Boiling of multicomponent working fluids used in refrigeration and cryogenic systems

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Abstract. Working fluids based on mixtures are widely used in cryogenic and refrigeration engineering. One of the main elements of low-temperature units is a recuperative heat exchanger where the return flow cools the direct (cold regeneration is carrying out) resulting in continuous boiling and condensation of the multicomponent working fluid in the channels. The temperature difference between the inlet and outlet of the heat exchanger can be more than 100K, which leads to a strong change in thermophysical properties along its length. In addition, the fraction of the liquid and vapor phases in the flow varies very much, which affects the observed flow regimes in the heat exchanger channels. At the moment there are not so many experimental data and analytical correlations that would allow to estimate the heat transfer coefficient during the flow of a two-phase mixture flow at low temperatures. The work is devoted to the study of the boiling process of multicomponent working fluids used in refrigeration and cryogenic engineering. The description of the method of determination of heat transfer coefficient during boiling of mixtures in horizontal heated channel is given as well as the design of the experimental stand allowing to make such measurements. This stand is designed on the basis of a refrigeration unit operating on the Joule-Thomson throttle cycle and makes it possible to measure the heat transfer coefficient with a good accuracy. Also, the calculated values of the heat transfer coefficient, obtained with the use of various correlations, are compared with the existing experimental data. Knowing of the heat transfer coefficient will be very useful in the design of heat exchangers for low-temperature units operating on a mixture refrigerant.

1. Introduction
In the cryogenic and refrigeration engineering systems based on throttle cycle are widely used, due to reliability, simplicity of the device and operation and, as a result, low cost. One of the promising ways to increase the efficiency of throttle cycles is the use of multicomponent working fluids (MWF), which are made by mixing traditional or new synthesized single-component substances. Many studies have shown that when operating low-temperature systems on such mixtures, in many cases, energy and mass-dimensional characteristics are improved, the resource and reliability of operation increases, and the starting period is shortened [1].

One of the main elements of the throttle cycle (in particular the Linde cycle) are heat exchangers. In one of them (recuperative heat exchanger), the reverse flow cools the direct flow (cold regeneration is carried out), resulting in continuous boiling and condensation of MWF in the channels. Another heat exchanger is an evaporator, to which the load from the cooled object is applied. Thus, a phase
transition is continuously carried out in the flows of heat exchangers, the working body is in a two-
phase state. The temperature difference (the change in the flow temperature between the inlet and
outlet of the heat exchanger) can be more than 100 K, which in turn affects the strong change of
thermophysical properties of the working fluid. The share of the liquid and vapor phases in the flow
also varies very much, which affects the observed flow regimes in the heat exchanger channels. As a
consequence, the use of MWF imposes certain requirements on the allowable flow rates in the
channels. If the flow rate is insufficient, the mixture can split up: the vapor phase will be at the top and
the liquid phase at the bottom, which will lead to a disruption of the phase equilibrium and, as a
consequence, to a worsening of the heat exchanger operation.

Currently in open literature there is no information about the universal methods of calculation of
processes of boiling and condensation when working on mixed refrigerants, even for relatively simple
and most frequently used in the throttle system heat exchangers type "tube in tube".

This paper describes the measurement techniques and construction of experimental stand, which
makes it possible to measure the heat transfer coefficient (HTC) of the two-phase MWF flow boiling
at low temperatures, and to compare the calculated HTC values obtained using different relationships
with experimental data.

2. Test facility
The experimental setup is assembled on the basis of a refrigeration machine operating on the Joule-
Thomson throttle cycle, the schematic diagram of which is shown in figure 1. The main parts of the

![Figure 1. Schematic of test facility: 1 - compressor; 2 - oil separator; 3 - condenser; 4 - recuperative
heat exchanger; 5 - throttle; 6 - heater; 7 - experimental section; 8 - evaporator; 9 - flow measurement
section; AIM - analog input module; IC - interface converter; PC - computer; T - temperature sensor;
P - pressure sensor; ΔP - differential pressure sensor.](image-url)
equipped with an automatic data collection system, which allows to save experimental data on the computer. Depending on the composition of the mixture and the required temperature level, the time for the installation to enter the regime is 60 to 90 minutes.

2.1. Test procedure
The method of determining the HTC should be simple from the point of view of technical implementation and at the same time be accurate enough. To the experimental section of known geometry a given amount of heat is supplied; by measuring the temperature distribution across the wall and the temperature of the flow of the working fluid, the HTC value can be determined from the heat transfer equation, as shown in figure 2.

![Diagram of heat transfer](image)

**Figure 2.** Method for determining the heat transfer coefficient: $r$ - radius; $T_{in}$ - inlet flow temperature; $T_{out}$ - outlet flow temperature; $T_{liq}$ - average value of the flow temperature; $T_1$, $T_2$, $T_3$ - measured temperature values used to obtain the extrapolation value of the wall temperature; $\Delta T_{wall}$ - wall temperature difference; $\Delta T_{wall-liq}$ - temperature difference between the wall surface and the flow.

The analytical expression for determining the HTC ($\alpha_{exp}$) has the form (1):

$$\alpha_{exp} = \frac{Q}{F \Delta T_{LMTD}} = f(Q, \Delta T_{LMTD}, F)$$  \hspace{1cm} (1)

$$F = \pi d_{in}L_{heat}$$

$$\Delta T_{LMTD} = \frac{\Delta T_{wall-in} - \Delta T_{wall-out}}{\ln(\frac{\Delta T_{wall-in}}{\Delta T_{wall-out}})}$$

$$\Delta T_{wall-in} = T_{wall} - T_{in}$$

$$\Delta T_{wall-out} = T_{wall} - T_{out}$$

$$T_{wall} = \frac{\ln \frac{T_1}{r_1}}{\ln \frac{T_3}{r_3}}$$

where $F$ is the area of the inner surface of the heated block to which heat is supplied; $d_{in}$ and $L_{heat}$ are the internal diameter and the length of the heated block; $\Delta T_{LMTD}$ is the log mean temperature difference; $\Delta T_{wall-in}$ and $\Delta T_{wall-out}$ are the inlet-to-wall and outlet-to-wall temperature differences; $T_{wall}$ is
the temperature of the inner wall surface of the heated block; $T_1$ and $T_3$ are the wall temperature of the heated block, measured by thermocouples at points 1 and 3; $r_1$ and $r_3$ are the coordinates of points 1 and 3; $Q$ is the thermal power released when an electric current passes through a conductor; $U$ and $I$ are the values of voltage and current.

The amount of heat input must be such as to cause a sufficient temperature gradient in the radial direction to fix the sensors and at the same time not lead to a strong change in the temperature of the flow of the working fluid between the inlet and outlet of the experimental section, which affects the strong change in its thermophysical properties, flow regime, mechanism of heat transfer. As a result, it was decided to make an experimental section of stainless steel since it provides the greatest temperature gradient in the radial direction from all the most widespread structural materials due to the fact that it has less thermal conductivity.

The heated block is schematically shown in figure 3 in the form of a thick-walled cylinder 50 mm high and 30 mm in diameter. For convenience of winding manganin wire on the outer surface of the unit there is a thread in the grooves of which the wire is laid. For passing a flow of the working fluid is provided a through hole with a diameter of 6 mm. In the thick-walled part of the cylinder with an indentation of 10 mm from each edge three 1 mm diameter holes are drilled radially in each direction to install thermocouples.

Figure 3. Heated block.

2.2. Test section
The experimental section consists of two identical, connected together heated blocks as shown in figures 4 - 6. They are connected using couplings having the same inner diameter and made of stainless steel. To measure the pressure and control the flow temperature at the inlet and outlet of the experimental section connectors are provided. Each connector has a hole on the top, into which the fitting for mounting the pressure sensor is soldered. Coaxially, at a slight distance, there is a cylindrical groove for the installation of platinum resistance thermometer (PRTs), which serve to measure the temperature of the working fluid flow. From one side the connector is connected to the heated unit and from the other side to the pipeline section of the working fluid flow. This section is installed after the throttle of the chiller directly right before the evaporator.
Figure 4. Solid model of the test section: 1 - fitting; 2 - connector; 3 – heated block; 4 - coupling; 5 - tube; Place of installation: a - PRTs; b - thermocouples; c - pressure sensors.

Figure 5. A longitudinal section of the experimental section.

Figure 6. Photo of the experimental section.
2.3. Flow measuring section

Knowledge of the flow is necessary to determine the flow rate, which is used in various correlations. The flow measurement section is a local hydraulic resistance: a straight, smooth, long tube \((L=310\, \text{mm})\) with an internal diameter of 4.75 mm to the inlet and outlet of which a differential pressure sensor is connected as shown in figures 7 - 9.

![Figure 7. Solid model of the flow measuring section: 1 - fitting; 2 - adapter; 3 – tube; 4 - connector.](image)

![Figure 8. A longitudinal section of the flow measuring section.](image)

The flowmeter is installed on the low pressure line, in front of the compressor. Thus, through this section a gas flow will pass and it will be possible to use the Darcy-Weisbach equation (2):

\[
\Delta P = \xi \frac{\rho u^2 L}{2d}
\]

where \(\Delta P\) is the pressure drop, \(\text{Pa}\); \(\rho\) is the density of the working fluid flow, \(\text{kg/m}^3\); \(u\) is the flow velocity in the section, \(\text{m/s}\); \(L, d\) are the length and the internal diameter of the section, \(\text{mm}\).

For gas flows of used working fluids with regard to their thermophysical properties and geometry of the section most likely a turbulent flow regime will be. In this case the friction coefficient \(\xi\) is determined as follows (3):

\[
\xi = \frac{0.3164}{\sqrt{Re}}
\]

The mass flow rate is determined from the expression (4):

\[
G = \rho u S
\]
where $S$ is the cross-sectional area of the section.

Figure 9. Photo of the flow measuring section.

Taking into account the known geometry of the section it is possible to obtain the final expression (5) for determining the mass flow through the pressure drop in the section and the thermophysical properties of the flow:

$$G = \frac{\pi^2 \Delta P \rho d^5}{8 \xi L}$$

(5)

This method of flow measurement can be considered quite accurate since it depends only on the pressure drop and the thermophysical properties. The error in determining the thermophysical properties can be ignored since the temperature and pressure are measured with sufficient accuracy. The error in measuring the differential pressure is also small and amounts to 1% of the measured value. Thus, the error in determining the flow rate by this method is within 1% of the measured value.

2.4. Test facility verification

To ensure reliability of the data obtained the test facility was verified. First, the heat balance of the experimental section was checked. For this purpose the installation was filled with gaseous nitrogen. The amount of heat applied was monitored as a product of the current to voltage. At the same time temperature sensors are installed at the entrance and exit of the experimental section. Thus, it is possible to determine the heat load as the product of the flow rate on the heat capacity and the temperature differential of the flow in the experimental section. Comparison of thermal balances obtained by different methods showed a maximum discrepancy of 3%. In parallel the HTC was measured at the section. The experimental values were compared with the calculated values obtained from the Dittus-Belter correlation for the forced single-phase flow in a pipe. The maximum deviation is 5.6%. After that the experimental stand was filled with R-22 and a comparison of the experimental data with the calculated ones was made. The maximum deviation is 17%. Convinced of the validity of experimental data it was possible to go to the main experiments.
3. Correlations for heat transfer coefficient of boiling mixtures

There are few papers devoted to the study of the processes of boiling and condensation of MWF in the low-temperature region (<240 K). Two studies are known in which mixtures consisting of three or more components have been studied [2], as well as a number of studies performed for binary mixtures [3]. Most of the techniques developed for mixtures based on ratios used for pure substances including amendments taking into account the multicomponent nature of working fluid. However, the data obtained with these correlations are of a particular nature and are applicable to a particular mixture and flow parameters.

In this paper the correlations presented in table 1 were used to obtain the calculated HTC values.

| Model          | Equation                                                                 | Comments                                      |
|----------------|--------------------------------------------------------------------------|-----------------------------------------------|
| Mishra [4]     | \( \alpha = C \alpha_l \left( \frac{1}{X_{tt}} \right)^m B o^n \)        | Horizontal test section.                      |
|                | \( B o = \frac{q}{Gr} \)                                                | Mixture 1: R-12 (23 – 27%) and R-22 (77 – 73%), C=5.62, m=0.23, n=0.5. Mixture 2: R-12 (41 – 48%) and R-22 (59 – 52%), C=21.75, m=0.29, n=0.23. |
| Granryd [5]    | \( \frac{1}{\alpha} = \frac{1}{\alpha_l F(X_{tt})} + \frac{xc_{pv}}{\alpha_v \frac{\partial h}{\partial T} P} \) \( \frac{x}{C_{lv}} \)   |                          |
|                | \( C_{lv} = 2 \)                                                       |                                |
|                | \( F(X_{tt}) = 2.37 (0.29 + \frac{1}{X_{tt}}) \)                         |                                |
|                | \( X_{tt} = \left( \frac{1-x}{x} \right)^{0.9} \frac{\rho_v (\mu_v)}{\rho_l (\mu_l)} \) \( x \) |                                |
|                | \( \alpha = 0.023 \frac{\lambda}{d_{in}} Re^{0.8} Pr^{0.4} \)          |                                |
| Little [6]     | \( \frac{1}{\alpha} = \frac{1}{\alpha_{wall-film}} + \frac{xc_{pv}}{\alpha_{film-v} \frac{\partial h}{\partial T} P} \) \( \frac{(1-x)c_l + xc_{pv}}{d_{in} (1-\varphi_l)} \) |                                |
|                | \( \alpha_{wall-film} = 0.023 \left( \frac{Re_{l_1}}{1 + \sqrt{\varphi_l}} \right)^{0.8} Pr_{l_1}^{0.4} \) \( \lambda_l \) \( d_{in} (1-\varphi_l) \) |                                |
|                | \( \alpha_{film-v} = 0.023 \left( \frac{Re_v}{\sqrt{\varphi_l}} \right)^{0.8} Pr_{v}^{0.4} \) \( \lambda_v \) \( d_{in} \sqrt{\varphi_l} \) |                                |
|                | \( \varphi_l = (1 + \frac{(1-x)\rho_v}{\rho_l})^{\frac{1}{2}} \)          |                                |
| Homogeneous    | \( \alpha_{hom} = 0.023 \frac{\lambda_{mix}}{d_{in}} Re_{mix}^{0.8} Pr_{mix}^{0.8} \) | Assumes the presence of phase equilibrium and the absence of phase slip |

Calculating the properties of mixtures in a two-phase region is a complex task. Some properties (density, enthalpy) obey the additivity property. However, the determination of viscosity and thermal conductivity is more complex. There are many relationships to determine these properties, but there is no consensus on recommendations for specific cases. Below are given the ratios used by the authors of this article to analyze the experimental data. The thermal conductivity of the mixture in the two-phase region \( \lambda_{mix} \) was calculated from the formula (6):

\[
\lambda_{mix} = (1-x)\lambda_l + x\lambda_v
\]
where \( \lambda_l \) and \( \lambda_v \) are the thermal conductivity of the liquid and vapor phases.

To calculate the dynamic viscosity of a mixture in a two-phase region \( \mu_{\text{mix}} \) the ratio (7) can be recommended:

\[
\frac{1}{\mu_{\text{mix}}} = \frac{x}{\mu_v} + \frac{(1-x)}{\mu_l}
\]

where \( \mu_l \) and \( \mu_v \) are the dynamic viscosity of the liquid and vapor phases.

The vapor content \( x \) was determined from equation (8):

\[
x = \frac{h_{\text{mix}} - h'}{h'' - h'}
\]

where \( h_{\text{mix}} \) is the enthalpy of flow at given temperature and pressure; \( h' \) and \( h'' \) are the enthalpy of the flow on the left and right boundary curves.

The properties of the mixture are determined using the REFPROP package.

4. Results

Experiments were carried out to determine HTC for freon and hydrocarbon mixtures. The parameters of the experiments are given in table 2.

Table 2. Parameters of the experiment Mogorychny/Dolzhikov.

| Run | Mixture composition (% mol) | Mass flow (kg/m²s) | Heat flow (kW/m²) | Pressure (kPa) |
|-----|-----------------------------|--------------------|-------------------|---------------|
| 1   | CH₄/C₂H₆/C₃H₈/(CH₃)₃CH (40/20/20/20) | 127 - 144          | 10                | 320 - 370     |
| 2   | R-14/R-23/R-22/R-236fa (40/20/20/20) | 297 - 343          | 10                | 340 - 395     |

Figure 10. The heat transfer coefficient during boiling of hydrocarbon mixtures.
Also known experimental data of Barraza who in his work [7] gives a description of the test facility and methods used to measure HTC during boiling nitrogen-hydrocarbon and argon-freon mixtures in a horizontal channel. As an experimental section he used a cylinder 105 mm high with a through hole of 2.871 mm in diameter for flowing the working fluid. The composition of the mixtures and the flow parameters are given in table 3.

Table 3. Parameters of the experiment Barraza.

| Run | Mixture composition (% mol) | Mass flow (kg/m²s) | Heat flow (kW/m²) | Pressure (kPa) |
|-----|-----------------------------|--------------------|------------------|---------------|
| 3   | CH₄/C₂H₆/C₃H₈ (45/35/20)    | 147                | 40               | 790           |
| 4   | R-14/R-23/R-32/R-134a (35/15/15/35) | 148              | 28               | 790           |

Figures 10 and 11 show the HTC values as a function of the average flow temperature obtained in the above experiments. A good qualitative and quantitative agreement of the experimental data is observed.

![Figure 11. The heat transfer coefficient during boiling of freon mixtures.](image)

5. Results analysis
Figures 12 - 15 show a comparison of the HTC calculated values obtained using the correlations presented in table 1 with the experimental data of Mogorychyn/Dolzhikov and Barraza.

The approximations average deviation (AAD) for the obtained values is estimated to understand their applicability.

\[
AAD = \frac{1}{N} \sum \frac{|\alpha_{exp} - \alpha_{calc}|}{\alpha_{exp}}
\]
Figure 12. Comparison of the experimental and calculated HTC values for Run 1: 1 - experiment Mogorychny/Dolzhikov; 2 - Mishra; 3 - Granryd; 4 - Little; 5 - Homogeneous model.

Figure 13. Comparison of the experimental and calculated HTC values for Run 2: 1 - experiment Mogorychny/Dolzhikov; 2 - Mishra; 3 - Granryd; 4 - Little; 5 - Homogeneous model.
Figure 14. Comparison of the experimental and calculated HTC values for Run 3: 1 - experiment Barraza; 2 - Mishra; 3 - Granryd; 4 - Little; 5 - Homogeneous model.

Figure 15. Comparison of the experimental and calculated HTC values for Run 4: 1 - experiment Barraza; 2 - Mishra; 3 - Granryd; 4 - Little; 5 - Homogeneous model.
A comparison of the deviations for all the experiments is given in table 4.

Table 4. Approximations average deviation (AAD), %.

| Run | Model     | Mishra | Granryd | Little | Homogeneous model |
|-----|-----------|--------|---------|--------|-------------------|
| 1   | 47.4      | 43.5   | 52.6    | 46.8   |                   |
| 2   | 14.5      | 9.5    | 14.4    | 11.8   |                   |
| 3   | 19.6      | 31.6   | 34.6    | 52.5   |                   |
| 4   | 25.9      | 29.8   | 33.2    | 48.3   |                   |

It can be seen from the comparison that none of the used correlations gives a good agreement with the experimental data especially in the region of high vapor contents ($x > 0.75$). This again confirms the fact that it is necessary to expand the base of experimental data on the boiling of multicomponent working fluids. The knowledge of HTC will be very useful in the design of heat exchangers for low temperature units operating on a mixed refrigerant.

6. Summary

The presented experimental stand allows measuring HTC with an error of less than 20%. Correlations were evaluated from the point of view of the possibility of predicting HTC during the boiling of zeotropic multicomponent hydrocarbon and freon mixtures. The calculated HTC values are compared with available experimental data. Since experimental data are available only for a small number of experiments it would be incorrect to make a final conclusion about the applicability of specific correlations. This analysis is a step towards a detailed understanding of the applicability of the correlations for multicomponent mixtures. More complete experimental data obtained for the same composition under different conditions are needed to understand the effect of each parameter on HTC.

The designed experimental stand allows to obtain with good accuracy the experimental HTC values during boiling of mixtures. This will make it possible in the near future to increase the amount of experimental data on the boiling of MWF and make a step towards a more accurate understanding of the course of this process.

References

[1] Lunin A I, Mogorychny V I and Kovalenko V N 2009 The use of multi-component working fluids in low-temperature technology (Moscow: Moscow Power Engineering Institute Press)

[2] Nellis G, Hughes C and Pfotenhauer J 2005 Cryogenics 45 546

[3] Greco A and Vanoli G 2005 Exp. Therm. And Fluid Science 29 189

Hsieh Y Y and Lin T F 2002 Int. J. of Heat and Mass Transfer 45 1033

[4] Mishra M P, Varma H K and Sharma C P 1981 Lett. Heat Mass Transfl. 8 127

[5] Granryd E 1991 Proc. 18th Int. Congr. Refrig. 1330

[6] Little W A 2008 AIP Conf. Proc. 606

[7] Barraza R and Nellis G 2016 Int. J. of Heat and Mass Transfer 97 683