Experimental research of low-temperature throttling refrigerator for cryoconservation of medical and biological objects

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Abstract. Results of experimental research of low-temperature multicomponent mixed-refrigerant throttling refrigerator are given. Appliance of refrigerator in new domestic-made ultralow-temperature freezer for cryoconservation and long-term storage of medical and biological objects at temperature minus 150°C is described. Within experimental research, test bench with development prototype of ultralow-temperature medical freezer is made. Experimental and theoretical studies are compared. Availability and amount of local composition shift is defined. Ways to improve simulation model and increase energy efficiency of cryostatting systems are determined.

1. Introduction and direction of research
Low-temperature multicomponent mixed-refrigerant throttling refrigerators (MRTR) designed for temperatures from -90°C to -160°C are central to many fields of science and technology. Low temperature refrigeration based on this type of cycle is high demanded and successfully applied for cooling radioelectronic and electrooptic sensors, high-temperature superconductors, gas purification for vacuum application, cryotreatment of construction materials in machinery and metal-working and for liquefaction of natural gas in energy industry. But particularly topical appliance of low-temperature is in medicine for cryoconservation of biomedical objects and for local cryotherapy.

At present, there is high demand for low-temperature equipment for long-term (over 10 years) and proper (amorphous state) storing of different biomedical objects such as hemic stem cells, marrow, packed red blood cells, cord blood, antibodies, bacteria and viruses in anabiosis at temperature minus 150°C. Advance of modern medicine in part of bio-insurance, donation, cancerous growth treatment, birth rate depends on development, researching and implementation in Russian medical centers of such low-temperature equipment.

Previously in articles [1-2] authors made cycle, which is patented [14], and developed simulation model of closed loop cycle of MRTR for medical ultralow-temperature chamber. In terms of choosing optimal cycle type, authors made calculations, optimization and comparing of refrigeration schemes used for cryostatting objects at temperatures from -90°C to -160°C [3]. Within carried out theoretical research of MRTR composition of multicomponent mixed refrigerant (MMR) was calculated, calculation and optimization approach of MMR was made. For adaptation of approach of calculation and optimization to modeling problem of MRTR in terms of MRTR simulation model, assumptions, constraints, variables and criteria of optimization, authors developed test installation for measuring dynamic parameters of MRTR and real composition of MMR in different points of the loop on various duties. Key feature of zeotropic MMR working in MRTR is composition shift of circulating refrigerant, which influences on dynamic parameters, in comparing with initial charge.

A lot of researching works are dedicated to that problem [4-11]. Experimental results show that the composition shift is caused by several reasons, including the differential hold-up in the two-phase flow, the differential solubility in the lubricant oil and the differential leakage of the mixed refrigerant. For example, in articles [4-6] scientists from Cryogenic Laboratory, Chinese Academy of Sciences M. Gong, W. Zhou, and J. Wu in 2002-2007 experimentally found that the mixture compositions vary at different operating periods of the cycle. The maximum change of the compositions is up to 6% for
different operating periods. The unevenness of the mixture compositions at different positions of the system may be up to 12%, or even more. Experiments with oil-free compressor were carried out providing different solubility in the lubricant oil. The results show that the dissolution in oil will greatly decrease the concentrations of those components with high-boiling point temperatures. The composition variation can reach about 74% for the highest-boiling component, and about 160% for the lowest-boiling component. Meanwhile, the solubility will increase when the temperature decreases or pressure increases. In addition, it is found that a variation in mixture composition was found to be caused by the liquid holdup in the low temperature two-phase flow. The composition of high-boiling components will decrease due to the liquid accumulation in the low temperature section: in recuperative heat exchangers, evaporators and so on. The initial charge refrigerant amount will influence the composition shift degree. The increase of the charged amount will decrease the degree of composition shift.

Scientists G. Venkatarathnam, H. Gurudath Nayak, Rajesh Reddy, Lakshmi Narasimhan N., Srinivasa Murthy, Bura Sreenivas from Indian Institute of Technology Madras, and W. Pang, J. Liu, X. Xu, J. Bao, L. Zhao from China institutes, in 2010-2016 in their articles [7-11] properly researched the relationship between the charged and circulating composition in a J-T refrigerator operating in the liquid refrigerant supply (LRS) and gas refrigerant supply (GRS) modes and an auto refrigerant cascade refrigerator (with a phase separator) operating in GRS mode (difference between GRS and LRS operating modes is mentioned in [12]). It was observed that the composition of the mixture charged when the compressor is switched off, and that in circulation during steady state are completely different in JT refrigerators operating with nitrogen-hydrocarbon mixtures. Apart from that, for a desired heat load, the number of refrigerant moles to be charged, and a given hardware, the charge composition of mixture components vary linearly with circulating composition [7]. However, refrigerant samples were extracted during steady state operation from the low pressure return line only. Consequently, it was not taken into account that the local compositions in other sampling points are also different, that illustrated in works M. Gong, W. Zhou, and J. Wu [5]. It is notable that relating to MRTR in systems for cryostatting objects it is important to find optimal composition of MMR, which circulates in evaporator in operating mode because during most of the time MRTR works in cryostatting regime at low temperature. In this case, circulating composition before evaporator needs to be defined, then calculate constants of relationship between the charge and circulating compositions for every component and make corrections to charged MMR, using step-by-step method described in [7]. Consequently, using experimental data of real circulating composition it’s possible to define optimal composition of MMR to provide cryostatting regime with any configuration such as the dead volumes in the system, amount of lubricating oil in the compressor, oil separator etc. Besides, difference of MRTR investigated in this article is cascade cycle with MMR on upper and lower stages. Upper stage is J-T refrigerator operating with zeotropic mixture – R404a. Lower stage – similar to J-T refrigerator operating with zeotropic mixture (R728/R50/R14/R170/R290) in GRS mode. Condensation of high-boiling components in high-pressure flow occurs in condenser-evaporator.

The main focus of this paper is to compare results achieved by theoretical and experimental research of MRTR of biomedical low-temperature chamber and to determine availability and amount of composition shift. The results obtained in this study are presented in this paper.

2. Theoretical model
Authors developed simulation model for MRTR used in biomedical ultralow temperature chamber for temperature level minus 150°C. Modeling and calculation of parameters for different compositions of MMR are made by oil and gas processing software «ASPEN HYSYS v.8.0». Problem of composition formation is solved by optimization of components’ fractions with chosen objective function including satisfying constraints. Method is implemented which based on interaction of programs «ASPEN HYSYS v.8.0» and numerical computer environment «Matlab v.R2016a». Method is realized in the certain way. Initial estimates of variables of MRTR model are assigned: mass fractions of the components of MMR, discharge and suction pressure of the compressor. «Matlab» realizes selection
of values of optimized parameters according to optimization algorithm, comparing of constraints and values of objective function with values of previous iteration and comparing with local extremes calculated before. Optimization method used in «Matlab» is nonlinear programming with search of global minimum. Generated variables send from «Matlab» to model in «ASPEN HYSYS v.8.0» by «ActXserver» and then used for calculation of objective and constraints. Then objective and constraints are sent back to «Matlab» and compared with previous iterations. Optimization algorithm considering received values and parameters changes initials of variables for the next iteration and then variable sent again. Optimization runs until global extreme will be found or maximum number of iterations will be made. Detailed description of model, optimization parameters and initials of variables of MRTR model and the results are mentioned in [1-3]. For model approbation, comparing of theoretical and experimental results of working parameters of MRTR for regime of minus 150°C is given in the article.

3. Experimental setup

MRTR test bench (figure 1) consists of thermally insulated body, low-temperature refrigerator, which provides cooling and cryostatting of insulated interior volume, control system. Lower stage cold section is situated in thermally insulated body. Type of body insulation – frosted polyurethane system Elastopor H2120/80/2 is used to reduce the radiation heat leak to the cold section. Detailed description of design of ultralow-temperature medical freezer is given in [1]. The main refrigeration system parts and control system are placed in machinery space. Low-temperature refrigerator includes two stages: upper stage – the single stage J-T refrigerator operating with azotropic mixture – R404a; lower stage - J-T refrigerator operating with zeotropic mixture (R728/R50/R14/R170/R290) in GRS mode. It consists of a hermetic single-acting reciprocating compressor (cylinder capacity 24.2 cm³) with polyester oil (charge amount 890 cm³) as lubricant, double-threaded fin-tube after cooler, thermal expansion valve and plate heat-exchanger as evaporator in high stage. Hot section of low stage is comprised of a hermetic single-acting reciprocating compressor (cylinder capacity 34.4 cm³) lubricated by polyester oil (charge amount 890 cm³), double-threaded fin-tube after cooler, dehydration filter and float type oil trap with a reservoir volume of 2 liters. Cold section of low stage consists of multi tubular heat exchanger of length 5.5 meters coiled in serpentinous shape, electronic expansion valve with step engine and a loop-type tubular evaporator. In multi tubular heat exchanger the high pressure refrigerant flows through eight capillary tubes with an internal diameter of 3.15 mm each, while the low pressure refrigerant flows on the shell side (internal diameter of 19 mm).

Resistant thermometers OWEN, calibrated on liquid nitrogen temperature level, measure the temperature of the working refrigerant in cold section. The pressure transducers of Emerson evaluate discharge and suction pressure of the compressors. In this test bench, the flow rate of the working refrigerant is indirect measured by the pressure-difference transducer Aplisens APR-2000 and then is calculated with Darcy–Weisbach equation. Refrigerant mass flow was determined on discharge pipeline of lower stage hermetic compressor. The friction factor evaluated by the use of various empirical relations for greater Reynolds number, which corresponds to a turbulent flow according to Blasius. Measured section is made in spiral form to increase pressure drop. Owing to the tube’s curvature, centrifugal forces are initiated during flow and give rise to a secondary current in the form of a double vortex, due to big curvature secondary flow is negligible. Thermophysical properties of refrigerant in measured section is calculated in «ASPEN HYSYS v.8.0» software for sample of the circulating stream in point 3 (figure 1). Eventually, mass flow is evaluated by method is given in [13].

Compressors power was measured separately using an electronic power meter - Mercury 230. Refrigerant samples were extracted during steady state operation from the several points (1-7 in figure1) and the composition of the mixture in circulation is determined using a gas chromatograph with thermal conductivity detector (TCD) and flame-ionization detector (FID) (Chromos GC-1000). The gas chromatograph (GC) is calibrated against standard mixtures and the uncertainty in the determination of composition was estimated to be ±0.2%. Method used in GC makes it possible to determine refrigerants on simultaneously sample injection in capillary and packed column. Whereby
R50, R728 and R14 are determined on TCD channel with packed column NaX and eight-port valve with backpurging function, and R170, R290 and repeated R50 on FID channel with capillary column St-PLOT/Si (50m x 0.32mm x 3.0μm) with six-port injection valve. For sample injection additional bench with expansion tanks and fine oil filters is installed. Carrier gases perform GC gas supply: helium for TCD column, nitrogen for FID column, hydrogen and air.

Taken together, the main instruments and their measuring range and accuracy presented in Table 1.

| Parameters                        | Instruments          | Measuring range | Accuracy       |
|-----------------------------------|----------------------|-----------------|----------------|
| Temperature (cold section)        | OWEN DTS-024-50P     | -196 – 250°C    | 0.3+0.005t%    |
| Temperature (hot section)         | Elemer TS-1388 (Pt-100) | -50 – 200°C    |                |
| High pressure                     | Emerson PT5-30M      | 0 – 30 bar      | ≤±1%           |
| Low Pressure                      | Emerson PT5-07M      | -0.5 – 7 bar    | ≤±1%           |
| Pressure difference               | Aplisens APR-2000    | 0 – 3000 Pa     | 0.075%         |
| Power                             | Mercury 230 ART02    | 0 – 10 A        | 1%             |
| Weighing balance                  | CAS SWII-05          | 0.02-5 kg       | e=d=1g         |
| Chromatography                    | Chromos GC-1000      | –               | 0.2%           |

**Figure 1.** MRTR test bench circuit diagram, T1-T15 and P1-P4 indicate measuring points for temperature and pressure respectively, the numbers from 1 to 7 indicate different sampling points.

Experimental procedure was carried out as follows. Upper and lower stages of the test bench circuit were initially flushed with dry nitrogen gas. The compressor oil was heated to a temperature of about 80°C to remove all dissolved gases. A vacuum pump was used to evacuate the desorbed mixture components before charging refrigerants. The required quantity of each of the components was
charged, starting from the highest boiling point component to the lowest boiling point component. At a steady state operation samples of the circulating stream were extracted and analyzed in a gas chromatograph to measure the concentration in circulation. All measuring parameters were recorded in SCADA system.

4. Results and discussion

Figure 2a, b show the test results of the MRTR while starting and steady-state operation supplied with the mixture R728/R50/R14/R170/R290 for working discharge pressure of 23 barg and suction pressure of 2.2 barg. Pressures are regulated by pressure and composition control system, which are patented [15,16]. Special attention should be focused on the temperature and pressure disturbance after the temperature in interior volume of minus 148 °C is reached. Fluctuations are related to sudden change of density of low-temperature boiling components in processes which happen in recuperative heat exchanger and evaporator while condensation and evaporation. Fluctuations could be smoothened by fine tune of opening of electronic expansion valve, but this would lead to time increase of starting operation and increase of consumed power. The measurements were done for the variable cooling capacity while starting operation (cool down chamber construction) and constant heat load at steady state operation – wall heat flux.

![Figure 2. Test results of the MRTR while starting and steady-state operation: temperature changing in cold section (T9-T12 - temperatures according to the circuit diagram, Tvol – temperature in the center of insulated interior volume), compressors’ powers (LSP – lower stage, USP – upper stage) and pressure difference on discharge pipeline.](image)

Based on figure 3, main reasons of composition shift in tested MRTR are solubility of high-temperature boiling components on lubricating oil, hold-up and two-phase hold-up of condensed components in trapped zones of heat exchangers (due to features of construction). Sample from point 3 shows real composition of MMR before evaporator. The relations between the charge and circulating compositions show significant increase of low-temperature boiling components and decrease of high-temperature boiling components. By the way, the results are correlating with experiments in [7-8]: the composition deviation of component with middle boiling point is not large while composition deviation of R728 is about 45% and R290 - 25%. For cryostatting systems optimization of
composition in this point define energy efficiency of whole refrigeration system. However, composition change in starting regime, because of dynamically changing of heat load and design of refrigeration system, should be considered in optimization on certain temperature level.

![Figure 3. Variation of local composition distribution during circulation in steady-state operation.](image)

There were made calculations as part of MRTR simulation model approbation for the actual circulating composition of the MMR obtained from experimental data. After that there was made a comparison of the calculated and experimentally obtained MRTR parameters, including multi tubular heat exchanger LMTD, power consumption, evaporator heat load, COP and etc. It was concluded that the parameters obtained as a result of theoretical studies cannot be fully applicable to real MRTR, because the simulation model doesn't consider a number of real MRTR operation and design features. So, the difference in the theoretically and experimentally definition of evaporator heat load was about 32%, and recuperative HE LMTD - 11%. It follows that the optimal parameters part search for the MRTR operation should be carried out from an array of local optimization extrema with additional constraints in the simulation model of MRTR operation in the cryostatting mode (steady-state operation) and also in the cooling mode.

5. Conclusions
Composition shift is an important parameter for MRTR on zeotropic mixture. The occurrence of composition shift depends on the design and operation of thermodynamic system.

In this paper MRTR test bench as part of medical ultralow-temperature chamber is established to investigate availability, amount of composition shift and local variation of the compositions.

A comparison was made between the results obtained theoretically and experimentally. The results obtained in this research will be helpful to modify the simulation model of MRTR and adjust the preparation of MMR composition to reach the maximum energy efficiency.

This research presents a preliminary attempt to measure the mixture composition at steady-state operation. A lot of efforts in the future need to be made in the investigation of the dynamic performance of the real MRTR for cryostatting including determination of changing the circulating MMR composition while starting operation and correction the composition to be charged to get the desired circulating composition in the evaporator.

This work will be helpful in understanding the work mechanism of this type MRTR and will be useful in the design of similar systems.

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