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Thermodynamic Analysis of Flammability and Ecological Safety Levels of Liquefied Petroleum Gas and R134a Refrigerant Mixtures for Refrigeration Applications

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Abstract

The flammability of hydrocarbons such as propane, butane, and liquefied petroleum gas (LPG) affects their wide acceptance as working fluids in air-conditioning, refrigeration, and heat pump systems. Mixing LPG with non-flammable R134a will produce a mixture with lower flammability. Therefore, this paper investigated the physical properties, flammability levels, and global warming potentials of LPG and R134a mixtures. The results obtained revealed that the mixtures of LPG and R134a exhibit low critical pressures and higher critical temperatures that are reasonably higher than the ambient temperature, which are beneficial to the cooling effect per unit volume of the refrigeration system. The mixture's boiling point temperature and global warming potential (GWP) reduce as its flammability increases, which will require a diligent trade-off among these three properties of the mixture. The percentages of LPG in the mixture that produced satisfactory GWPs with respect to the EU's F-gas regulation and those that falls into the safety class of mild flammable range from 45 to 100% and 5 to 90%, respectively. Conclusively, this study reveals that novel ecologically acceptable refrigerants with reduced flammability could be obtained from the mixtures of LPG and R134a refrigerants within the range of 45 to 90% content of LPG.

Nomenclature

$a_1, a_2, ., a_n$:	Percentage of mole of each refrigerant (%)
BPT:	Boiling point temperature (°C)
C_{xC}:	Number of carbon atoms in the compound
CC:	Cooling capacity per unit volume (kJ/m^3)
CE:	Cooling effect (kJ/kg)
COP:	Coefficient of performance
CPT:	Critical point temperature
GWP:	Global warming potential

h_1, h_2 and h_4:	Enthalpies at the compressor, condenser, and evaporator inlets, respectively
H_{xH}:	Number of monoatomic hydrogen atoms in the compound HC: Hydrocarbon
LEL:	Lower explosive limit (% Vol.)
LFL:	Lower flammability limit (% Vol.)
LPG:	Liquefied petroleum gas
M_i:	Mass Fraction of the component ‘i’ in mixture
n:	Number of refrigerants or component in the mixture
NBP:	Normal boiling point (^oC)
ODP:	Ozone depletion potential
PRPTC:	Power required per tonne of cooling (kW/TR)
R134a:	1,1,1,2-Tetrafluoroethane refrigerant
SWI:	Specific work input (kJ/kg)
V_{air}:	Coefficient of air quantity at the stoichiometric composition
X:	Mole fraction of LPG
ρ_1:	Vapour density at the compressor’s inlet (kg/m^3)

1. Introduction

Refrigerants are the operating fluids in the heat pumps, air-conditioners and refrigerators. In vapour compression refrigeration systems, they undergo a repeated phase transition from liquid to vapour and back again to liquid. Due to their flammability, toxicity, ozone depletion and global warming, refrigerants are subject to strict regulation. Since about five decades ago, concerns about how refrigerants affect the environment and climate change have been on the increase. This has led to the rising worldwide pressure to ban ozone depleting substances and minimize the use of high global warming potential (GWP) refrigerants in a variety of refrigeration applications [1, 2].

Hydrocarbons have made a comeback as working fluids in refrigeration applications, attributed to rising issues about the ecological effect of CFC, HCFC and HFC refrigerants. Hydrocarbons 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. They are now commonly used in vapour compression systems in recent years [3]. Together with their mixtures, they show good potentials as replacements for high-GWP conventional refrigerants (R134a and R22).

According to Razzaq et al. [4], hydrocarbon (HC) refrigerants such as ethane (R170), ethylene (R1150), propane (R290), butane (R600), iso-butane (R600a), Liquefied Petroleum Gas (LPG), and their mixtures have excellent thermodynamic properties, good system performance, zero ODP, no fluorine (thus low GWP), high latent heat of vaporization and are readily available. However, these ecological advantages come with their own drawbacks. Hydrocarbons are highly flammable; safety might be quickly undermined if proper precautions are not taken. The safety risk of hydrocarbons in refrigeration applications is a concern that must be addressed.

The use of refrigerant mixtures is gaining popularity in refrigeration and air-conditioning applications because they allow for greater flexibility in the search for new ecologically safe alternatives that match the desirable qualities of existing ecologically unfavourable refrigerants [5]. Mixtures of two or more pure HC refrigerants have exhibited good performance in the existing refrigeration systems without modifications. Oyelami and Bolaji [6], investigated the performance of a household refrigerator using liquefied petroleum gas as working fluid. The study showed that the liquefied petroleum gas has superior results in terms of power use and COP than R134a refrigerant in the system. However, liquefied petroleum gas also known as LPG refrigerant is a combination of hydrocarbon gases and is highly flammable which has limited its broad acceptance and usage in refrigeration systems [7].

Flammability refers to a substance’s ability to self-propagate a flame over a particular distance. A refrigerant's flammability refers to its ability to ignite and produce fire. Once flammable refrigerants are discharged into the air, they pose an immediate threat and at atmospheric pressure, the refrigerant can react with air, burn or combust and resulting in a flame or explosion [8]. The use of flammable refrigerants is limited to controlled conditions with monitors, sufficient ventilation and explosion-proof equipment due to the obvious dangers [9]. The lower explosive limit (LEL) or lower flammability limit (LFL) of any flammable material is a quantity in air that is commonly given as a percentage of volume. Any combination of fuel and air below the LFL is too lean to allow combustion [10].

One of the properties that make R134a a popular alternative refrigerant in heat pump, air-conditioning and refrigeration systems is that it is non-flammable. However, LPG consists of extremely combustible hydrocarbon gases. In order to overcome this problem, several approaches are being proposed by some researchers [11-13] to address the potential for combustion and safety issues with hydrocarbon refrigerants, including the use of improved compact heat exchangers, system design optimisation, system charge reduction, and the establishment of guidelines and standards for safety measures.

Hydrocarbon refrigerants can be mixed with HFC refrigerants such as tetrafluoroethane (R134a) and pentafluoroethane (R125) to adjust their properties and produce new ecologically acceptable refrigerants with reduced flammability. Some other flame-retardant refrigerants are carbon-dioxide (CO_2), heptafluoropropane (R227ea), and trifluoroiodomethane (R13I1) [14, 15]. For safety concerns in storage, processing and handling, refrigerant flammability properties are crucial. Also, to determine the working qualities of the system's refrigerant and to establish the suitability of refrigerant according to the intended working conditions, physical features of refrigerants are very essential. Therefore, this paper evaluates the physical properties, flammability levels and the global warming potentials of LPG and R134a mixtures.

2. Materials and Methods

2.1 Components of the Investigated Refrigerant Mixtures

Mixtures are made up of components that are evenly spread and cannot be separated physically. They allow for the modification of capacity by altering the element makeup. This paper theoretically investigated the mixing of different percentages of R134a refrigerant with LPG in order to lower flammability and global warming potentials of the resulting mixtures. The flammability limits of LPG and R134a refrigerant mixtures were computed at different composition ratios using appropriate equations and REFPROP software version 10.0 [16]. The investigated mixtures are those with a 5 percent increase in LPG mass ratio. These mixtures are further designated as RMLPG05, RMLPG10, RMLPG15 up to RMLPG95 making a total number of nineteen mixtures. The last two digits referred to the percentage of LPG in the mixture.

2.2 Determination of Boiling Point of Refrigerants

Normal boiling point (NBP) of some common refrigerants can be obtained from literature [16, 17]. The average boiling point (ABP) of refrigerant mixtures at a pressure of 1.013 bar is given in Equation 1:

$$ABP = \sum_{i=1}^{n} x_i T_{bi} \tag{1}$$

where, x_i stands for the mass portion of the constituent 'i' in the blend; T_{bi} stands for the normal boiling point of the constituent 'i' in the mixture; and n stands for the number of the constituent in the mixture.

2.3 Computation of Lower Flammability Limit (LFL) of Hydrocarbon Refrigerants

A reaction of one-step stoichometric combustion of a hydrocarbon refrigerant with chemical formula of $C_{xC}H_{xH}$ is given in Equation 2 and 3 [18]:

$$C_{x_C}H_{x_H} + V_{air}\left(O_2 + 3.75N_2\right) \rightarrow x_C CO_2 + \frac{x_H}{2}H_2O + 3.76V_{air}N_2 \tag{2}$$

$$V_{air} = x_C + 0.25x_H \tag{3}$$

where V_{air} stands for the coefficient of air quantity at the stoichiometric composition; C_{xC} stands for the number of carbon atoms; and H_{xH} stands for the number of monoatomic hydrogen atoms in the hydrocarbon refrigerant. The lower flammability limit (LFL) is given as in Equation 4:

$$LFL = \frac{1}{1 + 4.76V_{air}} \tag{4}$$

2.4 Determine the Lower Flammability Limits of Refrigerant Mixtures

After the flammability limits of all the components in the mixture have been computed, the mixture's lower flammability limit (LFL_{Mix}) is determined using Equation 5, 6 and 7 (Le Chatelier's rule). When handling different mixes of fuel, oxygen, and solutions, this technique offers a greater degree of adaptability [10, 19, 20].

$$\frac{1}{LFL_{Mix}} = \frac{a_1}{LFL_1} + \frac{a_2}{LFL_2} + \ldots\ldots + \frac{a_n}{LFL_n} \tag{5}$$

$$\frac{1}{LFL_{Mix}} = \sum_{i=1}^{n} \frac{a_i}{LFL_i} \tag{6}$$

$$LFL_{Mix} = \left(\sum_{i=1}^{n} \frac{a_i}{LFL_i} \right)^{-1} \tag{7}$$

where a_1, a_2, ….., a_n are the percentages of mole of each refrigerant that formed the blend; n stands for the number of refrigerants that formed the blend; and LFL_i stands for the lower flammability limit of the component 'i' in the mixture.

2.5 Safety Classifications of Refrigerants

The three major classes of flammability extracted from ASHRAE Standard 34 are shown in Table 1: Class 1 includes incombustible refrigerants that do not spread a flame when tested in accordance with the standard; Class 2 includes refrigerants with low flammability; another group is carved out of this class, which includes refrigerants with mild flammability (Class 2L); and Class 3 consists of extremely flammable refrigerants such as hydrocarbons [21]. Toxicity of refrigerants is indicated in two ways; lower and higher toxicities designated as Class A and Class B, respectively.

Table 1 *Refrigerant flammability classifications [21]*

Flammability class		Lower flammability limit (% Vol.)	Toxicity	
			Lower (A)	Higher (B)
Class 1	Non-Flammable	No flame propagation	A1	B1
Class 2L	Mildly flammable	≥ 6.2	A2L	B2L
Class 2	Low flammable	≥ 3.5	A2	B2
Class 3	Super flammable	≤ 3.5	A3	B3

2.6 Theoretical Determination of the GWP of Refrigerant Mixtures

The Kyoto Protocol and the EU's F-gas regulations call for the use of refrigerants with GWPs of less than 150 for refrigerators, freezers and mobile air-conditioners, and less than 750 for split refrigeration systems under 3 kg refrigerant charge [22]. Table 2 shows the extract from the EU F-Gas Policy review 2022 that rule out the application of high-GWP chemicals in various equipment [23]. Refrigerant mixture is produced by combining two or more pure fluid refrigerants. Adding the GWP of each component in proportion to their mass yields the GWP of the mixture. Therefore, the mixture's global warming potential (GWP_{Mix}) is given in Equation 8 [24]:

$$GWP_{Mix} = \sum_{i=1}^{n} M_i \left(GWP_i \right) \tag{8}$$

where, M_i stands for the mass portion of the constituent 'i' in the blend; GWP_i stands for the global warming potential of the constituent 'i' in the mixture; and n stands for the number of the constituent in the mixture. The GWPs of the components of the mixtures are obtained from literature [25].

Table 2 *Excerpt from the EU F-Gas Policy review 2022 [23]*

Equipment	**GWP**	**Prohibition date** (Day/Month/Year)
Refrigerators and freezers for commercial use	≥150	01/01/2024
Small refrigerators and freezers	≥150	01/01/2025
Plug-in room air-conditioning and heat pump	≥150	01/01/2025
Split systems under 3 kg refrigerant charge	≥750	01/01/2025
Split systems of up to 12 kW rated capacity	≥150	01/01/2027

3. Results and Discussion

The average boiling point (ABP) of the refrigerant mixture at varying percentage content of LPG in the blend is shown in Figure 1. As shown in the figure, the curve obtained followed a linear trend in which the ABP of the LPG and R134a mixture reduces as the percentage of LPG in the mixture increases. However, increase in the quantity of LPG will increase the flammability of the mixture. This required diligent trade-off between flammability and low boiling point of the refrigerant blend. This result is in line with that of Molnarne and Schroeder [26], which showed a linear relationship between the boiling point temperature of hydrogen/air mixtures and the mole fraction of hydrogen in the mixtures. The linear equation is also in agreement with the model equation obtained by Hristova [27] for the flash point temperature of binary mixtures of flammable and non-flammable gases. The model, as given in Equation 9 was used to compute the boiling point temperature of the mixture at varying percentages of LPG in the mixture, and the comparison of the data obtained with the initial data showed $\pm 6.7 \times 10^{-4}$ deviation.

$$ABP = -0.5949x - 26.006 \tag{9}$$

where ABP is the average boiling point temperature of the refrigerant blend; and x stands for the fraction of LPG in the blend.

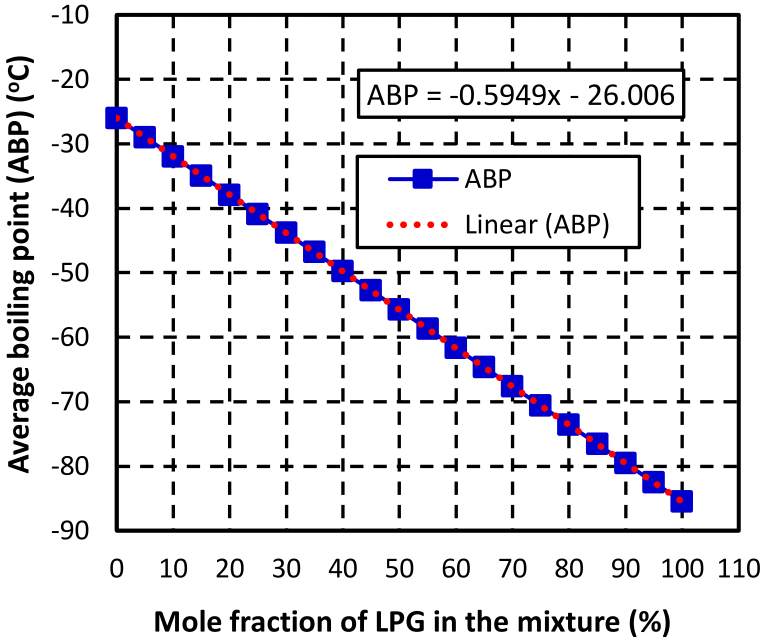

Fig. 1 *The variation of the mixture's boiling point temperature with the mole fraction of LPG in the blend*

The variation of the mixture's critical point temperature with respect the percentage content of LPG in the blend is shown in Figure 2. A refrigerant's critical point temperature is the optimum temperature in its saturation curve at which it cannot be liquefied regardless of its vapour pressure. As illustrated in the figure, the curve obtained followed a polynomial pattern of the second order and the critical point temperature of the refrigerant mixtures reduces as the percentage content of LPG in the blend increases. This interpretation is consistent with the findings of Bell et al. [28], which showed a quadratic curve for the critical point temperature versus mole fraction of R1234yf in the binary mixtures of carbon-dioxide with R1234yf, and R32 with R1234yf. The model, as given in Equation 10 was used to compute the critical point temperature of the mixture at varying percentages of LPG in the mixture, and the comparison of the data obtained with the initial data showed ±0.099 deviation. Lower critical point temperature but reasonably higher than ambient temperature is advantageous in refrigeration systems [29].

$$CPT = 0.0136x^2 - 1.9103x + 98.586 \tag{10}$$

where CPT is the critical point temperature of the refrigerant mixture.

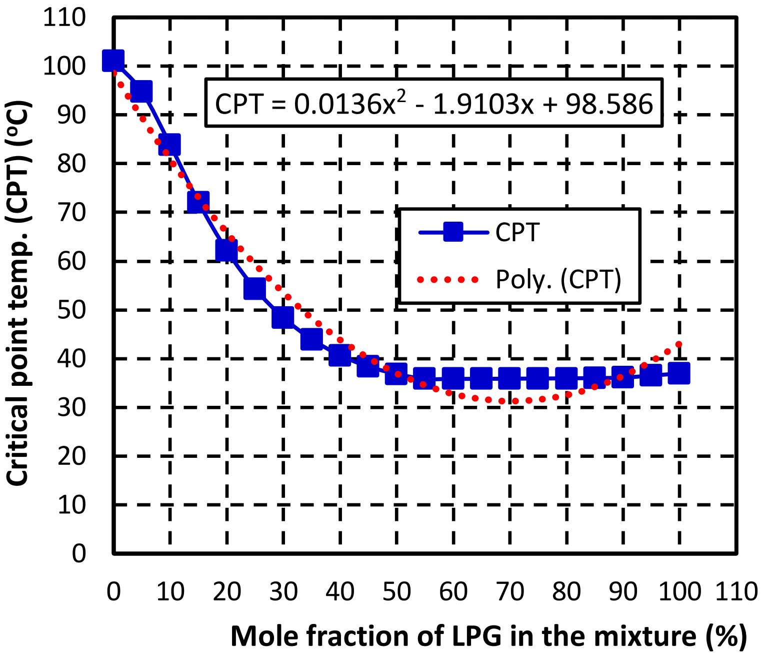

Fig. 2 *The variation of the critical point temperature of the refrigerant blend with the percentage content of LPG in the blend*

The variation of the blend's critical point pressure with respect to the percentage content of LPG in the blend is shown in Figure 3. According to El-Banbi et al. [30], the refrigerant's critical point pressure should be greater than the condenser pressure, if not, the condensation zone will shrink and there will be heat rejection. As shown in Figure 3, the critical pressure of mixture increased sharply up to its maximum point at 15 % LPG content with gradual reduction after the optimum critical pressure. The relationship between LPG mole fraction and critical pressure of the mixtures produced a best fit curve that followed a polynomial pattern of the fifth order as given in Equation 11. This result aligns with that of Zhang et al. [31], which showed a polynomial relationship curve for the critical point pressure versus mole fraction in binary mixtures containing hydrocarbon. At different proportions of LPG in the mixture, the critical point pressures were computed using the mathematical model, and the results obtained were compared with the baseline data, which revealed a ±0.045 deviation.

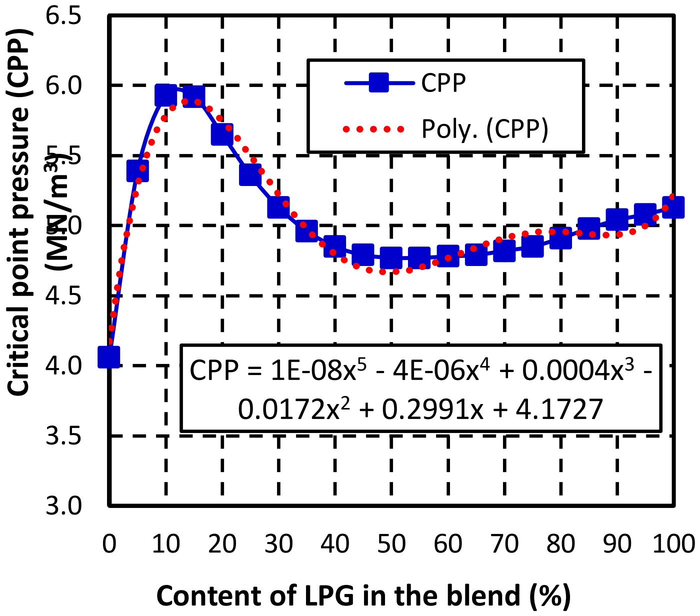

Fig. 3 *The variation of the critical point pressure of the refrigerant blend with the mole fraction of LPG*

$$CPP = (1 \times 10^{-8})x^5 - (4 \times 10^{-6})x^4 + 0.0004\,x^3 - 0.0172\,x^2 + 0.2991\,x + 4.1727 \qquad (11)$$

where CPP is the critical point pressure of the refrigerant mixture.

Figure 4 shows the curves of saturation pressure versus saturation temperature for the investigated refrigerant blends in the operating temperature range of -20 to 20 °C. Temperature-related changes in saturation vapour pressure are shown according to the graphic. A decrease in pressure will decrease the compressor work input, but it will also decrease the specific cooling capacity; hence, the extent of the decrease in these two parameters should be considered to determine which one will yield a better coefficient of performance (COP). This required diligent trade-off between the desirable low pressure and low GWP of the refrigerant mixture. The physical, safety and environmental properties of the studied refrigerant mixtures are shown in Table 3.

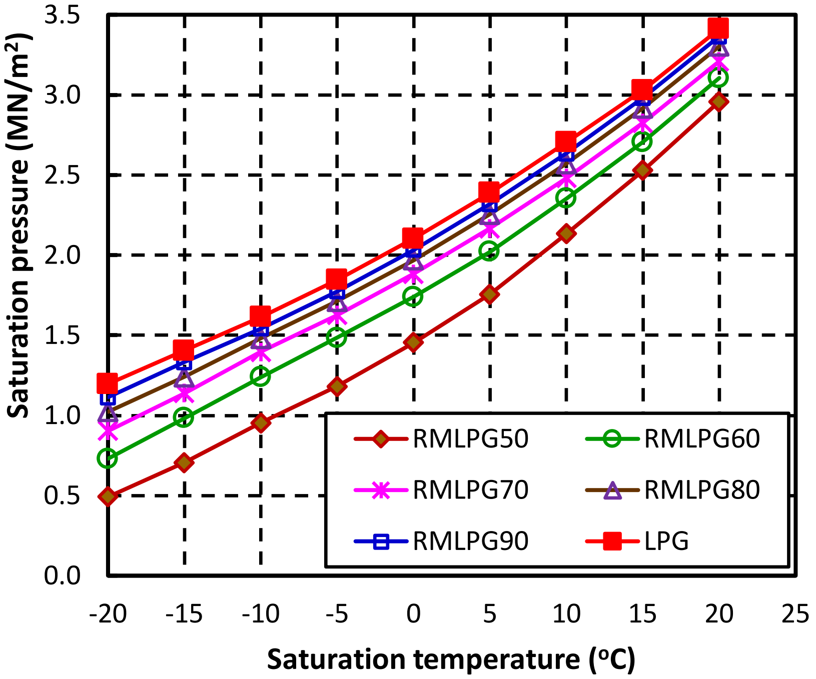

Fig. 4 *Saturation pressure versus saturation temperature of the refrigerant blends*

Table 3 *Physical and environmental characteristics of the investigated refrigerant blends*

Properties	Refrigerants					
	LPG	RMLPG50	RMLPG60	RMLPG70	RMLPG80	RMLPG90
Molar mass (kg/kmol)	30.83	47.35	42.77	38.99	35.83	33.14
Density at critical point (kg/m³)	206.07	314.57	291.82	271.02	252.18	235.29
Pressure at critical point (MN/m²)	5.13	4.77	4.78	4.79	4.91	5.04
Temperature at critical point (°C)	41.96	41.84	40.85	40.89	40.96	41.11
Boiling point (°C)	-85.46	-55.75	-61.70	-67.65	-73.60	-79.55
Ozone depleting potential (ODP)	0	0	0	0	0	0
Global warming potential (GWP)	3.10	651.50	521.82	391.14	261.46	132.78

Figure 5 shows the global warming potential (GWP) of the refrigerant mixture at varying mole fraction of LPG in the mixture. GWP of a refrigerant is a typical way to express its effect on the climate. It is the measure of the amount of infrared radiation absorbed by a refrigerant which is compared to the impact of carbon dioxide with GWP of 1. Figure 5 exhibits a linear trend, as given in Equation 12 between GWP and the percentage of LPG in the mixture. This outcome is consistent with that of Zheng et al. [32], who demonstrated a linear relationship between the variation of GWP value and the mass fraction of R32 in a combination of R32 and R1234yf. The model equation's output was compared to the baseline data, and the findings indicated a ±0.017 deviation. As shown in Figure 5, the percentages of LPG in the mixture that produced GWPs which satisfy the EU's F-gas regulations (GWPs of less than 150 for refrigerators, freezers and mobile air-conditioners, and less than 750 for split air-conditioning systems under 3 kg refrigerant charge) range from 45 to 100 %.

$$GWP = -12.968\,x + 1299.9 \tag{12}$$

where GWP stands for the mixture's global warming potential.

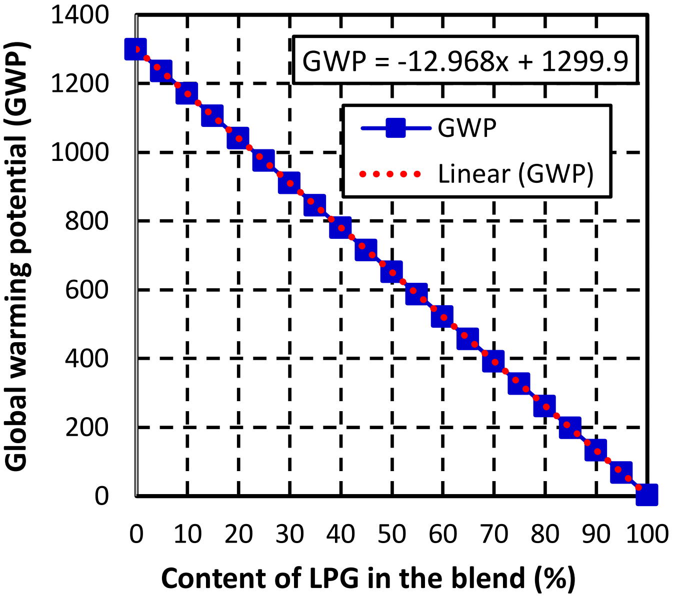

Fig. 5 *The GWP of the refrigerant blend with the mole fraction of LPG in the blend*

The variation of the blend's lower flammability limit (LFL) with respect to the percentage content of LPG in the blend is shown in Figure 6. The fact that R134a is non-flammable is one of the qualities that make it a good combination with highly flammable LPG. The mixtures of LPG and R134a are considered to produce new low-GWP refrigerants with reduced flammability [33]. The relationship between LPG mole fraction and LFL of the mixtures produced a best fit curve that followed a polynomial trend of the fifth order as in Equation 13. This result agrees with of Lv et al. [34], which established a polynomial curve for the LFL in relation to the mole percent of R32 in the binary combinations of R134a and R32. The model equation showed a ±0.017 deviation in comparison with baseline data. With reference to Table 1 and Figure 6, the percentages of LPG in the mixture that fall into safety class of "mild flammable" (LFL > 3.5 % vol.) range from 5 to 90%. Compare with the results in Figure 4 and 5, ecologically acceptable refrigerants with reduced flammability could be sought for in the mixtures of LPG and R134a refrigerants within the range of 45 to 90 % content of LPG.

$$LFL = \left(-2\times10^{-7}\right)x^5 + \left(6\times10^{-5}\right)x^4 - 0.0066\,x^3 + 0.3546\,x^2 - 9.0467\,x + 97.616 \tag{13}$$

where LFL stands for the lower flammability limit of the refrigerant blend.

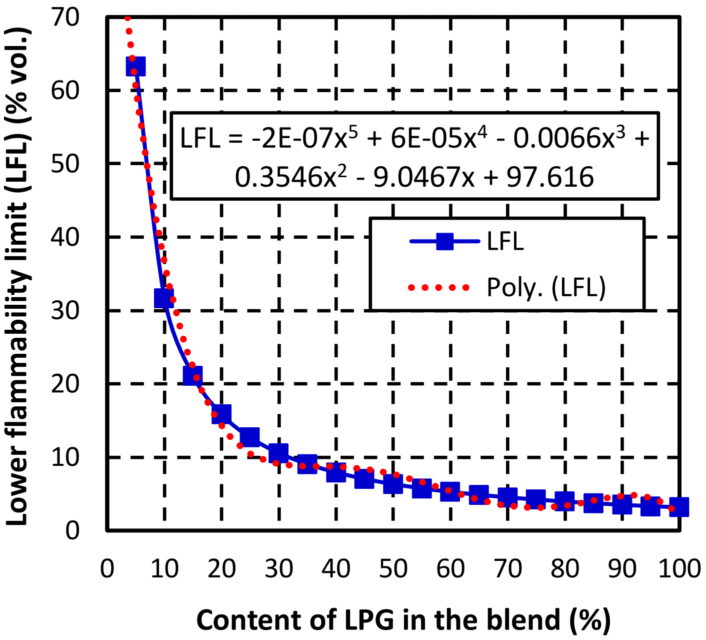

Fig. 6 *The variation of blend's LFL with the percentage content of LPG in the blend*

4. Conclusion

Liquefied petroleum gas known as LPG is a combination of highly combustible hydrocarbon refrigerants which affected its wide acceptance as working fluid in air-conditioning, refrigeration and heat pump systems. This paper investigated the mixing of different percentages of R134a refrigerant with LPG in order to lower flammability and global warming potentials. The physical properties, flammability levels and the global warming potentials (GWPs) of the resulting mixtures were evaluated and the followings are the conclusion drawn from the computational analysis:

(i) The average boiling point of the LPG and R134a mixture reduces as the flammability of the mixture increases which required diligent trade-off between flammability and low boiling point temperature of the refrigerant mixture.
(ii) The mixtures of LPG and R134a yielded lower critical point temperatures which are reasonably higher than ambient temperature. The low critical point temperature will increase the cooling effect per unit volume.
(iii) The mixtures produced higher critical pressures that are advantageous to the refrigeration system and the curve of critical pressure versus LPG mole fraction followed a fifth order polynomial trend.
(iv) The GWP of the mixture reduces as its flammability increases which also required diligent trade-off between flammability and low-GWP of the refrigerant mixture.
(v) The percentages of LPG in the mixture that produced satisfactory GWPs with respect to the EU's F-gas regulation range from 45 to 100 %.
(vi) The higher the percentage of LPG in the mixture, the lower the flammability limit (LFL) and the more flammable the mixture is. The relationship between LFL and LPG mole fraction produced a best fit curve that followed a fifth order polynomial trend.
(vii) The percentages of LPG in the mixture that fall into safety class of "mild flammable" (LFL > 3.5 % vol.) range from 5 to 90 %.

In general, the novel ecologically acceptable refrigerants with reduced flammability could be obtained from the mixtures of LPG and R134a refrigerants within the range of 45 to 90 % content of LPG. The development of these non-flammable environmentally friendly refrigerant mixtures will promote personal and environmental safety, protect our eco-system, and enhance the quality of life and well-being of citizens.

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Conflict of Interest

The authors state that their publishing of the paper does not include any conflicts of interest.

Author Contribution

The authors confirm contribution to the paper as follows: ***study conception and design:*** *first and third author;* ***study supervision and administration:*** *second and fourth author;* ***data collection:*** *first, fourth and fifth authors;* ***analysis and interpretation of results:*** *first, third and sixth authors;* ***draft manuscript preparation****: first, third and fifth authors. After reviewing the research results, each author gave their approval to the manuscript's final draft.*

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