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Thermodynamic comparison of three small-scale gas liquefaction systems

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Abstract

Natural gas liquefaction systems are based on refrigeration cycles, which can be subdivided into the cascade, mixed refrigerant and expander-based processes. They differ by their configurations, components and working fluids, and have therefore various operating conditions and equipment inventory. The present work investigates three configurations suitable for small-scale applications because of their simplicity and compactness: the single-mixed refrigerant, single and dual reverse Brayton cycles. The impact of different feed compositions and refrigerant properties is analysed. A detailed assessment of the energy and exergy flows is conducted, and the most promising cycle layouts are identified by performing multi-objective optimisation procedures. The findings illustrate the resulting trade-offs between the system performance and size in different operating conditions. Mixed-refrigerant processes prove to be more efficient (1000-2000 kJ/kg_{LNG}) than expander-based ones (2500-5000 kJ/kg_{LNG}) over larger ranges of operating conditions, at the expense of a greater system complexity and higher thermal conductance (250-500 kW/K against 80-160 kW/K). The results show that the use of different thermodynamic models leads to relative deviations of up to 1% for the power consumption and 20% for the network conductance. Particular caution should thus be exercised when extrapolating the results of process models to the design of actual gas liquefaction systems.

Keywords: Gas liquefaction, process optimisation, process modelling, multi-objective optimisation, exergy analysis

1. Introduction

Liquefied natural gas (LNG) is a liquid mixture of hydrocarbons consisting mainly of methane, generally produced at high pressure (20 to 50 bar) and stored at about -160 °C and near atmospheric conditions. LNG is a cleaner fuel than conventional fossil fuels such as black oil because of the smaller emissions of nitrogen and sulphur oxides. For this reason, it is suggested as a substitute for heavy oil as a marine fuel. LNG presents other advantageous properties, such as high energy content - about 45-50 MJ/kg - and density - around 20-23 MJ/L. It can also be used for delivering natural gas to remote areas not connected to pipeline infrastructures.

The installed capacity of LNG production facilities exceeds 320 mtpa (million tonnes per annum) worldwide, of which around 6% corresponds to small-scale facilities, i.e. with a capacity lower than 1 mtpa per unit [1]. The development of small-scale plants has gained interest in the last decades, because of the possible environmental, geopolitical and economic benefits. Some of the main issues of small-scale LNG facilities are: (i) the higher production cost per unit of LNG, (ii) the global profitability of the supply chain, (iii) the limitations in terms of system design. Improving the performance of such systems, while maintaining them compact, is key to a successful development and deployment. The requirements for small- and large-scale

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Nomenclature

T	Temperature, K or °C	PR	Peng-Robinson
\dot{E}	Exergy rate, W	RB	Reverse Brayton
\dot{Q}	Heat rate, W	SMR	Single Mixed Refrigerant
\dot{W}	Power, W	SRK	Soave-Redlich-Kwong
p	Pressure, Pa	WI	Wobbe Index
<i>Abbreviations</i>		<i>Subscripts</i>	
DRB	Dual Reverse Brayton	0	dead state
EOS	Equation of State	D	destruction
GERG	Groupe Européen de Recherches Gazières	F	fuel
LNG	Liquefied Natural Gas	L	loss
NG	Natural Gas	P	product

LNG plants are different: the performance (power consumption) of the gas liquefaction process is of key importance in all cases, but other factors such as the equipment count, dynamic behaviour and compactness are of bigger importance for the former. For example, processes such as the propane precooled mixed refrigerant system (C3MR) and the pure-refrigerant cascade with nine or ten pressure levels are preferred for large-scale applications because of their high efficiency, but they are not suitable for small-scale because of their high equipment count and capital cost [2].

For this reason, the present paper investigates the thermodynamic performance of three refrigeration systems for production of liquefied natural gas, which are suitable for small-scale applications. Several research groups have investigated the efficiency of these processes, but the literature on small-scale LNG systems is scarcer. Barclay and Denton[3] compare mixed refrigerant and expander-based processes for offshore applications, aiming at listing selection criteria. Their work suggests that expander-based processes are well-suited because of their compactness and high inherent safety. It also pinpoints the low performance of such cycles compared to state-of-the-art ones, but does not include a thorough analysis of the space requirements and cost performances. Finn[4] compares mixed-refrigerant and expander-based processes for small-scale applications and concludes that (i) the former can be cost-effective if plate-fin exchangers and single compressors are used, and (ii) such cycles can compete with expander cycles for such sizes. Cao et al.[5] evaluate the performance of two types of small-scale LNG processes in skid-mounted packages: a nitrogen-methane cycle and a single mixed-refrigerant (SMR) process. They conclude that the first can be superior to the second in the absence of propane pre-cooling. Remelje and Hoadley[6] analyse the performance of four refrigeration cycles for small-scale applications. The single mixed-refrigerant process was taken as a reference, to which three other processes, belonging to the expander-based category, are compared. They state that the SMR process displays the smallest power consumption per unit of LNG produced, and that open-loop expansion processes are highly sensitive to the feed gas composition. Pérez and Díez[7] present the current situation of small to medium-scale LNG facilities, and discuss the technical factors that impact the selection of the refrigeration process. They propose qualitative parameters to help decision-makers. Castillo and Dorao[8] analyse the cost formation for expander and mixed-refrigerant technologies, with a focus on the heat transfer area costs. He and Ju[9] evaluate sixteen configurations of expansion cycles for distributed-scale systems from a thermodynamic point of view and optimise these setups based on their second-law efficiency. The literature survey shows that there is no clear agreement on the most suitable liquefaction process for small-scale applications - mixed-refrigerant processes are more efficient, but harder to control because of the refrigerant composition and possible maldistribution issues. The single-mixed

refrigerant process, as well as the nitrogen single- and dual-expander systems, have at present attracted most attention.

Several of these works use the exergy analysis method to assess the thermodynamic inefficiencies of liquefaction processes. Remelje and Hoadley[6] show that the SMR process is characterised by a high share of exergy destruction within the cryogenic heat exchanger, followed by the aftercoolers and the compressors. They also prove that the distribution of the thermodynamic inefficiencies is different from mixture processes to expander ones. Shirazi and Mowla[10] present the simulation results of SMR concepts and pinpoint the aftercoolers as the most exergy-destroying components, followed by the compressors and the LNG heat exchanger. Similar conclusions are drawn by Xu et al.[11]. More recently, Morosuk et al.[12] apply exergy-based methods (exergetic, exergeo-economic and exergo-environmental) to the PRICO process, which is a SMR process. They suggest to improve the cryogenic heat exchanger. It is the component responsible for most irreversibilities and the associated cost is the highest. The numerical differences between those results are imputable to the assumptions on the component efficiencies, refrigerant composition and feed properties.

The optimisation of these liquefaction systems has also been the subject of many studies, as shown with the annotated bibliography of Austbø et al.[13]. Most works deal with a single objective function, such as the minimisation of the power consumption, capital costs, operational expenses, or the maximisation of the net profit. Boulougouris and Papanikolaou[14] optimise the design of a LNG terminal with focus on the motion response and sea surface evaluation. Shah et al.[15] present a multi-objective optimisation approach of a two-stage reverse Brayton cycle. Shirazi and Mowla[10] use a genetic algorithm to optimise SMR layouts to minimise the specific power consumption, which can be decreased from the 1500 kJ/kg_{LNG}-range to the 1100 one. Castillo and Dorao[16] deal with the single-mixed refrigerant process and emphasise the importance of the market prices (gas and electricity) on the quality of the optimisation solutions. Khan and Lee[17] optimise the SMR process efficiency using a particle swarm approach, achieving up to 10% savings in power consumption. Few studies [18] compare several cycles by conducting multi-objective optimisation routines, analysing the trade-off of e.g. the network conductance and power consumption.

The recent research in this field investigates numerous aspects of LNG production systems, from exergy analysis to dynamic performance and system optimisation. Yuan et al.[19] simulate and optimise a single nitrogen expansion process with carbon dioxide pre-cooling, suitable for small-scale applications, and perform an exergy analysis to depict the sources of inefficiencies. Fazlollahi et al.[20] compare alternative liquefaction options in the frame of cryogenic carbon capture systems in different operation modes. Khan et al.[21] focus on the dual mixed-refrigerant process, with the objective of minimising simultaneously the heat transfer conductance and compression power. Mehrpooya et al.[22] introduce a novel configuration for gas liquefaction systems at large scale, using absorption refrigeration systems. Fazlollahi et al.[23] investigate the performance of two liquefaction processes in transient conditions, during load variations. New control methods and alternative heat exchanger designs are suggested to improve the system performance. Pham et al.[24] propose an alternative decision-making method to improve the performance of single mixed-refrigerant processes, with a focus on the compression power. Song et al.[25] conduct single- and multi-objective optimisations of a single nitrogen expansion process with carbon dioxide pre-cooling to maximise the system efficiency and liquefaction rate, using a genetic algorithm.

All these works consider only one thermodynamic model in the optimisation procedure. The annotated bibliography of Austbø et al.[13] shows that the equation of state (EOS) of Peng-Robinson (PR) [26] is the most widely used, followed by the Redlich-Kwong [27] EOS with Soave modifications (SRK) [28]. Few use the Reference Fluid Thermodynamic and Transport Properties (REFPROP) database developed by the National Institute of Standards and Technology (NIST) [29]. Cubic equations of state are known for the poor prediction of the liquid properties [30,31], especially in the case of methane. These inaccuracies can result in large discrepancies between computational results and experimental data, as underlined by Dauber and Span[32] and Yuan et al.[33].

The present paper aims at completing the current state-of-the-art. First, it deals with the thermodynamic performance of three potential small-scale gas liquefaction processes. At the difference of most papers, several natural gas feeds from European countries (five) are taken as case studies, to investigate the relation between the feed and refrigerant compositions. Secondly, three different thermodynamic models are used to compute the refrigerant properties and the power consumption of each cycle, to investigate how the selection of one or

another impacts the final results. Finally, the trade-off between the system performance and size is assessed by performing multi-objective optimisation routines on the mixed-refrigerant and expander-based processes.

2. Methods

2.1. System description

The processes investigated in this work are the following: the single mixed-refrigerant (SMR) (Figure 1), the single- (Figure 2) and the dual-expander (Figure 3) systems. The SMR process has attracted interest because of the low number of equipment items, and is considered for small-scale applications in several research works [6,8,11,12,34]. Single- and dual-expander cycles for small-scale LNG production has also gained attraction among researchers, because of their simplicity. They are suitable for e.g. offshore applications with exploitation of remote gas sources [6,8,9,35,36]. They have been developed and analysed extensively by liquefaction technology providers [2,4,37,38]. The first one belongs to the category of mixed-refrigerant processes, in which a mixture of hydrocarbons (e.g. methane, ethane, ethylene, propane, butanes, pentanes) and nitrogen is used as a refrigerant, and the temperature level of -165 to -162 °C is achieved by the Joule-Thomson effect (adiabatic expansion through a valve device). The working fluid is in two-phase in the coldest parts of the system and latent heat can therefore be exploited throughout most of the process. Natural gas is cooled, liquefied and subcooled down from state 1 to state 2 in a cryogenic three-flow heat exchanger. The refrigerant is cooled down in parallel (6 to 7), is expanded through an adiabatic valve (7 to 8), entering the two-phase region. It is then redirected to the cool box where it is fully evaporated and superheated (8 to 9), compressed (9 to 10) and condensed back to liquid state (10 to 6).

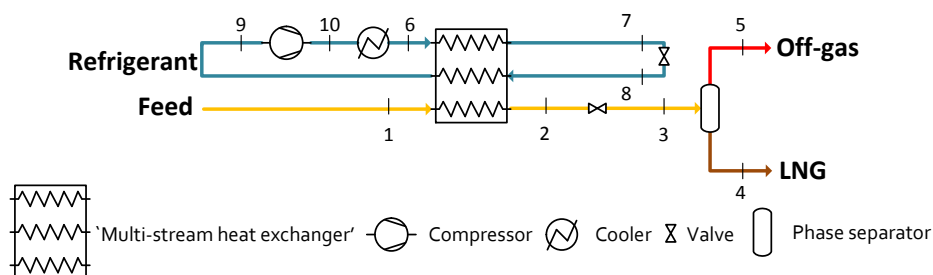


Figure 1: Process flowsheet of the single mixed-refrigerant process

The two other processes belong to the class of expander-based systems, in which one (single) or two (dual) fluids act as refrigerants. The main concept is the use of a reverse Brayton cycle. The temperature level of -165 to -162 °C is achieved by decreasing the vapour pressure through an expander in which work is extracted, and heat is rejected to the environment in the intercooling and aftercooling steps. The working fluid is only in vapour phase throughout the complete cycle, which results in larger flows than in the mixed refrigerant process, as only sensible heat can be exploited. In general, the refrigerant is a pure fluid (nitrogen or methane) but may be a mixture of these two. Natural gas is cooled, liquefied and subcooled from state 1 to state 3 in two cryogenic heat exchangers. The refrigerant is pre-cooled in parallel (7 to 8), expanded (8 to 9), and is then heated near the ambient conditions (9 to 10 and 11). It is finally compressed (12 to 13) and cooled back (13 to 7). One- to two-stage compression processes are common for mixed-refrigerant processes because of the small pressure ratio, while three to four stages are usual for expander-based systems.

The advantage of the mixed-refrigerant process is two-folded. First, the match of the temperature profiles on the hot and cold sides is improved because both natural gas and refrigerant are zeotropic mixtures. In other words, they condense and evaporate over a range of temperatures, and not at a fixed temperature level as for pure fluids. The refrigerant composition and temperature glide can thus be tuned to thermally match different feed compositions. Secondly, the refrigerant is mostly in two-phase conditions, which leads

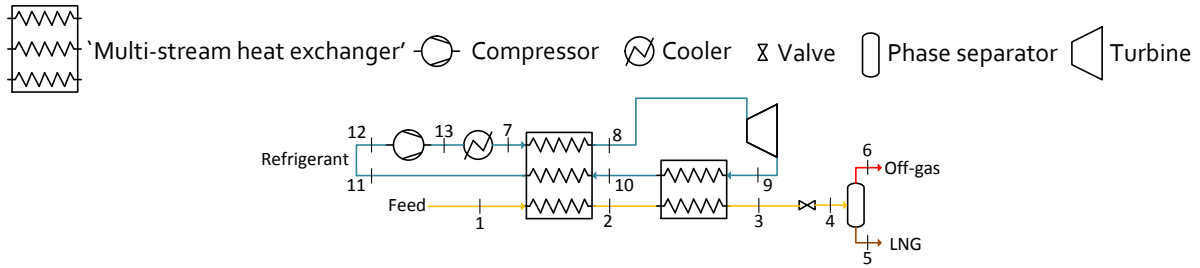


Figure 2: Process flowsheet of the single expander-based process

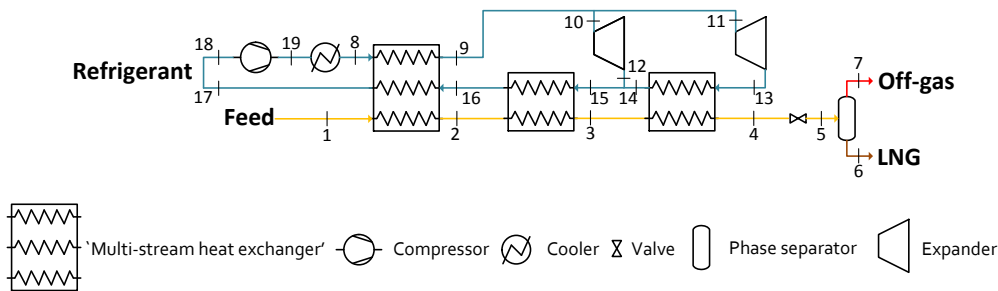


Figure 3: Process flowsheet of the dual expander-based process

to high heat transfer coefficients. The advantage of the expander-based process is the system simplicity: the refrigerant is in gaseous phase, which avoids instabilities and maldistribution issues in the heat exchangers. Few equipment items are required, and the system may be inherently safe if inert refrigerants, such as nitrogen, are used.

2.2. System modelling

The process models were developed in Aspen Plus version 7.2 using the Peng-Robinson equation of state (EOS) [26], which is a thermodynamic model widely used in the oil and gas industry for simulating hydrocarbon processes in which few (if any) polar compounds are present. The selection of other thermodynamic models such as the Redlich-Kwong [27] with Soave modifications [28] EOS, or the multi-parameter GERG [39] EOS, is discussed later. The present work considers natural gas compositions from five European countries (Table 1), which differ in the proportions of each hydrocarbon and nitrogen. One of the goals of the present study is to assess how different feed compositions affect the power demand and system efficiency. The models build on the following assumptions:

- the natural gas feed has an initial temperature and pressure of 20 °C and 32 bar.
- the produced LNG has a temperature of -160 °C after subcooling and is delivered at 1.7 bar;
- the recovered off-gas after subcooling and expansion is not re-liquefied;
- heat losses and pressure drops inside the heat exchangers are neglected;
- the compressors have a polytropic efficiency of 72 %, which corresponds to the lower bound of today's LNG compressors [6];

- intercooling to a temperature of 20 °C within the compression process can be achieved; intercooling temperatures of 40 °C may be preferred in some countries because of the initial temperature of the cooling water
- the turbines have an isentropic efficiency of 80 % [12];
- for the dual cycle, the temperatures at the outlets of each heat exchanger are fixed to the dew point, bubble point and subcooled temperatures of the natural gas (desuperheating in the first heat exchanger, condensing in the second one and subcooling in the third one);
- cooling water is available at a temperature of 10 °C.

Table 1: Natural gas grid composition for Denmark, Italy and Spain after removal of carbon dioxide, water and heavy hydrocarbons, given in molar fractions [40]

	CH ₄	C ₂ H ₆	C ₃ H ₈	n-C ₄ H ₁₀	i-C ₄ H ₁₀	N ₂
Denmark	0.903	0.060	0.024	0.006	0.004	0.003
Italy	0.980	0.007	0.002	0.001	0.000	0.009
Spain	0.816	0.134	0.037	0.004	0.003	0.007
France	0.904	0.062	0.015	0.005	0.002	0.011
England	0.924	0.038	0.008	0.002	0.001	0.027

2.3. System analysis

The gas liquefaction processes were evaluated by conducting energy and exergy analyses. *Energy* can be stored, transformed from one form to another (e.g. heat to power) and transferred between systems, but can neither be created nor destroyed (conservation law). *Exergy* is defined as the ‘maximum theoretical useful work (shaft work or electrical work) as the system is brought into complete thermodynamic equilibrium with the thermodynamic environment while the system interacts with it only’ [41]. Unlike energy, exergy is not conserved but is destroyed to a certain extent because of irreversible phenomena taking place in real processes (e.g. heat transfer).

$$\sum_{\text{in}} \dot{E}_{\text{in}} = \sum_{\text{out}} \dot{E}_{\text{out}} + \dot{E}_{\text{D}} \quad (1)$$

$$\dot{E}_{\text{P}} = \dot{E}_{\text{F}} - \dot{E}_{\text{D}} - \dot{E}_{\text{L}} \quad (2)$$

where:

- \dot{E}_{in} and \dot{E}_{out} are the exergy of a stream (matter, heat or power) entering or exiting the process;
- \dot{E}_{D} is the destroyed exergy, which accounts for the performance losses within the liquefaction system;
- \dot{E}_{P} is the product exergy, which denotes the desired effect in exergy terms, in this case, the increase of exergy of the liquefied natural gas;
- \dot{E}_{F} is the fuel exergy, which represents the resources spent to liquefy the natural gas, in that case, the net power consumption;
- \dot{E}_{L} is the lost exergy, which characterises the exergy discharged into the environment without any practical use, such as the heat discharged in the intercooling and aftercooling processes.

An energy analysis allows therefore for tracking the energy flows and the conversion from one form to another. An exergy analysis identifies the location, magnitude, and causes of thermodynamic inefficiencies, and gives hints on possible system improvements.

2.4. System performance

The performance of an LNG production system can be evaluated based on technical, economic and environmental indicators, and the present work focuses on thermodynamic aspects:

- the specific power consumption per unit of liquefied natural gas w ;
- the coefficient of performance (cooling) COP of the refrigeration cycle;
- the exergetic efficiency ε , which relates the minimum theoretical power consumed to produce the cooling capacity to the actual one;
- the exergy destruction ratio y_D^* , which shows the contribution of each individual equipment to the total amount of irreversibilities

The first two indicators can be deduced from the energy balance of the overall system, while the two latter are a direct application of the second law of thermodynamics.

2.5. System optimisation

The system was optimised in two steps (Table 2). First, a *single-objective optimisation* was performed, with the aim of maximising the system performance. Secondly, *multi-objective optimisations* were conducted, to minimise both the power consumption and system size. The latter is correlated to the size of the heat exchanger network and thus to the overall heat transfer conductance. These objective functions (maximising system performance and minimising its size) are conflicting. For example, higher cycle efficiency can be achieved by minimising the temperature difference between the hot and cold sides in the LNG/refrigerant heat exchanger. However, the required heat transfer area (A) and the conductance of the overall heat exchanger conductance (UA) will increase accordingly. These trade-off are of particular interest for this study and the results are shown as Pareto frontiers [42], where any improvement in one objective results in deterioration of another one.

Table 2: Optimisation method of the gas liquefaction processes

Objective functions:

- Minimise the specific power consumption w
- Minimise the overall heat network conductance UA

Subject to constraints:

- Minimum temperature approach 3 K
- Minimum vapour fraction in expanders 92 %
- No two-phase conditions in expander-based cycles

Decision (design) variables:

- Operating pressures (low- and high-levels)
 - Precooling temperature
 - Mixture flowrate
 - Mixture composition
-

The decision variables of the optimisation problem differ with the process under study (Table 3). They can be grouped into the cycle parameters (e.g. temperature and pressure levels) and fluid properties (e.g. chemical composition). We considered eight possible chemical compounds in the optimisation routines, which leads to the generation of mixture compositions that can be challenging to handle in practice (mixture stability, miscibility, distribution). The results give therefore a lower bound of the power consumption of the liquefaction system, which would be greater if only five chemical components such as methane, ethane,

Table 3: List of decision variables - the partial flowrates are expressed in kmol per kg of processed natural gas

Parameter	Process	Range
High-level pressure (bar)	SMR	[10,50]
	N ₂	[40,100]
	Dual N ₂	[40,100]
	Dual CH ₄	[40,100]
Low-level pressure (bar)	SMR	[1,10]
	N ₂	[1,40]
	Dual N ₂	[1,40]
	Dual CH ₄	[0,1]
Precooling temperature (°C)	N ₂	[-60,-40]
Methane (kmol/kg _{NG})	SMR	[0.03,0.08]
	Dual CH ₄	[0.1,0.9]
Ethane (kmol/kg _{NG})	SMR	[0.03,0.08]
Propane (kmol/kg _{NG})	SMR	[0,0.05]
n-Butane (kmol/kg _{NG})	SMR	[0,0.02]
i-Butane (kmol/kg _{NG})	SMR	[0,0.02]
n-Pentane (kmol/kg _{NG})	SMR	[0,0.02]
i-Pentane (kmol/kg _{NG})	SMR	[0,0.02]
Nitrogen (kmol/kg _{NG})	SMR	[0,0.05]
	N ₂	[0.1,0.9]
	Dual N ₂	[0.1,0.9]

propane, *n*-butane and nitrogen are considered. In practice, pentanes may be avoided because of possible freezing issues in the subcooling process.

LNG systems, especially the ones using mixtures, are thermodynamically complex and there may be numerous interactions between the refrigeration cycles (case of dual). The optimisation problem displays therefore severe non-linearities and may present several local optima. Several works use evolutionary algorithms such as genetic ones, and the genetic algorithm developed by Molyneaux[43] is used in the present work, using the tuning parameters (e.g. population size) presented in Mokhatab and Poe[2] when applicable. Other techniques such as gradient-based techniques can be powerful for optimising SMR processes, as shown in Wahl et al.[44].

3. Results

3.1. System modelling

The present systems were simulated using the Peng-Robinson equation of state [26], which is a cubic equation of state well-suited for simulations of hydrocarbon processes. A literature survey shows that all optimisation studies of gas liquefaction processes were performed considering only one thermodynamic model. Only a few works, such as the one of Dauber and Span[32], discuss the impact of using different equations of state on the predictions of the power consumption and temperature conditions. The multi-parameter model of the ‘Groupe Européen de Recherches Gazières’ (GERG) may be seen as the most accurate model, as it builds on fundamental derivations of the Helmholtz free energy and includes reference equations of state such as the models of Span and Wagner[45]. It is computationally-costly but returns results within the uncertainty range of measurements. Other thermodynamic models may be used, such as the virial equations of Benedict et al.[46,47] and of Lee and Kesler[48], which lie on empirical measurements correlations. Models based on the perturbed chain model in the statistical theories Gross and Sadowski[49] have also gained interest in the last decade and their accuracy is significantly better for highly polar mixtures, as emphasised in Diamantonis et al.[50]. The carbon dioxide and water content of the natural gas and

refrigerant mixtures is negligible in the present work, and cubic equations of state are deemed satisfactory in comparison.

This work compares the accuracy of the PR and SRK models to the GERG one for the SMR and RB case studies. A comparison for the SMR process suggests that the PR EOS is generally more accurate than the SRK EOS in the prediction of the heat transfer rates and conductance UA. A similar conclusion can be drawn for the estimation of the temperature approach in the heat exchanger (Figure 4). This suggests that the prediction of the temperature profiles within the cryogenic heat exchangers is on overall more accurate with the PR model.

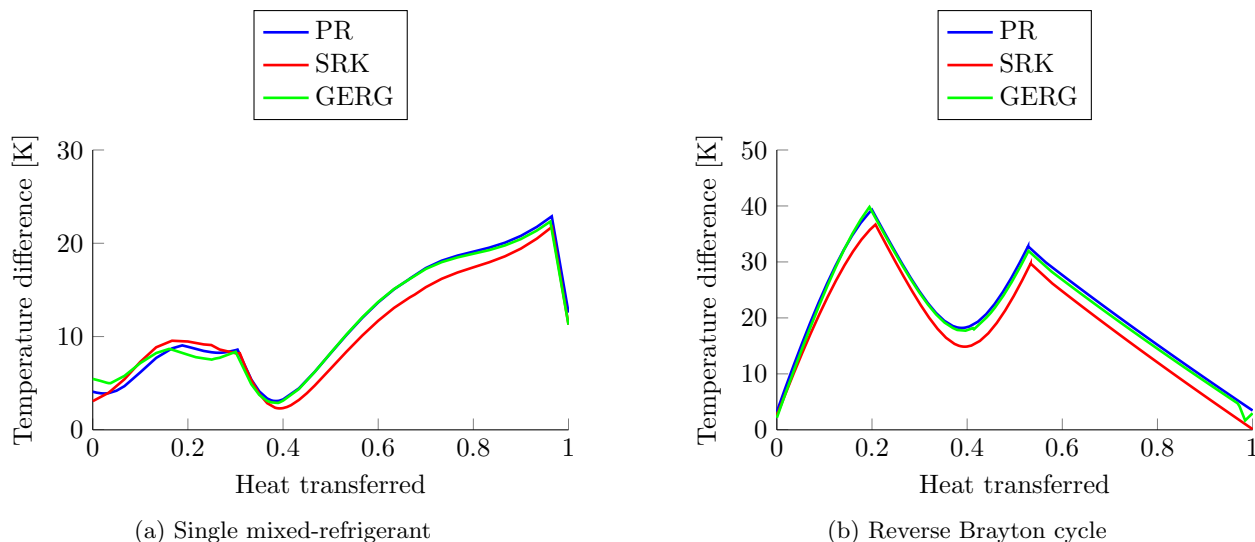


Figure 4: Comparison of the temperature differences within the cryogenic heat exchangers for different thermodynamic models

However, these conclusions cannot be generalised for all gas liquefaction processes and refrigerant compositions. The deviation in the prediction of the heat duty reaches up to 6% for the subcooling heat exchanger in the dual expansion system, but is only about 1.9% when considering the total amount of heat removed from 20 °C to -162 °C. This suggests that the Peng-Robinson equation of state presents deviations in the prediction of the dew and bubble points, and in the derivation of the specific heat capacity in the liquid phase, which is confirmed by numerous works in this field [32].

3.2. System analysis

3.2.1. Single mixed-refrigerant process (SMR)

The minimum power consumption of the SMR process is around 1500 kJ/kg_{LNG}. An analysis of the temperature profiles within the cryogenic heat exchangers (Figure 5) shows the close temperature match between the cold and hot sides in the heat transfer process. The minimum temperature difference (pinch point) is generally located at the dew point of the natural gas mixture, of around -75 to -80 °C. It may be at the hot end of the heat exchanger if the refrigerant undergoes significant superheating.

No direct correlations can be found between the natural gas properties and the refrigerant composition (Figure 6) that would lead to the minimum power consumption. Several trends are nevertheless observable. Firstly, a large share (70 to 90%) of the heat exchanger duty is actually associated with the refrigeration demand of the refrigerant itself. Large amounts of methane and ethane, which have the greatest cooling capacity, are thus not necessarily beneficial. Secondly, the refrigerant exiting the internal heat exchangers should be in superheated conditions. Small amounts of butanes and pentanes are preferable as they are the least volatile components, to ensure that the suction temperature is higher than the dew point. Thirdly, the Joule-Thomson coefficient of the refrigerant mixture must be positive overall, over the expansion process, to ensure a temperature drop. The use of nitrogen is thus advisable because greater nitrogen flow rates lead to bigger temperature drops.

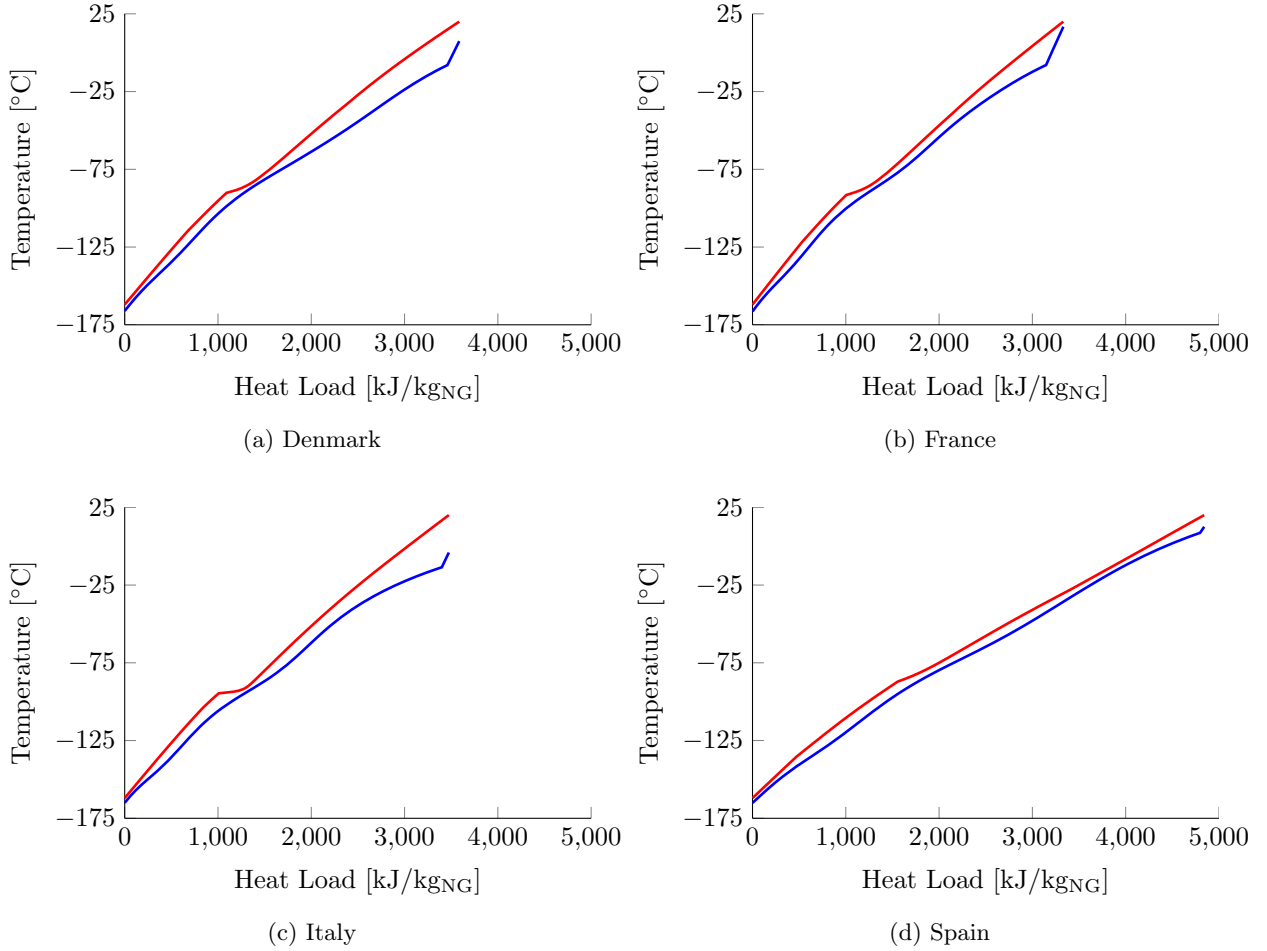


Figure 5: Temperature profiles within the cryogenic heat exchangers in the single mixed-refrigerant process

3.2.2. Reverse single and dual Brayton cycle

The minimum power consumption is around $3300 \text{ kJ/kg}_{\text{LNG}}$ for the RB process with nitrogen and $2500 \text{ kJ/kg}_{\text{LNG}}$ for the DRB system. Gaseous nitrogen has a much smaller cooling capacity than phase-changing hydrocarbons, implying greater flow rates. The heat duty of the cryogenic heat exchanger is nearly two times smaller than in mixture-based processes, and the temperature-enthalpy profiles are much more marked by the properties of the natural gas feed (Figure 7). The match between the hot and cold composite curves for natural gas feeds with the highest methane content (Italian and British cases), as the liquefaction process takes place over a smaller temperature glide.

The comparison of the temperature-heat profiles for the dual cycle shows that less heat is transferred, because of the lower refrigerant rates in the precooling and liquefaction heat exchangers. Substituting nitrogen by methane (Figure 8) process results in higher system performance. The power consumption decreases by about 7% for both single and dual configurations.

However, a main issue with methane is the sub-atmospheric pressure in the subcooling stage of the liquefaction process, which is not advisable because of the possible leakage issues. This issue can be circumvented if (i) higher subcooling temperatures are desired for the liquefied gas (for example, if the desired LNG temperature is -150°C instead of -160°C , the refrigerant temperature after expansion can be increased and the vapour pressure would not be sub-atmospheric), (ii) methane is used in a topping cycle and nitrogen in a bottoming one.

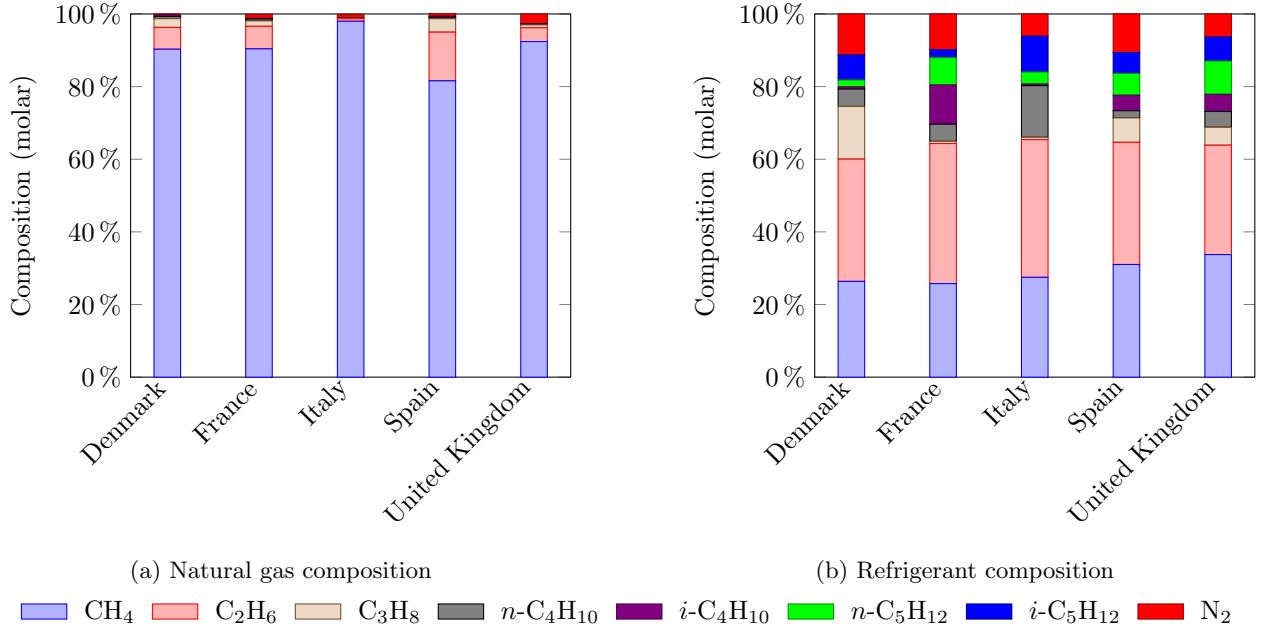


Figure 6: Chemical compositions of the natural gas and refrigerant for the single mixed-refrigerant process

3.3. Performance comparison

A comparison of the three processes after the single-objective optimisations shows that mixed-refrigerant processes feature smaller net power consumption than expander-based systems (Table 4).

Table 4: Power consumption of the studied gas liquefaction processes from the single-objective optimisations

	Mixed-refrigerant	Expander-based
w	1500 - 1800 kJ/kg _{LNG} 0.4 - 0.5 kWh/kg _{LNG}	2500 - 5000 kJ/kg _{LNG} 0.7 - 1.4 kWh/kg _{LNG}
$\frac{w}{LHV}$	3.5 % (Denmark)	5.4 % (Denmark)
$\frac{w}{WI}$	3.8 % (Denmark)	5.9 % (Denmark)

A detailed analysis of the results (Table 5) shows that the mixed-refrigerant processes are characterised by lower pressure levels and higher efficiencies. Two categories of optimum refrigerant compositions are depicted for the SMR process, the first with a large fraction of low-weight hydrocarbons, the second with a large fraction of medium-weight ones. However, the saturation pressure is lower for heavy than for light hydrocarbons at equal temperature. A lower value of the low-pressure level is required for mixtures with higher contents of medium- and high-weight hydrocarbons to prevent liquid formation at the compressor inlet.

3.4. Exergy analysis

A comparison of the three systems, based on an exergy analysis (Figure 9) of the Danish case, shows that the expander-based systems are less performant than mixture-based ones. The exergy destruction in the cryogenic heat exchangers is greater in the SMR process, despite the better temperature match, because of the greater cooling duty. On the contrary, the exergy destruction in the turbomachineries is greater for the reverse Brayton configurations, because of the higher refrigerant flowrate and higher pressure ratios.

The present analysis demonstrates the importance of the compression efficiency and pressure ratios for the performance of reverse Brayton processes, since the turbomachinery components represent up to

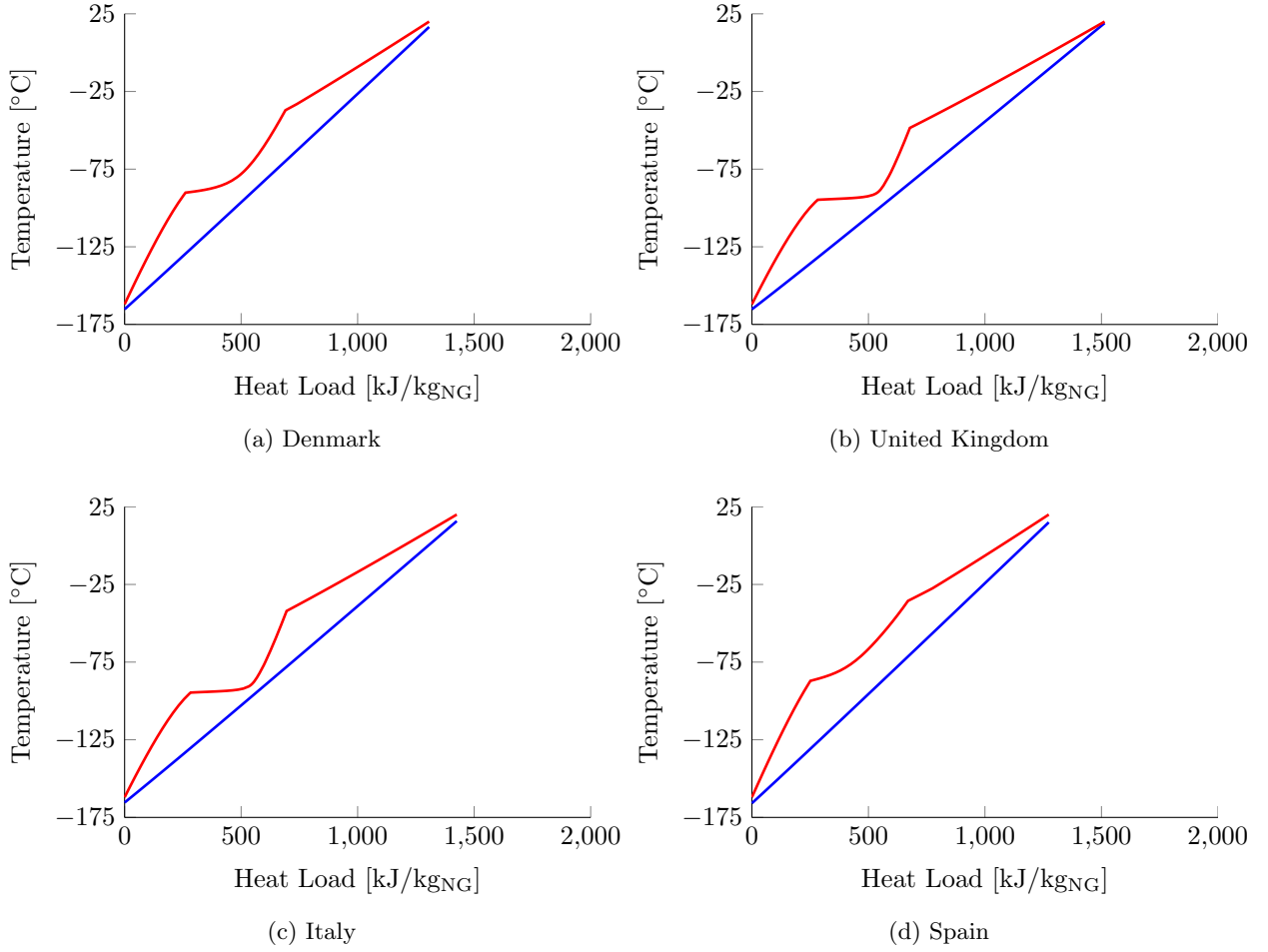


Figure 7: Temperature profiles within the cryogenic heat exchangers in the reverse Brayton process

Table 5: Performance of the mixed-refrigerant and expander-based processes

	SMR-1	SMR-2	RB-1	RB-2
Refrigerant composition				
Nitrogen (mol-%)	$\simeq 14$	$\simeq 7$	100	100
Methane (mol-%)	$\simeq 30$	$\simeq 21$	-	-
Ethane and propane (mol-%)	$\simeq 38$	$\simeq 51$	-	-
Butanes and pentanes (mol-%)	$\simeq 14$	$\simeq 19$	-	-
Pressure and temperature levels				
High level (bar)	$\simeq 33$	$\simeq 14$	$\simeq 65$	$\simeq 95$
Low level (bar)	$\simeq 4$	$\simeq 1.5$	$\simeq 3$	$\simeq 6$
Precooling ($^{\circ}\text{C}$)	-	-	$\simeq -48$	$\simeq -55$
Performance indicators				
w (kJ/kg _{LNG})	$\simeq 1500 \pm 1.7\%$		$\simeq 3300 \pm 1.5\%$	
COP	$\simeq 0.85$		$\simeq 0.46$	
ε (%)	$\simeq 33$		$\simeq 15$	

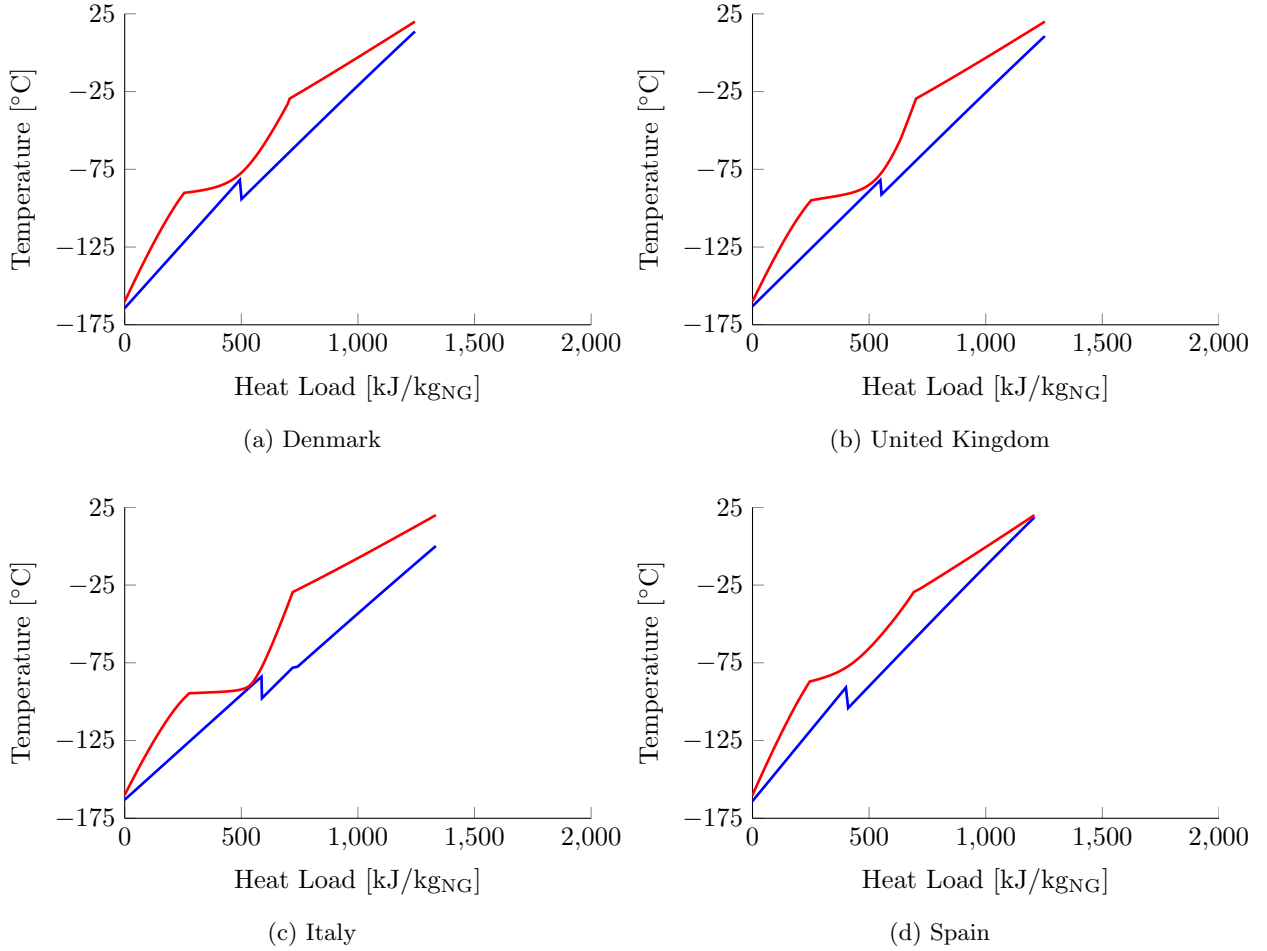


Figure 8: Temperature profiles within the cryogenic heat exchangers in the dual reverse Brayton process with methane as working fluid

60% of the total system irreversibilities. The use of more performant compressors would result into a smaller power consumption and cooling water demand, and in turn into smaller exergy destruction within these components. For mixed-refrigerant processes, improving both the cryogenic heat exchangers and the compressors appears worthy, as each is responsible for about 35% of the total exergy destruction. This suggests to invest into more efficient compressors and to put efforts on tuning the refrigerant composition adequately. Higher intercooling temperatures, as expected in warm countries developing LNG, would result in greater power consumption, exergy destruction in the compressors and coolers, and exergy losses with the cooling water.

3.5. System optimisation

A comparison of the gas liquefaction processes based on the minimisation of the net power consumption suggests that mixed-refrigerant processes are preferable against expander-based ones for small-scale liquefaction purposes. However, this single-objective approach does not consider other important factors such as the process size and the associated capital costs. A multi-objective optimisation was performed to assess the possibilities to minimise the conductance of the heat exchanger network and the total power consumption. The expander-based process displays the smallest values of heat exchanger conductances but greater power consumption (Figure 10), as a result of larger temperature differences between the hot and cold streams in the cryogenic heat exchangers.

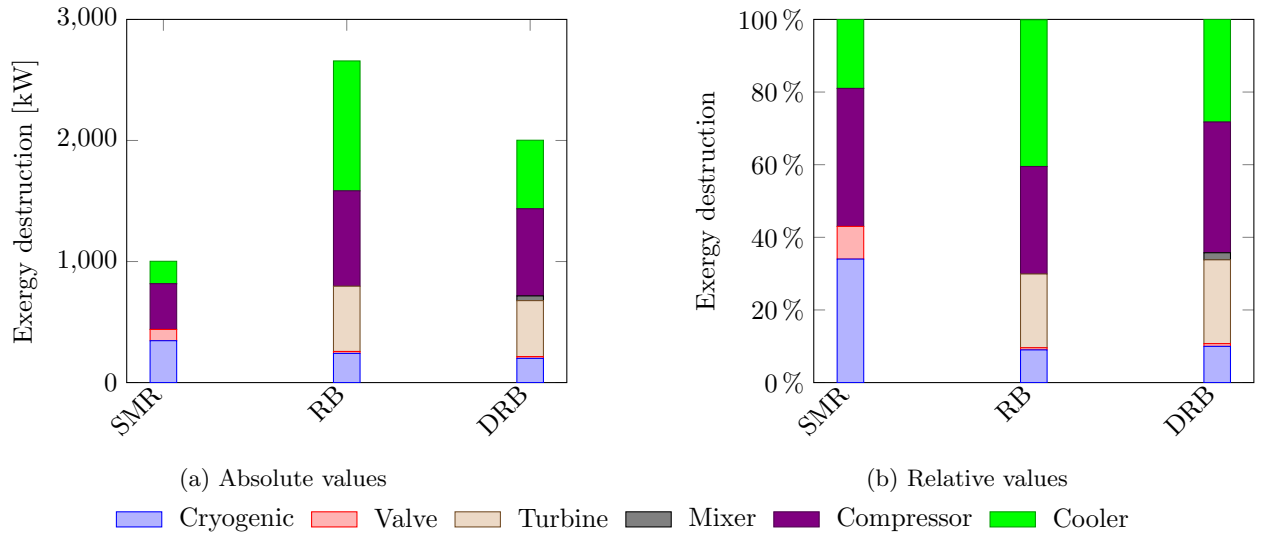


Figure 9: Distribution of exergy destruction in the LNG production systems

Table 6: Minimum power consumption and conductance of the studied liquefaction processes from the multi-objective optimisations

	Mixed-refrigerant	Expander-based
w	1500 kJ/kg _{LNG}	2500 kJ/kg _{LNG}
UA	250 kW/K	75 kW/K

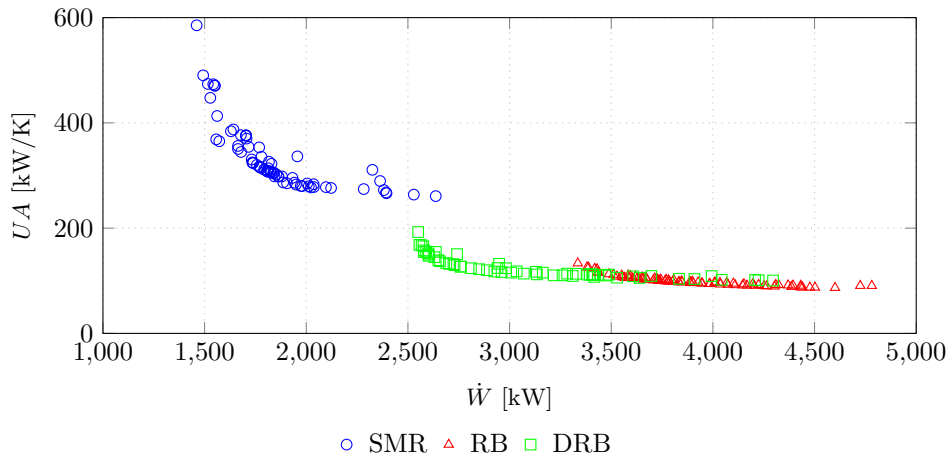


Figure 10: Trade-off between the power consumption and heat network conductance (UA) for the three small-scale systems, for the Danish composition

For the SMR process, the main differences between the solutions characterised by a minimum power consumption or by a minimum overall conductance differ mainly in the operating pressures (30 bar against 37 bar for the high-level, and 4 against 1 for the low-level) and contents of medium-weight hydrocarbons (5% against 7% for propane, and 4% against 2% for *n*-butane). The contents of methane, ethane and nitrogen vary in a range of ($\pm 1\%$). When sorted by family of chemical compounds (light- and medium-weight alkanes, heavy hydrocarbons, nitrogen), no significant differences could be found between the several

mixture compositions.

For the single-stage expansion process, the main differences lie in the refrigerant flowrate, the low-pressure level and the precooling temperature. Higher flowrates (0.3 kmol/kg_{LNG} against 0.2 kmol/kg_{LNG}), combined with a lower precooling temperature (-48 °C against -12 °C) and a moderate low-pressure level (around 7 bar against 1 bar) are associated with smaller power consumption. This illustrates that: (i) higher flowrates are required for lower expansion ratios to ensure enough heat transfer driving force in the multi-stream heat exchanger, (ii) lower precooling temperatures (state 2) result in smaller net power consumption, at the expense of larger heat transfer areas. Similar flowrates (0.22 kmol/kg_{LNG}) and high-pressure levels are found for all these configurations, and the only difference is found for the low-pressure level. Compared to the conventional reverse Brayton cycle, the dual process is systematically featured by smaller flowrates of refrigerant. The use of methane instead of nitrogen is beneficial, as it results either in a reduction of the specific power consumption by up to 400 kJ/kg_{LNG} or in a smaller conductance of the entire heat exchanger network by up to 50 kW/K.

Differences in the feed composition result in different net power consumptions (Figure 11) in the optimum solutions. The smallest values are found in the Spanish case, whilst the highest correspond to the Italian one. These figures are expected, as methane has a greater thermal capacity than heavy-weight hydrocarbons. The thresholds in power consumption are similar for all gas compositions if a reverse Brayton process is used, while they differ by (± 250 -400 kW) for the two other processes. This illustrates that the SMR and DRB layouts have a bigger room for optimisation due to the higher number of degrees of freedom (mixture composition and pressure levels).

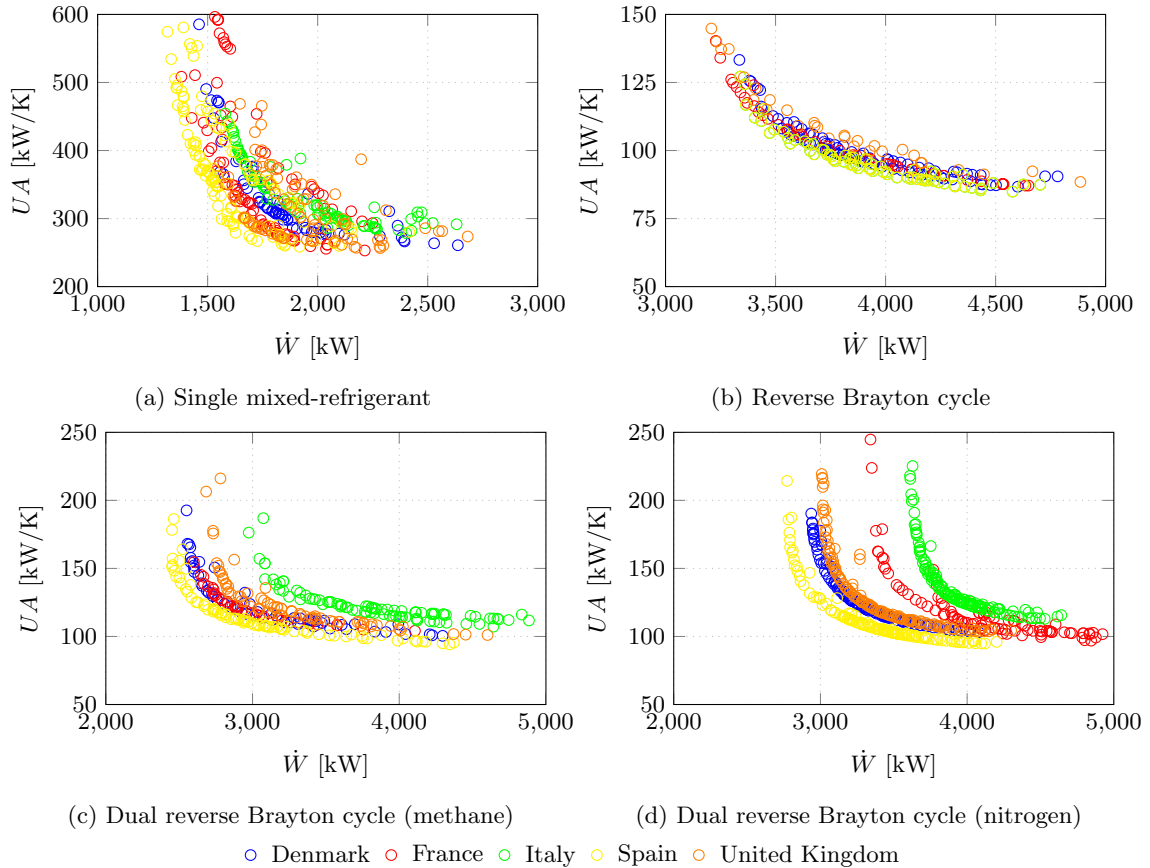


Figure 11: Trade-off between the power consumption and heat network conductance (UA) for five feed compositions

4. Discussion

4.1. Performance of the single mixed-refrigerant process (SMR)

The results presented in the existing literature cannot be compared directly with the present findings. Different values of the temperature and pressure levels, allowable temperature differences and component efficiencies are used. The specific power consumption ranges between 1300 and 1500 kJ/kg_{LNG} in the present work, depending on the gas feed composition. It falls in the range of 1100 [10] to 1600 kJ/kg_{LNG} [17] displayed in the literature, after optimisation of the operating conditions.

The distribution of the exergy destruction among the SMR components is similar to the findings presented in research works as well. Remelje and Hoadley[6] show the high share of exergy destruction within the cryogenic heat exchanger (46%), followed by the aftercoolers (30%) and the compressors (21%). Shirazi and Mowla[10] pinpoint the aftercoolers as the most exergy-destroying components (33%), followed by the compressors (31%) and the LNG heat exchanger (27%). Similar figures are found by Xu et al.[11], although they are 5 %-points higher for the compressors and 6 %-points lower for the aftercoolers. Morosuk et al.[12] emphasise that the cryogenic heat exchanger is the component responsible for most irreversibilities (45.1%). The numerical differences between those results are imputable to the simulation assumptions.

Chang[51] analyse an optimised and ideal SMR process (i.e. with a minimum temperature difference of 0 K in the heat exchangers and 100% efficiency of the compressors) for a similar gas composition and state that the maximum figure of merit for such cycles is about 57%. The simulation of the proposed optimal solutions, assuming an efficiency of 100% of the compressors, gives a figure of merit of about 51%. This suggests that a further optimisation of the mixture composition assuming a minimum temperature approach of 0 K would return results in the same magnitude. The conclusions of the present work on the effects of varying the refrigerant composition (contents of methane, nitrogen, butanes and pentanes) are also supported by the works of Austbø[52], where it is claimed that the system performance, in the optimal cases, is mostly affected by the mixture content of light ends.

4.2. Performance of the expander-based processes (single and dual)

As suggested in the literature, expander-based processes present higher power consumption than mixed-refrigerant processes, and this finding is supported in the present work in the case of small-scale applications. In consequence, these systems also present a lower coefficient of performance and second-law efficiency. A direct comparison with the work of Remelje and Hoadley[6] is not possible since they do not analyse a simple or dual reverse Brayton cycle, but most advanced configurations. However, they also conclude that mixed-refrigerant processes are generally more efficient.

Chang[51] also investigate the performance of an optimised reverse Brayton process and find an ideal limit (figure of merit) of 60%. These figures are, however, difficult to compare directly because of the dissimilarities of the assumptions. The simulation of the present solution, assuming an efficiency of 100% of the turbomachines, gives a figure of merit of about 49%. This indicates that further optimisation of the pressure levels, to achieve a minimum temperature approach of 0 K in all heat exchangers, would give results in the same magnitude.

4.3. Overall conductance of the heat exchanger network

No numerical figures have been found in the literature about the UA values of the heat exchanger network for mixed-refrigerant and expander-based processes. Most remarks are qualitative, stating for example that the heat exchangers for mixed-refrigerant processes are bigger as a result of a closer temperature match over the entire liquefaction process, compared to expander-based ones. The overall conductance of the heat exchanger network can be used as an indicator for the system cost only when comparing different solutions where the heat transfer coefficients (U-value) is similar. This is the case when comparing different solutions of a given configuration or for similar types (e.g. the two expander-based systems). The refrigerant in the expander based systems is in gaseous state in all heat exchangers, which usually leads to relatively low U-values. In the mixed-refrigerant system, the refrigerant is changing phase in both heat exchangers. Heat transfer coefficients during phase change can easily be up to an order of magnitude higher during phase

change than for gas flows, which means that the required area for transferring the same amount of heat is significantly smaller. The overall network conductance can therefore not directly be used to compare system costs of the different systems. A more thorough analysis of the needed heat transfer areas should be carried out to give a clearer comparison of the heat exchanger sizes of the two different system types.

4.4. Future work

Future work within this topic may include performance comparisons for a higher number of mixed-refrigerant and expander-based processes, from nitrogen to methane reverse Brayton cycles (open and closed) to dual mixture-refrigerant systems. Moreover, a more thorough comparison of the thermodynamic models used for simulating such systems would be beneficial. It is indeed shown that the use of cubic or fundamental equations of state potentially gives noticeable discrepancies. This may be important to consider in the light of the small temperature approaches that are found in the cryogenic heat exchangers.

5. Conclusion

This paper presents a comparison of three small-scale processes for the liquefaction of natural gas. The first one is the single mixed-refrigerant process, which builds on the use of up to eight chemical compounds, whilst the two latter are the single and dual expander-based cycles, using nitrogen or methane as pure refrigerant. Under the given set of assumptions and considering natural gas from the Danish grid after removal of carbon dioxide and heavy hydrocarbons, the SMR process is characterised by a power consumption of less than 1800 kJ/kg_{LNG}, while this reaches more than 2600 kJ/kg_{LNG} for the N₂ or CH₄ single-stage expansion processes. These numbers were estimated by conducting a single-objective optimisation. A further comparison was then performed by a multi-objective optimisation, considering the minimisation of the thermal conductance (UA), which illustrates the system size, and the net power consumption. Such an approach is beneficial for assessing technical, practical and possibly economic trade-offs. The extension of this work to a bigger group of system configurations can set up a basis for comparing more consistently gas liquefaction processes.

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