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New HFC/HFO Blends as Refrigerants for the Vapor-Compression Refrigeration System (VCRS)

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Abstract: In this work, which is related to the current European Parliament Regulation on restrictions affecting refrigeration, four new three-component refrigerants have been proposed; all were created using low Global Warming Potential(GWP) synthetic and natural refrigerants. The considered mixtures consisted of R32, R41, R161, R152a, R1234ze (E), R1234yf, R1243zf, and RE170. These mixtures were theoretically tested with a 10% step in mass fraction using a triangular design. The analysis covered two theoretical cooling cycles at evaporating temperatures of 0 and −30 °C, and a 30 °C constant condensing temperature. The final stage of the work was the determination of the best mixture compositions by thermodynamic and operational parameters. R1234yf–R152a–RE170 with a weight share of 0.1/0.5/0.4 was determined to be the optimal mixture for potentially replacing the existing refrigerants.

Keywords: hydrofluoroolefins; hydrofluorocarbons; refrigerants; low GWP

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1. Introduction

It was only a few years ago that most refrigeration appliances used refrigerants containing chlorine; these had a destructive effect on the ozone layer. The use of chlorofluorocarbons (CFCs) and hydrochlorofluorocarbons (HCFCs), which actually possess very good thermodynamic properties, is now prohibited. Apart from the destructive impact on the ozone layer, refrigerants also affect the natural environment by contributing to the greenhouse effect. Bearing in mind the changes taking place in the environment, the European Parliament approved Regulation No. 517/2014 [1] on fluorinated greenhouse gases, which significantly limits the possibility of using the refrigerants currently available on the market, especially those with a high Global Warming Potential (GWP). The purpose of legal regulations is to limit global climate change in accordance with the Paris Agreements and to prevent the adverse effects of this change. In response, greater use of natural refrigerants and hydrocarbon derivatives is necessary, as they have a lower environmental impact. The extensive use of carbon dioxide or hydrocarbons, as well as hydrofluoroolefins (HFOs) and hydrofluorocarbons (HFCs) with a GWP not exceeding 150, is a key challenge in transforming the refrigerant market.

HFO substances quickly decompose in the lower atmosphere due to the double carbon bond in the molecule; this guarantees very low GWP, but also results in flammability [2]. The most popular refrigerant in this group is R1234yf, which is used as a replacement for R134a in automotive air-conditioning systems. The second leading representative of the new generation of refrigerants is R1234ze(E), which has zero Ozone Depletion Potential (ODP) and a low GWP. It has been proposed as a replacement for R32, R410A, and R134a. However, R1234ze(E) is not an ideal substitute for the phased-out refrigerants, as it has a lower specific cooling capacity [3], a lower coefficient of performance (COP) [4], and a lower heat transfer coefficient [5,6]. According to EN 378-1 [7], both HFO refrigerants have a very low level of GWP, at only 4 and 7, respectively. In addition to compressor cycles, HFOs are also considered for operation in other types of refrigeration equipment, such as ejector devices [8,9] or combined systems [10,11].

Combining HFO refrigerants with HFCs has become popular as a way to improve their properties, especially low cooling capacity, due to low latent heat of vaporization and specific refrigerating effect. In scientific research, it has often been combined with R32, a high-pressure refrigerant with satisfactory thermodynamic properties, high latent heat, and a relatively low GWP of 675 [12]. The R32–R1234ze(E) mixture was tested in the proportions of 0.5/0.5 and 0.2/0.8 to determine the heat transfer coefficient obtained in a horizontal tube [13] and the isobaric heat capacity (a mole fraction of R32 from 0.226 to 0.946) [2]. Another proposed combination of HFO/HFC refrigerants is the R32–R1234yf mixture, at different weight proportions, tested to determine a truncated virial equation of state [14].

Akasaka [15] presented models of thermodynamic properties for the mixtures R32–R1234ze(E) and R32/R1234yf, the uncertainties of which are 1% for the bubble point pressure and 0.25% for the liquid density. The author states that while the models show slightly greater uncertainties than the typical Helmholtz energy state equations for pure fluids, they are applicable to preliminary analysis of refrigeration equipment and heat pumps. R1234ze(E), like R1234yf, was also combined with R134a to determine the vapor–liquid balance of the mixtures [2] and to test the possibility of replacing pure R134a with new substances in home refrigerators.

Aprea et al. [16] proposed a mixture of R1234yf–R134a with a 10% HFC weight share, defined by the GWP limit of 150. This mixture achieved a 17% lower life-cycle climate performance (LCCP) index than pure R134a.

Other experimental results have shown that for R134a–R1234ze(E) mixture, despite the larger required refrigerant charge, a shorter daily operating time was obtained while maintaining the set temperature level, which resulted in a reduction of energy consumption by 14% compared to R134a [17]. Another combination is the R152a–R1234ze(E) mixture, for which the equilibrium (vapor + liquid) was tested; this is one of the most important parameters used to calculate and optimize cooling cycle efficiency, as well as the organic Rankine cycle and other chemical processes [18].

Ternary mixtures are another way to combine the new generation of refrigerants with HFCs. Various mixtures containing HFO are currently commercially available. These include, among others:

- R447A, which has a favorable GWP of 572, and, in terms of heat exchange, can be treated as a potential alternative to R410A (GWP = 2088) [19];
- R457A and R459B, with a low GWP (less than 150), which can provide a smaller refrigerant charge and improve energy efficiency in relation to R404A [20];

It is not only synthetic refrigerants that are combined together. Natural substances, such as carbon dioxide and hydrocarbons, are also used to create refrigeration blends. Kondou et al. [21] examined the heat transfer coefficient during evaporation and condensation of the ternary mixture R32–R1234yf–R744 with different weight shares (0.29/0.62/0.09, 0.43/0.53/0.04 and 0.06/0.34/0.6) and compared them with the binary R32–R1234ze(E) mixture (with weight shares of 0.4/0.6 and 0.73/0.27). Saengsikhiao et al. [22] considered using R463A as a replacement for R404A. It has been shown that R463A may operate at a higher ambient temperature, achieving higher COP in a low-temperature application, and lower GWP compared to R404A.

A further example of combining HFCs with HFOs and natural substances is R134a–R1234yf–R600a. Isobutane has good latent heat and excellent thermodynamic performance but is highly flammable and explosive. R1234yf is also slightly flammable, so non-flammable R134a has been added to reduce this flammability. The vapor–liquid equilibrium data were collected for the mixture [23]. Research has also been carried out on implementing a model for forecasting vapor + liquid balance data in a binary mixture of CO2, HFC, or HC, with two low GWP refrigerants (R1234yf and R1234ze (E)) [24]. Another flammable natural refrigerant that has attracted interest in recent years is dimethyl ether

(DME or RE170), which is widely used in the chemical industry, medicine, etc., and is therefore considered an alternative refrigerant. It has a low boiling point, high latent heat, and is non-toxic, slightly corrosive, and more environmentally friendly, because its ODP and GWP are 0 and 1, respectively. The study investigated the flammability of the RE170–R1234yf–R134a mixture (with weight fractions of 0.1/0.8/0.1) [25].

There are many possible refrigerant combinations; HFO is usually combined with well-known substances from the HFC group due to their excellent environmental parameters. As shown above, it is mainly R1234yf and R1234ze(E) that are selected for analysis. Others are not as popular for a variety of reasons, including a normal boiling points above 0 °C or even non-zero ODP. In this study, along with R1234ze(E) and R1234yf, it was decided to use one of the rarer hydrofluoroolefins as a substrate for the newly defined mixture, namely R1243zf (a GWP of approx. 1, an ODP of 0).

Finding the perfect refrigerant, which has favorable thermodynamic properties and can work efficiently in the refrigeration cycle, while also being cheap, easily accessible, and safe for the environment, is a very difficult task, if only because of the numerous legal and technical changes taking place in the refrigeration sector. There are many possibilities for combining and mixing pure refrigerants together to achieve the desired properties and to have a negligible effect on the atmosphere. In this work, four ternary mixtures were proposed and theoretically tested for their cooling capacities in theoretical cooling circuits to determine the optimal composition for the new blends. Significant effort has gone into developing and testing different refrigerant blends. However, there are still many unresolved issues and opportunities that need to be investigated. In light of the state of the art in this field, combining and mixing pure refrigerants together to achieve the desired properties while having a negligible effect on the atmosphere is still an open problem and a meaningful endeavor. This combining and mixing took several aspects into account, such as the use of the rarer hydrofluoroolefins (instead of commonly used refrigerants) as a substrate for the newly defined mixture, the triangular design, which involves the use of three refrigerant components, and the thermodynamic and operational parameters. Such an approach is hardly visible in the literature, which confirms the novelty of the presented research. Its realization required the application of new approaches that employed advanced analysis of the four ternary mixtures, along with their experimental verification. This allowed for the advanced testing of their cooling capacities in theoretical cooling circuits to determine the optimal composition for the new blends. It should also be highlighted that the weight fraction of the presented blends was determined (and not fixed, as a frequent goal of scientific articles) to find the optimal mixture for potentially replacing the existing refrigerants. Consequently, this article contributes to the state of the art in the new HFC/HFO blend selection by developing appropriate methods that can be feasibly implemented.

2. Materials and Methods

2.1. Methodology and Prerequisites for the Mixture Components

The selection of the optimal zeotropic or azeotropic mixture components was based on a triangular-basis plan, which involves the use of three refrigerant components. The sum of the weight shares of the selected substances must always be equal to 1 (100%), and the single share must be zero or positive. Thus, each blend will be defined by three independent variables. The shares of individual components (pure refrigerants) in the mixture change from 0 to 1 (from 0% to 100%) in steps of 0.1 (10%). The basic properties and operating parameters of each mixture, depending on their composition, are presented on ternary charts. In the corners of the graph are the pure substances selected as mixture components. The edges of the triangle depict binary mixtures, while the ternary mixtures are inside the triangle.

The basic criteria for selecting the constituent substances were the environmental parameters of a zero ODP and a low GWP. A condition was also set regarding the normal boiling point, the value of which had to be at least −25 °C to avoid under-pressure conditions. Given the final evaporation temperature levels obtained, all the proposed mixtures were assigned to the implementation of air-conditioning or freezing cycles and compared to the corresponding reference refrigerants. An additional criterion was the small distribution of individual components in already existing and commercially available mixtures. It was decided that the newly created working fluid should contain at least one alternative refrigerant (natural or from the HFO group), the rest being HFCs. The criteria were ranked according to their weight and checked in accordance with the diagram presented in Figure 1. The main initial consideration when creating mixtures and selecting their components was to limit the GWP value to 750 for air-conditioning systems (or 150 if possible) and 150 for low-temperature refrigeration devices. Obtaining low temperatures for the assumed operating conditions of the condenser may be associated with obtaining correspondingly higher pressure ratios in the system (compression ratio). In this case, the operation of the system may be disadvantageous due to a number of phenomena, such as low volumetric efficiency of the compressor and decrease in refrigerant mass flow due to an increase in specific vapor volume, increase in the compressor power consumption, high discharge temperature, and deterioration of lubricating properties of oils. In order to avoid the above-mentioned problems, it was assumed that the maximum value of compression ratio of the analyzed mixtures should not exceed 8.0.

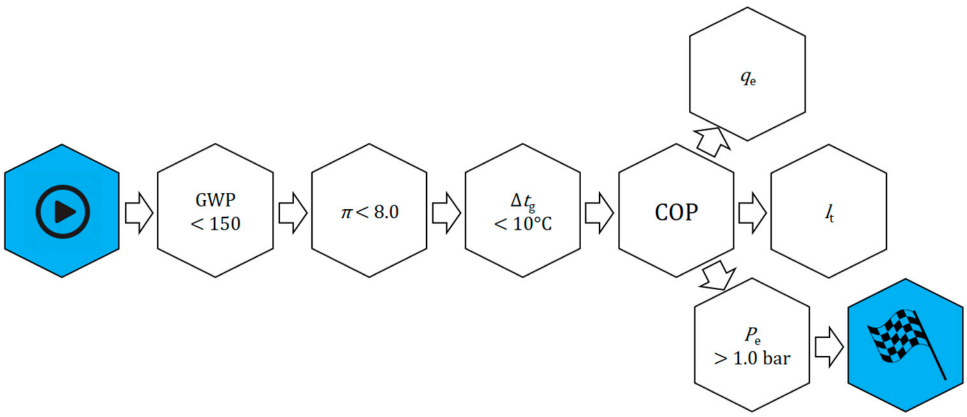

Figure 1. Selection criteria flow chart.

An equally important issue in the operation of real cooling cycles is the temperature glide, which may cause evaporator malfunctions. Components with extremely different vapour pressures can cause frosting to the initial sections of the evaporator due to the evaporation of low-boiling components. On the other hand, components with a high boiling point may not completely evaporate, which can lead to fractionation of the refrigerant inside the system and change its operating parameters. In case of extreme temperature glides, it is necessary to increase the vapour superheat set point to prevent the compressor from sucking in liquid refrigerant. In this analysis, the temperature glide of the mixtures was limited to 10 K. After meeting the initial criteria, the working parameters of the mixtures were analyzed.

Four mixture types have been presented and tested in this paper:

- R32–R41–R1234ze(E)—a mixture combining R32 that is currently gaining popularity, the R41, which has a low normal boiling point and a very low GWP, and R1234ze(E), the second most-studied HFO refrigerant;
- R32–R161–R1234ze(E)—a mixture similar to the previous one. However, the second component has been changed to R161, which has a slightly higher boiling point at 1 bar pressure but an almost eight-times lower GWP;
- R1234yf–R152a–RE170—this mixture combines the most popular refrigerant from the HFO group—R1234yf, with R152a often used in HFC/HFO mixtures, and a natural

substance RE170, which has been increasingly used in refrigeration mixtures, since the early 2000s;

- R1243zf–R152a–RE170—a mixture related to the previous one. However, the HFO group has been replaced by another, R1243zf, which thermodynamic properties are the most similar to widely used R134a, and which has a higher heat of vaporization than R1234yf.

All have been included in the European Patent (EP3309233A1) [26], which relates to compositions for use in refrigeration and air-conditioning. However, we would like to highlight that this does not negate the possibility of studying them from the scientific standpoint, with no intention of using them commercially. Basic information on the pure components used is presented in Table 1.

Table 1. Basic properties of pure components [27].

Refrigerant	Unit	R1234yf	R1234ze(E)	R1243zf	R152a	R161	R32	R41	RE170
Name	-	2,3,3,3-Tetrafluoroprop-1-ene	trans-1,3,3,3-Tetrafluoroprop-1-ene	3,3,3-Trifluoropropene	1,1-Difluoroethane	Fluoroethane	Difluoro-methane	Fluoro-methane	Dimethyl ether
CAS No.	-	754-12-1	29118-24-9	677-21-4	75-37-6	353-36-6	75-10-5	593-53-3	115-10-6
GWP [1]	-	4	7	1	124	12	675	92	1
Critical temperature	°C	94.7	109.4	103.8	113.3	102.1	78.11	44.13	127.2
Critical pressure	bar	33.82	36.35	35.18	45.17	50.46	57.82	58.97	53.37
Normal boiling point	°C	−29.48	−18.97	−25.42	−24.02	−37.54	−51.65	−78.31	−24.78
Molar mass	g/mol	114.0	114.0	96.05	66.05	48.06	52.02	34.03	46.07
Flamability class [7]	-	2 L	2 L	2 L	2	3 *	2 L	3 *	3

* flammable compounds, not included in EN 378; determined based on NFPA 704.

2.2. Theoretical Refrigeration Cycle—Assumptions

Two theoretical single-stage refrigeration cycles were determined for comparing the mixtures. In the first cycle, the evaporating temperature (t_e) was set at 0 °C, with the condensing temperature (t_c) equal to 30 °C. This cycle corresponds to the work done by air-conditioning systems. The evaporating temperature in the second cycle was set to −30 °C, with the condensing temperature unchanged. This allowed to examine the behavior of mixtures in low-temperature systems. In both cases, it was assumed that the compression isentropic efficiency was $\eta = 0.7$. For the purposes of theoretical analysis, it was assumed that the liquid subcooling in the condenser and the vapour superheating in the evaporator were equal to zero, as presented in Figure 2. Pressure drops in the heat exchangers and in the pipeline flow were also omitted.

The theoretical analysis of comparative cycles allowed us to determine the basic operating parameters of the proposed mixtures. With the help of enthalpy at the characteristic operating points, the specific cooling capacity (q_e), the specific work of the cycle (l_t), the volumetric cooling capacity (defined as $q_e/v_{suction}$), and the COP of each of the proposed mixtures were defined in terms of the assumed variability in the weight shares of the individual components. On the basis of the evaporation and condensation pressures obtained, the compression ratio and the temperature glide were determined. The specific refrigeration system parameters were determined for both the high-temperature and the low-temperature cycles. In addition to the operating parameters, the basic properties were also determined, such as the GWP, the critical point temperature and pressure, the normal boiling point (1 bar), and the molar mass.

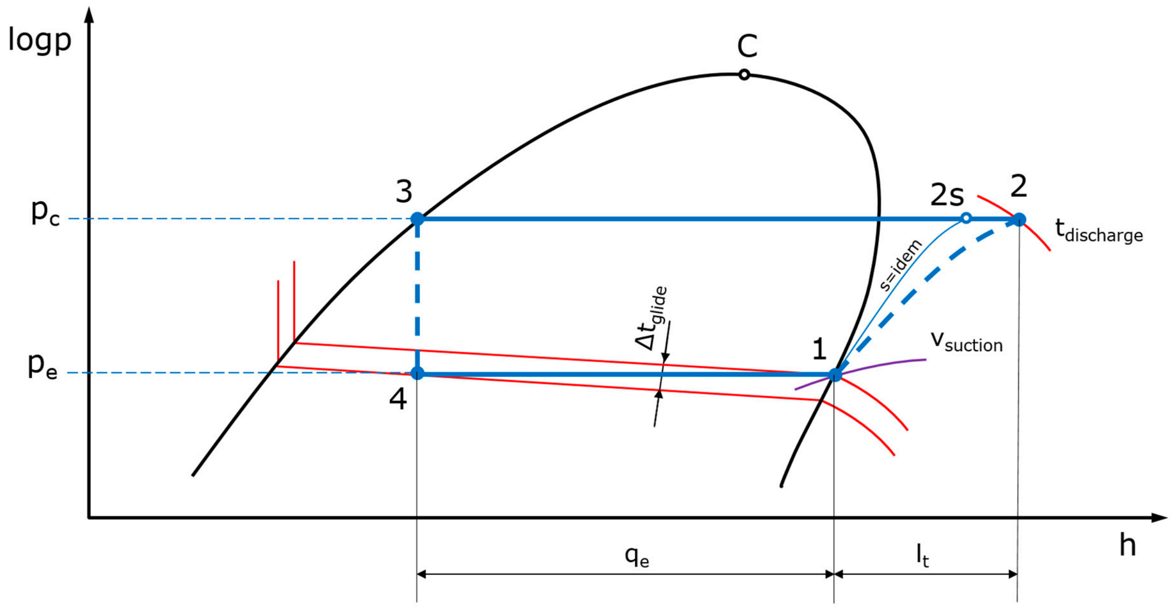

Figure 2. Single stage refrigeration cycle analyzed.

The theoretical analysis of the newly defined zeotropic or azeotropic mixtures was carried out using the REFPROP 10.0 program [27], from which the thermodynamic and transport properties of the fluids relevant for the study and their mixtures were taken, such as the critical temperature and pressure, the normal boiling point, and the molar mass. The thermodynamic properties of the mixtures were determined by employing a model that applies mixing rules to the Helmholtz energy of the mixture components, along with a departure function to account for the departure from ideal mixing. The same "XR0" mixing rule was used in REFPROP for all analyzed mixtures. It should be emphasized that in order to determine the properties of mixtures, it is necessary to have properties for binary subsystems, and among the mixtures under consideration, not all subsystems were tested experimentally. Therefore, it should be borne in mind that despite the generally good accuracy of determining thermal-flow properties, some of the presented values may differ from the actual ones, which requires confirmation in the field of further experimental studies.

The newly defined zeotropic or azeotropic mixtures will be compared (at work) to the refrigerants being withdrawn from cooling equipment as a result of the European Parliament regulation. It is important to find the parameters that will be favored by the proposed compositions. Taking into account the scope of application, the benchmark refrigerants for low temperature circuits will be R404A and R507A, and for high temperature circuits, R410A, R134a, R32, and R429A. The R429A mixture was used as a benchmark due to its similar composition to the two newly defined blends tested. This refrigerant consists of RE170, R152a, and R600a in the corresponding weight proportions of 60%, 10%, and 30%.

3. Results

Presented paper analyzes the results, which aimed to define the optimal composition of a new refrigerant mixture. Four lists of refrigerants were considered.

3.1. R32–R41–R1234ze(E)

The first of the mixtures contains the now popular R32 refrigerant, which is treated as a substitute for the R410A refrigerant and has very good thermodynamic properties, as well as a relatively low GWP; nevertheless, it is in the A2L flammability class. The second HFC component is R41, which has a very low global warming potential. The representative from the HFO group is the well-known R1234ze(E), which is freely available on the market.

Analyzing the composition of the discussed mixture, it is most likely to belong to the A2L flammability class.

The paper presents the value gradients determined for the R32–R41–R1234ze(E) mixture. This started from the basic values: the GWP and the normal boiling point, presented in Figure 3.

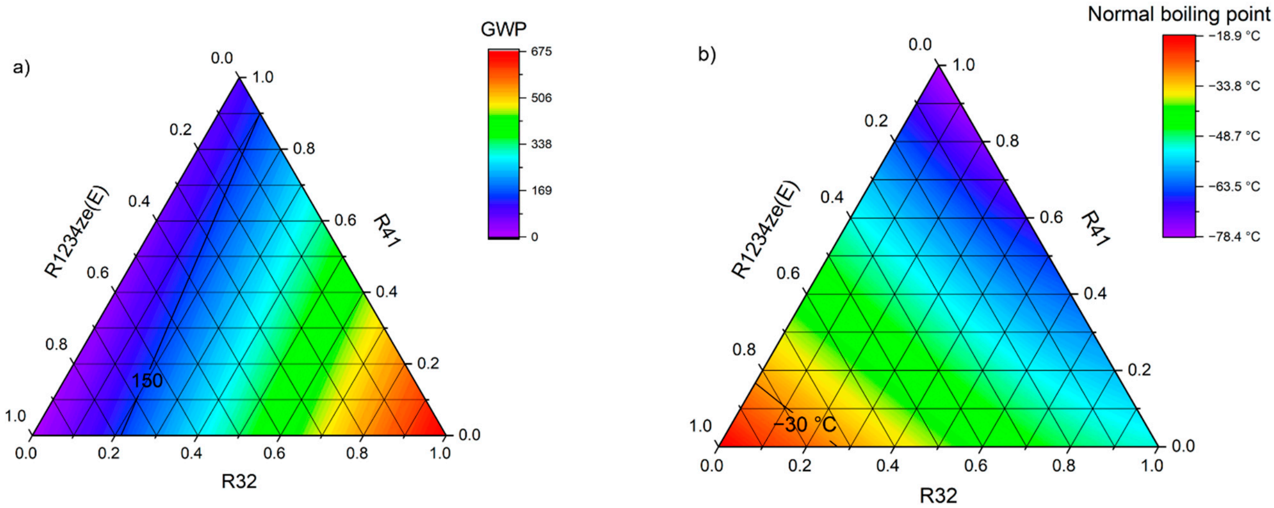

Figure 3. Summary of the basic properties for the R32–R41–R1234ze(E) mixture: (**a**) GWP and (**b**) normal boiling point.

The mixture, regardless of the shares of individual components, has a GWP below 750, as none of the components exceed this threshold. However, in order to meet the criterion for low-temperature cycles, it is necessary to limit the share of R32 to a maximum of 20%. Thus, only the left side of the triangle shown in Figure 3a is potentially usable. Attention should also be paid to the lower left corner of the normal boiling point diagram (Figure 3b). The boiling points at normal pressure in this area are higher than −30 °C, which means that the system will work under pressure. In the case of failure and leakage, it may lead to the appearance of air inside the system, which in the case of flammable refrigerants may pose a real risk of ignition of the installation due to the possible exceeding of the lower flammability limit of the mixture. Moreover, analyzing Figure 4b, it can be seen that the area at the left edge of the triangle is almost entirely covered by high temperature glides, reaching even over 25 K. In practice, only the mixtures located in the upper corner and in the right corner should be considered as suitable for low-temperature cycles.

For air conditioning cycles, the GWP limit according to [1] is 750, which is not exceeded at any point in the graph. The only limit here is the temperature glide, the value of which should not exceed the assumed threshold of 10 K (compare Figure 4a). Figure 5a,b show that it is impossible to select the mixture in such a way as to ensure both the possible high COP and the high volumetric cooling capacity of the mixture. Optimizing the mixture for high volumetric cooling capacity will provide benefits in terms of reducing the compressor (piston) displacement, which will translate into the number of pistons, compressor weight, dimensions, and price. Optimization for the possible high COP will reduce operating costs due to the minimization of the energy needed to compress the refrigerant. Regardless of the chosen direction of optimization, only a few mixtures are possible to use. Taking into account that GWP should be limited to 150, only two ternary mixture are available with mass fractions 0.1/0.8/0.1 and 0.1/0.7/0.2. Unfortunately, both of these mixtures contain a high proportion of R41, a refrigerant that is currently hardly available on the market due to the lack of widespread use in refrigeration. At the same time, it can be seen that there is no other R41-free mixture that meets these criteria. Extending the analysis to the threshold of 750 adopted by Regulation (EU) 517/2014, it is possible to select two binary mixtures R32–R1234ze(E) with the weight shares of 0.9/0.1 and 0.8/0.2. Of the two mixtures, the first is the more promising, as it is characterized by an increase in volumetric cooling capacity by 8%, with an almost identical COP. It should be remembered that the GWP threshold of 750 applies only to single split air-conditioning systems containing less than 3 kg of F-gases and only in this group of devices can the mixture proposed above be used.

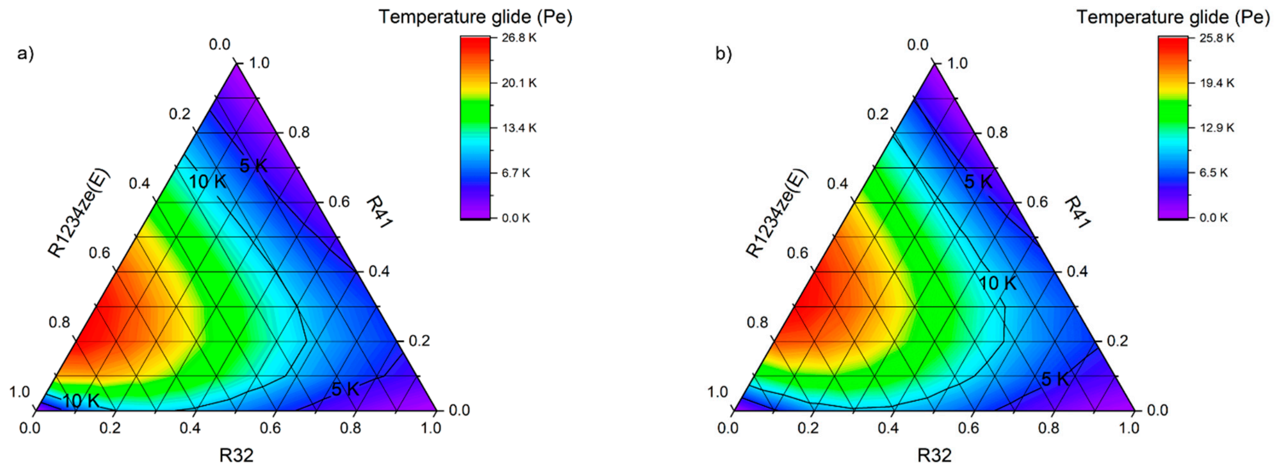

Figure 4. Temperature glide at the evaporation pressure for cooling cycles with R32–R41–R1234ze(E) mixture as refrigerant: (**a**) AC system (t_e/t_c = 0/30 °C); (**b**) low-temperature system (t_e/t_c = −30/30 °C).

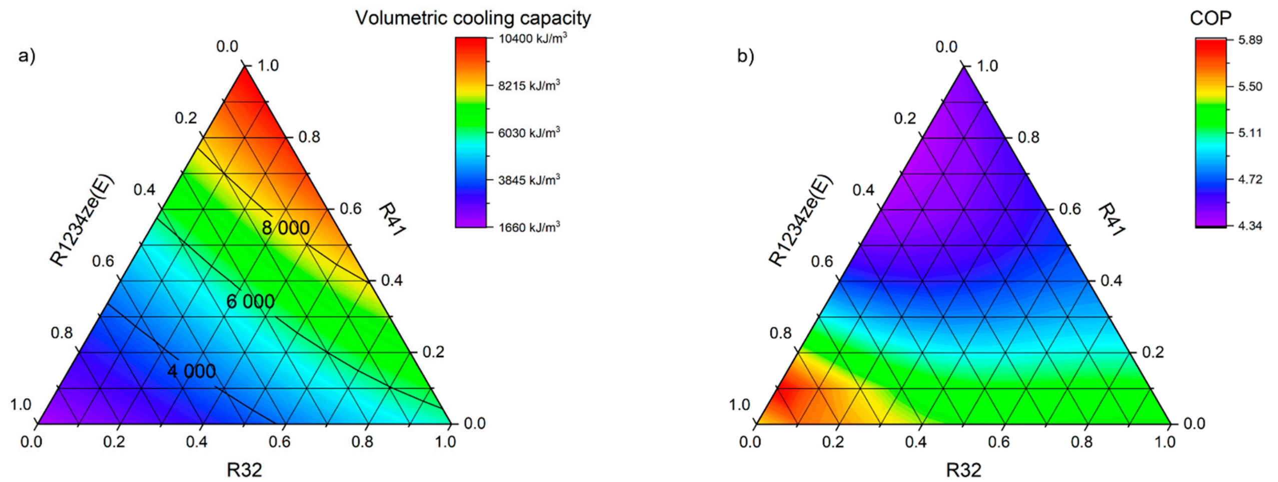

Figure 5. Volumetric cooling capacity (**a**) and (**b**) the COP for air-conditioning system (t_e/t_c = 0/30 °C) with R32–R41–R1234ze(E) mixture as refrigerant.

In the case of low-temperature cycles, additional problems in finding the proper mixture are the areas of high pressure ratio and high discharge temperature (Figure 6a,b). The use of a mixture with a mass fraction of R1234ze(E) greater than or equal to 0.4 leads to exceeding the assumed permissible level of the compression ratio for a single-stage cycle. In addition, a fraction greater than 0.8 largely leads to an evaporation pressure below 1 bar. For freezing circuits, it is not possible to select a mixture that does not contain R41, as the normal boiling point and GWP limitations effectively exclude the use of the R32–R1234ze(E) binary mixture. It is necessary to introduce a third component that will lower both these parameters at the same time. Similarly to the case of air-conditioning cycles, for the considered ternary mixture, there is only one composition that meets all the assumptions—0.1/0.8/0.1. However, further analysis of Figure 6a–d shows that by abandoning the R1234ze(E) component, all the operating parameters of the circuit are improved. The use of the binary mixture R32–R41 (0.1/0.9) allow the increase of both the COP and the volumetric cooling capacity, by 3% and 20.6%, respectively. A 6 K reduction in discharge temperature is also achieved, while the pressure ratio value drops below 5.5.

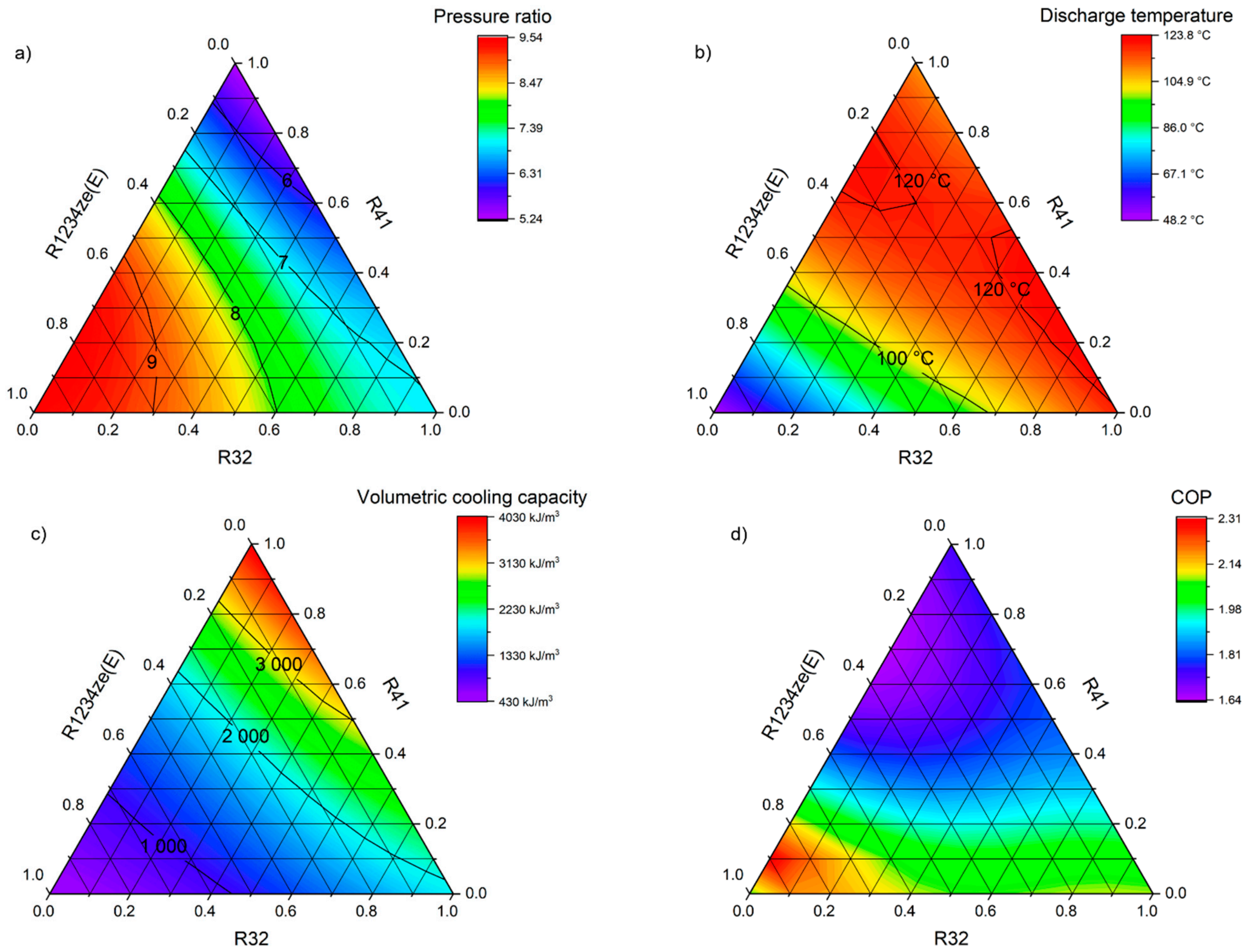

Figure 6. Low-temperature system parameters ($t_e/t_c = -30/30$ °C) of the R32–R41–R1234ze(E) mixture: (**a**) pressure ratio; (**b**) temperature of the refrigerant vapour at the compressor discharge; (**c**) volumetric cooling capacity; and (**d**) COP.

3.2. R161–R41–R1234ze(E)

The second mixture analyzed is a combination of R161, R41, and R1234ze(E). This composition is similar to the first mixture, but the R32 refrigerant has been replaced with R161, which also belongs to the HFC group. It has a very low GWP (equal to 12). In this mixture, R161 and R41 are highly flammable, while R1234ze(E) belongs to the 2 L class; therefore, it can be assumed that ternary mixtures will also belong to the highest flammability class. Each mixture component has a GWP below 150 (Figure 7a), so none of the points exceeded the strictest limit. Similar to the previously considered mixture, the normal boiling point of mixtures with high proportions of R1234ze(E) precludes the use of part of the composition in low temperature systems (Figure 7b). The heterogeneity of the mixtures is a serious problem in both the high- and low-temperature systems. Due to the very high temperature glides of this mixture for both analyzed cycles, only the narrow range of compositions meet the criterion of the maximum Δt_{glide} of 10 K, as presented in Figure 8.

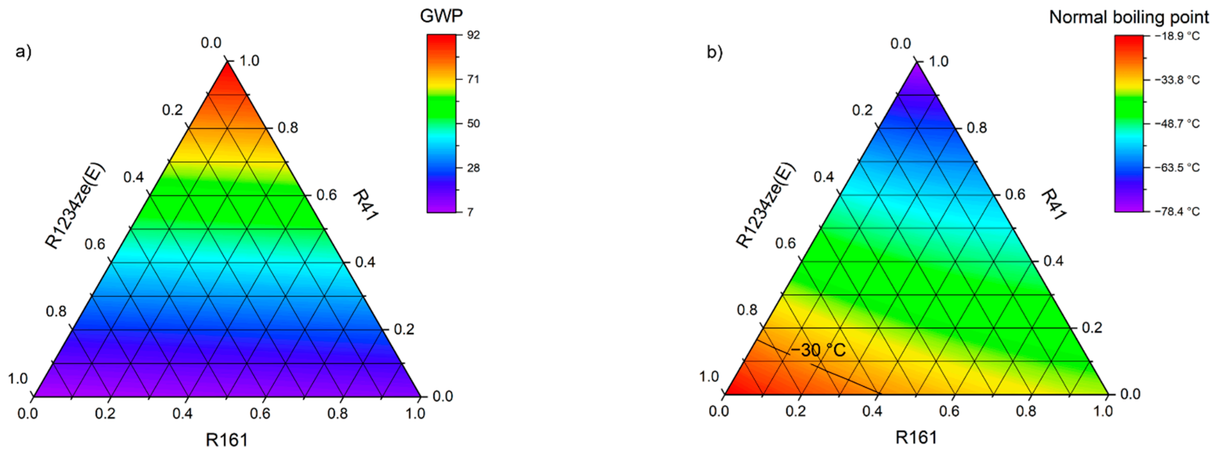

Figure 7. Summary of the basic properties for the R161–R41–R1234ze(E) mixture: (**a**) GWP and (**b**) normal boiling point.

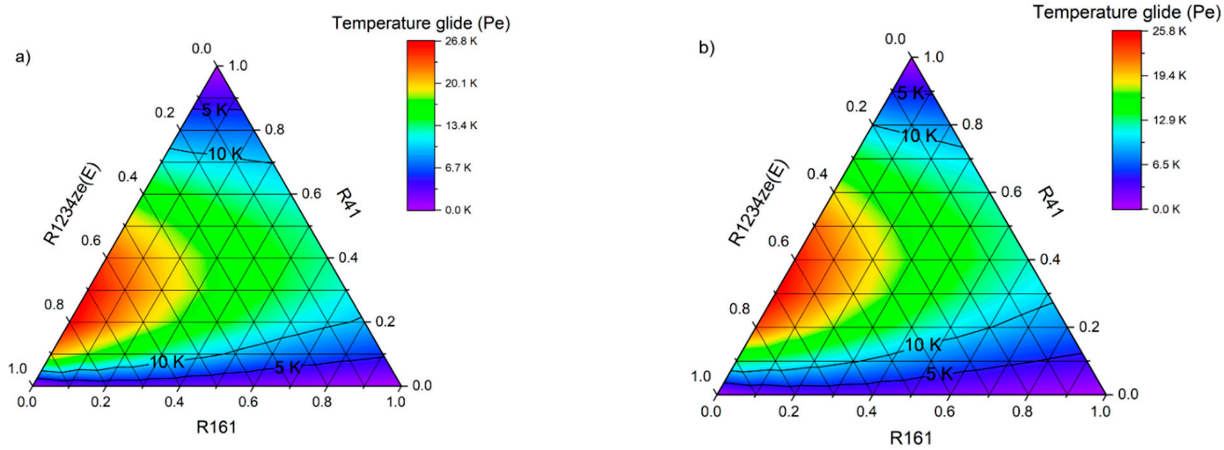

Figure 8. Temperature glide at the evaporation pressure for cooling cycles with R161–R41–R1234ze(E) mixture as refrigerant: (**a**) AC system (t_e/t_c = 0/30 °C); (**b**) low-temperature system (t_e/t_c = −30/30 °C).

The similarity to the previous mixture can also be seen in the COP and volumetric capacity charts (Figure 9). The areas of the highest values of these parameters are mutually exclusive, so it is necessary to consider which of the values will be more important for the end user. Considering the low cooling capacity of split air-conditioning devices, it can be concluded that higher energy efficiency will be more beneficial. Choosing a mixture with mass fractions of 0.8/0.1/0.1 will result in a much higher COP compared to a mixture with mass fractions of 0.1/0.8/0.1. An additional advantage is also a lower temperature glide.

In the case of mixtures predestined for operation in low-temperature circuits, the choice of a mixture with the 0.8/0.1/0.1 composition gives an additional advantage resulting from a significantly lower temperature of the medium after the compression process (see Figure 10b). Lowering this temperature by more than 30 K will be crucial for the operation of the system, especially in the summer. Discharge temperature drop will result in the lack of restrictions in terms of thermal stability of oils or the need for additional cooling of the compressor working elements. Compared to the 0.1/0.8/0.1 composition, the pressure ratio does increase, but its value remains at an acceptable level of 7.55 (Figure 10a). The mixture R161–R41–R1234ze(E) with mass fractions of 0.8/0.1/0.1 seems to be optimal also for low-temperature cycles, where for the assumed operating parameters it obtains COP = 2.2, with a volumetric cooling capacity of 1129 kJ/m^3 (Figure 10c,d).

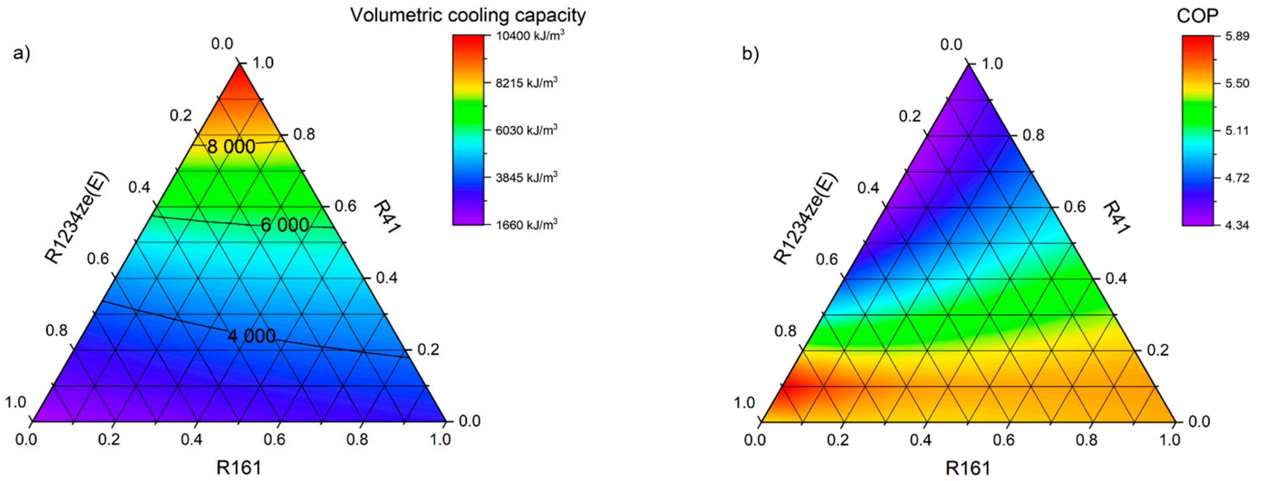

Figure 9. Volumetric cooling capacity (**a**) and (**b**) the COP for air-conditioning system (t_e/t_c = 0/30 °C) with R161–R41–R1234ze(E) mixture as refrigerant.

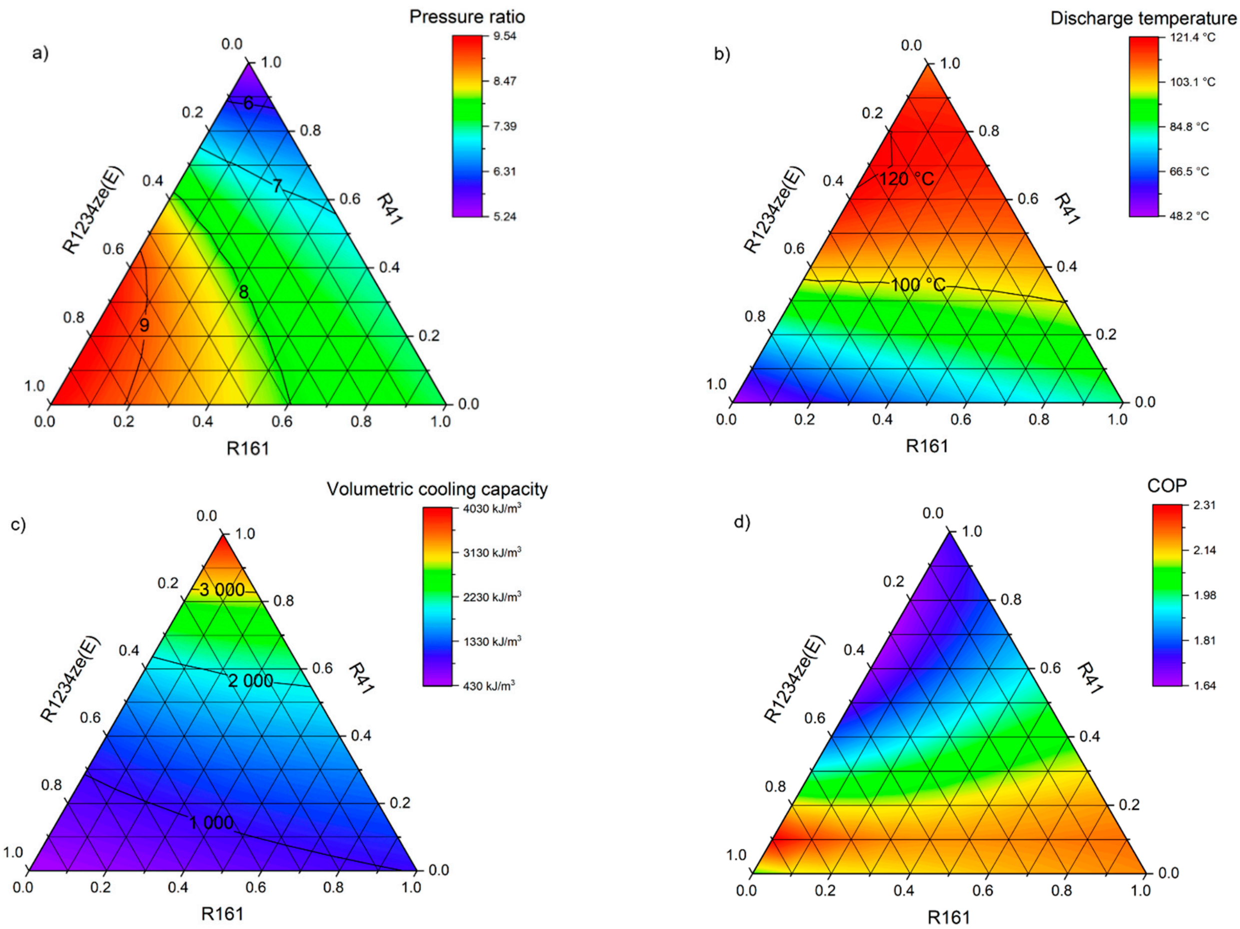

Figure 10. Low-temperature system parameters (t_e/t_c = −30/30 °C) of the R161–R41–R1234ze(E) mixture: (**a**) pressure ratio; (**b**) temperature of the refrigerant vapour at the compressor discharge; (**c**) volumetric cooling capacity; and (**d**) COP.

3.3. R1234yf–R152a–RE170

The third mixture has a completely different composition to the previous ones. It was decided to check the working parameters of the mixture, which in addition to fluorinated HFC/HFO refrigerants also contains dimethyl ether, known as RE170. The proportion of dimethyl ether in the mixture is likely to influence the toxicity and flammability class of the new refrigerant. The mixture's flammability class is assumed to be 3, as to be expected from a mixture with a high proportion of RE170. The representative from the HFC group is R152a. This is often used in existing mixtures because of its low GWP and its relatively high normal boiling point, thus prompting its use in medium to high temperature circuits. The last mixture component is the most common and most easily available fluid from the HFO group—R1234yf. As in the R161–R41–R1234ze(E) mixture, all the constituents have a GWP lower than 150; therefore, the GWP of the mixture itself does not exceed this value (Figure 11a). The flaws of this blend is that normal boiling point never reached −30 °C, which proves third mixture is not predisposed for use in low-temperature units (Figure 11b). The device's operation under negative pressure is associated with certain dangers and requires the use of a larger compressor. Considering all of the above, in the case of the R1234yf–R152a–RE170 mixture, it was decided to analyze only the high-temperature cycle.

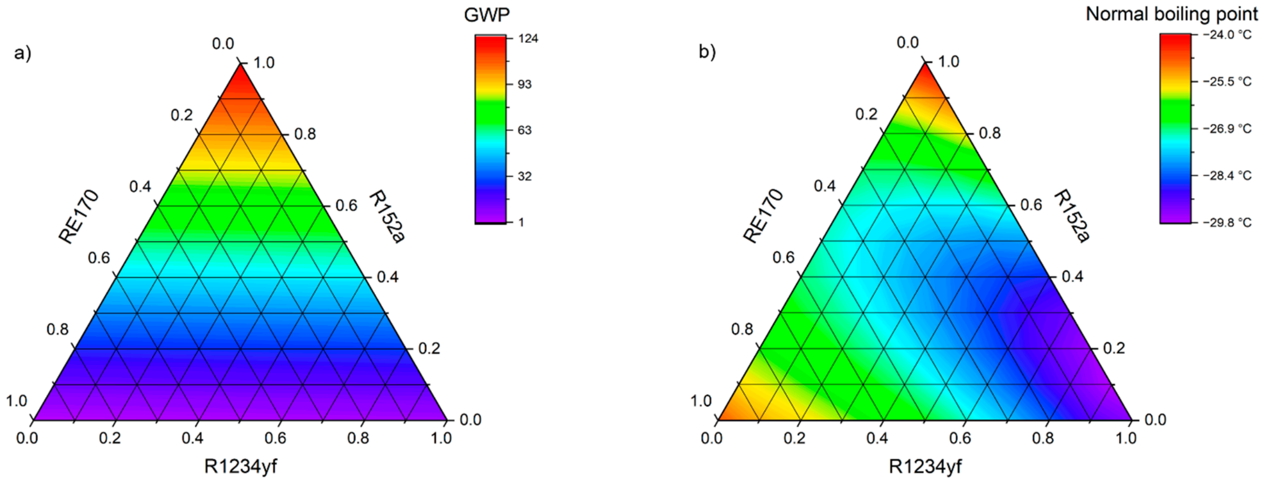

Figure 11. Summary of the basic properties for the R1234yf–R152a–RE170 mixture: (**a**) GWP and (**b**) normal boiling point.

When analyzing the third mixture (R1234yf–R152a–RE170) and the temperature glides obtained (Figure 12), the conclusion is that the temperature difference during the phase change does not exceed 1 K for the assumed evaporating pressure level regardless of the composition; thus, it can be considered at least as near-azeotropic. The achieved volumetric cooling capacity and COP play the greatest role in the assessment (Figure 13a,b). The highest COP value determined was 5.67, but it was for pure RE170. Given the importance of the fluid share from the A3 group being as small as possible, this point was not analyzed. It is worth noting that, with a binary and ternary mixture, as many as 54 points out of 63 have a COP > 5.48, which is the reference value obtained by R134a. There are two ternary mixtures that meet all the assumptions of the work and appear to be good potential substitutes for phased-out fluorinated greenhouse gases. The first is the 0.1/0.1/0.8 mixture, which has a COP of 5.65. The second mixture with a very similar COP has a mass fraction of 0.1/0.5/0.4, but is additionally characterized by a lower normal boiling point and temperature glide well below 0.1 K. In this term, it can be considered as azeotropic. The second composition also has a volumetric cooling capacity that is 124 kJ/m^3 higher than that of the first mixture.

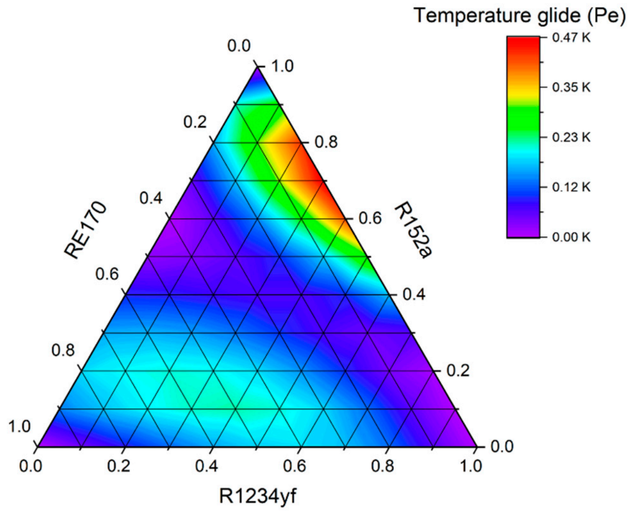

Figure 12. Temperature glide at the evaporation pressure for air-conditioning system (t_e/t_c = 0/30 °C) with R1234yf–R152a–RE170 mixture as refrigerant.

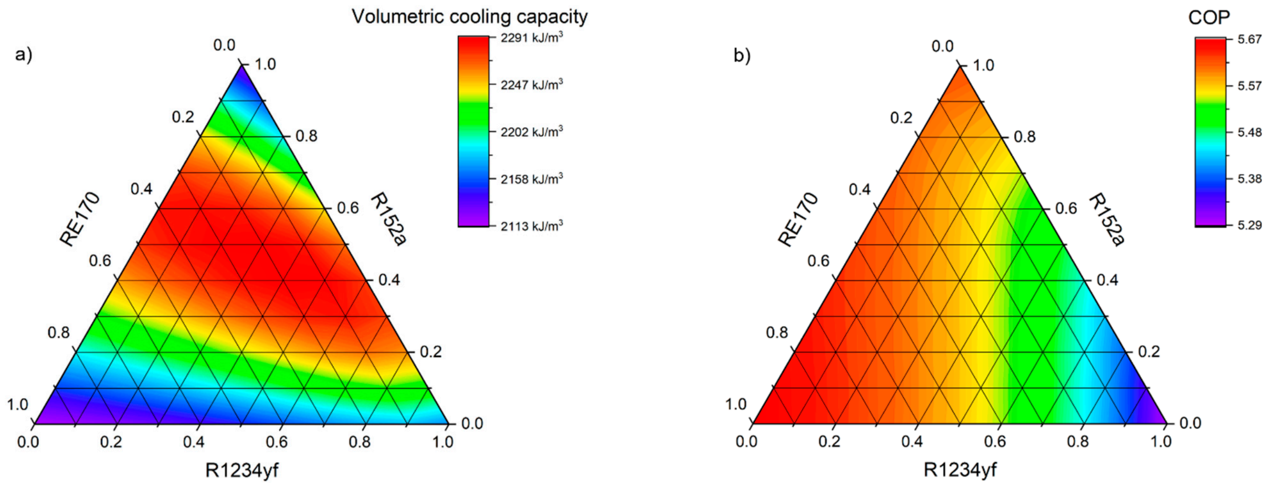

Figure 13. Volumetric cooling capacity (**a**) and (**b**) the COP for air-conditioning system (t_e/t_c = 0/30 °C) with R1234yf–R152a–RE170 mixture as refrigerant.

3.4. R1243zf–R152a–RE170

The last analyzed mixture is similar in composition to the third mixture, because it combines the two previously used refrigerants, RE170 and R152a. The HFO group refrigerant has been changed to a less popular R1243zf, as yet unused in any of the encountered mixtures. The compositions discussed, like those in the previous point, will most likely belong to the higher flammability group due to the presence of dimethyl ether. As with the previous combination of fluids, the GWP does not exceed that of R152a, and due to the normal boiling point, the mixture is considered only in high-temperature circuits (Figure 14).

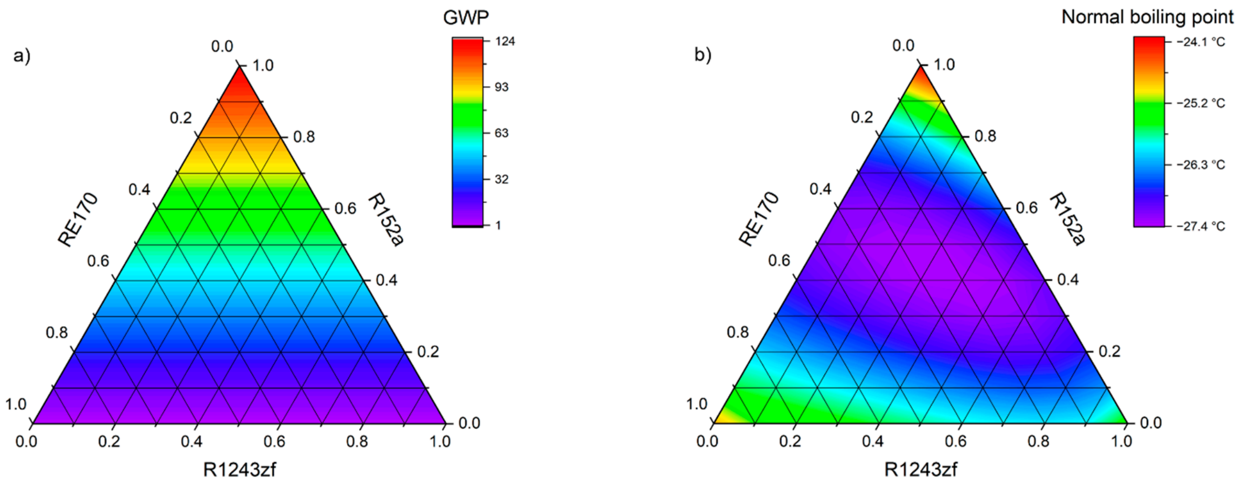

Figure 14. Summary of the basic properties for the R1243zf–R152a–RE170 mixture: (**a**) GWP and (**b**) normal boiling point.

The mixture R1243zf–R152a–RE170, regardless the composition, fulfils the requirements for the Δt_{glide} and π limits. For this mixture, an interesting parameter is the normal boiling point (Figure 14b), which reaches its minimum in the center of the triangle, not at the corners or sides. The lowest temperature achieved was −27.4 °C for the composition of 0.4/0.4/0.2. This phenomenon did not occur in the previously tested mixtures. Although all the points reached a negative normal boiling point, it is not low enough to use this mixture safely in freezing equipment. The temperature glide presented in Figure 15 is definitely below 1 K and has the lowest values in the center of the graph.

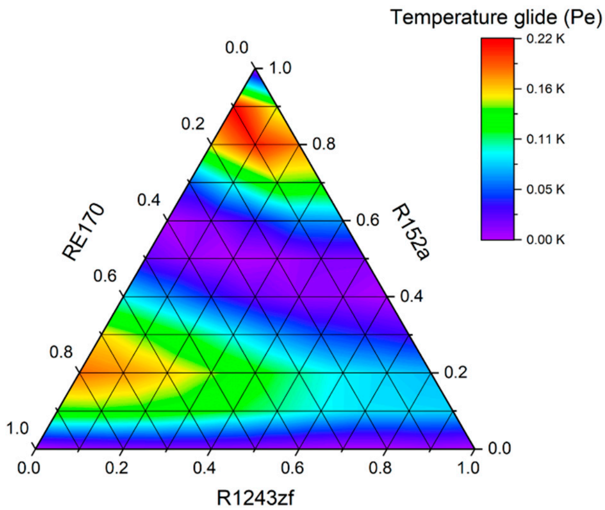

Figure 15. Temperature glide at the evaporation pressure for air-conditioning system (t_e/t_c = 0/30 °C) with R1243zf–R152a–RE170 mixture as refrigerant.

Moreover, a significant number of mixtures with a weight fraction of R152a in the range of 40–60% show a temperature glide significantly below 0.1 K, which makes them azeotropic. Considering the volumetric cooling capacity and the COP (Figure 16), as well as the previously mentioned normal boiling point and temperature glide, the best composition turns out to be 0.2/0.5/0.3. This is a compromise between a moderately high COP and a nearly zero temperature glide. Furthermore, it is a ternary mixture, and the RE170 belonging to the A3 flammability class does not make up the largest share. The second optimal substance, with a slightly higher COP, but also Δt_{glide}, is the mixture with a weight composition of 0.1/0.5/0.4. At the same time, it is a composition that achieves almost the highest volumetric cooling capacity of 2284 kJ/m^3.

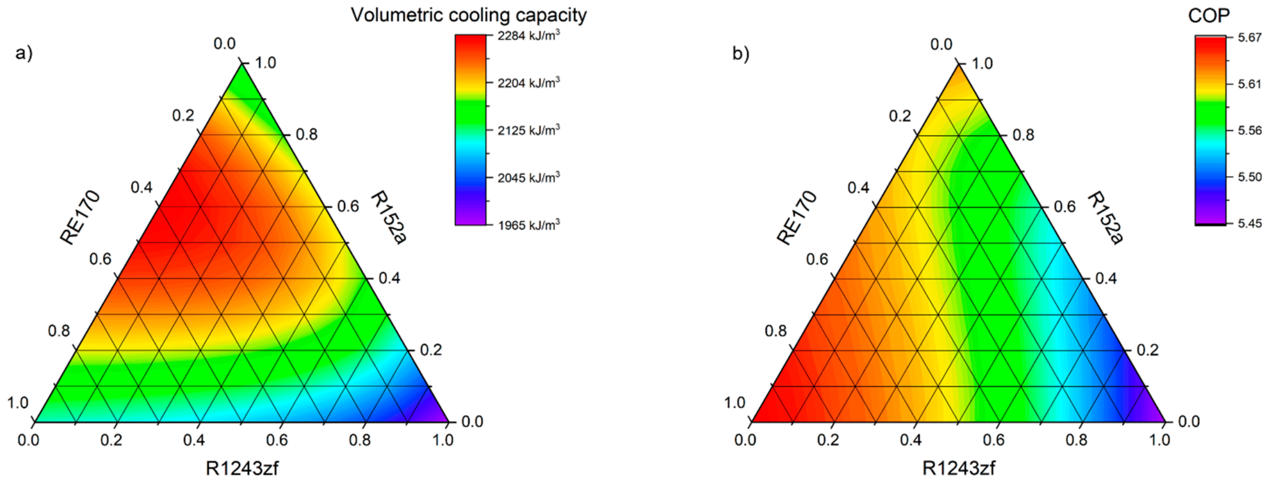

Figure 16. Volumetric cooling capacity (**a**) and (**b**) the COP for air-conditioning system (t_e/t_c = 0/30 °C) with R1243zf–R152a–RE170 mixture as refrigerant.

4. Discussion

Summarizing the results presented in this study, seven optimal mixture compositions were selected, of which six are ternary mixtures and two are binary:

- R32–R41–R1234ze(E) 0.9/0/0.1 air-conditioning cycle
- R32–R41–R1234ze(E) 0.1/0.9/0 low-temperature cycle
- R161–R41–R1234ze(E) 0.8/0.1/0.1 both cycles
- R1234yf–R152a–RE170 0.1/0.1/0.8 air-conditioning cycle
- R1234yf–R152a–RE170 0.1/0.5/0.4 air-conditioning cycle
- R1243zf–R152a–RE170 0.2/0.5/0.3 air-conditioning cycle
- R1243zf–R152a–RE170 0.1/0.5/0.4 air-conditioning cycle

Their distribution on the experiment plan is shown in Figure 17.

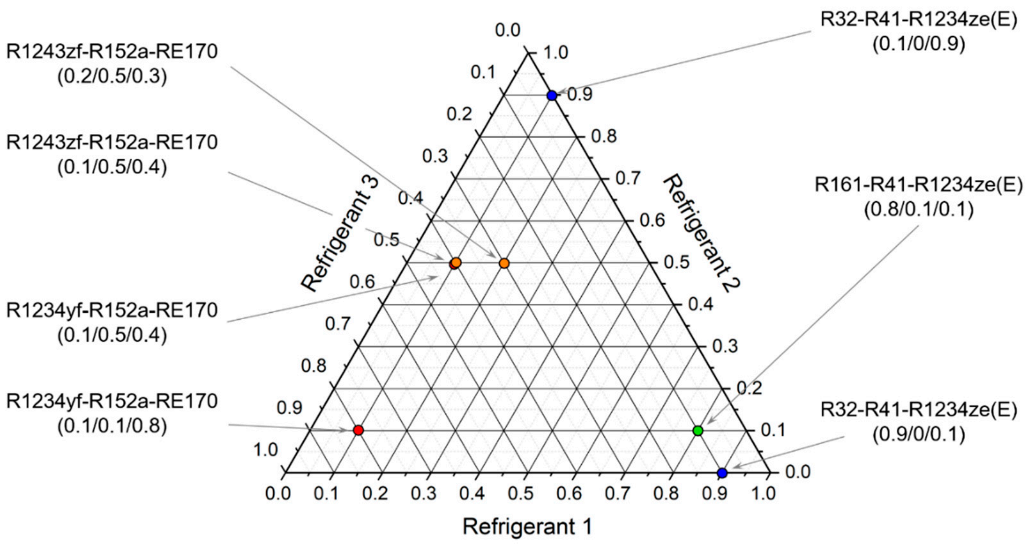

Figure 17. Marking the optimal mixtures in a triangular pattern.

There were many criteria for proving the applicability of a given refrigerant. The main ones were the temperature glide values achieved at p_e and within the whole range of working pressures. Based on the temperature differences obtained for the evaporation process, for which the limit value was set as 10 K, a significant proportion of the points for the first two mixtures were rejected. The temperature drop in the evaporation process for the assumed cooling cycle and variable evaporation temperatures for selected mixtures are presented in Figure 18. The figure shows that only for the mixture R161–R41–R1234ze(E), significant changes in the evaporation temperature are obtained, and therefore this mixture must be classified as zeotropic (ZEO). At the same time, it is clearly visible that with the increase in the evaporation temperature, the temperature glide increases significantly.

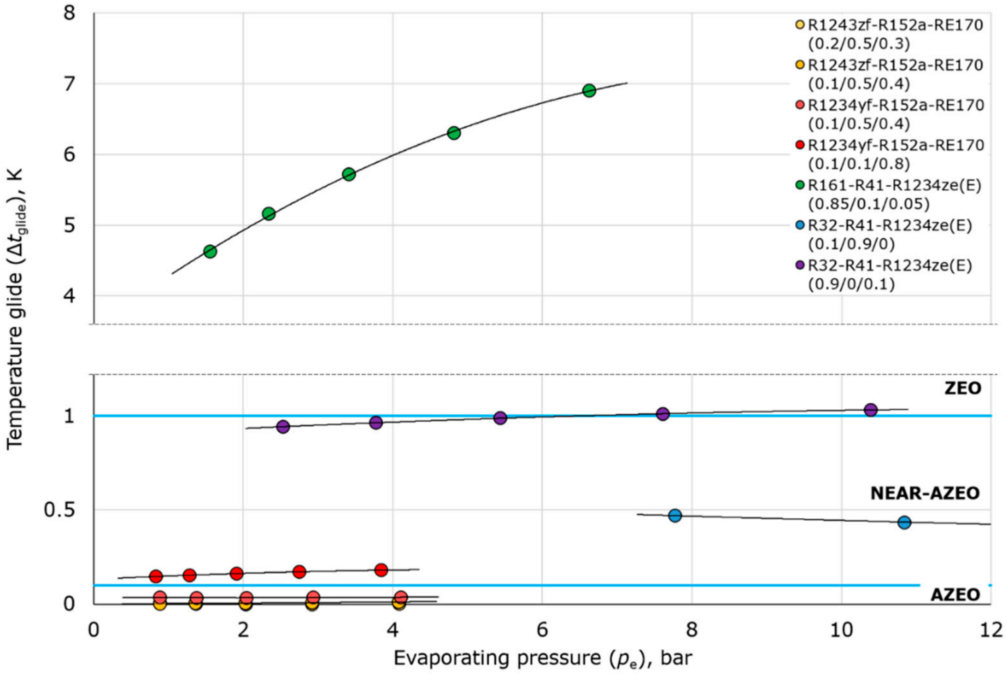

Figure 18. Temperature glide of the new mixtures depending on the evaporating pressure.

On the other hand, all the mixtures containing the R152a and RE170 fluids are characterized by a small temperature glide, the value of which does not exceed 0.5 K. The binary mixture R32–R1234ze(E) shows a temperature glide of almost exactly 1 K (limit for near-azeotropic mixtures). Figure 18 also shows that among the selected mixtures, R32–R41 achieves much higher evaporating pressures.

Table 2 shows a comparison of the newly defined mixtures with the reference refrigerants dedicated to low-temperature installations. The parameters that are favorable to the new mixtures are the GWP and specific cooling capacity. Compared to currently used refrigerants, it was found that the most optimal is a R32–R41 binary mixture with a mass fraction of 0.9/0.1, for which the specific cooling capacity is 243 kJ/kg and is more than twice as high as for the R404A or R507A. The great advantage of the R32–R41 binary mixture is the increase in volumetric cooling capacity by over 2700 kJ/m^3. The disadvantages are the slightly higher temperature glide and high working pressures. The biggest problem seems to be the much higher discharge temperature exceeding 110 °C, which is typical for the currently implemented and used R404A substitutes. Conversely, R161–R41–R1234ze(E) obtains a lower volumetric cooling capacity, which is dictated by a much higher specific vapor volume, at the same time showing a much higher temperature glide, which can eliminate this refrigerant from systems requiring precise evaporator temperature. The use of this mixture, however, allows to significantly improve the COP; compared to R404A, the increase is as much as 15.8%. However, the use of these mixtures requires a profound change in the refrigerant market, as currently R41 is not widely available.

Table 2. Comparison of the newly defined mixtures to the reference refrigerants suitable for low-temperature systems; properties based on [27].

Refrigerant	Unit	R404A (R125–R143a–R134a)	R507A (R125/R143a)	R32–R41-R1234ze(E)	R161–R41-R1234ze(E)
Weight share	-	0.44/0.52/0.04	0.5/0.5	0.1/0.9/0	0.8/0.1/0.1
GWP [1]	-	3922	3985	150	20
Critical temperature	°C	72.12	70.62	45.32	95.99
Critical pressure	bar	37.35	37.05	58.93	53.10
Normal boiling point	°C	−45.74	−47.01	−76.80	−39.77
Molar mass	g/mol	97.60	98.86	35.25	48.87
Evaporating temperature	°C	−30	−30	−30	−30
Evaporating pressure	bar	2.02	2.13	7.77	1.55
Condensing temperature	°C	30	30	30	30
Condensing pressure	bar	14.14	14.59	41.66	11.73
Pressure ratio	-	6.99	6.85	5.36	7.55
Compressor Discharge temperature	°C	52.86	51.75	110.63	87.43
Specific suction vapour volume	m^3/kg	0.095	0.089	0.062	0.252
Liquid density	kg/m^3	1019.4	1022.6	957.6	708.6
Temperature glide for p_e	K	0.48	0.003	0.47	4.63
Volumetric cooling capacity	kJ/m^3	1118.2	1151.6	3894.1	1128.9
Specific cooling capacity	kJ/kg	106.01	102.09	243.28	284.94
Specific heating capacity	kJ/kg	161.86	156.24	381.77	414.37
Specific work of the cycle	kJ/kg	55.85	54.15	138.49	129.43
COP	-	1.90	1.88	1.76	2.20

Table 3 shows the properties of the mixtures considered as refrigerants in air conditioning cycle. R410A, R134a, R32, and R429A were used as reference. An indicator that definitely favors the new refrigerants is the GWP, the value of which for R32-free mixtures does not exceed 63 and is less than half the permissible limit. Of the reference substances, only R429A has the same low GWP. The COP for the new mixtures are also favorable, since they all exceed a value of 5.34. When comparing the temperature glide, it can be observed that it is almost zero for the mixtures containing RE170, which is a slight advantage over R429A. These mixtures achieve a volumetric cooling capacity almost identical to that of R134a and approximately 6–12% higher than that of R429A. However, they are not in competition with R32 or mixtures containing it, so a higher amount of refrigerant in the system will be required. In terms of the obtained volumetric cooling capacity, the mixtures R32–R1234ze(E) and R161–R41–R1234ze (E) may be an interesting proposition. For both mixtures, the obtained values are much higher than for R134a, and in the case of a binary mixture R32–R1234ze(E) also at a similar level as for R32 and R410A. By analyzing all the variables, it can be summarized that the mixture R1234yf–R152a–RE170 with the weight shares of 0.1/0.5/0.4 seems to be the most promising for the implementation in air-conditioning cycles.

The conclusions drawn from the theoretical analysis should be confirmed by means of experimental studies of the various evaporation and condensation temperatures. To do this, mixtures with similar compositions should be tested, with a smaller jump in the weight shares of the individual components. However, this is future research work, as the scope of this work only included theoretical considerations regarding the new mixtures.

Table 3. Comparison of the newly defined mixtures to the reference air-conditioning refrigerants; properties based on [27].

Refrigerant	Unit	R134a	R32	R410A (R32–R125)	R429A (RE170–R152a–R600a)	R1234yf-R152a–RE170	R1234yf-R152a–RE170	R1243zf-R152a–RE170	R1243zf-R152a–RE170	R32–R41-R1234ze(E)	R161–R41-R1234ze(E)
Weight share/name	-	1,1,1,2-Tetrafluoro-ethane	Difluoro-methane	0.5/0.5	0.6/0.1/0.3	0.1/0.1/0.8	0.1/0.5/0.4	0.2/0.5/0.3	0.1/0.5/0.4	0.9/0/0.1	0.8/0.1/0.1
GWP [1]	-	1430	675	2088	14	14	63	63	63	608	20
Critical temperature	°C	101.06	78.10	71.34	121.95	123.28	114.90	113.04	115.22	80.58	95.99
Critical pressure	bar	40.59	57.82	49.01	47.30	51.41	47.82	46.07	47.77	58.07	53.10
Normal boiling point	°C	−26.07	−51.65	−51.62	−25.37	−25.88	−27.44	−27.33	−27.27	−50.23	−39.77
Molar mass	g/mol	102.03	52.02	72.58	50.76	50.62	58.38	61.86	57.82	55.02	48.87
Evaporating temperature	°C	0	0	0	0	0	0	0	0	0	0
Evaporating pressure	bar	2.93	8.13	7.98	2.64	2.75	2.92	2.86	2.89	7.61	4.82
Condensing temperature	°C	30	30	30	30	30	30	30	30	30	30
Condensing pressure	bar	7.70	19.28	18.84	6.64	6.99	7.42	7.30	7.36	18.16	11.73
Pressure ratio	-	2.63	2.37	2.36	2.52	2.54	2.54	2.55	2.54	2.39	2.43
Compressor Discharge temperature	°C	41.97	64.99	51.26	44.62	48.78	48.56	47.94	48.84	62.75	53.36
Specific suction vapor volume	m^3/kg	0.069	0.045	0.033	0.157	0.151	0.122	0.118	0.125	0.046	0.086
Liquid density	kg/m^3	1187.5	939.6	1032.7	633.9	697.8	780.6	804.9	773.4	957.6	688.5
Temperature glide for p_e	K	0.00	0.00	0.08	0.64	0.15	0.00	0.00	0.02	1.01	6.30
Volumetric cooling capacity	kJ/m^3	2263.5	5743.2	5292.9	2026.9	2158.4	2271.7	2226.5	2255.3	5393.2	3572.2
Specific cooling capacity	kJ/kg	156.88	259.98	173.11	318.32	326.24	277.84	261.77	281.34	248.74	307.00
Specific heating capacity	kJ/kg	185.53	308.70	206.27	374.80	384.02	327.34	308.48	331.43	295.30	362.11
Specific work of the cycle	kJ/kg	28.65	48.72	33.17	56.48	57.78	49.51	46.71	50.08	46.55	55.11
COP	-	5.48	5.34	5.22	5.64	5.65	5.61	5.60	5.62	5.34	5.57

5. Conclusions

Referring to the applicable environmental parameter limits set on refrigerants, four new refrigerant mixtures have been proposed in this work. The optimal weight shares of the individual components were estimated by analyzing the GWP, thermodynamic, and operational parameters.

Theoretical tests were performed using the REFPROP 10.0 program, and the collected data allowed for a preliminary estimation to be made. On the basis of theoretical analyzes, it was shown that all the proposed compositions, except for the R161–R41–R1234ze(E) mixture, can be classified as near-azeotropes or even azeotropes, because their temperature glide in a wide range of evaporation pressure does not exceed 1K. At optimal compositions, the share of HFOs in all mixtures does not exceed 20%. After considering the advantages and disadvantages of the refrigerants proposed, it was determined that the most optimal composition in high-temperature (air conditioning) systems was the R1234yf–R152a–RE170 mixture with a weight share of 0.1/0.5/0.4. This is argued by its low GWP, equal to 63, the relatively high COP of 5.61, the relatively low normal boiling point of -27.27 °C, and a lower weight share of the most flammable components (flammability class 3) than in the case of R429A.

The analyzes also show that it is extremely difficult to find a blend with a negligible impact on the greenhouse effect and at the same time good thermodynamic properties, which could be used as a replacement for R404A or R507A in low-temperature systems. Both of the proposed mixtures have disadvantages compared to currently used refrigerants. Although the R32–R41 achieves high volumetric cooling capacity, it is also characterized by high evaporating and condensing pressures and high discharge temperatures, which will result in higher thermal and force loads of the compressor working elements and may lead to their shorter life span. On the other hand, the R161–R41–R1234ze(E) mixture, despite the high coefficient of performance, shows nearly ten times higher temperature glide than R404A, which may cause evaporator malfunctions. Components with extremely different vapor pressures can cause excessive frosting to the initial sections of the evaporator due to the evaporation of low-boiling components. On the other hand, components with a high boiling point may not completely evaporate, which can lead to fractionation of the refrigerant inside the system and change its operating parameters. In case of extreme temperature glides, it is necessary to increase the vapor superheat set point to prevent the compressor from sucking in liquid refrigerant, which obviously affects the efficiency of the system. Therefore, further research should be directed towards this application. It should be emphasized that the presented analyzes do not explore the issue of using these mixtures in cooling cycles completely. Above all, further studies of the flammability and safe use of the presented mixtures are required, as all the components used are flammable, and a significant part of them belong to the highest flammability class.

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Conflicts of Interest: The authors declare no conflict of interest.

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