

Potential Application of Commercial Refrigerants as Adsorbate in Adsorption Refrigeration System

Michael John¹, Cuthbert Z.M. Kimambo¹, Trygve M. Eikevik², Ole J. Nydal² and Joseph Kihedu¹

¹ University of Dar es Salaam, Dar es Salaam (Tanzania)

² Norwegian University of Science and Technology, Trondheim (Norway)

Abstract

Commercial refrigerants used for vapour compression cycle show positive adsorption capacity with various activated carbon adsorbents however most of them appeared to have high global warming potential (GWP) values. Nevertheless, refrigerants such as R1234yf, R1234ze (E), R450A, R290, R600a, R744 and R717 have been proposed as low GWP alternatives in vapour compression cycles.

Properties of some vapour compression cycle refrigerants qualify them as the good alternative adsorbate for solar adsorption refrigeration systems. Maximum adsorption capacity of 2kg/kg-adsorbent reported for commercial refrigerant R134a on activated carbon, while the maximum for classical adsorption pairs is 0.259 kg/kg-adsorbent which appears for activated carbon-methanol pair. These refrigerants will add the availability of the adsorbates, reduced scale of problems associated with material compatibility and leakages. These systems have good potential applications in off grid areas in developing countries with abundant availability of solar energy including food preservation, vaccine and medicine storage.

Keywords: Refrigerant, GWP, Adsorption refrigeration, off grid application,

1. Introduction

Refrigeration is the cooling effect of the process of extracting heat from a lower temperature heat source, a substance or cooling medium, and transferring it to a higher temperature heat sink, probably atmospheric air and surface water, to maintain the temperature of the heat source below that of the surroundings for purposes of comfort or preservation (Kreith et al., 1999). Refrigeration technology includes the classical cycle with a compressor and an expansion valve, absorption processes, adsorption processes and desiccant systems. Vapour compression cycle is the common refrigeration technology however, significant implications of refrigerants to the environment such as ozone layer depletion, greenhouse effect, global warming and huge electrical energy consumption have forced scientists to develop environmentally friendly refrigerants and refrigeration technologies (Anupam et al., 2016).

Refrigerants are used in a wide variety of heating, ventilation, air conditioning and refrigeration (HVAC&R) equipment in both industrial and domestic equipment from household refrigerator, cooling chambers and air conditioners (Benhadid-Dib and Benzaoui, 2011; Goetzler et al., 2014). The first generation of refrigerants involved whatever worked and was available, including familiar solvents and other volatile fluids of which majority were flammable, toxic, or both and some were also highly reactive included substances such as hydrocarbons, ammonia and carbon dioxide. The second generation refrigerants shifted to fluorochemicals for safety and durability which included CFCs and HCFCs which became widely used because were efficient, non-flammable and non-toxic. However, in the 1980s CFCs and HCFCs were determined to play a major role in depleting the stratospheric ozone layer, therefore phased out in the 1990s in favor of a third generation of refrigerants HFCs which presented zero ODP but along with CFCs and HCFCs were greenhouse gases that contribute to the radiative forcing of climate with significantly GWP when released to the atmosphere (Calm, 2008; Goetzler et al., 2014; Velders et al., 2009). This shifts ignited renewed interest in natural refrigerants particularly ammonia, carbon dioxide, hydrocarbons and water (Calm, 2008).

The growing international emphasis on global warming mitigation stimulated interest for the fourth generation of

low GWP refrigerants with countries like the United States, Canada and Mexico proposed an amendment to the Montreal Protocol in 2014 to reduce production and consumption of HFCs by 85% during the period 2016-2035 for Non A5 (developed) countries and the European F-gas legislation issued in 2014 which will reduce HFC consumption by 79% over the period 2016-2030 (Goetzler et al., 2014).

On other hand, adsorption refrigeration systems can utilize environment friendly refrigerants and low temperature waste heat and renewable energy sources like solar radiation and biomass derived thermal energy to generate the cooling effect (Anyanwu, 2004; El-Sharkawy et al., 2008).

2. Low GWP refrigerants for vapour compression cycle refrigeration

Various researchers are on board searching for new refrigerants which will be both efficient and environmental friendly. Velders et al. (2009) reported the hydrocarbons, ammonia and CO₂ as the suitable low GWPs refrigerants if compared to HFCs for small refrigerant charges systems where a refrigerant leak would not pose unacceptable flammability or toxicity risk, however for industrial systems with large refrigerant charges expert to managed fire and toxicity risk were required. Koyama et al. (2010) carried out drop-in experiments in order to investigate the possibility to introduce R1234ze (E) and its mixture with R32 as low GWP alternatives for vapour compression heat pump/refrigeration systems. The results obtained proved that the heating effect and COP of R1234ze (E) could be improved by adding R32 as the second component into R1234ze (E) and the mixtures found to be strong candidates for replacing R410A in domestic heat pump system. The mixture of 50% R1234ze (E) and R32 produced the COP of about 7.5% lower than that of R410A at the same heating load of 2.8 kW. Pure R1234ze (E) at 1.6 kW produced the COP value of 20% lower than that of R410A at 2.8 kW. Evaporator and condenser sides pressure drops in both HFO-1234ze(E) and its mixture with R32 were almost at the same level as R410A though the refrigerant flow rates were lower, enlargement of diameter of heat transfer tubes and connecting tubes required as compared with the case of R410A. Drop-in replacements of R1234yf and R1234ze to R134a refrigerants were also tested with no performance enhancing modifications to the refrigerators to evaluate energy consumption, results indicates that R1234yf was a suitable drop-in replacement for R134a in domestic refrigeration applications however the lower capacity of R1234ze in domestic refrigerators would need to be addressed therefore R1234ze might not be suitable for drop-in replacement (Karber et al., 2012). Table 1 show low GWP refrigerants for vapour compression cycle.

Tab. 1: Alternative Vapour Compression Refrigerant (Low GWP refrigerants)

S/N	Refrigerant	Chemical formula	ODP	GWP 100 yrs.
1	R1234yf	CH ₂ =CF ₂ CF ₃	0	<4.4
2	R1234ze (E)	CHF=CHCF ₃	0	6
3	R450A	(R134a/R1234ze commercial mixture)	NA	NA
4	R290	CH ₃ CH ₂ CH ₃ -propane	0	20
5	R600a	CH(CH ₃) ₂ CH ₃ -isobutane	0	20
6	R744	CO ₂ -carbon dioxide	0	1
7	R717	NH ₃ -ammonia	0	<1

Navarro-Esbrí et al. (2013) did an experimental analysis of a vapour compression system using R1234yf as a drop-in replacement for R134a in vapor compression system. The tests were performed by varying the condensing and evaporating pressure, superheating degree, the compressor speed and the use of IHX. The energetic comparison was performed on the basis of the cooling capacity, volumetric efficiency, compressor power consumption and the COP. The energy performance parameters of R1234yf in a drop-in replacement were close to those obtained with R134a at high condensing temperatures and making use of an IHX. The cooling capacity of R1234yf was 9% lower in an R134a refrigerant facility in the tested range of which difference decreased when the condensing temperature increases and when an IHX was used. The volumetric efficiency was about 5% lower for R1234yf compared with R134a. Also, the compressor volumetric efficiency using R1234yf showed a greater dependence on the compressor speed. The values of the COP obtained using R1234yf were between 5% and 30%

lower than those obtained with R134a. However, when the condensing temperature raises from 313.15 K to 333.15 K the difference decreases from 25% until 8%, even more when using an IHX.

Mota-Babiloni et al. (2014) did energy performance evaluation of two low GWP refrigerants, R1234yf and R1234ze (E) as drop-in replacements for R134a. Tests were carried out in a monitored vapour compression system combining different values of evaporation and condensation temperature without/with the adoption of an internal heat exchanger. The volumetric efficiency, cooling capacity and COP taking R134a as baseline were analyzed with results showed that without IHX the average volumetric efficiency for R1234yf and R1234ze was 4% and 5% lower compared with R134a. The cooling capacity obtained with R1234yf and R1234ze was reduced, with an average difference of 9% and 30% without IHX. Also, COP values were about 7% lower for R1234yf and 6% lower for R1234ze than those obtained using R134a. The use of an internal heat exchanger reduced the COP differences for both replacements as shown in Table 2.

Tab. 2: R1234yf and R1234ze (E) as drop-in replacements for R134a without IHX (Mota-Babiloni et al., 2014)

Refrigerant	Volumetric efficiency	Cooling Capacity	COP
R134a (baseline)	100%	100%	100%
R1234yf	4% lower (96%)	Reduced 9%	7% lower (93%)
R1234ze(E)	5% lower (95%)	Reduced 30%	6% lower (94%)

Mota-Babiloni et al. (2015a) presented a drop-in analysis of an internal heat exchanger (IHX) used in a monitored vapour compression system. They compared R134a and two of its alternatives, R1234ze and R450A (R134a/R1234ze commercial mixture). Study, concluded that IHX produced benefits for all refrigerants tested and could be considered its use for R450A as drop-in or retrofit R134a replacement and new design installations using R1234ze. However, without IHX in an R134a system, alternative R450A would overcome the R134a COP results but for R1234ze more system modifications would be needed. Experimental performance of non-flammable refrigerant R450A (contrary to R1234ze and R1234yf) as R134a replacement in a vapour compression system under a wide range of operating conditions were studied by Mota-Babiloni et al. (2015b). The cooling capacity of R450A found to be slightly lower than those obtained with R134a (6% lower as average). The COP of R450A found to be 1% average higher than R134a due to compressor power consumption for R450A being much lower than R134a. The IHX affected positively the R450A energy efficiency in a similar proportion to R134a. The discharge temperature of the alternative was found to be lower, 2K as average than of R134a. R450A could be used directly in R134a systems with slightly lower cooling capacity and similar COP, however redesign and optimisation would lead to better energy performance and hence, lower power consumption.

Janković et al. (2015) reported the analysis of R1234yf and R1234ze (E) as drop-in replacements for R134a in a small power refrigeration system. The first analysis was based on equal evaporation and condensation temperatures before and after the refrigerant replacement. The second analysis was carried out for equal cooling medium conditions in the condenser, so that the transport properties and the heat transfer features in the condenser were considered for the three refrigerants. Results showed that different conclusions may be drawn if the drop-in analysis was carried out for equal condensation temperatures or for equal temperatures of the cooling medium in the condenser, as well as that these results were affected by the condenser design. R1234yf was found as an adequate drop-in refrigerant for R134a, but R1234ze (E) may perform better when used an overridden compressor to match the refrigerating system cooling power. Sethi et al. (2016) evaluated the performance of R1234yf and R1234ze (E) in a representative vending machine system and compared against the baseline refrigerant R134a as promising replacements for new small refrigeration systems. In the theoretical analysis, it was estimated that R1234yf has pressures similar to R134a whereas R1234ze (E) has pressures lower than R134a. Based on actual drop-in system testing, it was found that R1234yf shows capacity and efficiency similar to R134a as estimated based on thermodynamic properties. R1234ze (E) was estimated to have about 25% lower capacity. To match the capacity of R134a larger compressor was needed as result lower efficiency observed due to higher pressure drop in the system. R1234yf may show even better performance by using a slightly larger diameter suction line and employing a suction line liquid line heat exchanger in new systems. R1234ze (E) may show performance similar to R134a by use of a compressor with larger displacement, a slightly larger diameter suction line and by increasing the number of refrigerant circuits in the heat exchangers.

Brown et al. (2010) reported thermodynamic properties of eight fluorinated olefins, namely: R-1225ye(E), R-1225ye(Z), R-1225zc, R-1234ye(E), R-1234yf, R-1234ze(E), R-1234ze(Z), and R-1243zf. The group contribution method were used to predict the critical temperatures, critical pressures, critical densities, acentric factors, and ideal gas specific heats at constant pressure. The Peng-Robinson equation of state was used to predict thermodynamic properties and the results were presented in pressure-enthalpy and temperature-entropy state diagrams. Higashi (2010) presented the thermo physical properties of R1234yf and R1234ze (E). He predicted the difficulties of finding the next generation refrigerant from pure substances hinted that, the combination among HFCs, HFOs, and HCs should be expected in the next generation refrigerant. Lai (2014) described the thermodynamic properties of the R1234ze (E) with the molecular based BACKONE and PC-SAFT equations of state (EOS). The investigation indicated that the relative deviations among corresponding cycles' characteristics were mostly within 1% compared with multi-parameter EOSs. Thus, molecular based EOSs could be used in the investigation of a refrigeration cycle in case of no multi-parameter EOS due to the insufficient numbers of accurate experimental data in full fluid region. For the refrigeration cycle using R1234ze (E), R1234yf, R22, R134a, and R32 as refrigerants, R1234ze (E) and R1234yf were reported to be potential alternative refrigerants for the replacement of R134a.

Mota-Babiloni et al. (2015c) reviewed the current status of commercial refrigeration for food freezing and conservation in retail stores and supermarkets as one of the relevant energy consumption sectors. They reported that new GHG regulations impose strong GWP limitations that were going to phase out currently used HFC refrigerants in commercial refrigeration (R404A and R507A). In low charge applications propane, showed a good performance, while low GWP HFC or HFC/HFO mixtures as drop-in or retrofit replacements (with little system modifications), and CO₂ systems in trans-critical systems or at the low-stage of cascade systems were proposed as the different alternatives. Mota-Babiloni et al. (2016) reviewed thermo physical and compatibility properties, heat transfer, pressure drop characteristics and vapour compression system performance of R1234ze (E) HFO refrigerant. Pure R1234ze (E) was found to be a good option only in new HVACR systems. Nevertheless, if it was combined with other refrigerants, the final GWP value was also considerably reduced and maintaining the efficiency parameters at levels that allow them to replace R134a, R404A or R410A in existing systems with minor modifications.

There are importance of proper selection of refrigerant (Calm, 2012). Most refrigerant were selected as per the application, the environmental and physical properties together with performance parameters. Global warming issues suggested to phase out presently most used refrigerant R134a and use natural refrigerants such as ammonia, carbon dioxide and hydro carbons (isobutene R600a and propene R290) in vapour compression refrigeration system for sustainable environment. R1234yf also reported to be a suitable candidate for refrigeration and air conditioning systems. However eco-friendly technologies like thermo-electric, magnetic and adsorption refrigeration will receiving more attention as energy environmental problems keep increasing (Bhatkar et al., 2013)

3. Performance of commercial refrigerants as adsorbate in adsorption refrigeration system

Several researchers have reported the performances of commercial vapour compression refrigerant in adsorption studies. Akkimaradi et al. (2001) presented adsorption isotherms for R134a on activated charcoal, in the temperature range of 273-353 K and pressures up to 0.65 MPa using volumetric method. Experimental isotherm data were obtained for three specimens of activated charcoal (Chemviron, Fluka and Maxsorb Charcoal) adsorbing R134a. A mathematical description of the isotherms was provided by using the D-A equation and the isosteric enthalpy of adsorption evaluated from the transformed isotherm data. Habib et al. (2010) measured adsorption rates of R134a and R507A onto pitch based activated carbon of type Maxsorb III with temperatures varying from 20-60 °C. The experimental data were found to match fairly with the Fickian diffusion model while the surface diffusions followed the classical Arrhenius trend for both pairs. It was noted that Maxsorb III can adsorb R134a as high as 1.6 kg/kg within an adsorption time interval of 1200 s at an adsorption temperature of 25 °C, while R507A can adsorb as high as 1.3 kg/kg within an adsorption time interval of 1100 s at adsorption temperature of 20 °C.

Askalany et al. (2013a) designed and built an adsorption cooling system using granular activated carbon GAC-R134a pair. The theoretical mathematical model results presented good agreements with experimental results. COP was theoretical 0.35 at 373 K driving temperature and 293 K evaporator temperature. The system operated between the pressures of 3.3 bar and 7 bar where the difference in the adsorption uptake reached up to 0.3 kg/kg and time of the intermittent cycle of about 900 s. The experimental SCE was up to 70 kJ/kg, while the theoretical SCE was 83 kJ/kg at 373 K driving temperature and 295 K evaporator temperature. SCE and COP was increased by raising the driving or evaporator temperatures. Increase of cycle time increased COP and SCE at constant evaporator temperature and decreased COP and SCE at constant driving temperature. Attalla et al. (2014) investigated experimentally the adsorption capacity of R134a on a GAC for adsorption/desorption process. The bed was designed and built with finned tubes heat exchanger to increase the heat transfer area. The experiments were conducted over a temperature range from 20 °C to 60 °C and pressure up to 10 bars. The data were correlated with D–A equation corresponding to adsorption/desorption process. The maximum adsorption capacity uptake found to be 1.92 kg/kg at 20 °C after 1200 s, later Attalla and Sadek (2014) found the isosteric heat of adsorption which varied from 120 to 340 kJ/kg for adsorption capacities ranging from 0.2 to 1.8 kg/kg-adsorbent. AquaSorb 2000-R407C pair was tested over a temperature range of 25-75 °C, maximum adsorption capacity of 0.43 kg·kg-1 of adsorbent was obtained (El-Sharkawy et al., 2016).

Askalany et al. (2013b) reported adsorption isotherms for refrigerant R32 on activated carbon powder (ACP) and fiber (ACF) forms over the range of 25-75°C and pressures up to 1400 kPa, measured using constant volume apparatus. The data were correlated by Toth and Dubinin-Astakhov with the adsorbed phase volume correction isotherm models. Adsorption parameters were evaluated from the least-squares fit of experimental data, whereas the D-A equation with volume correction fitted the data better. The uptake and pressure dependence of the isosteric heat of adsorption was extracted from the D-A equation using the Clapeyron equation. ACP observed to have better adsorption than ACF. Adsorption isotherms and kinetics of R410A onto ACP (Maxsorb III) and ACF (A-20) studied by Askalany et al. (2014), the maximum adsorption capacity of the ACP was found to be 1.6 times higher than that of ACF at same adsorption temperature. Both investigated adsorption isotherms (Tóth and D-A) models and kinetics (LDF) models have been found to fit the experimental data within ±5% deviations. D-A equation was used to determine the isosteric heat of adsorption pairs. Adsorption isotherms and kinetics were also investigated experimentally and theoretically over a temperature range of 25-75 °C for Maxsorb III-R152a and AquaSorb 2000-R404A pairs (Ghazy et al., 2016a; Ghazy et al., 2016c). Table 3 shows the commercial vapour compression refrigerant reported on adsorption studies.

Tab. 3: Commercial Vapour Compression Refrigerants used in Adsorption Research

S/N	Refrigerant	Chemical formula	Adsorption capacity kg/kg	ODP	GWP 100 yrs. (CO ₂ =1)
1	R134a	CH ₂ FCF ₃	1.6-2.0 with ACP	0	1,370
2	R32	CH ₂ F ₂ -methylene fluoride	1.29 ACP/0.94 with ACF	0	716
3	R404A	R-125/143a/134a (44.0/52.0/4.0)	0.52 with AquaSorb 2000	0	3,700
4	R507A	R-125/143a (50.0/50.0)	1.3 with ACP	0	3,800
5	R410A	R-32/125 (50.0/50.0)	NA	0	2,100
6	R152a	CH ₃ CHF ₂	1.3 with ACP	0	133
7	R407C	R-32/125/134a (23.0/25.0/52.0)	0.43 with AquaSorb 2000	0	1,700

Askalany and Saha (2015) investigated experimentally and theoretically the adsorption kinetics of Difluoromethane (R32) onto activated carbon powder (Maxsorb III) and activated carbon fiber (A-20) at various adsorption temperatures ranging from 25-65 °C. Two commonly used theoretical models (LDF and FD) fitted experimental data well within ±5% error. Jribi et al. (2013) reported a transient mathematical model of a 4-bed adsorption chiller using Maxsorb III as the adsorbent and low GWP refrigerant R1234ze (E). The performance of

the cyclic-steady state of the system for different heating and cooling water inlet temperatures presented. With 80 kg of Maxsorb III the chiller produced 2 kW of cooling power at driving heat source temperature of 85 °C along with a cooling temperature of 30 °C, the performance which compared slightly higher than that of the Maxsorb III-R134a based adsorption cooling cycle. Consequently propose R1234ze (E) as potential replacement of R134a based adsorption cooling system. Ghazy et al. (2016d) reported adsorption isotherms and kinetics of HFC-152a onto activated carbon of type Maxsorb III over a temperature range of (25 to 75) °C. The D-A and Toth equations were used to correlate the adsorption isotherms with D-A found to be more suitable. Maxsorb III adsorbed up to 1.3 kg of HFC-152a per kg of adsorbent. Adsorption kinetics data were correlated using LDF and FD models and isosteric heat of adsorption was estimated. Activated carbon/CO₂ pair reported to show the maximum COP of 0.16 for evaporation and desorption temperatures of 15°C and 80°C (Jribi et al., 2010).

Ghazy et al. (2016b) investigated experimentally and theoretically over a temperature range of 25-75 °C adsorption isotherms and kinetics of AquaSorb 2000-R404A pair. D-A and Tóth equations were used to fit equilibrium uptake while adsorption kinetics was correlated using LDF and FD models. The experimental maximum adsorption capacity of AquaSorb 2000-R404A pair was about 0.52 kg/kg. The activation energy and the pre-exponential coefficient were estimated to be 10488.49 J/mol and 1.11 respectively. Using the Clausius-Clapeyron equation, Isosteric heat of adsorption found to vary from 145 to 330 kJ/kg depending on the adsorbate loading. Both LDF and FD models found suitable to simulate the adsorption kinetics of AquaSorb 2000-R404A with an acceptable error of ±5%. The D-A equation was used to fit the P-T-C diagram of AquaSorb 2000-R404A.

Classical pairs which includes activated carbon-methanol, activated carbon-ethanol, activated carbon-ammonia, silica gel-water and zeolite-water reported to have the maximum adsorption capacity of 0.259 kg/kg-adsorbent which occurs on activated carbon-methanol pair, while the commercial refrigerant R134a presented maximum adsorption capacity of 2 kg/kg-adsorbent with activated carbon adsorbent (Shmroukh et al., 2015). The application of commercial refrigerants as adsorbates will positively contribute to adsorption refrigeration systems as it will add the availability of the adsorbate, reduce the problems associated with material compatibility like on ammonia with copper and reduced leakage problems associated with the negative pressure systems (Zhong et al., 2006). These adsorption refrigeration systems have good potential for off grid applications in developing countries with abundant availability of solar energy including food preservation, vaccine and medicine storages.

4. Conclusions

The potential for adsorption refrigeration system using the commercial refrigerants is evident. The maximum adsorption capacity for the classical working pairs (activated carbon-methanol, activated carbon-ethanol, activated carbon-ammonia, silica gel-water and zeolite-water) of 0.259 kg/kg which appeared for activated carbon-methanol is small compared to adsorption capacity of 2 kg/kg-adsorbent which appeared for modern adsorption working pair of activated carbon-R134a (Shmroukh et al., 2015).

Commercial refrigerants such as R134a, R32, R404A, R507A, R410A, R152a and R407C have been studied as the potential adsorbates for adsorption cooling systems or for mixture separation in the blended refrigerant (Akkimaradi et al., 2001; Askalany and Saha, 2015; Askalany et al., 2013a; Askalany et al., 2014; Askalany et al., 2013b; Attalla and Sadek, 2014; Ghazy et al., 2016b; Ghazy et al., 2016d; Habib et al., 2010). Though they have shown the positive adsorption capacity with various type of activated carbon adsorbents, they also possess high GWP values (Calm and Hourahan, 2007).

Several refrigerants including R1234yf, R1234ze (E), R450A, R290, R600a, R744 and R717, have been proposed to be possible alternatives or drop in replacement of commercial high GWP refrigerants in vapour compression cycles (Bhatkar et al., 2013; Brown et al., 2010; Higashi, 2010; Koyama et al., 2010; Lai, 2014; Mota-Babiloni et al., 2015a, 2015b; Mota-Babiloni et al., 2015c; Navarro-Esbrí et al., 2013; Sethi et al., 2016). These refrigerant could be the good alternative for adsorption pairs for solar adsorption refrigeration system. And therefore increase the availability and options for the adsorption refrigerants, reduced scale of problem associated with material compatibility as is there ammonia system and copper, positive pressure system therefore avoiding the leakage problem associated with the vacuum systems.

Besides the adsorption properties, it is also important to understand the kinetics of these adsorption pairs before

the conclusions of best adsorption pairs as the kinetics data are essential and useful in designing adsorption refrigeration systems.

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