Chapter

Energy efficient technologies for ultra-low temperature refrigeration

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Abstract: Sustainable transition is ongoing in many refrigeration and heat pump applications following what is established in national and international regulations. However, many of them have not controlled ultra-low temperature refrigeration (at –80 °C) because of the challenges associated with these systems' operation. This chapter presents the main characteristics of several technologies (vapor compression cycle with element variations, sublimation, or absorption cycle) that can be applied for this range of temperatures, focusing on the constructing elements, advantages and drawbacks. Then, recently developed configurations that can appear in commercial systems in the coming years are explored. These configurations are based on vapor compression cascade cycles, including an intermediate heat exchanger, ejector and three-stage. Apart from the additional elements or stages, working fluids used in these configurations are critical parameters for increasing the resulting energy performance and cooling capacity, ending with more sustainable ultra-low temperature freezers.

1 Impact, requirements, and challenges of low and ultra-low temperature refrigeration on biomedical research

Biomedical research and medical clinics need refrigerated storage to conserve critical samples, medications, vaccines, organs etc., at very precisely controlled temperatures. Regular refrigerators and freezers are not appropriate as they fail to provide uniform temperature stability. Therefore, they compromise the integrity of the stored samples and hence reproducible results or even life-threatening scenarios. Also, biomedical research can often require explosion-proof refrigerators and freezers for flammable materials. In addition to negligible temperature fluctuations, these refrigerators are highly recommended to have quick temperature recoveries, given that their doors are continually opened and closed. Moreover, room temperature samples are sometimes stored directly at ultra-low refrigerators, avoiding the thawing of neighboring samples at the unit. Also, when this equipment fails to keep the optimal temperature range, audible and visual alarms are needed to ensure the contents are protected. These readouts are often pointed reads, but bulk data recording is highly recommended for monitoring if any abnormality is found in the samples at a certain point during the long-term storage.

Regarding ~4 °C refrigerators, they store most of the daily chemical reagents used in biomedical research. They span from cell culture compounds (culture medium, L-glutamine, non-essential amino acids, gelatin, etc.) to biomolecular reagents such as antibodies, enzyme-linked immunoassays (ELISAs), flow cytometry compounds, protein and ribonucleic (RNA) extraction reagents, etc.

Regular ~–20 °C freezers host many other different samples such as stable dilutions of desoxyribonucleic acid (DNA), complementary DNA (cDNA), primers, polymerase chain reaction (PCR) reagents, enzymes, some drugs, and vaccines. However, specific samples and more complex compounds, due to their nature, need ultra-low temperatures according to thermostability studies. Examples of these types of compounds are unstable RNA solutions, liquid nitrogen (LN_2) snap-frozen tissues and protein extracts. All these samples contain a vast amount and variety of destructive enzymes called hydrolases. For instance, RNase (destruction of RNA), proteases (destruction of proteins) or phosphatases (dephosphorylation) can threaten the integrity of the samples. Conversely, at very low temperatures (around ~–80 °C), all these catalyzed degradation reactions happen more slowly; it is the same idea as freezing food in the food industry to keep it from spoiling.

Finally, on the other extreme of the temperature range, cell lines need temperatures under –135 °C for long-term storage to keep their properties and grow and multiply correctly when thawed. Cells are probably some of the most delicate tools used in a biomedicine laboratory. They need a controlled freezing gradient of –1ºC per min until they reach –80 °C to avoid ice formation injury and then transferred to freezers that reach between –196 to –135 °C temperatures. All methods used for this ultra-low temperature storage have advantages and disadvantages that must be assessed by the research center, such as the electric freezer with LN_2 back-up, liquid phase nitrogen freezers and vapor phase nitrogen freezers. Unfortunately, this type of freezer is not the most common in research centers or hospitals because not many samples require these ultra-low temperatures. Moreover, they encompass many special safety issues such as the risk of asphyxiation, thus dedicated liquid nitrogen storage areas get dangerous restricted access rooms.

As an example, recently, with the appearance of the Sars-CoV-2 pandemic and vaccines, a problem that has been present in our society for a long time has become evident, deep freezing. Vaccines can be produced by numerous different mechanisms giving rise to the wide offer of effective, safe, and lasting vaccines. They can be inactivated, live-attenuated, viral vectors, toxoids, messenger RNA (mRNA) and subunits/recombinant/polysaccharides/conjugates vaccines. Depending on the development mechanism, their storage requirements and shelf lives may be different. Storage and logistical transport at refrigerators or freezers from –20 to 8 °C are virtually feasible everywhere, except in countries with limited access to refrigerators. This is the case of Johnson & Jonhson (2 to 8 °C), Astra Zeneca (2 to 8 °C) and Moderna (from –25 to –15 °C, but it can be stored during one month at 2 to 8 °C) vaccines, which can face better the already established infrastructure of hospitals and vaccination centers and fit better the current logistical cold-chains. However, Pfizer-BioNTech vaccine based on mRNA fails to be kept at –20 °C like

Moderna, which has some stabilizing technologies such as lipid nanoparticles that allow better conservation and prevents RNA degradation as the temperature rises.

However, these technologies, together with modified stabilized nucleosides, are far from solving the cold-chain problem now after the stress testing drugmaker companies have performed and reported. Also, –80 °C freezers (Fig. 1) are not a daily basis device that can be found in hospitals at huge numbers, hence hindering, even more, the administration of these doses. The so-called ‘pizza boxes’ have been designed by Pfizer to transport 195 vials in each box in payloads with dry ice, which can store vials in the package at -80 °C for some days. This transport method is widely used to ship frozen items. Nevertheless, dry ice production and transport logistics cannot harbor these massive deliveries of doses. Additionally, dry ice cannot provide a constant and uniform temperature around the parcel, jeopardizing the integrity of the mRNA and hence provoking a life-threatening situation in a global pandemic.

Fig. 1 Biomedical refrigerator

The following chapter will recapitulate the main characteristics, advantages, and drawbacks of different configurations of ultra-low temperature refrigerators. The aim is to shed some light on not widely used but existing technologies that can provide better and cost-effective efficiencies in low-temperature freezers, aiming to provide alternatives to gold-standard refrigeration devices.

2 Configurations

There are numerous technologies in refrigeration, and they are classified then according to configurations. The concept of configurations refers to arranging the components into a circuit, obtaining different groups with typical peculiarities.

The most common configurations in refrigeration are the following: simple, indirect expansion, cascade, intermediate exchanger, ejector, multi-stage, absorption, and auto-cascade.

2.1 Single-stage

This configuration is the basis for all configurations and contains the essential elements of the refrigeration machine for a reliable and safe operation. Fig. 2 shows its four main elements.

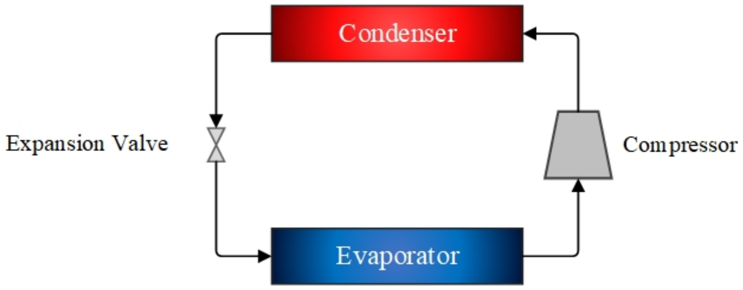

Fig. 2 Scheme of a single-stage cycle

The compressor has the function of compressing the refrigerant, absorbing the energy supplied externally. After the compressor, the refrigerant passes through the condenser, an exchanger that generally expels the heat to the environment, space or fluid. In contrast, the refrigerant is cooled down by changing the phase from superheated gas to subcooled liquid to pass through the throttling device mechanism, which expands to generate a pressure drop, be it an expansion valve, capillary tube or another. Finally, the refrigerant passes through the evaporator, which is another exchanger. Still, this process simultaneously transfers heat from a space or fluid to the refrigerant, turning it into a vapor state (with a certain degree of overheating) and returning to the compressor.

2.2 Indirect expansion

This configuration requires an additional heat transfer fluid (at low temperatures, mixtures based on glycol or CO_2), whose only function is carrying the cooling effect to an evaporator located in another position. When using a flammable, highly toxic or high global warming potential (GWP) refrigerant, the refrigerant charge is reduced and contained in the primary circuit. In its most basic configuration, apart from the evaporator of the primary vapor compression circuit, it has an evaporator that exchanges heat between the two circuits, a pump for the secondary fluid and the external heat exchanger, as shown in Fig. 3.

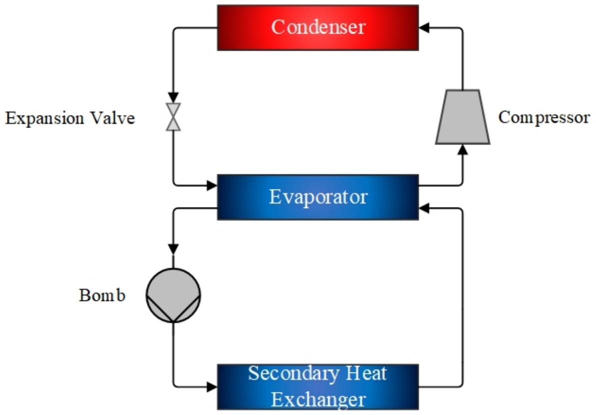

Fig. 3 Scheme of an indirect expansion cycle

2.3 Cascade

The cascade system consists of different vapor compression circuits linked by an additional exchanger (cascade heat exchanger in Fig. 3). Different refrigerants may be used in each circuit without mass exchange according to the most suitable temperature range, as shown in Fig. 4.

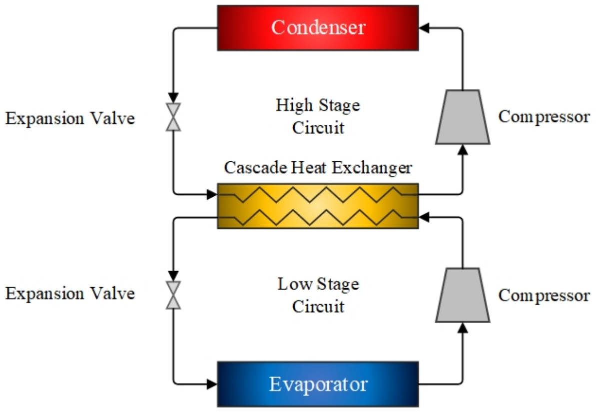

Fig. 4 Scheme of a cascade cycle

According to the optimal temperature range for performance (in operational and energetic terms), one of the most significant benefits of this system is using different refrigerants in each circuit. The higher stage circuit is subjected to higher pressures and temperatures. In contrast, the refrigerant in the low stage circuit covers lower temperatures at acceptable pressures (they are known as the high and low-temperature circuits, respectively). The compression pressure ratio and discharge temperatures are significantly reduced for high-temperature lifts, but their construction is more complex than single-stage cycles. Therefore, it offers several possibilities for optimization and increasing energy efficiency.

Usually, the interest is in the low stage circuit's refrigeration, for which an evaporator is used in the cooling process. Contrary to indirect refrigeration systems, both circuits are vapor compression cycles, so the fluid goes through the four main processes typically observed.

2.4 Intermediate heat exchanger (IHX)

As shown in Fig. 5, this configuration introduces an additional heat exchanger (different from those mentioned in the indirect expansion and cascade cycles, Section 2.2 and 2.3, respectively), achieving two fundamental effects. On the one hand, it superheats the refrigerant before the compressor inlet, ensuring it is in vapor phase. On the other hand, it subcools the refrigerant in the liquid line, increasing the refrigeration effect (evaporator enthalpy difference) and ensuring it is in the liquid phase. However, it increases the compressor suction temperature, decreases the mass flow rate, and increases discharge temperature.

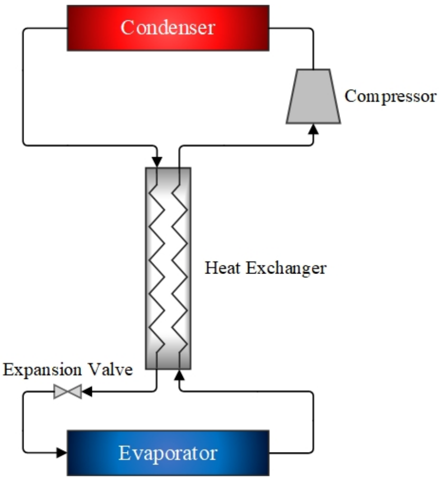

Fig. 5 Scheme of a cycle with intermediate heat exchanger

The use of the intermediate heat exchanger (IHX, also known as liquid-to-suction heat exchanger) does not guarantee an increase in COP (coefficient of performance). It is only observed when the increase in the refrigeration effect is more significant than resulting in specific compression work. The final effect depends on the refrigerant and the operating conditions, and it should be studied on a case-by-case basis. It is commonly seen in other applications due to its simple modification. In small capacity systems, it can be observed in the form of a capillary tube-suction line heat exchanger. The capillary tube is rolled with the suction line, so pressure drop and subcooling in one side and superheating are co-occurring.

2.5 Ejector

The ejectors have the role of occupying the function of the expander component of the circuit, as shown in Fig. 6.

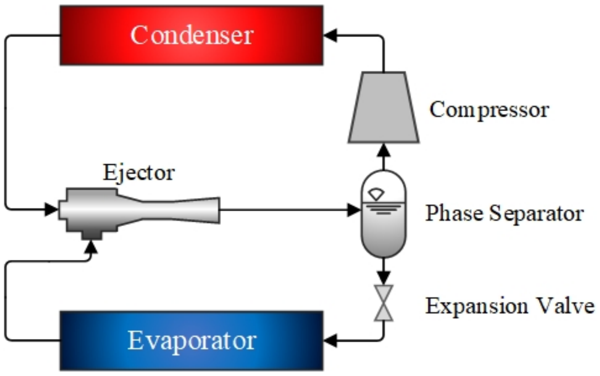

Fig. 6 Scheme of a cycle with ejector

The ejector consists of different parts described in Fig. 7. The first part is the nozzle, which is responsible for converting the potential energy of the primary fluid (the one that comes from the condenser) into kinetic energy, a divergent-convergent transformation zone.

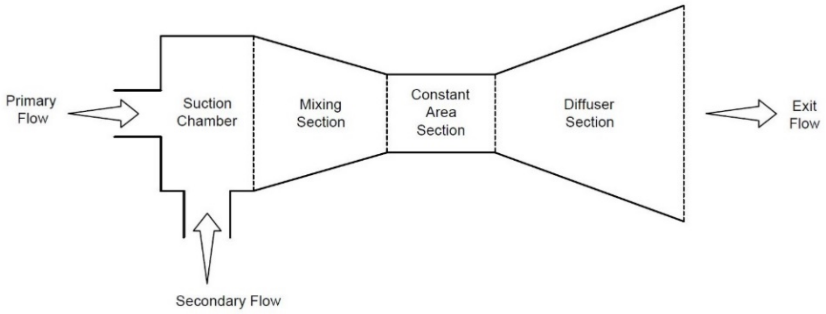

Fig. 7 Schematic representation of an ejector and its main parts

In turn, the rest of the ejector can be divided into three zones: a converging section that works as a mixing chamber, a constant area section and finally, a diffuser as a diverging section. In the convergent part, the primary and secondary fluids are mixed and directed to the second part, taking advantage of the kinetic energy. The fluid passes through the second part, a zone of a constant area where the pressure increases and the flow becomes subsonic to pass to the diffuser, an inverted cone.

In this zone, the high pressure and temperature flow stream (that of the primary fluid) is transferred to the low pressure and temperature stream (secondary), suctioning the secondary flow so that it mixes in the step described above. The fluid experiences a significant pressure loss in this last stage until it reaches the target outlet pressure.

2.6 Multi-stage

It uses several compressors, dividing the compression process into several stages, allowing a higher temperature lift. The refrigerant discharged by the low stage compressor is desuperheated by mixing it with the refrigerant from another part of the circuit in the liquid or vapor phase using an additional heat exchanger or a phase separator. Subsequently, it goes through the high stage compressor, thus decreasing the partial pressure ratio and final discharge temperature more than if it just comprised a compressor. The total electricity consumption is usually reduced for higher temperature lifts. Fig. 8 shows an example of a vapor injection two stage cycle, in which a part of the refrigerant is injected at an intermediate pressure in vapor phase. The main drawback is that the total mass flow rate compressed by the high stage compressor is higher than single-stage cycles.

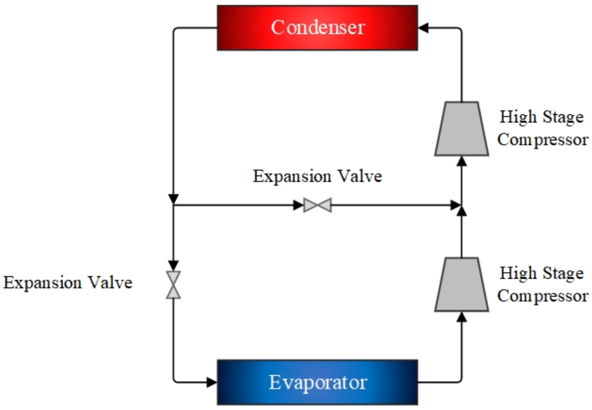

Fig. 8 Scheme of a multi-stage cycle

2.7 Absorption

In this configuration, like in the ejector, one of the main components of the cycle is replaced, in this case, the compressor. For this configuration, the compressor is substituted by a subcircuit called the absorption system.

The absorption system can work based on two processes, as explained by Srikhirin et al. [1]. In the first process, two vessels are connected, having a binary solution of a working fluid formed by one part of the refrigerant and the other absorbent, each in a container. The absorbent in the container takes the refrigerant vapor from the other container provoking a pressure drop. While the refrigerant vapor is being absorbed, the temperature of the remaining refrigerant decreases due to its vaporization. Consequently, it cools the refrigerant container. At the same time, the solution within the container that previously contained only the absorbent becomes more diluted due to the higher content of absorbed refrigerant.

When the solution becomes saturated, the refrigerant is extracted from the diluted solution, heat is applied to the vessel, and the refrigerant vapor returns to the other container. It condenses when transferring heat to the surroundings.

Nevertheless, these two processes do not have to occur in the same vessels. Since the first process occurs at a higher pressure than the second, an expansion valve and a pump are necessary, Fig. 9.

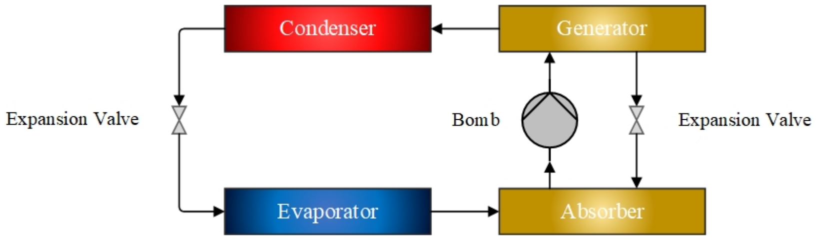

Fig. 9 Absorption cycle

As a result of this system, vessels on the right side have the role of a condenser and evaporator, yielding and absorbing energy, respectively. Thus, it has the same effects as in the simple cycle described above in this chapter.

This system has the disadvantage of offering very low COPs because of being an inefficient system, partly due to the need to use thermal energy in the compression system, which is inconvenient. However, it is not based on synthetic refrigerants as most vapor compression cycles neither require a compressor, which consumes electricity and produces noise and vibrations and increases the maintenance cost of the installation.

2.8 Auto-cascade

The auto-cascade configuration follows the premise of the cascade of separating the refrigerants for different temperatures-pressures required. A cycle is designed in which the refrigerants flow through separate lines due to their thermodynamic properties and later put them back together. Still, this system is not achieved by separating them in different cycles but by causing their separation in a single physical cycle. Fig. 10 shows a basic scheme of a simple auto-cascade cycle configuration.

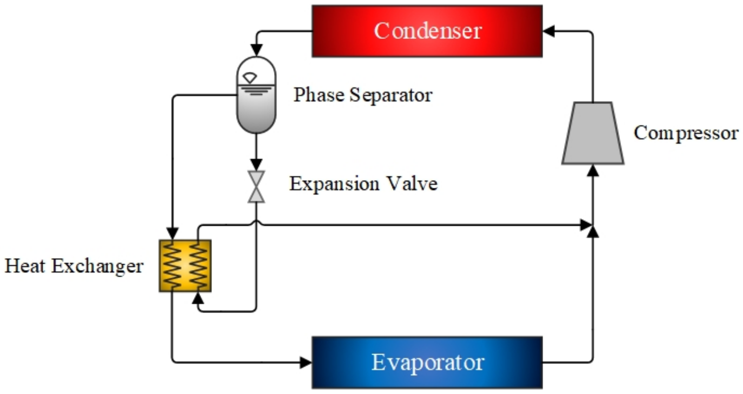

Fig. 10 Scheme of an auto-cascade cycle

The benefits of the cascade system are varied, allowing, for example, to have a more significant temperature difference without the need of separating the refrigerants in different cycles. The main drawbacks are the choice of refrigerants and the difficulty when designing the cycles so that refrigerants separate.

2.9 CO_2 sublimation

Recently a new type of cycle appears that allows CO_2 to work at temperatures below its triple point. This point occurs when the CO_2 reaches a temperature of –57.57 ºC and 5.1 bar of pressure, converging the three states simultaneously. Below this point, the refrigerant would theoretically go from the gaseous state directly to the solid-state. But as sows Di Nicola et al. [3], CO_2 would be able to reach up to approximately –78ºC while maintaining a metastable liquid phase.

This can be achieved through cycle modifications such as those already exposed above, for example, double compression. Additionally, the introduction of an intermediate exchanger or the use of cascade systems can also be considered, according to Mickoleit et al. [4]. Another example cycle was proposed by Sobieraj [5], allowing to reach –72 ºC. There is also the case of Xu et al. [6], who developed a specific component for this purpose using it as an evaporator.

3. Ultra-low temperature configurations recent findings

A review of the systems used in ultra-low temperature refrigeration and refrigerants can be read in the article by Mota-Babiloni et al. [2]. Cascade and auto-cascade systems are the most widely used in commercial low or ultra-low temperature freezers. This is because they present considerable advantages when tackling high-temperature lifts, as shown by Mumanachit et al. [3], who compared the cascade system with the double stage. They concluded that the cascade is more efficient below the optimal point of the COP and more cost-effective below –46.2 ºC.

After that, all the configurations reviewed come from modifications of the basic cascade refrigeration cycle.

3.1 Two-stage cascade

Relative to simple cascade cycles, Di Nicola et al. [4] compared different hydrofluorocarbons (HFCs) to the natural refrigerant R-717 at temperatures of –70 ºC, concluding that ammonia is around 5 % superior in terms of COP.

Lee et al. [5] investigated the optimum condensation temperature to maximize COP and decrease energetic losses. They observed that COP increases with increasing evaporation but decreases with increasing condensation temperature and temperature variation. Dopazo et al. [6] confirmed the influence of operating temperature variations in the COP. They quantified a 70 % increase in the COP when the evaporator temperature goes from –55 to –30 ºC. When the condenser temperature rises from 25 to 50 ºC, it causes a 45 % drop in the COP. In the same way, they found that an IHX temperature increase from 3 ºC to 6 ºC reduces COP by 9 %. A proper IHX optimization is essential to have an optimal operating temperature, according to the research of Sun et al. [8]. It also observed that R-41 is a good substitute for R-23.

Experimentation has also been carried out in cascades with not-in-kind refrigerants such as HFE-7000 and HFE-7100. Adebayo et al. [7] concluded that the pair that obtains the highest COP is R-717/R-744, while the opposite is observed with HFE-7100/R-744.

3.2 Cascade with intermediate heat exchanger

The same research mentioned above by Di Nicola et al. proposes using an IHX in the low stage (LS) circuit, concluding that it could benefit energy performance. It is not until years later, when Bhattacharyya et al. [8] studied a cascade system with IHXs, positioning one in the high stage circuit (HS) and the other in LT, trying to optimize it and first observed that the performance of the other system is independent of the exchangers' performance.

Liu et al. [9] carried out a theoretical and experimental study of a cascade system with an IHX in the LS cycle but could turn it into dual. They observed that the COP was lower if only the LS IHX worked, so the dual cycle has a higher potential for energy performance improvement. Also, Dubey et al. [10] proposed a cascade system with two IHXs, observing that the COP with HS IHX was greater than that of the LS. They proved that the higher the temperature difference in the cascade heat exchanger, the lower the COP. Also, the lower the temperature of the LS evaporator, the lower the COP.

3.3 Cascade with ejector

Dokandari et al. [11] proposed two ejectors in a two-stage cascade, placing one in each subcircuit. They reported a 7 % COP increase in comparison with a standard cascade system. Yunxiang et al. [12] compared a cascade circuit with an LS IHX and the same circuit but adding an LS ejector. They concluded that the ejector reduces energy consumption by 4.8 %, but they will extend this study in the future.

3.4 Cascade with three stages

Another way to cascade is to use even more than two stages. Johnson et al. [13] developed a three-stage cascade with dynamic control, showing effectiveness against flow disturbances in the secondary fluid. A comprehensive comparison of refrigerants is found in the article by Sun et al. [14] in which the following groups of refrigerants are recommended: R-1150/41/717, R-1150/41/152a, R-1150/41/161, R-1150/170/717, R-1150/170/152a and R-1150/170/161.

4 Conclusions

The slower development of ultra-low temperature refrigeration compared to other refrigeration and heat pump applications make evident the gap existing in this field yet to be discovered and analyzed. Existing challenges are related to difficulties in proper and long-life system operation and improvement in energy efficiency. The need of preserving vaccines at –80 °C is increasing the attention devoted to refrigeration at extreme or not-in-kind conditions, such as ultra-low temperature.

Several projects have concluded over the last few years and pointed out that refrigeration at these temperatures should be based on cascade cycles. However, the sector has not been developed and studied in-depth beyond other configurations, three-stage cascades, or more complex cycles, optimizing each circuit's working fluids.

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