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Novel reactive refrigerants to make vapour compression heat pumps more efficient and suitable for industrial applications

Silvia Lasala[a,*], Julien Joliat[a], Olivier Herbinet[a], Luis Pinilla Monsalve[a], Jean-François Portha[a]

[a] Université de Lorraine, CNRS, LRGP, 54000 Nancy, France

Abstract

Thermodynamic cycles represent the backbone of our current energy systems: thermal power plants, which underpin the global electricity production; and heat pumps, which are expected to triple their installed capacity within 2030 in the residential sector and be deployed in the industrial one. Thermodynamic cycles are currently known to operate with chemically inert working fluids throughout the cycle. To substantially increase the performance of thermodynamic cycles, and to allow the more feasible use of heat pumps at industrial level, the original approach, which we are investigating through different research projects, proposes the use of working fluids being the site of a reversible dimerization reaction. These chemical reactions evolve across each unit operation, as a consequence of the impact that the imposed changing of temperature and pressure has on the reaction's chemical equilibrium. In a preliminary study based on the use of fictitious reactive fluids, we have demonstrated that using reactive rather than inert working fluids more than doubles the heat pump's coefficient of performance. This is mainly explained by the occurrence of an exothermic reaction in the hot heat exchanger (releasing more heat in the environment), and of an endothermic reaction in the compression process (requiring less energy to compress the fluid). After introducing the concept of "reactive refrigerant", this work validates those preliminary results, using a real reactive refrigerant. Focusing on industrial heat pump applications, we present the behaviour of dimerizing carboxylic acids (e.g., formic acid), which we have identified as a suitable working fluid for residential and industrial heat pumps.

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Keywords: Industrial and residential heat pumps; Vapour compression heat pump; Reactive working fluids.

1. Introduction

The market penetration and further development of heat pumps (HPs) play a key role in meeting sustainability goals, enabling the efficient electrification of both residential and industrial heating and cooling. Indeed, according to the IEA, in 2022, residential heating and space cooling contributed to about 14% of global CO_2 emissions, with 4.2 Gt and 0.9 Gt of CO_2 emissions for heating and cooling, respectively [1]; industry was responsible for the 25% of global CO_2 emissions, with 9.2 Gt of CO_2 emitted mainly due to heating needs [2].

Focusing on the industrial sector, the decarbonization of industry is thus not possible without efficient and reliable solutions for heat production at medium temperature, i.e. 150-300 °C. High-temperature heat pumps are already available on the market [3], with a maximum achievable temperature of about 80 °C – 240 °C, depending on the considered technology and application. Systems achieving highest temperatures (200 – 240 °C) are based on open water-based cycles, Mechanical Vapor Recompression (MVR), which are applied to evaporation, drying, distillation, and desalination. However, single-stage compression MVR HPs currently operate with a temperature lift limited to 30-50 °C, depending on the compression technology, mainly due to the much higher temperature increase of water during its compression. In these heat pumps, the production of high-temperature heat is thus limited to the availability of a high-temperature heat source.

* Corresponding author. *E-mail address:* silvia.lasala@univ-lorraine.fr

Another promising mechanical industrial heat pump technology is the vapour compression heat pump, which is based on a closed loop and processes specifically selected refrigerants. Commercial systems currently achieve 100 - 120 °C, while prototypes and less mature technologies predict the upcoming achievement of heat sink temperatures higher than 140 °C [4]. However, due to an inefficient compression process and to the thermodynamic behavior of refrigerants during compression: (i) the second law efficiency is low, which is about 30-50% [4], depending on the HP's complexity, (ii) the outlet temperature of the compressor, which is the hottest point of the machine, may approach the thermal stability limit of the refrigerant (whether organic) [5] and of the lubricating oil [6], or the mechanical resistance of the compressor, thus limiting the HP's temperature lift. The reliable achievement of higher temperatures at acceptable high efficiencies and costs is the challenge which is currently faced.

The most efficient commercial technology for residential heating is based on vapour compression heat pumps, and the European sustainability target is to triple the installed heat pumps capacity by 2030 [7]. However, the second law efficiency of current residential heat pumps is limited to the 30-40%.

For both industrial and residential heat pumps, their relatively low performance can be attributed to two main factors: the inefficiencies in the compression process and the use of working fluids whose thermodynamic transformations, though ideally reversible, do not align with the maximum efficiency identified in the thermodynamic limit. Hence, the ways for improvement currently undertaken tackle both the development of more efficient compressors and the selection of thermodynamically optimal fluids [8]. However, the selection of the working fluid is limited both by existing regulations [9] to fluids having low global warming potential (GWP) and zero ozone depletion potential (ODP), and by impositions on safety (toxicity and flammability) characteristics [10]. The selection of a working fluid is a matter of trade-off mainly because [11]: (i) most performant fluids are flammable and explosive; (ii) the acceptance of fluids characterized by non-zero but small ODP allows for an important reduction of GWP.

The increasing priority of complying with efficiency and environmental constraints is smothering previous limitations: (i) non-flammable and non-toxic refrigerants, now accepting "mildly flammable" gases; (ii) zero-ODP fluids [12,13], with the current acceptance of small ODP fluids. In the long term, fluorinated refrigerants will likely be banned in new and existing systems, and will be totally replaced by natural refrigerants (mainly, R290 (C_3H_8), R744 (CO_2), R717 (NH_3) and R718 (H_2O)). However, multiple concerns affect the use of these fluids too: safety (NH_3 is toxic and C_3H_8 is flammable), technical (e.g., low-pressures for R718, high-pressures for R744) and low-performance issues.

In order to contribute to overcome these problems, in the context of three ongoing research projects, we are developing a first-of-a-kind novel vapour compression heat pump based on the innovative use of working fluids that are no more inert media, but fluids being the place of a reversible dimerization reaction ($A_2 \rightleftarrows 2A$, with A_2 being a dimer and A the relative monomer). This reaction obeys chemical equilibrium thus evolving either in the forward or backward direction under the effect of temperature and pressure modification in each unit operation of the machine.

Preliminary calculations performed by Barakat et al. [14] are based on fictitious gaseous dimerizing fluids $A_2 \rightleftarrows 2A$. Although based on simple hypotheses, these calculations show how chemical reactions evolve along the thermodynamic transformations of the fluid in each heat pump unit (see Fig. 1).

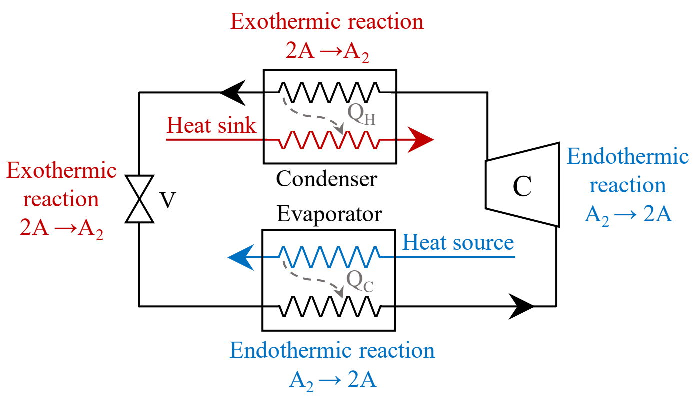

Figure 1. Evolution of chemically reactive working fluids along a vapour compression heat pump. Symbols: C: compressor; V: expansion valve; Q_H: thermal power delivered to the heat sink; Q_C: input thermal power to the heat pump, from an external source.

More specifically, Barakat et al. [14] show that compressing a reactive fluid in the vapor phase shifts the reaction toward its endothermic direction ($A_2 \rightarrow 2A$), thereby reducing the fluid's temperature rise during compression due to the cooling effect of the reaction. The compression work is thus reduced and the temperature of the fluid at the compressor outlet is drastically decreased with respect to inert fluid compression. Moreover, since the chemical reaction during the cooling in the condenser results to be exothermic ($2A \rightarrow A_2$), an additional amount of heat (named Q_H, in Fig. 1) is finally released to the environment, which constitutes the useful effect of the heat pump. These preliminary conclusions led us to clarify how reactive working fluids can substantially increase heat-pump efficiency - defined as the ratio of heat rejected to the environment to the compression work - and to quantify a second law efficiency exceeding 70%.

Based on these results, we identified the family of carboxylic acids as particularly promising among the available fluids capable of undergoing reversible reactions. As an example, the dimerization reaction of formic acid is shown in Fig. 2 and considered in this study. Formic acid, the simplest carboxylic acid, is a versatile compound with a variety of industrial, agricultural, and laboratory uses due to its acidic properties and reactivity. However, differently from more complex carboxylic acids (i.e., acetic acid), formic acid exhibits toxic properties which must be considered. Nonetheless, considering that R717 (ammonia) [15], known for its significantly higher acute toxicity than formic acid [16], is already used as one of the most promising natural refrigerants in industrial heat pump systems, we aim to explore the potential advantages of formic acid as an alternative natural refrigerant. It is also important to note that its ODP and GWP are both zero.

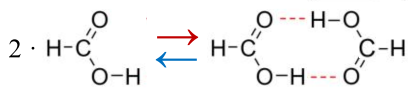

Figure 2. Reversible reaction occurring in pure formic acid.

This paper presents preliminary calculations for vapour compression heat pumps operating with formic acid, considering both residential and industrial applications, with the aim to validate our preliminary results obtained on fictitious fluids and to present a comparative analysis -although simple- with respect to inert refrigerants. Section 2 presents the methodology, while section 3 introduces the results and their analysis.

2. Methodology

This section outlines the methodology applied in this work to perform thermodynamic cycle calculations, with a focus on residential and industrial heat pump applications.

2.1. Thermodynamic calculations

Thermodynamic cycle calculations were carried out assuming a simple vapour compression heat pump cycle, illustrated in Fig. 3. The calculation procedure is summarized below and is based on the input data reported in Table 1. The different working fluids considered, together with the corresponding thermodynamic models, are listed in Table 2. A common simplifying assumption adopted throughout the analysis is that the heat source and the heat sink operate at constant temperatures, denoted as T_C and T_H, respectively. The evaporation and condensation temperature of the working fluid in the evaporator and in the condenser are reported in Table 1 as T_{eva} and T_{cond}.

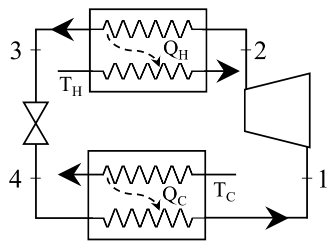

Figure 3. Simple heat pump cycle under investigation.

Table 1. Assumptions for residential heat pump calculations.

Residential heat pump – case study A				Industrial heat pump – case study B			
Heat source temperature, T_C (K)	293.15	Subcooling temperature difference, ΔT_{SC}, i.e. the difference T_{cond} - T_3	5	T_C (K)	353.15	ΔT_{SC}	5
Heat sink temperature, T_H (K)	338.15	Superheating temperature difference, ΔT_{SH}, i.e. the difference T_1 - T_{eva}	5	T_H (K)	413.15	ΔT_{SH}	13
Compressor isentropic efficiency, $\eta_{c,is}$	0.75	Pinch point in the evaporator and condenser, ΔT_{PP}	5	$\eta_{c,is}$	0.75	ΔT_{PP}	5

Table 2. Working fluids analyzed in the two case studies, and the applied thermodynamic model. Calculations for formic acid were carried out using an in-house Fortran code, while calculations for the other fluids were performed using Refprop [17].

Residential heat pump – case study A		Industrial heat pump – case study B	
Fluid	*Thermodynamic model*	*Fluid*	*Thermodynamic model*
Formic acid	Wasik et al, 2025 [18]	Formic acid	Wasik et al, 2025 [18]
R290 (propane)	Lemmon et al, 2009 [19]	R1233zd(E)	Mondéjar et al., 2015 [20]
		R718 (water)	Wagner and Pruß, 2002 [21]

With reference to the architecture shown in Fig. 3 and to the thermodynamic state points 1-4, the corresponding properties (P: pressure, T: temperature, h: specific enthalpy, *s*: specific entropy, *v*: specific volume) were calculated as described below. The subscript “sat”, “sat-l” and “sat-v” denote saturation conditions, and saturated liquid and vapour states, respectively. The determination of properties as shown in Equations (1)-(9) allows the calculation of all the other properties.

- Point 1: compressor inlet (superheated vapour)

$$T_1 = T_C - \Delta T_{\mathrm{PP}} \tag{1}$$

$$P_1 = P_{\mathrm{sat}}(T_C - \Delta T_{\mathrm{PP}} - \Delta T_{\mathrm{SH}}) \tag{2}$$

- Point 3: valve inlet (subcooled liquid)

$$T_3 = T_H + \Delta T_{\mathrm{PP}} \tag{3}$$

$$P_3 = P_{\mathrm{sat}}(T_H + \Delta T_{\mathrm{PP}} + \Delta T_{\mathrm{SC}}) \tag{4}$$

- Point 2: compressor outlet (superheated vapour)

$$P_2 = P_3 \tag{5}$$

$$h_2 = h_1 + \frac{h_{2s}(P_2, s_1) - h_1}{\eta_{c,\mathrm{is}}} \tag{6}$$

- Point 4: valve outlet (two-phase region, with quality τ)

$$P_4 = P_1 \tag{7}$$

$$h_4 = h_3 \tag{8}$$

$$\tau_4 = \frac{h_4 - h_{4,\mathrm{sat-l}}}{h_{4,\mathrm{sat-v}} - h_{4,\mathrm{sat-l}}} \tag{9}$$

2.2. Performance and preliminary-design assessment

The coefficient of performance of the heat pump is calculated as:

$$COP = \frac{Q_H}{W_c} = \frac{h_2 - h_3}{h_2 - h_1} \quad (10)$$

where W_c is the compression work; and the theoretical limit, which in this specific case where constant-temperature heat sink and heat source have been considered, coincides with the Carnot efficiency:

$$COP_{max} = \left(1 - \frac{T_C}{T_H}\right)^{-1} \quad (11)$$

The compressor size is evaluated by calculating the volumetric ratio, defined as the ratio of specific volumes at the compressor inlet and outlet: v_1/v_2.

3. Results and Discussion

3.1. Residential heat pumps calculations

The results obtained applying the methodology outlined in section 2, specifically for the residential case study, are reported in Table 3 and represented in Fig. 4 and Fig. 5. The analysis of these results confirms that using formic acid significantly increases cycle efficiency compared to propane, yielding a 27% improvement in COP. Fig. 4 further illustrates that the cycle operating with formic acid has a shape closer to that of the Carnot cycle, compared to propane. This improvement is mainly attributed to the entropic and thermal effects of the chemical reaction on the expansion and compression processes. Indeed, the reaction is exothermic during expansion and endothermic during compression. The exothermic reaction during expansion contributes to a negative entropy change in the expanding reactive system; the endothermic cooling effect during compression, indicated by the increase in monomer molar fraction from point 1 to point 2 in Table 3, leads to a reduction in entropy.

Furthermore, the endothermic behavior observed during the compression of formic acid results in a significant decrease in fluid temperature at the compressor outlet, which represents a potential advantage for high-temperature heat pump applications.

Table 3. Residential heat pump results.

	Formic acid			Propane – R290	
	T (K)	P (MPa)	Monomer molar fraction, x_A (-)	T (K)	P (MPa)
1	288.15	0.001(*)	0.123	288.15	0.64
2	355.67	0.022(*)	0.261	367.13	2.85
3	343.15	0.022(*)	0.001	343.15	2.85
4	283.15	0.001(*)	Two-phase	283.15	0.64
$h_2 - h_3$ (kJ/kg)	503.1			289.0	
$h_2 - h_1$ (kJ/kg)	128.2			94.8	
COP/COP_{max}	**52%**			**41%**	
v_1/v_2	16.6			4.4	

(*) The thermodynamic model of formic acid underestimates saturation pressure. For example, DIPPR's [22] empirical correlations show that the saturation pressure of formic acid is 0.003 MPa at T_{eva} = 283.15 K and 0.044 MPa at T_{cond} = 348.15 K.

However, formic acid exhibits a much higher compression volumetric ratio and a low minimum pressure, making its use challenging in low-temperature residential applications. To address this issue, one solution which we propose and is currently investigated consists in the use of blends of formic acid and CO_2.

In addition, Fig. 5 shows that the current superheating degree, ΔT_{SH} = 5 °C, is insufficient for formic acid as it leads to its wet compression. In this case, a minimum ΔT_{SH} of 12 °C is required, to avoid this issue, resulting in a second low efficiency (COP/COP_{max}) equal to 47%, or lower for higher values of ΔT_{SH}.

While the results from other HFO refrigerants are not presented here, they generally lead to significantly lower efficiencies compared to propane.

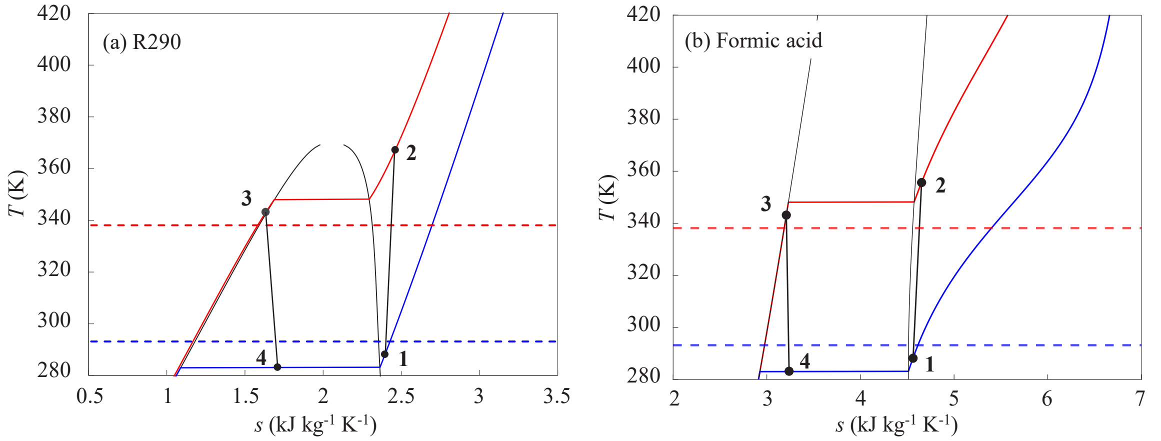

Figure 4. Temperature-entropy diagram of two heat pumps operating with propane (Fig. (a)) and with formic acid (Fig. (b)), relative to the case study A.

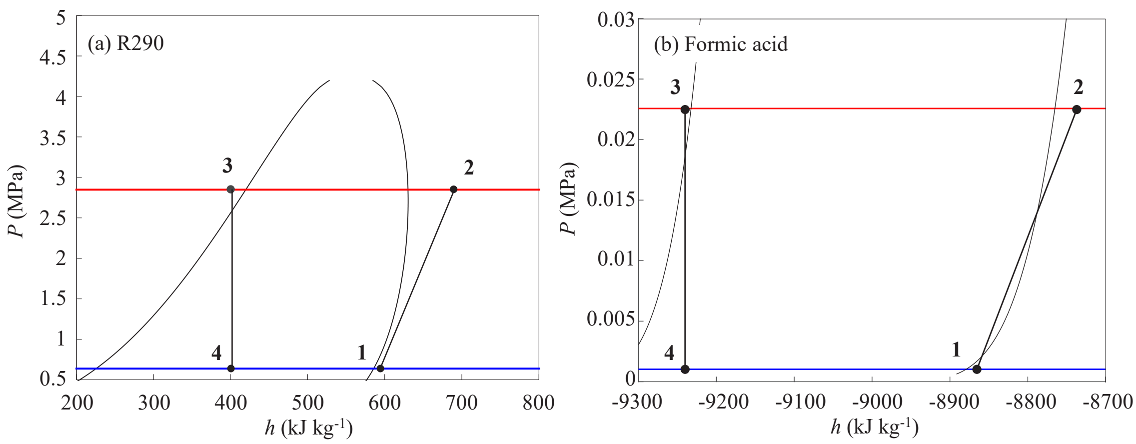

Figure 5. Pressure-enthalpy diagram of two heat pumps operating with R290-propane (Fig. (a)) and with formic acid (Fig. (b)), relative to the case study A.

3.2. Industrial heat pumps calculations

This section reports the numerical results obtained for the industrial heat pump applications considered in this study. In particular, Table 4, Fig. 6 and Fig. 7 confirm what was observed in section 3.1: the use of formic acid allows the notable increase of the heat pump efficiency, compared to both R1233zd(E) and R718 (water). Although R718 yields high efficiencies, it is important to note that operation with formic acid significantly reduces the maximum cycle temperature, from 829.60 K (R718) to 430.10 K (formic acid). This result is highly promising, as formic acid not only improves efficiency compared to one of the most efficient refrigerants (water), but also facilitates the practical implementation of high-temperature heat pumps by substantially lowering the cycle's peak temperature.

As mentioned in the Introduction, both are classified as "toxic" substances, making it important to compare our results for formic acid with those for R717. However, R717 is not suitable for constant-temperature heat sink industrial applications, as the system's maximum pressure reaches supercritical levels, leading to significant entropy losses during heat exchange and a substantial decrease in efficiency.

Finally, Fig. 7 shows that increasing the selected superheating degree is necessary, as it helps to prevent wet compression for both R1233zd(E) and formic acid. Notably, in this case study, the compression of R1233zd(E) is wetter than that of formic acid.

Table 4. Industrial heat pump results

	Formic acid			R1233zd(E)		R718	
	T (K)	P (MPa)	Monomer molar fraction, x_A	T (K)	P (MPa)	T (K)	P (MPa)
1	348.15	0.013	0.271	348.15	0.41	348.15	0.02
2	**430.10**	0.24	0.001	**433.9**	2.74	**829.60**	0.48
3	418.15	0.24	0.401	418.15	2.74	418.15	0.48
4	335.15	0.013	Two-phase	335.15	0.41	335.15	0.02
$h_2 - h_3$ (kJ/kg)		528.88		109.7		2996.2	
$h_2 - h_1$ (kJ/kg)		155.12		47.2		969.2	
$COP/COP_{\max}$		**50%**		**34%**		**45%**	
v_1/v_2		14.2		7.7		9.1	

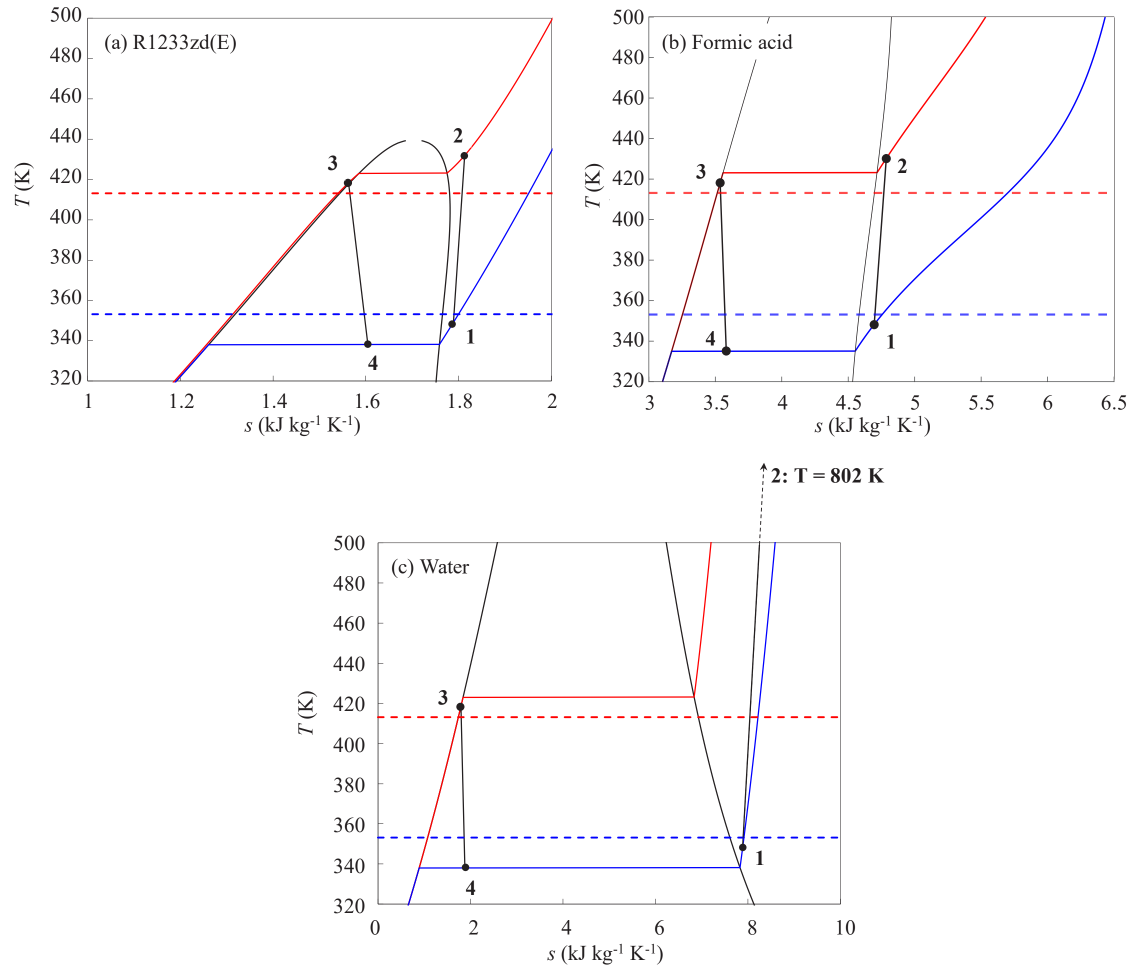

Figure 6. Temperature-entropy diagram of two heat pumps operating with R1233zd(E) (Fig. (a)), formic acid (Fig. (b)), and water, (Fig. (c)), relative to the case study B.

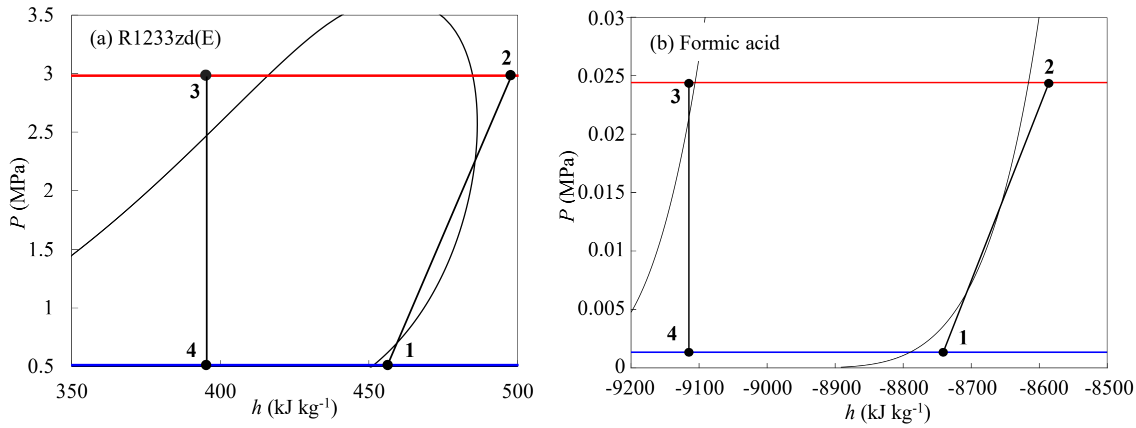

Figure 7. Pressure-enthalpy diagram of two heat pumps operating with R1233zd(E) (Fig. (a)), formic acid (Fig. (b)), relative to the case study B.

4. Conclusions

This study has demonstrated that the preliminary results obtained by Barakat et al. [14] considering gaseous fictitious reactive working fluids are confirmed by the use of real working fluids, with higher efficiencies and reduced outlet temperatures of the compression process. For residential heat pump applications, the use of pure formic acid is strongly limited by its low operating pressure and high volumetric compression ratio. However, this drawback may be overcome by adding inert CO_2 to the reactive fluid. In contrast, industrial heat pump applications using formic acid appear highly promising, as this fluid not only increases cycle performance but also reduces the maximum cycle temperature or, alternatively, enables operation at higher heat sink temperatures. In future work, our group is investigating an alternative non-toxic carboxylic acid, acetic acid, which is a dimerizing fluid with comparable efficiency to formic acid, but with enhanced safety properties. Additionally, we are proposing a more extensive study with the aim of expanding our preliminary findings on constant-temperature heat sink and source systems, as well as simple heat pump configurations. This extended research will explore the potential for variable temperature systems and more optimized configurations to improve overall performance and efficiency. Finally, we are evaluating the compatibility of formic acid and acetic acid with potential materials, and developing a high-temperature prototype operating with formic acid to validate theoretical calculations.

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