO2_casc)

c) CO_2-sublimation cycle as cascade with two-stage compression in the lower stage

(CO2_casc_2s)

d) CO_2-sublimation cycle as cascade with two-stage compression and internal heat exchanger

(CO2_casc_2s_iHX)



e) Pure CO_2-sublimation cycle with intermediate pressure receiver

(CO2_pure)

f) Pure CO_2-sublimation cycle with intermediate pressure receiver and two-stage compression in the lower stage

(CO2_pure_l2s)

g) Pure CO_2-sublimation cycle with intermediate pressure receiver and two-stage compression in the upper stage

(CO2_pure_u2s)

h) Pure CO_2-sublimation cycle with intermediate pressure receiver, two-stage compression and internal heat exchanger in the lower stage

(CO2_pure_l2s_HX)

Figure 3: Flow diagrams of investigated cycles and designation

For CO_2-sublimation systems a single-stage cascade machine (Figure 3b), practically analog to the reference system, was initially investigated. In this case, a sublimation heat exchanger is used instead of a conventional evaporator. Further investigations were conducted with two modified systems, featuring a two-stage compression with an intercooling in the lower stage of the cascade system and an internal heat exchanger (Figure 3c+d). Pure CO_2

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[1] $GWP_{100,\ R\text{-}452A}$=1945;
[2] $GWP_{100,\ R\text{-}404A}$=3922; $GWP_{100,\ R\text{-}507A}$=3985

systems were also investigated for comparison, including the basic form with a single intermediate pressure level and liquid receiver (Figure 3e), as well as three modified variants with multi-stage compression and an internal heat exchanger (Figure 3f - h). At high ambient temperature levels the pure CO_2 systems have to be operated as a transcritical process.

3.2 Boundary conditions and calculation environment

For preparing the theoretical analyses of the systems, the values of different boundary conditions have to be assumed as listed in Table 2.

Table 2: Boundary conditions

Evaporation (Sublimation) temperature T_0	-80 – -60 °C
Ambient temperature T_{amb}	12 – 32 °C
Subcooling at condenser / gas cooler T_{SC}	3 K
Superheating at evaporator /sublimator T_{SH}	5 K
Minimal temperature difference in air cooled / cascade / internal heat exchangers ΔT_{air} / ΔT_{casc} / ΔT_{iHX}	10 K / 7 K / 5 K

Regarding the high compression ratio, its influence on isentropic efficiency should be considered. However, there is no existing universal correlation between these two parameters, especially considering that the operating conditions of the CO_2 compressor in this case are very different from existing tests. For a first estimation, a simple linear approximation according to the data from Hrnjak (2006) was used:

$$\eta_{is} = 0.875 - 0.0375\,\pi \tag{1}$$

The discharge temperature is limited to a maximum of about 150 °C to avoid wrecking impacts on the refrigerant oil. Pressure losses in pipes, connections and heat exchangers are neglected. For the CO_2 sublimation heat exchanger it is assumed that solid CO_2 can be fully sublimated in the heat exchanger. Stationary simulations of the systems were carried out with the software Engineering Equation Solver (EES). The pressure levels or compression ratios were optimized in order to achieve the highest possible coefficient of performance (COP) with the help of an in EES implemented genetic algorithm. The fluid properties used in the calculation were adopted from Refprop-library (Lemmon *et al.*, 2010) and LibCO$_2$-library (Kretzschmar *et al.*, 2014). The LibCO$_2$-library is based on the Helmholtz EOS from Jäger and Span (2012) and enables the properties calculation of CO_2 below the triple point.

3.3 Results and discussion

At first two basic CO_2-sublimation cycles (CO2_casc / CO2_pure) were compared to each other. The calculated discharge temperatures and COP-values are presented in Figure 4 and Figure 5, respectively. It can be seen that the discharge temperature of the CO_2 compressor in the cascade system has already reached the maximum of 150 °C at a sublimation temperature of -65 °C. With decreasing sublimation temperature the upper stage of the cascade has to be operated at an evaporation temperature lower than the optimal point, so that the overall system efficiency decreases significantly. Similarly, the optimal high pressure for the pure CO_2 system could not be reached at a sublimation temperature below -70 °C. Both systems cannot be operated at temperatures lower than approximately -72.5 °C at all. Compared to the R-23 system, the CO_2 cascade system - despite of having a much higher discharge temperature - could achieve similar COP-values at a sublimation temperature of -60 °C and all investigated ambient temperatures ranging from 12 °C to 32 °C. The pure CO_2 system has higher efficiency at a lower ambient temperature and lower efficiency at a higher ambient temperature compared to the baseline, as a result of switching from subcritical to transcritical operation.

Applying an extra compression stage with intercooling could greatly reduce the compressor discharge temperature and makes it possible to operate at a low sublimation temperature down to -80 °C. Also the overall system efficiency increases sufficiently. Compared to the reference system, an increase in COP of 9 – 20 % for the CO_2 systems could be achieved depending on the modification and temperature range. A comparison of COPs as a function of temperature among the modified sublimation cycles with an extra compression stage (CO2_casc_2s / CO2_pure_l2s / CO2_pure_u2s) is shown in figure 6. It can also be seen that the liquid separation at a higher pressure level

(CO2_casc_l2s against CO2_pure_u2s) provides higher efficiency over the entire temperature range, however at an expense of cooling capacity (compare Figure 9).

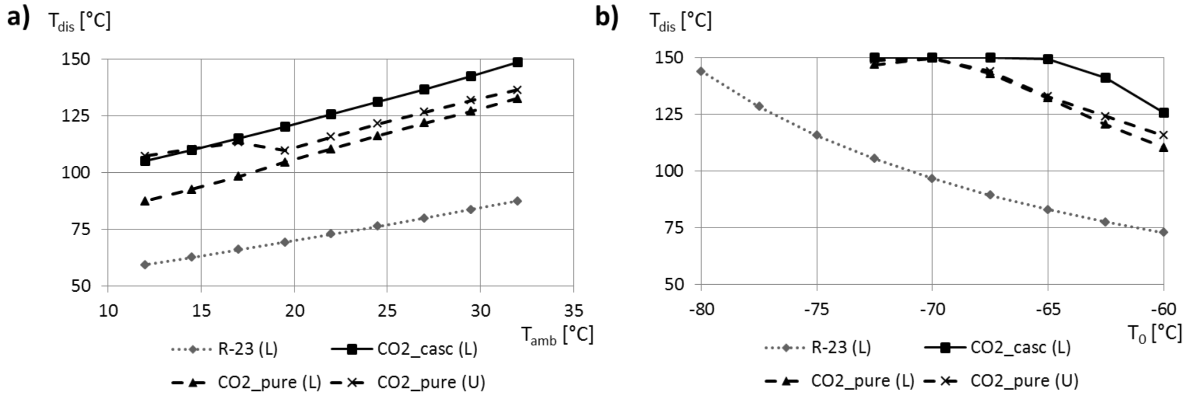

Figure 4: Discharge temperature of two basic sublimation cycles in relation to: a) the ambient temperature T_{amb} (at T_0 = -60 °C); b) sublimation / evaporation temperature T_0 (at T_{amb} = 22 °C); "U" for upper stage; "L" for lower stage

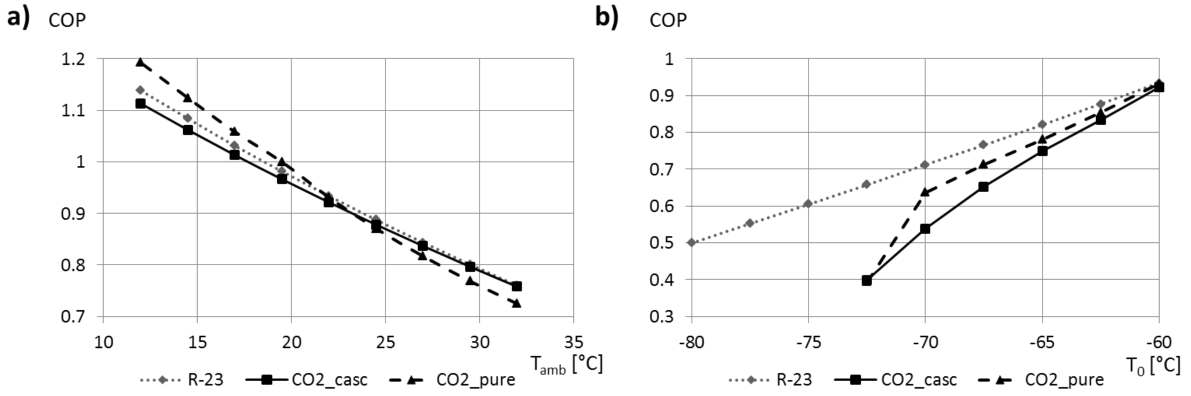

Figure 5: COP of two basic sublimation cycles in relation to: a) the ambient temperature T_{amb} (at T_0 = -60 °C); b) sublimation / evaporation temperature T_0 (at T_{amb} = 22 °C)

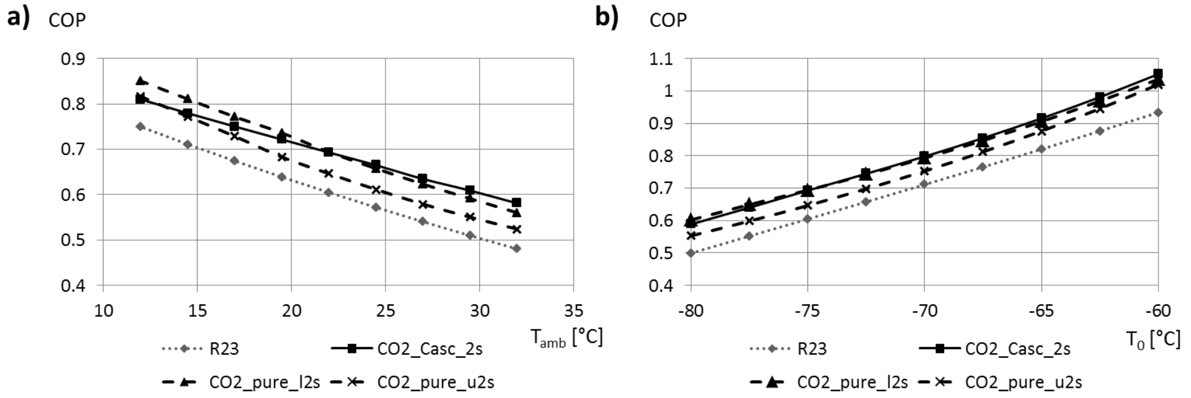

Figure 6: COP of modified sublimation cycles in relation to: a) the ambient temperature T_{amb} (at T_0 = -75 °C); b) sublimation / evaporation temperature T_0 (at T_{amb} = 22 °C)

Providing an internal heat exchanger in the CO_2 system could bring an additional contribution to increase COP in a wide ambient temperature range, as is shown in Figure 7. However, it might have little or even negative impact on the system efficiency at extremely low sublimation temperatures or high ambient temperatures. This could be explained in the following way: The compression ratio and the discharge temperature are as high under these temperature conditions that a further increase in the suction temperature will prevent reaching the optimal pressure level. In addition, the suction volume flow will be increased with the internal heat exchanger and as a further result, the volumetric cooling capacity will be slightly reduced (see Figure 9).

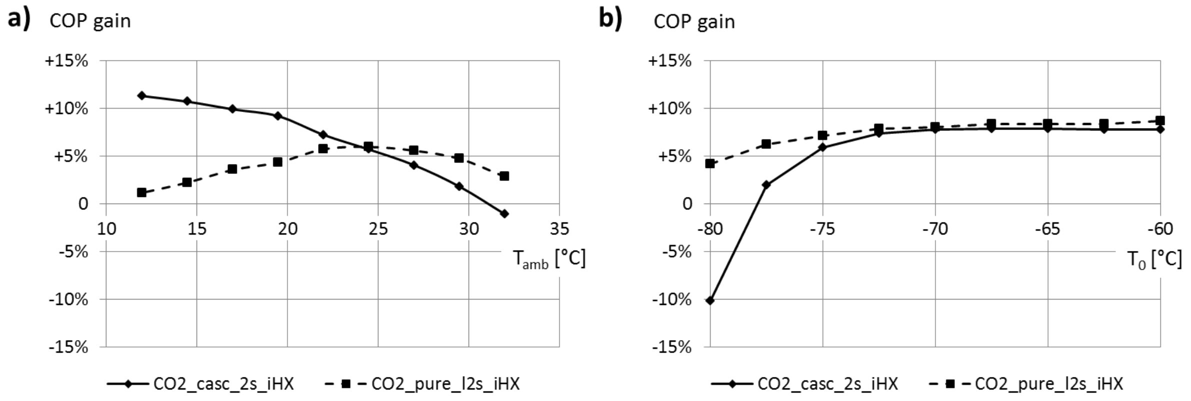

Figure 7: Improvements of COP with the use of internal heat exchanger in relation to: a) the ambient temperature T_{amb} (at $T_0 = -75$ °C); b) sublimation / evaporation temperature T_0 (at $T_{amb} = 22$ °C)

Finally, the overall system efficiency and the volumetric cooling capacity of all investigated systems are presented in Figure 8 and 9, respectively. The most complex systems (CO2_casc_2s_iHX / CO2_pure_l2s_iHX) can achieve a COP-improvement of over 20 % at both evaporation temperatures of -75 °C and -60 °C, compared to the R-23 system. The CO_2-sublimation systems have generally advantages over the R-23 system of having larger volumetric cooling capacities at a sublimation temperature of -60 °C; however, they could drop noticeably as the sublimation temperature decreases. There is no significant difference in COP or cooling capacity between the cascade system and the pure CO_2 system. It should be mentioned here that so far the calculated COP-improvements are related to the defined reference system (R-23). A modified and optimized R-23 cascade system, for example with two-stage compression (R-23_2s) and additional internal heat exchanger (R-23_2s_iHX) could achieve an similar COP-value compared to the modified CO_2-sublimation systems at sublimation / evaporation temperature of -60 °C and even higher efficiency at a lower evaporation temperature of -75 °C. At this point, one important fact should be noted: A two-stage compression in the lower stage is not common in present R-23 cascade systems for certain reasons. R-23 systems usually do not have the problem of a critical compression ratio and high discharge temperature and finally additional costs can be avoided. On the contrary, a multi-stage compression for CO_2-sublimation systems is generally necessary for feasibility considerations. Despite of an increase in costs, the switchover to CO_2 could enforce the use of a more efficient system.

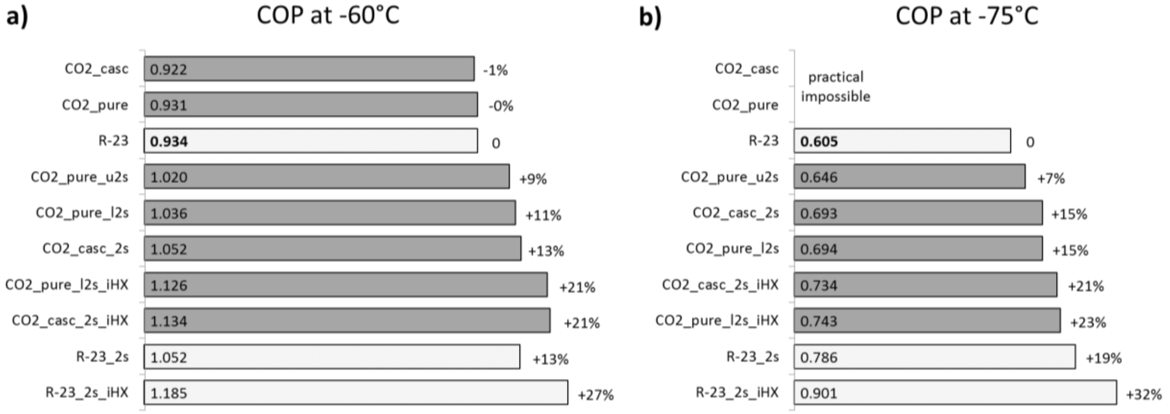

Figure 8: Comparison of COP at ambient temperature of 22 °C and evaporation/sublimation temperature of: a) -60 °C; b) -75 °C

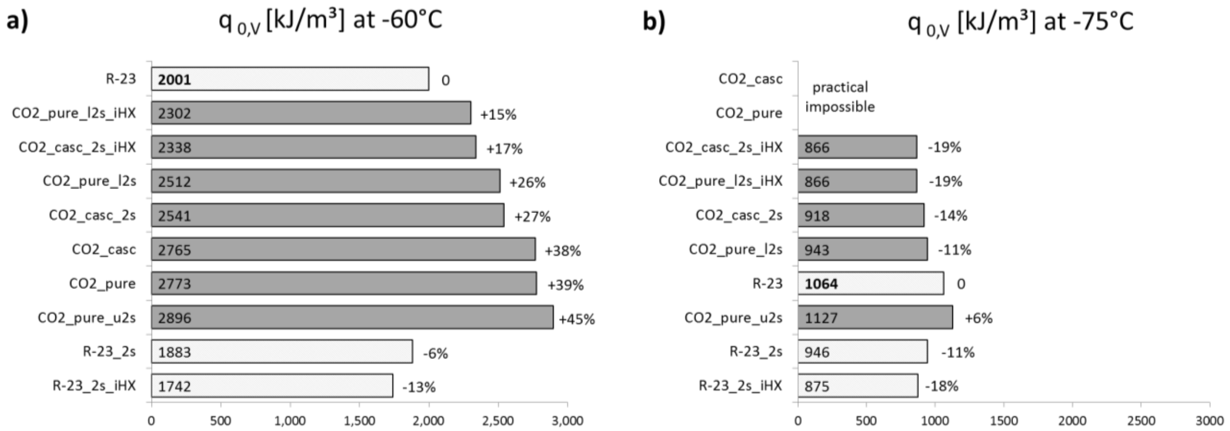

Figure 9: Comparison of volumetric cooling capacity at ambient temperature of 22 °C and evaporation/sublimation temperature of: a) -60 °C; b) -75 °C

3.4 Selection

Taking account of the calculation results and other technical and practical factors, the cascade design with a two-stage compression and an internal heat exchanger is finally chosen. Key arguments for the selection are listed as follows:

- It has been shown that both the pure CO_2 sublimation cycle with intermediate pressure receiver as well as the cascade system could achieve similar COP and cooling capacity under most operation conditions.
- At a high ambient temperature, the pure CO_2 systems have to be operated in transcritical conditions, which lead to a drop in efficiency according to the calculation. Additionally, it is practically difficult to control and optimize three pressure levels.
- A Cascade-system is currently the most popular design in market. It also allows the upper and the lower stage of the system to be adjusted separately. For the use of the CO_2-sublimation cycle in an existing plant, adjustments have to be made only at the lower stage.
- Multi-stage compression in CO_2-sublimation cycles is necessary for a sublimation temperature of -70 °C or lower.
- The internal heat exchanger has a conditionally extra contribution to system efficiency. More important for operation safety issues is the fact, that an extra superheating can prevent rest solid particles from entering the first stage of the compressor.

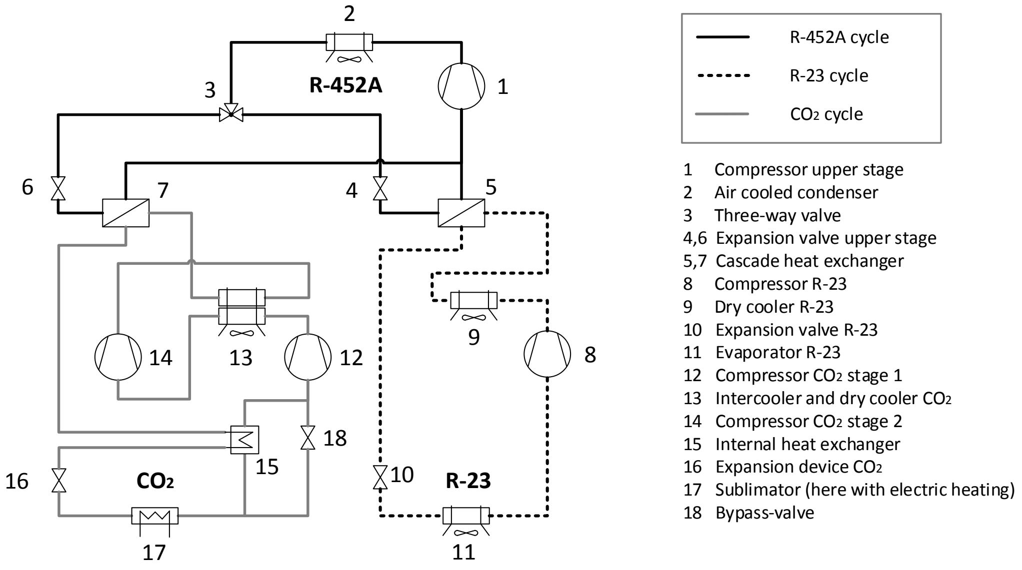

Figure 10: Flow chart for the final design of the cascade machine for comparison of CO_2 with R-23

The final system layout is shown in figure 10. The cascade machine is equipped with a conventional R-23 and a CO_2-sublimation cycle. These two cycles were designed to operate with the same upper stage and the operating mode can be switched by a three-way valve. However, a parallel operation is not planned, simply because the optimal operating conditions of these two cycles are too different. The machine is designed to have a cooling capacity of 1 kW at a sublimation / evaporation temperature of -75 °C for both lower stages. The performance of the machine was calculated, as shown in Table 3.

Table 3: Performance prediction at design point

Cycle		**R-23**	**CO_2**	
Upper Stage (R-452A)				
Evaporation temperature [a]	°C	-24.8	-21.0	
Condensation temperature [b]	°C	35.0	35.0	
Suction pressure	MPa	0.216	0.252	
Suction temperature	°C	-20.0	-16.0	
Suction flowrate	m³/h	3.954	2.649	
Discharge pressure	MPa	1.622	1.622	
Discharge temperature	°C	76.3	69.8	
Compression Ratio	-	7.507	6.436	
Compressor power consumption	kW	0.846	0.567	
Cascade heat exchanger power	kW	1.356	1.074	
Condensation power	kW	2.202	1.641	
Mass flow rate	kg/h	4.50	3.49	
Lower Stage			*1st Stage*	*2nd Stage*
Evaporation / Sublimation temperature	°C	-75.0	-75.0	
Condensation temperature	°C	-17.3	-13.5	
Suction pressure	MPa	0.150	0.134	0.728
Suction temperature	°C	-70.0	-21.5	32.0
Suction flowrate	m³/h	3.385	4.158	0.9052
Discharge pressure	MPa	1.517	0.728	2.392
Discharge temperature	°C	116.0	144.4	147.5
Compression Ratio	-	10.14	5.432	3.285
Cooling Capacity	kW	1	1	
Compressor power consumption	kW	0.807	0.473	0.329
Gas cooler power	kW	0.451	0.344	0.384
Mass flow rate	kW	-	126	
Evaporation temperature [a]	kg/h	22.05	11.84	

[a] referred to dew point; [b] referred to boiling point

4. COMPONENTS DESIGN AND SELECTION

In this section the component design and selection for the cascade refrigeration machine is discussed. Most of the components for the demonstration unit are standard equipment and therefore available on the market. This concerns all components in the R-452A cycle, the R-23 cycle and most of the components in the CO_2 cycle, which operate exclusively with gaseous and liquid CO_2. Therefore, the discussion mainly focuses on the special CO_2 components.

Although there is currently no CO_2 compressor which has been specially developed for the sublimation applications, a conventional subcritical CO_2 compressor can be used without any further modifications. The suction temperatures of -21.5 °C and 32 °C as well as the compression ratios of 5.4 and 3.3, for the first and second stage respectively, are within the normal scope. However it should be mentioned that the absolute pressure levels for sublimation applications are considerably lower than for conventional uses. As a result, the compressor motors, especially for the first stage, are significant underloaded. The necessary volume flow of the compressor can be determined by the theoretical volume flow (according to the calculation) and the volumetric efficiency for a specific compressor (according to the manufacturer). An uncertainty here is the isentropic efficiency of the compressor, which has direct influence on the discharge temperature and the system efficiency. In real operations, the discharge temperature could be higher than the calculated value during high compression ratios. According to the first experimental data from Zhang and Yamaguchi (2009), the discharge temperature is not as critical as expected even with fewer compression stages. This might be explained by the lower motor load of the compressor compared to the normal test conditions. In worst cases, the machine can still be operated with a lower intermediate temperature of the cascade and has lower efficiency as a consequence.

Considering that the CO_2 intercooler and dry cooler have similar cooling capacity, they are designed to be placed parallel to each other as a single heat exchanger unit, in order to reduce space requirements and costs. The air supply is realized with a single fan.

In the throttling device for a CO_2-sublimation system the two-phase state changes from liquid-gas to solid-gas in the narrowest cross sections. This could cause clogging inside the throttling unit itself and should be generally prevented. Thus, geometry without complex flow redirection is preferred, such as an orifice plate, a short capillary tube or special nozzle designs. Initial achievements and experience have been gained in internal tests at TU Dresden using capillary tubes in open cycles. However further investigations under different boundary conditions are still required, in particular in a closed cycle. The construction of the cascade machine gives the flexibility to test different expansion devices.

For the design of an appropriate sublimation evaporator, the knowledge of heat transfer and flow characteristic of the solid-gaseous flow is necessary. First experiments show that the heat transfer coefficient for CO_2 flow sublimation in a single straight pipe could range from 60 to 1800 W/(m² K), depending on the pipe geometry and test conditions (Balduhn and Engelhorn, 1994, Zhang and Yamaguchi, 2011, Iwamoto *et al.*, 2015). However, a general correlation for sublimation heat transfer is not available. Additional studies for heat transfer and flow characteristics are still required. In this design, a U-shaped pipe is used as sublimation evaporator, which is equipped with 2 electric heating sections. The heat transfer in the pipe and flow characteristic, especially through the pipe bend, could be further investigated. The results should serve as a basis for a further sublimation heat exchanger design.

5. CONCLUSIONS

Within this paper, we tried to develop an efficient CO_2-sublimation system for refrigeration applications from -50 °C to -80 °C, in order to replace the conventional, greenhouse-promoting R-23 system. Selection of the cycles and components were discussed based on simulation result of steady state operation. Following conclusions can be made:

- With respect to the high isentropic expansion coefficient and steep vapor pressure curve in the sublimation range of CO_2, a multi-stage compression with intercooling is generally required, in particular for sublimation temperatures below -70 °C.
- The internal heat exchanger could provide protection of the first compression stage against low temperatures and solid particles. While an internal exchanger can have an extra contribution to the system efficiency at most operating conditions, it slightly increases the suction volume flow.
- Both the pure CO_2 system with intermediate pressure receiver and cascade design could achieve a COP-improvement of up to 20 % over the baseline R-23 system, depending on the modification and operating condition. However, a similarly modified R-23 system still has advantages in volumetric cooling capacity and COP in low evaporation temperatures below -75 °C.
- Regarding the calculation results and practical usability, the cascade design with two-stage compression in the lower stage and internal heat exchanger is finally chosen. It is used to build up a demonstration machine, which allows switching the operating mode between the CO_2-sublimation and a conventional R-23 lower stage cycle, so that these two cycles can be compared directly to each other under the same ambient conditions in practice.
- All components for the cascade machine that operates exclusively with gaseous and liquid refrigerants are standard equipment which is available on the market. A normal subcritical CO_2 compressor can be used in the sublimation cycle, while the default motor is oversized.

- Further investigations in expansion devices and heat exchangers for CO_2-sublimation applications are still required.

NOMENCLATURE

COP	coefficient of performance	(–)
GWP_{100}	global warming potential over 100 years	(–)
p	pressure	(MPa)
$q_{0,V}$	volumetric cooling capacity	(kJ/m³)
T	temperature	(°C or K)
ΔT	temperature difference in heat exchanger	(K)
η_{is}	isentropic efficiency of compression	(–)
π	compression ratio	(–)

Subscript

0	evaporation / sublimation
air	air cooled
amb	ambient
casc	cascade
iHX	internal heat exchanger
SC, SH	superheating, subcooling
dis	discharge

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