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Novel Reduced GWP Refrigerant Compositions for Stationary Air Conditioning

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ABSTRACT

The working fluids most widely used for small and mid-sized air conditioning systems globally are R-410A and R-22. While these fluids have many positive attributes for cooling and heating, they are the subject of valid criticisms regarding their high direct global warming potential (GWP) and, in the case of R-22, ozone depletion potential (ODP) also.

In the interest of improved environmental sustainability, a new class of refrigerant molecule has been developed, the hydrofluoroolefin, or HFO. While the very low direct GWP values of these molecules are attractive, none of the HFOs by themselves are fully satisfactory for use in conventional stationary AC system designs, for reasons of concern about low capacity and flammability. Blended refrigerant candidates have been developed to provide better overall safety and performance, while retaining significant environmental sustainability properties versus the legacy refrigerants. This paper discusses the considerations for designing viable refrigerant blends and the process for developing useful refrigerant compositions.

There is as yet no universal agreement on what are the most critical properties for air conditioning fluids. Proposed legislation and industry discussions are proposing such features as high energy efficiency, low Life Cycle Climate Performance (LCCP), low direct GWP, performance to match current commercial fluids, non flammability, and more. No single candidate has yet been identified that meets every proposed requirement for a refrigerant fluid. This paper discusses three candidate fluids that have been developed to meet three of the most often cited sets of environmental, physical and performance properties for air conditioning (AC). Each of these candidate fluids possesses desirable, but different sets of properties. The trade-offs and relative performance and environmental merits of each will be discussed.

The three fluids vary in direct GWP values, flammability, capacity, and critical temperature. As these fluids are not yet commercial, they will be designated by laboratory identification DR codes that are being used in the AHRI (American Heating and Refrigeration Institute) AREP (Alternative Refrigerant Evaluation Program). One is a 2L flammable gas that gives capacity performance near that of R-410A, designated DR-5A. One is a nonflammable composition for use to replace R-22 in high ambient temperature environments, designated DR-91. The third is a 2L flammable replacement for R-22 with a direct GWP of less than 150, designated DR-3. In designing and evaluating these new compositions, the trade-offs that exist between refrigeration capacity, efficiency (COP), temperature glide, GWP value, and flammability have been explored and assessed. We report on some comparisons of these compositions in AC measurement and modeling work. The new compositions have been evaluated with thermodynamic refrigeration cycle models at standard AC conditions, and at high ambient temperature operating conditions, and show good performance. Testing is underway to evaluate performance of these new compositions in laboratory equipment and in actual operating systems, as compared to R-410A or R-22 performance in the same or similar systems. The status of the testing will be reported in this paper. These new

compositions should provide useful options to help maintain the quality of life and health benefits that accrue from air conditioning and refrigeration, but in an energy efficient and environmentally sustainable manner.

1. INTRODUCTION

Under the terms of the Montreal Protocol, CFCs like R-12 and HCFCs including R-22 have been scheduled for phase out. In fact at this time most use of these gases as refrigerants has been phased out, with exception of some legacy equipment and equipment in Article 5 countries, as allowed by the Protocol. There is still significant work being done to identify the most suitable replacement refrigeration and air conditioning fluid options. The first generation of work has been to replace CFCs and HCFCs with HFCs. With recognition of global climate change concerns, work has transitioned to development of replacements with lower direct GWP values. R-134a was a successful development because it had physical properties that were similar enough to R-12 to allow an efficient transition from R-12 in compliance with the regulatory and environmental needs of stratospheric ozone protection. The new molecule HFO-1234yf is now on track to replace R-134a. For most of the other commercial halogenated refrigerants it is a greater challenge to identify simple replacements. In the case of R-22, for example some AC application work transitioned to R-407C. Eventually OEMs transitioned to the higher pressure blend R-410A in order to meet safety and energy use standards for cooling occupied spaces and comply with the Montreal Protocol. In temperate climate zones this has been a workable solution, again with the caveat that R-410A has a moderately high direct GWP. The application of R-410A in hot climates has not been as successful. R-410A has a critical temperature that is about 25 K lower than that of R-22, and the two phase operating zone of R-410A is sufficiently narrow at hot ambient condensing conditions that both, volumetric cooling capacity and energy efficiency are diminished. Because of these concerns, many building owners, system manufacturers, and users in hot climate regions have been reluctant to stop using R-22. As cooling for food preservation, human comfort, and related quality of life matters is highly valued by society, research has been conducted in order to identify cooling solutions that have less impact on the earth's environment, and at the same time do not create negative consequences to human health or safety. Cooling technologies based on physical effects such as thermoelectric, magneto caloric, acoustic wave compression, etc. are being evaluated, but none is as energy efficient for moving heat from one place to another as the vapor recompression cycle. There are several potential refrigerant gas candidates that can be beneficially used in a vapor compression cycle, including CO2, ammonia, and hydrocarbons. Each offers some advantages environmentally, but each also has disadvantages that limit their use to specific areas of application. For every end use application it is necessary to weigh the refrigerant properties and select the gas that provides the best balance of properties to meet the cooling need while also besting meeting environmental and safety needs.

2. REDUCED GWP REFRIGERANT GAS

The concerns about contribution of leaked or otherwise released refrigerant gases to global climate change have resulted in substantial research and investment to find ways to provide cooling and at the same time reduce potential impact on the environment. Measures to reduce climate change impact include developing new working fluids for cooling and heat pump systems which, if inadvertently leaked to the atmosphere, have low environmental impact. Additional measures involve reducing the energy used to operate these machines. Design of cooling equipment to facilitate installation, operation, and service without loss of working fluids is necessary. More attention is being given to service practices, especially decommissioning at the end of the useful life of cooling equipment, to recover and recycle refrigerant gas, and thereby reduce atmospheric contamination. Much more can and should be done in all of these areas.

2.1 Direct GWP Values vs. LCCP

The nonflammable fluorocarbon refrigerant gases have high chemical and thermal stability, both of which are desirable for use as safe working fluids in closed systems. They offer efficient operation in vapor compression cooling and heating cycles. The nonflammability of these gases has also been a valued property to protect life and

property. The same high chemical stability that is desirable in a closed refrigeration machine however contributes to long atmospheric lifetimes if these fluids are released. Long atmospheric lifetime together with the characteristic infrared energy absorption characteristic of most aliphatic fluorocarbons gives these HFC molecules a high direct GWP value. It must be stated that the direct GWP value of a refrigerant gas is not the only predictor of environmental or climate change impact for any refrigerant. All refrigeration and AC systems consume energy, usually electricity, in order to operate their respective compressors, pumps and fans. The generation of that electricity is typically by burning fossil fuel, such as coal, oil, or natural gas. The combustion of those fuels generates the global warming gas carbon dioxide (CO2). Over the lifetime of a typical AC or refrigeration system substantial quantities of CO2 from power generation are produced and emitted to the atmosphere. A cooling system that operates more efficiently, using less power over its lifetime will have less climate change or global warming impact than a less efficient system that produces the same amount of cooling. It is therefore more meaningful to consider overall life cycle climate performance (LCCP) instead of only the direct GWP value of a working fluid. However, at the present time legislation is being drafted and promulgated in terms of direct GWP values, so for this paper direct GWP values will be values will be discussed. Note also that in this paper GWP values are from the IPCC Fifth Assessment Report (AR5)

An extensive search for new materials to use as refrigerants has identified another family of molecules, the hydrofluoroolefin (HFO). These molecules, most notably the tetrafluoropropenes HFO-1234yf and HFO-1234zeE offer benefits for some cooling applications. Surprisingly these isomers have physical and cooling properties that differ significantly from each other. In the pure form, each has limited ranges of efficient application. Both are 2L flammable, producing feeble and unstable flames. The HFO-1234 isomers have very low heat of combustion if burned, and yield very little pressure rise as a consequence of their very slow flame propagation speed. It is important to note that some HFO molecules such as 1,4-hexafluoro-2-butene (HFO-1336mzz) are entirely nonflammable, and other HFOs such as trifluoroethene have been used as monomers in the plastics industry for making polymers and is known to be highly reactive. Therefore it is important to be very specific when addressing properties of members of this specific class of molecule. HFO properties can vary over a large range, as do HFC properties, examples as shown in Table 1.

Some fluorinated olefin molecule alternatives with short atmospheric lifetimes are being evaluated for use as components of working fluids in AC and refrigeration systems. Such solutions may reduce global climate change impact, but other practicality and safety questions need to be addressed. Work has been done to show that some refrigerants with short atmospheric lifetime have adequate stability to give necessary product life in sealed refrigeration and air conditioning systems. Others, including the monofluoro alkanes have unexpected thermal instability or toxicity (Leck, 2012), (Hydutsky 2014), (Allgood 2014).

HFO-1234yf has been shown to be a good replacement for R-134a in medium temperature applications including automobile air conditioning (SAE, 2009), and is proving to be as least as effective as R-134a in some commercial refrigeration applications such as bottle coolers (Minor *et al* 2010), and vending machines (Okoshi *et al* 2010). However, air conditioning and heat pump systems most often are designed to use refrigerants with higher volumetric capacity than that of R-134a or HFO-1234yf. It has been reported that blend compositions containing HFCs and HFO-1234yf can work well in room and building air conditioning equipment (Leck and Yamaguchi, 2010). Such a blended refrigerant can take advantage of the higher capacity of some HFCs like R-32, while benefitting from the higher efficiency and lower GWP of HFO-1234yf, and gain other benefits as well. The other benefits of the blends include better isentropic compression characteristics and more robust lubricant compatibility.

3. REDUCED GWP BLENDS FOR R22 REPLACEMENT

The design of viable refrigerant blends can be challenging. ASHRAE Standard 34 lists more than fifty zeotropic refrigerant mixtures, and more than a dozen azeotropic refrigerant blends that have been submitted for safety classification and designation. Some of those refrigerant blends have proven to be useful to the industry, and many have not seen broad commercial acceptance. This illustrates the fact that creating a refrigerant composition that is truly useful to the cooling industry is not trivial. Dissimilar molecules may interact with each other in ways that are

sometimes unexpected to form mixtures that exhibit varying degrees of non ideal solution behavior. Simple mixture models that assume ideal mixing properties can yield unexpected and inaccurate results. (Minor and Leck 2008).

Inspection of the molecules listed in Table 1 shows that boiling point, GWP value, and flammability properties can vary widely within a family of, for example, similar two carbon and three carbon molecules. It would appear easy to select ingredients with similar boiling points, similar degrees of flammability, and GWP values to mix together. However, inspection of the table shows that such properties as low GWP, non flammability, and low boiling points are often exclusive. It is difficult to select ingredients to form a mixture that will match the boiling point of, say, R-22, at -40.9 C, retain low GWP, and non flammability. One finds candidate low GWP formulae can be flammable, or have high temperature glide.. Add to that the need to approach the critical temperature and cooling performance of R-22 in a system and it is more challenging. One finds that some compromise and property balance is necessary. The goal becomes minimizing the compromises while maximizing the desirable properties.

Modeling the performance of blends will yield correct results only if accurate binary interaction parameters and appropriate mixture modeling techniques are employed. We are working to develop a new family of refrigerant compositions that use low GWP ingredients, including HFO molecules, in order to achieve a higher degree of environmental sustainability. In our laboratory it is standard practice to begin by measuring many properties, including interaction coefficients for all of the binary pairs considered for mixtures. In addition high order modeling techniques have been developed and validated to facilitate accurate modeling of candidate mixtures. Modeled results are ultimately validated by equipment tests in order to determine impacts of pressure drop and heat transfer characteristics of the candidate refrigerant blends, as well as power consumption by the compressor and motor. The results in this paper are based on such thermodynamic modeling. Combinations of R-32, R-125, R-134a, R-1234yf, and in some cases other HFOs and other HFCs are ultimately needed. The subject refrigerant candidates have been submitted for AREP or PRAHA and other OEM test programs to get final analysis of the usefulness of the overall blended refrigerant.

Refrigerant Number	Chemical Name	Boiling Point °C	Direct GWP	Flammability Class (ASHRAE/ISO)	
			(AR5)		
R-32	difluoromethane	-51.7	677	A 2L	
R-116	hexafluoroethane	-78.2	11,100	A 1	
R-125	pentafluoroethane	-48.1	3170	A 1	
R-134a	1,1,1,2-tetrafluoroethane	-26.1	1300	A 1	
R-143a	1,1,2-trifluoroethane	-47.5	4800	A 2	
R-152a	1,1,difluoroethane	-27	138	A 2	
R-161	monofluoroethane	-37.6	4	A 3*	
R-170	ethane	-89	5.5	A 3	
R-218	octafluoropropane	-36.8	8900	A 1	
R-227ea	1,1,1,2,3,3,3-heptafluoropropane	-16	3350	A 1	
R-236fa	1,1,1,3,3,3-hexafluoropropane	-1.4	8060	A 1	
R-245fa	1,1,3,3,3-pentafluoropropane	14.9	858	B 1	
R-290	propane	-42.1	3.3	A 3	
R-1234yf	2,3,3,3-tetrofluoropropene	-29.4	<1	A 2L	
R-1234zeE	1,3,3,3-tetrafluoropropene	-19	<1	A 2L	
HC-600	n-butane	-0.5	4	A 3	
HFO-1336mzz	1,1,1,4,4,4-hexafluorobutene	33.4	2	A 1*	

Table 1: Selected fluorocarbon and hydrocarbon refrigerant ingredient properties;

 "*" indicates anticipated classification.

3.1: HFO Based Refrigerant Blends

There are already HFO based refrigerant blends being commercialized to replace the very higher GWP gases. As the topic here is air conditioning, for simplicity, I am showing only some AC refrigerant candidates. Note, on this chart, the GWP values shown are based on the recently released data from the IPCC Fifth Assessment Report, AR-5.

In Table 2 are summarized a broad range of reduced GWP refrigerant candidates for comfort cooling, along with comparison to the refrigerant each is intended to replace. This table is expanded to show replacement candidates for R-134a, which is used is some heat pumping and cooling equipment, due to its higher critical temperature. The higher critical temperature facilitated use in high ambient operating temperature regions, where it is widely used in large chillers driven by centrifugal or screw compressors. Also shown is R-123, which is a low pressure HCFC fluid widely used in centrifugal chillers due to the higher COP that can be achieved by these machines. The nonflammable HFO-1336 and compositions based on HFO-1336 are being evaluated for use in chillers and heat pumps for large commercial and industrial scale systems. Note that the reduced GWP candidates fall into two categories, nonflammable - ASHRAE Class 1 - and slightly flammable – ASHRAE Class 2L. In order to achieve non flammability and R-22 capacity levels, the DR-91 composition has a higher GWP, but still less than 900. To meet the GWP cap of 150 and still have R-22 level capacity the refrigerant DR-3 will fall into the ASHRAE classification of 2L due to its mild flammability.

		Lower GWP Candidates for Air Conditioning			oning	
Current	GWP	Non Flammable		Mildly Flammable		
Refrigerant	(AR-5)	(ASHRA)	E Class 1)	(ASHRAE Class 2L)		
		Name	GWP	Name	GWP	
R-134a	1300	XP10	<600	HFO-1234yf	1	
R-22	1760	DR-91	<870	DR-3	<150	
R-410A	1924			DR-5A	460	
R-123	79	HFO-1336	2			

 Table 2: Air Conditioning candidate property comparison

For clarity, the AC fluids that are being evaluated for domestic and light commercial use are highlighted in Table 3 relative to the legacy cooling system platform they are intended to replace. Here domestic refers to room air conditioners and ducted AC units sized for single family homes or small office buildings and apartments. R-32 is in a slightly different category as it uses technology for mitigation of discharge temperatures and special lubricants to accommodate lower solubility in conventional compressor lubricants. The refrigerants in Tables 2 and 3 need no such considerations and use the same lubricants and construction materials and techniques as conventional HFC refrigerants. With any of these replacements, especially for evaluation purposes, it may be necessary to adjust compressor speed, or to select a compressor with slightly different displacement in order to exactly match the volumetric capacity of a legacy system for comparison testing. In many cases it may not be important to exactly match the capacity of a new system to legacy platform capacities, but comparison of overall energy efficiency performance may be facilitated by operating a test system at a capacity level that matches the legacy baseline system.

3.2: Cooling Performance Comparisons

In Figure 1 is shown the Pressure vs. Enthalpy phase diagram (PH Diagram) for R-22, overlaid with the PH two phase envelopes that have been developed for the two R-22 replacements, DR-3 and DR-91.

Refrigerant	Replaces	Flammability	GWP	
DR-91	R-22	NO	< 870	
DR-3	R-22	Yes – 2L	< 150	
DR-5A	R-410A	Yes – 2L	< 470	

Table 3: Air conditioning refrigerant candidates for domestic & light commercial

From these diagrams it can be seen that the widths of the two phase region of the domes are similar to while slightly less than that of R-22, consistent with the small variations in relative volumetric capacity, as enumerated in the cycle calculation results shown in Table 4. The PH diagrams also illustrate that the critical pressures of DR-3 and DR-91 are close to that of R-22. Similar comparisons have been made for R-410A and DR-5A.



Figure 1: Pressure vs. enthalpy compared to R-22

Thermodynamic modeling analysis has been employed to demonstrate the relative COP and Capacity of these refrigerant compositions. The cycle conditions for the results in Table 4 are as follows:

Evaporator: 7 °C, Condenser: 47 °C, compressor efficiency: 70 %, Liquid sub cooling: 10 K, Evaporator superheat: 5 K

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All of the results are normalized relative to R-22, as there is high interest in many hot climate regions for direct replacement of R-22, especially if appropriate nonflammable refrigerant gas can be identified. DR-91 was developed to address the need for non flammability. The critical point temperatures (T_{cr}) of the various refrigerants are noted in the bottom line of Table 4. The relatively high T_{cr} in addition to the shape of the PH dome can explain how R-22 is able to maintain satisfactory cooling performance in hot climate conditions, as compared to R-410A. R-407C was included for reference in the comparisons as it is a known zero ODP and nonflammable AC refrigerant.

Property	R-22	R-407C	DR-3	DR-91	R-410A	DR-5A	R-32
GWP (IPCC AR5)	1760	1624	146	870	1924	465	677
Flamm. Class	1	1	2L	1	1	2L	2L
Capacity vs. R-22		+1.7 %	- 4.5 %	-11 %	+45 %	+42 %	+59 %
COPc vs. R-22		-2 %	-2 %	-1 %	-6 %	-5 %	-6 %
Glide, Kelvins	0	4.8	7	4.2	< 0.2	1.5	0
Discharge Temp. °C	83	75	69	67	81	87	103
Critical Temp. °C	96.1	86	82.5	86	71.3	76.5	78.1

 Table 4: Detailed comparisons of candidate replacements vs. R-22

The relatively higher critical temperatures of the R-22 replacement candidates suggest that operation at hot ambient conditions may be possible with lower loss of performance than the higher capacity R-410A and its replacements. To test this, cycle models were run at two conditions, typical AC conditions, and hot climate conditions. The results of that analysis are shown graphically in Figure 2.



Figure 2: Results for Normal vs Hot Climate conditions - COP and Capacity Normalized to R-22

4. DISCUSSION

Operating a cooling cycle at hot temperatures results in a reduction in capacity and in COP. Increasing the condensing temperature from 35 C to 50 C resulted in a capacity reduction of about 11 %, and a COP reduction of about 40 % for the R-22 base case.. It is not too surprising that potential alternatives to R-22 also exhibit decreases in capacity and COP. The amount of decrease will vary with the characteristics of each individual refrigerant. In the case illustrated in Figure 2, capacity reductions for R-410A were 14.3 %, and COP loss was 42.3 %, the largest

reduction of all of the refrigerants compared. One of the concerns in hot climate areas, and especially at even higher discharge temperature conditions, is that R-410A, with its lower critical temperature of 71 C, versus 96 C for R-22. Also the more narrow Pressure –Enthalpy dome and hence the reduced latent heat of phase change is smaller. One goal for developing a replacement for R-22 is to retain as much efficiency and capacity as possible in the hot climates. For this reason, a replacement refrigerant should ideally have a high T_{cr} and a wide two phase PH dome at operating conditions, indicating greater latent heat capacity from condensation and evaporation.

While all refrigerants, like R-22 and R-410A lose performance at the higher temperature condition, the degree of loss varies with the refrigerant. However as the data in the bar chart shows, at the high condensing temperature, the reduced GWP candidates DR-3, DR-91, and DR-5A all retain more of their COP than does R-410 A. As energy efficiency is the most import factor in reduction of the overall carbon footprint, this is very significant. Reduction in capacity is not so critical, as using a larger displacement compressor can establish volumetric capacity at a desired point. That being said, DR-5 shows less capacity loss at the hot condition than the other candidates.

5. CONCLUSIONS

Some considerations of the process by which a refrigerant composition for air conditioning is developed are described. Shown are refrigerant compositions with properties suitable for use in hot climates. Physical property and cycle modeling results have been outlined. By using a detailed development process, a set of reduced GWP compositions have been developed that can be used in new air conditioning systems that are similar to current commercial AC and heat pump designs. Two candidates were developed for use in R-22 AC system designs, and a separate composition has been developed for use in system design types nominally optimized for use with R-410A. One of the described R-22 replacement (DR-91) is nonflammable, has energy efficiency that is very close to that of R-22. Its moderate temperature glide makes it suitable for use in conventional system designs with DX evaporators and condensers. The volumetric cooling capacity is about 11 % less than that of R-22, so performance may be achieved by using a slightly higher speed compressor, or a compressor with slightly more displacement. The other described R-22 replacement candidate (DR-3) is optimized to meet more stringent direct GWP limits of 150. That candidate has capacity near that of R-22, but is slightly (ISO /ASHRAE Class 2L) flammable. A third refrigerant (DR-5A) is an improvement over a previous developmental candidate refrigerant for use in higher capacity R-410A type systems.

This work describes and documents some of the factors that affect the choice of refrigerant and the refrigerant properties that need to be considered in order to minimize impact on the earth's climate. It is pointed out that use of simplified concepts like direct GWP value, can lead to non optimal choices, or to development of regulations that may not be best for limiting overall carbon emissions into the environment and hence, on limiting global climate change. The proliferation of proposed refrigerant compositions shows that there is a need for systematic, data based approaches to developing environmentally and commercially viable refrigerant solutions for air conditioning in all climate zones. This work illustrates some of the considerations involved in developing refrigerant solutions that simultaneously meet the environmental and industrial needs.

NOMENCLATURE

AC Air Conditioning

- CAP Volumetric cooling capacity; the amount of heat transferred respectively at the evaporator or condenser (including the compressed vapor superheat and liquid sub-cooling) per unit volume of the working fluid entering the compressor
- COP Coefficient of Performance for cooling; ratio of the heat absorbed at the evaporator or condenser (including the compressed vapor superheat and the liquid sub-cooling) and the work of compression
- GWP Global Warming Potential (one hundred year integrated time horizon)
- HFO Hydro-Fluoro-Olefin
- P_{cond} Condenser pressure
- T_{disch} Compressor discharge temperature

T _{cond}	Condenser temperature
T _{cr}	Critical temperature
T _{evap}	Evaporator temperature

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