Evaluation of a two-stage mixed refrigerant cascade for HTS cooling below 60 K

Thomas M. Kochenburgera,∗, Steffen Grohmanna,b, Lothar R. Oellricha

aInstitute for Technical Thermodynamics and Refrigeration, Karlsruhe Institute of Technology, Engler-Bunte-Ring 21, 76131 Karlsruhe, Germany
bInstitute for Technical Physics, Karlsruhe Institute of Technology, Hermann-von-Helmholtz-Platz 1, 76344 Eggenstein-Leopoldshafen, Germany

Abstract

A mixed refrigerant cascade presents a potential solution for cooling of high-temperature superconductors between 55 K and 70 K. The envisioned process consists of a pre-cooling and a low-temperature stage, where pre-cooling to 120 K is achieved by a conventional mixed refrigerant cycle. The low-temperature stage operates with a mixture of neon, nitrogen and oxygen at high pressure. Process simulation predicts an overall efficiency of about 8 % of Carnot at 55 K. Simulation results for the pre-cooling stage were validated by experiments with an existing test stand.

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1. Introduction

High-temperature superconductors (HTS) have no electrical resistance and can thus conduct large current densities with almost no losses. Their application in the power grid in cables, transformers and fault current limiters, as well as in superconducting motors and generators has been studied extensively in recent years. So far, cooling of HTS applications has mostly been realized by sub-atmospheric boiling of liquid nitrogen, where the low-temperature limit is given by the triple point of $T_{tr,N_2} = 63.15$ K. Lower operating temperatures allow higher current densities, but require different cooling methods. Besides temperature, the choice of cooling largely affects the design of HTS applications, their overall efficiency, reliability, practicability and cost. This motivates the study and development of closed-cycle solutions beyond liquid nitrogen, in order to promote the commercialization of HTS applications.

Commercial cryocoolers can be divided into regenerative cycles (Stirling, pulse-tube and Gifford-McMahon) and recuperative cycles (reverse-Brayton, Linde-Hampson and Claude). Stirling cryocoolers are available in a cooling power range up to 4 kW at 70 K. Because of frequent maintenance [1], these machines require redundant installations. Pulse-tube and Gifford-McMahon cryocoolers provide a few 100 W of cooling power at most and are thus unsuited...
for larger HTS applications. Reverse-Brayton and Claude coolers usually have cooling powers above 10 kW because of the difficulties in downscaling the cold turbo-machinery. Linde-Hampson coolers with pure refrigerants, which can operate in the whole range of cooling power, usually disqualify because of low efficiency.

As an alternative, this paper discusses the potential of Linde-Hampson mixed refrigerant cycles (MRCs) at cryogenic temperatures. Since cooling powers from several 100 W to a few kW at 55 K – 70 K are not well covered by present cooling technology, the envisioned process was designed for this range. The functional principles and applications of MRCs are discussed in the subsequent Section 2. Section 3 describes both implementation and results of a process simulation. In Section 4, an experimental validation for modeling the pre-cooling stage is presented. The final Section 5 provides conclusions and an outlook to future activities.

2. Mixed refrigerant cycles

2.1. State-of-the-art

Linde-Hampson coolers operating with pure refrigerantshave low efficiency, caused by the mismatch of heat capacity flows in the counter-flow heat exchanger (CFHX) and consequential losses in the Joule-Thomson (JT) valve. By using wide-boiling refrigerant mixtures (usually nitrogen plus several hydrocarbons), the heat capacity profiles can be adjusted through contributions of latent heat of the components. This shifts the CFHX operation into the two-phase region and yields considerable performance improvements [2]. Besides efficiency, main advantages are the simple setup with off-the-shelf refrigeration components and the scalability. MRCs present a proven and reliable technology for the temperature range of 300 K – 90 K, covering cooling capacities from small cryocoolers for electronic and medical devices up to baseload natural gas liquefaction plants.

Single-stage MRCs become increasingly inefficient below 90 K, because higher boiling components (hydrocarbons) must be avoided due to their risk of freezing. Technical options for separating the high-boilers from the coldest part include cascade (two separate cycles) or auto-cascade configurations (one cycle with phase separation). In a recent publication, Lee et al. [3] described such a two-stage MRC for HTS cooling below 70 K. Pre-cooling was achieved with liquid nitrogen, eventually to be substituted by a MRC cycle. The experiments revealed difficulties with freezing of nitrogen in the second stage, which was operated with a neon-nitrogen mixture. The concept discussed in the following section can solve this particular problem.

![Fig. 1. Binary nitrogen-oxygen phase diagram (adapted from Barrett et al. [4], experimental data by Ruhemann et al. [5]).](image-url)
2.2. MRC operation below the triple point of nitrogen

Lower operating temperatures enable the increase of current densities in superconductors. As liquid nitrogen freezes at 63.15 K, several authors advocate liquid air as alternative for achieving lower temperatures. In the literature, however, there seems to be confusion as to the exact freezing temperature of air. Fig. 1 shows the phase diagram of a binary oxygen-nitrogen mixture. It forms an eutectic system, in which the freezing temperature of the mixture is decreased significantly below that of the pure components. An air-like mixture consisting of 79 % nitrogen and 21 % oxygen (mole fractions) has its liquidus temperature (at which it begins to freeze) at approx. 61 K and its solidus temperature (at which it is completely frozen) at approx. 57 K. Increasing the oxygen content further decreases the liquidus temperature, reaching a minimum of 50.1 K at the eutectic composition with 77 % oxygen. On this basis, it is possible to adjust the oxygen content of the refrigerant to a certain operating temperature in order to avoid freezing.

2.3. Cascade layout

The layout of our two-stage mixed refrigerant cascade is shown in Fig. 2. The first stage is a classical MRC that pre-cools the high-pressure flow of the second stage to about 120 K. It utilizes a refrigerant mixture of nitrogen and hydrocarbons. The second stage operates with a high-pressure mixed refrigerant that contains the low boilers nitrogen, oxygen and neon. The inclusion of oxygen lowers the freezing temperature of the mixture and allows cooling temperatures down to 55 K. The modeling of mixture properties and the optimization of the composition are discussed in the following section.

3. Simulation of the two-stage mixed refrigerant cascade

3.1. Property model

For the evaluation of the concept, the entire process was simulated with the chemical engineering software Aspen Plus®. In the literature, the Peng-Robinson equation of state (PR-BM in Aspen Plus®) is commonly used for the calculation of the thermophysical properties in MRC simulations. However, the software database does not provide binary interaction parameters for neon-nitrogen and neon-oxygen systems. Therefore, experimental binary isothermal vapor-liquid-equilibrium data of neon-nitrogen by Streett [6, 7] and neon-oxygen by Streett and Jones [8] were used to fit the binary interaction parameter $k_{ij}$ in the van-der-Waals mixing rules of the PR-BM model. The results were compared to the Predictive Soave-Redlich-Kwong (PSRK) model, which uses UNIFAC activity coefficients in the mixing rules and does not require additional interaction parameters. As shown in the right chart of Fig. 3, it was found that both models performed satisfactorily in the neon-oxygen system. For neon-nitrogen, on the other hand, the Peng-Robinson model exhibited difficulties with the form of the boiling line, while the PSRK prediction was sufficiently accurate up to about 100 bar (left chart in Fig. 3). As a result, the PSRK model was applied in the simulations.
3.2. Operating conditions

With the aim of optimal performance conditions, the total compressor power was minimized using the Aspen Plus® SQP optimization module. The variables during optimization were the refrigerant compositions and the mass flows in either stage, as well as the temperature of the high pressure stream after the counter-flow heat exchanger CFHX2 (Fig. 2).

The process simulations revealed the best performance at compressor suction/discharge pressures of 5/20 bar in the pre-cooling stage, and 40/100 bar in the second stage. The high pressures in the second stage increase the efficiency considerably, and the small pressure ratio can be managed by a single-stage compressor. 120 K was chosen as the pre-cooling temperature at the cold end of the counter-flow heat exchanger CFHX1. The evaporator HX3 had a design capacity of 300 W cooling power at 55 K. The oxygen/nitrogen mass ratio in the second stage was constrained to 2:1 in order to keep the freezing point of the mixture at about 52 K (cf. Fig. 1). Minimum temperature differences of $\Delta T_{\text{min}} = 4$ K between the fluid streams were assumed in CFHX1, while $\Delta T_{\text{min}} = 2$ K was set for CFHX2. In addition, a pressure drop of $\Delta p = 2$ bar was assumed in each fluid stream. The temperatures at the exit of the aftercoolers were fixed to 300 K in a superheated state, the so-called gas refrigerant supply mode. This simplifies the refrigerant filling and the start-up of a mixed refrigerant cycle. The isentropic efficiency of the compressors was taken as $\eta_{\text{is}} = 0.6$.

3.3. Simulation results

The results of the optimization are summarized in Table 1. The main heat exchangers CFHX1 and CFHX2 have design specifications of $U A = 4700$ W/K and $U A = 2000$ W/K respectively, where $U$ is the overall heat transfer coefficient with regard to the contact area $A$. The total compressor power was $P_{\text{tot}} = 16.5$ kW, which results in a coefficient of performance of $\text{COP} = 1.8\%$. This corresponds to a relative efficiency of 8 % with regard to the Carnot cycle operating between 55 K and 300 K.

4. Experimental validation of the pre-cooling stage

4.1. Description of the setup

A single stage MRC test stand available at the Institute [9] was used to experimentally validate the simulation concept for the first stage. The process flow diagram of the test stand is shown in Fig. 4. The refrigerant mixture was compressed to 15 bar – 20 bar by a 800 W oil-lubricated rolling piston compressor, which was followed by an oil
Table 1. Results of the Aspen Plus® optimization.

|                      | First stage | Second stage |
|----------------------|-------------|--------------|
| Suction pressure (bar) | 5           | 40           |
| Discharge pressure (bar) | 20         | 100          |
| Compressor power (kW)  | 6           | 10.5         |
| Refrigerant composition (mass fraction) | | |
| 15 % nitrogen         |             | 82 % neon    |
| 15 % methane          |             | 12 % oxygen  |
| 5 % ethane            |             | 6 % nitrogen |
| 65 % propane          |             |              |
| 82 % neon             |             |              |
| 12 % oxygen           |             |              |
| 6 % nitrogen          |             |              |
| Total massflow (g/s)  | 31          | 49           |
| Temperature before JT valve (K) | 120       | 60           |
| Temperature after JT valve (K)  | 114.8    | 52.8         |

separation stage consisting of a liquid separator, two coalescers and an activated charcoal adsorber. In the coldbox, the refrigerant mixture was cooled and gradually condensed in a multi-tubes-in-tube heat exchanger that was equivalent to the CFHX1 in Fig. 2. Subsequently, the mixture was expanded to 4 bar in a pressure-regulated Joule-Thomson valve. The low-pressure stream was warmed up and re-evaporated against the high-pressure stream in the heat exchanger, and then returned to the compressor. A separate nitrogen gas stream, which simulated the heat load of the second stage, was regulated by a mass flow controller (MFC) and supplied into a copper capillary that was wound around the heat exchanger CFHX1. Temperatures and pressures were recorded at several locations of the cycle.

For the validation of the pre-cooling simulation, the Aspen Plus® model had to be adapted to the different pressure levels of the test stand. The existing heat exchanger was assumed to have a performance of $UA = 200 \text{ W/K}$, which proved to be a reasonable number in previous experiments. The optimized refrigerant composition in mass-% was 9.6% nitrogen, 7.5% methane, 11.6% ethane and 71.3% propane.

![Fig. 4. Simplified process flow diagram of the experimental test stand.](image)

4.2. Experimental results

After switching on the compressor, the temperatures decreased rapidly and approached steady state conditions with a no-load temperature of 97.5 K. When the lowest temperature was reached, the additional nitrogen stream was set to 0.25 g/s, which was the maximum capacity of the mass flow controller. As a consequence, the temperature behind the Joule-Thomson valve increased to 112.1 K, while the temperature of the nitrogen stream leaving the heat exchanger was 125.7 K.

In Fig. 5, the temperatures measured along the heat exchanger are compared to the predictions of the Aspen Plus® model. For this representation, it was assumed that the $UA$-value of the heat exchanger increased linearly with its length. The temperatures in the mixed refrigerant cycle agreed reasonably well with the model. The nitrogen temperatures, however, differed noticeably from the refrigerant temperatures and the predicted cold exit temperature
was not reached. The reason was found in the poor heat transfer to the nitrogen capillary, which was wound around the main heat exchanger tube and brazed to it just in a few places. Nonetheless, this solution fulfilled its purpose for providing a continuous heat load along the heat exchanger, and to verify the simulation of the pre-cooling stage.

5. Conclusion

The results of this work show that cooling below 60 K can safely be achieved with a two-stage mixed refrigerant cascade. The concept promises economic cryogenic cooling systems with reasonable efficiencies, based on standard refrigeration components. This implies reliability and a flexible adaptation of cooling power to the needs of HTS applications. The construction of an experimental test stand for the whole cascade is planned at the Institute.

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