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High Temperature CO_2 Heat Pumps for Industrial Process Heat Applications

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ABSTRACT

Electrification of heat-generating processes is a key method to decarbonize industrial emissions. Carbon dioxide is one of the oldest known refrigerants, but its use is presently limited to low temperature (< 120 °C) applications, and when operating in a transcritical mode, for heating liquid water and similar non-phase-change materials. Conventional subcritical HFC or PFAS refrigerants also have temperature limitations due to thermal degradation. Finally, most smaller heat pump systems use oil-lubricated positive displacement compressors, which also impose temperature limitations due to the thermal stability of the entrained oil in the refrigerant.

By taking advantage of oil-free, turbomachinery-based equipment and using CO_2 as the refrigerant, the temperature limitations of existing heat pump solutions can be eliminated. Novel cycle architectures, partially derived from sCO_2 power cycle concepts, have been developed that significantly improve the performance of CO_2 heat pumps relative to conventional vapor compression architectures, both for non-phase-change material heating and for medium pressure steam generation. Cycle simulations of these new heat pump cycles that cover a wide range of conditions and applications have been completed. Laboratory-scale (< 50 kW_{th}) demonstration systems have been built and tested, and their measured performance provides validating data for the simulated results.

INTRODUCTION

In many industrial processes, heat is applied to one or more materials at a relatively high temperature. The most common method of creating this heat today is through the combustion of fossil fuels. However, as the world moves towards a carbon-free energy system, alternative methods to provide industrial heat will be required. While direct electrical heating with devices such as resistance, arc or induction heaters can attain the necessary temperatures, the coefficient of performance (COP), defined as the amount of heat transferred to the process divided by the electrical power input, of these processes can never be greater than 1.0.

In contrast, thermodynamic heat pump cycles can attain COP values well in excess of 1.0. However, cycle and working fluid limitations typically only permit modest heating temperatures in heat pumps. For instance, traditional HFC and HFO refrigerants thermally decompose at high temperatures, making them unsuitable for higher temperature heat pump applications (and are facing regulatory bans in many markets due to their high ozone depletion and global warming potentials). A transcritical CO_2 heat pump can provide higher temperature than other refrigerants due to its thermal stability and supercritical state during the transfer of heat from the compressed CO_2 to the heat sink material. Larger scale heat pumps that use oil-free centrifugal compression can also avoid thermal decomposition of the entrained lubricants that typically are used with smaller scale positive displacement compressors.

The thermophysical properties of CO_2 have a profound impact on the heat transfer processes both within and external to the heat pump cycle. Optimal performance of a CO_2 heat pump requires careful matching of the heat capacity characteristics of the working fluid and the heat source / sink fluids and innovative cycle designs. In this paper, we describe CO_2 heat pump cycles that serve two important but diametrically opposed applications. Many applications involve heating a nearly constant specific heat capacity fluid such as air or a single-phase chemical feedstock from a low to a high temperature. The second widely used process heating application is steam generation, which occurs at constant temperature. In these two applications, the heat transfer between the working and process fluids is fundamentally different, and implies that different heat pump cycles will likely be required to optimize performance.

BACKGROUND

In a conventional vapor compression heat pump cycle, the working fluid is compressed from a relatively low temperature, low pressure state to one of higher temperature and pressure. This heat can then be transferred to a medium that receives and either uses or stores that heat. During the process of heating the medium, the working fluid is cooled. The fluid is then expanded to the low-side system pressure (either through an adiabatic expansion valve or a mechanical expander that extracts enthalpy from the fluid by performing shaft work). The temperature and pressure of the fluid decrease at the outlet of the expansion device. Low-temperature heat is then added to the fluid from an external source, in many cases from the environment, or from a medium such as water.

For lower temperature applications, a conventional heat pump cycle provides a good combination of performance and simplicity. Most commercial heat pumps today use hydrofluorocarbon (HFC) or hydrofluoroolefin (HFO) working fluids in a completely subcritical operating state. During the process of transferring heat to the sink, the working fluid is condensing at constant temperature ($T_{range} = T_{outlet} - T_{inlet}$). This type of heat pump/working fluid combination can supply heat at temperatures up to approximately 150 °C [1], limited by working fluid and lubricant thermal stability. The working fluids that are usable at higher temperatures typically have low vapor pressures, which requires vacuum evaporation and risks air entrainment when ambient temperature heat sources are used. Other refrigerants, such as R600/R601 (butane and pentane) have high flammability risks, and others like R717 (anhydrous ammonia) have high toxicity, limiting their potential use in many environments. CO_2, or R744, is a low cost, non-flammable, zero ODP and low GWP refrigerant with low toxicity, making it an attractive refrigerant for large-scale industrial heat pumps.

CO_2 Transcritical Heat Pumps

CO_2 heat pumps typically operate in transcritical mode, where the fluid is supercritical from the compressor exit to the expander inlet, and subcritical over the remainder of the cycle. The high vapor pressure of CO_2 allows for efficient extraction of heat from low temperature sources during its evaporation process, including ambient air or water. During the supercritical pressure process of transferring heat to the sink, the working fluid temperature continuously decreases. This makes CO_2 transcritical cycles ideal candidates for applications where the heat sink temperature needs to be increased over a wide T_{range}, such as domestic hot water heating, and where high temperature waste heat sources are not available.

By using a transcritical CO_2 heat pump, limitations regarding working fluid stability are avoided, and a turbomachinery-based system can be designed without lubricant entrained in the working fluid. However, practical considerations regarding pressure ratio and temperature glide-matching in the high-temperature heat exchanger impact the attainable performance of a conventional transcritical CO_2 heat pump for high temperature range and temperature lift. For instance, the required compressor pressure ratio of a simple non-recuperated transcritical CO_2 heat pump to achieve 300 °C heat sink temperature is greater than 20:1 (Fig. 1).

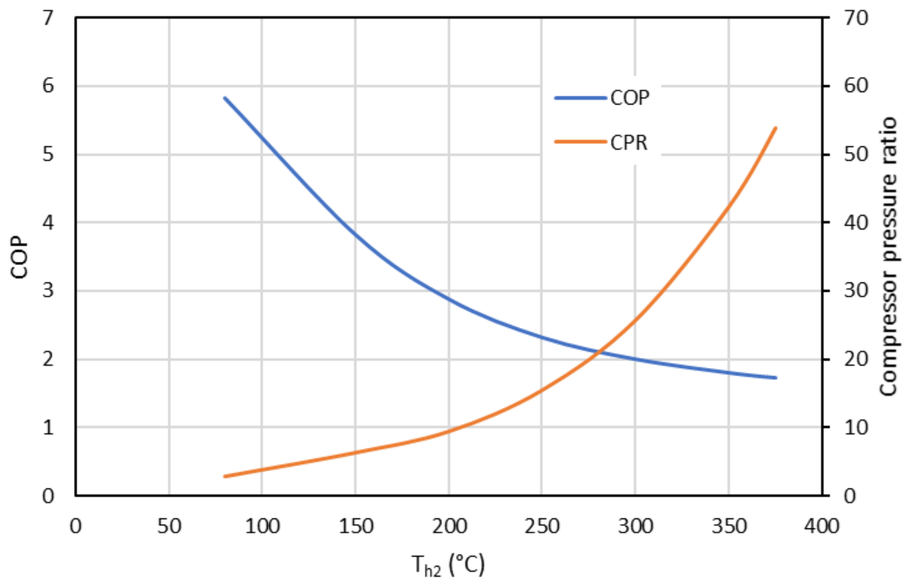


Figure 1: Coefficient of Performance (COP) and required compressor pressure ratio as a function of the maximum temperature of the heated medium (T_{h2}). For these calculations, the initial temperature of the medium is fixed at 20 °C, and the heat source is 15 °C.

HIGH TEMPERATURE CO_2 HEAT PUMPS

In this paper, we are focused on applications at higher temperatures than conventional heat pumps can supply, up to the limits of practicability from a performance and compressor exit temperature limitations, 150 to 400 °C. These higher temperature applications fall into two main categories:

1. Large T_{range}: In these cases, the heat sink temperature needs to be raised from a relatively low to a high value, and the specific heat capacity ($c_p = \partial h/\partial T|_p$) is essentially constant. Examples of the include air heating for drying applications, or crude oil heating for viscosity reduction.

2. Zero T_{range}: When the heat sink undergoes a phase change, the heat sink temperature is constant during the phase transition process. The most common application is steam production.

High temperature process fluid heating

To reduce the compressor pressure ratio required to achieve high heat sink temperatures, a recuperator can be used to recover some of the residual enthalpy exiting the primary heat exchanger to preheat the fluid prior to entering the compressor. The performance of the recuperated heat pump cycle (Fig. 2) is better than the simple heat pump cycle for T_{h2} values greater than approximately 200 to 250 °C (Fig. 3). For these simulations, the compressor pressure ratio was limited to be no greater than 10:1.

While the simple recuperated system can achieve high lift, it is limited by the recuperation process itself to relatively high working fluid temperature exiting the HTX. As a result, the TQ curves have

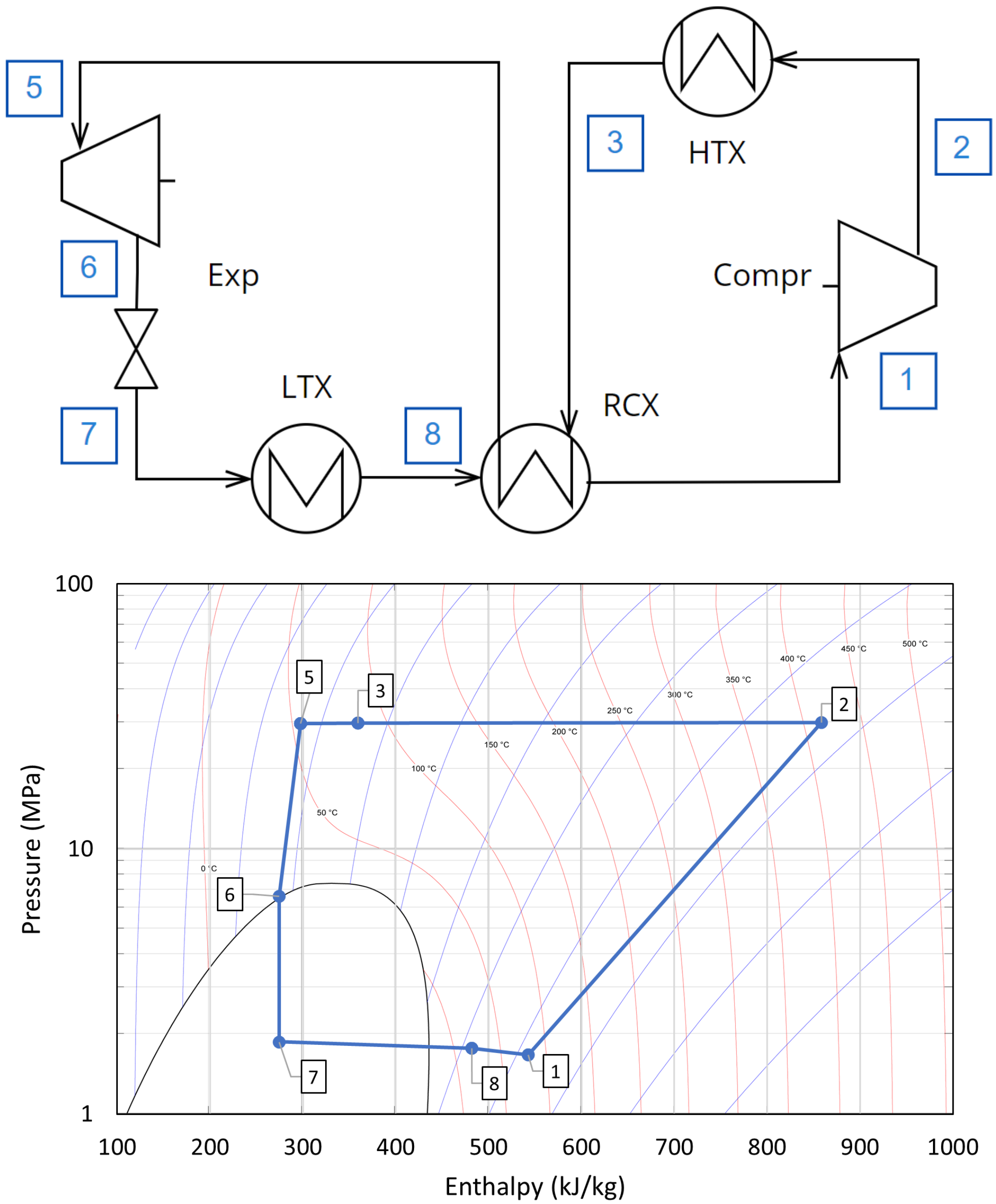


Figure 2: Simple recuperated cycle process flow and pressure-enthalpy diagrams.

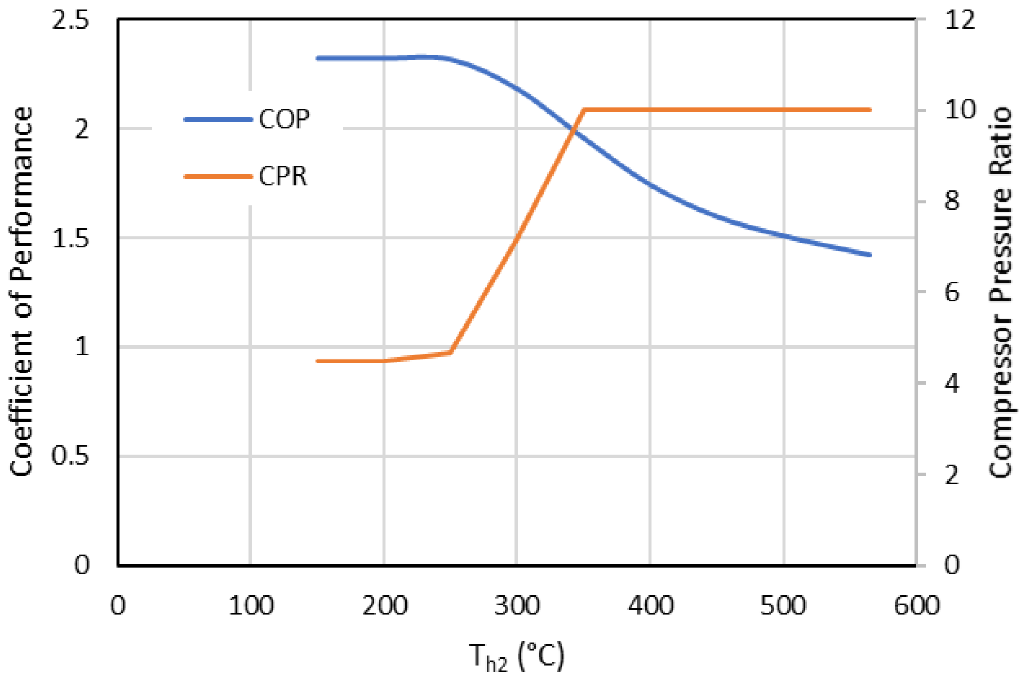


Figure 3: Performance and compressor pressure ratio for the simple recuperated cycle as a function of the thermal medium temperature.

markedly different slopes, which represent exergy loss in the HTX. In addition, the large mismatch of specific heat capacity values at the different pressures within the system also results in a major slope difference in the recuperator TQ plot, further adding exergy destruction to the cycle and reducing its performance (Fig. 4).

To address these limitations, Echogen has developed a new high temperature heat pump (HTHP) cycle [2] (Fig. 5) based on the architecture of its waste heat recovery cycle [3], but operating in the reverse direction. In one version of this cycle, the HTX is divided into two stages. In the first stage, the working fluid transfers heat to the medium, heating the medium and cooling the working fluid. Upon exiting the first stage of the HTX (State 3), the working fluid is then divided into two portions. The first portion (3A) enters a second stage of the HTX, where it is further cooled, and preheats the medium, which is flowing in the opposite direction of the working fluid. The second portion (3B) enters a recuperator (RCX2), where it also is cooled and preheats the working fluid on the low-pressure side of the system (States 8 to 1). Upon exiting the second stage of the HTX (4A) and the recuperator (4B), the flow may recombine (4) and enter into an additional recuperator for further cooling of the high-pressure working fluid (4 to 5) and preheating of the low-pressure working fluid (7 to 8). It then enters the expander (5) as in the simple recuperated cycle, where work is extracted, and the working fluid pressure and temperature are reduced (6). Heat is then added from the environment or similar source (6 to 7).

The division of the working fluid into two portions allows for better matching of the heat capacities in the two sides of the recuperator. In addition, the working fluid can then be cooled to a lower

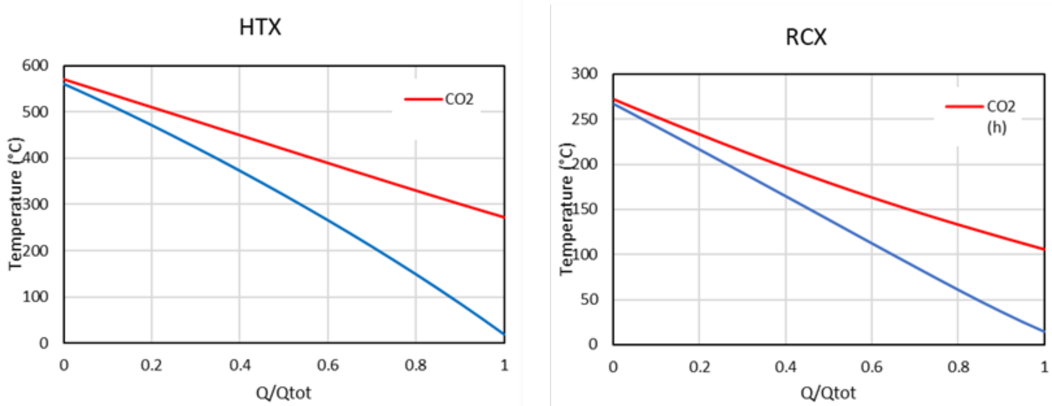


Figure 4: TQ plots for the simple recuperated cycle, high-temperature heat exchanger (HTX) and recuperator (RCX). *** note, need to replace with 400 °C cases ***

temperature than it can in the single recuperated cycle, which also allows its TQ slope to better match the medium thermal slope. Both effects reduce the exergy destruction of the cycle and improve its performance.

To better understand the implication of the refrigerant state during the heat transfer process to the heat sink, we need to consider the fundamental performance parameters associated with the classical heat pump cycle. The limiting performance for a theoretical heat pump transferring heat between two infinite, constant temperature thermal reservoirs is determined by the well-known Carnot equation:

$$COP = (1 - T_c/T_h)^{-1} \tag{1}$$

where T_c and T_h are the temperatures of the infinite cold and hot reservoirs respectively. However, in the case of a finite reservoir, the reservoir temperature will vary as heat is extracted or added. By integrating over a range of infinitesimal reservoirs between the initial and final temperatures of the reservoir, one can derive that for a constant specific heat capacity reservoir material, Eq. (1) still applies with the reservoir temperatures replaced by the thermodynamic (or Lorenz [4]) mean temperatures:

$$\bar{T} = \frac{T_{max} - T_{min}}{\ln\left(T_{max}/T_{min}\right)} \tag{2}$$

where T_{max} and T_{min} represent the temperatures of the reservoirs at the beginning and end of the heat transfer process. Interestingly, the Lorenz COP will exceed the Carnot COP for cases where the heat sink temperature range (defined as $T_{h,max} - T_{h,min}$) increases, leading to the perhaps counter-intuitive result that the ideal COP increases as the heat sink temperature range increases.

While the preceding analysis was defined from the perspective of the thermal reservoir temperatures, it can be helpful to think of the heat pump performance as two separate processes. In the first, we presume a thermal reservoir that exchanges heat from the refrigerant perfectly—that is, with zero temperature difference. In this case, the mean refrigerant temperature is equal to the mean reservoir temperature, in which case the idealized COP is represented by the Lorenz expression above.

In the truly idealized case, the refrigerant temperature is equal to the reservoir temperature

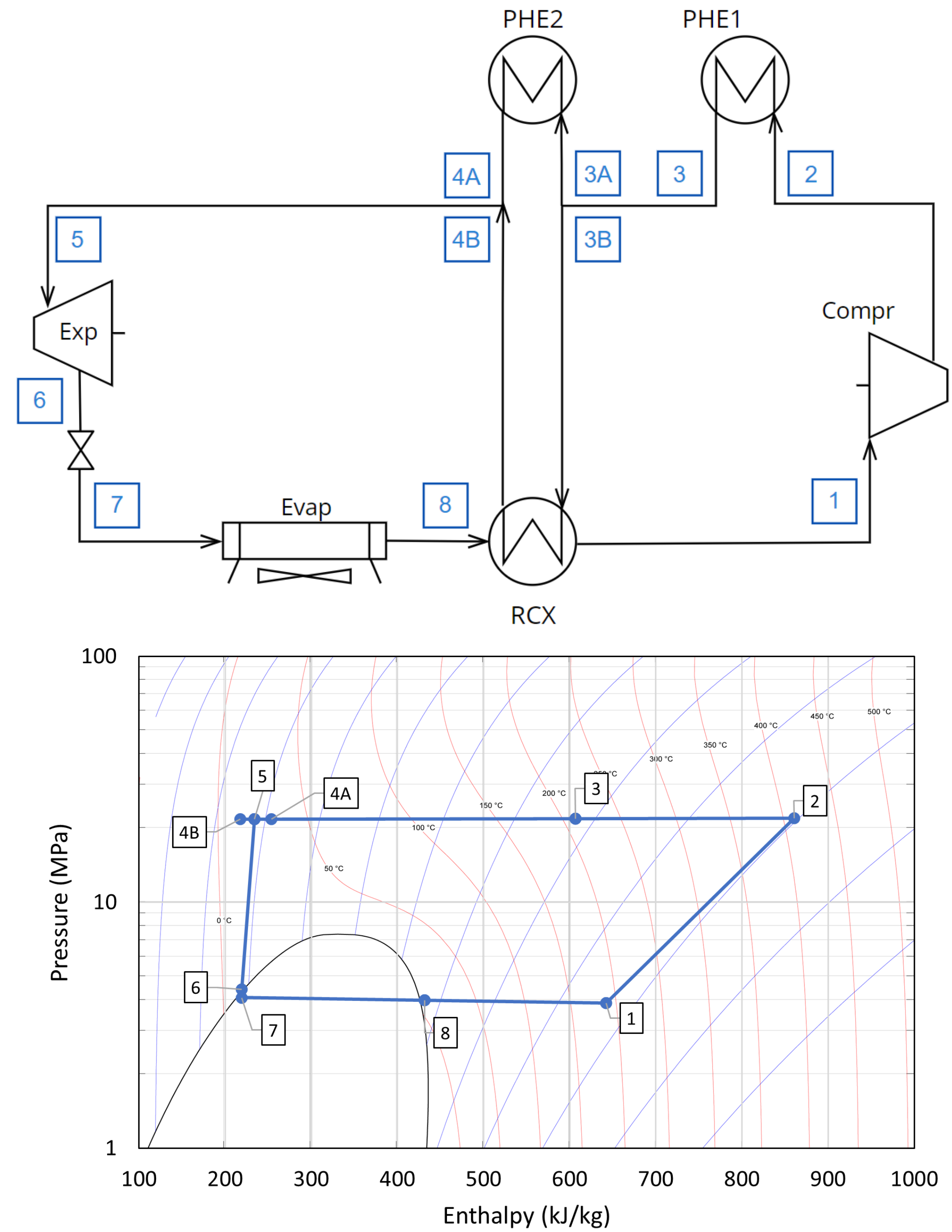


Figure 5: Advanced recuperated cycle process flow and pressure-enthalpy diagrams.

throughout the heat transfer process, and thus we could calculate the COP with the Lorenz equation above using the refrigerant fluid mean temperatures ($\bar{T}_{f,h}$ and $\bar{T}_{f,c}$). In the less idealized case where finite temperature differences exist between the refrigerant and the reservoirs[1], the maximum practical COP is still represented by the Lorenz expression with the refrigerant mean temperatures, which will necessarily be lower than the COP defined by the reservoir temperature. One can show that the fractional reduction in COP due to the minimum temperature difference between refrigerant and reservoir ($\Delta T_{min} = \bar{T}_{f,h} - \bar{T}_{r,h}$) can be approximated as:

$$\frac{\Delta COP}{COP} = \Delta T_{min} \left(T_{lift}^{-1} - T_{r,h}^{-1} \right) \tag{3}$$

where $T_{lift} = \bar{T}_{r,h} - \bar{T}_c$. The minimum temperature difference ΔT_{min} can be reduced by increasing the heat transfer area of the heat exchanger, but only to the point where the fluid and reservoir temperatures are equal at any point in the heat exchanger (known as the "pinch" point). Further improvements can only be achieved by better matching the fluid and reservoir temperatures as a function of the amount of heat transferred, a process known as "glide-matching".

The application we are targeting here is single-phase process fluid heating—air in particular, but the same thermodynamic principles apply for any single-phase fluid (or solid, for that matter, in the case of particle heating). In this case, the TQ plot of the heat sink is a straight line, with a slope of $1/\left(\dot{m}c_p\right)$. For a subcritical heat pump, the refrigerant curve on the TQ plot is a straight line with zero slope, since the refrigerant condenses at constant temperature as it transfers heat to the heat sink. As a result, even with an infinitely large heat exchanger, we can show that

$$\Delta T_{min} > T_f - \frac{T_f - T_{r,min}}{ln(T_f/T_{r,min})} \tag{4}$$

Since a supercritical fluid has a non-zero c_p, no such restriction exists for the CO_2 HTHP, providing it a fundamental performance advantage.

However, the specific heat capacity of CO_2 at the supercritical state proposed here is a moderately strong function of temperature and pressure. Exergy analysis [5] shows that the most efficient heat transfer process in a counterflow heat exchanger is one in which the TQ curves of the two materials are parallel. The flexibility of the HTHP cycle allows the CO_2 TQ curve to be divided into multiple segments by varying the proportion of the CO_2 flow rate through each section of the PHE. As the slope is proportional to $1/\left(\dot{m}c_p\right)$, this variation in $\dot{m}_{CO_2}$ allows close matching of the TQ slopes of the two materials. This cycle innovation results in significant improvement in COP, as well as reduces the required compressor pressure ratio to achieve a target heat sink temperature.

The performance of the HTHP is calculated by system modeling as a function of the heat source temperature and heat sink peak temperature (Fig. 6). Over the range of conditions simulated, the system COP ranges from a low of 2.0 at −20 °C heat source temperature and 400 °C discharge temperature to over 4.0 at 40 °C and 200 °C.

Steam generation

The process of steam generation has a fundamentally different character from the process fluid heating case above, and requires a different approach to heat pump design. In this case, the heat

[1] We will confine our attention to the heat sink, or "hot reservoir", for the remainder of the analysis.

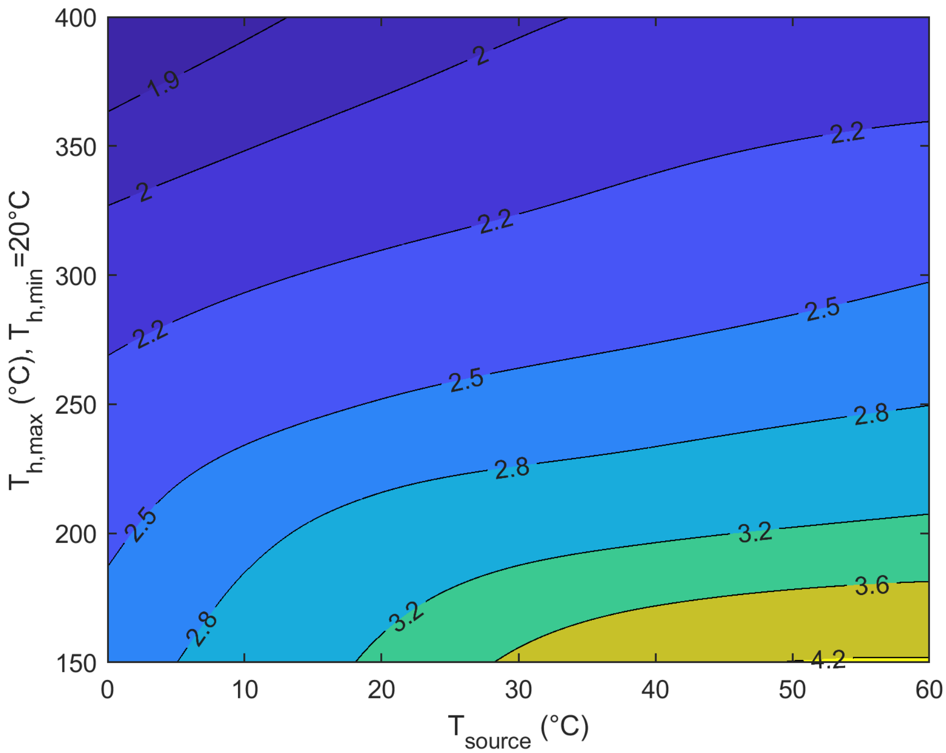


Figure 6: COP map for CO_2 HTHP, when heating air from an initial temperature of 20 °C.

transfer takes place at $T_{range} = 0$, while the CO_2 working fluid, being in the supercritical state, continuously decreases in temperature. As noted in the previous section, the addition of a recuperator to the simple cycle lowers the temperature range of the working fluid. However, for the steam generator application, this approach is of limited value. For instance, a simple recuperated heat pump, when generating 16.5 bar steam (T_{sat}=202 °C), is required to operate at a very high compressor discharge temperature to avoid a pinch restriction at the right-hand side of the TQ plot shown below:

Another potential issue with this configuration is the discharge state of the expander (state 5 in **??**), which can be seen on the PH diagram below to fall squarely within the vapor dome (need figure). Thus, a significant fraction of the expanded fluid will have flashed to the vapor state in the later stages of the expander, which could substantially affect both the performance and the durability of this device and is generally avoided in practice.

Finally, the COP of this cycle is limited as the Lorenz mean temperature of the high temperature heat addition process is relatively high. For this example, $T_{m,h}$=321 °C and the predicted COP is 1.48. To reduce $T_{m,h}$, one can split the compression processes into two or more separate stages and extract heat between each. While this approach does reduce $T_{m,h}$ to 261 °C, the COP only raises slightly to 1.50, and the expansion into the dome persists. The fundamental problem with this cycle (at least while using CO_2 as the working fluid) is that the large dependence of heat capacity on temperature and pressure causes a mismatch in the temperature "glide" between the

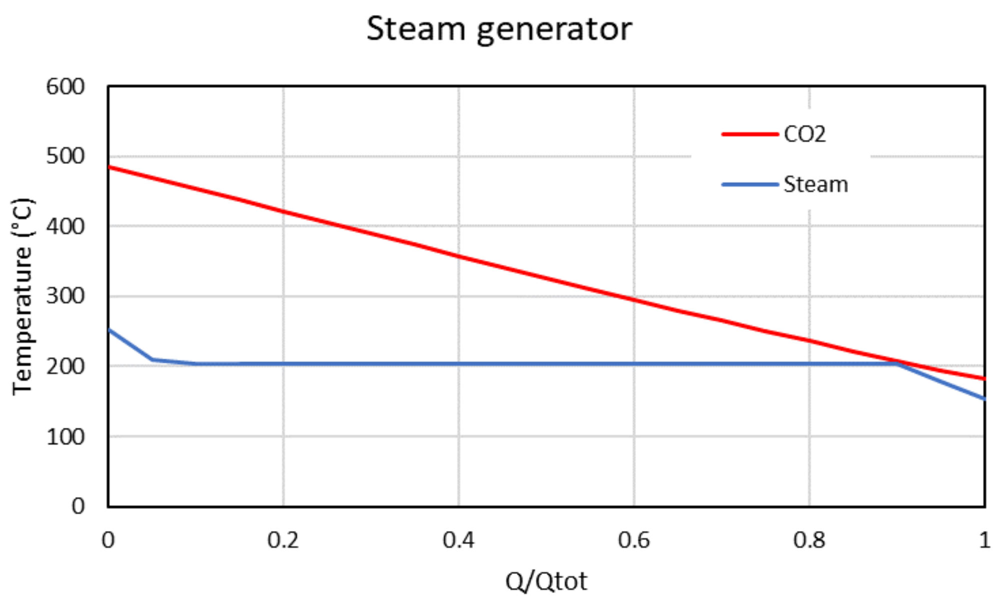


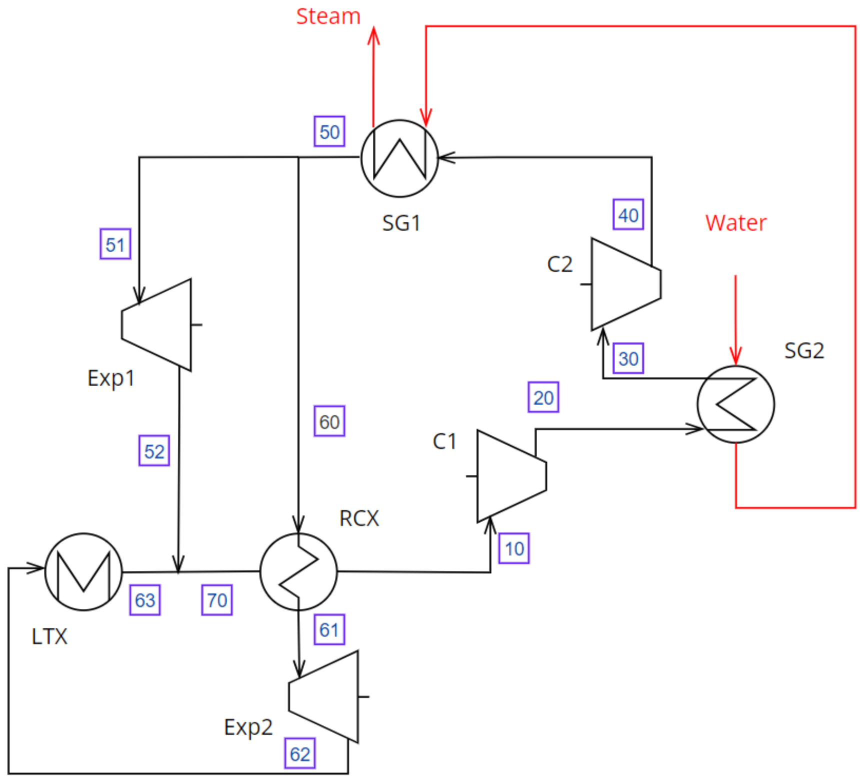
Figure 7: TQ plot for the steam generator on a simple recuperated CO_2 heat pump cycle.

low-pressure and high-pressure sides of the recuperator. This causes a large temperature difference between the high-pressure CO_2 exit and the low-pressure CO_2 entrance of the recuperator, which in turn represents a large exergy destruction in the recuperator, and thus a loss in performance (need figure).

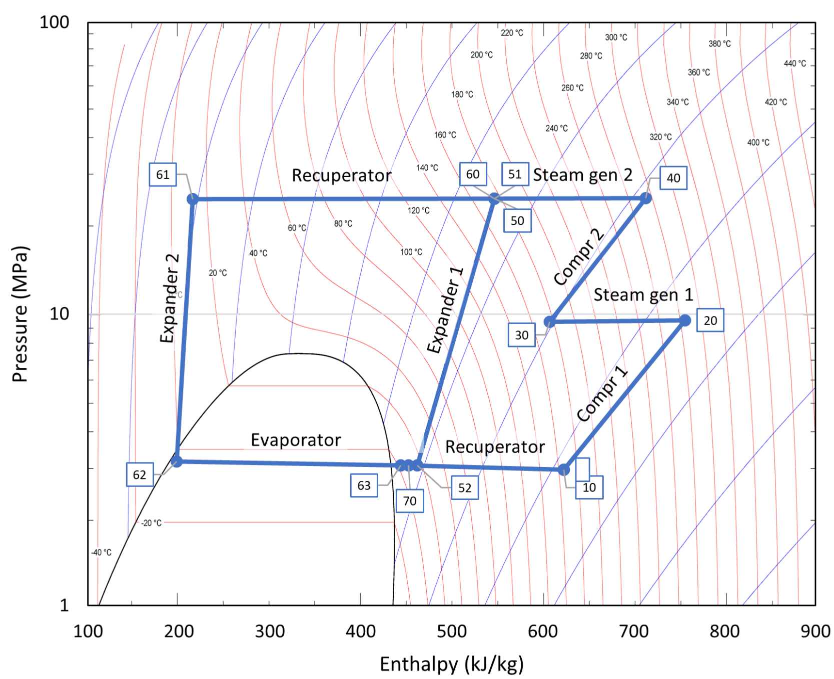
The new heat pump steam generator (HPSG) cycle [6] (Fig. 8) addresses the heat capacity mismatch between the two fluid streams by dividing the flow at the outlet of Steam Generator 2 and expanding a portion directly through a turbine (expander). The fraction of the total mass flow rate at state 60 is roughly proportional to the ratio of the specific heat capacities at states 60 and 70, such that $\dot{m}_{60}c_{p,60} = \dot{m}_{70}c_{p,70}$. This cycle increases the COP markedly over the previous two cycles. For this set of assumptions, the COP is 1.63, vs 1.50 for the previous best prediction. The improvement in performance is due to several factors:

1. The mean temperature of the high-temperature process $T_{m,h}$ is further reduced to 256 °C using the same steam conditions as the previous analysis.
2. The heat capacity of the two flow streams in the recuperator is now well-matched, significantly reducing the exergy destruction in that heat exchanger.
3. The work recovered in the expander offsets a portion of the compressor work.
4. By better matching the heat capacities of the two streams, a much lower temperature at state 51 can be achieved. Thus, the lower-temperature expander (T1) exit is now a single-phase liquid, or low-vapor level mixture of liquid and vapor. The higher-temperature expander (T2) exit is also single-phase, on the vapor side of the 2-phase region.

For comparison, a model of a 2-stage subcritical vapor compression heat pump was created us-



(a) Process flow diagram



(b) Pressure-enthalpy diagram

Figure 8: CO_2 HPSG cycle. Boxed numbers refer to state points

ing the same modeling approach as for the CO_2 HPSG, with R1233zd(E) as the refrigerant and a 40 °C heat source (lower heat source temperatures required subatmospheric low-side pressure, or a separate cascaded heat pump with a lower normal boiling point refrigerant). Atmospheric pressure steam was generated by the vapor compression heat pump, and compressed to higher pressure in a 2-stage steam compressor with boiler feedwater injection between stages. The COP calculated using this cycle was 2.24, or 12% higher than the HPSG at the same conditions. Although from a strict performance perspective, the HPSG cannot match the vapor compression plus steam compression system, the other advantages of the HPSG are significant:

- Operational simplicity: Steam can be generated directly in a single heat pump at pressures well above 20 barg.
- Low-temperature heat source capability: CO_2 heat pumps can effectively use heat from sources that are well below 0 °C due to the high vapor pressure of CO_2.
- Thermal storage integration: Due to the large temperature differential in the SG1 and SG2 heat exchangers, conventional sensible enthalpy thermal energy storage materials can be used.
- Refrigerant cost and safety: Unlike HFC and HFO refrigerants, CO_2 is not likely to be banned imminently. And unlike butane or ammonia, the flammability and toxicity risks of CO_2 are low.

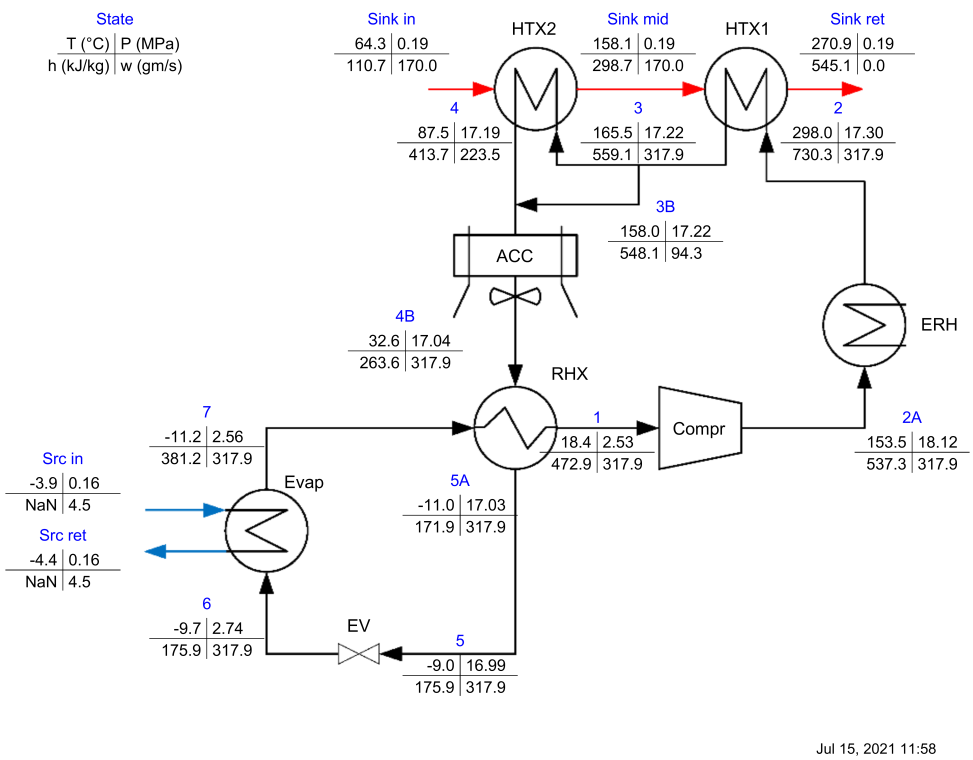
PILOT-SCALE TESTING

The feasibility of the both cycles have been further demonstrated experimentally at a laboratory scale. A 2-stage, water-cooled, oil-free reciprocating compressor (Hydro-Pac LX-series) forms the basis of the laboratory-scale system (Fig. 11), with the two stages acting in series. Due to piston seal temperature limitations, the compressor cylinders are water cooled. For the HTHP cycle, the non-adiabatic nature of the compressor is compensated by electric boost heaters that simulate the temperature at the exit of an adiabatic compression process that would be typical of the larger, non-cooled machines that will be used on the pilot and commercial scale HTHPs. At this point, the flow is then directed to a 2-stage heat exchanger that transfers heat to a single-phase heat transfer fluid (HTF). At an intermediate point in this heat exchanger, a portion of the CO_2 flow is diverted to a recuperator to preheat the low-pressure CO_2 flow prior to being compressed, while the remainder transfers additional heat to the HTF. The two flows then merge and expand through an adiabatic valve to the low pressure side of the system. At this point, the CO_2 is in the 2-phase regime, with a vapor content of less than 20% by mass. The 2-phase CO_2 is then fully evaporated at −2 °C, then preheated in the recuperator before closing the cycle.

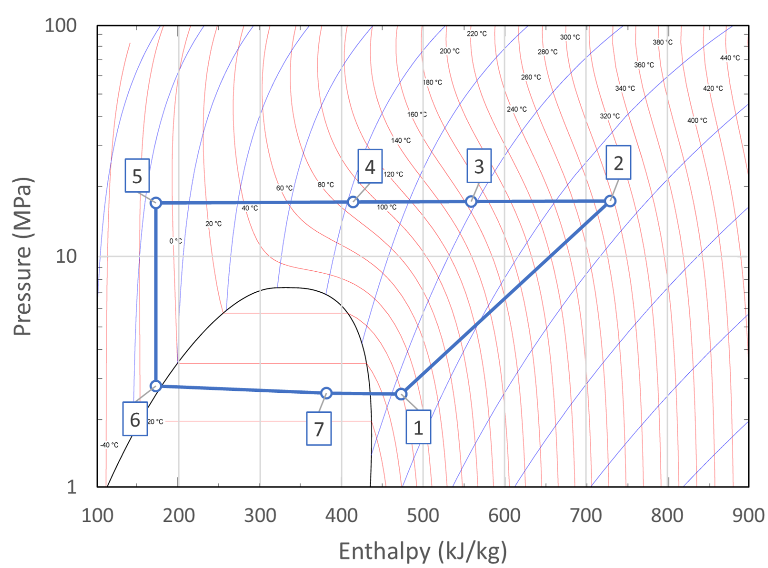
The portion of CO_2 flow diverted to the high-pressure side of the recuperator was controlled to approximately match the heat capacity ($\dot{m}c_p$) of the fluid on the two sides of the recuperator, minimizing the exergy destruction and subsequent performance loss in the recuperator.

Due to the small scale of the system, the expander in the full-scale system is simulated with a simple control valve. In addition, the inlet temperature limitation of the reciprocating compressor is non-representative for a full scale system. However, the fundamental process was demonstrated, and process heat generation achieved at a laboratory scale, providing a high degree of confidence in the system design principles.

The same compressor was used to demonstrate the HPSG cycle at laboratory scale. Based on the



(a) Process flow diagram



(b) Pressure-enthalpy diagram

Figure 9: Example pressure-enthalpy diagram of operational laboratory CO_2 HTHP. Dashed line indicates simulated adiabatic compression path.

earlier testing, the original equipment manufacturer piston ring and rider rings were replaced with a set of graphite-filled PTFE components, which were capable of significantly higher temperature operation. In addition, the cylinder water cooling and the auxiliary heat rejection used in the previous testing were eliminated.

Also for the heat pump steam generator testing, a set of tube-in-tube heat exchangers, through which heated CO_2 flowed in the inner tube, and water/steam through the annular gap between tubes, were installed after each compressor stage. After the first compressor stage, the heated CO_2 transferred heat to liquid water in the first set of tube-in-tube heat exchangers (SG2) then returned to the second compressor stage (C2) for further compression and heating, before returning to a second set of tube-in-tube heat exchangers (SG1) to complete the steam generation and partially cool the CO_2.

Downstream of SG1, the CO_2 flow was split into two parallel paths. Part of the flow was passed to the recuperator (RCX) to preheat the low-pressure CO_2 prior to entering C1. Following the recuperator, this CO_2 was expanded in a control valve to the system low-side pressure, reducing its temperature. The 2-phase CO_2 was then vaporized in a water-to-CO_2 heat exchanger.

The remainder of the CO_2 was expanded directly through a parallel control valve, and mixed with the vaporized CO_2 from the evaporator discharge, at which point the mixed fluid was then heated by the first portion of the CO_2 in the recuperator. By appropriate flow split modulation, the heat capacity ($\dot{m}c_p$) of the fluid on the two sides of the recuperator is matched, minimizing the exergy destruction and subsequent performance loss in the recuperator. The effectiveness of this approach to heat capacity matching was demonstrated during the test program, as shown in Fig. 12

Due to the small scale of the system, the two expanders in the full-scale system were simulated with simple control valves. In addition, inlet temperature limitations of the reciprocating compressor limited the system performance. However, the fundamental process was demonstrated, and steam generation achieved at a laboratory scale, providing a high degree of confidence in the system design principles.

SUMMARY

Industrial heating processes require electrified processes to reduce carbon emissions from fossil-fired heating sources. With the new CO_2 heat pump cycles described in this paper, two significant gaps in temperature capability can be closed. Echogen is presently developing two projects to demonstrate both cycles at pilot scale (500 kW_{th}) for the HTHP cycle, and commercial scale (10 MW_{th}) for the HPSG system.

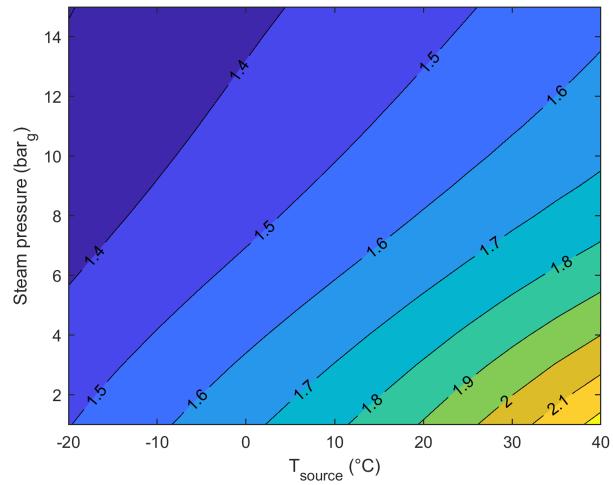


Figure 10: COP map for CO_2 HPSG.

Figure 11: Laboratory-scale HPSG using low-speed reciprocating compressor.

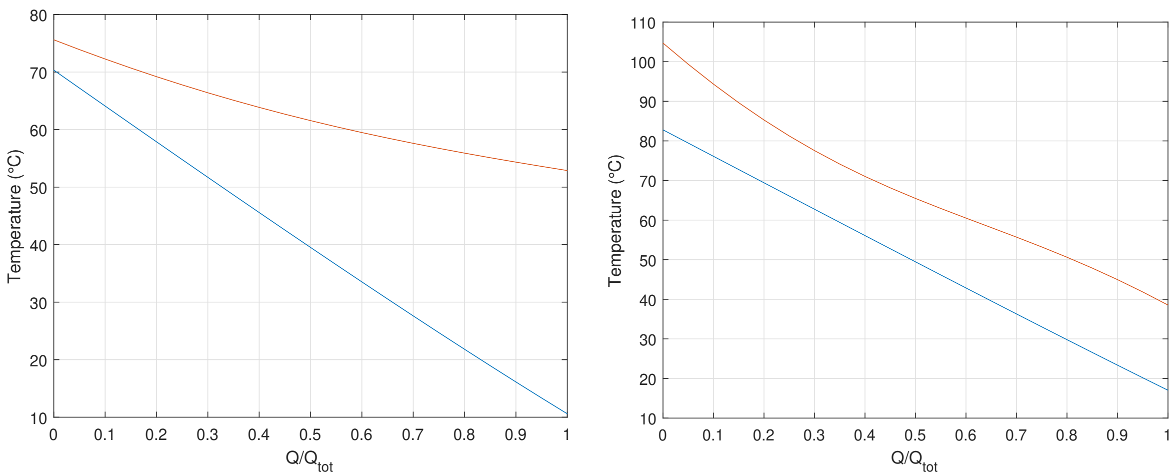
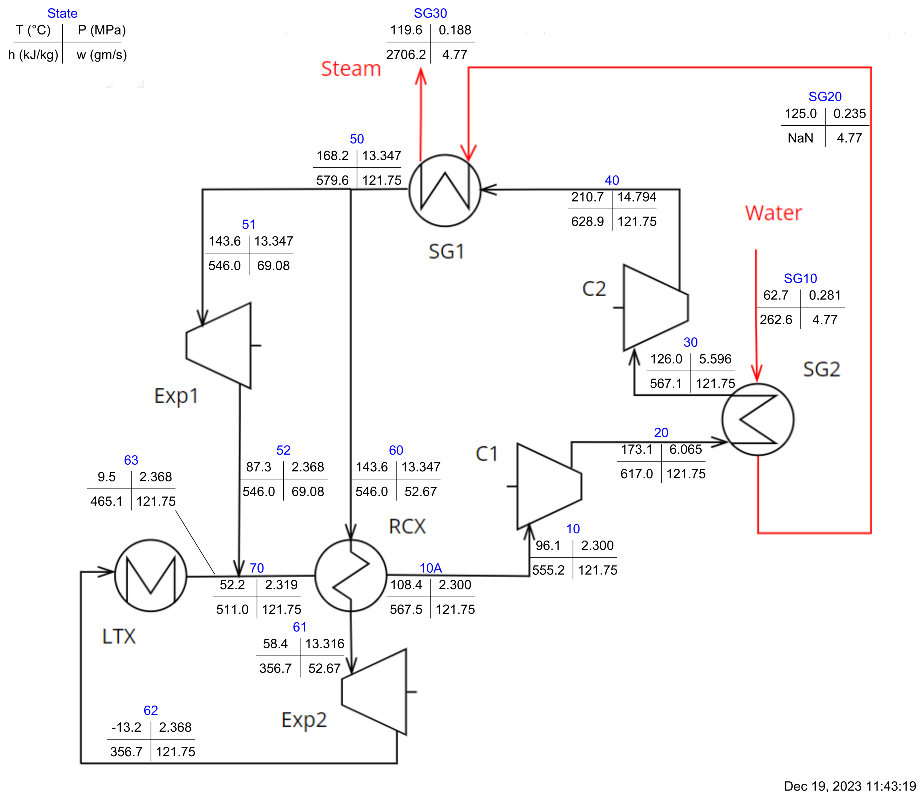
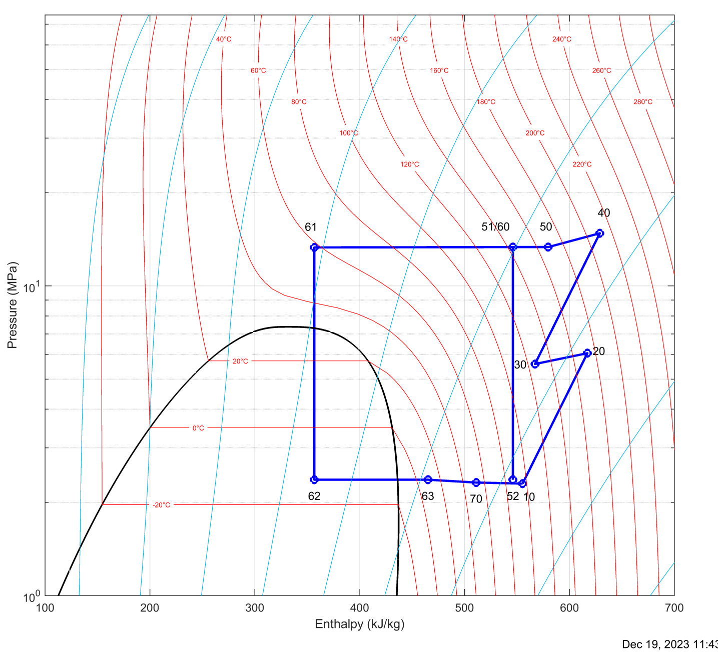


Figure 12: TQ plots for the recuperator based on measured inlet/outlet temperatures during laboratory-scale testing. The plot on the left is representative of a 2-stage heat pump without the high-temperature exander, while the other plot shows the TQ behavior with approximately 53% of the flow extracted through the high-temperature expander. The difference in slopes in the first figure represents a large loss in thermodynamic potential, or exergy.



(a) Process flow diagram



(b) Pressure-enthalpy diagram

Figure 13: [[Example pressure-enthalpy diagram of operational laboratory CO_2 HPSG.]]

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