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THEORETICAL ANALYSIS OF LOW FLAMMABLE AND ECOLOGICAL-SAVE MIXTURES OF LIQUEFIED PETROLEUM GAS AND TETRAFLUOROETHANE AS REFRIGERANTS

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SUMMARY: Liquefied petroleum gas (LPG), like other hydrocarbon gases, has good thermo-physical and ecological properties, but its high-flammability has limited its direct use in refrigeration applications. Therefore, this paper investigates the thermodynamic performance of low flammable and ecological-save mixtures of LPG and tetrafluoroethane refrigerant in a refrigeration system. The computational results of global warming potential (GWP) and low flammability limit shown that the blends with 50 – 90 % LPG contents have GWP below 750 and fall within the lower flammability safety class of which five were selected for further analysis. The results showed that the thermal conductivity and the refrigerating effects produced by the selected blends are greater than those of the reference refrigerant (R134a). Also, the selected blends exhibited appropriate lower pressure ratio, discharge temperature and specific power than R134a refrigerant. Four of the blends required low power input and exhibited higher coefficient of performance which are clear indication that they are more energy efficient than R134a in the refrigeration systems.

Key words: *blends, ecological-save refrigerants, flammability, LPG, R134a*

INTRODUCTION

Most residential, commercial and industrial refrigeration and air-conditioning systems used for creating conducive environments, preserve and store products in homes, restaurants and large storage warehouses are commonly operated on the principles of vapour compression cycle. In this cycle, the working fluids (refrigerants), rapidly evaporates and condenses as it alternates between the vapour and liquid phases without leaving the refrigerating system. The refrigerant receives heat from the cold body during evaporation and uses this heat as its required latent heat to transform from liquid to vapour, whereas it rejects heat to external bodies during condensation *(Banjo et al., 2019).*

Halogenated refrigerants, especially chlorofluorocarbons (CFCs) and hydrochloroflurocarbons (HCFC), are the commonly used refrigerants in vapour compression refrigeration systems over the past century, Because of their outstanding thermodynamic and thermo-physical qualities. However, they possess poor environmental qualities due to their undesirable contributions to the ozone layer destruction and climate change by reason of their high global warming potential (GWP) *(Hmood et al., 2021)*. In 1987, Montreal Protocol regulated the use of both the partially and fully halogenated chemicals including hydro-chlorofluorocarbons (HCFCs) and CFCs in air-conditioning and refrigeration systems as a result of their high ozone depletion potential (ODP) *(Bolaji, 2014)*.

Once ozone depleting chemicals were outlawed, their emissions significantly decreased and the refrigeration industry moved its focus to

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the use of hydrofluorocarbons (HFCs) as substitute refrigerants *(Mota-Babiloni et al., 2017)*. HFC refrigerants were proposed as replacement refrigerants for CFCs and HCFCs in many refrigeration, air-conditioning and heat pump applications. In these applications, HFC refrigerants such as R134a, R404A, R407C and R410A, were developed as alternatives working fluids *(Bolaji et al., 2017, Roy and Halder, 2020)*.

HFC refrigerants are a desirable option for refrigeration applications since most of the refrigerants are non-flammable. Equipment like air conditioning and refrigeration systems are extremely simple to produce, install and operate when the working fluids are non-flammable and when such refrigerants spill, they pose no fire risk. Even though these refrigerants are virtually inert to the ozone layer, they are greenhouse gases with high-GWP because of their extremely stable chemical composition. Majority of the HFC refrigerants are not combustible *(Purohit et al., 2020)*.

HFC usage is presently expanding at a rapid rate, posing a severe threat to global warming and climate change. It will be difficult to protect against the emissions of these HFC refrigerants from refrigeration equipment during operations, services and repairs. Ecological-save practices can only obtainable by complete prohibition of the high-GWP refrigerants *(Mgbemene et al., 2016)*. Currently, there are greater demands than ever before to minimize greenhouse gas (GHG) emissions to safeguard the environment, as well as to develop a new generation of low-GWP and ecologically harmless refrigerants *(Flerlage et al., 2021)*.

Natural refrigerants have made a comeback as working fluids in refrigeration applications, attributed to rising issues about the environmental effect of synthetic refrigerants. Natural refrigerants are chemical compounds that are naturally present in the environment and are created by biochemical functions in nature. Some of these naturally occurring substances are hydrocarbons, ammonia, carbon dioxide and water. They have a very low or negligible potential for global warming and are not ozone-depleting. These compounds were formerly utilized as refrigerants in the 19th century and are now being reintroduced to the refrigeration systems due to their insignificant environmental effect *(Razzaq et al., 2018)*. As a natural substance, hydrocarbon refrigerants have no destructive effect on ozone and has nearly zero impact on greenhouse effect.

In the air-conditioning and refrigeration industries, the utilisation of hydrocarbons (HCs) and their blends as working fluids has gained interest in recent times due to their benefits in terms of their good ecological properties. Butane (R600), isobutane (R600a), propane (R290), dimethylether (RE170), ethylene (R1150) and propylene (R1270), are the most often utilized hydrocarbon refrigerants in refrigeration applications. Several publications have examined the suitability of these refrigerants and their blends. Bolaji and Huan *(2012)* studied the performance of propylene (R1270), propane (R290) and iso-butane (R600a) as R22 replacements in a refrigeration system incorporated with an internal heat exchanger. The study found that the thermo-physical parameters of R290 and R1270 matched those of R22, and the two refrigerants outperformed R22 in the system. However, the specific volume and saturation pressure of R600a differed greatly from those of R22, and the system will require extensive redesign before it can be used as R22's alternative.

Chen et al. *(2019)* investigated how well a zeotropic blend of R290 and R600a performed in a home refrigeration system. In order to improve the system's performance, the study used a phase separator and an internal heat exchanger. The outcomes show that the system outperformed the conventional household refrigeration system with a 13.5 percent improvement in volumetric refrigeration capacity under the same working conditions. Thavamani and Senthil (2022) also investigated the performance of R290 and R600a refrigerant blend as R134a's substitute in an existing refrigerator. The experiment demonstrated that due to the high latent heat of the hydrocarbon mixture, it produced a lower temperature in the evaporator than R134a. Under the same operating conditions, the coefficient of performance value of the system operating with hydrocarbon mixture is higher than that of R134a system.

Ghanbarpour et al. *(2021)* evaluated the environmental effect and energy performance of R1270, R600a, and R290 as R134a replacements in three types of vapour compression refrigeration systems comprise of a cycle incorporating a liquid-suction heat exchanger, a two-stage and a single-stage cycles. The three hydrocarbon refrigerants performed better than R134a in all the three refrigeration systems tested. The use of hydrocarbons yielded 50% reduction in carbon footprint, with an additional 8% reduction possible by adding a liquid-suction heat exchanger.

Liquefied Petroleum Gas (LPG), a blends of hydrocarbon refrigerants, has also been investigated in vapour compression refrigeration systems. Ahmad et al. *(2020)* conducted experiments to determine whether LPG could be used to replace R134a in a home refrigeration system under subtropical temperatures. The findings of the study showed that LPG outperformed the reference refrigerant in terms of reduced mass flow rate, compressor outlet temperature and pull-down time. The study confirmed LPG as a possible replacement for R134a in household refrigeration systems. Likewise, Babarinde et al. *(2018)* investigated energy performance of evaporator, refrigerant control, condenser and compressor of a household refrigeration system using LPG and iso-butane refrigerants. The study showed that the coefficient of performance of the refrigerator working with LPG was 9.5 % higher than that of iso-butane.

All the reviewed studies have demonstrated that LPG and hydrocarbon refrigerants are not only ecologically friendly, but they also exhibit desirable thermal properties and performance characteristics in the refrigeration systems. However, the extreme flammability of hydrocarbon and LPG refrigerants has prevented their widespread adoption and use in refrigeration systems. In order to overcome this problem, hydrocarbon refrigerants can be mixed with HFC refrigerants to adjust their properties and produce new low-GWP refrigerants with lower flammability. Therefore, this paper investigates the thermodynamic performance of low flammable and ecological-save mixtures of LPG and R134a refrigerant in a refrigeration system.

MATERIALS AND METHODS

Flammability limits and GWPs of LPG and R134a blends

The flammability limits of each refrigerant (LFLi) in the blend are obtained from the literature *(Zlochower and Green, 2009)* and the blend's lower flammability limit (LFL_{blend}) is computed using LeChatelier's rule *(Kondo et al., 2008)*:

$$\frac{1}{LFL_{blend}}=\sum_{i=1}^{n}\frac{a_i}{LFL_i} \qquad [1]$$

where a_i is the mass fraction of each refrigerant in the blend and 'n' stands for the number of refrigerants that formed the blend.

Similarly, the blend's global warming potential (GWPblend) is given as (Meng et al., 2016):

$$GWP_{blend}=\sum_{i=1}^{n}a_i(GWP_i) \qquad [2]$$

where, GWP_i stands for the global warming potential of each refrigerant in the blend. GWPs of common refrigerants are available in literature *(Bitzer, 2020)*.

LPG and R134a Blends

Liquefied petroleum gas (LPG) has excellent thermo-physical and ecological properties that are suitable for ideal refrigerant except that it is a highly flammable gas. To reduce the flammability of LPG, it is mixed with 1, 1, 1, 2-tetrafluoroethane (R134a) at different ratios. The lower flammability limits (LFL) of LPG and R134a refrigerant blends at various compositions were computed using suitable equations and REFPROP software. The five mixtures that fall into the lower flammability group (LFL > 3.5 % vol.) and GWP less than 750 were selected. The ASHRAE safety classification of refrigerants is shown in Table 1. The investigated refrigerant blends are designated as RLPG50, RLPG60, RLPG70, RLPG80 and RLPG90 where the last two numbers indicated the percentage of LPG in the mixture.

Table 1. Refrigerant flammability classifications

Tablica 1. Klasifikacija zapaljivosti rashladnog sredstva

Flammability class		**Lower flammability limit (% Vol.)**	**Toxicity**	
			Lower (A)	**Higher (B)**
Class 1	Non-Flammable	No flame propagation	A1	B1
Class 2	Mildly flammable	> 3.5	A2	B2
Class 3	Highly flammable	≤ 3.5	A3	B3

The performance of these refrigerants in a standard vapour compression refrigeration cycle is analysed theoretically. The computer software employed for this study is known as REFPROP. It is the most extensively utilized refrigerant database software package *(Lemmon et al., 2013)*. It has capacity of computing the thermo-physical parameters of pure fluids and mixtures throughout a wide variety of fluid states, including liquid, gas at their saturated, subcritical and supercritical phases.

Refrigeration System

Refrigeration is a process of transferring heat from a lower temperature medium to another medium outside the system at a higher temperature. In agreement with the Thermodynamics' Second Law, such transfer of energy cannot take place unless work is done on the system. Schematic diagram of refrigeration operating on a vapour compression system is shown in Figure 1. It essentially consists of an expansion valve, an evaporator, a reciprocating compressor and a condenser. These four major components are connected by appropriate size of tubes to circulate the refrigerant with suitable thermodynamic and ecological-save properties. The pressure–volume (p–h) diagram for the cycle is shown in Figure 2. As shown in the figure, processes 1-2 is compression, 2-3 is condensation, 3-4 is expansion or throttling, and 4-1 is evaporation.

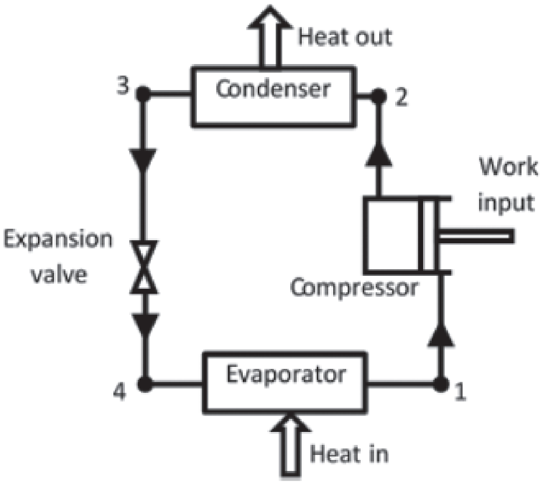

Figure 1. Schematic illustration of vapour compression refrigeration system

Slika 1. Shematski prikaz kompresijskog rashladnog sustava pare

The heat transfer across the major components in the refrigeration system were determine and used to compute the performance parameters of the refrigerant blends:

Refrigerating effect (RE) is computed using Eq. [3]:

$$RE = h_1 - h_4 \qquad [3]$$

Compressor work input (CWI) per unit mass of refrigerant is computed using Eq. [4]:

$$CWI = h_2 - h_1 \qquad [4]$$

where h_1, h_2 and h_4 are the specific enthalpies of refrigerant at inlet to the compressor, condenser and evaporator, respectively.

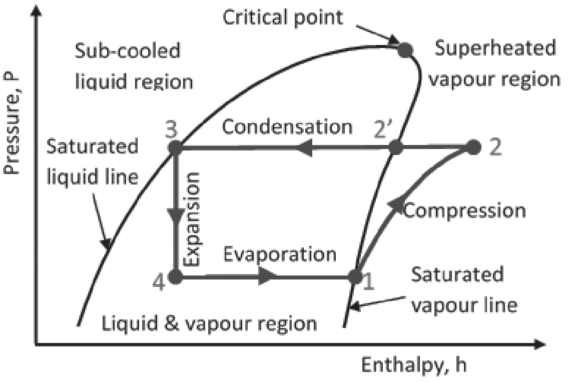

Figure 2. Simple refrigeration cycle on pressure-enthalpy diagram

Slika 2. Jednostavan ciklus hlađenja na dijagramu tlak-entalpija

The coefficient of performance for refrigeration process (COP) is calculated using Eq. [5]:

$$COP = \frac{RE}{CWI} \quad [5]$$

The pressure ratio is calculated using Eq. (6):

$$PR = \frac{P_{out}}{P_{in}} \quad [6]$$

where P_{out} and P_{in} are the pressures of refrigerant at outlet and inlet of the compressor, respectively. The Specific Power Required (SPR) per ton of refrigeration is determined using Eq. [7]:

$$SPR = \frac{3.5CWI}{RE} \quad [7]$$

RESULTS AND DISCUSSION

The results of the global warming potential (GWP) and the lower flammability limit (LFL) of the refrigerant blends at varying mass fraction of LPG are shown in Figure 3. As revealed in the figure, GWP and LFL reduce as the mass fraction of LPG increases. The refrigerant blends that have GWP below 750 and fall within the safety class of lower flammability (LFL > 3.5 % vol.) are the blends with 50 – 90 % LPG contents (RLPG50, RLPG60, RLPG70, RLPG80 and RLPG90). The results of the thermo-physical properties, LFLs and GWPs of these five refrigerant mixtures and the conventional refrigerant (R134a) are shown in Table 2.

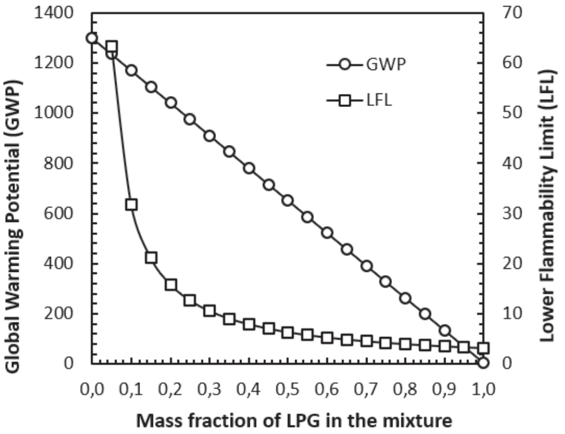

Figure 3. The GWP and LFL at varying mass fraction of LPG

Slika 3. GWP i LFL pri različitim masenim udjelima UNP-a

Table 2. Thermo-physical and ecological properties of the studied refrigerant blends

Tablica 2. Termičko-fizikalna i ekološka svojstva ispitivanih mješavina rashladnih sredstava

Properties	Refrigerants					
	R134a	**RLPG50**	**RLPG60**	**RLPG70**	**RLPG80**	**RLPG90**
Molar mass (kg/kmol)	102.03	68.51	64.28	60.54	57.22	54.25
Boiling point (oC)	-26.07	-24.98	-24.76	-24.55	-24.34	-24.11
Temperature at critical point (oC)	101.06	110.28	112.36	113.96	115.81	117.65
Pressure at critical point (MN/m2)	4.06	3.97	3.95	3.93	3.92	3.90
Density at critical point (kg/m3)	511.90	314.57	291.82	271.02	252.18	235.29
Lower flammability limit (% vol.)	–	6.33	5.28	4.53	3.96	3.52
Global warming potential (GWP)	1300	652	522	392	262	133

The plots of vapour pressure of the studied refrigerant blends at varying saturation temperature is shown in Figure 4. Similarity in saturation temperature and pressure of refrigerants are essential factors that determine their appropriateness as substitute for one another in a refrigeration system. As presented in the figure, vapour pressure rises with temperature for all the six refrigerants. The vapour pressure profiles for the five refrigerant blends are quite near and identical to that of reference refrigerant (R134a). The similarity of the temperature and pressure properties of the refrigerant blends with those of R134a proved that they can be utilized as replacements for R134a in heat pump, air-conditioning and refrigeration systems.

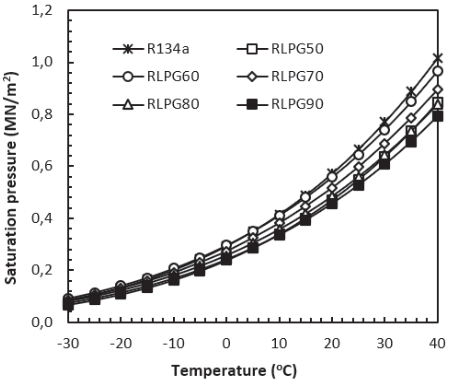

Figure 4. The pressure-temperature relation for the studied refrigerant

Slika 4. Odnos tlak-temperatura za ispitivano rashladno sredstvo

Figure 5 depicts the curves of the discharge temperature at varying evaporating temperature for the five refrigerant blends and the reference refrigerant. Low discharge temperatures are very desirable in refrigeration systems because excessive discharge (compressor's exit) temperature considerably reduce refrigeration system's performance. As presented in Figure 5, the temperature of the refrigerant at the compressor's exit decreases as the evaporating temperature rises. Relative to R134a, all the evaluated refrigerant blends maintained low discharge temperature which is advantageous to the vapour compression refrigeration system.

The trends for changes in pressure ratio of R134a and its studied alternate refrigerant blends with respect to the evaporating temperature are presented in Figure 6. As evidenced by the figure, increase in evaporating temperature lowers the pressure ratio for all refrigerants under consideration. High pressure ratio has a negative impact on how well a refrigeration system works because an increase in pressure ratio will require more work from the compressor and lower the coefficient of performance of the system. Compared to R134a, RLPG50 has an average pressure ratio of 3.4% greater while the average values of RLPG60, RLPG70, RLPG80 and RLPG90 are 5.7, 8.0, 11.6 and 15.7 % lower, respectively.

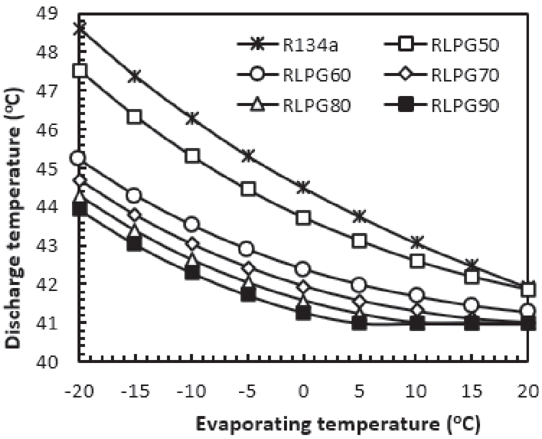

Figure 5. The curves of the discharge temperature at varying evaporation temperature

Slika 5. Krivulje temperature pražnjenja pri promjeni temperature isparavanja

The curves for variations in refrigerating effect of R134a and its studied alternate refrigerant blends with respect to the evaporating temperature are illustrated in Figure 7. The main objective of every refrigeration system is to provide a high refrigerating capacity. As presented in the figure, the refrigerating effect steadily rises with an increase in evaporating temperature. The refrigerating effects produced by the refrigerant blends are greater than that of the reference refrigerant by the average values of 4.5, 6.7, 15.8, 25.3 and 35.2 % for RLPG50, RLPG60, RLPG70, RLPG80, and RLPG90 mixtures, respectively.

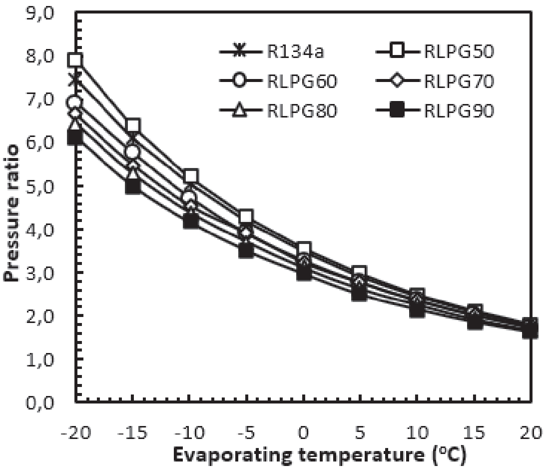

Figure 6. The influence of evaporating temperature on the pressure ratio

Slika 6. Utjecaj temperature isparavanja na omjer tlaka

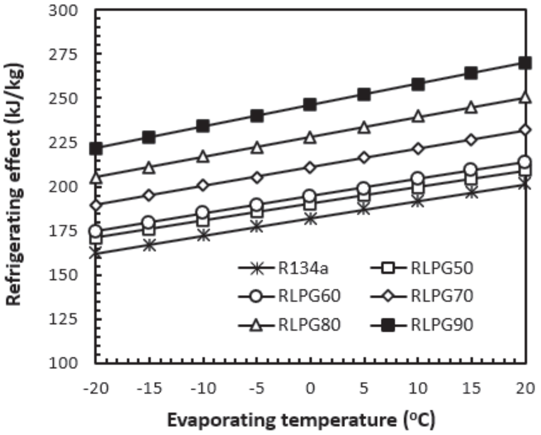

Figure 7. The changes in refrigerating effect with respect to evaporating temperature

Slika 7. Promjene u rashladnom učinku s obzirom na temperaturu isparavanja

Figure 8 shows the curves of thermal conductivity for the five investigated refrigerant blends and R134a at varying evaporating temperature. As presented in the figure, the thermal conductivity reduces as evaporating temperature increases. This is because when evaporating temperature rises, refrigerant viscosity drops and thermal conductivity is proportional to viscosity. Hence, thermal conductivity is inversely related to evaporating temperature. Compared to R134a, all the five refrigerant blends exhibited higher thermal conductivity with average values of 3.7, 1.6, 2.4, 5.4 and 8.3 % for RLPG50, RLPG60, RLPG70, RLPG80, and RLPG90 mixtures, respectively.

The curves of the specific power required (SPR) per ton of refrigeration for the assessed refrigerants at varying evaporating temperature is shown in Figure 9. Refrigerants with less power required per ton of refrigeration are energy efficient and advantageous to the environment and the refrigeration system. Similar trends in the SPR curves were seen for all the six refrigerants and the curves showed that power required decreases as evaporating temperature rises. The SPR for the five assessed refrigerant blends are quite near and identical to that of R134a. However, relative to R134a, the refrigerant blends required low power input and they are therefore more energy efficient.

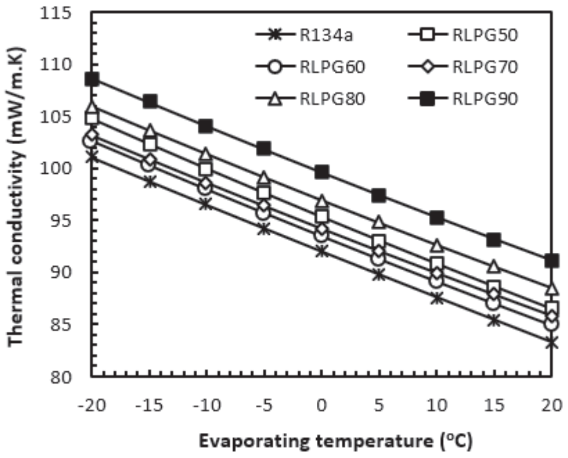

Figure 8. The changes in thermal conductivity with respect to evaporating temperature

Slika 8. Promjene toplinske vodljivosti u odnosu na temperaturu isparavanja

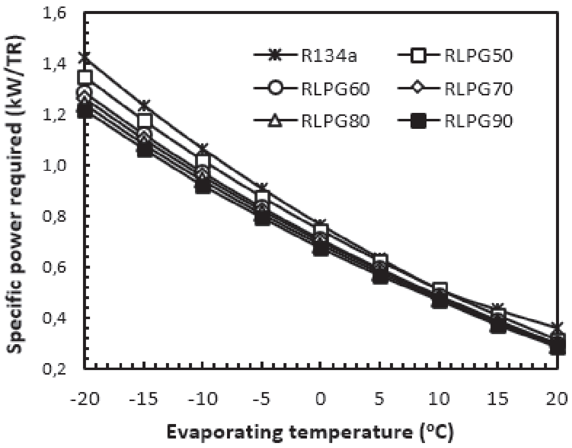

Figure 9. The specific power required at varying evaporating temperature

Slika 9. Specifična snaga potrebna pri različitim temperaturama isparavanja

Figure 10 shows how evaporation temperature affects the coefficient of performance (COP). The total cycle performance is represented by the COP. The system's COP rises as the evaporation temperature rises because the compressor work input (CWI) is reduced (Equation 5). The COPs of the investigated refrigerants are relatively comparable, as shown in Figure 10, with a little greater deviation for four of the refrigerant mixtures. The COP increases as the LPG content of the mixtures increases. The average COP of RLPG50 blend is 1.2 % lower in comparison with that of R134a, while those of RLPG60, RLPG70, RLPG80 and RLPG90 blends are higher by 3.6, 4.9, 6.7 and 8.7 % respectively.

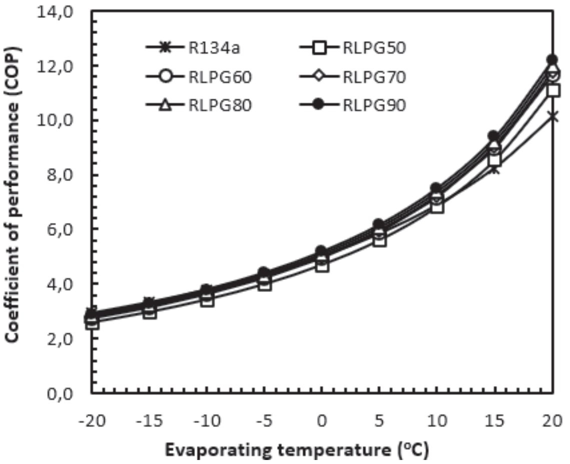

Figure 10. The coefficient of performance at varying evaporating temperature

Slika 10. Koeficijent učinka pri različitim temperaturama isparavanja

CONCLUSION

The current direct use of the highly flammable hydrocarbon gases as working fluid in refrigeration systems is not ideal and not safe for many air-conditioning and refrigeration applications. Liquefied petroleum gas (LPG), like other hydrocarbon gases, has good thermo-physical and ecological properties but its high flammability has hindered its wide acceptance as refrigerant. LPG is mixed with R134a at various composition ratios in order to adjust the properties of the components and to produce new low-GWP refrigerants with lower flammability. The theoretical performance of low flammable and ecological-save mixtures of LPG and R134a as working fluid in refrigeration systems is investigated in this paper and the followings are the conclusions drawn:

- The results of GWP and LFL showed that the refrigerant blends that fall within the safety class of lower flammability (LFL > 3.5 % vol.) and have GWP below 750 are RLPG50, RLPG60, RLPG70, RLPG80 and RLPG90. These five blends are selected for performance analysis in a refrigeration system.

- The temperature-pressure profiles of the five refrigerant blends are quite identical to that of the reference refrigerant (R134a) between the temperature ranges of -30 to 40 oC which showed that they can be utilized as its replacements.
- The thermal conductivity and the refrigerating effects produced by the refrigerant blends are greater than those of the reference refrigerant (R134a) and average refrigerating effects of 4.5, 6.7, 15.8, 25.3 and 34.9 % higher were obtained for RLPG50, RLPG60, RLPG70, RLPG80 and RLPG90 blends, respectively.
- Relative to R134a, all the five selected refrigerant blends exhibited advantageous lower pressure ratio, discharge temperature and specific power required.
- The coefficient of performance (COP) increases as the LPG content of the mixtures increases. With respect to R134a, the average COP of the RLPG50 mixture is 1.2% lower, while the COPs of RLPG60, RLPG70, RLPG80, and RLPG90 mixtures are higher by 3.6, 4.9, 6.7, and 8.7%, respectively.
- In general, the study has shown that the five evaluated refrigerant mixtures are ecologically save with relatively low global warming potential, low flammability, low specific power required and four of the mixtures exhibited high COP. These are clear indication that they are more energy efficient than the reference refrigerants (R134a) in refrigeration systems.

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TEORIJSKA ANALIZA NISKOZAPALJIVIH I EKOLOŠKI PRIHVATLJIVIH SMJESA UKAPLJENOG NAFTNOG PLINA I TETRAFLUORETANA KAO HLADILA

SAŽETAK: Ukapljeni naftni plin, poput drugih plinovitih ugljikovodika, ima dobra termičko-fizikalna i okolišna svojstva, ali visoka zapaljivost ograničava njegovu izravnu uporabu za hlađenje. Studija istražuje termodinamička svojstva nisko zapaljivih i ekološki prihvatljivih smjesa ukapljenog plina i tetrafluoretana kao hladila u sustavu hlađenja. Izračuni rezultata potencijnog globalnog zatopljenja (GWP) i niske zapaljivosti pokazuju da smjese koje sadrže 50 – 90 % ukapljenog plina imaju GWP ispod 750 te su u sigurnosnom razredu niže zapaljivosti, od kojih je pet odabrano za daljnju analizu. Rezultati pokazuju da su toplinska vodljivost i učinci hlađenja pri uporabi odabranih smjesa veći od referentnog hladila (R134a). Nadalje, odabrane smjese imaju prikladno niži tlak, niže izlazne temperature i specifične snage nego R134a hladilo. Četiri smjese zahtijevale su nižu snagu i pokazale su viši koeficijent učinkovitosti što jasno dokazuje da su energetski učinkovitije od R134a u sustavima hlađenja.

Ključne riječi: *smjese, ekološki prihvatljiva hladila, zapaljivost, ukapljeni naftni plin, R134a*

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