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Thermodynamic Competitiveness of High Temperature Vapour Compression Heat Pumps for Boiler Substitution

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Abstract

Replacement of fossil fuel-fired boilers for generation of high-temperature water and steam is of paramount importance for reaching a fossil-free economy. Heat pumps could be possible substitutes, but the presently marketed solutions are mainly limited to low-temperature applications. This paper compares the thermodynamic performance of various High-Temperature Heat Pumps (HTHP) based on the vapor compression cycle with natural gas boilers for assessing their technical feasibility and competitiveness as future replacements. Different cycle configurations and natural refrigerants were compared based on the coefficient of system performance and exergetic efficiency. Moreover, the technical feasibility of the most interesting cycles was discussed based on current technologies. The results showed that HTHP are promising substitutes for gas boilers for heat production of temperatures up to 180 °C. In particular, ammonia cycles were preferable for source and sink temperatures lower than 60 °C and 110 °C, respectively, while water was a better-working fluid at higher temperatures. The complete exergy analysis showed different behaviors for various natural refrigerants in the working domain but confirmed the results identified by the energy analysis. In any case, the technological analysis showed that further improvement of compressor technologies is required for making these solutions competitive.

Keywords: Boiler substitution, electrification, heat pumps, industrial energy use, natural refrigerants.

1. Introduction

Climate change is a major threat to society. In response, many countries are trying to phase out fossil fuels, moving towards a more electrified society, which would be suitable for a high penetration of renewables in the energy system [1]. One of the challenges this shift entails is the substitution of a large share of industrial energy use, e.g., for generation of steam by boilers. A possible solution is the usage of High-Temperature Heat Pumps (HTHP). These devices have the potential of producing heat by means of electricity in an efficient way, while also reducing the energy use of industrial processes by upgrading the temperature of waste heat [2]. Unfortunately, state of the art solutions are limited by the maximum possible supply temperature, which makes it difficult to substitute boilers for applications above 100 °C. According to Arpagaus et al. [3], only 20 marketed models, all employing synthetic refrigerants, can supply temperatures up to 150 °C. However, substantial barriers limit their application in the industrial practice, such as lack of experience in their integration and large payback periods, caused by high component cost [4]. In this way, the large demand of medium temperature heat (between 100 °C and 400 °C), responsible for 19 % of the overall European industrial heat demand and generally supplied via steam systems [5], cannot be addressed by this technology. Brunin et al. [6] and Ommen et al. [7] investigated the working domain of vapor compression HTHP utilizing various refrigerants for temperatures of up to 200 °C. They highlighted

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Nomenclature

T	Temperature, K	F	Fuel
$\dot{E}$	Exergy flow rate, kW	H	High stage
e	specific exergy, kJ/kg	i	i -th state point
h	Specific enthalpy, kJ/kg	L	Low stage
s	Specific entropy, kJ/kgK	m	Intermediate stage
<i>Abbreviations</i>			
COP	Coefficient Of Performance	P	Product
COSP	Coefficient Of System Performance	boiler	Boiler
ME	Marginal Exergy	crit	Critical point
<i>Greek letters</i>			
η	Efficiency	el	electric
π	Pressure ratio	in	Inlet
ε	Exergetic efficiency	max	Maximum
<i>Subscripts</i>			
0	Ambient state point	PH	Physical
D	Destruction	sink	Sink
		source	Source
		th	Thermal

the technical and economic limitations of current technologies, agreeing that the most limiting component appears to be the compressor. In fact, traditionally utilized positive displacement compressors present harsh operational constraints in terms of achievable temperature and pressure. In particular, two factors establish the most stringent limitations: (i) refrigerant critical point and (ii) compressor operating envelope. While the former is an intrinsic property of the working fluid and can be handled by changing the medium, the latter is a technological issue that has to be addressed by means of technical development. Different compression technologies are used depending on the working fluid considering economic constraints. In particular, oil lubricated positive displacement compressors are commonly utilized for HFC, HFO, hydrocarbons, and ammonia [3]. However, marketed solutions used for refrigeration are limited in the maximum pressure and temperature achievable. A survey conducted by Ommen et al.[7] revealed a pressure upper limit of 28 bar for hydrocarbon compressors, while according to Emerson Climate Technologies[8] a pressure of 76 bar is achievable for ammonia compressors. In all the cases, lubrication by means of oil sets a temperature limit at the compressor discharge. According to Neksa et al.[9] the safety limit for avoiding oil denaturation is 180 °C for most of the commonly utilized oils.

Possible solutions for achieving higher temperatures with natural refrigerants are the utilization of hybrid compression-absorption heat pumps [10] or the usage of water as working fluid, coupled with enhanced compression technologies. The latter has been of high interest in the past years, for developing compressors able to provide high displacements and withstand high temperatures, while still attaining reasonable capital investments. Positive displacement compressors could be utilized, as proved by Chamoun et al.[11], but centrifugal compressors also seem attractive, as they may achieve higher isentropic efficiencies and have moderate external dimensions [12,13], resulting in lower specific capital investment [14]. The technical limiting point in this technology is the compression ratio per stage, due to a limitation in the peripheral speed of the blades. According to Šarevski and Šarevski[15], a limit of 4.7 is found for the pressure ratio. Similar values are also confirmed by Madsbøll et al.[16], while GEA[17] reports a limit of 2.5 for marketed solutions of compressors used for Mechanical Vapour Recompression in drying processes. Multi-stage compression

configurations are required for achieving high temperature lift, increasing the capital investment of the system. A reduction in cost is paramount for their application in heat pumps, as discussed by Lachner et al. [18]. To this end, Bantle et al. [19], Bantle [20], Bantle et al. [21] and Madsbøll et al. [16] recently modelled and tested centrifugal steam compressors based on turbo-compressor units mass produced for the automotive industry, showing promising performance for the application in cost-effective HTHP. However, these units are in the prototyping phase and a marketed solution is yet to come.

The objective of this study was to investigate the performance of possible vapor compression HTHP working with various natural refrigerants in order to assess their thermodynamic competitiveness as a replacement for natural gas boilers. Ultimately, this may serve as a rational basis for evaluating R&D investments for achieving higher supply temperatures at affordable costs. Subcritical single stage, two-stage compression with open intercooler and cascade systems were analyzed utilizing four different natural refrigerants: propane (R290), isobutane (R600a), ammonia (R717) and water (R718). They were compared based on both Coefficient of System Performance and exergetic efficiency, analyzing their actual working domain based on currently available technologies, and their potential working domain considering a technological improvement. Lastly, a detailed exergy analysis at component level was performed, introducing a novel set of exergy-based indicators, named *Marginal Exergy* product, fuel and destruction.

2. Methods

Examination of the vapor compression heat pump performance compared to natural gas boilers was carried out for three different cycles and four natural refrigerants. A model of each heat pump, as well as of the natural gas boiler, was implemented in Engineering Equation Solver [22].

2.1. Vapor compression heat pump

Schematics of the considered natural gas boiler system (a) and the three heat pump cycles are presented in Fig. 1. The single stage (b) and two-stage compression (c) cycles were investigated considering four natural refrigerants (R290, R600a, R717 and R718), while the cascade system (d) was evaluated in the sole configuration utilising R717 in the low-temperature cycle and R718 in the high-temperature cycle, although other possible fluid combinations might be possible. This choice was made for enlarging the heat pump working domain and is further discussed in Section 3.

Similar assumptions were considered for all the cycles and working fluids, in order to ease their comparison. In industrial applications, heat is usually transferred by means of fluids, e.g, water, steam or oil. Both source and sink media were considered to be pure water in liquid state, which was at ambient pressure in case of the source and at a pressure of 15 bar for the sink, in order to avoid vaporization in the evaluated temperature range. Fixed temperature glides were applied to the two media and heat exchange was modeled in all heat exchangers by a defined pinch point temperature difference.

The boiler performance was calculated by assuming constant efficiencies, while the heat pump models were based on constant efficiencies for the compressor and electric motor. Pressure losses in the heat pump system were accounted for by assigning fixed drops of saturation temperature. This is a typical approach utilized in the refrigeration industry, as presented by Granryd [23]. Constant superheat after evaporators was assumed. Subcooling was allowed, ensuring to locate a pinch point at the condenser outlet. Compressor heat losses were disregarded. The used values are presented in Table 1.

Finally, the intermediate pressure in both the two-stage and cascade cycles was assumed. For the former, it was set according to Equation 1 as discussed by Jiang et al. [24], while for the latter Equation 2 [24] was preferred, setting the bottom cycle condensation temperature, due to the usage of two different working fluids.

$$p_m = \sqrt{p_H \cdot p_L} \quad (1)$$

$$T_m = \sqrt{T_H \cdot T_L} \quad (2)$$

Where p is pressure, T is absolute temperature, H refers to high stage condenser, L to low stage evaporation and m to intermediate stage conditions.

Figure 1: Principle sketch of: (a) boiler, (b) single stage HTHP, (c) double stage HTHP and (d) cascade HTHP.

2.2. Operation limits

As presented in Section 1, different compression technologies are generally utilised based on the adopted working fluid. In this work, positive displacement compressors were considered in case of R290, R600a, and R717, while turbo-compressors were adopted for R718. Operation limits were assumed according to Ommen et al.[7] and Šarevski and Šarevski[15]. In case of R718, due to the low compression ratio it was decided to consider two water compressors in series also in the single stage and the cascade systems. The utilized limiting operation data are presented in Table 2. The heat pumps were assumed to operate subcritically.

2.3. Thermodynamic analysis

Heat pumps and natural gas boilers were compared based on thermodynamic performance. Two main indicators were utilized as a base of comparison, one based on the First Law of thermodynamics, namely Coefficient of System Performance (COSP) and one based on the Second Law of thermodynamics, namely exergetic efficiency (ε). The COSP was defined as the ratio of useful energy supplied as heat and the primary energy necessary to supply it, or in other terms, the product of Coefficient of Performance (COP) of the heat pump and primary energy to electricity conversion efficiency (η_{el}) as described in Equation 3:

$$\text{COSP} = \eta_{\text{el}} \cdot \text{COP} \quad (3)$$

Following its definition, this performance indicator can be compared consistently to the boiler thermal efficiency.

The exergy analysis was performed as suggested by Adrian Bejan, George Tsatsaronis[25], assuming an ambient temperature of $T_0 = 20^\circ\text{C}$ and ambient pressure of $p_0 = 1.013 \text{ bar}$. The kinetic and potential

Table 1: Modelling assumptions for heat pumps and gas boiler.

<i>Type of data</i>	<i>Value</i>	<i>Unit</i>	<i>Designation</i>
Efficiency	0.8	-	Compressor isentropic efficiency
	0.95	-	Compressor motor electric efficiency
	0.9	-	Natural gas boiler efficiency
	0.4	-	Primary energy to electricity conversion efficiency
Temperature	7	K	Compression suction superheat
	8	K	Pinch point temperature difference in heat exchangers
	10	K	Source temperature lift
	20	K	Sink temperature lift
Pressure	0.3	K	Saturation temperature drop due to pressure drops in pipes
	1	K	Saturation temperature drop due to pressure drops in heat exchangers
	1	bar	Source water pressure
	15	bar	Sink water pressure

Table 2: Applied operation limits and critical point of various working fluids.

<i>Working fluid</i>	<i>Pressure limit, bar</i>	<i>Temperature limit, °C</i>	<i>Pressure ratio limit, -</i>	<i>Critical pressure, bar</i>	<i>Critical temperature, °C</i>
R290	28	180	-	42.5	96.7
R600a	28	180	-	36.5	134.6
R717	76	180	-	111.3	132.4
R718	-	-	22.1	217.7	373.9

components of exergy were neglected. Moreover, chemical reactions occur only in the boiler system. Hence, the chemical component of exergy was considered only for the natural gas stream in the boiler, for which the specific exergy content was assumed equal to 51,384 kJ/kg [25]. Finally, the physical component of exergy at the i -th state point was calculated as Equation 4:

$$e_i^{\text{PH}} = (h_i - h_0) - T_0 \cdot (s_i - s_0) \quad (4)$$

Where e is the specific exergy content, h is the specific enthalpy, and s is the specific entropy of the i -th state point (i) and the ambient (0). The superscript PH refers to the physical component of exergy. The exergetic efficiency of each system was calculated as Equation 5:

$$\varepsilon = \frac{\dot{E}_P}{\dot{E}_F} \quad (5)$$

Where $\dot{E}$ is the exergy flow of product (P) and fuel (F) streams. The term product refers to the exergy provided to the sink, while fuel relates to the exergy supplied by both heat source and compressor. Finally, a detailed exergy analysis was conducted on the heat pumps at components level. Two further parameters were considered, namely Exergy destruction flow ($\dot{E}_D$) (Equation 6) and Marginal exergy (ME_k) (Equation 7) product, fuel and destruction, with respect to parameter k .

$$\dot{E}_D = \dot{E}_F - \dot{E}_P \quad (6)$$

$$\text{ME}_{k,i} = \frac{\frac{d}{dk}(\dot{E}_i)}{\dot{E}_i} \quad \text{for } i \in \{P, F, D\} \quad (7)$$

In this study, marginal exergy was calculated with respect to the sink supply temperature. The derivative was approximated by the difference quotient.

As defined in Equation 7, marginal exergy represents the ratio between the local derivative of exergy flow with respect to parameter k (in this case sink supply temperature), and the exergy flow itself. This indicator is particularly useful for describing the exergy efficiency trend of a system under varying conditions, both internal to the studied system, and at its boundary (as in this study). It can be demonstrated (Appendix A), that the derivative of the exergetic efficiency of a system, with respect to parameter k is proportional to $(ME_{k,P} - ME_{k,D})$. Hence, wherever the so defined marginal exergy product is lower than the marginal exergy destruction, the derivative of exergetic efficiency is negative, wherever they are equal, the derivative is nil, and wherever the marginal exergy product is higher than the marginal exergy destruction, the derivative is positive. Thanks to this property, the marginal exergy concept can analytically characterize the exergetic efficiency trend and, most importantly, identify the components exerting the highest influence on it. A high marginal exergy product in a system component detects a positive influence on the trend of exergetic efficiency, while a high marginal exergy destruction at component level points out a negative influence on it.

3. Results

The comparison of HTHP and natural gas boilers was firstly assessed based on First Law performance. Fig. 2 and Fig. 3 present the results of the investigation, showing technical limitations and COSP compared with boiler efficiency. The first figure is for single stage and two-stage cycles, while the latter presents the cascade system results.

Hatched areas in the two figures depict working domains for which the operation of the heat pump was considered unfeasible. In this respect, the upper line represents supercritical conditions ($p_H > p_{crit}$), while the lower one shows conditions for which heat recovery by means of direct heat exchange would be possible ($T_{sink,in} < T_{source,in}$). This is because the sink inlet temperature (sink,in) is lower than the source inlet temperature (source,in). In this case, the potential for direct heat exchange should be exploited prior to a heat pump installation [2]. Red dashed lines illustrate operation limits of marketed compressors, in terms of pressure ($p_H > p_{max}$), temperature ($T_H > T_{max}$) or pressure ratio ($\pi > \pi_{max}$). Based on this, the bright-colored area represents the working domain achievable with available technologies, while the shaded one shows the operation points reachable contingent upon technological improvement is attained. Finally, the isolines depict the heat pump COSP and should be compared with the dashed-dotted line representing the boiler efficiency ($\eta_{th,boiler}$): for all the operation points with COSP higher than $\eta_{th,boiler}$, the installation of a heat pump in replacement of a natural gas boiler would be beneficial in terms of primary energy consumption.

By examining Fig. 2, it can be noted that all the studied systems reached better first-law based efficiencies compared to natural gas boilers in most of their working domain. Boilers performed better only at high temperature lifts, above 60 °C for R600a single stage cycle, which showed the lowest performance, up to more than 100 °C for R718. However, it is clear how sink temperatures above 110 °C are difficult to reach with current technologies (bright-colored area), no matter the cycle. The only fluid capable of such high temperatures was water, whose usage was unfortunately limited by the availability of high-temperature heat sources. Temperatures higher than 110 °C were achievable only with heat source available at more than 60 °C.

Concerning the other fluids, they have severe limitations at high temperatures, mainly caused by the high pressures required, which exceed the critical pressure and would require a transcritical cycle to be considered. This is especially the case of R290, which could not supply sink temperatures above 100 °C in subcritical conditions.

Exploring the feasible working domains, it was also clear that water was the working fluid with the highest potential for future applications at high temperatures. In fact, no other fluid was capable of achieving temperatures higher than 135 °C in subcritical conditions. This consideration was also valid analyzing the thermodynamic performance. R290 and R600a showed the lowest COSP throughout the whole working domain, while R717 and R718 raised it considerably, especially in the single stage cycle configuration. The last two had very close values, with R717 performing better at source temperatures below 60 °C, while R718 performed better at high source temperatures. Finally, it could be noted how the two-stage cycle provided a general improvement in COSP and, in the case of R717, it considerably enlarged the feasible working domain

relying on current technologies. This was thanks to a reduction of compressor discharge temperature at high temperature lifts, which limited the feasible operating envelope in the single stage configuration. The same benefit was not reported for the other operating fluids, as compressor discharge temperature did not result to be a limiting factor.

Figure 2: Working domain and COSP of different fluids and cycles. Isolines represent COSP, dotted lines operation limits and dashed-dotted lines boiler efficiency.

This result inspired the investigation of a cascade system using R717 as the low temperature working fluid and R718 as the high temperature working fluid, trying to exploit their respective strengths, both in terms of achievable lift and COSP, for enlarging the working domain using currently available technology. The results are presented in Fig. 3.

Figure 3: Working domain and COSP of R717/R718 cascade cycle. Isolines represent COSP, dotted lines operation limits, and dashed-dotted lines boiler efficiency.

As it can be noted, this configuration significantly enlarged the working domain, with respect to all the previously presented cycles, allowing to reach sink temperatures above 140 °C with source temperatures as low as 30 °C. The performance reduction in terms of COSP was appreciable, but still limited to around 0.5 as maximum value, ensuring a better performance compared to natural gas boilers for temperature lifts as high as 95 °C. This established the R717/R718 cascade system as a valid substitute for natural gas boilers relying on marketed compression technology.

In order to further investigate the benefits of replacing gas boilers by means of high-temperature heat pumps, a Second Law approach was utilized, comparing the exergy efficiencies of the various devices. Fig. 4 presents the results of this analysis. Exergy efficiencies of different cycles and working fluids were compared with the exergy efficiencies of natural gas boilers for varying sink and source temperatures. The source temperature was varied uniformly between 60 °C and 90 °C. Markers in the figure pinpoint the exergetic efficiency maximum of the drawn curves. Values for R290 are not shown for clarity, due to the fact that they lay slightly below R600a ones creating confusion.

As it can be noted, all the heat pump cycles performed better than natural gas boilers in terms of exergetic efficiency, no matter the used working fluid. They could reach efficiencies close to 70 % and a difference of almost 40 % if compared to natural gas boilers. Comparing different working fluids, R600a showed lower performance than R717 and R718, in accordance with the analysis of COSP. Moreover, also for exergetic efficiency, R717 and R718 were close in values, with R717 performing better for low source and sink temperatures, while R718 reached higher values in case of high sink and source temperatures. As expected, the double stage cycle was capable of higher exergetic efficiencies if compared to the others, while the cascade cycle had performance similar to the single stage heat pump running with R718. Finally, all the working fluids showed convex exergetic efficiency trends, suggesting that a maximum existed. This was spotted for R600a in both single stage and double stage cycles, and for R717 in case of single stage cycle (denoted by circles in Fig. 4). R718 did not show an absolute maximum in the investigated temperature domain, suggesting that its performance could be further improved by raising the sink temperature. The exergetic efficiency of the various HTHP in the full investigated working domain is reported in Appendix B.

The convex trend raising the sink temperature spotted for exergetic efficiency profoundly differed from the monotonically decreasing one of COSP. In order to further understand the reason of this result, a detailed

Figure 4: Exergetic efficiency varying sink and source temperature, for different fluids and cycle configurations, compared to natural gas boilers. Source temperature was varied from 60 °C to 90 °C.

exergy analysis was performed for one case, corresponding to the R717 single stage cycle with a source inlet temperature of 30 °C. The results are shown in Fig. 5. Full lines represent marginal exergy of fuel, product, and destruction, while dotted lines depict exergy flows for the same components. As it can be noted, the marginal exergy was always positive for all the components, proving the monotonically increasing trend of exergy flows. However, below 121 °C the marginal exergy destruction was lower than the marginal exergy of product, whereas above this sink temperature it became higher. This justifies the convex trend of the exergetic efficiency. Moreover, it can be proven that the maximum of exergetic efficiency appeared exactly where the marginal exergy destruction equalled the marginal exergy of product, as analytically proven in [Appendix A](#). This is graphically confirmed by comparing Fig. 4a and Fig. 5.

In order to understand the cause of this higher increase in marginal exergy destruction if compared to marginal exergy of product, a detailed exergy analysis was conducted at the component level for the same heat pump. Fig. 6 shows that the components responsible for the convex exergetic efficiency trend were mainly the expansion valve and the condenser. In fact, in the former, the highest marginal exergy destruction took place, while the latter showed the biggest marginal exergy of product. However, although both of them dropped by increasing the sink outlet temperature, the second one dropped much faster. In this way, the cumulated marginal exergy of products became smaller than the marginal exergy destruction, as spotted in Fig. 5. Furthermore, by looking at exergy flows, Fig. 6 shows that the expansion valve was the only component with concave trends. This generated the lower decay in marginal exergy destruction, with respect to all the other components. Evaporator and compressor did not appear to have a significant

Figure 5: Marginal exergy and exergy flow of fuel, product and destruction for the R717 single stage heat pump with an inlet source temperature of 30 °C.

impact on the overall marginal exergy variation, as the former showed very low values for all marginal exergy components, while the latter did not show relevant variations in these indicators, compared to expansion valve and condenser. It should be hereby noted how the limited influence of the compressor, with respect to exergetic efficiency trends would not be evident without considering marginal exergy components. On the contrary, by only looking at exergy flows, this would appear as the most influent element, showing the highest exergy product flow and the highest difference between exergy product and exergy destruction flows.

4. Discussion

Heat pumps and natural gas boilers were compared based on a First Law (COSP) and a Second Law (ε) performance. However, the best base for comparison of devices using different energy sources was not unanimously identified and was, as today, still object of debate. Both of the used indexes present, in fact, some drawbacks.

COSP was a good attempt at comparing the two systems based on primary energy consumption, considering the average conversion efficiency from primary to secondary energy (natural gas to electricity). However, this parameter does not account for the concept of energy quality, not considering how energy sources were utilized if compared to their maximum potential. Moreover, the definition of the average conversion efficiency is of primary importance for retrieving reliable results, as proven by the work of Bless et al.[26]. This is not trivial, as this factor embeds many uncertainties related to the definition of relevant system boundaries. Enlarging geographical boundaries (e.g. from a single plant, to a regional electricity grid, up to a national or continental grid) it is increasingly difficult to determine a reliable average primary energy to electricity conversion efficiency, due to the usage of different conversion technologies in various ambient conditions. Moreover, the definition of such parameter was not straightforward for all the production plants. This is the case, for example, of plants with multiple products (e.g. Combined Heat and Power and trigeneration plants) and devices able to convert renewable energies to electricity (e.g. wind turbines, photovoltaic panels, concentrated solar plants). Finally, this value was likely to change in time, due to technological shift and improvements. The chosen value of 40 % represented a conservative estimate, as many fossil fuel-fired power plants reached higher conversion efficiency [27]. As aforementioned, already in this case heat pumps performed better than natural gas boilers in most of the investigated temperature domain. Therefore, they presented themselves as good solutions for natural gas boilers replacement. Moreover, in an

Figure 6: Marginal exergy and exergy flow of fuel, product and destruction for different components of the R717 single stage heat pump with an inlet source temperature of 30 °C.

economy moving towards a higher penetration of renewable energy technologies, the conversion efficiency (as defined in this paper) would raise up to 100 % or close to that value. In this case, the heat pump COSP would be much higher and equal to their COP, making heat pumps favourable over natural gas boilers in all the considered conditions. This finds agreement in other reported studies [3,26,28]. The COP performance in the investigated working domain is reported in Appendix B.

Exergetic efficiency, on the other hand, accounts for the quality of energy and showed higher values for heat pumps than for natural gas boilers in the totality of the investigated working domain. It was considered the fairest basis of comparison from a thermodynamic perspective because it combines the First and Second Laws of thermodynamics. However, it did not compare the two devices based on primary energy, due to the fact that the considered heat pumps were electrically driven, which was commonly retained to be a secondary energy source [29]. In a future where electricity is totally produced by means of renewable energy sources, electricity could arguably be considered a primary energy source, increasing the consistency of exergy as the basis of comparison. In any other case, life cycle concepts would enter the discussion in order to fairly compare distributed energy sources (e.g. sunlight, wind energy) with concentrated energy sources (e.g. fossil fuels), making other types of analysis necessary, such as Life Cycle Assessment (LCA) [30], exergoenvironmental analysis [31] or emergy analysis [32]. They were however accompanied by high uncertainties in their formulation, for which reason the results of such analyses need careful investigation and might be challenged.

No economic parameter was taken into consideration, performing an analysis purely based on thermodynamics. Despite comparing different heat supply technologies under energy and exergy point of view, filling a reported gap in the literature [33], these results could be of less interest from an industrial perspective,

concerning the working domains in which a boiler substitution would be beneficial from an investor viewpoint. However, the aim of the work was to state a basis for taking research and development decisions based on sustainability, with a long-term perspective. In this context, economic evaluation might involve considerable uncertainty related to the estimate of prices for non-marketed items. Nonetheless, future work could assess investment costs for such devices or required price levels in order to make them acceptable in the current/future energy market.

Finally, a novel set of indicators based on the exergy concept, named *Marginal Exergy* (Equation 7) was proposed. Its usefulness was two folded: (i) it described the exergetic efficiency trend, and (ii) when applied at component level, it analytically identified the components responsible for it. These features proved to be particularly useful in the performed analysis, as the exergetic efficiency of the system was analysed under varying boundary conditions. An analysis based on the sole exergy flows, as traditionally performed, would not be conclusive in this case. For instance, the expansion valve was found to be the main cause for the exergetic efficiency degradation increasing the temperature lift. However, the analysis of the exergy flows would not prove this point, as the exergy destruction flow in the expansion valve was lower than, or comparable to, the one of the other components (Figure 6). On the contrary, when considering the marginal exergy destruction, it is apparent how the highest value was attributable to the expansion valve. Similarly, the marginal exergy concept allowed to get deeper insight for the other components, as discussed in Section 3. To this end, the marginal exergy concept could be a useful integration to the already well established indicators proper of exergy analysis, in order to better characterize the sources of inefficiencies in thermodynamic systems and their role under varying operative conditions.

5. Conclusion

Three different High-Temperature Heat Pump (HTHP) cycles, employing four different natural refrigerants, were compared to natural gas boilers for heat production, from a thermodynamic perspective. Two performance parameters were considered, namely: (i) Coefficient Of System Performance and (ii) exergetic efficiency. HTHP was found to be competitive for boiler substitution in delivering heat of up to 180 °C with temperature lifts of up to 160 °C. In this temperature domain, they can perform better in terms of both COSP and ε . R717 was the best performing working fluid for low source and sink temperatures, below 60 °C and 110 °C, respectively, while R718 performed better at a higher temperature. Moreover, two-stage cycles were able to achieve the highest efficiencies.

Despite providing the same results in terms of HTHP substitution competitiveness, exergetic efficiency showed different trends than COSP, when raising the sink temperature. While the former followed a convex trend with a maximum, the latter decreased monotonically. A detailed exergy analysis proved that the trend with maximum was caused by the slower decrease in marginal exergy destruction in the expansion valve if compared to the marginal exergy of product in the condenser when the condensation temperature (and pressure) increased. In this way, the aggregated marginal exergy destruction of the HTHP became greater than the one of product, generating the maximum trend.

Finally, the examination of the working domain utilizing available technologies suggested that reaching very high temperatures might be challenging with the currently available technology. However, the results showed a promising potential for future technological development, especially for devices employing R718 as working fluid.

Appendix A. Derivation of marginal exergy

The following provides an insight in the derivation and definition of marginal exergy, as presented in Equation 7.

Directly from the definition of exergetic efficiency (Equation 5) and exergy destruction (Equation 6), Equation 5 can be rearranged as:

$$\varepsilon = \frac{\dot{E}_P}{\dot{E}_P + \dot{E}_D} \quad (\text{A.1})$$

The derivative of exergetic efficiency with respect to parameter k is then:

$$\frac{d}{dk}(\varepsilon) = \frac{d}{dk} \left(\frac{\dot{E}_P}{\dot{E}_P + \dot{E}_D} \right) \quad (\text{A.2})$$

Or, elaborating on Equation A.2 by applying the quotient rule:

$$\begin{aligned} \frac{d}{dk}(\varepsilon) &= \frac{\frac{d}{dk}(\dot{E}_P) \cdot (\dot{E}_P + \dot{E}_D) - \dot{E}_P \cdot \left(\frac{d}{dk}(\dot{E}_P) + \frac{d}{dk}(\dot{E}_D) \right)}{(\dot{E}_P + \dot{E}_D)^2} \\ &= \frac{\frac{d}{dk}(\dot{E}_P) \cdot \dot{E}_P + \frac{d}{dk}(\dot{E}_P) \cdot \dot{E}_D - \frac{d}{dk}(\dot{E}_P) \cdot \dot{E}_P - \frac{d}{dk}(\dot{E}_D) \cdot \dot{E}_P}{(\dot{E}_P + \dot{E}_D)^2} \\ &= \frac{\frac{d}{dk}(\dot{E}_P) \cdot \dot{E}_D - \frac{d}{dk}(\dot{E}_D) \cdot \dot{E}_P}{(\dot{E}_P + \dot{E}_D)^2} \\ &= \frac{\frac{\frac{d}{dk}(\dot{E}_P)}{\dot{E}_P} - \frac{\frac{d}{dk}(\dot{E}_D)}{\dot{E}_D}}{\frac{(\dot{E}_P + \dot{E}_D)^2}{\dot{E}_P \cdot \dot{E}_D}} \\ &= \left(\frac{\frac{d}{dk}(\dot{E}_P)}{\dot{E}_P} - \frac{\frac{d}{dk}(\dot{E}_D)}{\dot{E}_D} \right) \cdot \frac{\dot{E}_P \cdot \dot{E}_D}{(\dot{E}_P + \dot{E}_D)^2} \end{aligned} \quad (\text{A.3})$$

Considering the definition of *marginal exergy* given in Eq. 7, Eq. A.3 can be written as:

$$\frac{d}{dk}(\varepsilon) = (\text{ME}_{k,P} - \text{ME}_{k,D}) \cdot \frac{\dot{E}_P \cdot \dot{E}_D}{(\dot{E}_P + \dot{E}_D)^2} \quad (\text{A.4})$$

The right-hand term of the above equation is always positive, as $\dot{E}_D \geq 0$ due to the second law of thermodynamics, and $\dot{E}_P \geq 0$ if the exergy product is correctly defined. Consequently, the sign of the derivative depends only on the left-hand term and can be completely described by considering marginal exergy product and destruction. More specifically:

$$\frac{d}{dk}(\varepsilon) \begin{cases} < 0, & \text{for } \text{ME}_{k,P} < \text{ME}_{k,D} \\ = 0, & \text{for } \text{ME}_{k,P} = \text{ME}_{k,D} \\ > 0, & \text{for } \text{ME}_{k,P} > \text{ME}_{k,D} \end{cases} \quad (\text{A.5})$$

Appendix B. Additional cycles performance

The following presents additional information on the performance of HTHP in the studied working domain. Fig.B.7 and Fig.B.8 respectively depict the COP and exergetic efficiency of the R717/R718 cascade system, while Fig.B.9 and Fig.B.10 report the same performance indicators for the remaining cycles and working fluids. The graphs are analogous to the ones reported in Fig.2 and Fig.3.

As aforementioned, from the analysis of Fig. B.7 and Fig. B.9, it is apparent how HTHPs perform much better than natural gas boilers, when the COP is the base of comparison. The spotted COP is higher than 1 in the entirety of the investigated working domain. Based on this, it can be argued that HTHPs will be more and more favourable substitutes for natural gas boilers, as renewable energy sources will increase their penetration in the electricity generation sector.

Figure B.7: Working domain and COP of R717/R718 cascade cycle. Isolines represent COP and dotted lines operation limits.

Figure B.8: Working domain and η_{ex} of R717/R718 cascade cycle. Isolines represent η_{ex} and dotted lines operation limits.

Figure B.9: Working domain and COP of different fluids and cycles. Isolines represent COP and dotted lines operation limits.

Figure B.10: Working domain and η_{ex} of different fluids and cycles. Isolines represent η_{ex} and dotted lines operation limits.

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