International Journal of Applied and Natural Sciences
ISSN: 3005-3471
DOI: 10.61424/ijans
Journal Homepage: www.bluemarkpublishers.com/index.php/IJANS

IJANS
BLUEMARK PUBLISHERS

| RESEARCH ARTICLE

A Comprehensive Review of Inorganic Refrigerants in LNG Liquefaction Cycles: Thermodynamic Performance and Environmental Impact

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| ABSTRACT

As the demand for Liquefied Natural Gas (LNG) continues to rise, the need for efficient and environmentally friendly refrigeration technologies has become more critical. This study presents a comprehensive review of inorganic refrigerants used in Liquefied Natural Gas (LNG) liquefaction cycles, concentrating on their thermodynamic performance and environmental effects. Using a thorough literature study, important refrigerants such as nitrogen, argon, krypton, xenon, and ammonia were examined in terms of efficiency, energy consumption, and sustainability. The findings show that inorganic refrigerants can improve energy efficiency by lowering power consumption and increasing exergy performance. Nitrogen was found to require the least amount of energy, whereas ammonia significantly increased the coefficient of performance (COP) in mixed refrigerant applications. Krypton and xenon both demonstrated great exergy efficiency, making them attractive candidates for future LNG operations. While these refrigerants have a lesser environmental effect than standard hydrocarbons, more advances are needed. The study recommends optimizing hybrid refrigerant systems, including renewable energy, and improving safety measures. Advancing these strategies can make LNG production more sustainable, reducing its carbon footprint while maintaining efficiency.

| KEYWORDS

Inorganic refrigerants, Liquefaction cycles, Thermodynamic performance, Environmental impact, Energy efficiency

| ARTICLE INFORMATION

ACCEPTED: 10 March 2025 **PUBLISHED:** 11 April 2025 **DOI:** 10.61424/ijans.v3.i1.258

1. Introduction

Natural gas, widely regarded as the cleanest fossil fuel, has emerged as one of the world's fastest expanding primary energy sources (Zou, 2020). This tendency has established natural gas as an important component of the global energy industry (Blose *et al.*, 2023). Natural gas has numerous qualities that make it a highly efficient, relatively clean-burning, and economical energy source (Wang *et al.*, 2022). As stated by EIA (2024), it has become a significant player in the global energy market, supplying around 23% of the world's energy and contributing nearly a quarter of the electricity generated worldwide. According to Shell's 2021 'Liquefied Natural Gas (LNG) Outlook Report', the global demand for LNG will reach 700 million tons by 2040. The increasing reliance on natural gas is not just about meeting energy demands but also about addressing environmental concerns (Azam *et al.*, 2021).

When compared to other fossil fuels, natural gas stands out for its comparatively lower emissions, both in terms of traditional air pollutants and greenhouse gases (nitrogen oxides (NOx), which are known to contribute to both global warming and acid rain), which has contributed to its widespread growth and adoption (Zuhuri, Mossavar-Rahmani and Behgounia, 2022; Arshad *et al.*, 2024). Fonquergne and Balch (2024) highlighted that in many regions, the replacement of coal with natural gas has led to significant declines in CO_2 emissions, thus demonstrating its effectiveness as an interim solution in meeting climate objectives.

2. Review of Liquefied Natural Gas Liquefaction Cycles

A unique characteristic that natural gas possesses is its ability to be transformed into Liquefied Natural Gas (LNG). 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 (Galimova et al., 2023). After extraction, the natural gas undergoes liquefaction at LNG plants (Peter Simpa *et al.*, 2024). Liquefaction involves cooling the gas to extremely low temperatures (typically around -162 degrees Celsius), which causes it to condense into a liquid state (Zhang, Meerman, René Benders, *et al.*, 2020; Yang, Li and Tan, 2023). A variety of refrigerants can be used in these cycles, with each type of refrigerant suited to different operational conditions. In selecting a refrigerant, companies consider factors such as environmental impact, thermodynamic efficiency, and compatibility with the desired operating temperatures (Savitha *et al.*, 2022).

These cycles utilize both natural and synthetic refrigerants, each with distinct properties and environmental impacts. Natural refrigerants such as hydrocarbons, ammonia, and carbon dioxide are increasingly favoured for their low environmental impact, while synthetic refrigerants like chlorofluorocarbons (CFCs), hydrochlorofluorocarbons (HCFCs), and hydrofluorocarbons (HFCs) have faced restrictions due to their ozone-depleting effects under the Montreal Protocol (Oshodin, Bolaji and Olorunfemi, 2023). In low-temperature applications such as LNG production, refrigerants like pure ethylene, methane, propane, and ethane are commonly employed due to their optimal thermodynamic properties at cryogenic temperatures (Ozueh, Ajienka and Joel, 2022).

There are only a few extensive reviews on the use of inorganic refrigerants in LNG liquefaction cycles, with an emphasis on their thermodynamic performance and environmental impacts. Ozueh, Ajienka and Joel (2022) studied the characteristics of inorganic refrigerants, including argon, krypton, xenon, and Nitrogen, in the light of an organic refrigerant - propane pre-cooled mixed refrigerants (C3MR). Refrigerants used in LNG processing influence the environmental footprint of the industry and impact operational costs. For example, Hydrocarbons such as methane and propane are particularly successful in achieving the low temperatures required for LNG production while maintaining energy efficiency. This is clarified in the research made by (Mazyan *et al.*, 2020), mixing the propane refrigerant with ammonia, sulfur dioxide and carbon dioxide, on the performance and the work of the compressor is studied. It was shown that the mixture of ammonia-propane and sulfur dioxide-propane enhances the overall COP by 7% and 9%, respectively, of the performance and the work of the compressor. Moreover, these refrigerants can be sourced from natural gas itself, creating a sustainable loop within the LNG production process. (Peter Simpa *et al.*, 2024) examined current trends and future opportunities for enhancing sustainability and reducing environmental impact across the LNG value chain.

The objective of this paper is to provide a comprehensive review of inorganic refrigerants used in LNG liquefaction cycles, assessing both their environmental impact and thermodynamic performance. This review is important because it has the potential to advance environmental science, refrigeration technology, and sustainable energy systems.

3. Methodology

This study employed a systematic literature review methodology to critically examine the environmental effects and thermodynamic performance of inorganic refrigerants in LNG liquefaction cycles. Process modelling, energy analysis, COP evaluation, and environmental effect assessment comprise the framework of the methodology. A comprehensive search strategy was implemented using the Scopus database, selected for its extensive coverage of manufacturing technology literature and high-quality peer-reviewed publications. The search focused on publications between 2020-2025, employing a carefully constructed Boolean search string combining key terms "Inorganic refrigerants" AND "Liquefaction cycles" AND "Liquefied Natural Gas" AND "Thermodynamic performance" AND “Environmental impact”. The initial search yielded 164 papers. The inclusion criteria prioritized peer-reviewed journals and high-impact conference proceedings, focusing specifically on empirical studies. Papers were excluded if they lacked clear methodological frameworks or focused solely on conceptual models without practical applications. However, this rigorous approach resulted in a final selection of 30 papers.

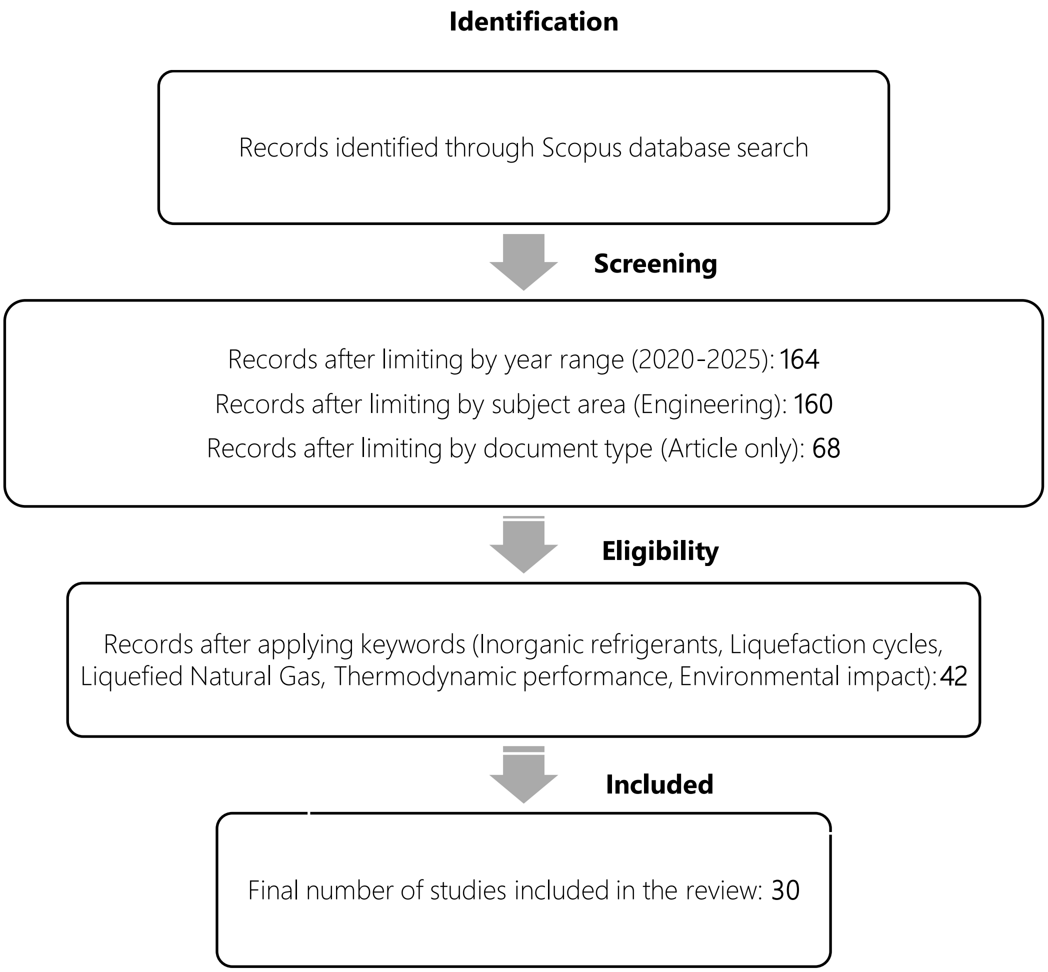

Figure 1: PRISMA Flow Diagram for Literature Selection Process

3.1 Publication of Journals by Ranking

The publication trends revealed in Figure 2 and Table 1 demonstrate a significant surge in research interest surrounding smart manufacturing and data analytics, with a striking concentration of scholarly output in recent years. The year 2024 stands out, representing 37.77% of the total publications, indicating an accelerating academic discourse on visibility in smart manufacturing operations. This temporal distribution reflects the rapidly evolving technological landscape and the increasing urgency for organizations to integrate advanced analytics into their operational frameworks (Bello et al., 2024; Gladysz et al., 2024). The exponential growth from merely 3 publications in 2019 to 17 in 2024 suggests a critical paradigm shift in understanding visibility technologies' strategic importance (Lehmam and Hassani, 2024).

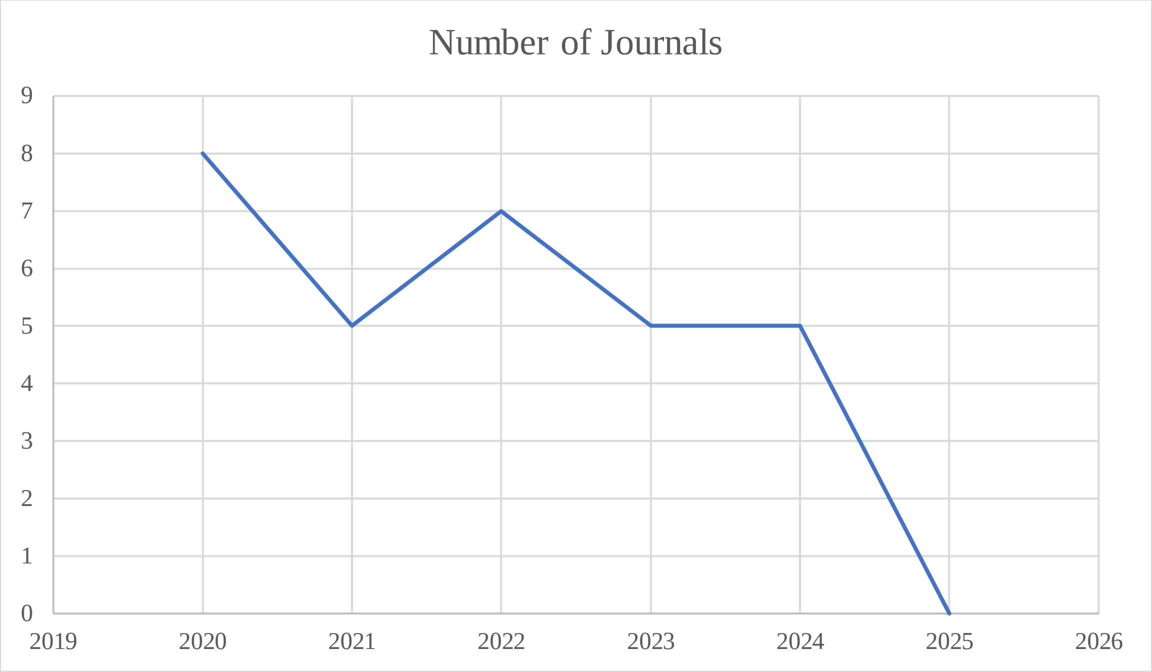

Figure 2: Journal Article by Year of Publication

Table 1: Year-wise distribution of articles

Year	Number of Journals	% of total
2025	0	0
2024	5	16.67
2023	5	16.67
2022	7	23.33
2021	5	16.67
2020	8	26.67
Total	30	100

3.2 Overview of LNG Liquefaction Cycles

Table 2 depicts a fundamental comparison framework in which multiple LNG liquefaction cycles emerge as important factors influencing energy efficiency, thermodynamic performance, and environmental effects. Each cycle, Cascade, Mixed Refrigerant, and Expander-based liquefaction presents distinct benefits and constraints, determining their application.

Table 2: Different cycles in Liquefied Natural Gas (LNG) liquefaction

S/N	**Theme**	**Author (s)**	**Key Finding**
1	Cascade Liquefaction Process	(He, Lin and Du, 2022)	"…In this 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). The proposed processes are combined with advanced distillation methods to reduce the energy consumption through process and energy integration…"
		(Wu, Wang, Dong, *et al.*, 2021)	They provided a comparative and critical analysis of the current status of natural gas liquefaction technology through examination of the advantages and disadvantages associated with three natural gas liquefaction processes (namely, the cascade liquefaction cycle, the expander-based cycle and the mixed refrigerant cycle). It was shown that the energy consumption related to the cascade refrigeration cycle is the lowest. Compared with it, the mixed refrigerant cycle has the advantages of being a simpler process, with fewer units, low investment cost and low requirements in terms of refrigerant purity. The expander-based cycle can be started and stopped quickly and simply, but the power consumption is relatively high.
2	Mixed Refrigerant Liquefaction Process	(Wu *et al.*, 2023)	"In this paper, this mixed refrigerant cycle liquefaction process is simulated using the HYSYS software and the main influential parameters involved in the process are varied to analyze their influence on the liquefaction rate and power consumption…"
		(Majeed *et al.*, 2020)	"Among all large-scale natural gas (NG) liquefaction processes, the mixed fluid cascade (MFC) process is recognized as the best alternative option for LNG production, mainly due to its competitive performance. However, from a thermodynamic point of view, the MFC process is still far from its potential maximum energy efficiency due to non-optimal execution of design variables."
		(Wu, Wang, Dai, *et al.*, 2021)	"The main objective of this study was to optimize the most commonly used mixed refrigerant process. The liquefaction performance of the optimized process was analyzed, and the influence of gas parameters on the power consumption, exergy loss, freezing mixture circulation, and cooling water load were investigated. The results show that compressor power consumption can be reduced by 29.8%, the cooling water load can be reduced by 21.3%, and the system exergy efficiency can be increased by 41% with the optimized process."

		(Tak *et al.*, 2023)	In this paper, a triple mixed-refrigerant (MR) cascade process with multi-stream heat exchangers (MSHEs) and compression system was compared to a single MR process with one MSHE, considered as the base case, and showed 19.0% energy reduction (805.6 kJ/kg LNG).
3	**Expander-based cycle**	(Qyyum, Qadeer, Ahmad and Lee, 2020)	"The use of nitrogen (N_2) expander-based liquefaction processes is prevalent in offshore sites for the production of floating liquefied natural gas. It is safe, has simple operability, and has a portable design with a small deck space requirement. However, the high operating cost that mainly accounts for the shaft work requirement in the compression units of the refrigeration cycle is still a major ongoing issue associated with nitrogen expander liquefaction processes. This high operating cost increases the total annualized costs of the N_2 expander liquefaction technology, and this ultimately reduces its global competitiveness of the process…."

3.3 Contribution of Inorganic Refrigerants to the Thermodynamic Efficiency of LNG Liquefaction Cycles

Table 3 reveals a vital comparison framework in which inorganic refrigerants play an important role in increasing thermodynamic efficiency, lowering energy consumption, and optimizing exergy performance in LNG liquefaction cycles. The findings emphasize the trade-offs between different refrigerants, with a focus on particular power consumption, coefficient of performance (COP), and total system efficiency.

Table 3: Thermodynamic Performance of Inorganic Refrigerants in LNG Cycles

S/N	Refrigerants	Author (s)	Key Finding
1	**Nitrogen (N_2)**	(Tobechi F. Ozueh, Ajienka and Joel, 2022)	"This paper comparatively studies the characteristics of four inorganic refrigerants, including argon, krypton, xenon, and Nitrogen, in the light of an organic refrigerant - propane pre-cooled mixed refrigerants (C_3MR) … The result shows that Nitrogen requires the least energy at the compressors…"
		(Qyyum, Qadeer, Ahmad, Ahmed, *et al.*, 2020)	"Accordingly, this paper proposes two-phase expansion using an innovative binary mixed refrigerant (MR) composed of ethane and nitrogen (C_2N)… Using the C_2N two-phase expander LNG process, 47.83% energy can be saved with 55.25% exergy destruction minimization and 24.12% total annualized costs (TAC) savings as compared to previously published nitrogen single expander process. Whereas, the C_3-precooled C_2N process gives higher energy savings, i.e., 52.45%, but low TAC savings, i.e., 1.6%, as compared to the nitrogen single expander LNG process."

		(Chan, Tam and Dev, 2022)	"…A small-scale liquefaction process with a refrigerant cycle is proposed in this study to meet these FLNG challenges. The Brayton refrigeration (BR) cycle is found to be most suitable for FLNG vessels, among other refrigerant cycles. The BR cycle using nitrogen as a refrigerant and a single expander is the focus of this study."
		(Chenchen *et al.*, 2022)	"This study introduces a pioneering Liquefied Natural Gas (LNG) cold energy-coupled Liquid Air Energy Storage (LAES) system, which incorporates an innovative nitrogen refrigeration cycle… The system's round-trip efficiency increases from 74.42% to 77.33% as the LNG temperature decreases from 135°C to -145°C, indicating a notable positive trend… The system's exergy efficiency is approximately 62.9%, surpassing recent research outcomes."
		(Chenchen *et al.*, 2022)	"In this paper, a liquefied natural gas cold energy coupled Liquid air energy storage (LNG-LAES) system containing nitrogen refrigeration is proposed, which uses nitrogen expansion to reduce the temperature and supplements high-grade cold energy for the cold box…. The results show that the round-trip efficiency of the system increases first and then decreases with the increase in charging pressure, and the optimal system performance is found to be at a charging pressure of 6.1 MPa… The LNG temperature is in the range of -135 ℃ to -145 ℃, and the system round-trip efficiency increases from 72.83% to 77.33%, demonstrating a gradual upward trend… the exergy efficiency of the system is about 62.9%, which is higher than the recent research results."
		(Yazdaninia *et al.*, 2021)	"In this study, a closed nitrogen expansion cycle (Niche) has been simulated with Aspen HYSYS V8.4. Energy and exergy analysis were applied to evaluate the process. Results of energy analysis indicated that the specific power consumption of this process is 0.68 kWh/kg LNG. The results of exergy analysis showed that exergy efficiency of Niche LNG is 35.51%."
		(Liu *et al.*, 2024)	"In this paper, based on the traditional BOG re-liquefaction process, a liquid nitrogen precooling circulation system is added and linked with the nitrogen purging system. Aspen HYSYS software was used to simulate and analyze the exergy efficiency, specific power consumption, and exergy efficiency in the proposed liquefaction process… According to the operation results, the optimized process has a specific power consumption of 1.028 kWh/kg LNG, which is 21.53 % lower than that of the original NH_3 and CO_2 precooling system."

		(Mehrpooya, Sadaghiani and Hedayat, 2020)	"The development and analysis of a combined hydrogen and natural gas liquefaction process are reported… Total power consumption of the process is 4.165 kWh/kg $_{LH2}$, kg $_{LNG}$. The energy analysis indicated that the coefficient of performance (COP) for the developed process is 0.2442, which is notably high compared with the COP of other identical processes (typically less than 0.1797). The exergy analysis revealed that the overall exergy efficiency of the liquefaction process is 62.54%."
2	**Argon**		"This study aimed to analyse five refrigerants in terms of energy requirement and cost, with consideration to maximize efficiency, minimize energy consumption… of our refrigerants – Nitrogen, Xenon, Argon and Krypton were analyzed and compared to the well-known APCI propane precooled mixed refrigerant (C3MR) Process. The exergy efficiency of Argon was 83%..."
		(Tobechi F. Ozueh, Ajienka and Joel, 2022)	"This paper comparatively studies the characteristics of four inorganic refrigerants, including argon, krypton, xenon, and Nitrogen, in the light of an organic refrigerant - propane pre-cooled mixed refrigerants (C3MR). This study was achieved by first simulating an already existing liquefaction plant using ASPEN HYSYS 11.0 and then analyzing the above-mentioned refrigerants with focus on Global Warming Potential, energy efficiency, exergy and coefficient of performance of the system. The result shows that Nitrogen requires the least energy at the compressors, and Krypton proves to be the best refrigerant for the chiller, while Xenon provided the best cooling effect, followed by C3MR, which has a high coefficient of performance."
3	**Xenon**	(Tobechi F. Ozueh, Ajienka and Joel, 2022)	"This study aimed to analyze five refrigerants in terms of energy requirement and cost, with consideration to maximize efficiency, minimize energy consumption… of our refrigerants – Nitrogen, Xenon, Argon and Krypton were analyzed and compared to the well-known APCI propane precooled mixed refrigerant (C3MR) Process. The exergy efficiency of Xenon was 36%..."
4	**Ammonia (NH_3)**	(Soujoudi and Manteufel, 2021)	"The objective of this paper is to investigate the feasibility of using an environmentally friendly refrigerant compound, ammonia, in the mixed refrigerant (MR) for the liquefied natural gas (LNG) pre-cooling cycle through thermodynamic performance, economic analyses and environmental impact… The thermodynamic analysis shows increasing ammonia's concentrations in the mixed refrigerant by 10% had the largest enhancement on the coefficient of performance (COP) of MR-1 by 0.67 and decreased the specific energy consumption of the pre-cooling cycle by 128 kJ/kg compared to the base case."

		(Zhang, Meerman, Rene Benders, *et al.*, 2020)	"Four expander-based processes were optimized and compared in this study, including conventional single nitrogen expansion process without (SN) and with ammonia absorption precooling (SNA), and single methane expansion process without (SM) and with ammonia absorption precooling (SMA)… The energy and cost analyses were performed for the four processes by comparing optimization results. Lastly, exergy losses in the main equipment were analyzed. The results show that the ammonia precooling cycle reduces energy consumption and production cost by 26–35% and 13–17%, respectively."
5	**Krypton**	(Tobechi F. Ozueh, Ajienka and Joel, 2022)	"This study aimed to analyse five refrigerants in terms of energy requirement and cost, with consideration to maximize efficiency, minimize energy consumption… of our refrigerants – Nitrogen, Xenon, Argon and Krypton were analyzed and compared to the well-known APCI propane precooled mixed refrigerant (C3MR) Process. The exergy efficiency of krypton was 82%…"

3.4 Environmental Impact of Inorganic Refrigerants

Table 4 provides a detailed assessment of the environmental effect of inorganic refrigerants in LNG liquefaction cycles, with an emphasis on global warming potential (GWP), ozone depletion potential (ODP), carbon footprint, and sustainability.

Table 4: Environmental Impact of Inorganic Refrigerants

Impact	Author(s)	Key Finding(s)
Global Warming Potential (GWP) and Ozone Depletion Potential (ODP)	(Bhatti *et al.*, 2024)	"Traditional refrigerants like chlorofluorocarbons (CFCs) and hydrochlorofluorocarbons (HCFCs) have caused significant environmental concerns because of their role in ozone depletion and global warming. This review explores the emerging field of environment-friendly refrigerants… Transitioning to environment-friendly refrigerants is essential for achieving sustainable refrigeration and air conditioning systems, mitigating climate change, and ensuring the long-term viability of cooling technologies while preserving the environment."
	(Tobechi F. Ozueh, Ajienka and Joel, 2022)	"This paper comparatively studies the characteristics of four inorganic refrigerants including argon, krypton, xenon, and Nitrogen … This study was achieved by first simulating an already existing liquefaction plant using ASPEN HYSYS 11.0 and then analyzing the above-mentioned refrigerants with focus on Global Warming Potential, energy efficiency, exergy and coefficient of performance of the system…"
	(Soujoudi and Manteufel, 2021)	"The objective of this paper is to investigate the feasibility of using an environmentally friendly refrigerant compound, ammonia, in the mixed refrigerant (MR) for the liquefied natural gas (LNG) pre-cooling cycle through thermodynamic performance, economic analyses and environmental impact… reducing the methane

		concentration in MR and replacing it with ammonia, decreased the amount of refrigerant leakage through compressor's seals and reduced the global warming potential index (GWPI) of mixed refrigerant up to 24.3%."
Carbon Footprint/Toxicity & Safety Risks	(Wang, He and He, 2024)	"The results show that ART integrated into the liquefaction processes could reduce the SPC and CE by 10~38% and 10~36% for NG liquefaction processes, and 2~24% and 5~24% for H_2 liquefaction processes. ART, which can achieve lower precooling temperatures and higher energy efficiency, shows more attractive perspectives in low carbon emissions of NG and H_2 liquefaction."

4. Discussion

The use of inorganic refrigerants in LNG liquefaction cycles has received a lot of interest because of their ability to improve thermodynamic efficiency while reducing environmental effects. Inorganic refrigerants such as nitrogen, argon, xenon, krypton, and ammonia have had extensive thermodynamic testing, with studies highlighting their ability to optimize energy consumption, improve exergy efficiency, and reduce specific power requirements (Qyyum, Qadeer, Ahmad and Lee, 2020; Tobechi F. Ozueh, Ajienka and Joel, 2022). Researchers such as Soujoudi and Manteufel (2021) have emphasized the importance of ammonia-based refrigerants in enhancing the coefficient of performance (COP) of liquefaction cycles while also lowering specific energy consumption. Despite these benefits, the widespread use of inorganic refrigerants in LNG liquefaction cycles is hampered by numerous significant obstacles. One of the main issues is the high operating costs of certain refrigerants, especially those that require substantial cooling and compression procedures (Mazyan *et al.*, 2020; Qyyum, Qadeer, Ahmad and Lee, 2020). Furthermore, toxicity and safety considerations, particularly with ammonia, raise regulatory and operational issues that must be addressed for large-scale usage (Soujoudi and Manteufel, 2021). Environmental issues influence the practicality of certain refrigerants. While nitrogen, argon, and krypton have zero ozone depletion potential (ODP) and a low global warming potential (GWP), their energy-intensive liquefaction procedures might negate these environmental benefits if not properly optimized (Bhatti *et al.*, 2024; Tobechi F. Ozueh, Ajienka and Joel, 2022). As a result, using sophisticated process optimization tactics, mixed refrigerant configurations, and renewable energy inputs might increase their sustainability while keeping LNG production economically feasible.

5. Conclusion

This study presents a complete analysis of the thermodynamic performance and environmental effect of inorganic refrigerants used in LNG liquefaction cycles. The findings highlight the importance of refrigerant selection in process efficiency, energy consumption, and environmental sustainability. While nitrogen, argon, xenon, krypton, and ammonia have distinct benefits in cryogenic applications, their viability is dependent on cost-effectiveness, safety concerns, and process integration techniques. It also emphasizes the necessity for ongoing research and technical innovation to improve the performance of inorganic refrigerants in LNG liquefaction.

5.1 Recommendations

Based on the findings of this study, the following recommendations are proposed to enhance the efficiency, sustainability, and feasibility of inorganic refrigerants in LNG liquefaction cycles:

- To comply with international environmental standards, the use of low-GWP and zero-ODP refrigerants ought to be given top priority (Bhatti *et al.*, 2024).
- To increase coefficient of performance (COP) and decrease energy destruction, LNG producers should investigate hybrid refrigeration cycles that include inorganic and organic refrigerants (Qyyum, Qadeer, Ahmad and Lee, 2020).
- To lower specific power usage, waste heat recovery and cooling aided by renewable energy sources should be explored.

- To guarantee operational safety, ammonia-based refrigerants need improved leak detection systems and regulatory compliance methods (Soujoudi and Manteufel, 2021).
- To reduce emissions from energy-intensive refrigeration cycles, carbon capture and storage (CCS) technology should be included in future liquefaction processes.
- Creating precooling technologies with minimal energy requirements will help cut down on carbon emissions related to the manufacture of LNG (Wang, He and He, 2024).

By addressing these issues, the LNG sector may move towards more environmentally friendly and effective liquefaction cycles, supporting international initiatives to lower greenhouse gas emissions and advance sustainable energy sources.

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