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**Theoretical study of working fluids used in Organic Rankine
Cycles and similar processes**

“Ph.D. Thesis Booklet”

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1. Introduction

The rapid increase in the price of electricity is a result of a decline in the availability of fossil fuels, which coincides with a rise in demand and numerous emerging environmental issues. These concerns are driving the search for clean, long-term energy alternatives to fossil fuels. Consequently, the world is increasingly attracted to utilizing power generation plants that operate with renewable energy sources, low-grade temperatures, waste heat recovery, and are economically sustainable. These qualities are present in the Organic Rankine Cycle (ORC) and the Trilateral Flash cycle (TFC). Furthermore, with the growth and development of many societies and decreasing reserves of traditional energy resources constantly, there is more need for energy sources and promising technologies [1,2]. Therefore, the ORCs are one of the best solutions to utilize various heat sources at low and medium temperatures [3–5], previously considered non-usable for power production. Unlike steam Rankine cycles, ORC and TFC are able to generate power at a low heat source temperature of 80–85 °C, while they will be more attractive at a high heat source temperature that exceeds 300 °C and has an extensive range of applications [6].

The performance and system design of the ORC system are fully reliant on the working fluid; thus, working fluid selection for ORCs is critical [7,8]. In a TFC and ORC system, hundreds of different organic materials can be used, which have low boiling point temperatures and can replace water as the working fluid [9]. The selection of the working fluids is difficult to challenge because it should have favorable thermodynamic performance and be environmentally friendly [10]. The physical parameters of working fluids (such as critical temperature, latent heat of vaporization, standard boiling temperature, specific heat, molecular complexity, and molecular weight) are crucial for power cycles to achieve excellent cyclic performance [11,12]. Therefore, the working fluids should have favourable thermophysical and transport properties at operating conditions and meet several other criteria, including being cost-effective, readily available, nontoxic, nonflammable, environmentally friendly, and chemically stable [13]. Important output parameters, like net power (W_{net}) and efficiency (η_{th}) are affected by the physical parameters of the cycles [14,15]. Moreover, both heavily depend on the thermodynamic properties of working fluids [16].

This dissertation examines the above-mentioned set of problems from several interrelated aspects. Two interrelated topics display the results according to pure and mixture working fluids:

1.1 Pure working fluids: This section involves studying the effect of some factors, output parameters, and properties of working fluids, such as the heat source temperature effect of the ORC and TFC performance, thermal efficiency comparison between ORC, and TFC by using basic ORC and recuperative ORC, the effect of working

fluids critical temperature (T_{cr}) on the thermal performance of ORC, and TFC, finding the subcategory of hydrofluoroethers (HFEs) working fluids, and the prediction that some of HFEs fluids can dominate the other fluids at specific conditions, and finally making a map of saturated alkanes fluid which help to predicate the category of working fluids. Regarding the pure working fluids, five journal papers, two pre-reviewed conference papers, and two conference abstracts were published, and one oral presentation was given in the workshop.

1.2 Mixture working fluids: This section involves studying the disadvantages of wet fluids and the costly system of using dry fluids. Consequently, we are looking for the possibility of finding the isentropic type of fluid by using mixed working fluids prepared from chemically similar structure materials to obtain almost ideal zeotropic mixtures because the isentropic fluids are preferred compared to others. Using an isentropic type, neither a superheater nor a recuperative and regenerative heat exchanger is required due to the fact that expansions are occurring and ending at the saturated vapor curve. Another part of this section focuses on the effect of the critical temperature of working fluids and the category of fluids on the ORC thermal performance by using four different thermodynamics models. Regarding the mixtures, two journal papers were published.

2. Research gap and theses

2.1. Pure working fluids

The increase in heat source temperature is thought to be always desirable in simple thermodynamic cycles when the goal is better performance. However, other constraints (like the cost of utilizing this added heat source) can overshadow the gain caused by the efficiency increase. Geothermal energy is often considered a low-grade energy source; therefore, it cannot independently support high-load applications. This is true even for countries with quite good geothermal potentials (like Hungary), where the well-head temperature of most of the existing geothermal wells is below 90 °C. However, the hybrid solar-geothermal installation is probably the most frequently used double-source design [17,18], where geothermal heat is used to preheat the compressed liquid, while solar heat is used to reach the maximum temperature and for evaporation. Still, engineers should know there is a narrow temperature range near the critical temperature, where thermodynamic efficiency has inverse maximum cycle temperature dependence. Therefore, there is a temperature range, which should be avoided during this application. Thus, this study aimed to prove that there is a subcritical temperature we cannot exceed; otherwise, the output parameters (thermal efficiency and net power) decrease.

Thesis 1

Increasing the maximal cycle temperature is an accepted method to increase thermodynamic efficiency. Using the basic layout (without superheating process) for several common power cycles (Organic Rankine cycle, steam Rankine cycle, and CO₂ cycle, but not for Trilateral Flash Cycle), an inverse effect has been found close to the critical point in the real and ideal cycle. A maximum of thermal efficiency and output energy appear at subcritical temperatures for most working fluids (except for the so-called super-dry ones where the T - s diagram has a very tilted dome). Above that value (which depends on the working fluids and the minimal cycle temperature), the increase in the maximal cycle temperature is counterproductive. Related publications [P1, P2, P10, P11].

Generally, heat recovery is useful with most of the power cycles, for example, the organic flash Rankine cycle [19]. Heat recovery can increase cycle efficiency in mixed and pure working fluids [20]. Therefore, some heat recovery devices (recuperator or regenerator) are recommended for all cycles with recoverable heat, which can be subcritical, transcritical, or supercritical [21,22]. One of our studies showed that, for the basic Rankine-like cycles (ORC, steam-Rankine cycle (RC), and subcritical CO₂ cycle), the thermal efficiency is generally higher than TFC. This statement is true for most working fluids, except in a few ones, like dodecane [23]. Therefore, we show that even by using super dry working fluids, ORC efficiency can overcome the TFC efficiency by using a recuperator regardless of the source temperature. The intersection points of the two efficiency lines (Equal Efficiency Point – EEP) are defined, and this point moves with the changing heat recovery ratio that has been studied. In this way, the maximum thermal efficiency in the function recuperator effectiveness can be found for ORC and TFC.

Thesis 2

The subcritical efficiency maximum cannot be seen by using super-dry (T - s diagram a very tilted dome) working fluids (like dodecane) with a basic (recuperator-free) layout. In these cases, the efficiency of Trilateral Flash Cycles (TFC) can exceed the efficiency of Organic Rankine Cycles (ORC), which is never seen with wet or moderately dry working fluid. By using super-dry working fluids, higher recuperator effectiveness increases the thermal efficiency of TFC and ORC. The increase is more significant for ORC; therefore, above a certain recuperator effectiveness, the efficiency of ORC always exceeds the efficiency of TFC, while in the recuperation-free (basic) case, the TFC can be only more efficient at higher maximal cycle temperatures. Related publications [P3, P8].

Numerous papers reported that the thermal performance, such as η_{th} and W_{net} , and some other parameters are strongly related to working fluids T_{cr} . The mentioned quantities increase with increasing the critical temperature of the working fluid used in the ORC

or TFC. Our assumption is that this is true, but only to a limited extent. Generally, one can say that the need to find any established correlation between the thermal properties of working fluid (especially the simple ones, such as critical temperature) and the thermodynamic performance of ORC and other cycles would be desired. Any of these correlations would be a great help in pre-screening working fluids. Unfortunately, the results are controversial; therefore, one can only say that for ORC, the critical temperature has strong limitations in working fluid evaluations [24]. One can assume that this statement is also valid for cycles similar to ORC. Thus, we aim to demonstrate the effect of working fluids T_{cr} on thermal performance for some subcritical cycles such as ORC and TFC by using the pure and mixture working fluids at the low-grade heat source. 13 alkanes and 10 halogenated alkanes were employed in this investigation. Finally, general recommendations and the effect of the working fluids T_{cr} on thermal performance have been summarized.

Thesis 3

Despite the critical temperature being an important factor for working fluids, the quantities describing the thermal performance are not strongly correlated with the higher critical temperature of working fluids. In contrast, it can only acceptably correlate with output parameters in a group of working fluids that chemically have a similar structure. Related publication [P4].

In recent years, many efforts have been made to substitute high Global warming potential (GWP) working fluids (WFs) with low-GWP. For example, hydrofluorocarbons (HFCs) were developed to replace hydrochlorofluorocarbons (HCFCs) with high ozone depletion potential (ODP) and GWP [25]. HFEs fluids are attractive due to their low GWP and zero ODP, which leads the researchers to pay specific attention to their utilization in the near future [26,27]. Despite the HFE being described as an attractive organic fluid, only a few theoretical and experimental investigations have been done to date; therefore, this type of fluid needs more studies.

Due to zero ODP, relatively low GWP, a very short atmospheric lifetime compared to HFC, non-flammable compared to Hydrocarbons (HCs) and Hydrofluoro-Olefins (HFO), and minimal toxicity, HFEs are considered a good candidate for use as working fluids in energy conversion systems and lead us to pay specific attention to their utilization in the near future; therefore, this study aims to analyze the η_{th} in the subcritical ORC, TFC, and PE-ORC by using three kinds of HFE (HFE7000, HFE7100, and HFE7500), with various T_{max} . Also, showing HFEs behaviors (normal dry or very dry working fluids) and comparing them to some of the other selected fluids in different categories such as HCs, HFCs, and HCFCs in η_{th} terms.

Thesis 4

The behavior of the most common HFEs (HFE7000, HFE7100, and HFE7500) fluids was obtained from the thermal efficiency analysis. HFE7000 is normal dry fluid, HFE7100 is moderately dry, and HFE7500 is super dry (T-s diagram a very tilted dome). Moreover, although under similar conditions, HFEs (HFE7000, HFE7100, and HFE7500) have lower thermal efficiency than conventional fluids (HCs, HFC, and HCFC) in ORC, but they are still a good competitor in TFC and PE-ORC under specific conditions, especially since they are environmentally friendly. Related publication [P5].

Due to the difference between working fluids, the optimal one should be chosen in almost every case separately. Obviously, there are other non-thermodynamic criteria during this optimization (flammability, price, etc.). GWP and ODP are considered two crucial points for working fluids, precisely when these two factors have a small value. Thus, they don't have a significant negative environmental impact. Several of the thermodynamically favorable working fluids have unacceptably high GWP and/or ODP; the use of most of these materials – mainly the halogenated alkanes, chloro-, fluoro-, and chloro-fluoro-hydrocarbons, frequently used as refrigerants and working fluids - are forbidden by the subsequent Montréal (1987) and Kyoto (1992) Protocols or by the Kigali Amendment (2016) [28].

The list of materials that are already forbidden or phased out is increasing; therefore, energy engineers have to play a very complex game to design ORC equipment using particular fluids that can be used for a sufficiently long time without banning their working fluid. One of the solutions is to use so-called “natural” working fluids; their presence in nature can be limited only in exceptional cases. The most well-known group of these natural working fluids are the alkanes (linear, branched, and cyclic ones) found in natural gas and/or crude oils. When finding an alkane as a working fluid for a given application, one can be sure that the lifetime of the given ORC installation will be influenced only by physical factors and will probably be unaffected by further regulative constraints.

Consequently, we presented the mapping of several alkanes concerning their working fluid characteristic. Compared to van der Waals fluids mapping, one can see good qualitative but poor quantitative agreements about the location of various working fluid classes. Using this mapping, one can either find a proper pure alkane to use as ORC working fluid for a given heat source – heat sink pair- or can obtain an idea of seeking a better-working fluid among the alkane mixtures.

Thesis 5

Reduced temperature – specific isochoric heat capacity map is a proper tool to help classify working fluids for model systems. Using this map, one can pre-classify all alkanes. Also, preliminary assumptions for the working fluid class for mixtures of close members of the homologue series can be obtained. This would help us check the applicability of pure and mixed alkanes previously unused as working fluids. Related publication [P9].

2.2. Mixture working fluids.

The shape of the T - s diagram significantly impacts the expansion properties and assists in predicting the final expanded state. The T - s diagrams of the categories of fluids classified as wet, isentropic, and dry [29], as shown in Figure 1.

Due to the wet type has liquid droplets (like droplet erosion [30,31], or superheated dry vapor in dry type (like the loss of residual heat or the need for an extra heat exchanger for heat recuperation [32,33]), looking for an isentropic type is required.

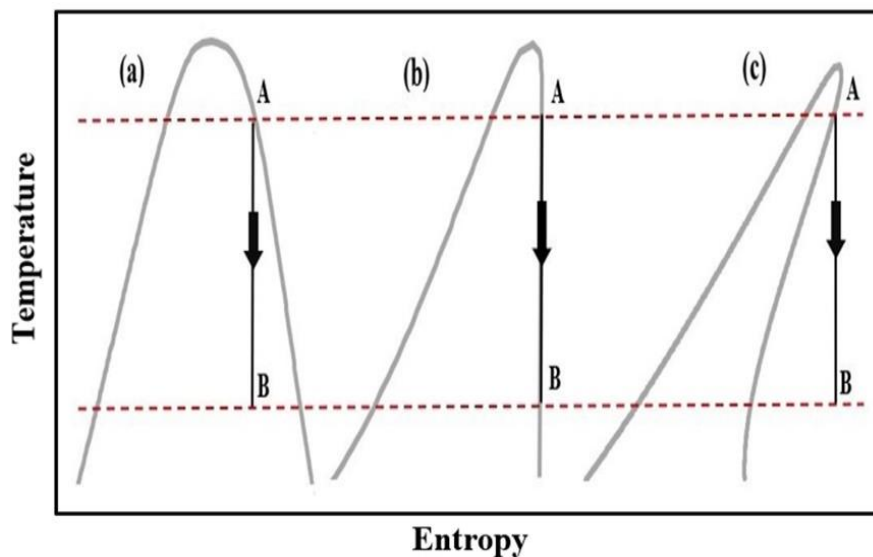


Figure 1. Traditional classification of working fluids, a) wet, b) isentropic, and c) dry category working fluids with ideal expansion between two different temperature levels, represented by points A and B.

Therefore, finding isentropic (or almost isentropic) working fluids is one of the goals for researchers dealing with ORC. Previous results indicated that by changing internal material properties (like molecular degree of freedom), one can go from the wet to dry category, and it is expected that the isentropic category might be seen during this transition). Because these internal properties cannot be changed continuously for pure materials, only discretely (like going from propane to butane), the investigation of mixtures was decided because, in these cases, fine changes can be reached by changing the composition. We aim to show the transition of mixed working fluids from the wet to dry category by mixing the hydrocarbons in various compositions and determining the appropriate transition point. propane, pentane, butane, and hexane were used as

working fluids to implement this. The exact nature of the transition has been determined, showing that even by using mixtures, strictly isentropic working fluids (ones having a part of the saturated vapour branch with infinite slope, spanning in a finite, nonzero temperature range) do not exist.

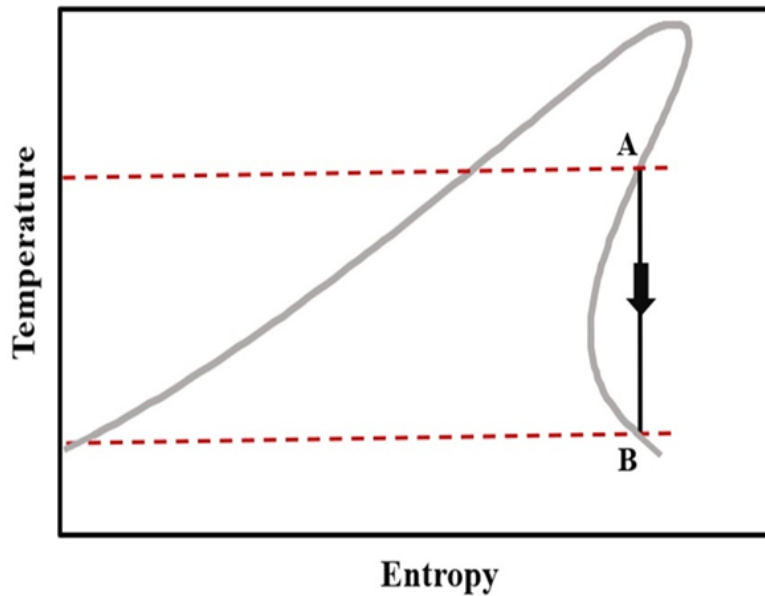


Figure 2. Real shape of working fluids, a) wet, b) “isentropic”, and c) dry category working fluids with ideal expansion between two different temperature levels, represented by points A and B.

Thesis 6

Despite the isentropic working fluids being more desirable than wet and dry, they are hard to find. The isentropic type (when the slope of the saturation curve on the T - s diagram is infinite in a nonzero temperature range) does not exist during the smooth transition from wet to dry using a mixture and up to four digits of compositions. Related publication [P6].

One of the studies concluded that the mixture of a low and high T_{cr} working fluid might have better thermal performance than the mixture of two highly critical temperature ones [32]. Additionally, Fan et al. [24] found that the zeotropic mixture of fluids from different categories (dry/wet) has a better performance compared to other mixtures made by the same categories, such as (dry/dry) and (wet/wet).

Also, some studies show that the best thermal performance of ORC was by using the zeotropic mixture compared to the pure fluids, and there is a good relation between thermal performance and the mixture T_{cr} and the categories of fluids. Therefore, the motivation of the present study is to clarify these points. In this part, we deal with the ideal process and aim to demonstrate the effect of the critical temperature of mixed working fluids on the thermal performance of subcritical ORC by using three scenarios

and four models at the low-grade heat source. General recommendations for choosing a properly mixed working fluid will be given, and the effect of the critical temperature of working fluids on thermal performance will be summarized. All of the working fluids investigated in this study are familiar to engineers and researchers, which others studied as well, for example [33].

Thesis 7

The impact of critical temperature and working fluid category on ORC thermal performance cannot be described straightforwardly. Consequently, the optimal values of thermal performance are not a monopoly to a specific category of the mixture (wet/wet, wet/dry, dry/dry) or the highest value of the critical temperature of working fluids, but rather depend directly on the proper selection of the mass fraction and thermodynamic model (setting the maximal and minimal temperature on the bubble and dew points line) for ORC application. Furthermore, using mixtures does not necessarily increase ORC system performance compared to pure fluids. Related publication [P7].

List of Publications

Journals publications

[P1] Ahmed AM, L. Kondor, A.R. Imre. Thermodynamic Efficiency Maximum of Simple Organic Rankine Cycles. Energies 2021;14: 307. <https://doi.org/10.3390/en14020307> 2021:14020307. (Q1, WoS, IF = 3.004), (Related to thesis).

[P2] Ahmed AM, Imre AR. Effect of high temperatures on the efficiency of sub-critical CO₂ cycle. Pollack Period 2021;16:73–9. <https://doi.org/10.1556/606.2021.00310>. (Q3, Scopus) (Related to thesis).

[P3] Ahmed AM, Imre AR. Investigation of thermal efficiency for subcritical ORC and TFC using super dry working fluids. Energy Sci Eng 2022;11:711–26. <https://doi.org/10.1002/ese3.1356>. (Q2, WoS, IF = 4.035), (Related to thesis).

[P4] Imre AR, Ahmed AM. Effect of the working fluids critical temperature on thermal performance for trilateral flash cycle and organic Rankine cycle. Int J Thermofluids 2023;20:100417. <https://doi.org/10.1016/j.ijft.2023.100417>. (Q1, Scopus), (Related to thesis).

[P5] Ahmed AM. Assessment of the thermal efficiency of subcritical power generation cycles using environmentally friendly fluids at various heat source temperatures. Energy Sci Eng 2022;10: 4768-4781. <https://doi.org/10.1002/ese3.1307>. (Q2, WoS, IF = 4.035), (Related to thesis).

[P6] Ahmed AM, Imre AR. Wet - to - dry transition description in the mixture of working fluids. *Discov Appl Sci (SN Applied Science)* 2024; 6:56. <https://doi.org/10.1007/s42452-024-05702-x>. (Q2, WoS, IF = 2.6), (Related to thesis).

[P7] Ahmed AM, Imre AR. Effect of critical temperature and category of zeotropic mixture working fluids on the thermal performance in subcritical ORC. *Int J Thermofluids* 2023;20:100400. <https://doi.org/10.1016/j.ijft.2023.100400>. (Q1, Scopus), (Related to thesis).

Conference proceeding and abstracts publications

[P8] Ahmed AM, Imre AR. The effect of recuperator on the efficiency of ORC and TFC with very dry working fluid. *MATEC Web of Conferences* 2021; 345: 00012, <https://doi.org/10.1051/mateconf/202134500012>. Pilsen (Czech Republic), (Related to thesis).

[P9] Ahmed AM, Kustán R, Groniewsky A, Imre AR. Alkanes as natural working fluids for organic rankine cycles. *AIP Conf Proc* 2021;2323. <https://doi.org/10.1063/5.0041439>. Pilsen (Czech Republic), (Related to thesis).

[P10] Ahmed AM, Imre AR. Imre, Critical temperature effect at the wet working fluids in the subcritical ORC performance. *The 13th International Exergy, Energy, and Environment Symposium (IEEES-13) Book of Proceeding*; 2022. https://www.researchgate.net/profile/Abdullah-Alzahrani-9/publication/370709806_Book_of_Proceedings_of_The_13th_International_Exergy_Energy_and_Environment_Symposium_IEEES13/links/645e54befbaf5b27a4c14df3/Book-of-Proceedings-of-The-13th-International-Exergy-Energy-and-Environment-Symposium-IEEES13.pdf , Makkah (Saudi Arabia). (Related to thesis).

Others

[P11] Ahmed AM, Imre AR. Effect of heat source temperature on ORC and TFC efficiency. 16th Miklós Iványi International PhD & DLA Symposium, University of Pecs, Oct 26-27.2020. https://issuu.com/pivanyi/docs/book_of_abstract_2020. Budapest (Hungary) (Book abstract), (Related to thesis).

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