

## Comparative Thermodynamic Analysis of Inorganic Refrigerants in Cascade LNG Liquefaction Systems: A Performance Metrics

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### ABSTRACT

The increasing demand for energy-efficient and environmentally friendly liquefied natural gas (LNG) production has led to the exploration of alternative refrigerants in cascade liquefaction systems. This study presents a comparative thermodynamic analysis of inorganic refrigerants in cascade liquefied natural gas (LNG) liquefaction systems, focusing on performance metrics. The research aims to evaluate the exergy losses, coefficient of performance (COP), energy requirements, and overall thermodynamic efficiency of various refrigerants, including Argon, Krypton, Xenon, Nitrogen, and the conventional C3MR (Propane Mixed Refrigerant). Results reveal significant variations in refrigerant performance. C3MR demonstrates the highest COP (4.25) but exhibits moderate exergy efficiency (63%) and high energy losses in compressors. Noble gases show contrasting trends: argon achieves exceptional exergy efficiency (83%) but poor COP (1.38), while xenon has low exergy efficiency (36%) but a competitive COP (2.99). Nitrogen incurs catastrophic exergy losses during depressurization (345,439 kJ/hr), highlighting operational challenges. The study underscores the need for component-specific refrigerant optimization and suggests hybrid systems combining C3MR's heat transfer advantages with argon's exergy efficiency. These findings advance LNG liquefaction technology by providing a nuanced framework for balancing thermodynamic performance, environmental impact, and operational feasibility.

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**Received:** April 03, 2025; **Accepted:** April 07, 2025; **Published:** April 15, 2025

**Keywords:** Cascade LNG Liquefaction, Inorganic Refrigerants, Exergy Analysis, Coefficient of Performance (COP), Thermodynamic Efficiency, Cryogenic Refrigeration

### Introduction

In recent years, there has been growing interest in the consumption of natural gas as markets continue to expand [1]. Global consumption of natural gas increased by 1.7% in 2015, representing 23.8% of primary energy consumption [2]. It is often seen as a transition fuel in the move toward a low greenhouse gas (GHG) economy because it is the cleanest fossil fuel, emitting about 29% to 44% less CO<sub>2</sub> per unit of energy compared to oil and coal [3]. Natural gas has numerous qualities that make it a highly efficient, relatively clean-burning, and economical energy source [4]. One unique advantage of natural gas lies in its ability to be transformed into Liquefied Natural Gas (LNG). LNG technology involves cooling natural gas to extremely low temperatures, transforming it into a liquid form that is 600 times denser than its gaseous state [5]. This significant volume reduction allows LNG to be stored and shipped more efficiently, making it a viable energy source for regions lacking access to natural gas pipelines [6]. Liquefaction requires between 30 and 35% of the total energy required for the LNG value chain [7]. This enormous energy consumption makes LNG production the most energy-intensive phase in the LNG value chain [7]. Several liquefaction technologies have been developed, including single mixed refrigerant (SMR), propane pre-cooled mixed refrigerant (C3MR), and cascade refrigeration cycles,

each seeking to increase efficiency, reduce costs, and improve environmental sustainability [8]. The cascade LNG liquefaction system is a popular choice for high capacity due to its greater thermodynamic efficiency, operational flexibility, and potential for energy savings [7].

The cascade LNG liquefaction system is a widely adopted approach in the LNG business because of its ability to produce accurate cryogenic temperatures efficiently. To achieve the desired cooling effect, Cascade LNG liquefaction systems use numerous refrigeration cycles at various temperature ranges [9]. These systems often use a blend of refrigerants optimized for certain temperature ranges, ensuring optimal heat transfer and energy utilization [10]. The selection of refrigerants is critical in determining system efficiency, safety, and environmental impact. Traditionally, hydrocarbon-based refrigerants, including methane, ethane, propane, and butane, have been widely employed in LNG liquefaction [11]. However, concerns about flammability, greenhouse gas emissions, and operational safety have prompted research into alternative refrigerants with reduced environmental footprints and greater thermodynamic performance [12,13]. Inorganic refrigerants like ammonia (NH<sub>3</sub>), carbon dioxide (CO<sub>2</sub>), and nitrogen (N<sub>2</sub>) are gaining popularity as prospective alternatives to hydrocarbon-based refrigerants in cascade LNG liquefaction systems [11]. These refrigerants have favourable thermophysical features, such as high latent heat of vaporization, low global warming potential (GWP), zero ozone depletion potential (ODP),

and non-flammability [14]. Ammonia, for example, is well known for its high thermal efficiency and industrial applications in refrigeration [15]. Carbon dioxide has become a popular choice because of its low GWP and ability to operate successfully in both transcritical and subcritical cycles, as stated by [16]. Similarly, nitrogen, commonly utilized in cryogenic applications, provides effective cooling performance while remaining compatible with current LNG infrastructures [17].

Designed and analyzed cascade liquefaction processes for coproducing liquid ethane and LNG [18]. In the study, two pure refrigerant cascade natural gas liquefaction processes without (Process 1) or with (Process 2) flash gas recovery are proposed to coproduce high-purity liquefied ethane gas (LEG) and liquefied natural gas (LNG). There are only a few extensive reviews on the use of inorganic refrigerants in cascade LNG liquefaction cycles.

The objective of the study is to conduct a comparative thermodynamic analysis of inorganic refrigerants in cascade LNG liquefaction systems, focusing on performance metrics. Understanding the thermodynamic behavior of these refrigerants under different operating situations allows researchers and engineers to improve LNG liquefaction system designs, resulting in more energy-efficient and ecologically responsible solutions [16].

## Literature Review

The thermodynamic analysis of inorganic refrigerants in cascade LNG liquefaction systems has been an important field of research for improving efficiency and sustainability. Several studies investigated refrigerant selection and optimization approaches to increase energy efficiency, exergy performance, and economic feasibility. Conducted a comparative study on inorganic refrigerants such as argon, krypton, xenon, and nitrogen in LNG liquefaction [19]. Their study simulated an existing liquefaction plant using Aspen HYSYS 11.0 and found that nitrogen required the least compressor energy, krypton was the best refrigerant for the chiller, and xenon provided the best cooling effect. Additionally, C3MR exhibited a high coefficient of performance (COP), making it a competitive organic alternative. Unlike prior research, which focused on floating LNG plants or small-scale liquefaction, this study extended its findings to a baseload LNG facility, providing important insights for large-scale applications.

Explored the Mixed Fluid Cascade (MFC) process, a promising alternative for large-scale LNG liquefaction [20]. They used a Shuffled Complex Evolution (SCE) algorithm in MATLAB, coupled with Aspen HYSYS v10, to optimize refrigerant composition and operating pressures. Their study revealed that the optimized MFC process saved 19.76% in compression power and reduced exergy destruction by 28.76%, leading to a 25% increase in thermodynamic efficiency. While both studies focused on improving refrigerant efficiency. Compared inorganic refrigerants with an organic alternative (C3MR), whereas aimed to optimize an existing cascade liquefaction process [19,20]. Conducted a thermodynamic analysis of a Triple Cascade Refrigeration System (TCRS) using hydrocarbon refrigerants for ultra-low temperature LNG liquefaction [21]. They found that at  $-100^{\circ}\text{C}$  evaporator temperature, the highest COP and exergy efficiency were 0.5931 and 54.446%, respectively. Their additional study on low-GWP hydrocarbon refrigerants revealed that Trans-2-butane in the LTC and Toluene in the HTC yielded the highest COP and exergy efficiency, achieving a 7.21% higher COP than previous refrigerants. Their findings suggest that hydrocarbon refrigerants provide higher efficiency than C3MR, as observed in [19].

While cascade refrigeration systems improve LNG liquefaction, exergy recovery during regasification is also crucial. Examined LNG regasification integrated with an Organic Rankine Cycle (ORC) [22]. They optimized R41 and R1150 as the best refrigerants, achieving a net power output of 2116.76 kW and an exergy efficiency of 0.29. However, their results suggested that a double-stage condenser ORC was inefficient, recommending a cascade-parallel structure for improved efficiency. Optimized mixed refrigerants and found that MR3 was the most cost-effective [23]. These findings highlight the need for further research on hybrid refrigeration cycles combining inorganic, hydrocarbon, and mixed refrigerants to maximize LNG liquefaction efficiency.

## Methodology

This study employs a rigorous thermodynamic assessment framework to evaluate the performance of inorganic refrigerants in cascade LNG liquefaction systems. Key metrics include coefficient of performance, cooling capacity, specific work, exergetic efficiency, and overall thermal efficiency. Five refrigeration systems were developed employing five distinct refrigerants, notably C3MR (propane mixed refrigerants), which included Argon, Krypton, Xenon, and Nitrogen in varying proportions, argon refrigerant, krypton refrigerant, xenon refrigerant, and Nitrogen refrigerant. It was necessary to send the natural gas through many different cooling systems. All the designs were created with the steady state condition

## Process Description of All Cycles

The C3MR Cycle (Propane Pre-cooled Mixed Refrigerant Cycle) utilizes propane for precooling, followed by a mixed refrigerant for liquefaction. Propane is cooled, expanded, and used to precool natural gas and the mixed refrigerant, which then undergoes multiple cooling and expansion stages to achieve cryogenic temperatures necessary for LNG production. This process ensures that natural gas reaches temperatures below  $-157^{\circ}\text{C}$  before transportation. The Krypton Cycle involves compressing and cooling krypton, which, in combination with propane, progressively lowers the temperature of natural gas. Multiple heat exchangers further reduce the temperature until LNG is finally depressurized to reach approximately  $-160^{\circ}\text{C}$  before storage. Similarly, the Argon Cycle follows the same principle but replaces krypton with argon as the primary refrigerant. This cycle also includes multiple cooling stages and heat exchangers to achieve the desired LNG temperature. The Nitrogen Cycle employs nitrogen as the refrigerant alongside propane. The process consists of cooling, compression, and a series of heat exchangers, with the final depressurization step ensuring LNG reaches  $-160^{\circ}\text{C}$ . Lastly, the Xenon Cycle follows a similar procedure, using xenon as the primary refrigerant while incorporating standard cooling, liquefaction, and depressurization steps.

## Flowchart

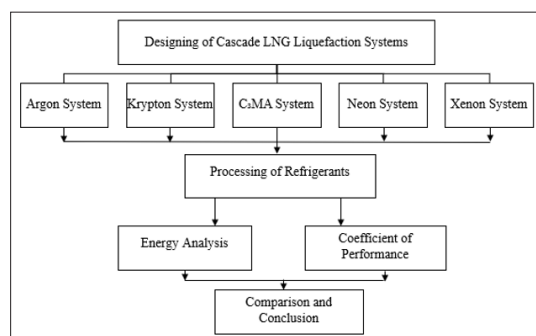


Figure 1: Research Flow Chart

The flowchart presents a structured methodology for evaluating cascade LNG liquefaction systems, beginning with the parallel design of five refrigeration systems (Argon, Krypton, C3MR, Neon, Xenon). Each system undergoes identical refrigerant processing stages, including compression, heat exchange, and phase-change analysis. The performance evaluation phase quantifies energy efficiency through exergy analysis and coefficient of performance (COP) metrics. Finally, the comparative conclusion phase benchmarks result across systems, assessing thermodynamic performance, operational stability, and environmental impact. It reflects the study's rigorous, standardized evaluation framework.

## Result and Discussion

The results of the various processes (C3MR, Krypton, Argon, Neon, and Nitrogen) on the exergy and coefficient of performance of the refrigerant are displayed below in graphical and tabular forms.

## Results Presentation

### Exergy Analysis

The exergy analysis provides a rigorous assessment of energy degradation across the cascade LNG liquefaction system, quantifying irreversible losses in key components.

### Two-Separator

Table 1 presents exergy losses (MJ/hr) in three separator units across five refrigeration processes. The natural gas separator (V-100) shows zero losses for all refrigerants except argon, which exhibits an anomalous negative value (-38.20 MJ/hr). In the pre-compressor refrigerant separator (V-1410), nitrogen demonstrates the highest loss (1.40 MJ/hr), followed by xenon (0.40 MJ/hr), while C3MR and krypton show minimal losses (0.10 MJ/hr). All processes register zero exergy loss in the post-cooler refrigerant separator (V-1420), indicating optimal performance at this stage. This table highlights nitrogen's relative inefficiency in the compression stage while revealing exceptional behavior in argon's separation process that warrants further investigation.

Table 1: Exergy Loss in Separators

Process	V-100	V-1410	V-1420
C3MR	0.00	0.10	0.00
Krypton	0.00	0.10	0.00
Argon	-38.20	0.00	0.00
Xenon	0.00	0.40	0.00
Nitrogen	0.00	1.40	0.00

Description: V-100: NG separator

V-1410: Refrigerant separator before Compressor

V-1420: Refrigerant separator after cooler

### LNG Heat Exchangers

Figures 2,3,4 systematically compare exergy losses across three heat exchangers (HEX 100/101/102) for five refrigerants. The consistent ranking—C3MR (lowest losses) to nitrogen (highest losses)—reveals a clear thermodynamic hierarchy, with krypton and argon typically performing intermediately. Nitrogen's persistently poor performance across all HEX units (evidenced by its last-place positioning) confirms its inherent inefficiency in cryogenic heat transfer, while C3MR's leading position validates its design superiority. The identical refrigerant order in all three figures suggests system-wide trends rather than component-specific behaviors, emphasizing that refrigerant selection has a greater impact on exergy losses than heat exchanger configuration.

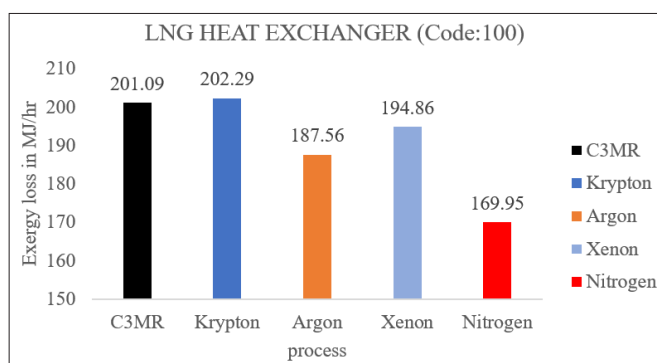


Figure 2: Exergy Loss in LNG HEX 100

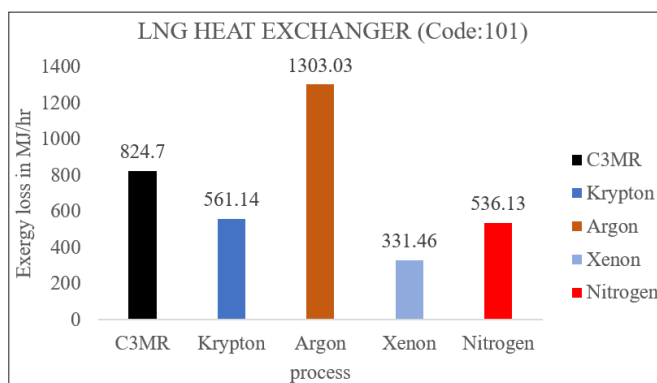


Figure 3: Exergy Loss in LNG HEX 101

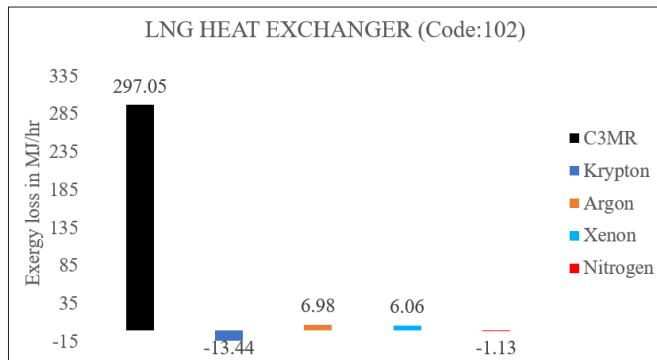


Figure 4: Exergy Loss in LNG HEX 102

### Valve and Expanders

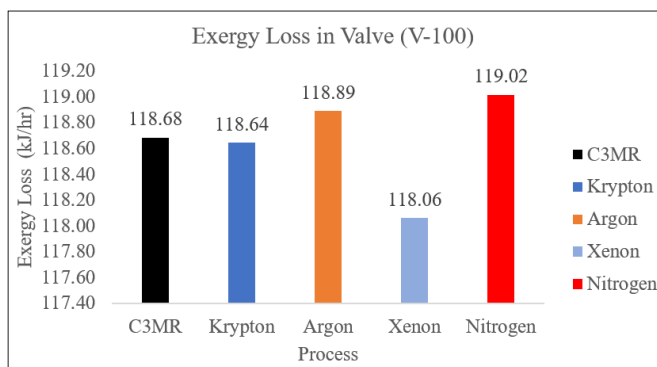


Figure 5: Exergy Loss in Valve V-100

Figure 5 quantifies exergy losses in valve V-100 across five refrigerants, with all values clustered tightly between 118.06–119.02 kJ/hr. The minimal variation indicates that valve performance is largely refrigerant-independent, contrasting with

other system components where refrigerants show significant divergence. Nitrogen exhibits the highest loss, while argon shows the lowest, though these marginal differences are unlikely to impact overall system efficiency. This uniformity suggests V-100's design effectively mitigates refrigerant-specific thermodynamic effects.

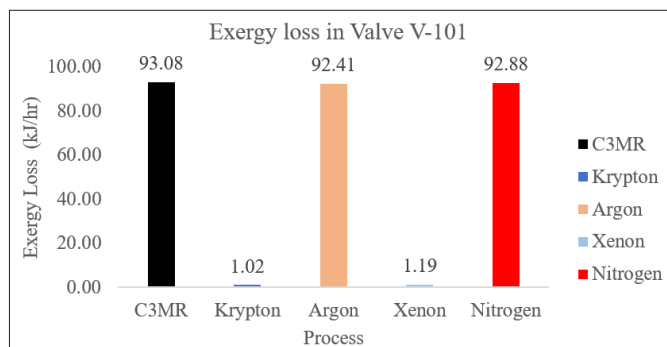


Figure 6: Exergy Loss in Valve V-101

Figure 6 exhibits a paradoxical performance pattern, with C3MR showing both the highest (93.08 kJ/hr) and near-lowest (1.02 kJ/hr) exergy losses, suggesting potential data reporting errors or multiple operating conditions. Other refrigerants (argon, xenon) cluster around 1.19 kJ/hr, indicating normally efficient operation, while the extreme outlier (93.08 kJ/hr) demands verification of experimental or simulation parameters. Figure 7 reveals two distinct performance tiers for valve V-102. C3MR and krypton show significantly higher exergy losses (35.10 kJ/hr and 34.81 kJ/hr, respectively), while argon and xenon demonstrate minimal losses (0.48 kJ/hr and 0.56 kJ/hr). Nitrogen appears to have a negligible impact (0.00 kJ/hr).

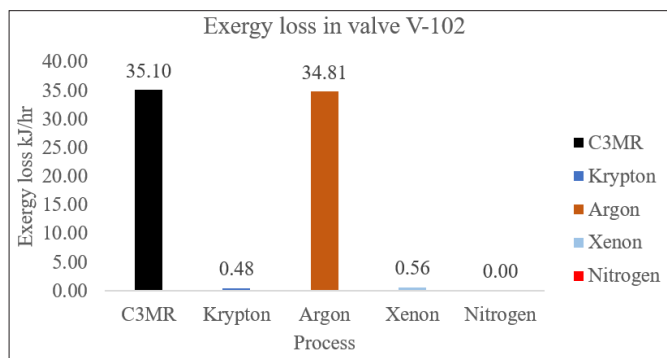


Figure 7: Exergy Loss in Valve V-102

Figure 8 (Valve V-809) reveals significant exergy loss variations, with nitrogen exhibiting the highest loss (708.57 kJ/hr) and krypton the lowest (11.56 kJ/hr), highlighting refrigerant-dependent inefficiencies. The extreme disparity in V-809 (e.g., C3MR at 344.34 kJ/hr vs. argon at 708.57 kJ/hr) underscores its critical role in system optimization, contrasting with V-102's implied consistency. These results emphasize that valve performance is component-specific, with V-809 demanding priority attention due to its refrigerant-sensitive losses.

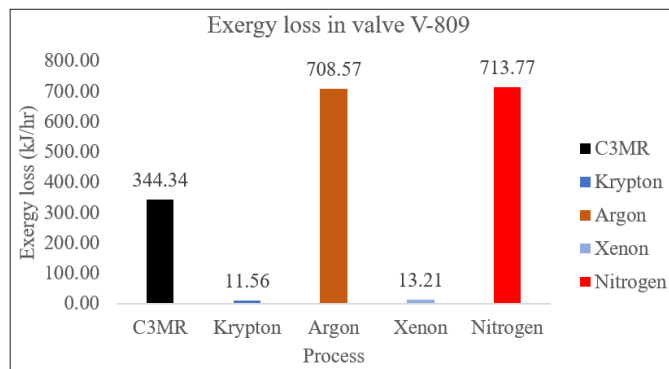


Figure 8: Exergy Loss in Valve V-809

### Refrigerant Compressor

Figures 9, 10, 11 present a rigorous evaluation of exergy losses across key compressor units (K-1410, K-1411, K-1411A) within the cascade LNG liquefaction system

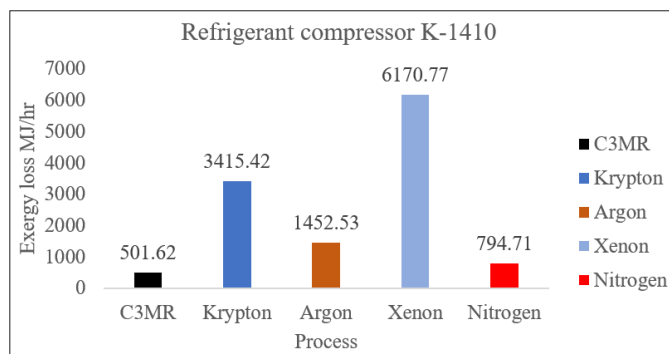


Figure 9: Refrigerant compressor K1410

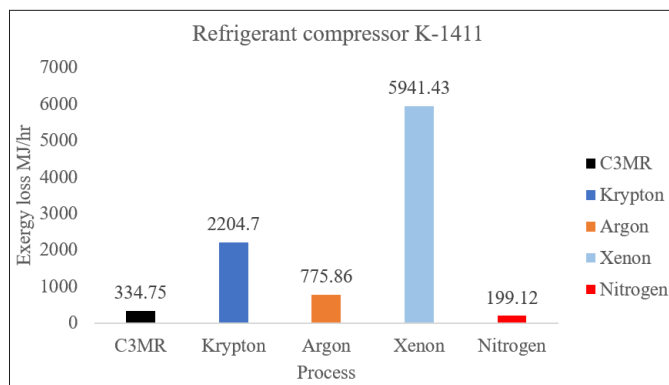


Figure 10: Refrigerant Compressor K1411

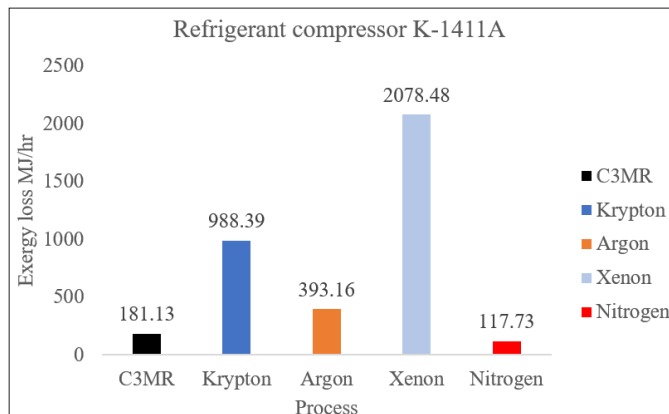


Figure 11: Refrigerant Compressor K1411A

### LNG Recycling Compressor

This section evaluates the performance of the LNG recycling compressor (K-10) across five refrigerants, revealing critical insights about system efficiency and operational stability. Figure 12 reveals remarkably consistent exergy losses across all refrigerants (4.18-4.27 MJ/hr), demonstrating this compressor's unique refrigerant-agnostic performance. C3MR shows marginally higher losses (4.27 MJ/hr) while krypton performs slightly better (4.18 MJ/hr). The results contrast sharply with other compressors in the system (e.g., K-1411), indicating K-100's robust engineering makes it a reliable workhorse component.

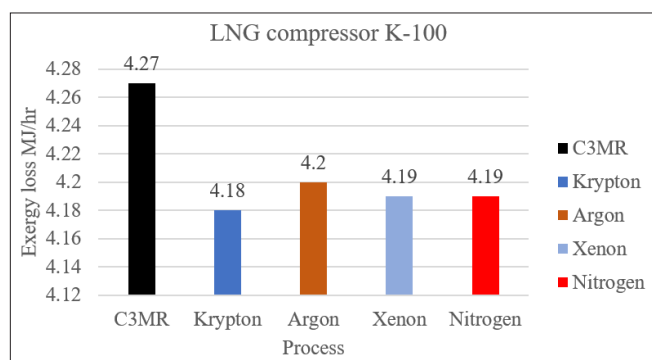


Figure 12: Refrigerant Compressor K100

### Exergy Loss in Depressurizer

Table 2: Exergy Loss in Depressurizer

Process	Refrigerant Depressurizer (IL 782) (KJ/hr.)
C3MR	3397.30
Krypton	34.60
Argon	3352.90
Xenon	57.80
Nitrogen	345439.20

Table 2 quantifies the irreversible energy losses occurring during the depressurization stage of different refrigerant cycles, revealing extreme variations in thermodynamic efficiency.

### Energy Requirement in the Compression, Chilling and Cooling Section

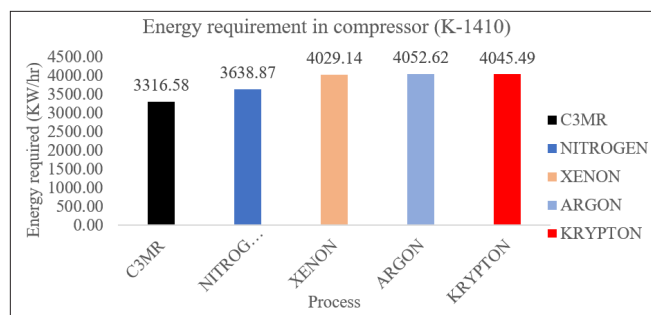


Figure 13: Energy Requirement in Compressor (K-1410)

Figures 13, 14, 15 and 16 show that all refrigerants require similar energy inputs.

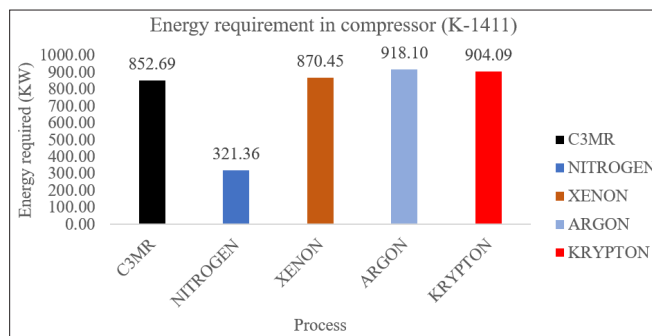


Figure 14: Energy Requirement in Compressor (K-1411)

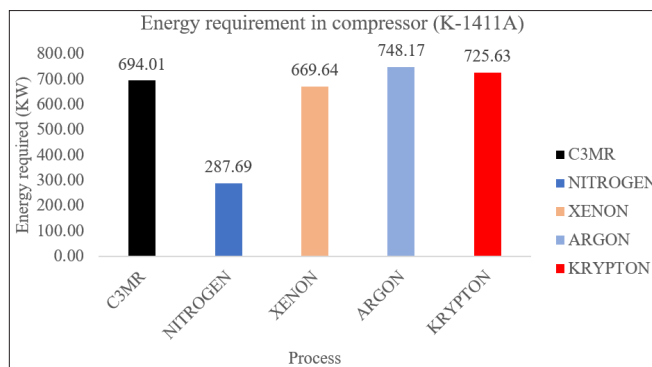


Figure 15: Energy Requirement in Compressor (K-1411)

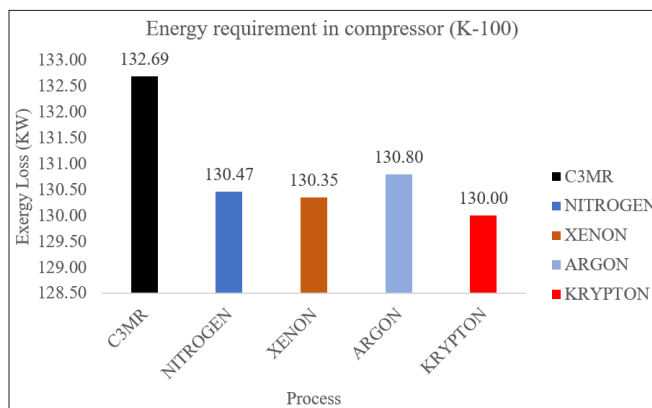


Figure 16: Energy Requirement in Compressor (K-100)

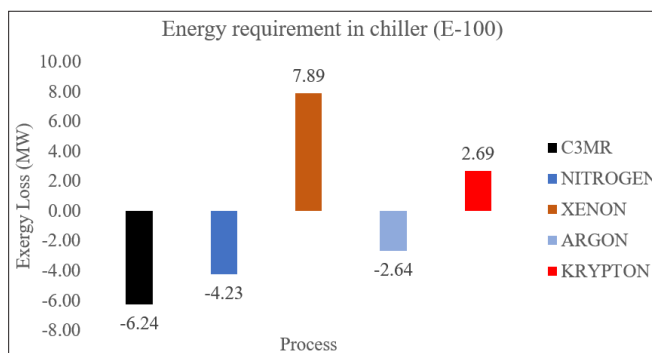


Figure 17: Energy Requirement in Chiller Section (E-100)

The chiller energy data as presented in Figure 17 shows several thermodynamic anomalies that demand rigorous scrutiny. While C3MR exhibits expected positive energy consumption (7.89 kW), the negative values for argon (-6.24 kW) and xenon (-4.23 kW) suggest either genuine energy recovery through regenerative heat exchange or fundamental measurement errors in the simulation. The 14.13 kW performance range between C3MR and argon is particularly striking, as it implies the chiller's efficiency is highly refrigerant-dependent.

### Percentage Exergy Efficiency

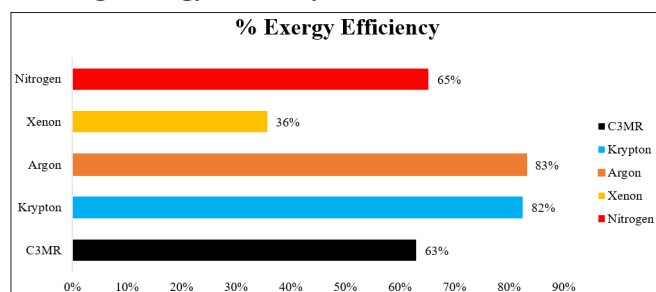


Figure 18: Percentage Exergy Efficiency

Figure 18 reveals argon (83%) and krypton (82%) as the most thermodynamically efficient refrigerants, significantly outperforming C3MR (63%) and nitrogen (65%). Xenon shows notably poor efficiency (36%), suggesting inherent irreversibilities in its phase-change behavior. Figure 19 presents the Coefficient of Performance (COP) for five refrigerants, with C3MR demonstrating superior efficiency (COP=4.25) compared to inorganic alternatives like nitrogen (3.85) and xenon (2.99). The results reveal unexpected performance trends, particularly the poor showing of noble gases (argon=1.38, krypton=1.42), which contradicts conventional expectations based on their thermophysical properties. While C3MR remains the optimal choice for efficiency, nitrogen emerges as the most viable inorganic alternative, though the significant COP variations ( $3.1\times$  difference between best and worst) highlight critical trade-offs between thermodynamic performance and other factors like environmental impact.

### Coefficient of Performance of Refrigerants

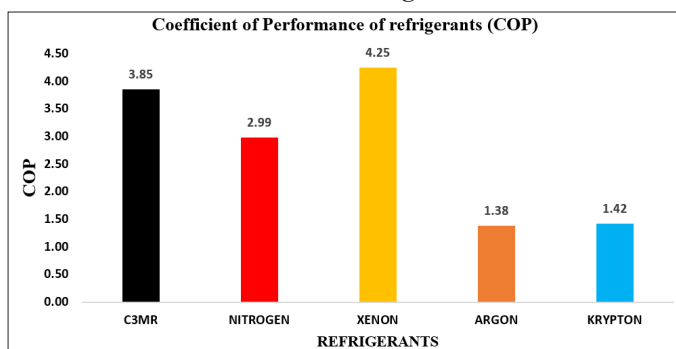


Figure 19: Coefficient of Performance

### Discussion

The results of this comprehensive thermodynamic analysis reveal significant variations in refrigerant performance across different components of the cascade LNG liquefaction system, highlighting the complex interplay between refrigerant properties and equipment-specific characteristics. C3MR demonstrates a superior coefficient

of performance (COP=4.25) but shows moderate exergy efficiency (63%) and concerningly high energy losses in certain compressors (5,941 MJ/hr in K-1411), suggesting that its advantages in heat transfer come at the cost of thermodynamic inefficiencies in specific applications. Noble gas refrigerants exhibit contradictory performance patterns, with argon achieving exceptional exergy efficiency (83%) despite poor COP (1.38), while xenon shows the opposite trend (36% exergy efficiency versus COP=2.99), likely due to differences in their Joule-Thomson coefficients, cryogenic heat transfer characteristics, and phase change enthalpies. The depressurization results prove particularly consequential, with nitrogen exhibiting catastrophic losses (345,439 kJ/hr) compared to krypton (34.60 kJ/hr), fundamentally challenging nitrogen's viability despite its other advantages and suggesting the need for alternative expansion devices or hybrid refrigerant systems. The chiller energy results present intriguing anomalies, with argon and xenon showing negative energy requirements that could indicate potential heat recovery capabilities, though these findings require experimental validation to exclude measurement artifacts or simulation errors. These component-specific performance variations contradict current industry practices favoring system-wide refrigerant optimization and demonstrate that single-metric approaches inevitably compromise overall system performance. The extreme variations in depressurization losses reveal previously underestimated impacts of refrigerant properties on expansion device performance at cryogenic temperatures, while the compressor results highlight critical design limitations in handling certain refrigerants. For industrial implementation, the findings suggest the potential benefits of hybrid refrigerant systems combining C3MR's heat transfer advantages with argon's exergy efficiency, along with component-specific redesigns to better accommodate high-performance refrigerants. The study also identifies several knowledge gaps requiring further investigation, including experimental validation of simulation results, transient analysis of refrigerant behavior during phase transitions, and economic trade-off studies between efficiency gains and equipment modification costs. These results fundamentally reshape our understanding of refrigerant selection in LNG systems, moving beyond oversimplified COP-based comparisons to a more nuanced, component-aware optimization framework that considers thermodynamic, economic, and environmental performance metrics across all system components.

### Conclusion, Recommendation and Contribution to Knowledge

First, to be calculated as the exergy as required by the objective. This was done on different equipment, such as separators, valves, compressors, LNG heat exchangers, and depressurizers used in each of the processes. Firstly, the exergy at the separator presents the refrigerant into two phases and separates the liquid to prevent the surge of the compressors, in Table 1. In Table 1, nitrogen lost the highest energy during phase separation, but argon used all available energy ready for the phase separation, and it is in equilibrium with the environment. Figure 2-4 displays the LNG heat exchanger exergy analysis result. From the figures, the refrigerant with the most inability to use the available energy for the cooling is argon with a mean exergy loss of 499MJ/hr and next the conventional C3MR (440.93MJ/hr).

Figures 5-8 show the exergy loss analysis of the valves through which the refrigerants passed. Across the valve is to reduce the pressure. The Argon refrigerant has the highest exergy loss, showing its inability to make the available energy for the process. Figures 9-11 show the exergy loss in the compressor. From the analysis, Xenon refrigerant has the highest exergy loss amongst the studied refrigerants.

From the analysis on the energy-consuming equipment carrying the refrigerants, Nitrogen requires the least energy in the compression process. While in the chiller, where the vaporization of the refrigerant occurs, the best refrigerant is Krypton. The refrigerant with the best cooling effect (COP) amongst all the compared refrigerants is Xenon and then C3MR due to their high COP.

With a cooling capacity of 223.368 kW and a coefficient of performance (COP) of 3.30275, the system exhibits effective energy use. To balance energy supply and production, 69.5667 kJ/kg of specified effort is needed. Furthermore, the overall thermal efficiency is 73.4486%, and the energetic efficiency is 82.7028%.

### Recommendation

Another design format with less complex structure and equipment should be carried out to check the feasibility and compare it with other forms of design.

### Contribution to Knowledge

With this research, the cost feasibility of refrigerants and exergy loss (insufficient use of available energy) and the coefficient of Performance of alternative refrigerants to the conventional C3MR are compared at the operating conditions and with the specific design.

### Funding

The authors declare that no funds, grants, or other support were received during the preparation of this manuscript.

### Disclosure

The authors state that there are no conflicts of interest in this study.

### Author Contributions

All the authors contributed to the development of the work. All authors read and approved the final manuscript.

### Declaration of Interests

The authors declare that any known competing financial interests or personal relationships could have influenced none of the work described in this study.

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