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Overview of the Use of Hydrocarbon Refrigerants in Air Conditioning Systems

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Refrigerant used in air conditioning and refrigeration (ACR) industries has come to a full circle since the beginning of the industrial revolution. With concerns on issues relating to the environment, such as the global warming and climate change, caused by the use of harmful gases as refrigerants. Therefore, it is of outmost importance to mankind, to find suitable alternative gases as refrigerants, which are not detrimental to the ozone layer and does not adversely affect the climate. In view of this, hydrocarbons (HC) which were previously used as refrigerants are being reintroduced gradually because its lower GWP and which does not inhibit ozone layer, as replacement refrigerants. ACR industry players have blended in by introducing some new equipment and components that specifically designed for HC use. Malaysia too has targeted a 10 % reduction in hydrochlorofluorocarbons (HCFC) consumption, commencing in 2016; with the banning of ACR equipment of 7.03 kW and below from using HCFC 22 [chlorodifluoromethane, ($CHClF_2$)]. Instead, hydrofluorocarbon (HFC) 410a [50 % difluoromethane (CH_2F_2) and 50 % pentafluoroethane (CHF_2CF_3)] is being introduced as replacement for HCFC 22. In fact, in other countries, the use of HFC, has already been phased down. HC has good potential to replace HCFC completely as refrigerant in the future; its performance, matches that of HCFC and the flammability issue can be overcome, with the use of effective and safety enhanced design. Their use can be facilitated further with the adaptation of a specific standard and by proper enacted legislation.

1. Introduction

Malaysia having committed to the reduction in the consumption of HCFC and other ozone depleting substances (Montreal Protocol) has to adhere to the Phase-out Management Plan (HPMP), set by Department of Environment (DOE) Malaysia, with the reduction of 10 %, 35 %, 67.5 % for the years, 2015, 2020, 2030 and 2040. A total phase out beyond 2040 has been planned, with only minimal quantities usage for servicing purposes (DOE, 2014). The local ACR industry too, must be in compliance to the ban imposed by DOE for the manufacture, importation and installation of split type air condition units of 7.03 kW and below.

The ACR players have resorted to convert from HCFC 22 to HFC 410a, due to its "0" ozone depletion potential (ODP) although having a Global Warming Potential (GWP) of 2100 as compared to HCFC 22, which is at 1790 (ASHRAE, 2013). Another suitable alternative to HCFC 22 is HFC 32 [difluoromethane, (CH_2F_2)], a mildly flammable refrigerant which has a low GWP (650) and need lower volume to produce the same refrigeration effect as HFC 410a (ASHRAE, 2013). However, the new equipment operates at higher pressures; incur additional operating costs, to the end user.

Domestic refrigerators sold in Malaysia are charged with iso-butane (HC 600a) due to its similar thermodynamic properties it has with the HFC 134a [1,1,1,2-tetrafluoroethane (CH_2FCF_3)] refrigerant. In the insulation materials cyclo-pentane is used as its blowing agent.

In the air conditioning sectors, there is no equipment manufactured or imported, that suited for the HC use. In the European Union (EU), HFC are currently being phased down due to its high GWP values. Elsewhere in China, Yang and Wu (2013) have reported that HC 290 (propane) split air conditioners (ACs) produced by GREE have certification by, Verband Deutscher Elektrotechnikere (VDA) and Technisher Uberwachungs Verein (TUV). These ACs are for export to the EU member nations, as for China, does not allow these equipment to be sold locally.

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In India, Rajadhyaksha et al. (2015) reported that since 2012, Indian ACR manufacturer, Godrej & Boyce has sold and installed around 100,000 units of HC 290 split ACs in Indian market. Complaints of insufficient cooling were about the same as that of split ACs using R 22, and HC leaks from outdoor units were about 0.3 % but none was ever reported indoors. In the EU the F gas regulations (EC Regulation No 517/2014) were enforced, and prompted the reduction in the use of HFCs. Malaysia encourages the new ACR equipment to use HFC 410a as replacement for HCFC 22. The Significant New Alternative Policy (SNAP) of the U.S. Environmental Protection Agency (EPA) on March of 2015 has also approved low GWPs refrigerants ranging from 3 to 675, to replace refrigerants with GWP ranging from 1,400 to 4,000 (US E.P.A., 2015). Malaysia has potential to utilize HC as refrigerants extensively in the future. This is because it is produced locally and available at relatively low cost. The only disadvantage being its safety aspects, which can be addressed by adopting safety standards and regulations practiced in developed countries.

2. A Brief History of Refrigerants

Stored ice was probably the very first attempt to provide refrigeration to prevent food from turning bad. Perkins created a vapour compression cycle using ethyl ether ($C_2H_5OC_2H_5$) (Calm and Didion, 1998). Other refrigerants in vapour compression cycles includes, methyl ether, carbon dioxide, carbon tetrachloride, ammonia, isobutene, propane, dielene and petrol (gasoline). Industrial accidents were prone due to leaks from refrigeration circuits.
Due to the combustibility and toxicity of early refrigerants, Thomas Midley Junior and Albert Leon synthesised, dichlorodifluoromethane (CFC12) in 1930s and marketed it through Fridgaire, of General Motors. It was known as Freon 12. The ozone depletion potential (ODP) of Freon12, is 1 and all other refrigerants use CFC12 as a reference point for ODP (Calm and Didion, 1998).
About 40 y later, Molina and Rowland (1974) published an article in Nature periodical regarding the depletion of the stratospheric ozone was caused by the use of CFC as refrigerant. This article prompted the quest for alternatives to CFC and HCFC, thus the commencement of the planned phase out of these refrigerants began through the ratification of the Montreal Protocol in 1989. (Calm and Didion 1998).
Chemical companies too provided solutions in the form of the use of HFCs, but were later curtailed due to their high GWP values and will also be phased down by the ratification of the Kyoto Protocol (1997). In the EU, the commonly known F-Gas regulations (EU Regulation No.517/2014) prohibits the use of refrigerants with high GWPs.
In the quest for low GWP and ‘zero’ ODP refrigerants, companies synthesised hydrofluoroolefins (HFO). HFO 1234 yf [2,3,3,3-tetrafluoro-1-propene ($CF_3CF{=}CH_2$)] (GWP =4) is to replace HFC 134a (GWP = 1430) in all vehicles manufactured in EU as of 1st January 2017 (EC Directive 2006/40/EC). Others blend existing HFC and HFO to overcome this directive. Daikin encourages the use of HFC 32 (GWP = 650) as the main refrigerant to replace HCFC 22. HFC 32 is categorised by ASHRAE standard 34 and ISO 817 as a mildly flammable refrigerant.
Most Scandinavian countries had already used natural refrigerants to replace synthesised ones, as early as in the 90s. Ammonia (R 717) used in ice making plants and water (R 718) is widely used as a refrigerant in absorption chillers, CO_2 (R 744) in trans-critical refrigeration cycles which was conceptualised by Lorentzen (1994). He also predicted correctly the reintroduction of natural refrigerants in the foreseeable future to replace synthesised substances. HC refrigerants which include HC 290 [propane, ($CH_3CH_2CH_3$)], HC 600a [isobutane, ($CH(CH_3)_3$)], HC 1270 [propylene, ($CH_3CH{=}CH_2$)] and HC 170 [ethane, (CH_3CH_3)] have now impacted as efficient alternative to replace HCFCs and HFCs, although HCs are categorised as A3 by ASHRAE 34 and ISO 817 because of their inherent flammable property.
Refrigerant usage in ACR industries had witnessed an about turn; during the early 1930s, natural refrigerants were widely used before the introduction of CFC, HCFC, HFC, HFO, and now are reintroduced due to climate change issue. The limitation of their use back in the 1930s was due to unavailability equipment, standards and legislation, which made these substances, considered unsafe to operators, buildings and occupants alike. Today we have ATEX (*Appareils destinés à être utilisés en ATmosphères EXplosibles*) certified hermetically sealed compressors, combustible gas leak detectors, standards and guides to provide a safer environment to use these flammable gases.

3. Hydrocarbon Refrigerants

Greenpeace has categorised HC as one of the ‘gentle five’ refrigerants, which include carbon dioxide (R-744), water (R-718), air (R-729) and ammonia (R-717). The most commonly used HC refrigerants are ethane (R-170), propane (R-290), iso-butane (R-600a), and propylene (R-1270). HC refrigerants are categorised as A3 by ASHRAE Standard 34 and ISO 817 which mean that these refrigerants are highly combustible with a lower flammability limit (LFL) of less or equal to 0.10 kg/m^3 and a heat of combustion of greater than or equal to 19,000 kJ/kg. Table 1 shows properties of the most commonly used HC refrigerants.

Table 1: Properties of Common HC Refrigerants (ASHRAE, 2013)

Refrigerant	Molecular mass M, (g/mol)	Critical temperature, t_{cr} (°C)	Critical pressure, P_{cr} (MPa)	Boiling point, T_{bp}(°C)	Atmospheric Lifetime, y	ODP	GWP
R 290	44.0	96.740	3.80	- 42.2	0.041	0	20
R 600a	58.1	134.660	3.64	- 0.5	0.016	0	20
R 1270	42.1	91.061	4.67	- 47.7	0.001	0	20
R 170	30.1	32.200	4.87	- 88.6	0.167	0	20

3.1 Flammability Issues and Classification of Hydrocarbon Refrigerants

Flammability of HC is one of the major factors in preventing its wide usage refrigerant in ACR industries. However, it must be noted that for HC to combust, it must meet three conditions. HC and air mixture ratio, must be within its flammability ranges of lower flammability limit (LFL) and upper flammability limit (UFL), ignition temperature must be high enough to start the combustion and finally, oxygen that air contains must be within the correct fuel/air mixture for combustion to occur.

Due to this inherent factor, many researchers have conducted experiments on the flammability issues of HC refrigerants used in vapour compression cycles by simulation with a number using actual equipment in test chambers.

Colbourne and Suen (2003) simulated the 'leak' scenario of HCs in an enclosed chamber and concluded that, if smaller amount HCs is mixed with the surrounding air the safety aspects of using HC refrigerants will be enhanced. Using the quantitative risk assessment (QRA) technique, Colbourne and Esperson (2013) established the ignition frequencies (IF) of ice cream cabinets with “safety optimised” design that uses R 290, placed in different room sizes and positions to be in a range of 2×10^{-11} to about 1×10^{-8} per y as compared to a typical frequency of fire in common household refrigerators of 1×10^{-5} per y. This shows a much less risk of fire in safety enhanced designed of R 290 cabinets as compared to common refrigerators.

Li (2014), used gas detectors set up at different locations to detect leaks encountered using R 290 from a split type air conditioner in an enclosed room. The concentrations of leaking HC were around 1/3 of LFL and HC will reach LFL at a region directly below the indoor unit. Zhang et al., (2013), ignited leaking refrigerants from a split unit air conditioner in a controlled environment, R 290 will ignite within a very close range of the indoor unit and combustion of R 290 leaking in an enclosed space will create a pressure of 6.5 kPa above atmospheric pressure. Other researchers used computational fluid dynamics (CFD) to ascertain the effects of leaking HC from a source in an enclosed space. Li et al. (2013) uses the ANSYS FLUENT V12 to ascertain the ratio of dangerous area volume of a simulated leaking refrigerant from a ceiling height of 3 meters using different flowrates over different time intervals. Nagoasa (2014) uses the open sourced OpenFOAM software to simulate leaking combustible gas from the height of 2.2 m on the wall. Girodroux et al. (2000) determined the critical flammability ratio (CFR) of HC with non-flammable refrigerants to find blends that can be accepted as non-flammable category by ASHRAE and Underwriter Laboratories (UL). Standards and comprehensive guides were also being developed to allow HCs to be used safely in different locations and environments.

3.2 Standards and Guides for Safe Operation of Hydrocarbon Refrigerants

Flammability issues of HC refrigerants can be overcome by good and safe design based on standards and safety guides. Adopted standards on refrigerants in particular HC refrigerants are published by authoritative bodies that are involved with the ACR industries and national standard bodies. ASHRAE (formerly known as The American Society of Heating Refrigerating and Air Conditioning Engineers), published several authoritative standards for the safe use of refrigerants. Standard 15, (Safety Standard for Refrigeration Systems), Standard 34, (Designation and Safety Classification of Refrigerants), and Standard 147, (Reducing the Release of Halogenated Refrigerants from Refrigerating and Air Conditioning Equipment.). The International Organisation for Standardization (ISO) publishes standards on the safe use of refrigerants. The ISO 5149 (2014) Refrigeration and Systems and Heat Pumps- Safety and Environmental Requirements which comprise three parts, Part 1 (Definitions, classification and selection criteria.), Part 2 (Design, construction, testing, marking and documentation) and Part 3 (Installation site). The ISO 817:2014 is a standard that deals with “Refrigerants-Designation and Safety Classification”. On the issues of refrigerant flammability, ASHRAE 34 (2010) and B.S.ISO 817 (2014), uses the flammability and toxicity levels classifications, as shown in Table 2. These existing standards pertaining to refrigerants safety and compatibility are available and widely adopted in most countries. Apart from the American and international standards, the E.U. have published the B.S. EN 378 (2014) which deals with “Refrigeration Systems and Heat Pumps–Safety and Environmental Requirements”. This standard comprises four parts which are in Part 1, (Basic requirements, definitions, classification and selection criteria), Part 2, (Design, construction, testing, marking and documentation), Part 3, (Installation site and personal protection.), and Part 3, (Operation, maintenance, repair and recovery)

Annex C of Part 1 EN 378 (2014) is for the calculation of maximum charge allowable for the HC used in different locations of a building. Part C.3.1 Annex C stated, factory sealed refrigeration systems with less than 150 g of A2 and A3 (flammable refrigerants) have “no location classification constraints” which means that the most domestic refrigerators having less than 150 g of HC refrigerants or blends can be placed anywhere in a building. The maximum charge allowable for air conditioning, heat pump and refrigeration equipment that does not comply to the factory sealed and charge HC of 150 g and below is shown in Eq(1) (ISO 5149-1, 2014; EN 378-1, 2013). Maximum charge allowable in human occupied spaces is given as:

$$M_{max} = 2.5 \times LFL^{5/4} \times h_0 \times A^{1/2} \quad (1)$$

When the refrigerant charge is known or ascertained than the minimum allowable area that the equipment is allowed to be installed in human occupied areas shall be determined using Eq(2).

$$A_{min} = m^2 \times (2.5 \times LFL^{5/4} \times h_0)^{\ 2} \quad (2)$$

where, M_{max} is the maximum charge allowable in a room (kg), *m* is the refrigerant charge amount in the system (kg), A_{min} is the required room area (m^2), *A* is the room area (m^2), LFL is the lower flammable limit (kg.m^{-3}) and h_0 is the height factor of appliance (m).
The British Refrigeration Association (BRA) have published a “Guide to Flammable Refrigerants” which contains useful information for safe use of HC from regulations and code of practices, transportation, applications and design to procedures in servicing and training. The Australian Institute of Refrigeration Air conditioning and Heating (AIRAH, 1998), “Refrigerant Selection Guide 1994”, includes a section on HC refrigerants.
Guides and standards are used to ensure that minimal amount of HC refrigerant is charged into systems to minimise the occurrence of mixture of HC and air meeting the LFL levels when HC leaks from the AC. Guides and standards do not provide the information needed to properly design systems that uses HC efficiently. The use of HC refrigerants must be safe and also efficient as inefficient HC systems contribute more CO_2 emissions than conventional HCFC or HFC refrigerants as 90 % of emissions are from are from indirect emissions. The energy efficiency ratios or coefficient of performances of HC is considered together with safety when designing and installing ACR systems.

Table 2: Classification of Refrigerants

Classifications	Explanations
Class A	Refrigerants that have an occupational exposure limit (OEL) of 400 ppm or greater
Class B	Refrigerants that have an OEL of less than 400 ppm, where the numeral 1, 2 and 3 denotes the following:
Class 1	No flame propagation in air at 60 °C and 101.325 kPa
Class 2	Exhibits flame propagation in air at 60° C and 101.3 kPa, lower flammability limit (LFL) greater than 0.10 kg/m^3 at 23 °C and 101.3 kPa, and heat of combustion less than 19,000 kJ/kg
Class 2L	Class 2 refrigerants may be classified as 2L if they exhibit maximum burning capacity of no more than 100 mm/s at 23 °C and 101.325 kPa
Class 3	Exhibits flame propagation in air at 60 °C and 101.325 kPa and lower flammability limits (LFL) less than or equal to 0.10 kg/m^3 at 23 °C and 101.325 kPa or heat of combustion greater than or equal 19,000 kJ/kg

4. Performance of Hydrocarbon Refrigerants

Energy Efficiency Ratio (EER) is the description of the performance of the refrigeration cycle.

$$EER = \frac{Useful\ refrigeration\ effect}{Net\ energy\ supplied\ by\ external\ sources} \quad (3)$$

EER as stated in Eq(3) is given as the ratio of useful refrigeration effect (in watts or British thermal units per hour) over the total energy (in watts) supplied to operate the ACR system.
Apart from this, the compression ratios, mass flow rate and temperature glides of the HC refrigerants must be taken into consideration in evaluating the full comparative analysis of the HC refrigerants. The molecular mass of R290 is about half of that of HCFC22 to have similar refrigeration effect (ASHRAE, 2013). Current ACR systems that uses R22 and R134a as refrigerants must change the charge volume and compressor displacement if R290 and R600a are used.
In actual experiments, numerous authors have reported different results in comparing HC refrigerants with HCFC and HFC refrigerants. Halimic et al. (2003) compared R290 with R12 and R410a in refrigeration cycles. They concluded that R 290 had highest cooling capacity and coefficient of performance (COP) of R 290 equals

that of R12. Joudi and Al Amir (2014) reported the highest COP for R290 as compared to R22, R407c and R410a in ambient temperature of between 35 °C to 55 °C. Devotta et al. (2005) tested a window type air conditioner according to Indian Standard 1391 which resulted in COP of R 290 higher that of R22 in ranges of 2.8 – 7.9 %. He et al. (2014) experimented using chest freezer concluded that COP of R290 is lower by 1.8 % as compared to R 134a. Rasti et al. (2014) blends of R 290/R 600a in mass ratio of 56 : 44 % in a domestic refrigerator resulting in reduction of energy consumption of 5.34 % as compared to R134a. Hwang et al. (2006) researched R 290 with R 404 and R 410a, where results indicated R 290 had COP improvements between 4 – 12 %. Jayaraj and Muraleedharan (2007), indicated that COP of R 290/ R 600 in ratio of 45.2 / 54.8 % is 3.6 % higher than R134a. Kim et al. (1998) uses the Korean Standard 9305 to compare R 600a and R 12 in domestic refrigerator. COP of R600a had 6 % improvements over R12. Teng et al., (2012) reported that COP of R 290 is higher than R 22 at 50 % of R22 charge. Wongwises and Chimres (2005) blended R290/ R600 at a rate of 60 / 40 % in a domestic refrigerator which results in reduced energy consumption of 4.86 % compared to R134a. Wu et al. (2012) concluded that a 20 % increase in compressor displacement will enhance the performance of split unit ACs.
The results gathered shows HCs have better performance than HCFC and HFC refrigerants in different set ups. HCs were used as refrigerant in the early 1900s and were replaced by the synthesised refrigerants to overcome the flammability issues. CFCs, HCFCs and HFCs were manufactured to mimic performances of natural refrigerants without the need to worry about toxicities and combustibility issues.

5. Conclusions

With the impeding phase-out and phase-down of ozone depleting and global warming refrigerants, Malaysia has started to impose a targeted reduction of HCFC in industries beginning of 2016. The ACR sector is the most affected, in particular the domestic AC markets, when DOE imposed the restrictions on the manufacture and installation of domestic AC of 7.03 kW and below which mainly use HCFC 22. Manufacturers and importers, switching to HFC 410a, as replacements for R22, would be making a wrong move, as the GWP and initial cost of R410a, are higher. Besides, components and oil used in the ACs are incompatible. R410a has 50 % higher operating pressures and uses polyolester oil (POE). Standards, guides and equipment (*ATEX* certified components) are already available for the safe and efficient use for HC based ACR equipment. It is about time that legislation on the use of HC be enacted to enhance its use in ACR industries, considering the widely available international and regional standards and guides for the adoption of HC and other natural refrigerants. In terms of performance, many researchers had unanimously concluded on the improved performances of HC and its blends compared to HCFC and HFC. These results would further enhance the selection of HC as the best choice of replacement for HCFC, instead of HFC.

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