Supplementary Information (SI) for Environmental Science: Processes & Impacts.

Supporting Information to "Finding non-fluorinated alternatives to fluorinated gases used as refrigerants"

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Contents

1) Definition of general terms

Air-conditioning: process of treating air to meet the requirements of a conditioned space by controlling its temperature, humidity, cleanliness or distribution (taken from EC 2024[1]).

Chiller: single system whose primary function is to cool a heat transfer fluid (such as water, glycol, brine or CO_2) for refrigeration, process, preservation or comfort purposes (taken from EC 2024[1]).

Heat pump: piece of equipment capable of using ambient heat or waste heat from air, water or ground sources to provide heat or cooling and is based on the interconnection of one or more components forming a closed cooling circuit in which a refrigerant circulates to extract and release heat (taken from EC 2024[1]).

Refrigeration: process of maintaining or lowering the temperature of a product, substance, system or other item purposes (taken from EC 2024[1]).

2) Non-fluorinated gases as refrigerants

2.1) Propane (R-290)

Propane has a GWP of 0.02 (reference [2]), zero ODP and does not form by-products or decomposition products in the atmosphere. The boiling point of propane is at -42.1 °C, the critical point at 96,81 °C and 4.2 MPa (reference [3]).

Propane is considered flammable with a safety group classification according to standard EN 378-1 of A3.[4] The safety classification means that certain measures have to be taken when handling propane and that there are charge limits for using propane in technical applications. The most relevant European standards for refrigeration, air conditioning, and heat pumps are EN 378, the product standard IEC EN 60335-2-40 for air-conditioning systems, and the product standard IEC EN 60335-2-89 for integral and remote commercial refrigeration applications.[5] The charge limits set in these standards are meant to handle the risk of the high flammability of propane and other A3 refrigerants.[6] Other measure that have to be taken into account are ensuring that there are no sources of heat at the workplace that could create flames, and no switches or devices that create sparks.[7] Moreover, static electricity should be avoided, and it must be guaranteed that the work area is ventilated properly.[7]

Arguments that propane is unsafe in transportation (e.g., cars or airplanes) seem unsubstantiated given that cars and airplanes carry dozens to hundreds of kilograms of other flammable fuels. However, the aviation industry has set strict regulations that do not allow the use of flammable refrigerants in airplanes.

2.2) (Iso)butane (R-600a)

Isobutane has a GWP of 0.006 (reference [2]), zero ODP and does not form by-products or decomposition products in the atmosphere. The boiling point of (iso)butane as at -11.7 °C, the critical point at 134,69 °C and 3.58 MPa (reference [3]). Like propane, it is considered flammable and has a safety classification of A3. Isobutane has been used widely in domestic refrigeration since 1993 and its use can be considered safe there.

2.3) Ammonia (R-717)

Ammonia is a strong-odor gas with zero GWP and zero ODP. Due to its toxicity, ammonia has a safety-group classification of B2L according to standard EN 378-1.[4] It has a short atmospheric lifetime and does not form any by-products or decomposition products with negative environmental impact.[8] The boiling point of ammonia is at -33.35 °C and the critical point at 132.4 °C and 11.15 MPa (reference [3]).

Compared to most other working fluids under most operating conditions, ammonia yields the highest energy efficiency.[9,10] However, efforts to further increase the energy efficiency of the vapor-compression cycle with ammonia have been limited in recent years. This is changing now and more research is currently under way to improve the energy efficiency of the systems and to reduce the specific refrigerant charge of ammonia systems.[9,11] Arguments that have been brought forward against the use of ammonia are that ammonia is flammable, corrosive, toxic if inhaled, and very toxic to aquatic life.[12]

The high toxicity of ammonia can indeed lead to hazardous situations in confined spaces and ammonia can also harm the health of workers and the public.[13] However, systems with ammonia can be designed to prevent leakage and minimize risks.[14] Measures include for example the use of welded joints, hermetic or semi-hermetic compressors, use of shell and plate heat exchangers as condensers and chillers, or installations of the system on a rooftop.[14] In addition, technical personnel need to have adequate training and follow basic safety procedures. It is also important that the system is frequently checked, that it is protected from external damage, and that the alarm system is well adjusted.[14]

When ammonia is shipped and stored as a liquid, any release into the atmosphere will begin evaporating immediately, resulting in a rapid lowering of concentration below the flammable range.[15] Ammonia that is mixed with water is also not corrosive to iron or steel. Care should be taken to not bring ammonia that includes moisture or water in contact with other metals such as zinc, copper or brass as ammonia (even mixed with water) as ammonia is corrosive to these metals. However, even the International Space Station is applying ammonia in its cooling system,[16] so it is possible to handle it safely.

2.4) Water (R-718)

Water is an odorless liquid without toxicity, zero ODP and a GWP < 1 (reference [17]). Water does not cause any safety issues and there is no disposal problem. The boiling point of water is at 100 °C, the critical point at 373.9 °C and 22.1 MPa (reference [17]). Water has a very large latent heat of vaporization which is positive when used as refrigerant.
However, systems with water vapor compressors have to satisfy the requirements of large pressure ratio, large volume flow rate and high exhaust temperature.[17,18] Also, systems with water phase problems of rust and corrosion of components as well as intrusion of lubricating oil which proposes higher requirements for the design and manufacture of water vapor compressors.[17]

Having a high exhaust temperature, water is a good working fluid in medium to high temperature heat pumps. Compared to CFCs, water has a high coefficient of performance.[19]

2.5) CO_2 (R-744)

CO_2 is an odorless, non-flammable gas with low toxicity, a GWP of 1 and zero ODP. It has an A1 safety classification according to standard EN 378-1. With a long atmospheric lifetime, CO_2 does not lead to any byproduct formation. The boiling point of CO_2 is at -78.46 °C (reference [3]), the critical point at 31.1°C and 7.38 MPa (reference [20]).

When used as a refrigerant, CO_2 typically operates at a higher pressure than fluorocarbons and other refrigerants, which makes it less suitable for large-scale applications.[21] This is also shown by the fact that e.g., only few large-scale heat pumps (with more than 1-2 MW) use CO_2.[21] However, more than 5 million Eco-Cute heat pumps, operating with CO_2 as working fluids, are installed in Japanese homes to produce the daily demand of hot water. Also, more than 100,000 transcritical refrigerant systems are installed globally in supermarkets and CO_2 is the preferred, energy efficient and safe fluid there. However, in the case of leaks in indoor installations, CO_2 accumulates on the ground and can lead to suffocation. Leak detection equipment with alarm functions is mandatory for machine rooms and cold storage rooms. Areas of application are food retail, deep-freezing in the industrial refrigeration sector, mobile air conditioning and service water heat pumps. System types include cascades, transcritical process control and cooling units.

2.6) Other Hydrocarbons

Other hydrocarbons such as ethane (R-170), ethene (R-1150), propene (R-1270), and methane (R-50) are also used as refrigerants. They have similar properties to propane and (iso)butane meaning that they have a very low GWP, high flammability and do not form by-products. The main problem with these refrigerants is the low critical temperature. Ethane has a critical temperature of 32.2°C, ethene of 9.19°C, methane of -82.3 °C. Only propene has a relatively high critical temperature with 96.8°C. This means that the system has to operate with a transcritical cycle which leads to high pressures and high component costs. However, these refrigerants are very suitable for smaller systems, as they have a greater volumetric cooling capacity than the other refrigerants.

2.7) Helium and nitrogen

Helium and nitrogen are non-combustible gases that have zero ODP and zero GWP. Both gases do not form by-products or decomposition products. The boiling point of helium is at –268.93°C, the critical point at -267.95 °C and 0.228 MPa (reference [3]). The boiling point of nitrogen is at -195.79 °C, the critical point at -147.1 °C and 3.39 MPa (reference [3]). Exposure to very high levels of pure nitrogen or helium can lead to suffocation.[22,23]

3) Refrigerant cycle and criteria to select a refrigerant

3.1) Refrigerant cycle

A simple refrigerant cycle includes the following phases:

Compression: The compressor sucks in the refrigerant in gaseous form and at a low temperature and low pressure level and compresses it. During this compression process, the pressure and temperature of the gas increase significantly.

Condensation: The compressed, hot refrigerant flows through the condenser, a heat exchanger that releases the heat of the refrigerant into the environment (e.g. room to be heated). This process takes place at approximately constant pressure. As the heat is released, the refrigerant condenses and becomes liquid, although its temperature is still above the ambient temperature.

Expansion: The liquid refrigerant then enters an expansion valve where it experiences an abrupt drop in pressure. This throttling leads to an approximate adiabatic expansion in which the refrigerant partially evaporates and cools down significantly. After this process, the refrigerant is a low-pressure, low-temperature mixture of liquid and vapor.

Evaporation: The cold refrigerant mixture circulates in the evaporator, another heat exchanger. Here it absorbs heat from the environment (e.g. from the air to be cooled or a product). This heat absorption causes the refrigerant to completely evaporate and transform into a lower pressure, higher temperature gas. The refrigerant then returns to the compressor and the cycle begins again.

The determination of which heat exchanger serves as the 'useful' side in the cycle depends on the specific application – whether the goal is to impart heat (via the condenser) or absorb it (via the evaporator). For enhanced efficiency and performance in various applications, the cycle may incorporate additional components like economizers, subcoolers, or variable speed drives.

3.2) Choice of refrigerant

Finding a suitable refrigerant for the individual system is complex because chemical, ecological and thermodynamic properties must be weighed balanced.[24] Considerations that should be taken into account are: a) The refrigerant must be chemically stable and soluble in oil so that the system can be operated. b) The refrigerant must not have an OPD and the GWP should be very low. The refrigerant should also not be toxic or special safety precautions must be taken that still enable its use. c) The use of HFOs should be avoided as HFOs can form TFA and the rising levels of TFA may lead to unforeseeable consequences in the future. d) The highest possible efficiency should be achieved to minimize the environmental impact. e) Temperature and pressure level of the refrigerant must be suitable for the respective application. A pressure level below atmospheric pressure will result in seal failure and diffusion of outside air into the circuit, significantly reducing efficiency.[24]

In regard to point d) – It has been shown that the electricity demand of a heat pump or refrigeration machine has the largest share of the total environmental impact of the system in most of the impact LCA categories.[25] A comparison of the efficiency of systems with different refrigerants showed that the highest efficiency can be expected with natural refrigerants.[24]

4) Technical description of the refrigerant systems

4.1) Refrigeration

Domestic refrigeration

Refrigerators and freezers for private households are operated at almost stationary operating points. The secondary side of the condenser is around 20°C (room temperature) and the secondary side of the evaporator is around 8°C or –18°C (depending on whether it is a refrigerator or freezer).

Stand-alone refrigeration systems in commercial stores

Stand-alone refrigeration systems are hermetically sealed refrigeration circuits that are simply plugged to electrical power without any further installation.[26] Stand-alone systems are often used in smaller stores but have been proven technically feasible in larger stores as well.[26] The secondary side of the condenser is around 20°C (room temperature) and the secondary side of the evaporator (inside the refrigerator) is between 0°C and 10°C.[27]

Central refrigeration systems in commercial stores

Central refrigeration systems provide the refrigeration capacity for the entire store centrally in one location, usually a machine room. Heat is extracted from the refrigerated goods via the evaporator. The temperature on the secondary side of the evaporator is between -18 °C and 10 °C, depending on the refrigerated goods. The condenser transfers the heat extracted from the refrigerated goods to the environment. Consequently, the condenser is often located on the roof of the building. Thus, the temperature of the secondary side is the same as the ambient temperature and therefore fluctuates depending on the location. [27] These kinds of system architectures are the majority of refrigeration systems currently installed in larger supermarkets.[26] Depending on the system, the refrigerant can be used for direct evaporation or with a heat transferring system such a glycol loop.

Industrial refrigeration

Industrial refrigeration includes cold storage in warehouses, cooling onboard ships, and process cooling. The temperature of the secondary side of the condenser depends on the source used (water or air). The secondary site of the evaporator could be as low as -60 °C.[28]

Transport refrigeration of goods

Transport refrigeration units are often installed in trucks, trailers, containers, or trains to keep goods within a compartment at a desired temperature level. The secondary side of the condenser can vary (ambient temperatures may range from -40 °C to 40 °C), while the secondary side of the evaporator should always be around 8 °C or -18 °C (depending on if used as refrigerator or freezer).

Ultra-low and low temperature freezer (temperatures below −20 °C)

Ultra-low temperature freezer (secondary site of evaporator: −100 °C to −50 °C) and low temperature refrigeration (secondary site of evaporator: −20 °C to −50 °C)[29] is often required to store biological samples as well as vaccines and biopharmaceuticals products. The working fluids typically used are those that can reach evaporation temperatures below −80 °C while operating above the atmospheric pressure at the suction point.[29]

4.2) Indoor Climate

Heat pumps in building energy systems

A heat pump, which is used for heating in buildings, works with a condenser inside the building and an evaporator outside. By integrating a four-way valve, the heat pump circuit can be reversed, swapping the functionalities of the condenser and evaporator. This allows to switch between heating and cooling mode, depending on the respective requirements.

The secondary fluids are selected depending on the specific requirements of the system and the respective environmental conditions. The choice of secondary fluid significantly influences the operating range and temperature levels of the heat pump.

Within building interiors, water is predominantly used as a secondary fluid. The required temperatures vary based on the application and country:

- Underfloor Heating: This requires supply temperatures between 35°C and 40°C. These lower temperatures enhance efficiency and contribute to thermal comfort.
- Radiators: Radiators necessitate higher temperatures ranging from 45°C to 90°C, depending on the heating system and structural considerations.
- Domestic Hot Water: Temperatures between 50°C and 70°C are necessary for heating domestic hot water to meet hygiene standards.

As an alternative to water, some systems employ air, especially for supply air systems, with temperatures ranging from 25°C to 52°C.[30]

Outside the building, various sources are utilized for heat or cold generation:

- Outdoor Air: With temperatures ranging from –20°C to 40°C, it exhibits the most significant fluctuations. Utilizing outdoor air is the least efficient, especially when there is high heat demand during low external temperatures, creating a counterintuitive relationship.
- Geothermal/Sole: Typically ranging from 0°C to 10°C, a significant drawback lies in the extensive retrofitting and higher investment costs due to substantial groundwork.
- District Heating Networks: These provide a constant temperature of around 8°C to 10°C.
- Groundwater: With temperatures between 8°C and 12°C, it offers a stable heat source, but its utilization involves a complex approval process.

In the field of building heating and cooling, heat pumps are commonly configured as either monobloc or split units. Monobloc units are compact systems in which all essential components are combined into a single unit. This typically includes the compressor, heat exchanger, expansion valve, and fan. [31]

- Installation: The installation is relatively simple as all components are integrated into one unit. They only require a connection to the electrical network and, depending on the application, to air ducts or the water circuit.
- Applications: Commonly used in portable air conditioning units, smaller heat pumps, and compact air conditioning systems.
- Advantages: Easy installation and maintenance, no need for refrigerant lines between indoor and outdoor units.
- Disadvantages: Often less efficient than split systems, especially in larger applications. Noise levels can be higher since all components are concentrated in one area.

Split units divide the main components of the system into two separate units: the indoor and the outdoor unit.

- Installation: More complex, as it requires a connection between the indoor and outdoor units through refrigerant lines.
- Applications: Frequently used in residential and commercial buildings for air conditioning and heat pump systems.
- Advantages: Higher efficiency and better performance, especially in larger systems. Lower noise pollution indoors, as the compressor and other noisy components are located outside.
- Disadvantages: More complex installation and maintenance. Refrigerant lines between the units are necessary.

District heating network

Large heat pumps can provide hot water (60°C to 70°C) and space heating (35°C to 45°C) for entire districts and neighborhoods. The heat pumps can use air, water, geothermal energy or waste heat as energy sources.[32]

Air conditioning

In air conditioning systems, the evaporator is inside to cool the room by transferring the heat to the refrigerant cycle while the condenser is outside to release the heat.

Plug-in room air-conditioning
Movable air conditioners are hermetically sealed and can be moved between rooms by the user. They are mostly used in private households.[33]

Stationary single and multiple split air conditioning
Single-split air conditioning systems consist of one outdoor and one indoor unit linked by refrigerant piping. These systems are installed on site and are used predominantly in private households.[33] We differentiate here between single-split systems with less than 3 kg of refrigerant and those with more than 3 kg of refrigerant, according to the current F-Gas regulation.[1] Multiple-split systems consist of one outdoor unit and multiple indoor units and are mainly used in commercial facilities.[33] Multiple-split systems include also Variable Refrigerant Flow (VRF) systems and Variable Refrigerant Volume (VRV) systems.[34]

Mobile air conditioning in vehicles on the ground
Mobile air conditioning are systems that cool the air in cars, buses, and trains.

Mobile air conditioning in aircrafts
There are two types of air conditioning systems commonly used on aircraft. Air-cycle air conditioning systems are used on most turbine-powered aircraft.[35] They use atmospheric air as working fluid and no phase change is required during the thermodynamic cycle.[36] Vapor-cycle air conditioning systems, on the other hand, are often used on reciprocating aircraft (and sometimes in turbine-powered aircraft).[35] Vapor-compression cycle systems are based on the closed vapor-compression refrigeration cycle, the working fluid is a refrigerant that changes phase during the cycle processes.[36]

Cooling units for data center
Data centers generate a lot of heat that needs to be transferred outside of the building.

Chillers

A chiller, typically a vapor compression or absorption unit, removes heat from a secondary fluid circulated towards the equipment to provide any kind of cooling. The heat from the chiller is either transferred towards a secondary loop, which enables heat recovery, or directly transferred to the ambient air via a heat exchanger. In addition to maintaining the temperature of various industrial devices and laboratory instruments, equipment and apparatuses at a constant level, chillers are also used for air conditioning in buildings and factories.[37]

4.3) Miscellaneous

Heat pumps in industrial energy systems

Industrial heat pumps are here defined as having ≥ 100 kW heating (and or cooling) capacity. They are used in various industries including the food industry, metal working industry, chemical industry or machienery.[38] In the food industry, for example, heat pumps are used to produce hot water, hot air or process heat.[32] Heat pumps in the metal working industry are used to generate heat for the treatment of metal parts and the production of hot water.[32]

Heat pump tumble dryers

Heat pump tumble dryers use hot air to absorb moisture from laundry to get them dry after a wash.

Refrigerant air dryers and mobile dehumidifiers

Refrigerant air dryers and mobile dehumidifiers remove moisture from (compressed) air. The system works by cooling down compressed air to approximately 3°C where the water vapor condenses into water. The liquid water can then be removed from the system, and the air can be reheated to room temperature.[39] Refrigerant air dryers are very common in laundry rooms in apartment buildings e.g., in Switzerland. Mobile dehumidifiers are used to maintain a defined humidity in production processes or warehouses and to dry buildings after water damage.

Ice rinks

Refrigerant cycles for ice rinks are used to produce the ice and keep it cool. Thus, the evaporator is under the ice surface. This means that heat from the ice is transferred to the refrigerant circuit. In the condenser, waste heat is transferred to the environment or used to heat buildings.

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