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РАСТВОРЫ ХЛАДАГЕНТ/КОМПРЕССОРНОЕ МАСЛО: ПРОБЛЕМЫ ПРИМЕНЕНИЯ И ИССЛЕДОВАНИЯ СВОЙСТВ

В работе проведен анализ и обобщение результатов исследования теплофизических свойств растворов хладагент/компрессорное масло. Рассмотрены методические аспекты экспериментального исследования термических свойств реальных рабочих тел для пароконденсационных холодильных машин и общие принципы прогнозирования термодинамических свойств подобных систем. Показано, что примеси компрессорного масла в хладагенте значительно снижают параметры эффективности компрессорной системы. Рассмотрен вопрос селективной растворимости многокомпонентных хладагентов в компрессорных маслах.

Ключевые слова: Раствор хладагент/компрессорное масло – Термические свойства – Параметры эффективности – Селективная растворимость.

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РОЗЧИНИ ХОЛОДОАГЕНТ/КОМПРЕССОРНЕ МАСТИЛО: ПРОБЛЕМИ ЗАСТОСУВАННЯ ТА ДОСЛІДЖЕННЯ ВЛАСТИВОСТЕЙ

У роботі проведено аналіз та узагальнення результатів дослідження теплофізичних властивостей розчинів холодоагент/компрессорне мастило. Розглянуті методичні аспекти експериментального дослідження термічних властивостей реальних робочих тіл пароконденсационних холодильних машин і загальні принципи прогнозування термодинамічних властивостей подібних систем. Показано, що домішки компрессорного мастила в холодоагенті значно знижують параметри ефективності компресійної системи. Розглянуто питання селективної розчинності багатокомпонентних холодоагентів в компрессорних мастилах.

Ключові слова: Розчин холодоагент/компрессорне мастило – Термічні властивості – Параметри ефективності – Селективна розчинність.

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REFRIGERANT/LUBRICANT MIXTURES: PROBLEMS OF APPLICATION AND PROPERTY RESEARCH

The results and generalizations of thermophysical property research for the refrigerant/lubricant mixtures are summarized. The methodological aspects of the experimental studies of the thermal properties of real working media for vapor compression refrigeration machines and the general principles of the thermodynamic properties simulation for such solutions are analyzed. It is shown that the admixtures of compressor oil in the refrigerant make the efficiency parameters of compressor systems much lower. The question of a selective solubility of the multicomponent refrigerants in compressor oils is discussed.

Keywords: Refrigerant/lubricant mixtures – Thermodynamic properties – Efficiency parameters – Selective solubility

I. INTRODUCTION

The transfer of refrigeration equipment to environmentally safe working fluids is still remaining unresolved. Currently, multicomponent refrigerants of fourth generation are used as a replacements for refrigerant R22. These refrigerants generally contain some hydrocarbons, which should provide satisfactory miscibility of mixed refrigerants with the lubricant oils. For example, azeotropic mixture of refrigerants

NH₃/DME (designated as R723 and consisting of 60 wt. % NH₃ and 40 wt. % RE170) is proposed as the alternative to ammonia [1, 2].

It should be emphasized that the real working fluid (RWF) in the vapor compression refrigeration systems is a solution of the refrigerant and the compressor oil. The presence of the compressor oil in the refrigerant leads to significant change in the thermophysical properties of the RWF. Therefore, the energy

efficiency of the refrigeration cycle, the intensity of heat transfer in the evaporator and condenser will be determined relating to the selected refrigerant and compressor oil [3 - 5].

RWFs are complex thermodynamic systems, since they consist of components which significantly differ in their physical and chemical properties. Difficulties in obtaining experimental information for the RWF are related primarily to the duration to establish thermodynamic equilibrium in the system, the zeotrope nature of the behavior of these solutions, and the concentration gradient in their surface layer [6].

The problems of modeling the thermophysical properties of RWF are connected to significant deviations of the behavior for such systems from the behavior of the ideal solutions, to the difficulties in using Maxwell rule to calculate the liquid-vapor phase equilibrium, to the lack of critical parameters, as well as the molecular weight for compressor lubricants. The question of the difference between the concentration of the surface layer and the concentration in the liquid phase of the RWF is not studied. The used models require a significant amount of empirical information in order to achieve an acceptable degree of accuracy in the description of the experimental data. Therefore, problem of calculating the caloric properties and developing the pressure-enthalpy ($P-h$) diagrams for the RWF in this case can not be solved. Thus, the experimental studies remain to be the primary source of the reliable information on the thermophysical properties of the RWF. Hence, further development of calculating and predicting methods for the thermophysical properties of the RWF is still the actual scientific direction.

II. ANALYSIS OF THE RESULTS OF THE PROPERTY RESEARCH FOR THE REFRIGERANT/OIL MIXTURES

The main objectives of this paper were to study the methodological aspects of measuring the thermophysical properties of the RWF and the development of the general principles of the RWF thermodynamic modeling. The recent studies show that the time necessary to establish the thermodynamic equilibrium in the measuring cell with the sample of RWF is an important factor [6]. This factor determines the accuracy of the information about the properties of RWF. This time depends on the physical properties of the components, the thermodynamic parameters of the mixture and can reach several hours. The results of our experimental studies show that the time periods during which the vapor pressure, density, capillary constant, and the concentration of RWF take the values close to equilibrium, are approximately the same and usually do not exceed 6 hours at the constant parameters of the RWF. The results of research [6, 7, 8] show that the duration of establishing the thermodynamic equilibrium is determined by the rate of the diffusion processes between the vapor phase of the sample (consisting almost pure refrigerant) and the liquid solution. Therefore, the time of establishment of thermodynam-

ic equilibrium substantially depends on the temperature and the concentration of RWF and increases markedly with increasing concentration of oil in the RWF and decreasing temperature. In addition, the equilibration time for the RWF depends on the ratio of the densities of the components. The time to establish thermodynamic equilibrium becomes less when the density of the liquid refrigerant is higher than the density of oil.

The results of the capillary constant measurements are important in the of surface tension research. The time to establish equilibrium values for this property (in the capillary rising method) depends not only from the average parameters of the thermodynamic system (temperature and concentration), but also from the height of the meniscus level of the RWF liquid phase in relation to the levels of the liquid in the measuring cell [6]. At low temperatures and high oil concentration in the RWF, the time to establish equilibrium value of the capillary constant may takes at least from 3 to 5 days in our experiments [6]. The capillary constant depends not only on the composition of the RWF liquid phase, but also on the composition of the solution surface layer enriched with the component decreasing the surface tension [6, 8, 9]. It may be assumed that an excess of refrigerant in the surface layer leads to decreases of the capillary constant and surface tension of the. As follows from results reported in [6] small admixtures of refrigerant lead to significant decreases of the capillary constant of the ROS. The reason is that the adsorption of the low-boiling component will be so intensive that the surface layer of the ROS will consist almost entirely of molecules of refrigerant. As a result, the concentration dependence of the capillary constant of the ROS demonstrates flat behavior in the range of the oil mass concentrations from 0.4 to 1.

In [6, 8] showed that increasing the isochoric temperature of the RWF two-phase sample leads to the composition change of the solution liquid phase. This change should be taken into consideration, i.e. experimental data have to be attributed to the concentration of the liquid solution which is changing in the experiment. In the practice of the thermophysical experiment, mixing of the solution usually promotes the elimination of the temperature and concentration gradients over the sample volume. In this case, the measured parameters become the stable values. This fact is evaluated as an achievement of the equilibrium state of the thermodynamic system. However, the constant values of the thermodynamic parameters may significantly differ from the true equilibrium values. This is due to the fact that the sample is not an isolated thermodynamic system (the sample receives an additional energy from the mixing device) in conditions of intensive stirring. Hence, the equilibrium values of the thermodynamic properties can be obtained only in case the thermodynamic system is isolated. Thus, the dynamic method for the phase equilibria and surface tension can not be recommended for the property research of the RWF [6]. Only the static methods make

it possible to obtain reliable experimental information of the RWF phase equilibria and surface tension.

The experimental results obtained for the surface tension at the RWF liquid-gas interface enable to quantify the adsorption of the refrigerant in the surface layer [6, 8]. As it was shown, the refrigerant excess adsorption for the RWF is always positive. Consequently, the surface layer is enriched with the refrigerant (see Figure 1).

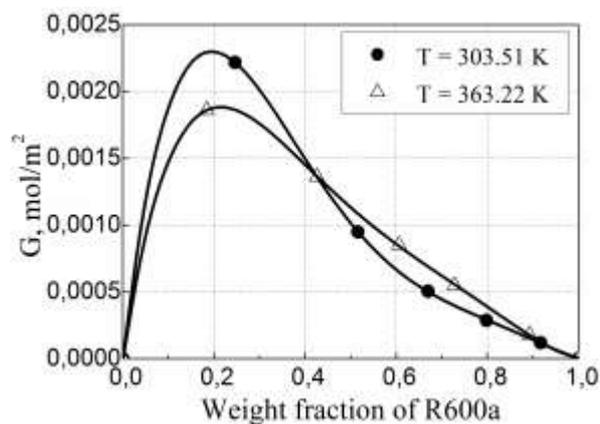


Figure 1 – Isotherms of the adsorption of isobutane in liquid phase of isobutane/oil mixture

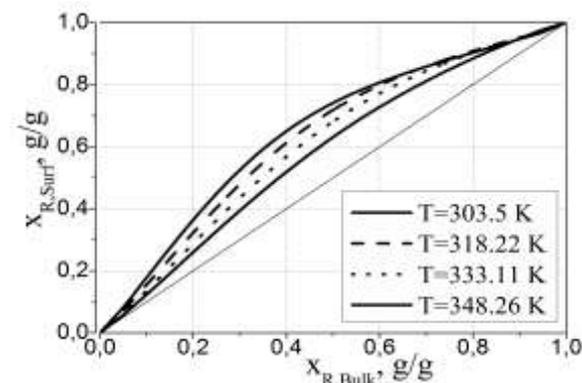


Figure 2 – Concentrations of the surface layer of the isobutane/oil mixture

The concentration of the surface layer differs significantly from the concentration of the liquid phase (see Figure 2). Thus, the concentration of the surface layer not only determines the value of the surface tension for the RWF, but also affects the behavior of the saturated vapor pressure isotherms.

Analyzing the methods for modeling the thermodynamic properties of the RWF some important problems should be considered. First of all, it is extremely difficult to find the data on the properties of compressor oils, their average molecular weight, and pseudocritical parameters. There are only few publications dedicated to the thermophysical properties of the compressor oils in a wide range of parameters and methods for determination the pseudo-critical parameters [10 - 12].

The determination of the RWF pseudocritical parameters is an important problem in modeling the

properties of the RWF. The potential opportunities of the corresponding states law for RWF depends on well-received values of the pseudo-critical parameters. Experimental determination of the RWF critical parameters using the stationary methods is not possible due to thermal instability of compressor oils. In [10] showed that the only exception is the dynamic method of pulse heating. The studies devoted to the research of the RWF pseudo-critical parameters are currently not available in the literature. In these circumstances, the priority is the calculation methods for determining the RWF pseudocritical parameters. New approaches for prediction the pseudocritical parameters that require a limited volume of the experimental information are given in present study.

As it was shown above, the surface layer of the RWF has a concentration that differs from the bulk concentration of the solution liquid phase. Therefore, the calculation of the pseudocritical temperature using the saturated vapor pressure of the RWF to describe the volumetric properties of solutions (density and viscosity) is not quite correct. Results of treatment of experimental data for the density and viscosity of the oils and the RWF lead to important conclusion: to describe the bulk properties of the RWF (density and viscosity) one should use its own set of pseudocritical parameters other than the values obtained in the framework of the theory of thermodynamic similarity based on investigation of the saturated vapor pressure.

Thus, the lack of information about the properties of the surface layer is one of the main problems which arise when a thermodynamic interpretation of the experimental data on the RWF surface tension, as well as for the vapor-liquid phase equilibrium. The composition of the layer differs from the concentrations of the solution coexisting phases. This circumstance makes a problem to develop methods of prediction the capillary constant, surface tension and vapor pressure of the RWF. For this reason, the deviations of the experimental data on saturated vapor pressure and surface tension from the values calculated by existing thermodynamic models very often many times larger than the uncertainty of the original experimental data.

From above follows, that Guggenheim model of the surface layer can be applied for analysis the thermal properties of the RWF. In this model, the liquid phase consists of the RWF bulk phase and the surface layer of finite thickness, enriched by volatile components compared to the bulk phase (see Figure 3). Each parts of the solution should have its own set of pseudocritical parameters which should be separately applied to describe the RWF surface properties (saturated vapor pressure and surface tension) and the RWF bulk properties (density and viscosity). Since questions about the thickness of the surface layer or its equivalent (average thickness) for the RWF are poorly understood, this hypothesis needs further development.

Successful simulation of the RWF thermophysical properties is impossible without information regarding the average molecular weight of the compressor oils. The best for experimental investigation of the

average molecular weight of the compressor oils are ebullioscopic and cryoscopic methods. An overview of published data for the pseudocritical parameters and an average molecular weight for some compressor oils and the results of our measurements are given in papers [11, 12].

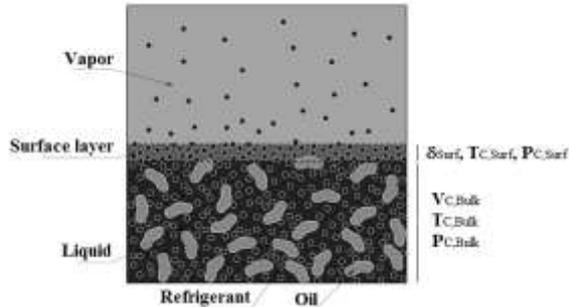


Figure 3 – Guggenheim model of the surface layer

III. IMPLEMENTATION IN INDUSTRY

It is well known that the energy efficiency, starting characteristics of the compressor, heat transfer in the apparatus, oil circulation along the compressor system, reliability of the compressor are determined by the RWF thermophysical properties.

An application of the “classical” ($P-h$) diagrams for the pure refrigerants does not allow taking into account the influence of compressor oil admixtures to the efficiency of the compressor system and intensity of the heat transfer in evaporator. Pressure-enthalpy ($P-h$) diagrams for RWF can be developed only by using information for the thermodynamic properties of the RWF [4, 13]. Creation of these diagrams should be performed for various concentrations x_g of the compressor oil flow circulating around the contour (x_g is the oil mass concentration before throttling device). In such diagrams, boundary curve for the liquid phase of the RWF practically coincides with the boundary curve for the pure liquid refrigerant. The isotherms near the condensation line have significant curvature and deviate from the isotherms for the pure refrigerant-

ant. The degree of deviation increases with increasing concentration of oil flowing through the contour of the compressor system. As the liquid phase of the RWF always has oil admixtures, a pair of boundary curve for the real working fluid is missing. The flows of the refrigerant, the oil and the RWF circulating on the circuit of the compressor system and calculated using the obtained diagrams are shown in Figure 4. This figure shows that the composition of the refrigerant/oil mixture differs significantly in different parts of the compressor system at $x_g = \text{const}$.

Figure 5 shows the change in the amount of the residual liquid in the evaporator (m_l) and the concentration of oil in the residual liquid (x_{oil}) in a parallel flow evaporator. This figure indicates that the most rapid increase in the oil concentration in the residual liquid in the evaporator occurs at high degrees of vapor quality of the RWF.

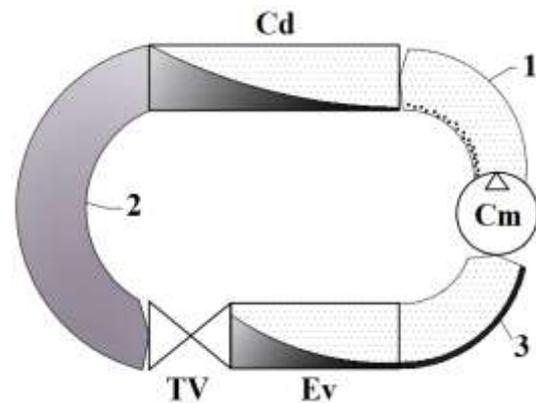


Figure 4 – Scheme of oil and RWF circulating in the circuit of the compressor system: TV = throttle valve, Ev = evaporator, Cd = condenser, Cm = comp-ressor. 1 = vapor of the refrigerant $m_{vap} = 96\%$, drops of refrigerant/oil mixture $m_{liq} = 4\%$, 2 = refrigerant/oil mixture $m_{liq} = 100\%$ ($x_R = 96\%$, $x_{oil} = 4\%$), 3 = wet steam of the refrigerant $m_{vap} = 91.7\%$, refrigerant/oil mixture $m_{liq} = 8.3\%$ ($x_R = 47\%$, $x_{oil} = 53\%$). Parameters in condenser: $T = 303 \text{ K}$, $P = 0.41 \text{ MPa}$; in evaporator: $T = 258 \text{ K}$, $P = 0.088 \text{ MPa}$.

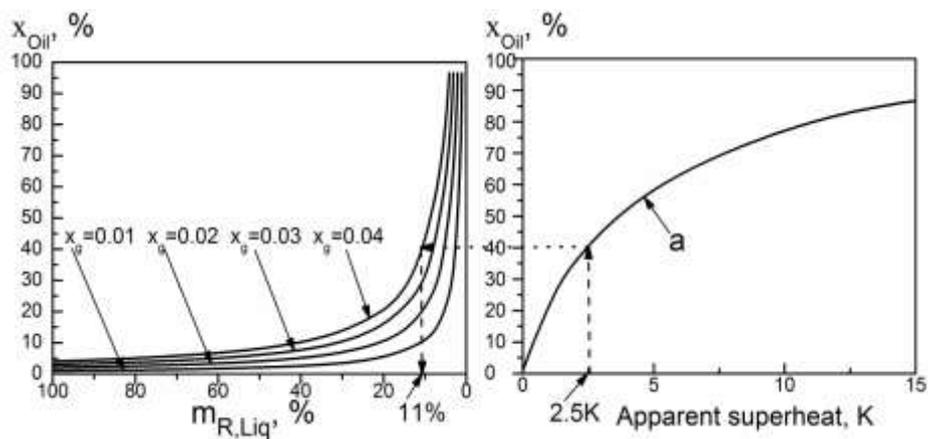


Figure 5 – Varying of remaining liquid quantity in evaporator $m_{R,Liq}$ and oil concentration in remaining liquid x_{oil} at parallel-flow evaporator (boiling temperature of pure refrigerant R600a = 270K). Auxiliary curve “a” yields detection of x_{oil} and $m_{R,Liq}$ at different values of apparent superheat ΔT .

Curve on the right side of the figure shows that the residual liquid m_l is equal to 8.3% from the total mass of working fluid for the apparent superheating to 2.5 K (as compared with the boiling point of the pure refrigerant R600a). This solution contains 53% of compressor oil and 47% of the ballast (not boiling out) refrigerant. Therefore, minimally sufficient value of the apparent superheat for that working fluid should be about 7.5 K. It is important to mention that the conclusion is valid only for that specific working fluid at the certain concentration (4%) of the compressor oil before throttling device.

Relative contribution of the oil admixtures in decreasing the specific refrigerating effect rate at optimum superheat increases with drop the boiling temperature in the evaporator and equal approximately 1.5 percent per 1 percent the concentration x_g . Overall negative impact of the oil admixtures to absolute value of the coefficient of performance depends on both the boiling point in the evaporator and the concentration x_g .

At the minimum permitted overheating of the working fluid (for the working fluid R600a/naphthenic mineral oil) relative contribution of the oil admixtures are within 5-7% to 1% of x_g . The overall impact of the compressor oil admixtures on performance of the compression system using RWF R600a/naphthenic mineral oil for the given cycle parameters is listed in the Table 1.

Table 1 – The impact of the oil admixtures on the performance of the compression system

Parameter, %	Concentration of the oil before throttle valve			
	1%	2%	3%	4%
q_{RWF}/q_{Ref}	97.88	96.39	94.89	93.40
$\varepsilon_{RWF}/\varepsilon_{Ref}$	91.44	85.03	79.32	74.17

The issue of withdrawal of HCFCs (R22, R142b, etc.) from application has acquired now even more importance. It should be noted the problem of transfer of the refrigeration equipment to the environmentally safety working fluids has still not been fully resolved. As a replacement for R22, the mixed refrigerants based on such components as: R32, R125, R143a and R134a (407C, 410A, etc.), as well as a number of new environmentally safety blends: R417A, R422A, R422D, R419A had been proposed and are widely used for today. A common feature of the fourth generation of mixed refrigerants is the presence of a small amount of hydrocarbons (R600a, R600) or dimethyl ether (RE170) in their composition (in addition to commonly used compounds R143a, R134a, R125). For design, the hydrocarbons and RE170 included in the working fluids have to ensure satisfactory miscibility of blended refrigerants with the polyester and the traditional use mineral or alkylbenzene oils.

It was many attempts to replace the refrigerant R22 by ammonia (R717), especially in the commercial refrigeration equipment [14]. However, in this way a number of restrictive factors which connected with technical difficulties, for example, such as selection of

compressor oil or additional costs for safety will be increased. Despite the fact that ammonia in some areas of application is almost an "ideal" refrigerant, the promising direction to develop substitutes is still actual. First of all, it is relevant for the commercial refrigeration, heat pumps and water-cooling plants of various capacities. For example, azeotropic mixture which consists 60 wt. % NH_3 and 40 wt. % RE170 is currently proposed as a substitute for the ammonia. The refrigerant (R723) has been tested on several industrial refrigeration system. Refrigerant R723 is dissolved in mineral oils. Therefore, the problem of compressor oil return to the compressor can be solved without troubles. Mass flow of the refrigerant is to 1.5 times greater than for ammonia. Consequently, at the same size of the compressor, the cooling capacity and cross section of the pipe refrigeration unit on the R723 will be less. Comparison of thermodynamic characteristics shows that the discharge temperature at the compressor system with the refrigerant R723 to less than the work on ammonia, whereas the specific cooling capacity increases slightly. COP for the equipment with R723 comparable and slightly higher for large degree of compression compared with the ammonia refrigeration machines. The tests show that the heat transfer coefficient in the evaporator for the mixture of R723/mineral oil higher than that for ammonia with mineral oil [1, 2].

Thus, a significant portion of the proposed alternative refrigerants are binary, ternary or multicomponent mixtures. Correspondingly, the application of such refrigerants may potentially cause to violation of the composition. This phenomenon leads primarily to change in performance of the compression system and may increase the fire hazard of refrigeration equipment. The reasons why the change of the composition of the working fluid might occur are evident: 1 - the redistribution of the components between phases of the working fluid in the process of evaporation and condensation, 2 - selective leak of the refrigerant components from the system or human error during the procedure of filling of the zeotropic refrigerant into refrigeration machine, 3 - different solubility of the components in the compressor oil, which is critically important even for azeotropic refrigerants. First two reasons mainly relevant for the zeotropic refrigerants. Unfortunately, the selective solubility of the component of multicomponent refrigerants the compressor oil still remains poorly studied. Only few publications dedicated to this important issue can be found in existing literature [15 - 17].

In order to demonstrate the importance of the question related to selective solubility of the multicomponent refrigerants in compressor oils we present the results of the experimental study of the concentrations of mixed refrigerant dissolved in the compressor oil and vapor composition, coexisting in equilibrium with these solutions. Experiments were conducted in a specially designed experimental setup [18]. The samples of R717 and RE170 with purity 99.5 wt. % have been used in the experiments. The results of solubility measurements of R723 and ammonia in oils are given in Tables 2 - 4.

Table 2 – Solubility of R723 and its components in mineral compressor oil ISO 68

x_R , wt. %	t , K	P_S , MPa	Composition of mixed refrigerant		Concentration difference $ x_{R717} - y_{R717} $, wt. %
			x_{R717} / x_{RE170} , wt. %	y_{R717} / y_{RE170} , wt. %	
9.81	333.13	2.695	51.2 / 48.8	67.1 / 32.9	15.9
9.44	355.54	3.801	50.6 / 49.4	73.1 / 26.9	22.5

Table 3 – Solubility of R723 and its components in mineral compressor oil ISO 46

x_R , wt. %	t , K	P_S , MPa	Composition of mixed refrigerant		Concentration difference $ x_{R717} - y_{R717} $, wt. %
			x_{R717} / x_{RE170} , wt. %	y_{R717} / y_{RE170} , wt. %	
8.28	333.15	2.704	43.8 / 56.2	69.0 / 31.0	25.2
11.95	355.54	4.051	51.5 / 48.5	74.1 / 25.9	22.6

Table 4 – Solubility of refrigerant R717 in compressor oils

Oil	x_R , wt. %	t , K	P_S , MPa
Mineral compressor oil ISO 68	3.99	333.18	2.614
Mineral compressor oil ISO 68	5.90	355.56	4.350
Mineral compressor oil ISO 46	4.27	333.13	2.606
Mineral compressor oil ISO 46	6.33	355.58	4.347

The data obtained for the selective solubility of the components for the mixed azeotropic refrigerant R723 in compressor oils indicate that formation of the RWF in the crankcase goes with violation of the initial concentration. Consequently, concentration of the multicomponent working fluid circulating in the compressor system changes as well. It is evident that problem of the selective solubility of the multicomponent working fluids is an important stage for the development of the alternative refrigerants and on their implementation into the industry. Finally, additional studies are required for the parameters of a certain refrigeration machine, such as: the volume ratio of the oil and the refrigerant, the coolant temperature and pressure in the compressor. Based on results [18] presented in Table 4, we can make a conclusion about limited solubility of the ammonia in compressor mineral oils. This effect leads to change of the RWF viscosity in the compressor and should be considered both for designing and exploiting of the refrigeration plant.

IV. CONCLUSIONS

Refrigerant-compressor oil solutions are complex thermodynamic systems of indefinite composition, consisting in thermally unstable components. This fact should be considered in the experimental study of thermophysical properties and in development of methods for the thermodynamic modeling. Large time to establish thermodynamic equilibrium, the restrictions on the intensity of stirring the solution, adsorption of the refrigerant in the surface layer of the RWF liquid phase, change in concentration of the liquid phase at the heating of the sample along the isochore affected the accuracy of the obtained experi-

mental data. Selective solubility of components of the mixed refrigerant in the compressor oils is not studied enough. Relatively low quality of experimental data on the thermodynamic properties of the RWF, lack of information on the molecular weight of oils and their thermophysical properties, lack of published data on the RWF pseudocritical parameters hampered progress in development of the prediction methods for the properties of the real working fluids for refrigeration equipment. Since the compressor oil admixtures in the refrigerant have significant influence in the energy efficiency of the compressor systems further studies the thermophysical properties of RWF still remain relevant.

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