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New High Pressure Low GWP Refrigerant Blends

by

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Abstract

This paper describes the latest findings of a project to identify azeotropic and near azeotropic mixtures with a normal boiling point between -40°C and -80°C. The aim was to match the saturated pressure-temperature characteristics and refrigerating capacity of existing refrigerants within this range (such as R-410A and R-744), whilst achieving higher critical temperatures and – where relevant – lower triple point temperatures. Thus, the intended outcome is an extended range of applicability and an improvement in energy efficiency. Of a large number of potential combinations, three azeotropic and near azeotropic mixtures have been selected, all formed from natural refrigerants or synthetic chemicals with a global warming potential (GWP) of less than 150. These mixtures were subject to a variety of analyses, including thermodynamic, performance and safety assessments. Patents have been granted and global licensees are now being sought.

1. Introduction

The amount of refrigerant in use around the world is 475 ktonnes. Global refrigerant leakage is 132 ktonnes per annum, i.e. a global annual emission rate of 27.8%. [1] There is growing pressure to limit the use of HFC refrigerants due to their global warming effect, which average more than 2000 times that of carbon dioxide. Refrigerant control matters because 13% of man-made global warming comes from halocarbons. [2]The EU has passed a directive and a regulation dealing with fluorinated gases. The directive will phase-out HFC-134a in vehicle air conditioning systems from 2011. The effectiveness of the regulation will be assessed in 2011, the authors believe that additional applications for F-gas restrictions will be identified, and the most likely outcome will be the phase out of refrigerants with a GWP above 150 in new refrigeration, air conditioning and heat pump applications. So the crucial question becomes the availability or otherwise of effective alternatives to HFCs. Many HFCs can be replaced by currently available natural refrigerants, with the notable exception of the higher pressure HFC, R-410A. [3]

There are distinct advantages of fluids that possess low normal boiling point (NBP) – or high saturation pressure – such as more compact systems, possibilities for achieving higher system efficiency, and advantages associated with operating above atmospheric pressure. However, the currently available refrigerants, such as R-410A, R-32, R-744 and R-170 suffer from negative characteristics such as high GWP and/or low critical temperature. There is no single-component refrigerant with low-GWP and high critical temperature and most mixtures that may achieve these criteria are zeotropes with high temperature glide. It was therefore concluded that azeotropes or near-azeotropes (less than 2 K temperature glide) with thermodynamic characteristics similar to R-410A and R-744 would be commercially attractive. A project was undertaken with the objective of identifying such blends. [4]

2. General Methodology

The development of the new refrigerant mixtures was carried out using a staged approach for analysing the various physical, chemical, environmental and thermodynamic characteristics of the fluids. The development process followed an iterative procedure starting from prioritisation of acceptance criteria through to identification of acceptable mixtures.

Initially, a set of criteria believed to represent the "ideal" refrigerant was established: zero ODP, negligible GWP, favourable thermodynamic and transport properties, sound chemical and material compatibility, high critical temperature, low triple point, good solubility with oils, low cost, low temperature glide, low

Refrigerant	**R-1270**	**R-161**	**R-170**	**R-41**	**R-717**	**R-744**
Chemical name	propene	ethyl fluoride	ethane	methyl fluoride	ammonia	carbon dioxide
Chemical formula	CH3CH=CH2	CH3CH2F	CH3CH3	CH3F	NH3	CO2
Molar Mass	42.08	48.06	30.1	34.03	17.03	44.01
NBP (°C)	-47.7	-37.6	-88.6	-78.3	-33.3	-78.4
Critical temp (°C)	92.4	102.1	32.2	44.1	132.3	31.1
ATEL (ppm)	1000	~1000	1000	~1000	25	5000
LFL (% vol)	2.7	3.8	3.2	7.1	14.8	none
Safety class	A3	A2	A3	A2	B2	A1
ODP	0	0	0	0	0	0
GWP (100)	3	12	3	97	0	1

Table 1: Characteristics of selected refrigerants

toxicity, and non-flammable. A number of pure substances of interest, primarily because of their zero ODP and low GWP, were identified: R-1270 (propene), R-161 (ethyl fluoride), R-170 (ethane), R-41 (methyl fluoride), R-717 (ammonia), R-744 (carbon dioxide). The basic characteristic data for these fluids is listed in Table 1.

Following the selection of these substances, the next stage involved identifying suitable mixtures and then evaluating their thermophysical, chemical and environmental aspects. A novel method based on multi-criteria approach to search the next generation refrigerants [5] was employed to select refrigerant pairs with azeotropic or near azeotropic behaviour, detailed in Table 2 with the selected mixtures highlighted.

Having selected mixtures, it was necessary to evaluate their performance within the intended refrigeration systems. This was carried out initially by system performance simulations, and latterly by experimental evaluation to precisely optimise the composition, obtaining the highest efficiencies across the range of likely applications.

The three primary mixtures that have arisen from this work are:

- **R-1270/R-161** hereafter referred to as ECP410A
- **R-170/R-717** hereafter referred to as ECP717
- **R-744/R-41** hereafter referred to as ECP744

Thermodynamic modelling and experimental studies confirm azeotropic behaviour for R-170/R-717 and R-1270/R-161 but near azeotropic behaviour for R-744/R-41. Each of these has distinct characteristics providing significant advantages over currently used refrigerants for specific applications. The following sections provide a detailed explanation of each refrigerant, addressing the development and validation of property models, system performance modelling and test results.

3. ECP410A – For Domestic and Commercial Air Conditioning and Heat Pumps

A basic set of criteria were used to identify the preferred composition range. These included:

- Refrigerating capacity similar to that of R-410A (based on typical air conditioning temperatures)
- Maximum working pressure not exceeding that of R-410A
- Low, or negligible temperature glide

Refprop [6] was used to generate the appropriate thermodynamic data for a range of compositions. Figure 1 shows the bubble point pressure and volumetric refrigerating capacity (relative to R-410A), and the temperature glide, plotted over a range of compositions at 15°C.

Refrigerant	R-744	R-717	R-41	R-170	R-161	R-1270
R-1270	Zeotrope	Azeotrope	Zeotrope	Zeotrope	**Azeotrope**†	
R-161	Zeotrope	Azeotrope	Azeotrope	Zeotrope		
R-170	Azeotrope	**Azeotrope**†	Azeotrope			
R-41	**Near azeotrope**†	Zeotrope				
R-717	Reaction ††					
R-744						

† pairs selected for this work

††Ammonia and carbon dioxide react to form ammonium carbamate, a white solid material

Table 2: Azeotropic low GWP refrigerant blends

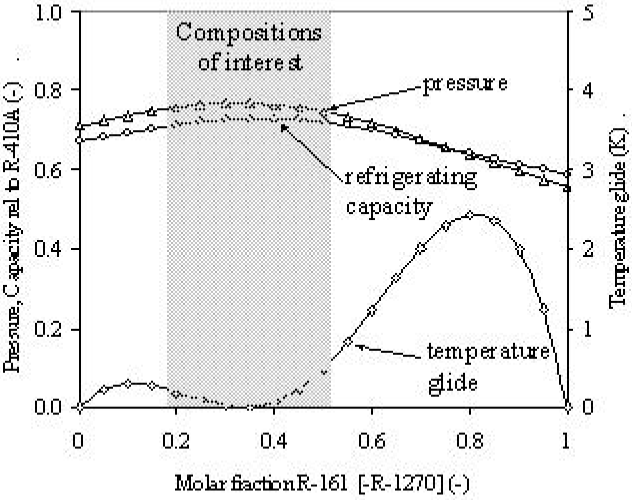

Figure 1: Bubble point pressure, volumetric refrigerating capacity and temperature glide of R-1270/R-161 at +15°C

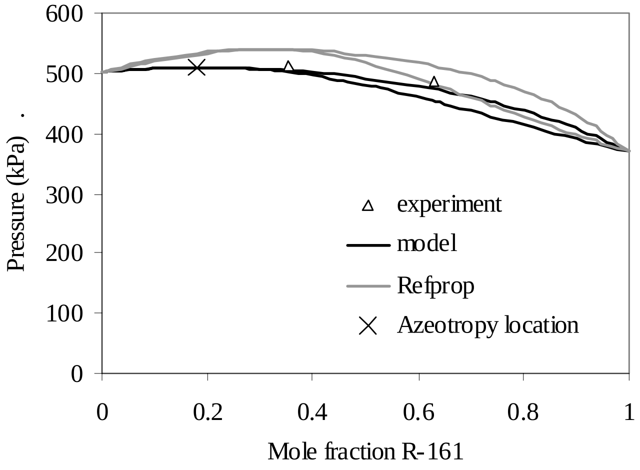

Figure 2: Comparison of measured saturation pressures of R-161/R-1270 mixtures against two models

It is seen that the mixtures have around 20 to 40% lower refrigerating capacities and pressures than R-410A, depending upon the composition. Further, the highest volumetric refrigerating capacity and saturation pressure, and smallest temperature glide occur over the same range of compositions; between 20% and 50% of R-161.

Since information on the properties of R-161 is sparse, a number of pressure-temperature equilibrium measurements were conducted at the Odessa State Academy of Refrigeration (OSAR), using both pure R-161 and R-1270/ R-161 mixtures over a variety of compositions and temperatures. Based on the results, a model was developed using the Peng – Robinson type equation of state (EoS) to predict the phase behaviour of the full range of compositions, pressures and temperatures. The liquid and vapour pressures for -5°C isotherms are presented in Figure 2 for both the developed model and values generated by Refprop. Some data-points are included, and identification of the azeotropy location.

In addition, a simulation exercise was carried out using a comprehensive system model to predict the real performance more closely. The results for refrigerating capacity and cooling COP from the simulation are presented in Figure 3. A number of observations were drawn from these results:

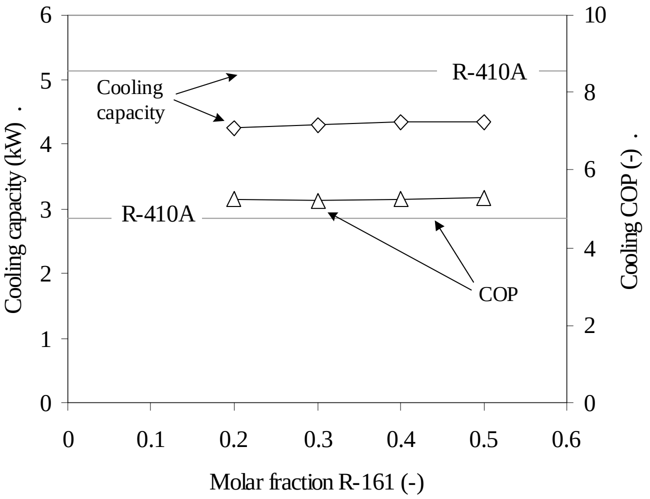

Figure 3: Evaporating capacity and cooling COP for mixtures of R-1270/R-161, and comparative R-410A values

- Cooling capacity is greater than the theoretical results (as Figure 1), between 83 – 85% of R-410A, whilst heating capacity is 80 – 82% of the R-410A values.
- Cooling COP is 10 – 11% higher than R-410A, whilst heating COP is 6 – 7% higher.
- Evaporating temperature is similar for all compositions and R-410A, condensing temperature is about 1 K lower for the

mixture and discharge temperature around 3 – 4 K lower than R-410A.

Significantly, it is seen that over the range of compositions evaluated there is little difference amongst most of the performance indicators (such as capacity and COP). This allowed selection of the preferred composition based on other criteria such as environmental, cost and safety.

The next stage was to carry out performance testing, which was done in a specially constructed rig at University College London. Measurements were made over a range of conditions: water inlet temperature from +7°C to +23°C, and condenser air-on temperature from +20°C to +35°C. This was first carried out using R-410A, then the preferred R-161/R-1270 mixture (25%/75% by mole), as well as two other compositions of R-161/R-1270, either side of this mixture. The results for the preferred composition are presented for a condenser air inlet temperature of +30°C; the results for other air temperatures were qualitatively comparable.

Cooling and heating capacity

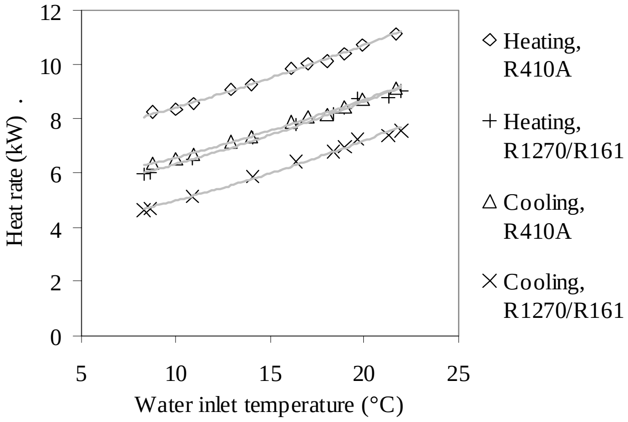

Figure 4: Evaporator and condenser capacity at +30°C condenser air inlet temperature

The cooling capacity was calculated based on the water flow rate and temperature measurements of the water circuit, and was cross-checked against the input power to the electrical heaters. Heating capacity was determined from calculated refrigerant mass flow and enthalpy change across the condenser, and checked against condenser airflow and air enthalpy difference. Figure 4 presents the comparative results for the cooling capacity and heating capacity over a range of water inlet temperatures. As expected, there is a general increase of both cooling and heating capacity with higher water inlet temperatures. The mixture shows an average cooling capacity about 20% lower than R-410A. The heating capacity is also about 20% lower that R-410A; a reduction in capacity which is comparable to that indicated by the simulation.

Cooling and heating COP

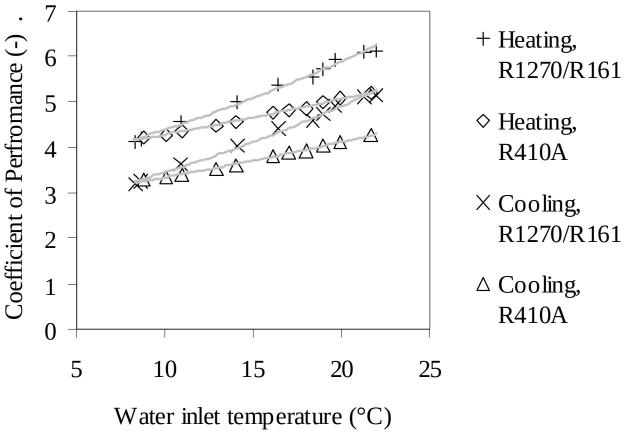

Figure 5: Heating and cooling COP at +30°C condenser air inlet temperature

The cooling and heating COP was calculated by taking the cooling or heating capacity as a ratio of the measured compressor power, and the results are presented in Figure 5. In general, there is a distinct decrease in both cooling and heating COP as water inlet temperature reduces, which is a typical characteristic. At higher water inlet temperatures the cooling and heating COP is around 15 – 20% greater than R-410A, whilst at lower water temperatures the COPs for the R-161/R-1270 mixture and R-410A start to approach unity. It is noted that the overall isentropic efficiency of the compressor (based solely on suction and discharge conditions) was approximately constant for R-410A over the entire range of water temperatures, at about 66 ±1%. However, with the R-161/R-1270 mixture, the isentropic efficiency fell from about 66% at the highest water inlet temperature to around 58% at the lowest water temperature.

The initial results for the R-161/R-1270 mixture indicated a drop in cooling and heating capacity of about 15 – 20%, compared to R-410A, based on standard rating conditions. Under the same conditions, cooling and heating COP was on average 10% higher than R-410A, although it tended to smaller values with lower water inlet temperatures. These higher efficiencies were supported by lower temperature differences and smaller pressure drops in the condenser and particularly the evaporator.

This refrigerant appears particularly suitable for domestic and commercial air conditioning and heat pump application where class A3 fluids may be safely used.

4. ECP717 – For Industrial Process, Food and Blast Freezing Applications

For a variety of refrigerating applications ammonia (R-717) is the natural refrigerant of choice. However, it possesses certain characteristics that are somewhat undesirable. In particular, the NBP of R-717 is -33°C, so lower temperatures result in sub-atmospheric pressures and the risk of ingress of air and moisture into the system, which can be detrimental to performance and reliability. Furthermore, R-717 has a low specific heat which leads to much higher discharge temperatures than experienced with other refrigerants; this is normally handled by costly oil inter-cooling or multi-stage compression. Lastly, it is immiscible with standard compressor oils, which in most systems results in the necessity for oil draining and also inhibits heat transfer, particularly at lower evaporating temperatures. In order to overcome these disadvantages, alternatives should offer a lower NBP, lower discharge temperatures and be miscible with conventional oils. This work identified the ammonia - ethane (R-170) mixture as an excellent azeotropic blend which overcomes many of these issues. The research found this azeotrope to be of a complex nature so the objective of this part of the investigation was to understand the thermodynamic behaviour of this mixture and to determine the optimum proportions.

The R-717/R-170 mixture forms positive azeotropes up to the critical region. Phase equilibria calculations based on existing information about pressure – temperature – composition measurements (p–T–x) data provided the phase diagram presented in Figure 6. With any ideal binary mixture, the bubble and dew-point lines are separate for the entire range of compositions, and only converge when the composition reaches 100% or 0% of the components. However, for the R-171/R-170 blend the two lines converge at other compositions to form an azeotropic region. A positive azeotrope exhibits a rise in the pressure-composition curve at low temperatures. At these compositions, the mixture behaves as if it were a pure, single component fluid. As temperature increases, the azeotropic vapour composition moves from the zone of the liquid – liquid miscibility gap in the direction of higher mole fractions of ammonia. At the high temperature limit, the homogeneous positive azeotropy disappears. The three–phase line terminates in the liquid-liquid upper critical end point (UCEP), which lies approximately 10 K above the critical temperature for pure ethane (+44.9°C). At low temperatures in the liquid-liquid-vapour three-phase range, the liquid phase is richer in ammonia. The R-170/R-717 blend also forms heterogeneous positive azeotropes (where the two components are not homogeneously mixed) up to the liquid-liquid UCEP where the occurrence of three fluid phases is observed as a liquid, vapour, and liquid sequence which is contrary to conventional three-phase equilibria with liquid-liquid-vapour sequence.

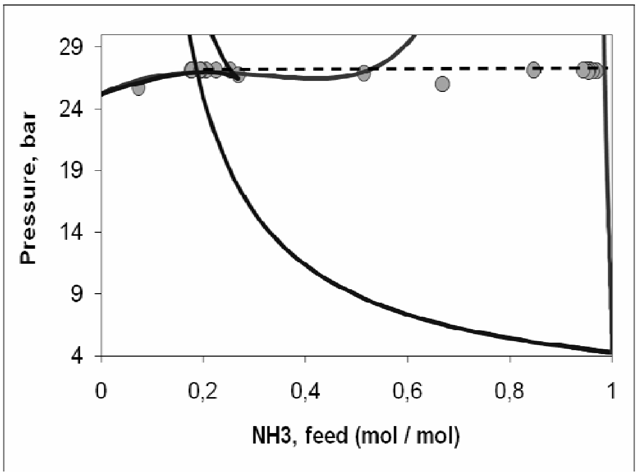

Figure 6: Pressure – composition diagram of heteroazeotropic R-170/R-717 blend at 0°C

Figure 6 illustrates the relationship between saturation pressure and composition for the R-170/R-717 mixture, showing a typical isotherm (line of constant temperature) including both azeotropic and Van Der Waals metastable states in the low-temperature region. The upper line indicates the pressure of the saturated liquid (i.e. the bubble-point) at the temperature, *T*, and the lower line indicates the pressure of the saturated vapour (i.e. the dew-point). The dashed lines correspond to the three-phase (liquid-liquid-vapour) equilibrium. The continuation of dew and bubble point curves above three-phase lines (isotherms at 0°C) reproduces the metastable states, that is, where the equilibrium conditions of the mixture may be sustained even if the external conditions, such as pressure or temperature, are changed.

The choice of preferred composition requires a balance of a number of different factors. These include system performance, operating pressures, critical points and safety classification. The property data has been used to analyse the performance with a cycle model, which provides quantitative indication of the performance over the range of compositions. Based on a condensing temperature of 27°C, an R-170 rich mixture tends to have a lower efficiency due to its low UCEP, as would be expected. Conversely, increasing the R-717 component tends to lead to a better COP, although this levels out once the composition increases the UCEP well above the condensing temperature. Considering these efficiency trends, the NBP of the mixture and limitation of discharge pressure, from a system performance perspective the composition may be within the range of 40% R-170/60% R-717, up to 70% R-170/30% R-717. It is also noted that there is sufficient R-170 within this composition range to adequately transport conventional low viscosity mineral oils, even at low evaporating temperatures.

Refrigerant safety classification is also an important consideration, since this dictates its applicability. According to ISO 817 [7], the toxicity class of R-170 is "A", whilst that of R-717 is "B". Depending upon the composition of the mixture, either an "A" or "B" classification may result. Using the criteria set out within ISO 817, an "A" classification may be achieved by ensuring a molar composition of least 21% of R-170. In terms of flammability, R-170 has a "3" classification, whereas R-717 has a classification of "2". Again, the ISO 817 criteria suggest that a flammability classification of "2" may be achieved with a molar composition of at least 27% of R-717. Therefore, in order for the mixture to achieve the more desirable "A2" classification, the molar composition should be between 21% R-170/79% R-717 and 73% R-170/27% R-717. It was observed that both the most desirable performance and safety classification coincide with similar compositions. The azeotropic blend optimised for below -33°C applications is 45% R-170/55% R-717; this achieves a sufficiently high critical temperature and an "A2" safety classification.

An illustration of the cycle performance for the chosen composition is given in Table 3. Here the cycle performance is calculated using an evaporating and condensing temperature of -55°C and +27°C, respectively, and a cooling capacity Q_0 = 10 kW for a single stage cycle. It is seen that the discharge temperature is significantly lower than pure R-717, as is the compression ratio. The theoretical COP shows a slight decline, although for a given system it is likely to exceed this due to the improved heat transfer because of better oil miscibility. The discharge pressure is 5403 kPa, and importantly, it is seen that the swept volume is 77 m^3/h.

This refrigerant has particular utility for industrial process, food and blast freezing applications and may displace liquid nitrogen as well as two stage ammonia systems.

Refrigerants	Q_0, *kW*	T_{out}, *°C*	P_0,*kPa*	P_k, *kPa*	P_k/P_0	*COP*	V_S,m^3/h	V_D, m^3/h
R-717/R-170(55/45)	10	146	580	5403	9.3	0.988	77	96
R-717	10	230	29.2	1086	37.2	1.235	124	155

V_S is volume flow in compressor suction inlet
V_D is compressor displacement rate

Table 3: An illustration of the cycle performance for the chosen composition

5. ECP744 – For Commercial Point-of-Sale Refrigeration and Vehicle Air Conditioning Equipment

Pure R-744 exhibits certain undesirable properties, such as high-side pressures considerably greater than most conventional refrigerants, and the evaporating temperature is limited by its freezing temperature (-56.6°C). One approach to overcome these shortcomings is to blend R-744. The R-744/R-41 (fluoromethane) mixture is considered to be a suitable option. R-744/R-41 belongs to the simplest type I of phase behaviour according to the Scott and van Konynenburg classification [8]. Figure 7 illustrates phase behaviour of R-744/ R-41 in the temperature range -50°C to +50°C. In the temperature-concentration range of interest a temperature glide of less than 2°C is observed, corresponding to near azeotropic (quasi-azeotropic) behaviour. This makes the blend potentially attractive as a practical refrigerant.

To construct a thermodynamic model of the R-744/R-41 blend, the Peng-Robinson equation of state (EoS) was applied. To define the accurate Peng-Robinson type EOS along the saturation curve for pure substances, the concept of local mapping was developed [9]. For the R-744 – R-41 system, experiments were carried out at OSAR. The results are superimposed on Figure 7. For each set of predicted isotherms the upper line indicates the pressure of the saturated liquid (also known as the bubble-point) at the temperature, T, and the lower line indicates the pressure of the saturated vapour (also known as the dew-point). Each set of curves are isotherms for -50°C, 0°C, +30°C, +35°C and +50°C. This temperature range represents the approximate limits of the anticipated operating conditions.

A performance evaluation was carried out in order to determine the preferred composition and to compare its efficiency and capacity. Further considerations include maintaining as high a critical temperature as possible, minimising temperature glide and achieving the highest COP. System performance evaluations with a detailed system model were carried out at European rating conditions, which revealed the following:

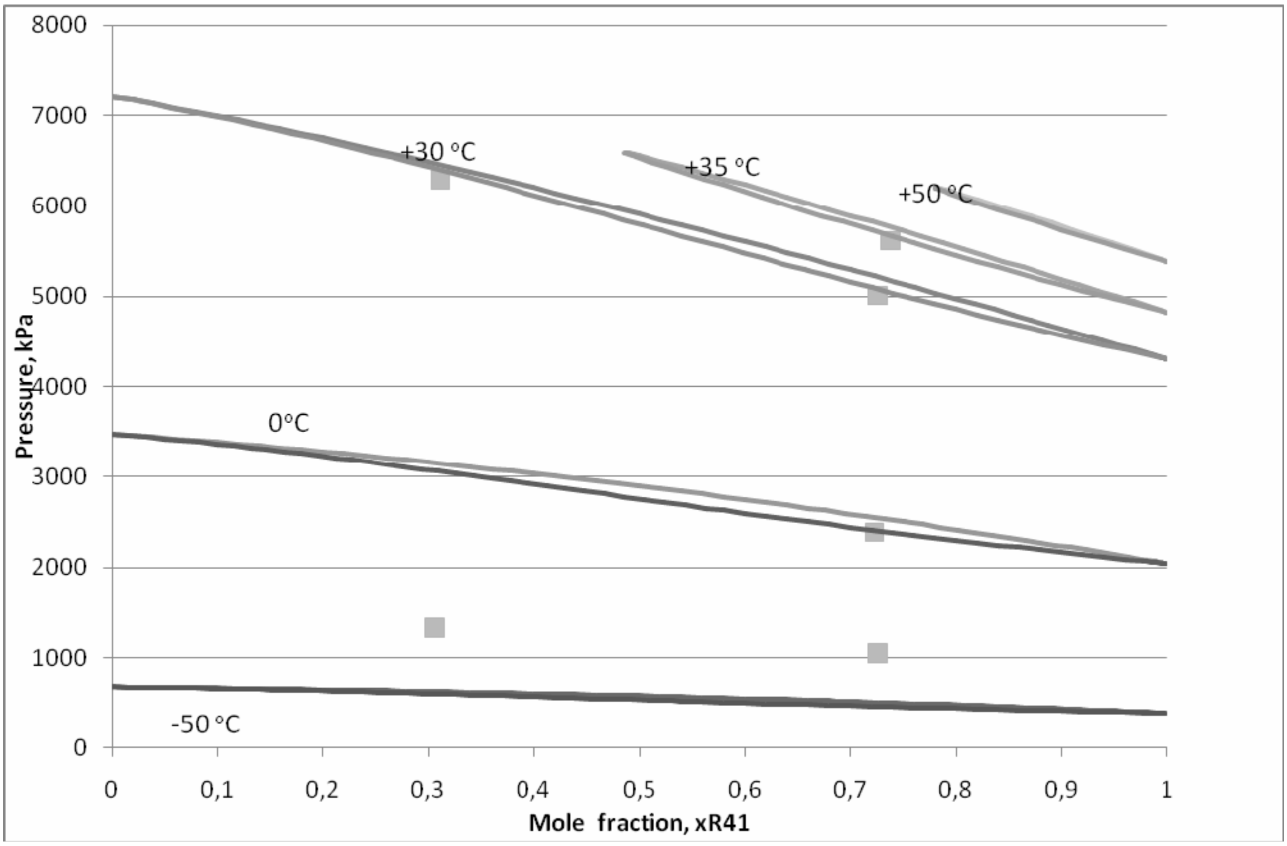

Figure 7: R-744 / R-41 phase equilibria

- In all cases, the refrigerating capacity and pressure was significantly greater than that of R-410A.
- Both evaporating and condensing capacity increase notably as R-744 composition increases.
- There is a notable reduction in both heating and cooling COP as R-744 composition increases.
- Evaporating and condensing temperatures show little variation across the range of compositions, whereas discharge temperature rises slightly with higher R-744 composition.

In order to evaluate the R-744/R-41 mixture, it was compared against pure R-744, since their thermodynamic characteristics are similar. Of primary interest is the COP, which was compared over a range of operating conditions (evaporator air-on temperature, and condenser air-on temperature). In particular, it was of interest to consider both sub-critical and trans-critical operation, so consideration was also given to the gas-cooler pressure.

Figure 8 presents the cooling COP (for air-conditioning application) of the R-41/R-744 mixture and pure R-744 against the gas cooler pressure, since the operation is mostly above the critical point. At a gas cooler air-on temperature of 39°C, it is seen that the R-41/R-744 mixture achieves its highest COP at about 8,000 kPa, whilst pure R-744 does not reach its peak CO_2 until some 12,000 kPa. Furthermore, the COP of the R-41/R-744 mixture is about 10% higher than that of the pure CO_2. When the entering temperature is at 27°C, the highest COP of pure R-744 is 1.7 at just under 8,000 kPa, but R-41/R-744 is still operating in sub-critical operating, achieving a COP of 2.0.

Figure 9 shows the variation in cycle COP over a wide range of condenser air-on temperatures, and in transcritical mode, the ideal high side pressure, i.e., the system controller is assumed to modulate the gas cooler pressure to optimise COP. For both refrigeration and air conditioning application temperatures, the R-41/R-744 mixture produces a notably higher COP than pure R-744, both under sub-critical and transcritical operating conditions. It is also noted that as the operating conditions approach the critical point of R-744, and until after the critical point of the R-41/R-744, there is a significantly greater difference between the COPs.

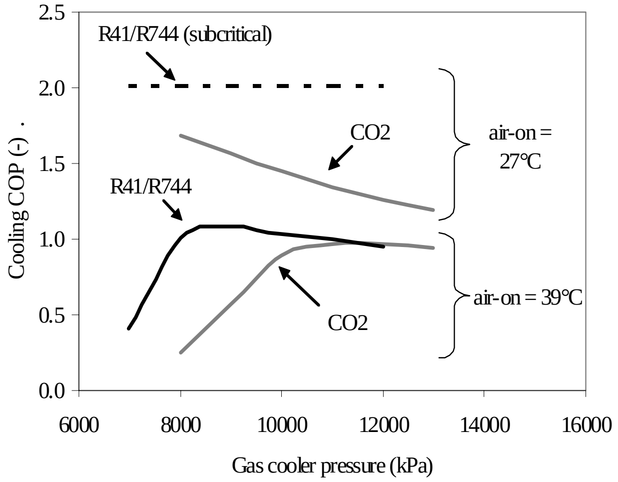

Figure 8: COP comparison between CO_2 and R-41/R-744 over a range of pressures and temperatures

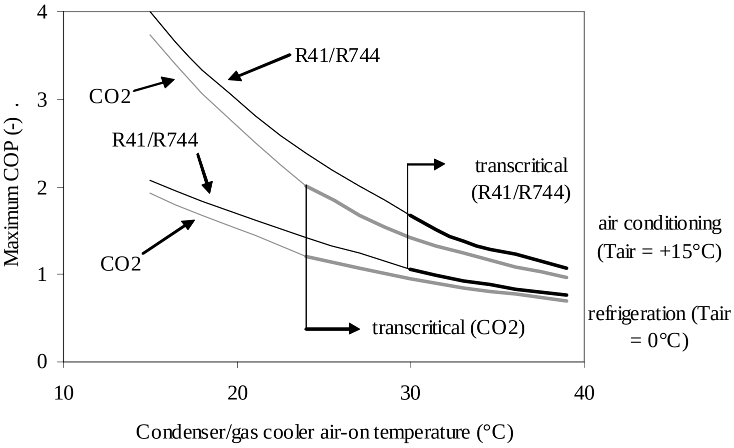

Figure 9: Comparison of maximum COP between CO_2 and R-41/R-744 under sub-critical and transcritical operation

When considering the difference between the optimum gas cooler pressure of the two refrigerants the important distinction is that the R-41/R-744 blend requires some 3,000 kPa lower operating pressure than pure R-744. A commercial benefit arises from this reduced high-side pressure, allowing a larger number of standard refrigeration components to be used, and the use of silver soldered joints with seamed mild steel pipe without incurring the penalty of the relatively high refrigerant leakage rates observed with most pure R-744 systems.

6. Final Remarks

Using a combination of novel property modelling, safety analyses and comprehensive system simulation, a number of previously unidentified azeotropic and near-azeotropic blends have been identified for use in certain applications where existing refrigerant options are subject to a variety of hindrances. A summary of the characteristics of these new blends is provided in Table 4.

ECP410A performance evaluations demonstrate that the chosen mixture provides an excellent alternative to R-410A, for applications where R-22 has been traditionally used. It has a low GWP of around 7. It has a normal boiling point of approximately -49°C. Significantly, this mixture possesses a notably high critical temperature approaching 95°C, which offers a major advantage when used in systems that operate under high ambient temperatures or for heat pump applications.

ECP717 is an azeotrope with a saturation pressure greater than R-717, higher refrigerating capacity, lower compression ratio, and reduced compressor discharge temperature. It is considered to be particularly applicable for industrial process, food and blast freezing applications

ECP744 is a near-azeotrope which shows promising improvements in system efficiency compared to pure R-744, in both sub-critical and transcritical operation, whilst also operating at significantly lower pressures. It is considered to be particularly applicable to commercial point-of-sale refrigeration and vehicle air conditioning equipment.

These new blends offer notable advantages over existing refrigerants, in particular:

- Zero ODP and low GWP, below 150, and mainly "naturally" occurring
- Improved thermodynamic properties (such as critical temperature and minimal temperature glide) over similar existing refrigerants
- Good solubility with oils
- Low toxicity, and reduced flammability
- Known and understood chemical and material compatibility

Name	**ECP410A**	**ECP717**	**ECP744**
Composition (molar)	75% R-1270, 25% R161	45% R-170, 55% R717	50% R-744, 50% R-41
Molar Mass	43.6	22.9	39.0
NBP (°C)	-49.2	-89.0	-84.5
Critical temp (°C)	94.9	41.9	37.9
Freezing temp (°C)	-160	-97	-121
ATEL (ppm)	No data	550 – 570	No data
LFL (% vol)	2.7 – 2.9	4.0 – 4.2	No data
Likely safety class	A3	A2	A1
ODP	0	0	0
GWP (100)	~7	~2	~46

Table 4: Characteristics of new blends

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Venue for the evening paper is THE ARDEN HOTEL in the West Midlands

Directions

By train: **Birmingham International Rail Station**. The hotel is located a short10 minute walk from the station (Walk through industrial park car park and footpath as shown) or 5 minute taxi ride.

By air: Birmingham International Airport. There are taxis available at the airport, alternatively a free shuttle train to the Birmingham International rail station is available from which the hotel is a short 10 minute walk.

By car: There is free parking at the hotel please use hotel website for directions.

THE ARDEN HOTEL & LEISURE CLUB
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