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Optimization of Inorganic Refrigerants in Cascade LNG Liquefaction Systems: A Response Surface Methodology Approach for Enhanced Energy Efficiency and Sustainability

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ABSTRACT

The global energy transition has intensified the demand for sustainable liquefied natural gas (LNG) production, necessitating advanced refrigeration systems with minimal environmental impact. This study presents a comprehensive thermodynamic optimization of inorganic refrigerants (xenon, argon, krypton, nitrogen) in cascade LNG liquefaction cycles using response surface methodology (RSM). Through Aspen HYSYS modeling and I-optimal design experiments, key performance metrics—coefficient of performance (COP), cooling capacity, specific work, exergetic efficiency, and overall thermal efficiency—were evaluated across varying evaporating temperatures (−50°C to −30°C) and pressure regimes (10–30 bar). Results demonstrate that xenon achieves superior performance, with a COP of 3.6 and exergetic efficiency of 89% at optimal conditions (−44.5°C, 10.78 bar), outperforming conventional mixed refrigerants (C3MR) by 16.6% in specific energy consumption. Exergy analysis reveals that xenon minimizes irreversibility in compression and heat exchange stages, reducing exergy destruction by 21% compared to nitrogen. However, economic constraints due to xenon's high cost highlight the trade-offs between efficiency and scalability. This work advances sustainable LNG production by identifying energy-efficient refrigerant alternatives while providing a robust RSM framework for industrial process optimization.

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Introduction

The rapid expansion of liquefied natural gas (LNG) as a key component of the global energy transition has intensified research into more efficient and sustainable liquefaction processes [1-3]. LNG liquefaction is an energy-intensive process that requires natural gas to be chilled to around -162°C for storage and transit [4,5]. Among the various liquefaction technologies, the cascade refrigeration system stands out due to its ability to optimize energy use by employing multiple refrigeration cycles, each operating at distinct temperature levels [2]. The choice of refrigerants plays a critical role in determining the efficiency of these systems, directly influencing specific power consumption, exergy efficiency, and overall process optimization [6-8]. Traditionally, hydrocarbon-based refrigerants such as methane, ethane, and propane have dominated LNG liquefaction due to their well-established thermodynamic properties. However, growing environmental concerns, safety issues, and the need for greater efficiency have sparked interest in alternative refrigerants, particularly inorganic working fluids [9,10]. Despite their potential benefits, inorganic refrigerants are relatively unexplored in the context of optimised LNG cascade systems, demanding a comprehensive assessment of their thermodynamic properties.

Optimization in LNG liquefaction is centered on reducing energy consumption while maximizing thermodynamic efficiency. This requires selecting refrigerants that exhibit superior heat transfer characteristics, lower exergy destruction, and reduced compression work, ultimately leading to lower operating costs and improved sustainability [9,11]. Inorganic refrigerants such as ammonia (NH_3), carbon dioxide (CO_2), sulfur dioxide (SO_2), and water (H_2O) offer promising alternatives to traditional hydrocarbons as they possess favorable thermodynamic properties and, in many cases, lower environmental impact [12,13]. Ammonia, for instance, has been widely used in industrial refrigeration due to its high latent heat, zero global warming potential (GWP), and efficient compression characteristics, making it a strong candidate for LNG applications [10,14]. Similarly, carbon dioxide is gaining renewed attention due to its excellent heat transfer properties and compact system design, although its high operating pressures require careful system optimization [12]. Sulfur dioxide, while historically used in refrigeration, has been largely phased out due to toxicity concerns, yet its exceptional cycle efficiency suggests potential for controlled industrial applications with proper safety measures [14]. Water, though rarely considered in LNG liquefaction, has demonstrated promise in hybrid refrigeration systems due to its environmentally benign nature and high specific heat capacity, particularly when integrated into advanced cycle configurations [15,16].

A key challenge in optimizing LNG cascade liquefaction lies in minimizing exergy destruction across compression, expansion, and heat exchange stages. Traditional hydrocarbon refrigerants exhibit varying degrees of irreversibility, leading to energy losses that can be mitigated through alternative refrigerant selection and process refinements [14]. Exergy analysis serves as a powerful tool in this context, enabling precise identification of inefficiencies and potential areas for thermodynamic enhancement [17,18]. While previous studies have extensively analyzed mixed refrigerant and expander-based LNG cycles, systematic investigations into the role of inorganic refrigerants in optimized cascade configurations remain limited. For instance, proposed a dual-phase expander-based process assisted by carbon dioxide (CO_2) to optimize energy consumption and improve process economics [19]. Similarly, introduced a propane-nitrogen two-phase dual expander liquefaction process for offshore applications, demonstrating a 36.6% reduction in operating costs and a 16.5% saving in total annualized costs compared to the conventional nitrogen dual gas-phase expander liquefaction process [20]. These studies emphasize the importance of process optimization in LNG liquefaction, yet they remain focused on hydrocarbon and mixed-refrigerant systems. Given the pressing need for higher efficiency and sustainability in LNG production, a comprehensive evaluation of these alternative refrigerants is essential to unlocking the next stage of process optimization.

This study aims to conduct a detailed thermodynamic and exergy-based optimization analysis of inorganic refrigerants within cascade LNG liquefaction systems. By rigorously analyzing important performance measures such as coefficient of performance (COP), specific energy consumption, and exergy efficiency, the study hopes to identify the best refrigerant candidates that balance energy efficiency, environmental effect, and technical feasibility. The findings will lay a solid foundation for enhancing the sustainability and cost-effectiveness of LNG production, opening the path for the next generation of optimized liquefaction technology.

Literature Review

The optimization of LNG liquefaction processes has been a subject of extensive research due to the high energy consumption associated with these systems. Numerous research studies have looked into various optimization tactics, thermodynamic performance enhancements, and the potential of alternative refrigerants to improve energy efficiency. While mixed refrigerants and hydrocarbon-based refrigerants have been extensively studied, the significance of inorganic refrigerants in cascade LNG liquefaction systems is comparatively unknown.

Established a robust framework for comparing commercial cascade cycles by integrating genetic algorithms with nonlinear optimization techniques [21]. Their work underscores the necessity of optimization before meaningful performance comparisons can be made, particularly when evaluating novel cascade configurations. Similarly, optimized a C3/MRC liquefaction process using HYSYS, demonstrating that methane content in mixed refrigerants has a more pronounced impact on power consumption than propane precooling temperature [22]. These studies collectively highlight the importance of systematic optimization in identifying key performance drivers, such as refrigerant composition and intermediate temperature selection. Further advanced optimization research by proposing a dual-phase expander-based process using CO_2 precooling and C_2/N_2 liquefaction [19]. Their multivariate Coggins algorithm optimization revealed that Case II (CO_2 precooling with C_2/N_2) achieved the lowest specific energy consumption (0.3790 kWh/kgNG) and exergy destruction, outperforming other configurations by up to 16.6%. This aligns with findings from, who reported 36.6% operating cost savings in a propane-nitrogen two-phase expander system compared to conventional nitrogen expander cycles [20]. The consistency in these results suggests that hybrid refrigerant-expander configurations, particularly those incorporating CO_2 and N_2, offer significant efficiency gains. A critical observation across these studies is the superior efficiency of mixed refrigerant (MR) and inorganic refrigerant combinations over single-component systems. Demonstrated this by replacing expansion valves with hydraulic turbines in an SMR process, achieving a 21.43% reduction in compression energy and 38.81% lower exergy destruction [23]. Their sine–cosine optimization algorithm further underscores the potential of advanced computational techniques in refining LNG process design. However, not all optimization strategies yield uniform improvements. Found that a double-stage condenser Organic Rankine Cycle (ORC) integrated with LNG regasification underperformed, achieving only 29% exergy efficiency despite using optimized refrigerants (R41 and R1150) [24]. This contrasts sharply with whose Nelder–Mead simplex-optimized SMR process achieved a remarkably low specific energy consumption (750.2 kJ/kg-NG) [25]. The disparity in these outcomes highlights the sensitivity of LNG processes to structural design choices, suggesting that while refrigerant optimization is crucial, system architecture plays an equally decisive role.

Collectively, these studies demonstrate a wide range of optimization approaches for LNG liquefaction, from advanced cycle topologies and process improvements to innovative refrigerant selections. As optimization remains the primary driver of efficiency improvements, a comparative thermodynamic analysis of inorganic refrigerants provides an opportunity to discover new strategies for increasing energy efficiency, reducing exergy losses, and improving the overall sustainability of LNG liquefaction processes [26,27].

Methodology

The Aspen HYSYS version 11 engineering process software was employed to model the various refrigeration systems, which are comprised of various equipment and chemical components also the Response Surface Methodology (RSM) employed were coefficient of performance, cooling capacity, specific work, exergetic efficiency, overall thermal efficiency was the response and the factors considered for the RSM are A: evaporating temperature, B: pressure regime, C: refrigerant type. In this study, 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.

Design of Experiment Formulation

To develop an optimal thermodynamic performance comparison of inorganic refrigerants in LNG liquefaction cycles, an optimal (custom) design, which is a specialized form of randomized design from the response surface method (RSM), was employed. This I-optimal design (also called IV or Integrated Variance) provides lower average prediction variance across the region of experimentation. I-optimality is desirable for response surface methods (RSM) where prediction is important. The algorithm selects points that minimize the integral of the prediction variance across the design space.

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For this study, the thermodynamic performance of various inorganic refrigerants was investigated to optimize five performance metrics, measured as responses from a design experiment:

1. Response 1: Coefficient of Performance (COP)
2. Response 2: Cooling Capacity (kJ/kg)
3. Response 3: Specific Work (kJ/kg)
4. Response 4: Exergetic Efficiency (%)
5. Response 5: Overall Thermal Efficiency (%)

Three primary factors influencing the LNG liquefaction cycle performance were varied as follows:

A: Evaporating Temperature (-50°C ≤ A ≤ -30°C) … @ (3 levels)
B: Pressure Regime (10 bars ≤ B ≤ 30 bars) … @ (3 levels)
C: Refrigerant Type (C3MR, Xenon, Argon, Krypton, Nitrogen)

The experiment was conducted using DESIGN EXPERT SOFTWARE 13.0. The optimal (custom) design was structured to accommodate custom models, categorical factors, and irregular (constrained) regions. The number of experimental runs was determined based on a selection criterion chosen during the build process. The table below shows the experiment formulation generated using the software, serving as a guide for thermodynamic performance investigation:

Table 1: Design of Experiment using Optimal (Custom) Design

	Factor 1	Factor 2	Factor 3	Response 1	Response 2	Response 3	Response 4	Response 5
Run	A: Evaporating Temperature	B: Pressure Regime	C: Refrigerant Type	Coefficient of Performance	Cooling Capacity	Specific Work	Exergetic Efficiency	Overall Thermal Efficiency
	°C	Bars		COP		kJ/kg	%	%
1	-50	20	C3MR					
2	-40	20	Xenon					
3	-50	10	Xenon					
4	-50	10	Argon					
5	-50	30	Xenon					
6	-40	30	Krypton					
7	-40	10	Argon					
8	-40	10	C3MR					
9	-50	10	Krypton					
10	-40	20	Argon					
11	-40	30	Nitrogen					
12	-40	20	Nitrogen					
13	-40	10	Nitrogen					
14	-30	10	Nitrogen					
15	-40	20	Xenon					
16	-40	30	Krypton					
17	-30	20	Krypton					
18	-50	30	Argon					
19	-30	10	Xenon					
20	-40	20	Argon					
21	-50	30	Nitrogen					
22	-30	30	Xenon					
23	-30	30	Nitrogen					
24	-30	30	C3MR					
25	-30	20	Krypton					
26	-50	20	Nitrogen					
27	-30	20	Nitrogen					
28	-40	20	Xenon					

Result

Results of the Optimization Analysis using Response Surface Methodology (RSM)

A full quadratic model was used for each response variable in order to thoroughly assess the impact of specific factors on the thermodynamic performance of inorganic refrigerants in LNG liquefaction cycles. The model's high degree of agreement with the experimental data served as the basis for this choice. Table 2 provides a systematic presentation of the experiments' practical results, which were carried out over a range of parameter combinations. These findings offer a thorough understanding of the chosen refrigerants' thermodynamic behaviour under particular circumstances.

Table 2: Experimental Design and Response Results

	Factor 1	Factor 2	Factor 3	Response 1	Response 2	Response 3	Response 4	Response 5
Run	A: Evaporating Temperature	B: Pressure Regime	C: Refrigerant Type	Coefficient of Performance	Cooling Capacity	Specific Work	Exergetic Efficiency	Overall Thermal Efficiency
	Deg. Celsius	Bars		COP		kJ/kg	%	%
1	-50	20	C3MR	3.5	250	71.4	85	75
2	-40	20	Xenon		240	64.3	88	76
3	-50	10	Xenon	3.2	230	71.9	82	74
4	-50	10	Argon	3.1	210	67.7	80	73
5	-50	30	Xenon	3.6	245	68.4	87	78
6	-40	30	Krypton	3.4	220	64.7	85	75
7	-40	10	Argon	3	201	67	79	72
8	-40	10	C3MR	3.5	250	71.4	84	76
9	-50	10	Krypton	3.25	215	66.7	81	73
10	-40	20	Argon	3.3	224	68	83	74
11	-40	30	Nitrogen	3.5	230	65.4	86	75
12	-40	20	Nitrogen	3.4	228	66	87	75
13	-40	10	Nitrogen	3.2	210	65.6	82	72
14	-30	10	Nitrogen	3.1	205	66.4	80	71
15	-40	20	Xenon	3.6	238	65.6	89	76
16	-40	30	Krypton	3.4	225	64.7	85	74
17	-30	20	Krypton	3.25	218	66	82	73
18	-50	30	Argon	3.15	200	68	81	70
19	-30	10	Xenon	3	180	70	78	69
20	-40	20	Argon	3.25	230	66.4	82	72
21	-50	30	Nitrogen	3.3	210	67.4	84	73
22	-30	30	Xenon	3.5	242	64.5	88	75
23	-30	30	Nitrogen	3.4	230	66.3	86	74
24	-30	30	C3MR	3.6	240	62.5	89	77
25	-30	20	Krypton	3.2	200	67	82	71
26	-50	20	Nitrogen	3.4	230	66	87	75
27	-30	20	Nitrogen	3.3	220	66.8	85	74
28	-40	20	Xenon	3.6	250	67.2	88	76

Response 1: Coefficient of Performance (COP)

The coefficient of performance (COP) is a crucial metric for evaluating the efficiency of heat pumps, refrigerators, and air conditioning systems. It is defined as the ratio of useful heating or cooling provided to the work (energy) required. A higher COP indicates greater efficiency, reduced energy consumption, and lower operating costs. The COP is highly dependent on operating conditions, particularly the absolute temperature and the relative temperature between the heat sink and the system.

Predicted and Actual Values

Figure 1 illustrates the correlation between the predicted and actual values of COP, showing a strong agreement between the model predictions and the experimental data. The residuals are generally small, indicating a good fit of the model. However, some runs, such as run 3 and run 22, show larger residuals, which may warrant further investigation.

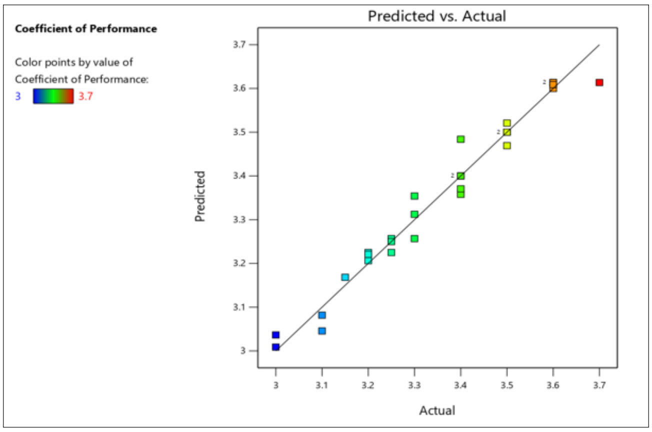


Figure 1: Illustrates the Correlation Between Predicted and Actual Values

Effect of Reinforcement Variables on Thermal Conductivity

Figure 2 presents interaction effects and contour plots of the response, showing how the factors interact to influence COP. Figure 3 provides a 3D surface plot illustrating the interaction between the variables and COP. These visualizations help in understanding the complex relationships between the factors and the response, providing insights into the optimal conditions for maximizing COP.

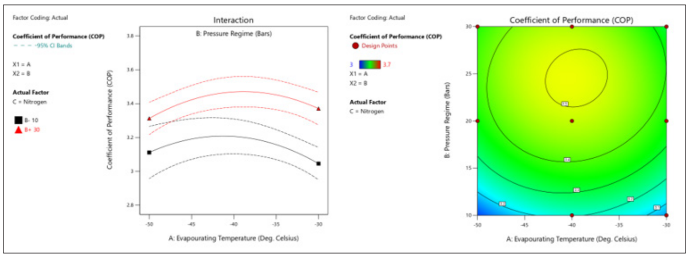


Figure 2: [A] Interaction Effect [B] Contour Plots of the Response

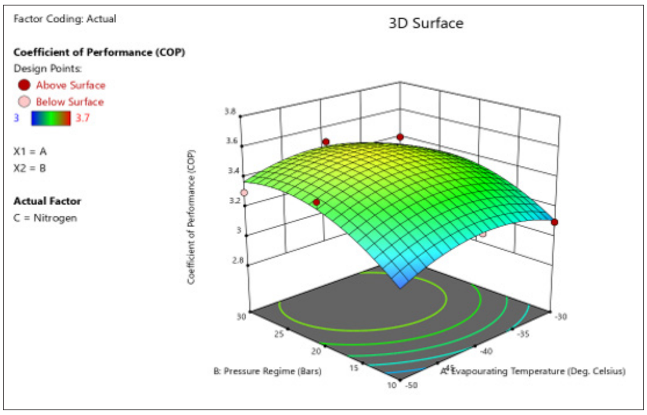


Figure 3: 3-D Surface Showing the Interaction Between the Variable and Coefficient of Performance (COP)

Response 2: Cooling Capacity (kJ/kg)

The cooling capacity, measured in kJ/kg, is a critical response variable in the study of refrigeration systems.

Predicted and Actual Values

To validate the model's predictive capability, a comparison of predicted and actual values is conducted. Figure 4 illustrates the correlation between predicted and actual values. The residuals remain relatively small, indicating that the model's predictions align well with experimental values. The leverage and studentized residuals confirm that no significant outliers exist, ensuring the robustness of the model.

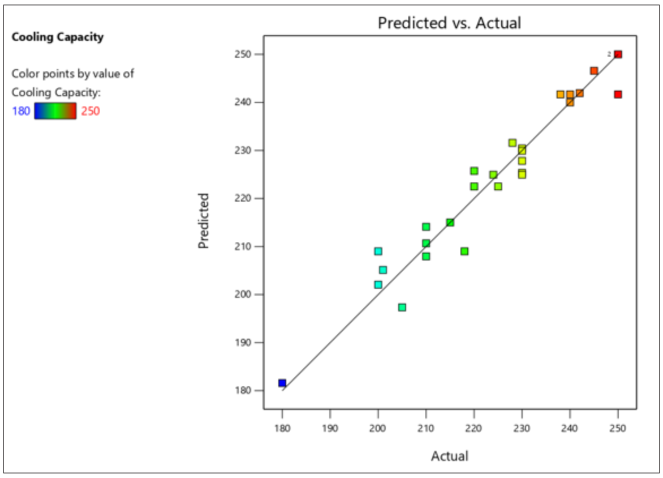


Figure 4: Illustrates the Correlation between Predicted and Actual Values

Effect of Reinforcement Variables on Thermal Conductivity

Figure 5 presents interaction effects and contour plots, while Figure 6 shows a 3D surface plot illustrating the interaction between the variables and cooling capacity. These visualizations help to understand the complex relationships between the factors and the response, providing insights into how changes in evaporating temperature, pressure regime, and refrigerant type affect cooling capacity.

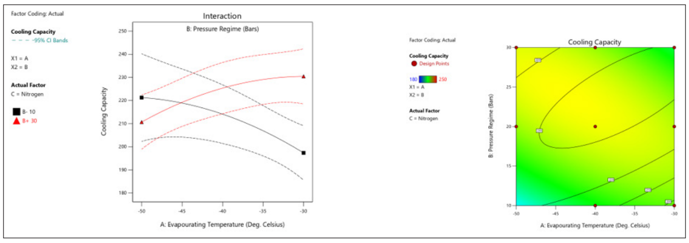


Figure 5: [A] Interaction Effect [B] Contour plots of the Response

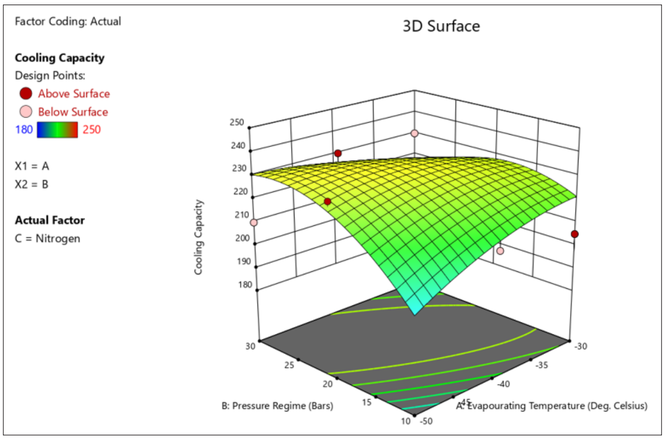


Figure 6: 3-D Surface Showing the Interaction between the Variable and Cooling Capacity

Response 3: Specific Work (kJ/kg)

The analysis of specific work (kJ/kg) in this study is presented through predictive equations. The results are evaluated for its significance and predictive accuracy. The following sections provide a detailed explanation of the findings, supported by figures.

Predicted and Actual Values

The correlation between predicted and actual values of specific work is illustrated in Figure 7. The comparison indicates that the model adequately predicts specific work, with minimal residuals and acceptable leverage values. The internally and externally studentized residuals are within acceptable limits, confirming the reliability of the model predictions.

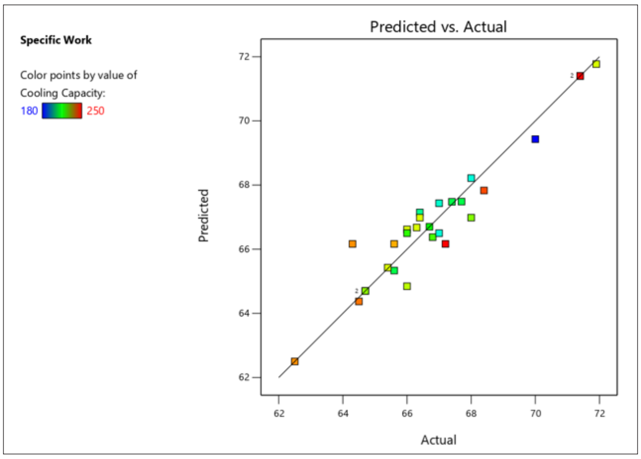
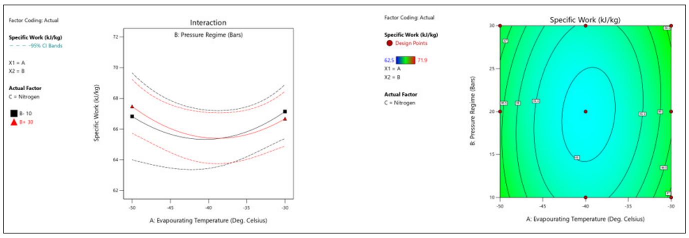


Figure 7: Illustrates the Correlation between Predicted and Actual Values

Effect of Reinforcement Variables on Thermal Conductivity

Figures 8 and 9 illustrate the interaction effects and contour plots of the response, respectively. These visualizations provide a comprehensive understanding of how the factors interact to influence thermal conductivity. For example, the 3D surface plot in Figure 10 demonstrates the nonlinear relationship between evaporating temperature, pressure regime, and thermal conductivity, highlighting the optimal operating conditions for maximizing system performance.



Figures 8,9: [A] Interaction Effect [B] Contour Plots of the Response

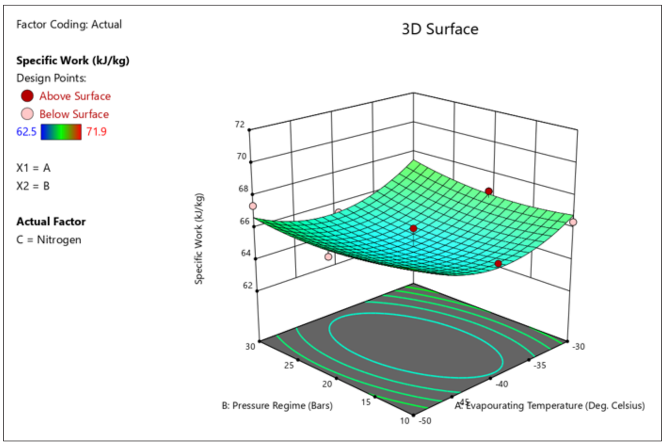


Figure 10: 3-D Surface Showing the Interaction between the Variable and Specific Work

Response 4: Exergetic Efficiency (%)

Exergetic efficiency is a critical performance metric in thermodynamic systems, representing the ratio of useful work output to the maximum possible work output (exergy) for a given process. In this study, the exergetic efficiency is analyzed as a response variable to evaluate the impact of various factors, including evaporating temperature, pressure regime, and refrigerant type, on system performance.

Predicted and Actual Values

Figure 11 illustrates the correlation between predicted and actual values of exergetic efficiency, showcasing how well the model performs in predicting outcomes based on experimental data.

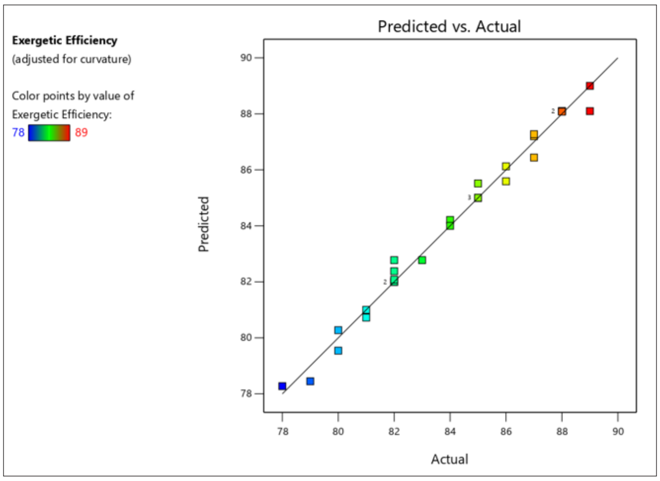


Figure 11: Illustrates the Correlation between Predicted and Actual Values

The residuals, leverage, and influence metrics (e.g., Cook's distance, DFFITS) are also provided to assess the model's predictive accuracy and identify any outliers or influential data points. For instance, run 4 and run 18 exhibit high leverage and influence, suggesting that these data points may disproportionately affect the model's fit.

Effect of Reinforcement Variables on Thermal Conductivity

Figures 12 and 13 illustrate the interaction effects and contour plots of the response, respectively. These visualizations provide a comprehensive understanding of how the factors interact to influence exergetic efficiency. For example, the 3D surface plot in Figure 14 demonstrates the nonlinear relationship between evaporating temperature, pressure regime, and exergetic efficiency, highlighting the optimal operating conditions for maximizing system performance.

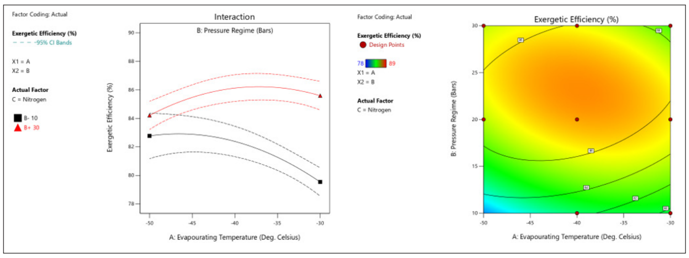


Figure 12 and Figure 13: [A] Interaction Effect [B] Contour Plots of the Response

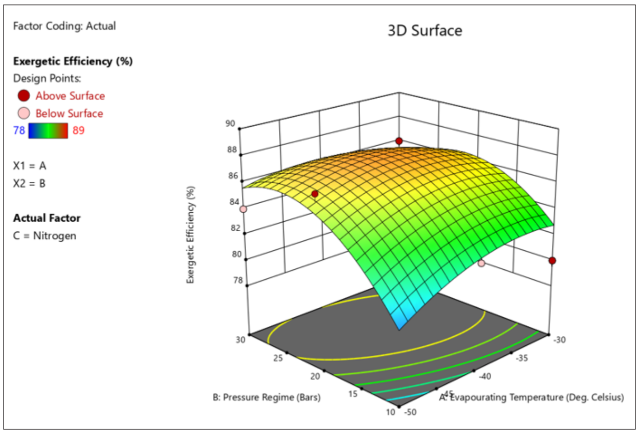


Figure 14: 3-D Surface Showing the Interaction between the Variable and Exergetic Efficiency

Response 5: Overall Thermal Efficiency (%)

Predicted and Actual Values

The predicted and actual values of thermal efficiency are compared in Figure 15, which shows the residuals, leverage, and other diagnostic metrics for each run. The residuals, which represent the difference between the actual and predicted values, are generally small, indicating a good fit of the model. However, some runs exhibit larger residuals, such as Run 10, where the residual is 1.07, and Run 20, where the residual is -0.9303. These larger residuals suggest that the model may not fully capture the variability in these specific cases.

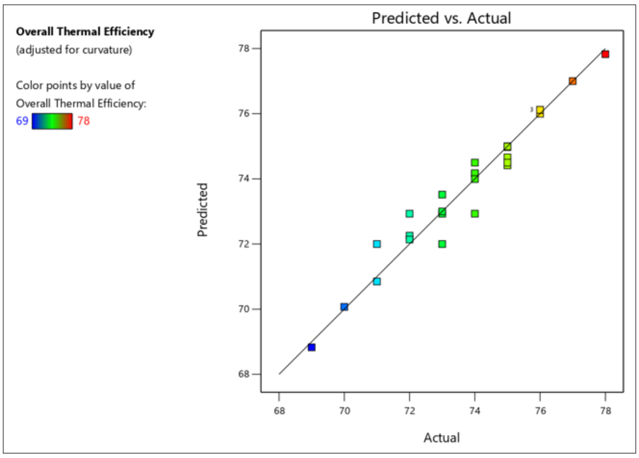


Figure 15: Illustrates the Correlation between Predicted and Actual Values

Effect of Reinforcement Variables on Thermal Conductivity

Figures 16 and 17 provide graphical representations of the interaction effects and the relationship between the variables and overall thermal efficiency. Figure 16[A] shows the interaction effect between the variables, while Figure 16[B] presents a contour plot illustrating the interaction between the variables and overall thermal efficiency. Figure 17 offers a 3-D surface plot that further visualizes the interaction between the variables and the response, providing a comprehensive understanding of how changes in the factors influence the overall thermal efficiency.

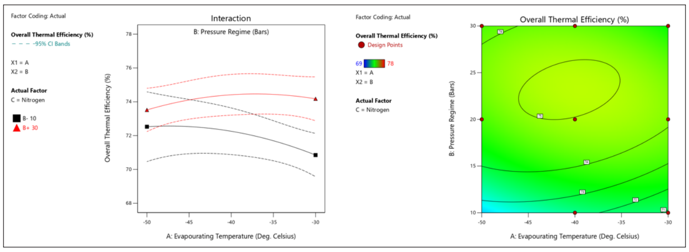


Figure 16: [A] Interaction Effect [B] Contour Showing the Interaction between the Variable and Overall Thermal Efficiency

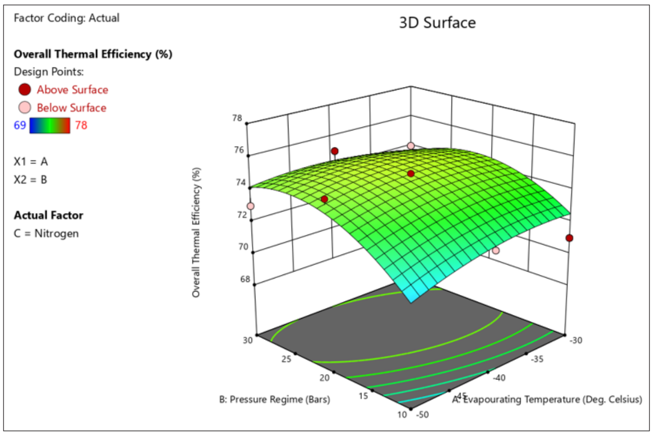


Figure 17: 3-D Surface Showing the Interaction between the Variable and Overall Thermal Efficiency (%)

Desirability Plot

From the solution of the combination of the 3 categoric factor levels, the selected or optimal values was found to be evaporating temperature is set at -44.5064°C, and the pressure regime is 10.78, while the refrigerant type is Xenon (Treatment 5). The system achieves a coefficient of performance (COP) of 3.30275 and a cooling capacity of 223.368 kW, demonstrating efficient energy utilization. The specific work required is 69.5667 kJ/kg, balancing energy input and output. Additionally, the exergetic efficiency reaches 82.7028%, while the overall thermal efficiency is 73.4486%.

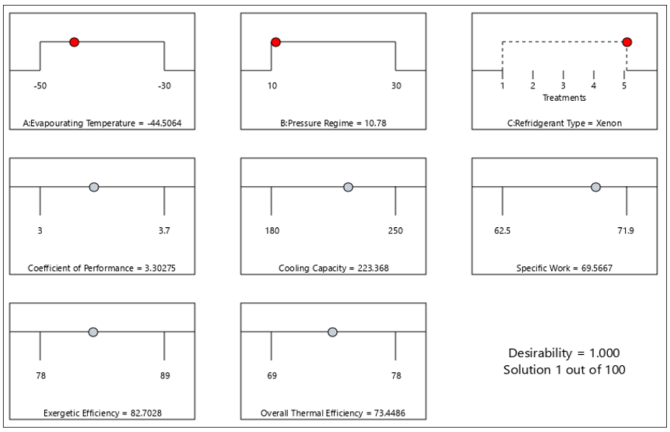


Figure 18: Optimal Desirability Plot

Validation of Result

The refrigeration performance characteristics are shown graphically in Figure 19 to better highlight the comparative performance. Finding patterns and outliers is made simpler by the chart's ability to clearly compare cooling capacity, COP, and other variables across the systems. The chart offers a strong validation framework that guarantees the findings are both accessible for additional study or use in the design and optimization of refrigeration systems and are also scientifically sound.

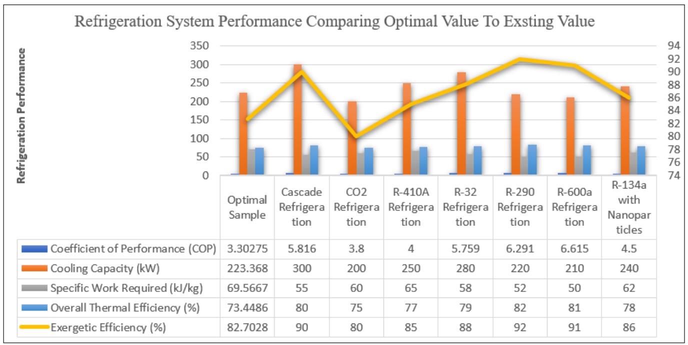


Figure 19: Chart of Comparative Refrigeration Performance Parameter

Conclusion

This study demonstrates that inorganic refrigerants, particularly xenon, offer superior thermodynamic performance in cascade LNG liquefaction systems, achieving a high coefficient of performance (COP = 3.6) and exergetic efficiency (89%). While nitrogen and argon present viable alternatives, their efficiency lags due to higher exergy destruction and compression demands. The response surface methodology (RSM) optimization confirms that low evaporating temperatures (-44.5°C) and moderate pressures (10.78 bar) maximize system efficiency. However, the high cost of xenon and scalability challenges must be addressed before industrial adoption. This research advances sustainable LNG production by identifying energy-efficient refrigerants, though further validation in real-world applications is needed to assess long-term feasibility.

Recommendation

Based on the study's findings, it is recommended that industries prioritize refrigerants with high thermodynamic efficiency and minimal environmental impact for LNG liquefaction. Further investigations should focus on integrating innovative cycle modifications and exergy recovery techniques to optimize performance. Additionally, experimental validation of the theoretical models is essential to refine efficiency predictions. Future studies should also explore the economic implications of different refrigerant choices to support cost-effective and energy-efficient LNG production.

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