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## Supersonic separation towards sustainable gas removal and carbon capture

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### ABSTRACT

Carbon capture and storage is recognized as one of the most promising solutions to mitigate climate change. Compared to conventional separation technologies, supersonic separation is considered a new generation of technology for gas separation and carbon capture thanks to its advantages of cleaning and efficient processes which are achieved using energy conversion in supersonic flows. The supersonic separation works on two principles which both occur in supersonic flows: the energy conversion to generate microdroplets and supersonic swirling flows to remove the generated droplets. This review seeks to offer a detailed examination of the cutting-edge technology for gas separation and carbon dioxide removal in the new-generation supersonic separation technology, which plays a role in carbon capture and storage. The evaluation discusses the design, performance, financial feasibility, and practical uses of supersonic separators, emphasizing the most recent progress in the industry. Theoretical analysis, experiments, and numerical simulations are reviewed to examine in detail the advances in the nucleation and condensation characteristics and the mechanisms of supersonic separation, as well as new applications of this technology including the liquefaction of natural gas. We also provide the perspective of the challenges and opportunities for further development of supersonic separation. This survey contributes to an improved understanding of sustainable gas removal and carbon capture by using the new-generation supersonic separation technology to mitigate climate change.

### 1. Introduction

Due to the need to improve energy efficiency and promote environmental sustainability, there has been an increased worldwide emphasis on natural gas as a cleaner energy alternative. Natural gas, acknowledged by the European Union as an eco-friendly option with reduced emissions in comparison to coal and oil, has played a key role in tackling the energy scarcity issue. Nevertheless, effective natural gas processing is still crucial for cutting emissions and securing the sustainable growth of the gas sector. Supersonic separators (SSs) have emerged as a promising technology for gas purification and carbon capture, offering advantages such as cleaner processing, simplicity,

reliability, safety, and cost-effectiveness [1]. These separators utilize converging-diverging nozzles to generate supersonic flows, resulting in lower temperatures during expansion and subsequent condensation of impurities [2].

As shown in Fig. 1, a typical supersonic separator comprises a swirl generator, a supersonic nozzle, and a diffuser [1, 74]. The supersonic nozzle induces low-pressure and low-temperature conditions, causing the phase change of CO<sub>2</sub> in supersonic flows. The swirl generator generates strong swirling flows with extensive centrifugal force (>500,000 g), facilitating the removal of droplets from the mixture. Shock waves are generated by the diffuser to decrease the flow velocity from supersonic to subsonic levels, which enhances the utilization of pressure energy, with a trade-off of some pressure reduction. In the context of CO<sub>2</sub>

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Nomenclature		Greek symbols	
<i>English</i>		$\alpha_r$	Heat transfer coefficient, W/(m <sup>2</sup> K)
$C_p, C_L$	Specific heat capacity for gas and liquid, (J/kg K)	$\Delta G$	Gibbs free energy (J)
$d$	Droplet diameter (m)	$\mu$	Dynamic viscosity (Pa s)
$dr/dt$	Droplet growth rate (m s <sup>-1</sup> )	$\gamma$	Specific heat ratio
$h_G, h_L$	Vapor and liquid enthalpy (Jkg <sup>-1</sup> )	$\rho_G, \rho_L$	Vapor and liquid density (kg/m <sup>3</sup> )
$J$	Nucleation rate (m <sup>-3</sup> s <sup>-1</sup> )	$\sigma_r$	Liquid surface tension (N/m)
$k_B$	Boltzmann constant	$\xi$	Function of temperature
$Kn$	Knudsen number	<i>Subscript</i>	
$L$	Latent heat (Jkg <sup>-1</sup> )	$c$	Critical
$l$	Mean free path of vapor molecules (m)	$G, L$	Vapor, liquid
$Ma$	Mach number	$s$	Saturation
$M_G, M_L$	Vapor and liquid mass (kg)	<i>Abbreviations</i>	
$p$	Pressure (Pa)	CCS	Carbon capture and storage
$p_s(T_G)$	Saturation pressure at $T_G$ (Pa)	CFD	Computational fluid dynamics
$q_c$	Condensation coefficient	CNT	Classical nucleation theory
$r$	Droplet radius (m)	EOS	Equations of state
$r^*$	Critical radius of droplets (m)	HCDPA	Hydrocarbon dew-point adjustment
$R$	Gas constant (J/kg K)	MD	Molecular dynamics
$S$	Supersaturation ratio	NG	Natural gas
$T_G, T_L$	Vapor and liquid temperature (K)	SRK	Soave-Redlich-Kwong
$T_s(p)$	Saturation temperature at $p$ (K)	SS	Supersonic separator
$t$	Time (s)	WDPA	Water dew-point adjustment
$v$	Velocity (m/s)		
$\omega$	Wetness fraction		

separation in flue gas containing N<sub>2</sub>, CO<sub>2</sub>, and water vapor, the optimized design of a supersonic separator ensures the occurrence of the phase change of CO<sub>2</sub> and water vapor in supersonic flows, with N<sub>2</sub> acting as the carrier gas [3].

This review provides a comprehensive overview of the current state of multiphase supersonic separation for natural gas purification. It highlights the key advances and challenges in the field. The insights gained from this review will be of interest to researchers and engineers working in the energy and combustion science field, as well as stakeholders in the natural gas industry.

Although the present study may suggest a predominant emphasis on natural gas cleaning, a technology recently reviewed by Cao and Bian [5], it's important to highlight that our current review article has been meticulously crafted to offer a comprehensive and thorough analysis of the subject matter. In this review, our efforts extend beyond the parallels with the work in Ref. [5]. **Firstly**, we have earnestly endeavoured to create a comprehensive and exhaustive review that delves into the core topic. Our article not only explores the intricacies of the supersonic separator mechanism in gas purification but also undertakes an in-depth examination of the broader landscape from technical study to economic

analysis. **Secondly**, our review encompasses a range of crucial aspects. Notably, we meticulously assess practical methods centred around gas purification and carbon dioxide removal, facilitating a comprehensive comparative analysis. **Furthermore**, we extend our scrutiny beyond the realm of mere thermodynamic evaluations. Our review incorporates a holistic approach, incorporating economic, environmental, and sustainability considerations, thereby painting a more complete picture of the subject. **Finally**, our review takes a stride towards innovation by exploring the advantages presented by gas liquefaction. We delve into the realm of gas liquefaction, highlighting the role of the supersonic separator in this context. Our article doesn't merely offer a cursory overview; instead, it endeavours to comprehensively compare and contrast the efficacy of the supersonic separator against alternative methodologies. **In essence**, while our article may share commonalities with the work in Ref. [1], it is meticulously tailored to offer a comprehensive and distinct review. By not only dissecting the supersonic separator mechanism but also providing a comprehensive exploration of practical methods, economic implications, and advancements like gas liquefaction, our review stands as a unique and valuable contribution to the subject matter at hand.

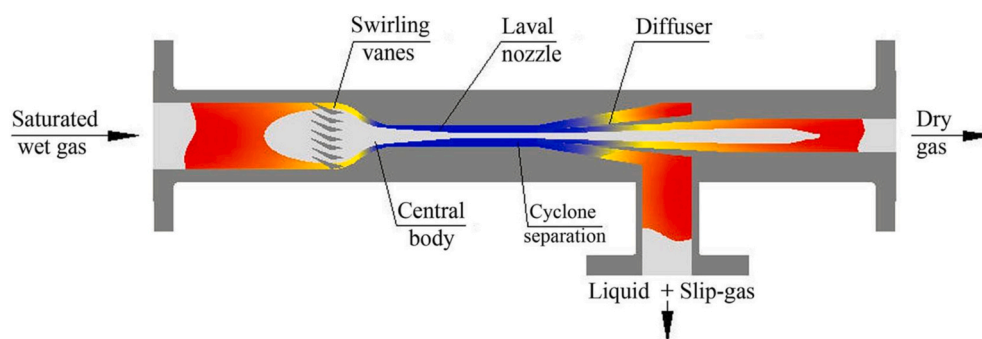


Fig. 1. A new concept of gas separation and CO<sub>2</sub> capture using energy conversion in supersonic flows [4] (Reprinted from Wen et al. [4], with permission from Elsevier).

Around 80% of the world's energy demands are currently met by power plants that rely on fossil fuels. Human actions have played a major role in boosting levels of greenhouse gases, leading to a one-degree Celsius uptick in Earth's average temperature from preindustrial times [6]. It is essential to control CO<sub>2</sub> emissions as these gases are the main cause of worldwide climate change. Carbon capture and storage (CCS) is becoming a popular method for addressing global warming [7]. The world's energy scene is changing significantly, with a greater focus on finding sustainable and eco-friendly energy options. The incorporation of CCS is seen as crucial in strategies to reduce climate change because it serves as a connection between conventional fossil fuels and sustainable energy options [8].

During the 1900s, the natural gas market saw major expansion and variety. Besides its usual role as a source of energy, new gas-to-liquid technology allows for the creation of various hydrocarbons, ranging from gasoline-like to Diesel-like substances. At present, unprocessed natural gas is obtained from three different types of wells: oil wells, gas wells, and condensate wells. Associated gas is the name commonly used for natural gas extracted from oil wells. This gas has the ability to exist on its own in the reservoir as free gas or to be mixed into the crude oil as dissolved gas. On the other hand, non-associated gas comes from wells that primarily produce gas and condensate without much or any crude oil. Gas wells normally produce just raw natural gas, whereas condensate wells yield both free natural gas and semi-liquid hydrocarbon condensates.

Regardless of where it comes from, natural gas is frequently discovered in combination with other hydrocarbons like ethane, propane, butane, and pentanes after being separated from crude oil. Moreover, unprocessed natural gas consists of a variety of other substances such as H<sub>2</sub>O, H<sub>2</sub>S, CO<sub>2</sub>, He, N<sub>2</sub>, among others. In this blend, natural gas liquids (NGLs) have substantial worth as byproducts of processing natural gas. NGLs, which include ethane, propane, butane, iso-butane, and natural gasoline, are sold individually and have various uses. These objectives consist of improving oil recovery in oil wells, serving as materials for oil refineries or petrochemical plants, and acting as sources of energy.

In the future, there are various possible methane sources, such as landfill gas, biogas, and methane hydrate [9]. Landfill gas is a specific form of biogas, with biogas typically being defined as gas produced from organic matter without being combined with additional waste. Certain areas currently use biogas, specifically landfill gas, but there is a considerable opportunity for more growth and use of this resource.

Advancements in technology have made it easier to explore remote locations, where efforts to meet the rising demand for natural gas are increasing. Nevertheless, the natural gas found in these reservoirs frequently includes high levels of impurities and heavy hydrocarbons. The gas that is taken out mainly contains methane, propane, and ethane.

Similar to petroleum, natural gas is a crucial component of the global hydrocarbon supply. However, natural gas directly obtained from the wellhead, while rich in methane, is not pure enough for various applications. To obtain pure methane and high-molecular-weight hydrocarbons suitable for diverse uses, a series of purification steps, known as gas processing or gas refining, are employed. These steps involve the separation of different hydrocarbons and fluids from the pure natural gas.

In addition, natural gases carry a certain concentration of water vapor, which increases with temperature or pressure [10]. The presence of water and the composition of the gases can lead to the formation of hydrates at different temperatures and pressures. It is crucial to develop techniques that minimize hydrate formation in order to ensure smooth operations and prevent potential issues.

Prior to transportation through gas pipelines, natural gas with impurities undergoes a drying and sweetening process. Dehydration is a crucial step during the gas pipeline gas-up phase to prevent the formation of hydrates and protect pipelines from fouling and corrosion.

Between 2002 and 2008, the Twister Bio-Engineering Company showcased the versatility of separators in applications ranging from gas

dehumidification to hydrocarbon condensation control and H<sub>2</sub>S elimination [11]. Building on this legacy, numerous studies have highlighted the efficacy of supersonic separators (SS) for CO<sub>2</sub> capture, dehydration, and liquefaction [12].

The supersonic separator's distinctive capability lies in its ability to achieve significantly lower temperatures during expansion. This is courtesy of the adiabatic cooling associated with the Laval nozzle. This unique feature enhances its effectiveness at extracting impurities from natural gas streams. The SS design has been thoroughly examined in scientific studies, delving into aspects such as its structural design, implementation methodologies, operational efficiency, economic viability, and potential industrial applications.

Supersonic separators stand out in several aspects compared to other purification technologies, such as adsorption, absorption, cryogenics, and membranes [13]. Unlike adsorption and absorption methods that often require substantial facilities, complex systems, and the use of chemicals with adverse environmental effects, supersonic separators offer a more streamlined and cost-effective solution. Due to their compact tube structure and the absence of rotating parts, the design of supersonic separators ensures stability and eliminates chemical discharge, making them environmentally friendly [3].

In summary, the comprehensive advantages offered by supersonic separators, coupled with their favorable comparison to other purification technologies in terms of cost-effectiveness, environmental impact, and operational flexibility, underscore their prominence as a promising and preferred technique for natural gas purification. The ongoing scientific exploration of their design and applications further solidifies the potential of supersonic separation in addressing the evolving needs of the natural gas industry. A summary of the reviewed literature, encompassing experimental work and numerical modeling of supersonic separators, is presented in Table 1.

A comprehensive review of the literature is presented in the subsequent chapters, organized as follows: Section 2 discusses the effect of heavy hydrocarbons, CO<sub>2</sub>, and other impurities on natural gas quality and utilization. Section 3 presents a review of the configuration and applications of supersonic separators. First, the type of design and its capability to separate gases are discussed. Then, its role in supersonic expansion and condensation is described. This section also includes a description of the mathematical modeling and simulation details, followed by a comparison with traditional separation techniques. Section 4 focuses on the applications of supersonic separation in natural gas purification. It places particular emphasis on the dehydration process, hydrocarbon and CO<sub>2</sub> removal processes, and studies related to natural gas liquefaction. In Section 5, the economic and environmental impact of supersonic separation is discussed. Lastly, Section 6 examines significant challenges and modifications to gas purification, supersonic separator simulations, the potential for further research and development, and the implications for the energy and combustion science field.

## 2. Characterization of natural gas impurities

### 2.1. Composition of impurities in natural gas

The increased need for natural gas has led to the finding of plentiful reserves in distant areas, facilitated by advancements in modern technology. During the extraction of natural gas from reservoirs, it frequently contains high amounts of pollutants and dense hydrocarbons [44]. The gas extracted is primarily made up of propane, methane, and ethane, making up most of the gas composition. Table 2 illustrates that natural gas is mainly made up of a combination of combustible hydrocarbons. Moreover, natural gases may also consist of nitrogen (N<sub>2</sub>), carbon dioxide (CO<sub>2</sub>), and hydrogen sulfide (H<sub>2</sub>S). There might also be trace quantities of argon, hydrogen, and helium [45].

Prior to being transported for further processing, natural gas with impurities undergoes a conditioning process, which includes drying and sweetening. The sweetening process is employed to remove hydrogen

**Table 1**  
Compilation of published works of supersonic separators.

Operational Fluid	Research Objectives	Year	Ref.
<b>Experimental studies</b>			
Natural gas (NG)	Gas Mixture Separation	1968	[14]
Water and ethanol/ propanol, Ethanol and propanol	Binary Mixture Condensation	2000	[15]
D <sub>2</sub> O and H <sub>2</sub> O	Binary Mixture Condensation	2002	[16]
Moist air	Dew Point Adjustment of Air	2005	[17]
Air and ethanol	Water and Ethanol Removal	2009	[18]
Air	Performance Exploration of SSSs under Various Conditions	2010	[19]
Moist air	Evaluation of SSSs' Dehumidification Performance	2011	[20, 21]
Water droplets and air	Investigation of Separation Efficiency	2014	[22]
Methane and Carbon dioxide	Carbon Dioxide Separation from Methane	2014	[23]
Ethanol/water and air	Condensation Mechanism Exploration in Supersonic Flow	2020	[24]
Argon and carbon dioxide	Homogeneous Nucleation of Carbon Dioxide in Argon Carrier Gas	2020	[25]
NG	Assessment of liquid film in a supersonic separator	2023	[1]
<b>Numerical modeling</b>			
NG	Formation and Expansion of Droplets: Multifaceted Modelin	2003	[26]
NG	Formation and Expansion of Droplets	2005	[27]
NG	Impact of Geometric Factors and Swirl on the Shockwave	2008	[28]
Air	Intense Swirling in a Laval Nozzle	2008	[29]
NG	Aid in Selecting an Appropriate Dehydration Method	2012	[30]
CO <sub>2</sub> + N <sub>2</sub> + O <sub>2</sub> + Ar + H <sub>2</sub> O	CO <sub>2</sub> Capture from Offshore Gas Turbines	2014	[31]
N <sub>2</sub> + water and H <sub>2</sub> O + CH <sub>4</sub>	Condensate Formation in Binary Mixtures	2015	[32]
NG	Relationship for Predicting the Speed of Sound in a Two-Phase System	2017	[33]
CH <sub>4</sub> , air, O <sub>2</sub> , CO <sub>2</sub> , SF <sub>6</sub>	Influence of Operational Parameters on Hydrodynamic Characteristics	2018	[34]
Ethane and methane	Predicting Nucleation by Investigating the Condensation Process	2018	[35]
NG	Influence of Inlet Operational Parameters on Condensation and Swirl Attributes	2019	[36]
Methane and air	Influence of the Inner Body's Geometry on the Shockwave Position	2019	[37]
Argon and carbon dioxide	Extensive Molecular Dynamics Simulations for Homogeneous Nucleation Study	2021	[38]
Water and methane	Explanation of Shock Wave Interaction with Boundary Layers and Discussion of Liquefaction Efficiency	2021	[39]
CO <sub>2</sub> -rich NG	Exergy Performance Comparison of Two Supersonic Gas Processing Alternatives with Conventional Methods	2021	[40]
Methane and water	Development of a New Eulerian-Lagrangian Method Coupled with Eulerian Wall Film Model to Enhance Separation Efficiency	2022	[41]
Methane and CO <sub>2</sub>	Proposal of a Mathematical Model for Predicting Phase Change	2023	[42]
Methane and water	Investigation of the Impact of Shock Wave/Boundary Layer Interaction on Condensation Flow	2023	[43]

sulfide and carbon dioxide from the natural gas stream.

## 2.2. Effect of heavy hydrocarbons, CO<sub>2</sub>, and other impurities on natural gas quality and utilization

The quality and usage of natural gas can be impacted by heavy hydrocarbons, carbon dioxide (CO<sub>2</sub>), and other impurities present in it. Ethane, propane, and butane, which are heavy hydrocarbons, contain

**Table 2**  
Components and sources of natural gas (Reprinted from Speight [45], with permission from Elsevier).

Component	Vol. %
Methane (CH <sub>4</sub> )	>85
Ethane (C <sub>2</sub> H <sub>6</sub> )	3–8
Propane (C <sub>3</sub> H <sub>8</sub> )	1–5
Butane (C <sub>4</sub> H <sub>10</sub> )	1–2
Pentane (C <sub>5</sub> H <sub>12</sub> )	1–5
Carbon dioxide (CO <sub>2</sub> )	1–2
Hydrogen sulfide (H <sub>2</sub> S)	1–2
Nitrogen (N <sub>2</sub> )	1–5
Helium (He)	<0.5

more energy per volume than methane, the main constituent of natural gas. The gas benefits from their presence as it boosts the overall energy content, which is particularly useful for applications needing greater heating values. Nevertheless, the combustion efficiency of natural gas can also be impacted by the presence of heavy hydrocarbons and impurities. Extra processing or treatment may be needed for these components to guarantee thorough combustion and prevent the creation of dangerous byproducts. In addition, contaminants such as heavy hydrocarbons and CO<sub>2</sub> can decrease the heating value of natural gas. The decrease in heating value can affect how efficiently appliances and equipment that use natural gas for heating function. Hence, the quality and usage of natural gas may be impacted by the existence of these impurities, necessitating the implementation of suitable procedures to enhance its burning effectiveness and heating capacity for different uses.

CO<sub>2</sub> and other impurities found in natural gas can lead to corrosion and harm pipelines during the transportation process. To maintain the durability and lifespan of pipeline infrastructure, extra steps may be required to eliminate or reduce these contaminants. Moreover, CO<sub>2</sub> plays a crucial role as a greenhouse gas in the process of global climate change. Rising levels of CO<sub>2</sub> from human activities are causing worry about the growing amount of greenhouse gases in the atmosphere [46]. The world's CO<sub>2</sub> levels play a direct role in causing climate change and result in increased global temperatures [47]. CO<sub>2</sub> in natural gas also contributes to its overall carbon footprint. Prioritizing efforts to reduce CO<sub>2</sub> emissions from natural gas is crucial, which can be achieved through implementing CCS or other mitigation technologies. These measures are essential in reducing the environmental effects of natural gas and addressing climate change [48].

Extra processing steps are needed to separate and purify natural gas due to heavy hydrocarbons and impurities, leading to complexity and increased costs for upstream processing facilities, ultimately affecting operational efficiency and economics. The effect of heavy hydrocarbons, CO<sub>2</sub>, and other impurities on natural gas quality and utilization can differ based on the specific application and regulatory standards. Continual attempts are being made to improve gas processing methods and create technologies to address these obstacles and enhance the overall efficiency and usage of natural gas.

In emerging economies, challenges like intermittent power distribution and costs still persist despite the increasing reliance on renewable energy for the future [49,50]. Compared to oil and coal, natural gas (NG) emits less carbon due to its higher hydrogen-to-carbon (H/C) ratio. As a result, the sector is working towards substituting carbon-powered systems with updated, eco-friendly options that have a lower carbon footprint. Natural gas continues to be a dependable option for energy in the medium term. Nevertheless, a notable problem emerges with 15–80% of verified NG reserves containing CO<sub>2</sub>, requiring creative approaches to exploration and extraction [51].

## 2.3. Importance of removing impurities in natural gas for efficient combustion and energy conversion

Fuel emissions play a crucial role in the offshore oil and gas industry

due to the processing and transportation of hydrocarbons on gas and oil platforms [52]. The majority of emissions from these platforms stem from gas-fired power generation [53]. Khesghi and Prince [54] as well as Xu et al. [55], conducted research on the CO<sub>2</sub> emissions associated with ethanol fermentation.

Water vapor is frequently included in natural gas streams due to changes in temperature and pressure. Corrosion in the presence of moisture is expedited by the existence of carbon dioxide and hydrogen sulfide in natural gas. In addition, hydrates have the potential to reduce flow capacity and create blockages within pipelines. Having water in a gas pipeline can cause slug flow and decrease efficiency. Moreover, particles and dense hydrocarbons can build up on heat transfer surfaces, reducing their efficiency. This leads to decreased rates of heat transfer and system performance in general. Cleaning natural gas is essential for keeping heat transfer surfaces clean, optimizing energy conversion, and increasing system productivity.

Removing impurities from natural gas and maintaining affordable electricity prices presents a major obstacle, particularly in light of the growing effects of climate change due to emissions such as CO<sub>2</sub>. Natural gas needs to go through various processing steps to become a dry, completely gaseous fuel appropriate for transportation and delivery through pipelines. Yet, impurities found in natural gas, like solids, moisture, and corrosive materials, may cause harm to combustion devices like burners, valves, and heat exchangers. By removing these impurities effectively, the lifespan of equipment can be prolonged, decreasing maintenance requirements and minimizing downtime.

During a standard natural gas processing procedure, various steps are included such as getting rid of hydrogen sulfide (H<sub>2</sub>S), dehydrating, modifying the water dew-point (WDPA) with propane, adjusting the hydrocarbon dew-point (HCDPA) with heavy hydrocarbons (C<sub>3</sub><sup>+</sup>), and removing carbon dioxide [56]. Sulfur compounds impurities can interrupt combustion by producing sulfur oxides (SO<sub>x</sub>) in the process. These substances add to air quality issues and are capable of corroding machinery. By efficiently eliminating sulfur compounds and other contaminants, the combustion process can be enhanced, leading to better combustion efficiency and decreased emissions.

Some impurities, like volatile organic compounds (VOCs) and hazardous air pollutants (HAPs), can lead to air pollution and present dangers to human health and the environment. Cleaning natural gas by removing impurities reduces the release of harmful substances, leading to more environmentally friendly and long-lasting energy conversion methods. In certain cases, natural gas is used as a raw material for catalytic processes in industries like refineries or petrochemical plants. Impurities in the gas flow can make catalysts less effective and reduce process efficiency. Ensuring the protection and longevity of catalysts is achieved by purifying natural gas, which optimizes the conversion processes.

Through the injection of CO<sub>2</sub> into oil fields, CO<sub>2</sub> can be removed from natural gas (NG) as well as used for enhanced oil recovery (EOR) [57]. In a related context, Reis et al. [58] conducted optimization studies on membrane permeation to address both bulk CO<sub>2</sub> removal and EOR requirements, considering time-dependent CO<sub>2</sub> contents to facilitate the absorption of final polishing chemicals. In a subsequent study, Reis et al. [59] further improved membrane permeation (MP) by considering time-varying CO<sub>2</sub> concentrations to overcome limitations associated with the exposure of bulk CO<sub>2</sub> removal, thereby allowing for the absorption of polishing chemicals. Anwar et al. [60] cited innovative CO<sub>2</sub> mitigation approaches such as algal CO<sub>2</sub> capture, nanotechnology CO<sub>2</sub> capture, and biochar CO<sub>2</sub> capture, among others.

In summary, the removal of impurities from natural gas plays a vital role in achieving efficient combustion, safeguarding equipment, enhancing heat transfer efficiency, controlling emissions, and maintaining the effectiveness of catalysts. These purification processes contribute to improved energy conversion, reduced environmental impact, and enhanced overall system performance.

### 3. Supersonic separation techniques

#### 3.1. Basic principle and design of supersonic separators

Supersonic separators (SS) are compact devices (as shown in Fig. 1) equipped with convergent-divergent Laval nozzles and stationary vanes at the inlet to generate swirls. Their refrigeration effect surpasses that of expanders, vortex tubes, and Joule-Thomson (J-T) devices [61]. One notable advantage of supersonic separators is the prevention of hydrate formation due to the short residence time, eliminating the need for regeneration and inhibitors. This process is environmentally friendly. Simulation studies of supersonic separators require a comprehensive approach that considers the formulation of the problem and incorporates all factors influencing performance. Designing or understanding the performance and operating conditions of supersonic separators involves making several decisions. Various variables must be taken into account, such as the number of phases, the equation of state, the type of shockwave, the nozzle geometry, and the working fluid selection. A good starting point is to identify the system and delve into the underlying theories and research methods.

The utilization of a Laval nozzle in the supersonic separator enables adiabatic cooling, resulting in significantly low temperatures during the separation phase. This capability enhances the extraction of impurities [62]. The separation performance and design of supersonic separators have been extensively investigated through scientific studies.

Microgravity-operated phase separators can be classified into stationary and centrifugal types. A static device, without moving parts, is more reliable for unmanned operations on a platform than one with moving parts. Particles are separated by their wetting properties [63]. Despite their short lifespan, static separators offer advantages such as simplicity, reliability, and low power consumption. However, excessive moisture can increase the presence of microorganisms, making the devices more susceptible to malfunctions. The hydrophobic-hydrophilic separator is a static separator that separates hydrophilic from hydrophobic materials by exploiting water's reversible attraction and repulsion. However, hydrophobic materials eventually lose their capacity to repel and trap particles. On the other hand, centrifugal separators require more power but have a longer operational life. These devices increase the centrifugal separation of condensed phases by converting axial velocity into angular velocity [64]. Spinning wings or vanes can be found either before or after the nozzle's throat in centrifugal separators.

Supersonic separators offer several advantages over conventional technologies. Some of these key advantages include.

- Compact size: Supersonic separators are relatively small in size, requiring less space for installation compared to traditional separators. This compactness makes them more convenient for transportation and handling.
- Cost-effective: The installation of supersonic separators is generally more affordable compared to larger and more complex separation systems. This cost-effectiveness makes them a favorable choice for various applications.
- Lower operating costs: Supersonic separators typically have lower operating costs due to their simplified design and lower energy consumption. These devices are efficient in separating heavy components like C<sub>3</sub><sup>+</sup> from the natural gas flow, leading to cost savings in the long run.
- Environmental friendliness: The use of supersonic separators contributes to environmental sustainability. These devices aid in the removal of impurities and heavy components from the gas stream, resulting in cleaner and more environmentally friendly gas for utilization.

Overall, supersonic separators provide benefits in terms of space efficiency, transportation, cost-effectiveness, lower operating expenses, and environmental considerations. These advantages make them an

attractive option for various industries and applications.

In a study conducted by Shoostari and Shahsavand [65], the impact of the diffuser angle on the location of normal shocks, pressure recovery coefficients, and shock onset positions was investigated. The researchers found that altering the divergence angle on the Laval nozzle did not have any effect on the temperature or pressure prior to the occurrence of the shock. However, by selecting an incline of  $8^\circ$ , they were able to achieve a pressure recovery coefficient of 0.88 with minimal energy loss. Moreover, the optimal value for the diverging section remained constant within the range of 290–310 K and 6–10 MPa, regardless of the initial gas temperature and pressure.

### 3.2. Converging-diverging nozzle and its role in supersonic expansion and condensation

A supersonic nozzle serves as a crucial component in the design and optimization of supersonic separators, playing a pivotal role in accelerating fluid to supersonic velocities by converting pressure and temperature into kinetic energy [66]. Supersonic separators, designed for the separation of gas and liquid phases in multiphase flows, offer distinct advantages, including reduced weight, smaller size, heightened efficiency, and lower maintenance costs compared to conventional separators [1].

The design and optimization of a supersonic nozzle tailored for specific separations involve careful consideration of various factors, including inlet and outlet conditions, fluid properties, nozzle shape, shock wave structure, and reliability requirements [67]. The selection and optimization of nozzle type and geometry play a crucial role in achieving efficient particle separation. Key details in this process include:

Converging-Diverging Nozzle (De Laval Nozzle) widely used nozzle type consists of a converging section followed by a diverging section, accelerating flow to sonic or supersonic speeds. For subsonic to sonic flow applications, a simpler converging nozzle may be employed. The shape of the converging section influences acceleration speed and efficiency, while the diverging section is critical for maintaining supersonic flow and preventing shock waves. Strategies for optimization involve tailoring the nozzle to achieve a specific Mach number at the exit, controlling shock waves, adjusting the divergence angle, optimizing throat dimensions, ensuring uniform flow distribution, and considering particle characteristics. Matching the nozzle design to the fluid properties, density, and size of particles is essential for efficient separation. Balancing nozzle dimensions with varying inlet conditions and particle characteristics is crucial for operational flexibility in separator design. Achieving the desired Mach number and pressure ratio at the exit is paramount for efficient separation, while avoiding flow separation, shock wave boundary layer interaction, and flow instability is essential for maintaining performance and reliability. The effects of fluid properties such as compressibility, viscosity, density, and phase change on flow behavior and separation efficiency must be accounted for in nozzle design. Moreover, historical contributions to the field by researchers like Oswatitsch [68] and recent studies by Wyslouzil and colleagues [69,70] have further advanced the understanding of supersonic nozzle principles.

Fig. 2 presents a schematic of a supersonic nozzle where a vapor-carrier gas mixture enters the nozzle with known temperatures, total pressures, and partial pressures of the condensable gas [71]. As the gas expands and cools through the nozzle, the supersaturation ratio ( $S$ ) rapidly increases. When the supersaturation exceeds a critical value ( $S_{crit}$ ) on a microsecond timescale, phase transition occurs. It has been observed that when phase transition occurs in a gas mixture, the temperatures and pressures are higher compared to the expansion of the same gas mixture without condensation, which can be attributed to isentropic expansion [72].

A De Laval nozzle is divided into three parts based on its speed: the subsonic, sonic, and supersonic sections. Following these sections are

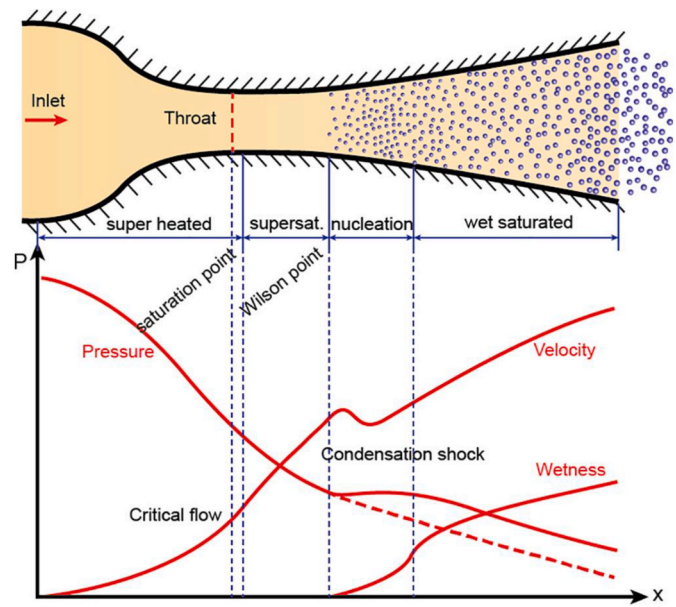


Fig. 2. Schematic of pressure, velocity and saturation treatment for condensation phenomenon in a supersonic nozzle [71] (Reprinted from Zhang et al. [71], with permission from Elsevier).

the liquid collector and the diffuser. The thermodynamic properties of a nozzle change along its axial length, making the placement of the nozzle an important aspect of simulations. However, each design of a nozzle may have a different geometry. The geometry of the nozzle is often determined by a series of converging-diverging angles and equations. Modeling nozzles is best done by establishing a universal positional relationship so that it can be applied to nozzles with different geometries [73].

In the Laval nozzle, condensable species undergo liquefaction due to the rapid temperature drop that occurs during the expansion of the fluid to supersonic speeds. The flow characteristics can be described using the Mach Number ( $Ma$ ), which is the ratio of the axial flow velocity ( $v$ ) to the sound speed property ( $c$ ) of the multiphase fluid. The flow initially starts as subsonic ( $Ma < 1$ ) in the Laval converging section, where the nozzle cross-section steadily decreases. As the flow approaches the nozzle throat, it reaches sound speed ( $Ma = 1$ ), and the cross-sectional area of the nozzle is at its minimum. This point is known as the maximum constriction. Beyond the throat, the flow rapidly expands in the Laval diverging section, resulting in an increasing cross-sectional area of the nozzle. In this section, the flow becomes supersonic ( $Ma > 1$ ) and accelerates to very high speeds.

The Laval diverging section can experience an irreversible normal shock adiabatic transition, which is a metastable phenomenon, when supersonic flow passes through it. In this case, the supersonic flow undergoes a transformation into subsonic flow, leading to increased entropy, pressure, and temperature while maintaining the same energy, momentum, and mass flow rate. To prevent the loss of separation due to re-evaporation, it is crucial to collect the Laval condensate upstream of the shock. Once the shock occurs, the subsonic flow recovers its temperature and pressure and continues through the SS exit.

In the SS process, the removal of the condensate from the supersonic flow results in an irreversible transition known as the SS shock. The outlet pressure is always lower than the inlet pressure, even during isentropic compression or expansion steps. The current state of the art in SS is represented by thermodynamic SS, as described by de Medeiros et al. [74], and the computational fluid dynamics (CFD) model developed by Wen et al. [4]. Yang et al. [75] have further expanded on these studies using the same nozzle design, representing the current state of the art in the SS literature.

A supersonic swirling separator offers several advantages. It operates without any dynamic parts, chemical additives, or human intervention [76]. Additionally, the high flow velocity of the device minimizes fouling or solid deposition, eliminating the need for cryogenic cooling units. The natural cooling effect in the device allows it to reach temperatures of up to 60 °C. Moreover, this apparatus can be used in various environments, including land, sea, and deep water applications [77].

### 3.3. Experiments in supersonic separation

There are three general types of research approaches [78]: experimental setup methods, thermodynamic methods, and computational fluid dynamics (CFD). Many academic studies have delved into these three topics concerning supersonic separations of natural gas, exploring experimental setups, thermodynamic aspects, and using CFD for modeling. Each approach contributes valuable insights and provides a comprehensive understanding of the phenomenon. In this section, we critically assess and draw conclusions from experimental results, offering a comprehensive overview of supersonic separator research advancements.

Examining the impact of reflux channels, drainage structures, and operational conditions on separation performance, one study found that using materials with minimal roughness and increasing pressure ratios improved the consistency between experimental and numerical simulations. Notably, reflux channels enhanced flow fields and separation performance. Additionally, innovative cylindrical drainage structures were identified as effective at mitigating the interaction between shock waves and boundary layers. This fostered a more balanced temperature field in the nozzle [79].

Another study examined the dehydration performance of an integrated cyclone front supersonic separator. By analyzing the impact of the pressure recovery coefficient on dew point depression and swirl strength on mass flow rates, it was concluded that the designed separator demonstrated high adaptability to variable mass flow rates and exhibited efficient dehydration of natural gas. This aligns with the broader notion that supersonic separators excel at natural gas dehydration and heavy hydrocarbon removal [80].

A supersonic separator with tilted blades at the entrance and a swirling stabilizer plus a nozzle was also designed in another study. This research supports the idea that supersonic separators are efficient at removing natural gas hydration and heavy hydrocarbons [81].

Investigating the particle paths and separation efficiency of a specific device, an experimental study employed the discrete particle method. The study highlighted the importance of a proposed annular nozzle, achieving a robust rotating flow field conducive to separation efficiency above 95%. The congruence between numerical and laboratory findings underscored the accuracy and stability of the discrete particle method in evaluating dehumidification characteristics [21].

In a comprehensive three-dimensional numerical and experimental study of air hydrodynamic behavior in supersonic separators, insights into shockwave locations were revealed. The study demonstrated that improving dehumidification performance is achievable by manipulating inlet and outlet parameters, such as increasing inlet pressure, decreasing inlet temperature, and elevating air humidity [15].

Another study explored water and ethanol vapor condensation using varying amounts of nitrogen. The investigation delved into the effects of carrier gas pressure on condensation onset, offering valuable information on the role of pressure in condensation processes within supersonic separators [82].

In a novel approach, an experimental study concluded that cyclone separator efficiency could be significantly improved by combining specific components. The optimized design, featuring a constant flow element, a leaf mill element, and a folding plate element, achieved a separation efficiency exceeding 95%. This finding holds significance in enhancing the efficiency and applicability of gas-liquid separators, particularly under challenging small flow conditions [83]. Another

experimental study investigated the homogeneous nucleation of carbon dioxide through ultrasonic nozzles. It explored nucleation processes, particle size distribution, and aerosol number density using a variety of experimental methods to gain a deeper understanding of fundamental processes in supersonic separators [25].

A practical investigation of various nozzle geometries led to the identification of an optimal nozzle shape for particle separation. Several geometries performed better than others at certain NPRs, including the triangular shape, while the conical shape performed better at low NPRs. Based on this research [84], practical guidance was provided for selecting nozzle geometries that maximize particle separation and pressure recovery. In summary, these studies collectively contribute valuable insights into supersonic separators, spanning structural improvements, dehydration performance, condensation processes, and nozzle geometries. These findings collectively advance the understanding of supersonic separator technology, offering a foundation for further innovations in natural gas purification and related applications.

### 3.4. Comparing of various simulation approaches

In complex systems, such as supersonic separation, where gas mixtures are separated at high pressures and speeds, simulation is extremely useful. However, there are different simulation approaches, each with its own advantages and disadvantages [78]. Here is a brief overview of some of the common simulation approaches and their pros and cons:

A thermodynamic model describes the behavior of a system using the principles of thermodynamics, including temperature, pressure, density, and composition. The focus of thermodynamic studies is on simplifying flow behavior, nucleation, and hydrodynamics to accurately depict thermodynamics. As multiphase flow equations become increasingly complex, these models require more rigorous calculations in order to understand phase equilibrium. A thermodynamic model is relatively simple and quick to implement, and can provide insight into the thermodynamic feasibility and efficiency of the separation process [85]; However, thermodynamic models do not account for fluid dynamics and transport phenomena in the system, such as turbulence, shock waves, diffusion, and heat transfer. Therefore, thermodynamic models may not be accurate enough to capture the detailed performance and design of the system [86]. Supersonic separation technology incorporates the principles of fluid dynamics, thermodynamics, and aerodynamics to achieve a breakthrough gas conditioning process [4]. Supersonic separation technology incorporates the principles of fluid dynamics, thermodynamics, and aerodynamics to achieve a breakthrough gas conditioning process [87]. Castier [88] developed a numerical simulation method using the Peng-Robinson equations to model supersonic separators. This method focuses on determining the thermodynamic parameters of the diverging-converging nozzle, including the speed of sound and phase equilibrium conditions for non-ideal multiphase systems. While this method does not provide detailed two-dimensional or three-dimensional parameter profiles like CFD models, it yields reliable results concerning shockwave position, phase behavior, and one-dimensional characteristics.

A CFD study, on the other hand, focuses on modeling multidimensional hydrodynamics more accurately, but often at the expense of simplifying thermodynamics. For simulations of the flow and thermal behavior of the system, numerical methods are used to solve the Navier-Stokes equations and other equations governing fluid mechanics and heat transfer. The CFD models can provide detailed information about the spatial and temporal distribution of gas mixture variables, such as velocity, pressure, temperature, and concentration [43]. Additionally, CFD models can simulate shock waves, turbulence, chemical reactions, and phase transitions in the system [89]. Wen et al. [20] utilized numerical simulations to investigate the distribution of parameters such as static temperature and tangential speed in supersonic separators. They highlighted the significant non-uniformity of radial distributions of dynamic gas properties, which has an impact on the separation



efficiency of components. Based on the conservation of angular momentum, they designed an ultrasonic rotary separator. Their analysis of the flow fields of natural gas with the presence of a shock wave revealed that the low temperature, high centrifugal field, and supersonic speed of the nozzle diffuser effectively separate water and heavy hydrocarbons from natural gas. The position of the shock wave determines the temperature distribution, which affects the re-evaporation and separation of liquid droplets. If a shock wave occurs in the separation part, it generates a high temperature that leads to droplet re-evaporation, hindering the gas-liquid separation process.

These studies highlight the importance of incorporating various factors such as viscous layer effects, condensation phenomena, and vorticity in modeling and understanding the behavior of two-phase flows and the performance of nozzles in supersonic separators. Numerical simulations offer valuable understanding of the intricate dynamics of such systems, aiding researchers in enhancing performance through optimized design and operation parameters. The issue lies in the fact that CFD models demand significant computational resources and time to execute. Moreover, the accuracy of CFD models depends on the numerical methods, mesh resolution, boundary conditions, and initial conditions. Therefore, CFD models may not be feasible or reliable for large-scale or long-term simulations [79].

Hybrid models combine different types of models to create a more comprehensive and efficient simulation of the system. Hybrid models can simulate the flow and temperature behavior of a system using an in-depth thermodynamic model and an in-depth computational fluid dynamics model, for instance Ref. [90]. By combining strengths and limitations of each model type, hybrid models can provide a balance between accuracy and computational cost. Developing and validating hybrid models, however, can be more complex and time-consuming, requiring a good understanding of and integration of the different types of models. Due to this, hybrid models may not be available or applicable to all systems and scenarios.

Optimal modeling strategies for supersonic separation depend on simulation objectives and constraints. An objective of evaluating the thermodynamic feasibility and efficiency of a separation process may justify the use of a thermodynamic model. An optimal design and operation of the system may be achieved through CFD modeling. Having both goals in mind may make a hybrid model the best choice. The choice of modeling strategy is also influenced by the model availability, computing resources and time, as well as results accuracy and reliability. Hence, optimal modeling strategies vary from case to case, and various factors may need to be taken into account.

#### 3.4.1. Computational fluid dynamics simulation for supersonic separation

Over the past two decades, thermodynamics and CFD have been used extensively to study gas separation using supersonic nozzles. CFD solvers, however, are still underdeveloped and struggle to capture complex transition behaviors and changes that occur at supersonic speeds. This is due to variations in fluid density and isothermal compressibility [91]. These difficulties arise from variations in fluid density and isothermal compressibility. Despite these challenges, CFD remains an essential tool, particularly in addressing critical design issues such as swirling motion and flow vane interaction [78].

#### 3.4.2. Limitations of simplified models in supersonic separators

The application of simplified models and idealized conditions in the study of supersonic separators provides computational advantages but introduces several limitations. This discussion highlights key constraints associated with these models and conditions in the context of supersonic separators.

The intricate high-speed expansion and condensation processes in supersonic separators challenge the adequacy of one-dimensional (1D) simulations [92]. To address this, researchers turn to two-dimensional (2D) and three-dimensional (3D) simulations, recognizing that factors like separator geometry, fluid properties, and operating conditions

intricately influence subsonic, transonic, and supersonic flows. Simplified geometries may fail to accurately represent a supersonic separator's complexities, leading to inaccurate predictions of flow patterns, shock wave interactions, and separation behaviors.

Simplified models frequently assume steady-state conditions, neglecting transient effects. While extending one-dimensional models to include steady-state supersonic separation with side streams, Castier [92] explored the influence of additional streams on nozzle dynamics. In reality, supersonic flows can exhibit unsteady behaviors, challenging the validity of assuming steady-state conditions. Idealized conditions often assume isothermal behavior, disregarding temperature variations. Supersonic flows typically involve significant temperature changes, and overlooking these effects can lead to inaccuracies, particularly in predicting shock wave properties.

Supersonic flows can have phase changes, such as vaporization or condensation, that affect separation dynamics [93,94]. However, some simplified models and idealized conditions may assume that the flow is single-phase, which means that the flow consists of only one phase, either gas or liquid. This may overlook the multiphase interactions that occur in the flow. To account for the multiphase interactions, some researchers use a Euler-Euler model, which models the gas and liquid phases as two interpenetrating continua, or a Euler-Lagrange model, which models the gas phase as a continuum and the liquid droplets as discrete particles, as shown in Fig. 3. For example, Matsuo et al. [95] used a 2D Euler-Euler model with viscous effects to simulate the behavior of two-phase flows in supersonic separators. They considered the interaction between the gas and liquid phases and the influence of viscous effects. In contrast, some CFD studies use a Euler-Lagrange model to track the behavior of droplets in the system [41]. However, to simplify the modeling process and reduce the computational cost, some studies use a simpler working fluid instead of natural gas, which has multiple components. For example, some CFD studies use pure air, methane, water, or a combination of these substances as the working fluid [32,96]. This may limit the applicability of the results to real-world scenarios.

Simplified models often simplify or limit boundary conditions, neglecting the impact of surface roughness, external disturbances, and inlet conditions. Salikaev and Gunnerov [97] investigated the influence of intake temperature, pressure, composition, and pressure drop on supersonic gas separator flow, emphasizing the importance of considering various factors in boundary conditions. Many simulations assume ideal gas behavior, disregarding compressibility effects. Jassim et al. [28] studied the impact of actual gas flow and nozzle shape on highly pressurized natural gas, highlighting inaccuracies in predicting flow fields due to ideal gas assumptions. The nozzle design was found to affect shock wave locations, emphasizing the need to consider actual gas behavior. Simpson and White [98] focused on condensation phenomena in nozzles but acknowledged that simplified models may not accurately capture turbulent effects. Neglecting turbulence in supersonic flows can lead to inaccurate predictions of mixing and separation, emphasizing the importance of accounting for turbulent behaviors.

In the pursuit of insights into the behavior of multiphase fluids within supersonic separators, researchers must navigate the complexities involved in simulation studies. The decisions made in selecting working fluids, nozzle geometry, and simulation software are crucial. Fig. 4 provides a comprehensive illustration, serving as a valuable guide for researchers throughout the simulation process [86]. Even though simplified models provide computational efficiency, their limitations, such as neglecting realistic geometry, steady-state assumptions, isothermal behavior, single-phase simplifications, limited boundary conditions, and ideal gas assumptions, highlight the difficulty of accurately representing the complex dynamics of supersonic separators. Designing and interpreting simulation studies in the field requires careful consideration of these limitations.

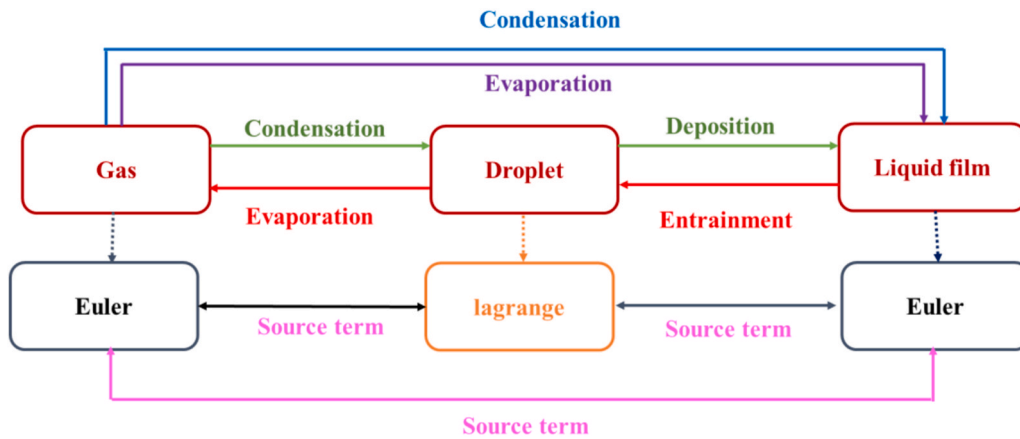


Fig. 3. Two-phase flow and the numerical simulation method [41] (Reprinted from Ding et al. [41], an open access article from Elsevier).

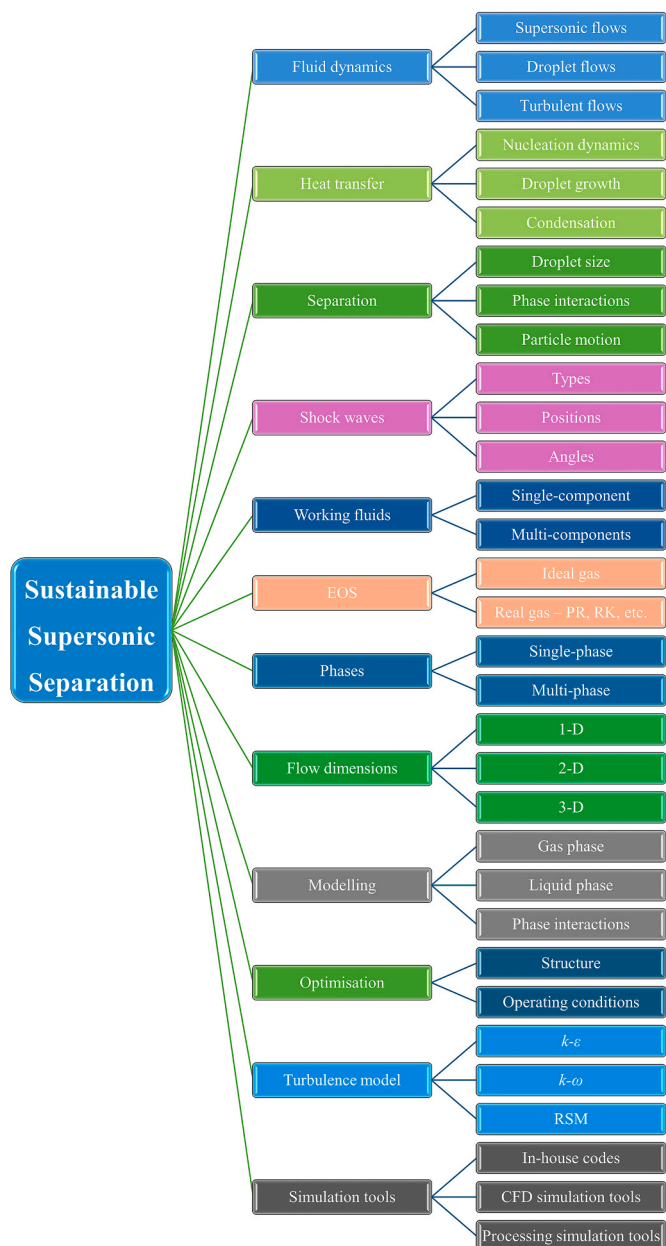


Fig. 4. Key considerations for supersonic separation.

### 3.4.3. Equation of state

When addressing pressurized gases, the use of ideal gas equations of state tends to introduce significant calculation errors, escalating up to 500% compared to a mere 2–3% at atmospheric pressure [99]. The deviation of real gas properties from the ideal gas law amplifies exponentially with pressure and temperature, displaying considerable variability based on gas compositions. To mitigate this deviation, a correction factor known as the gas compressibility factor is introduced into the ideal gas equation [100]. Various correlations have been proposed for the gas compressibility factor, expressed in terms of pseudo-reduced pressure or pseudo-reduced temperature, such as the Hankinson-Thomas-Phillips correlation [101] and the Hall-Yarborough equation of state [102]. In this regard, Arina [103] investigated the behavior of CO<sub>2</sub> as a supercritical fluid in a converging-diverging nozzle using the Redlich-Kwong, Carnahan-Starling-De Santis, and van der Waals equations of state. Their findings suggested that all three equations of state (EOS) provided reasonably accurate predictions of gas behavior in the supercritical fluid flow through the nozzle.

The SRKV (Soave-Redlich-Kwong) equation is known for its ability to account for the non-ideal behavior of real gases under specific conditions [104]. Additionally, to accurately predict thermodynamic properties of fluids, a real fluid equation of state is essential, especially in high-pressure and low-temperature conditions, considering factors like residual thermodynamic sound speed, residual enthalpy, and residual entropy. These additional factors play a significant role in accurately predicting the thermodynamic properties of the fluid under study. The use of cubic equations of state is prevalent for estimating vapor pressure, partial vapor pressure, and vapor-liquid equilibrium, providing a convenient framework for analyzing fluid behavior in different phases, as follows [28]:

$$p = \frac{RT}{V-b} - \frac{a\alpha(T)}{V(V+b)} \text{ with } \alpha(T) = \left(1 + (0.480 + 1.574\omega - 0.176\omega^2) \left(1 - \sqrt{T/T_c}\right)\right)^2 \quad (1)$$

where  $p$ ,  $T$ ,  $V$ , and  $R$  are pressure, temperature, volume, and gas constant, respectively.  $a$  and  $b$  are model parameters.  $\alpha(T)$  represents the cohesion factor which is a function of the acentric factor ( $\omega$ ), temperature, and the temperature at the critical point  $T_c$ .

To calculate the thermodynamic speed of sound in a multiphase fluid within supersonic separators, various approaches have been proposed. Nichita et al. [105], Firoozabadi and Pan [106], and Castier [107] have developed methods considering multiple phases to determine the speed of sound in complex systems. Secchi et al. [108] introduced a technique specific to multicomponent phases using the GERG (Groupe Européen de Recherches Gazières) equation of state, enabling the calculation of

thermodynamic sound velocity in multicomponent systems while considering interactions between different components. NIST REFPROP provides three equations of state for gas mixture calculations: GERG-2008 [109], AGA8 [110], and Peng-Robinson [111]. Despite the availability of these options, the Peng-Robinson equation is deemed less accurate and not recommended for general use in REFPROP, while the AGA8 equation is cautioned against, especially in the liquid phase or near the critical point. NIST REFPROP encourages the use of its default GERG equations of state, which, although more complex, exhibit lower uncertainties than the standard GERG-2008 equations developed by Kunz and Wagner [109].

The GERG-2008 equation of state, an extension of GERG-2004 [112], covers 21 natural gas components, offering wide applicability across temperature, pressure, and composition ranges, including gas phase, liquid phase, supercritical region, and vapor-liquid equilibrium states. The GERG-2008 equation of state is valid over a range of 90–450 K and up to 35 MPa, with an extended range of 60–700 K and up to 70 MPa. It accurately represents experimental binary and multicomponent data for gas-phase and gas-like supercritical densities, sound speeds, and enthalpy differences, making it suitable for diverse technical applications such as pipeline transport, natural gas storage, liquefied natural gas processes, and separation processes [99]. Despite its commendable accuracy, areas of improvement are acknowledged, such as limited available vapor-liquid equilibrium data, particularly for mixtures of CH<sub>4</sub> and C<sub>4</sub>H<sub>10</sub> at low temperatures. To utilize the GERG-2008 equation of state for mixtures, the following mixing rules are employed:

$$\frac{1}{\rho_r(\bar{x})} = \sum_{i=1}^n x_i^2 \frac{1}{\rho_{c,i}} + \sum_{i=1}^{n-1} \sum_{j=i+1}^n 2x_i x_j \beta_{v,ij} \gamma_{v,ij} \frac{x_i + x_j}{\beta_{v,ij}^2 x_i + x_j} \frac{1}{8} \left( \frac{1}{\rho_{c,i}^3} + \frac{1}{\rho_{c,j}^3} \right) \quad (2)$$

$$T_r(\bar{x}) = \sum_{i=1}^n x_i^2 T_{c,i} + \sum_{i=1}^{n-1} \sum_{j=i+1}^n 2x_i x_j \beta_{T,ij} \gamma_{T,ij} \frac{x_i + x_j}{\beta_{T,ij}^2 x_i + x_j} (T_{c,i} T_{c,j})^{0.5} \quad (3)$$

In these relationships,  $\rho_{c,i}$  and  $T_{c,i}$  represent the critical density and critical temperature of component  $i$ , while the four binary parameters  $\beta_{v,ij}$ ,  $\gamma_{v,ij}$ ,  $\beta_{T,ij}$ , and  $\gamma_{T,ij}$  are adjusted based on the binary mixtures data [113].

Comparing GERG-2008 with the Peng-Robinson equation of state,

studies by Baladao and Fernandes [114] indicate superior results for GERG-2008 in calculating pressure and density for various mixtures, although it requires a longer computational time due to iterative calculations for the vapor and liquid phases in the vapor-liquid equilibrium calculation. The GERG-2008 equation of state is expressed in a dimensionless reduced form, incorporating the Helmholtz free energy for the ideal gas mixture, residual Helmholtz free energy, and binary specific and generalized departure functions. Mixing rules are applied to adapt the GERG-2008 equation of state to mixtures, with consideration of the Michelsen-Kistenmacher syndrome.

#### 3.4.4. Simulation of phase transition in supersonic separator

In fast-expanding fluids, some regions reach saturation while the majority remains unsaturated. Gas condensation is not a simple equilibrium process [115]. In the flow of natural gas through a supersonic separator, a non-equilibrium phase transition occurs along with supersonic flow and mass transfer of condensing gases. After the rapid expansion of natural gas in the Laval nozzle, spontaneous condensation takes place due to the gas being supersaturated, leading to the formation of condensation nuclei and subsequent droplet growth.

The process of spontaneous condensation in supersonic settings involves two main stages: nucleation and droplet growth [116,117]. Fig. 5 illustrates the operational approach employed in the condensation model [71], providing an overview of the simulation methodology used in studying this phenomenon.

#### 3.4.5. Supersaturation phenomenon

Supersaturation serves as a measure of how far a fluid's properties deviate from thermodynamic equilibrium, represented by the saturation line. When steam flows through a nozzle, a phase change occurs, transitioning supercritical steam into subcooled water [118]. This phase transition involves spinodal decomposition, producing dry steam and liquid water from wet steam. It also involves phase separation, taking place in the unstable region depicted in Fig. 6. Spinodal decomposition occurs when a fluid enters the spinodal region of a phase diagram. In the case of wet steam within the metastable zone, between the binodal and spinodal lines, homogeneous nucleation occurs [119].

This phenomenon involves the formation of small droplets from supersaturated vapor. This initiates the condensation process and the

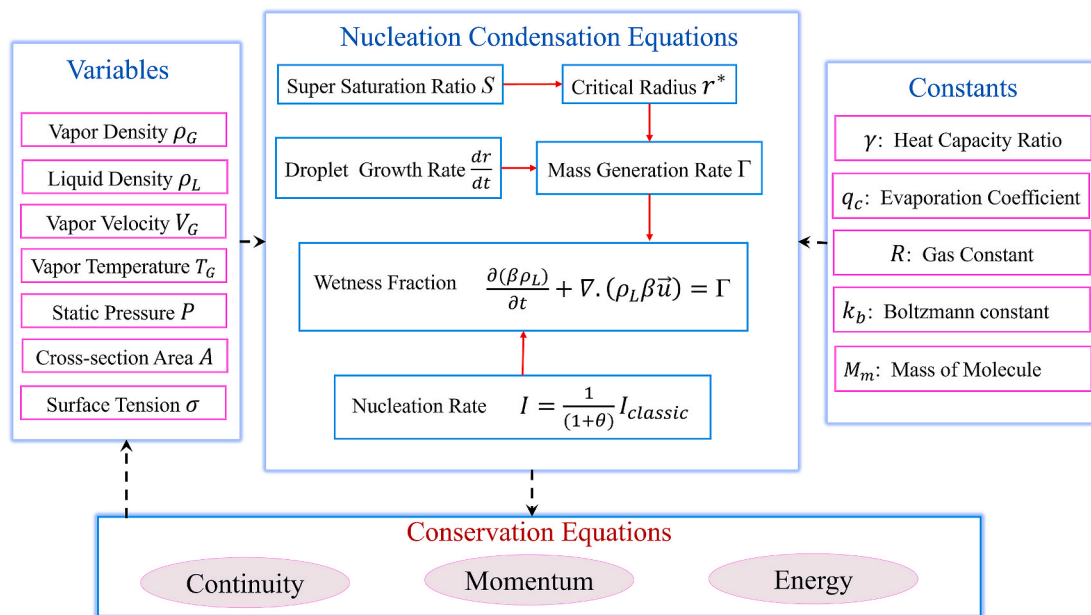


Fig. 5. Strategy for implementing the condensation model [71] (Reprinted and edited from Zhang et al. [71], with permission from Elsevier).

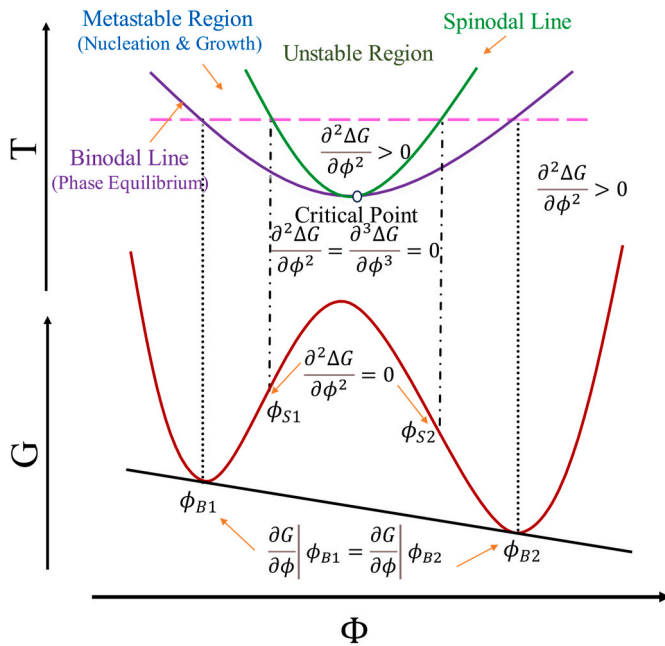


Fig. 6. Fluid transition path along the spinodal and binodal lines, metastable, and unstable regions [119] (Reprinted from Ochi et al. [119], an open access article from IOP Science).

subsequent phase transition from supercritical steam to subcooled water. Measurement of this non-equilibrium process poses significant challenges, and interpreting metastable thermodynamic properties remains a significant challenge. It is defined by Brazhkin [120] that metastable phases are non-equilibrium states of matter with reversible properties throughout an experiment. Metastable regions on phase diagrams, located between the spinodal line and phase boundary, can have a finite fluctuation that can make a solution unstable. It is this fluctuation, known as a nucleus, and how much energy is expended in creating such a nucleus that determines the phase's metastability. The theoretical calculations of this energy value show a decrement towards zero closer to the spinodal line. In the analysis of the macroscopic system's behavior, Kaplun and Meshalkin [121] find no fundamental difference between stable and metastable states, except for a limited lifetime in metastable states.

Several studies have investigated non-equilibrium condensation in high-velocity flows. Gyarmathy [122] and Duff [123] conducted research on condensation within supersonic nozzles, using water vapor, nitrogen, and CO<sub>2</sub>. In these studies, static pressure measurements were used to determine the onset of condensation. Condensation caused a pressure decrease in the converging section and an increase in the diverging section due to the energy released during condensation. It was demonstrated by Duff [123] that static pressure measurements in CO<sub>2</sub> can be used to detect condensation at 0.1% moisture content. Although the study lacked experimental data to complement the numerical calculations, Baltadjiev et al. [124] suggested mixed results could be possible away from the critical point of the gas.

### 3.4.6. Modeling supersonic condensation shockwaves

The accuracy of predicting water vapor condensation in supersonic flows with shock waves is intricately tied to the Gas Spontaneous Nucleation Rate Model [125]. This model dictates the rate of liquid droplet formation from the supersaturated gas phase and relies on thermodynamic state, flow properties, and system geometry. Modifying this model to better capture the phase transition process has the potential to significantly enhance prediction accuracy. Another pivotal factor influencing accuracy in simulating shock wave interactions and phase change processes in supersonic flow is the choice of the numerical

method and computational grid [126]. The numerical method must adeptly capture shock wave structures, boundary layer separation, vortex formation, and droplet dynamics [127].

Employing advanced shock-capturing schemes, such as Weighted Essentially Non-Oscillatory (WENO) schemes, can prove beneficial. Additionally, ensuring the computational grid is sufficiently fine to resolve flow features and droplet size distribution is imperative. Validation of numerical simulations against experimental data is essential for establishing accuracy and reliability. The type and composition of the gas mixture represent a third influential factor. Different natural gases can exert varying effects on fluid flow and thermodynamics. For instance, the CO<sub>2</sub>-CH<sub>4</sub> mixture may undergo supersonic condensation and swirling separation, impacting separation efficiency [126]. A comprehensive understanding necessitates a multi-component simulation to study the concentration effects of different components on flow and phase change. Experimental validation or simulation refinements in these areas are essential for advancing the understanding of supersonic separation processes.

### 3.4.7. Classical nucleation theory

In experiments, the formation of liquid droplets has been demonstrated not to occur under saturated conditions during rapid expansion [124]. Rather, under subcooled conditions, nucleation is induced within high-speed flows, overcoming the energy potential. The system then returns to near equilibrium conditions through the spontaneous condensation of the fluid.

The nucleation-driven condensation is initially preferred, leading to the creation of the first droplets of the liquid phase [128]. The subsequent phase transition is then governed by the growth of supercritical droplets, or droplets larger than a critical radius, denoted as  $r^*$ , which effectively suppresses nucleation, re-establishing equilibrium.

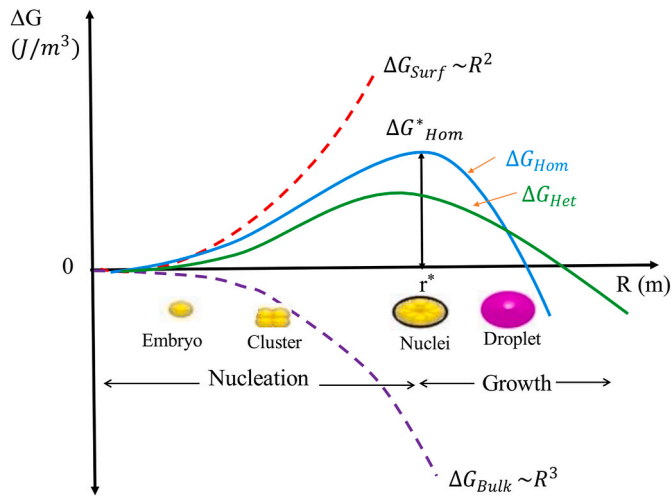
The Wilson line is a characteristic of the condensing vapor highly dependent on the expansion rate. Higher expansion rates result in a deeper excursion into the metastable region, shifting the Wilson line towards regions of higher subcooling [129]. Supersonic nozzles are favorable for studying these phenomena as the rate of expansion and nucleation can be altered by varying the length of the orifices while maintaining the same pressure ratio.

The thermodynamic non-equilibrium states of gas mixtures can be induced by drastic changes in temperature and pressure [130]. To restore thermodynamic equilibrium, nucleation within the gas mixture must lead to the growth of detectable-size droplets. The onset of nucleation, crystallization, droplet growth measurement, and determining the Wilson line or spinodal line can be studied using laser light transmission and scattering, such as shearing interferometer [131,132].

Spontaneous nucleation in supersonic nozzle flow condensation heavily relies on supersaturation [133]. Supersaturation determines the ability of a flow to form new nuclei. When the flow approaches saturation, vapor molecules do not instantly condense due to the presence of a free energy barrier (the sum of volume and surface free energies, as depicted in Fig. 7). Instead, they continue to expand as superheated steam. However, at a specific degree of supersaturation, the critical radius is reached, causing a predetermined number of condensation nuclei of a certain size to form in the steam [134]. Higher degrees of supersaturation lead to a faster rate of nuclei formation [135]. The intrinsic and average kinetic energies of vapor molecules are related to the likelihood of nucleation occurring. The rate of nucleation formation can be characterized by Gibbs free energy, which has specific dimensions [136]:

$$\Delta G = 4\pi r^2 \sigma_r - \frac{4}{3}\pi r^3 \rho_L R T_G \ln(S) \quad (4)$$

The liquid phase is denoted by the subscript  $L$ , while the gas phase is represented by the subscript  $G$ . The droplet radius is indicated by  $r$  is the droplet radius, surface tension by  $\sigma_r$ , droplet density by  $\rho_L$ , gas temperature by  $T_G$ , and supersaturation ratio by  $S$ .



**Fig. 7.** Radius of particles and the changes of Gibbs free energy. The figure also includes schematic representations of the nucleation stages, showing the reversible states of the embryo and cluster and the irreversible state of the nucleation. After the nuclei phase, the droplets start to grow [137] (Reprinted from Taqieddin et al. [137], an open access article from IOP Science).

Calculating the maximum free energy relative to the droplet radius makes it possible to calculate the critical radius in the nucleation process [138].

$$r^* = \frac{2\sigma_r}{\rho_L RT_G \ln(S)} \quad (5)$$

In this equation,  $r^*$  is the cluster's critical radius. A droplet with a radius smaller than the critical radius evaporates, while a droplet larger than the critical radius grows [139]. It is possible to estimate the rate of condensation and nucleation of supercooled vapor using the required radius. Two-phase flows form droplets based on their nucleation rates under supersaturation conditions. The nucleation rate can be calculated using the following formula [140]:

$$I = I_0 e^{-G_b} \quad (6)$$

There are various expressions and corrections for  $I_0$  and the exponential function in the relation (4).  $I_{\text{class}}$  is known as a basis for calculating the rate of classical nucleation (CNT):

$$I_{\text{class}} = \frac{\rho_G^2}{\rho_L} \sqrt{\left(\frac{2\sigma_r}{\pi M^3}\right)} \exp\left[-\frac{\Delta G^*}{KT_G}\right] \quad (7)$$

$\rho_G$  represents the vapor density, Boltzmann's constant by  $K$ , and molecule mass by  $M$  in this equation.

### 3.4.8. MD nucleation simulation

In the realm of supersonic gas separation techniques, unraveling the intricacies of non-equilibrium condensation, particularly in the nanoscale regime, is paramount for advancing our comprehension of processes like nucleation and droplet growth. Nucleation, the initial step in condensation, is significant in quantifying the condensation process. This is crucial for applications such as controlling CO<sub>2</sub> liquefaction [141]. However, despite experimental efforts, quantitative measurement of the nucleation rate of CO<sub>2</sub> remains a challenge due to limitations and discrepancies between experimental results and classical theoretical predictions [25].

In recent years, molecular dynamics (MD) simulations have emerged as a powerful tool to bridge the gap in our understanding of nucleation processes [142]. These simulations provide researchers with a microscopic view, allowing them to monitor the evolution of nonequilibrium dynamics over time [143]. There has been considerable research

focused on H<sub>2</sub>O models [144] and Lennard-Jones fluids [145] using MD simulations, but there is little research on CO<sub>2</sub> nucleation [38,146]. Understanding the nucleation process of CO<sub>2</sub> becomes crucial for the application of supersonic gas separation technology in carbon capture and storage CCS. With MD simulations, nucleation rates can be calculated using methods such as average first-pass times or thresholds [5]. Nucleation, however, is a stochastic process. Nucleation rate predictions require multiple independent MD simulations in order to gain comprehensive insights.

Extensive research has been conducted on supersonic separators, which promise to remove CO<sub>2</sub> from natural gas. It remains unclear, however, whether classical nucleation theory (CNT) is applicable to CO<sub>2</sub> condensation mechanisms in natural gas. Using a CH<sub>4</sub>/CO<sub>2</sub> mixture gas, recent studies have investigated the condensation characteristics of CO<sub>2</sub> in natural gas using CFD and MD simulations [147]. The investigation uncovered crucial insights into CO<sub>2</sub> nucleation and growth pathways at the molecular scale. A Laval nozzle creates conditions that facilitate CO<sub>2</sub> liquefaction at low temperatures. Condensation conditions can be optimized by manipulating the inlet temperature and pressure. A MD simulation of the nucleation stage revealed a complex interaction between CO<sub>2</sub> gas molecules, latent heat release, and cluster stability influenced by energy interactions with surrounding molecules. The study revealed a substantial deviation between the CNT results and MD simulations by orders of magnitude, emphasizing the need for corrections to the classical theory.

In conclusion, molecular simulations, particularly MD simulations, stand as a cornerstone in unraveling the complexities of non-equilibrium condensation in supersonic separators at the nanoscale. Aside from providing a microscopic understanding of nucleation processes, these simulations also provide a theoretical reference for optimizing the separation effect of CO<sub>2</sub>. We will undoubtedly contribute significantly to the advancement of supersonic gas separation and its applications in carbon capture and storage, by integrating various findings, correcting classical theories, and refining simulation techniques.

### 3.4.9. Droplet growth rate

During actual droplet growth, a lot of molecules surround the condensation nucleation [148]. As the molecules of vapor condense on the condensation nuclei's surfaces, the droplets keep growing [149]. The vapor molecules release latent heat into the surrounding gas, which is a normal mechanism for transferring heat and mass. Slip velocity between vapors and liquids is often ignored since liquid droplets have tiny formation diameters, on the order of nanometers. The Knudsen number is the ratio of the vapor molecules' mean free path to the diameter of the droplet [5]:

$$Kn = \frac{l}{2r} \quad (8)$$

This formula shows the interaction between droplets and vapor molecules; In Eq. (6) mean free length of vapor molecule ( $l$ ) is defined as:

$$l = 1.5\mu \sqrt{RT_G/p} \quad (9)$$

With a modest value of  $Kn$  and a large droplet diameter, the continuous flow model can be used to compute. A high  $Kn$  means the droplet diameter is smaller than the distance between vapor molecules, so the free molecular flow model can find the flow field.

Condensation is thought to occur only on the surface of existing droplets after the nucleation zone. One of the continuous droplet growth model was developed by Gyarmathy to simulate homogeneous condensation [150]. In the model, heat and mass are transmitted, capillary influence is considered, and vapor molecules are diffused through the surrounding media. A droplet's energy balance is written as follows [151]:

$$L \frac{dm_r}{dt} = m_r C_L \frac{dT_L}{dt} + 4\pi r^2 \frac{p}{\sqrt{2\pi R T_G}} \left( \frac{\gamma + 1}{2\gamma} \right) C_p (T_L - T_G) \quad (10)$$

$C_p$ ,  $C_L$ , represent the specific heat capacities in the vapor and liquid phases, respectively, and  $\gamma$  denotes the thermal conductivity of the vapor.

Eq. (7) shows that the condensation of molecules gives the droplet energy that can either be transferred back into the vapor or increase its temperature [152]. In many wet steam calculations, due to the smallness of the droplets, its thermal inertia can be ignored, so equation (7) is simplified as follows:

$$L \frac{dm_r}{dt} = 4\pi r^2 \frac{p}{\sqrt{2\pi R T_G}} \left( \frac{\gamma + 1}{2\gamma} \right) C_p (T_L - T_G) = 4\pi r^2 \alpha_r (T_L - T_G) \quad (11)$$

Gayrathy has presented a relationship in the following form to calculate the temperature of the droplet despite the change in surface tension coefficient and enthalpy of evaporation between  $T_L$  and  $T_G$ :

$$T_L = T_G + \left[ 1 - \frac{r^*}{r} \right] (T_s(p) - T_G) \quad (12)$$

where  $T_s(P)$  is the saturation temperature corresponding to the vapor pressure. New droplets do not form after nucleation, and condensation only occurs on the surfaces of existing droplets [153]. Mass and energy are exchanged between buds and their surroundings, and the buds absorb molecules. A variety of analysts have studied the droplet in growth rate so far. By combining the two relations (8 and (9), the following expression for the growth rate of the droplet is obtained [154]:

$$\frac{dr}{dt} = \alpha_r \frac{(T_L - T_G)}{\rho_L L} \quad (13)$$

In this equation,  $L$  is the latent heat of vaporization at the saturation temperature corresponding to the vapor pressure.  $\alpha_r$  is also the convective coefficient of heat transfer between a droplet and its environment (steam) and is expressed as the following relationship [155]:

$$\alpha_r = \frac{\lambda}{r(1 + 3.18Kn)} \quad (14)$$

The enthalpy ( $h$ ) and density ( $\rho$ ) of the mixture are calculated using the wetness fraction ( $w$ ) as follows [156]:

$$h = wh_L + (1 - w)h_G \quad (15)$$

$$\frac{1}{\rho} = \frac{w}{\rho_L} + \frac{(1 - w)}{\rho_G} \quad (16)$$

Further details on wet flow equations can be found in Ref. [155].

The application of pressure boundary conditions at the intake and exit of the supersonic separator, along with no-slip and adiabatic boundary conditions at the walls, is a common practice in numerical simulations of supersonic separation [157]. These boundary conditions help define the behavior of the fluid and ensure accurate representation of the system.

### 3.5. Factors influencing the efficiency of supersonic separators

The effectiveness of supersonic separators hinges on various factors, including inlet temperature, pressure, flow velocity, and impurity concentrations. These parameters impact fluid dynamics, thermodynamics, phase transitions, and droplet formation.

The separation efficiency of supersonic separators is notably affected by the inlet temperature. This parameter influences the saturation pressure and temperature of the gas mixture, determining the degree of supersaturation and the onset of condensation. A higher inlet temperature can postpone condensation, potentially diminishing separation efficiency. The optimum inlet temperature range is contingent on gas mixture type, composition, operating pressure, and separator design. Notably, an enhanced supersonic separator with a diversion cone is

recommended to operate within the temperature range of 300–320 K [158], although variations may exist for different separators and gas mixtures.

Inlet pressure plays a pivotal role in density and velocity modulation of the gas mixture, impacting expansion and shock wave formation in the nozzle [159]. Elevated inlet pressure may amplify the expansion ratio, consequently enhancing separation efficiency. The optimal inlet pressure range is subject to gas mixture characteristics, operating pressure, and separator design. For an improved supersonic separator with a diversion cone, the suggested inlet pressure range is 400–600 kPa [158]. However, this range may vary for different types of supersonic separators and gas mixtures.

Flow velocity is a critical parameter significantly influencing the separation efficiency of supersonic separators. The interaction between flow velocity, shock waves, expansion fans, and other flow features plays a decisive role in determining separation efficiency. Research by Liu and Ding [159] reveals that an increase in inlet port number and gas-liquid area ratio leads to a decrease in separation efficiency. Senfter et al. [160] further report that high inlet volume flow rates enhance particle separation but also result in higher pressure drops. Recordings indicate separation efficiencies ranging from 26.92% to 38.56%, accompanied by pressure drop variations between 0.218 bar and 0.413 bar.

Higher flow velocities in supersonic separators induce stronger shock waves and increased kinetic energy, potentially optimizing separation efficiency through efficient phase separation. An optimal range of flow velocities is crucial for effective operation, dependent on separator design, fluid characteristics, and separation objectives. The interaction of shock waves with the fluid stream necessitates meticulous design of critical components such as nozzles and diffusers, directly influencing overall separation efficiency. Optimization of the overall geometry, including diverging and converging sections, is essential to accommodate desired flow velocities for efficient separation. Certain supersonic separators offer adjustable operational ranges to cater to variations in flow conditions, enabling optimization for specific applications and fluid characteristics. However, trade-offs may exist between higher flow velocities and considerations such as energy consumption, equipment wear, and maintenance, requiring a delicate balance.

Recognizing that the optimal flow velocity range varies based on specific design, intended application, and substance characteristics, experimental studies, numerical simulations, and prototype testing are commonly employed to determine the most effective flow conditions for a given supersonic separator.

### 3.6. Comparison with traditional separation techniques

Various techniques can be used to separate impurities. Several factors need to be taken into account when choosing a specific sweetening process. These factors encompass: Types of impurities to be eliminated, such as  $H_2S$ , and  $CO_2$ , acid gas concentrations at the inlet and outlet of the process, gas flow rate, temperature, and pressure parameters, environmental considerations and compliance requirements, and evaluation of relative economics for the chosen process. As shown in Fig. 8, these techniques include chemical, physical, or hybrid absorption, adsorption, membrane separation, or a combination thereof [161].

With membrane separation technology [162], gas components are selectively passed from one end of the membrane to the other. On one side of the membrane barrier, a large partial pressure of the essential components maintains a concentration gradient [163]. This novel process relies heavily on membrane materials. An appropriate membrane material should have high permeability and selectivity, along with superior mechanical strength and chemical stability [164].

In the scenario of high-pressure natural gas (NG), it undergoes a process where it is introduced into a membrane unit. Within this unit, water vapor is effectively extracted through the membrane, resulting in a dehumidified gas remaining in the retentate [165]. The permeate, which consists of the separated water vapor, is subsequently

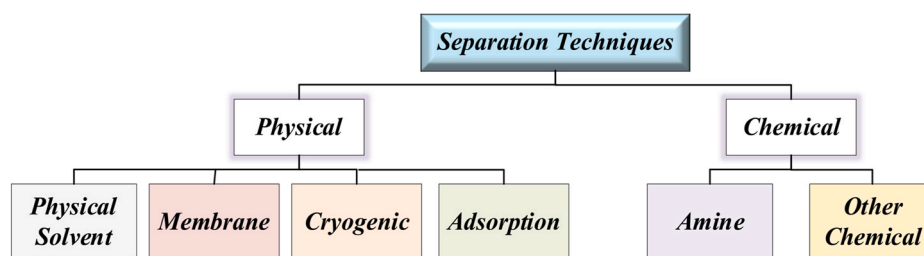


Fig. 8. The most commonly used sweetening processes.

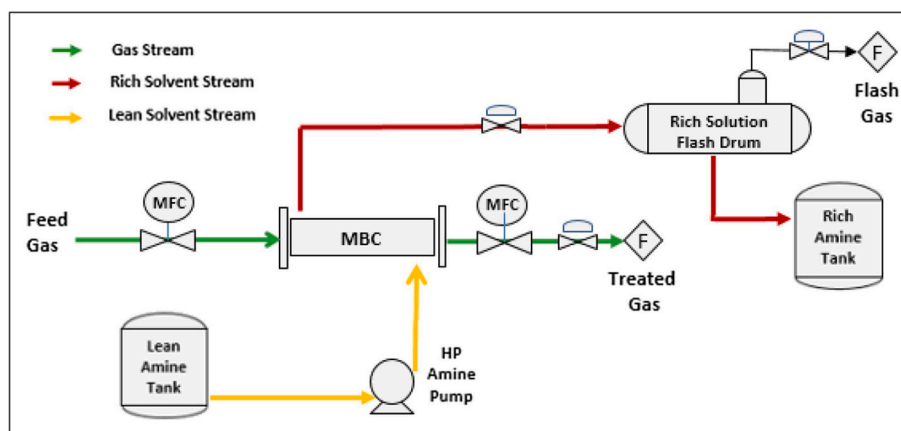


Fig. 9. Schematic of the pilot-plant setup for CO<sub>2</sub> removal from natural gas using high pressure membrane contactors [166] (Reprinted from Quek et al. [166], with permission from Elsevier).

recompressed. After isolating the condensate, it is combined with the initial gas flow to complete the process. In contrast, when dealing with low-pressure NG, the gas flow first undergoes compression. Following compression, the condensed water is separated from the gas using a separator. The gas, now free of the condensate, is then introduced into the membrane block. The permeate obtained from the membrane block is subsequently mixed with the original gas flow to conclude the process. A schematic of CO<sub>2</sub> removal from natural gas using high pressure membrane contactors is shown in Figure 9 [166].

The absorption technique relies on the idea that different gases dissolve to different extents in liquids [167,168]. This method could involve the chemical reaction that takes place during gas purification. The absorbing material, crucial for absorption processes, must possess both high absorption capabilities and thermal stability. Using a liquid desiccant contactor-regeneration process is a common way of drying natural gas in the gas industry. In this method, illustrated in Fig. 10 [30], the wet gas is exposed to a dry solvent with minimal water content. The liquid absorbs the water from the gas, leading to a concentrated liquid stream and a dehydrated gas stream. Before being recycled back to the first column for water removal from the feed gas, the solvent is regenerated in a second column [169]. Triethylene glycol is the most commonly used absorbent in the gas industry, followed by calcium chloride, ethylene glycol, diethylene glycol, and tetraethylene glycol. Because of their high hygroscopic nature, low vapor pressure, high boiling points, and low solubility in natural gas, glycols have been shown to be the most efficient liquid desiccants currently in use [170]. Because of its excellent ability to lower the dew point, cost-effectiveness, and reliability in operation, TEG has been widely accepted as the most economical glycol. Nevertheless, glycol dehydrators experience several operational problems. Contaminants in glycol solutions can come from suspended foreign matter, while the formation of decomposition products can occur due to overheating the solutions. The formation of foam in the solution can also lead to the transfer of liquid. Finally, environmental issues linked to fugitive emissions are being addressed through

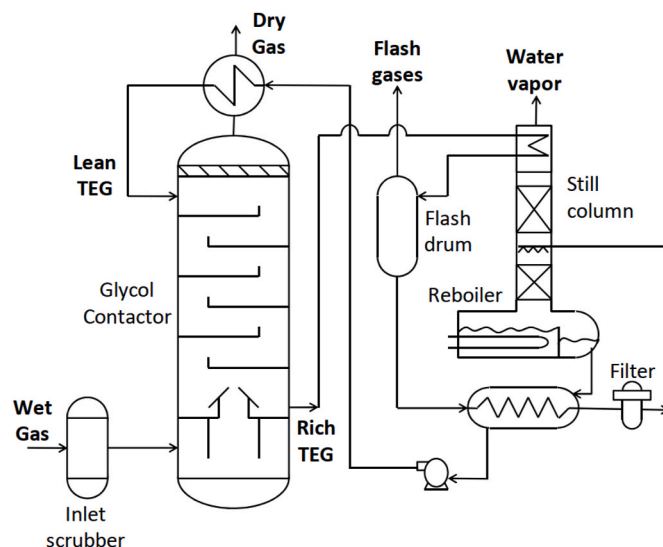


Fig. 10. Schematic of industrial absorption dehydration process using TEG [30] (Reprinted from Netušil Ditl [30], an open access book from IntechOpen).

efforts to minimize their impact. In addition to WDPA, NG contains liquids (NGL) that are typically removed to meet hydrocarbon dew-point specifications (HCDPA). In the majority of cases, NGL has greater value as separate products, and cryogenic processing, despite being a costly alternative, is the preferred method for separating NGL. Hydrocarbon dew-point of natural gas is operationally significant, and HCDPA is a quality criterion for gas sales. The extraction of NGL results in a decrease in the heating value of the gas product, which can reduce its market value. HCDPA specifications are typically met by low-temperature separation.

Adsorption is a type of mass transfer on solid surfaces [171]. Molecular attraction or chemical bonding is responsible for the adsorption gas molecules on porous solid surfaces. Chemical or physical processes can be used, depending on the surface forces, to achieve a very low concentration. Activated alumina, silica gel, and a molecular sieve are common solid adsorbents in the gas industry. The adsorption process of water molecules is dependent on the gas pressure and temperature, where higher pressure enhances adsorption, while higher temperature reduces it. These factors are carefully considered during the design of process parameters. To ensure continuous operation, a minimum of two bed systems is employed, with one bed dedicated to gas drying while the other undergoes regeneration [30]. Regeneration is accomplished using either preheated gas or a portion of the dehydrated natural gas, as depicted in Fig. 11 [30].

Scientists have shown that supersonic separators are more energy-efficient than conventional natural gas separators [39]. In comparison with traditional post-combustion capture technologies, supersonic separation offers several advantages, including a simple mechanism, simple equipment design, without moving parts, ease of maintenance, and no emissions [42]. Fig. 12 illustrates the configuration of a supersonic dehydration line [61]. A novel supersonic separator has been proven to be helpful in many gas conditioning applications, like dehumidifying and extracting heavy hydrocarbons from natural gases.

Dew points for water and heavy hydrocarbons need to be corrected for proper transportation and economics. In addition to their capability to separate carbon dioxide and hydrogen sulfide from natural gas, SS units offer versatility in various applications such as the production of liquefied natural gas (LNG), hydrogen generation, biogas upgrading, controlling industrial emissions, and enhancing processes in petrochemical refining. Refrigeration, membranes, adsorption, and absorption are traditional methods for correcting the dew point of water and hydrocarbons [172]. Processes like these usually cost a lot and require substantial facilities, including complex systems and plenty of chemicals with adverse effects on the environment [44]. Natural gas streams can be purified using supersonic separators by removing a variety of impurities. The lack of moving parts makes it incredibly reliable, up to 99% [173]. As compared to other conventional separators, the SS can accept a single-phase gas stream as input. With adiabatic expansion, two or more phases can form when water or heavy hydrocarbon species condense at low temperatures. After proper pressure recovery, this phenomenon can result in the gas and liquid phases separating, and a single stream of gas will leave the SS unit.

With its compact tube structure, the supersonic separator is extremely stable, low in space and weight, and is composed of no rotating parts. Due to the fact that this type of separator does not require any chemicals to discharge pollution, it is an environmentally friendly

device [174].

#### 4. Applications of supersonic separation in natural gas purification

The supersonic separator comprises several key components, including the swirling device, de Laval nozzle, cyclonic separator, and diffuser extension. Natural gas is released from high-pressure, low-velocity reservoirs. Its low temperature and pressure result from expansion to supersonic speeds in the Laval nozzle, causing the natural gas to drop below its dew point. Unwanted substances condense into liquid and are then separated by centrifugation before being collected in separate streams. Droplets of liquid form when water vapor and a heavy hydrocarbon component combine. The cyclone separates the liquid from the gas by centrifuging the liquid droplets onto the wall as the gas passes through the device [47]. After the dry gas's pressure is restored in the diffuser, it is sent down the transmission line for further processing [175].

The specific heat of the carrier gas plays a pivotal role in influencing condensation characteristics [32]. Additionally, as intake pressure and temperature increase, condensation commences closer to the nozzle throat, resulting in a decrease in nucleation rate and an elevation in outlet humidity. The rotational gas flow in the separator's horizontal axis is 1/3 slower compared to non-rotating flow [176]. The efficiency of SS purification is influenced by temperature, pressure, and flow rate, with lower temperature and higher gas flow Mach numbers inside the SS unit leading to increased liquid formation [177].

##### 4.1. Dehydration of natural gas for pipeline transport

Dehydration plays a crucial role in gas processing as it safeguards pipelines against corrosion and prevents hydrate formation. The water vapor-carrying capacity of a gas is limited and depends on its temperature and pressure. Hydrates, which can form on free water, have the potential to reduce flow capacity, hinder transmission efficiency, and even block transmission lines [86]. Additionally, water in the gas can lead to a loss in heating value and pipeline corrosion. When water molecules in the vapor phase within the pipeline begin to condense and aggregate, methane hydrate crystals form, causing the formation of larger particles [178]. When these particles come into contact with natural gas containing  $H_2S$  and  $CO_2$ , they can cause corrosion and erosion. The dew point temperature of water decreases as the ambient temperature drops. Gas hydrates have a physical appearance similar to snow. Several variables, including composition, water content, temperature, and pressure, influence the crystallization of hydrates and the associated issues. Therefore, the development of methods to prevent hydrate formation is of utmost importance.

In the gas industry, various techniques are available for gas dehydration, with absorption and direct cooling being the most prominent methods. Absorption involves the use of diethylene and glycol in the central section to remove water from the gas. In a study, Netušil et al. [179] compared three commonly used methods of dehydrating natural gas: adsorption using solid desiccants, absorption with triethylene glycol, and condensation. The comparisons were based on energy requirements and the suitability of the energy source. Under low pressures (NG from UGS at 13 MPa), the condensation method appeared to be the most demanding. As the pressure decreased linearly, its demand decreased to 145 kW at 13 MPa. The results demonstrated that condensation and adsorption both required approximately the same amount of energy. The energy demand for condensation decreased with increasing NG pressure, but with a reducing tendency when NG pressure was increased from 13 MPa to 16 MPa. Molecular-sieve adsorption and triethylene-glycol (TEG) absorption are conventional methods for dehydrating natural gas in offshore rigs [179]. However, these technologies for gas conditioning and NGL extraction require significant infrastructure investment and entail substantial capital and operational

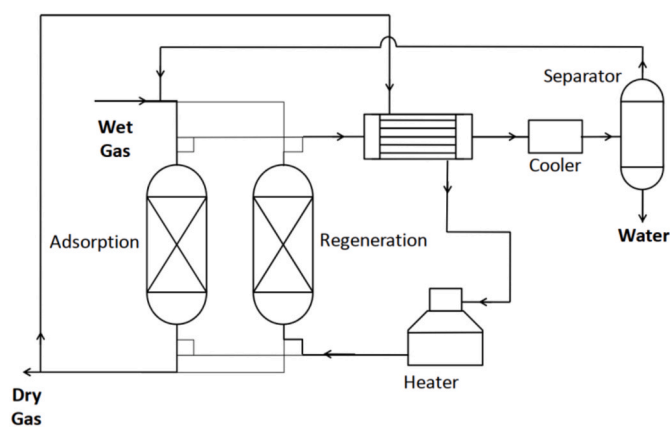


Fig. 11. Schematic of cyclic operation in adsorption dehydration columns for water removal [30] (Reprinted from Netušil Ditl [30], an open access book from IntechOpen).



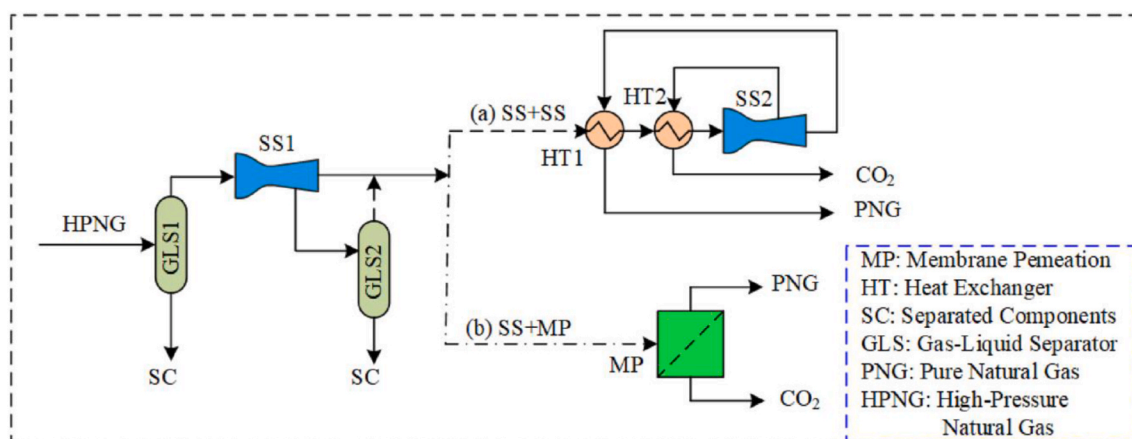


Fig. 12. Configuration of a supersonic dehydration line [61] (Reprinted from Wang et al. [61], with permission from Elsevier).

costs. They often involve spinning components, require complex human operations, pose safety concerns, and necessitate regular maintenance schedules. Moreover, traditional chemical additives used as hydrate inhibitors can be environmentally hazardous [180].

A promising high-tech innovation in the field is the use of supersonic technology to develop target components from natural gases. By employing a convergent-divergent Laval nozzle, supersonic flow is generated. The supersonic swirl separation technique is an emerging method for condensing and separating heavy hydrocarbons and water from natural gas. After comparing several gas dewatering methods, including the ultrasonic nozzle technique with others, it was determined that the ultrasonic separator method stands out as one of the most effective approaches for dehydrating natural gas due to its distinctive advantages [181].

#### 4.2. Removal of heavy hydrocarbons for improved combustion efficiency

Parameters such as population growth, economic and technology levels, and government policies affect the energy sector in a country. A sustainable future depends on both sustainable energy resources, and efficient energy systems which employ these resources. Therefore, enhancing the efficiency of energy systems is vital to reduce energy consumption. For this purpose, it is crucial to understand energy usage patterns such as the types of energy carriers used, and factors that influence their usage.

Natural gas contains heavy hydrocarbons that need to be removed to increase its heat capacity, prevent corrosion of liquefaction equipment, and avoid crystallization during the liquefaction process [182]. Teixeira et al. [183] explored a novel approach that utilizes supersonic separators to recover thermodynamic hydrate inhibitors from raw NG while reducing inhibition losses, as well as performing HCDPA and WDPA on the gas. Failure to separate heavy hydrocarbons from natural gas results in increased pipeline flow capacity and major challenges, including the need for larger pipeline diameters, expanded process facilities, increased power requirements, and significant cost escalation.

Several methods are available for separating heavy hydrocarbons, including refrigeration processes, absorption processes, cryogenic processes, surface absorption, membrane separation, and supersonic separators. Among these methods, the supersonic separator has gained popularity due to its simplicity, reliability, safety, lower installation and handling costs, minimal pressure drop, and suitability for coastal, offshore, and underwater operations.

#### 4.3. CO<sub>2</sub> capture for reduced emissions

The urgency of the climate crisis necessitates immediate action to address carbon emissions. While the full extent of the crisis is not yet

known, it is clear that continued emissions will have severe consequences. It is essential to pursue short-to medium-term solutions to aid in recovery while simultaneously developing sustainable strategies for long-term benefits [184]. The building, transportation, and industry sectors are the primary sources of global carbon emissions. Fig. 13 depicts the energy policy roadmap designed to achieve carbon neutrality by 2050 [185].

The roadmap includes several key elements: I) Subsidies for renewable energy: Encouraging the adoption and utilization of renewable energy sources through financial incentives and support. II) Energy storage and electric vehicles: Promoting the development and deployment of energy storage technologies and electric vehicles to reduce reliance on fossil fuel-based energy systems. III) Low-energy buildings: Implementing measures to construct and retrofit buildings with energy-efficient designs and technologies, aiming to minimize energy consumption and carbon emissions. IV) Low-carbon industries: Encouraging industries to adopt cleaner and more sustainable practices, technologies, and processes to reduce their carbon footprint. V) Carbon capture, utilization, and storage: Investing in and implementing technologies that capture and store carbon dioxide emissions to prevent their release into the atmosphere, and exploring ways to utilize captured carbon for

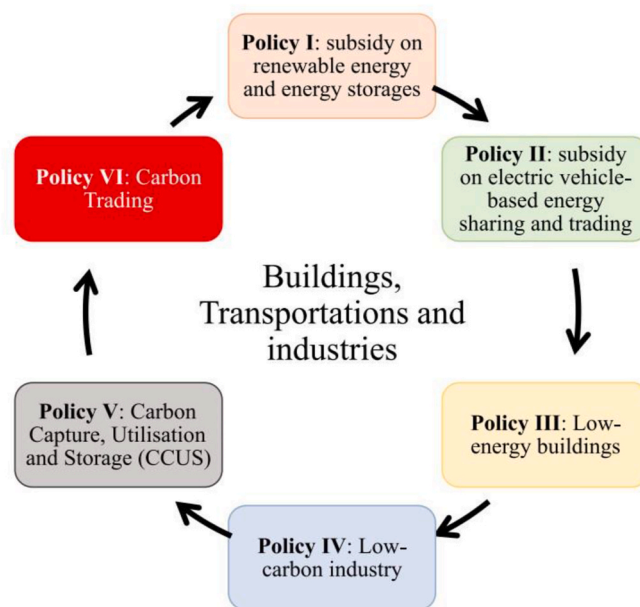


Fig. 13. Roadmap towards carbon neutrality by 2050: Energy Policy Perspective [185] (Reprinted from Zhou [185], an open access article from Elsevier).

various purposes. VI) Carbon trading: Establishing mechanisms for trading carbon credits or allowances to incentivize emission reductions and facilitate the transition to a low-carbon economy.

Researchers are currently studying how to reduce human-caused CO<sub>2</sub> emissions by capturing, transporting, and storing CO<sub>2</sub> [186]. The basic idea of CCS consists of gathering carbon dioxide mainly from industrial and electricity sources, compressing it, transporting it over extended distances, and depositing it deep underground for storage [187]. Fig. 14 illustrates various low-carbon emission options for energy supply, including natural gas, solar power, wind energy, ocean energy, nuclear power, and waste-to-energy solutions [185]. To ensure reliable and stable energy supply, different types of energy storage systems can be implemented, such as thermal storage, electrical storage, and hydrogen storage [188]. Energy distribution serves end-users, such as buildings, industry, and transportation [189]. Decarbonization roadmaps mainly concentrate on four main strategies: substituting carbon, decreasing carbon, storing carbon, and cycling carbon. The goal of these tactics is to substitute high-carbon energy sources with low-carbon options, decrease carbon emissions by enhancing efficiency and utilizing cleaner technologies, capture or store carbon emissions to avoid their release into the air, and create sustainable systems for utilizing and recycling carbon.

These roadmaps offer recommendations for reaching a carbon-free energy system and reducing the environmental effects of carbon emissions [185].

The research on oxy-combustion, pre-combustor, and post-combustor technologies to mitigate carbon dioxide emissions in energy production has gained significant attention [190]. The process involves separating CO<sub>2</sub> from industrial emissions, compressing it, drying it, and transporting it for geologic storage or enhanced oil recovery (EOR) [191]. Post-combustion capture (PCC) offers the advantage of low heat consumption, although the solvent regeneration process requires a substantial amount of energy [192]. However, chemical absorption for large-scale PCC is not as mature as chemical absorption in other applications. Industries involved in chemical production and cogeneration heavily rely on hydrogen purification and CO<sub>2</sub> capture [193]. High demand for hydrogen has also led to an increased supply of hydrogen, which powers gas turbines and fuel cells [194]. Zhu et al. [195] have highlighted recent advancements in the purification of hydrogen-rich gases in their research.

A combination of H<sub>2</sub>, CO<sub>2</sub>, CH<sub>4</sub>, CO, H<sub>2</sub>O, and hydrocarbons is created from syngas by first producing it by coal gasification or reforming of steam methane [81]. In offshore rigs, chemical, membrane,

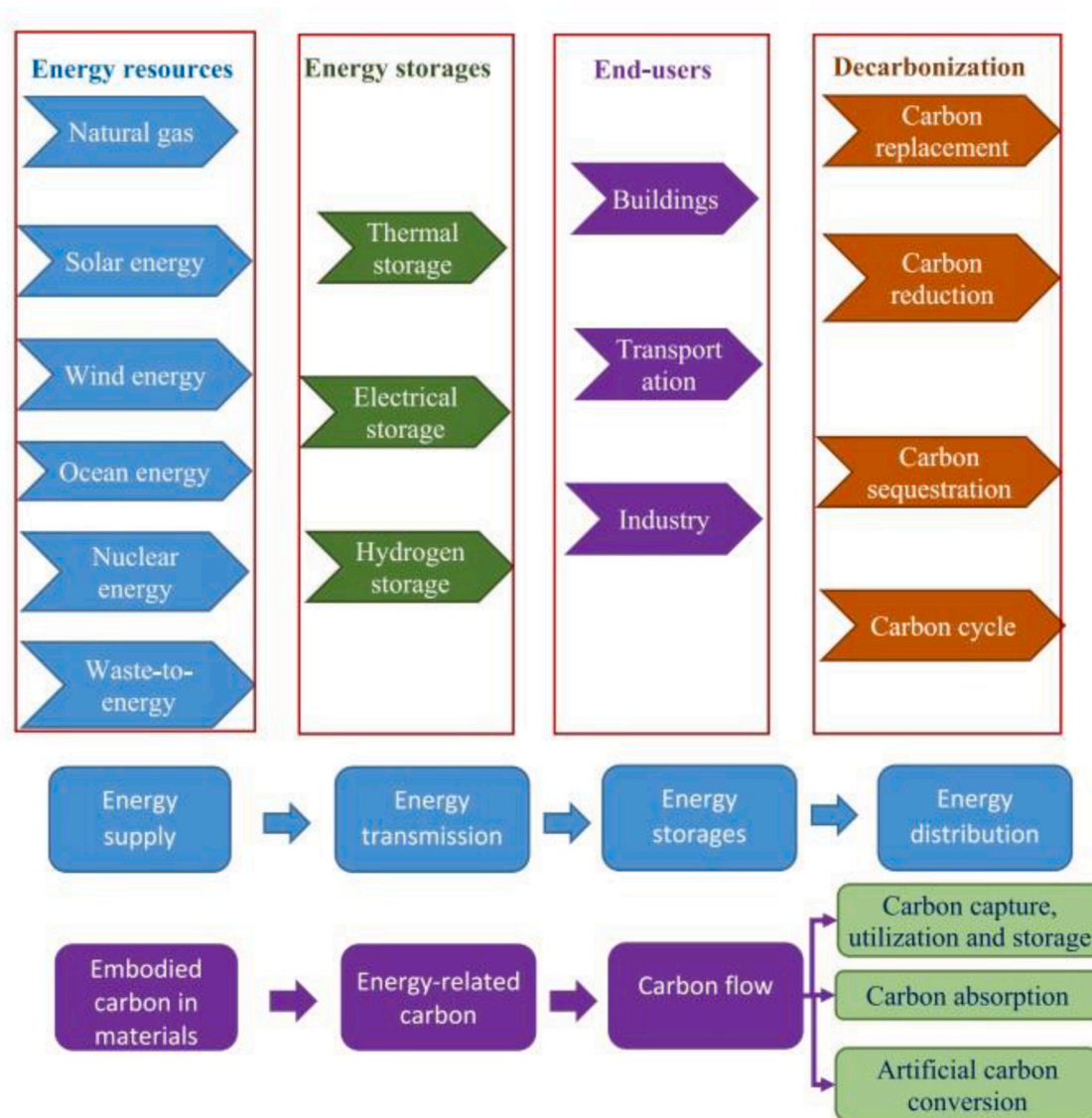


Fig. 14. Roadmap for decarbonization: Promising energy resources, storage systems, and end-User distribution [185] (Reprinted from Zhou [185], an open access article from Elsevier).

and physical absorption are the main methods used conventionally to extract CO<sub>2</sub> from NG [196].

The use of SS for CO<sub>2</sub> collection from NG with high CO<sub>2</sub> content has been extensively studied. Preparing NG with water dew-point adjustment (WDPA) and heavy hydrocarbon dew-point adjustment (HCDPA) is necessary to prevent water C<sub>3+</sub> condensation, while CO<sub>2</sub> condensation requires lower temperatures. Monitoring CO<sub>2</sub> freeze-out is crucial to avoid SS blockage. The flow path of the SS for CO<sub>2</sub> must adhere to the solid-vapor-liquid equilibrium freeze-out barrier [74]. Sun et al. [197] proposed a model for nucleation and droplet growth of CO<sub>2</sub> condensation from a CH<sub>4</sub>-CO<sub>2</sub> feed under high pressure.

Jiang et al. [35,198] have recently employed a separator that separates carbon dioxide from NG. Based on the mechanism of droplet and gas separation, they investigated condensation parameters. In addition, they investigated the effects of carbon dioxide percentages, input pressure, and intake temperature.

#### 4.4. Natural gas liquefaction for easier transport and storage

Natural gas liquefaction plays a vital role in facilitating the transport and storage of natural gas by converting it into liquefied natural gas. Recovering cold energy from LNG can significantly reduce the refrigeration requirements and save energy [199]. In addition to its capabilities in separating carbon dioxide and hydrogen sulfide from natural gas, SS units can also be used to generate LNG. Fig. 15 illustrates a compact gas liquefaction process, where the natural gas is pre-cooled using a heat exchanger to lower its temperature [2]. The cooled natural gas is then directed into a Laval nozzle, where it undergoes a rapid transformation into a liquid state. The combination of high velocity and low temperature in the Laval nozzle facilitates this transformation [2].

After passing through the Laval nozzle, the gas-liquid mixture moves into a gas-liquid separator. In this separator, the liquids separate from the gas and are directed towards the LNG storage tank. The separated liquids, now in the form LNG, are stored in the LNG storage tank for further use or distribution. At the same time, the low-temperature natural gas from the separator is combined with the boil-off gas (BOG) generated by the LNG storage tank. BOG is the gas that evaporates from the LNG due to heat gain or other factors. The mixture of low-temperature natural gas and BOG is then passed through a heat exchanger. In the heat exchanger, the incoming natural gas is warmed

using the heat energy from the low-temperature gas mixture. This process improves energy efficiency by utilizing the heat from the BOG and low-temperature gas.

For transporting LNG on ships to receiving terminals, the required temperature and atmospheric pressure are typically around 110 °C [200]. The liquefaction of natural gas requires approximately 500 kW/h of electric energy per ton of LNG at 161 °C, which includes a substantial amount of cold energy [201]. Energy requirements for the liquefaction of natural gas range between 0.45 and 0.55 kWh/kg [202]. Due to the strong relationship between these parameters, enhancing the performance of the liquefaction process is both difficult and constrained. After being distilled to make liquefied gas, LNG is kept in insulated tanks. The liquid is subsequently compressed to the required pressure for pipeline transit and evaporated to ambient temperature [203].

Bian et al. [35] proposed a revolutionary technique for liquefying gases, specifically natural gas. In their process, natural gas is pre-cooled in a heat exchanger and then passed through a Laval nozzle, where it undergoes liquefaction at high velocity and low temperature. The gas-liquid mixture is then directed to a gas-liquid separator, and the liquid component is stored in an LNG storage tank. The gas-liquid separator also combines low-temperature natural gas from the LNG storage tank with boil-off gas before entering the heat exchanger.

Gas liquefaction requires the separation of impurities such as nitrogen, mercury, moisture, acid gases, and heavy hydrocarbons. Liquefaction is a physical process of converting natural gas into a liquid state by condensation phenomenon [204]. Liquefaction of gases is used for scientific, industrial, and commercial purposes, and its volume is 600 times less than natural gas under normal conditions.

LNG is the cleanest fossil fuel economically and environmentally preferable to liquid fuels such as diesel, fuel oil, and fuel oil in many countries. This product is much safer compared to Compressed Natural Gas (CNG) due to the maintenance conditions (low operating pressure). The pressure or temperature can be increased or decreased to liquefy many gases. Nowadays, to facilitate the storage of NG and its transportation, they often use the process of liquefaction of natural gas and converting it into LPG and LNG [205]. A supersonic separator is an efficient tool for producing liquefied natural gas at the lowest possible cost. Converging-diverging nozzles in these separators cause the flow to become supersonic, causing the temperature to drop drastically and finally condensation to occur.

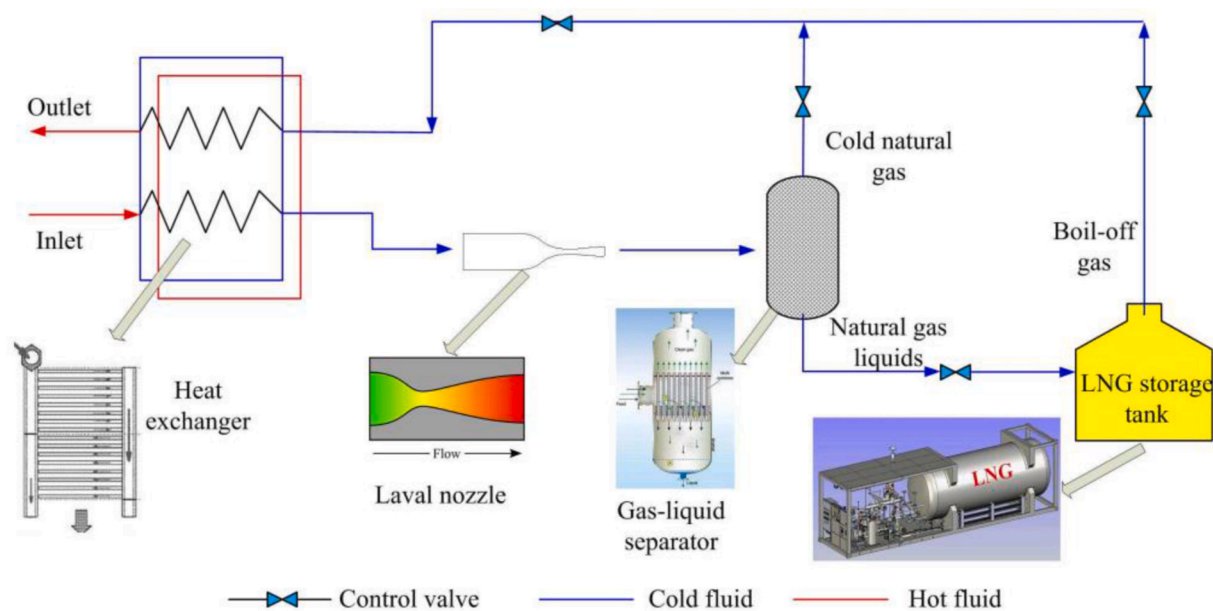


Fig. 15. Schematic of a compact gas liquefaction process: simplicity, efficiency, and environmental friendliness [2] (Reprinted from Bian et al. [2], with permission from Elsevier).

#### 4.5. Impurities removal for improved energy conversion efficiency

Natural gas separation has other advantages, aside from purifying and reducing emissions. Although fossil fuels will eventually be replaced by renewable energy sources [206], there has been a relatively low adoption rate of renewable and emission-reducing technologies [207], but achieving the Paris Agreement's greenhouse gas mitigation targets requires these technologies. Due to its low levels of emissions, NG has received the most attention due to its role in making energy sustainable [208]. CCS and CO<sub>2</sub> capture and utilization (CCU) are important technologies for minimizing environmental impacts. The CCU also converts CO<sub>2</sub> into fuels and chemicals, contributing to carbon recycling [209].

There are a number of industries that could benefit from separated CO<sub>2</sub> [210]. The most popular method is to convert CO<sub>2</sub> directly into methanol and indirectly through bi-reformation [211]. Upon reaction between CO<sub>2</sub> and methane, syngas is produced which is further converted into methanol by water gas shift reactions. Using carbon dioxide as a raw material in chemical processes is the main driving force behind these alternative methanol syntheses [212]. Being the main raw material for chemical industries, methanol operates on a global demand market. Aside from reducing oil dependency, methanol can also serve as an alternative source of fuel or raw material to manufacture hydrocarbons [213]. This could reduce industrial dependence on natural gas and crude oil [214].

### 5. Economic and environmental impact

The supersonic separator technology surpasses the limitations of traditional technologies. They do not need external power to operate, which makes them a more environmentally friendly choice compared to other technologies. Separators, too, lack moving components, making them more dependable and durable. In addition to being non-leaking and non-polluting, separators have a minimal ecological footprint, making them a favored option for industries focusing on sustainability. Separators are also safer to handle because they do not need chemicals to function. This is particularly crucial for sectors with rigorous safety requirements. Separators make dehydration easier due to their uncomplicated structure lacking retaining components. Hence, it boasts low operational expenses and the ability to function without supervision, making it highly beneficial for sectors needing constant operation [215]. Figure 16 contrasts the sustainability of supersonic separation and conventional technologies in processing engineering, examining four-dimensional indicators: environmental impact, efficiency, health and safety, and economic viability [216]. The findings indicate that supersonic separation outperforms conventional methods in terms of environmental friendliness, efficiency, and economic benefits. Specifically, the Sustainable Plant-Wide Index for supersonic separation is 0.99, significantly higher than the conventional route's 0.86. This underscores the superior sustainability of supersonic separation over conventional technologies.

#### 5.1. Cost analysis of supersonic separation compared to other techniques

High-purity gases find widespread application in diverse industries such as pharmaceuticals, analytics, electronics, and petrochemicals. The imperative to increase yields, reduce costs, and optimize performance in these sectors underscores the necessity for process-specialty gases devoid of trace impurities [217].

CCS stands out as a pivotal eco-friendly technology essential for minimizing economically feasible CO<sub>2</sub> emissions from power plants [218]. Despite recent validation of full-scale amine-based CO<sub>2</sub> capture systems [219], the persistent hurdle of costly CO<sub>2</sub> emission reductions has spurred the exploration of innovative technologies [220]. Among these technologies are molten carbonate fuel cells (MCFCs) [221], membranes [222], pressured combustion capture [223], supersonic separator [147], and flow-driven anti-sublimation.

As observed by AlNouss et al. [224], the simplest HCDPA alternative is Joule-Thomson expansion (JTE) comprising heat exchanger, isenthalpic valve, and vessel for natural gas liquids extraction. These authors economically/environmentally assessed more complex HCDPA systems considering six turboexpander configurations for lower power consumption and CO<sub>2</sub> emissions, not surprisingly identifying an economic-environmental trade-off. The complexity of implementing and operating supersonic separation technology should be considered in the cost analysis. Other techniques may have different complexities, which can impact installation, training, and ongoing operational requirements.

Teixeira et al. [52] showed that SS-methanol-recovery entails an economic leverage that affords a post-capture plant abating 43% of emitted CO<sub>2</sub>; i.e., such SS processing is a cleaner gas production compared to the conventional counterpart. For safe transportation and to preserve the heating value of natural gas streams, dew point corrections of water and heavy hydrocarbons are essential. A supersonic separator can achieve both tasks with a high degree of reliability, minimal operating costs, and minimal requirements for operational facilities. The ability of supersonic separation to handle a wide range of impurities and operating conditions can be advantageous in certain applications. Flexibility in adapting to varying feed gas compositions and impurity levels may offer cost benefits compared to other purification techniques that are more specialized.

Comparing the SS process with a conventional sequence of operations—WDPA through water absorption by triethylene glycol (TEG) followed by HCDPA with Joule-Thomson (JT) and low-temperature separator (LTS)—revealed nuanced findings [169]. The SS process exhibited higher capital expenditure (CAPEX) when compared to the TEG + JT/LTS process.

Further results dedicated that the operational expenses (OPEX) slightly favored the conventional process due to its superior availability, resulting in higher annual natural gas (NG) supply costs [169]. However, the SS process outperformed the TEG + JT/LTS process in terms of revenues owing to its higher availability and superior natural gas liquid (NGL) recovery, thereby enhancing overall economic performance. The net present value (NPV) of the SS process surpassed that of the TEG + JT/LTS process. Fig. 17 provides a comparison of fixed capital investment and discounted cumulative cash flow between conventional and supersonic separation gas plants. The findings reveal that while supersonic separation entails higher fixed capital investment compared to conventional routes, it delivers superior economic performance in terms of discounted cumulative cash flow compared to conventional gas plants [225].

Fig. 18 shows the incremental cumulative discounted cash flow (MMUSD) for the SS process, displayed incrementally relative to the conventional process (TEG + JT/LTS), reflected the higher CAPEX of the

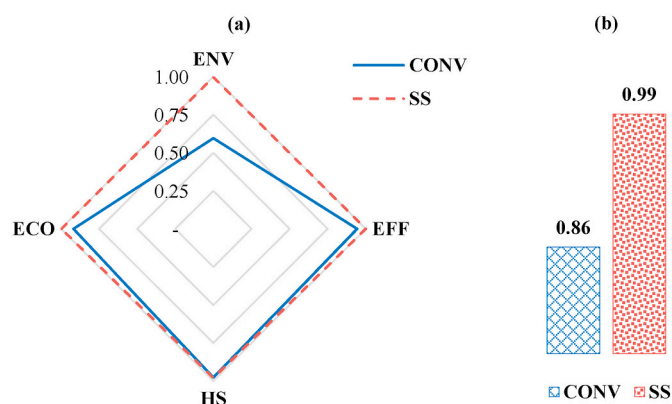


Fig. 16. Sustainable Plant-Wide Index results (CONV: Conventional, SS: Supersonic separation, ENV: Environment, ECO: Economic, EFF: Efficiency, HS: Health and Safety) [216] (Reprinted from de Faria et al. [216], with permission from Elsevier).

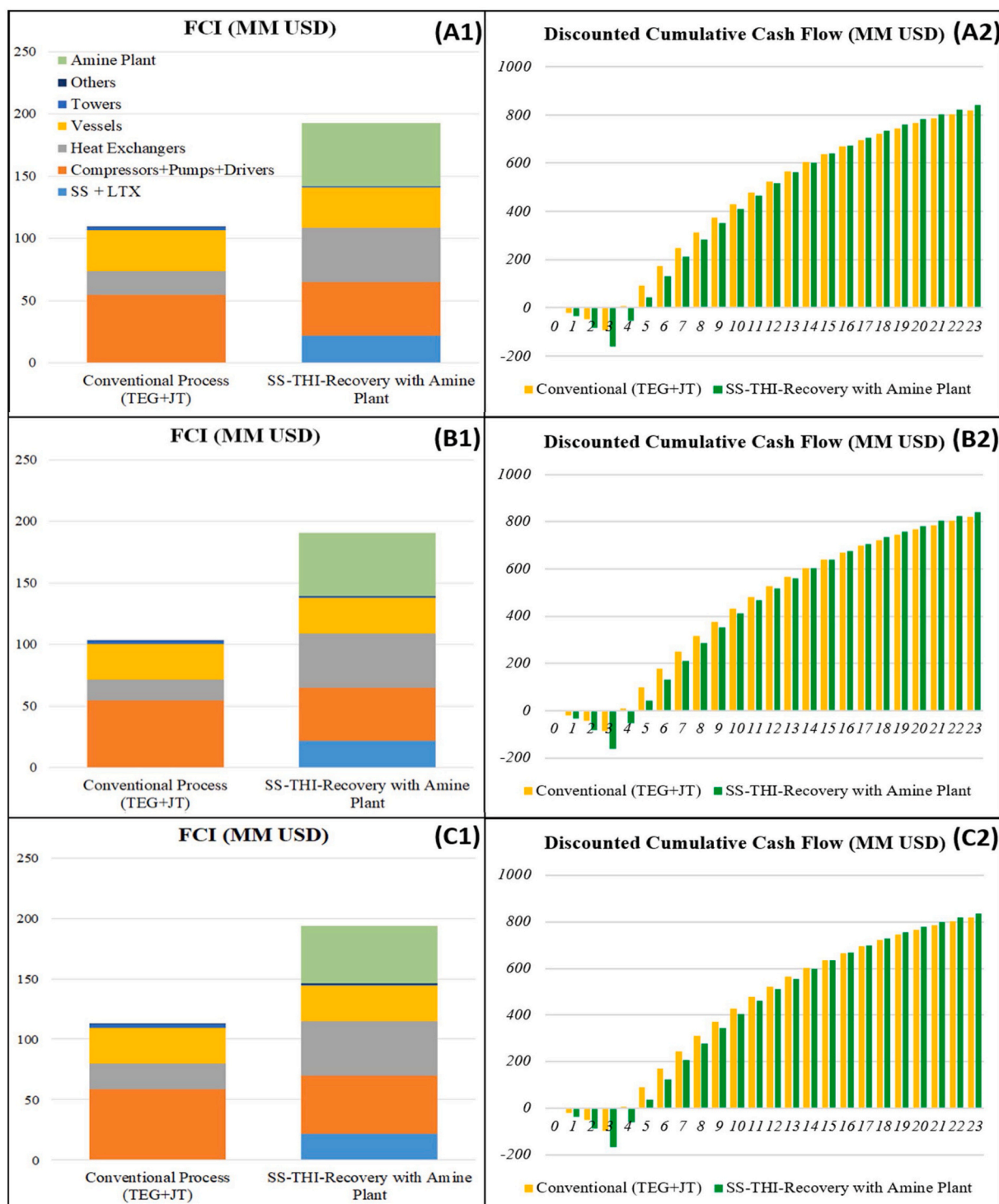


Fig. 17. Fixed capital investment [(A1), (B1), (C1)] and Discounted Cumulative Cash Flow vs. year [(A2), (B2), (C2)] for Conventional and SS (supersonic separation) Gas Plants: (A1)–(A2) THI means methanol; (B1)–(B2) THI means Ethanol; (C1)–(C2)THI means monoethylene-glycol [225] (Reprinted from Teixeira et al. [225], with permission from Elsevier).

SS process primarily attributed to required compression [77]. This compression, in turn, offered an additional benefit of export gas pressure 12.5 bar higher than the counterpart of the TEG + JT/LTS case. Furthermore, the SS process augmented NGL production, leading to increased revenues, early payback of the investment, and a higher NPV. A crucial aspect in investment considerations is depreciation, and the demonstrated 99% uptime of supersonic separators over six years, coupled with near-zero maintenance costs and inspections once every

six years, positions this technology as having lower opportunity costs compared to traditional TEG + JT/LTS technology. The glycol regeneration process in traditional methods, involving the release of a stripping gas to the atmosphere after flaring, contrasts with the SS process, which reduces major equipment requirements in NG dew-pointing and eliminates the need for chemicals. Notably, the stripping gas in traditional methods, containing C3+ alkanes and potentially hazardous benzene–toluene–xylene (BTX) aromatic components, contributes to

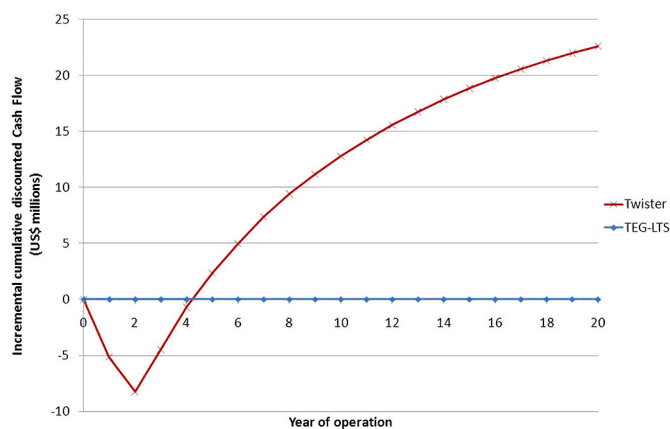


Fig. 18. Incremental cumulative discounted cash flow comparison between SS process (Twister) and TEG + JT/LTS dew-point control process [77] (Reprinted from Machado et al. [77], with permission from Elsevier).

CO<sub>2</sub> emissions due to flaring and poses risks in case of incomplete flaring.

In summary, the comprehensive cost-benefit analysis of supersonic separation compared to conventional methods reveals a multifaceted landscape. While initial costs may be higher, the long-term economic advantages, operational efficiency, and environmental benefits position supersonic separators as a viable and sustainable solution across a spectrum of industrial applications.

## 5.2. Energy consumption and sustainability considerations

There are several definitions of sustainable development, but one of the most commonly accepted is: development that satisfies current demands without jeopardizing the demands of the future [226]. Although though the future of sustainable energy relies on alternative energy sources, challenges remain, such as intermittent, location, transmission, and pricing concerns, particularly in developing nations. As evidenced by the substitution of fossil-fuels with a low H/C (heat-to-carbon) ratio (such as oil and coal) with natural gas that has a higher H/C ratio and emits less carbon dioxide per unit of energy produced, there is an industry-wide effort to replace carbon-fired power plants with efficient alternatives that leave a smaller carbon imprint. Hence, natural gas is a safe option for medium-term energy solutions. Nevertheless, more than 10% of confirmed NG deposits include 15–80% mol CO<sub>2</sub>, posing problems and necessitating the development of novel NG exploration-and-production method [91].

Most people agree that a stable supply of energy resources is a necessary but inadequate precondition to civilization's development. Moreover, a reliable supply of energy resources is necessary for sustainable development. Depending on how sustainability is defined, these statements have several effects. These assertions have an essential meaning, which is that society cannot grow sustainably without an energy supply that is easily accessible, affordable, and capable of being used for all activities without having a detrimental influence on society. The utilization of energy resources as efficiently as possible is necessary for sustainable development, which is the second implication of the opening sentence in this section [227]. In this way, society makes the most of the advantages of using its energy resources while avoiding the drawbacks (including environmental harm) related to their usage. Thus, more efficient use of these resources allows them to contribute to sustainable development over a longer period of time. This conclusion acknowledges that all energy supplies are limited to some extent. It is likely that efforts to increase energy efficiency will continue even if energy sources eventually become affordable and widely available. This is because doing so will reduce the number of resources (such as energy and materials) needed to build and maintain energy harvesting systems

and equipment and lessen any negative effects on the environment. The first implication, which is obviously relevant to sustainable development, has been and is still the topic of much debate. Secondly, energy efficiency plays a critical role in sustainable development, yet is less widely recognized and comprehended.

Rising energy consumption puts a burden on existing infrastructures and negatively impacts the environment via emissions of carbon monoxide, CO<sub>2</sub>, nitrogen oxide, and sulfur monoxide. Sustainable development is a long-term strategy for solving current environmental challenges.

Until 2050, natural gas is expected to grow by 1.1% per year, while petroleum-based liquid fuels, the most widely used source of energy, are expected to decline. NG's competitiveness is supported by its enormous resources and growing production, including NG hydrate [228], which is still in its infancy. Additionally, among fossil fuels, natural gas emits the least carbon dioxide (CO<sub>2</sub>) per unit of energy produced, which results in a cleaner burning process. NG is a crucial energy source for the world, and it is expected to continue playing a significant role in this century. Yet, extraction and production of NG can be challenging, particularly in new remote offshore fields. These fields are often characterized by low-efficiency power generation via gas-fired turbines that emit hot flue gas, resulting in high resource depletion, high carbon emissions, and low sustainability. To increase NG exploration and production efficiency, it is imperative to find solutions to these challenges [229]. This is essential because oil-and-gas offshore rigs have a major effect on the environment, producing CO<sub>2</sub> and CH<sub>4</sub> via on-site power generation, flare systems, and processing facilities. In addition, these consequences are significantly more significant under platform end-of-life situations [230]. Offshore oil rigs have a significant environmental impact due to generators, flares, and handling facilities on-site that produce CH<sub>4</sub> and CO<sub>2</sub>. In order to meet the rising demand for natural gas and global sustainability requirements [40], new offshore processing designs are needed to maximize resource utilization.

## 5.3. Environmental impact of supersonic separation and natural gas utilization

Sustainable development depends on taking environmental factors into account. Continuous environmental harm is not sustainable over time for a variety of reasons, including the cumulative impact of such actions on the ecosystem, which over time may result in a variety of health, ecological, and other difficulties. The quantity of energy used by a civilization has a tremendous impact on the environment. A society aiming for sustainable development would ideally only use energy sources that have no impact on the environment. Efforts to improve energy efficiency may alleviate some (but not all) of the concerns about restrictions on sustainable growth due to environmental emissions. There is an obvious connection between energy efficiency and the environment since reduced resource use and pollution often occur with greater energy efficiency for the same services or commodities. Higher energy efficiency reduces energy losses. With the bulk of efficiency gains, the environment benefits in two ways. Pollutant emissions are reduced first by reducing the amount of energy needed for each unit of manufacturing. When the whole lifetime of energy resources and technology is taken into account, it becomes clear that enhanced efficiency decreases the environmental impact at the majority of life cycle stages.

The mitigation of carbon dioxide emissions (CO<sub>2</sub>) has been a major concern in the past few decades, and both chemical and physical solutions have been developed. However, these solutions typically impose a significant economic cost on emitting processes. It is possible to solve this problem by converting CO<sub>2</sub> into valuable products, such as polymers, methanol, and chemical commodities. In addition to reducing emissions, the project will be economically beneficial as well.

While natural gas plays a different role since it can both be used as a mode for reducing carbon emissions and a replacement target for cleaner alternatives, depending on the sector, strategy, and operation

dynamics being analyzed. In a sustainable energy economy, natural gas infrastructure will likely play a less prominent role, despite the consensus that moving away from coal and petroleum is essential (or implementing additional measures such as CCS). The combustion of natural gas releases pollutants and greenhouse gases (GHG), while natural gas itself emits GHG, primarily methane. The shift towards natural gas as a primary fuel source is becoming increasingly popular among industrialized nations. This is due to the concern for all three kinds of consequences, including environmental, economic, and social impacts. Recent increases in gas production have been accompanied by declines in total domestic GHG emissions, indicating that natural gas may be a cleaner and more efficient alternative to conventional fuels (like petroleum or coal). As a consequence, natural gas has been pushed as a "bridge" fuel to reduce carbon emissions, particularly as a cheaper alternative to coal for power generation [231]. A well-established & low-cost source of energy, natural gas can be applied to a wide range of industries including electricity generation, transportation, and manufacturing. However, it is important to note that natural gas extracted from reservoirs contains high amounts of contaminants and heavy hydrocarbons. This means that the majority of the extracted gas is composed of these materials. It has been argued that natural gas should not be used as a bridge fuel, attributing the concerns that it may impede the development of advanced, "terminal" technologies or pose an unacceptable environmental risk if it is sourced from unconventional sources [232].

Natural gas is a cleaner and more efficient fuel source than oil and coal. However, it still contains contaminants and hydrocarbons that can cause issues when used as fuel. Fortunately, technologies have been developed to remove these contaminants and hydrocarbons from natural gas before fuel use. This process is known as gas processing and involves several stages such as separation, dehydration, sweetening, and fractionation. Overall, the shift towards natural gas as a primary fuel source is a positive step towards reducing fossil fuel negative impacts on the environment and human health. However, it is crucial to ensure that proper gas processing techniques are employed to minimize contaminants and hydrocarbons in the gas. Supersonic separation in gas purification and the utilization of natural gas can have several environmental impacts. Some key considerations are as follows.

- 1) **Reduced Emissions:** Supersonic separation helps remove impurities from natural gas, including particulate matter, liquid droplets, and potentially harmful substances. By purifying the gas stream, supersonic separation reduces the emissions of pollutants during combustion or utilization processes. This contributes to improved air quality and reduces the environmental impact associated with gas utilization.
- 2) **Greenhouse Gas Emissions:** Natural gas, primarily composed of methane, is a cleaner-burning fossil fuel compared to coal or oil. When natural gas is efficiently utilized and its impurities are effectively removed through processes like supersonic separation, the resulting combustion or utilization processes emit fewer greenhouse gases such as carbon dioxide (CO<sub>2</sub>) and methane (CH<sub>4</sub>). This can help mitigate climate change impacts.
- 3) **Air Quality Improvement:** Supersonic separation plays a crucial role in improving the quality of natural gas utilized for energy production. By removing impurities, it reduces the emission of harmful pollutants such as sulfur dioxide (SO<sub>2</sub>), nitrogen oxides (NO<sub>x</sub>), and volatile organic compounds (VOCs). This leads to improved air quality, reduced smog formation, and decreased negative health impacts.
- 4) **Reduced Environmental Contamination:** Natural gas extracted from the wellhead may contain impurities like heavy hydrocarbons, sulfur compounds, and other contaminants that can contaminate the environment if released. Supersonic separation aids in the removal of these impurities, reducing the potential for soil, water, and ecosystem contamination during gas processing and utilization.
- 5) **Conservation of Natural Resources:** Efficient utilization of natural gas, facilitated by supersonic separation, contributes to the conservation of natural resources. By extracting the maximum energy content from purified natural gas, less gas is wasted or lost during the utilization process. This maximizes the energy value obtained from each unit of natural gas extracted and reduces the need for additional resource extraction.
- 6) **Water Resource Conservation:** Supersonic separation reduces the water content in natural gas by removing water vapor and liquid droplets. This can help conserve water resources by minimizing the water needed for gas processing and reducing the potential for water contamination during gas utilization.

## 6. Conclusion and future perspectives

Supersonic separation in gas purification and the utilization of natural gas offers several important benefits and considerations for the environment. By effectively removing impurities from natural gas, supersonic separation helps reduce emissions, improve air quality, and minimize environmental contamination. The utilization of natural gas, when combined with efficient purification techniques, contributes to reduced greenhouse gas emissions and conservation of natural resources.

Because of the impressive results this technology provides, several studies have been conducted at designing, functionality, economic viability, as well as industrial uses of supersonic separators. However, it is essential to acknowledge that continued efforts are needed to further optimize these processes and address environmental challenges. This paper, which is an evaluation of the literature, gives a concise overview of recent developments in the field of supersonic separation technology. It identifies potential directions for further research.

### 6.1. Summary of advances and limitations in supersonic separation for natural gas purification

In general, the purification procedure include removing water, oil, and chemicals (e.g. hydrogen sulphide, carbon dioxide, and mercury) from a substance [233]. Conventional techniques for natural gas conditioning and NGL extraction include facilities with high capital costs and high operating expenses. The design and operation of these facilities are heavily influenced by the characteristics of wells and natural gas. Conventional sweetening and dehydration systems feature rotary components, need complicated human operations, safety concerns, and regular maintenance schedules, and create off-specification gas upon startup. Traditional chemical additions, such as hydrate inhibitors, represent significant environmental risks. Using a contactor and regenerator equipped with a hygroscopic liquid desiccant is a standard method of dehydrating gases. SS is a revolutionary separation technique. It is based on a theoretical design that combines aerodynamics, thermodynamics, physical separation, and fluid dynamics in a novel way, resulting in a new gas conditioning process.

Supersonic separator is a relatively new technique that has lately found prominence in the dehydration of natural gas. The process combines condensation and separation into a single device that controls water and dew point of hydrocarbons while improving Natural Gas to Liquid processing.

These machines have shown their viability as pipeline conditioners, particularly in unattended situations. Natural gas still needs further research to fully comprehend its properties in supersonic conditions. Further studies are required on the effects of input parameters (temperature, pressure, and composition) on boundary layer separation, as well as the effects of counterclockwise vortex generation on nozzle performance.

#### 6.1.1. Challenges in scaling supersonic separation for offshore platforms

The application of supersonic separation technology in offshore gas

turbine environments presents unique challenges beyond those encountered in onshore industries. In addition to the standard issues faced by onshore installations, the specific requirements of offshore platforms introduce additional obstacles related to size (footprint), weight, and stabilization in the presence of wave motion. Installations for CO<sub>2</sub> capture must navigate not only the constraints of limited space but also the imperative of maintaining process efficiency [31].

Compared to traditional dehydration plants, supersonic separators offer distinct advantages in terms of size, weight, cost-effectiveness, and environmental impact. Notably, they lack spinning components, making them suitable for autonomous operation offshore [234]. The technology exhibits higher energy efficiency in comparison to conventional natural gas processing methods, as supported by academic research. Despite being a relatively recent innovation, commercial test units have been operational since 1998 at various global sites, including Nigeria, the Netherlands, and Norway, accumulating substantial operational experience. Since 2003, Malaysia has employed supersonic separation technology for dehydration, with installations on the B11 platform owned by Petronas and Sarawak Shell Berhad (SSB) processing up to 300 MMSCFD [11].

The transition from lab/pilot-scale supersonic separators to industrial-scale application on offshore platforms introduces formidable challenges. Addressing these challenges necessitates a multidisciplinary approach, integrating expertise in fluid dynamics, material science, and offshore engineering. Successfully overcoming these hurdles holds the key not only to unlocking the potential of supersonic separation technology but also to contributing to more efficient and sustainable gas processing in the demanding offshore environment. Translating the success of supersonic separators from lab environments to offshore platforms introduces challenges in scaling throughput. The vast quantity of gas encountered in industrial settings requires a reevaluation of separator capacity without compromising efficiency.

The fluid dynamics governing supersonic separation at smaller scales may not fully encapsulate the complexities of offshore environments. Offshore platforms are subjected to varying sea states and weather conditions, necessitating a thorough understanding of how these factors affect the performance and stability of supersonic separators. Offshore platforms expose equipment to corrosive elements, challenging the material integrity of supersonic separators. Selecting materials that withstand the corrosive nature of the offshore atmosphere without compromising efficiency is a critical consideration. Adapting lab-scale separators to offshore platforms requires seamless integration with existing infrastructure. Challenges arise in aligning the separator with other processing units and maintaining compatibility with pipelines, demanding careful planning and engineering. Offshore platforms operate within confined spaces, necessitating a careful balance between scaling up separator size to handle industrial throughput and maintaining a compact footprint. Efficient space utilization is crucial for practical implementation.

Offshore environments pose unique challenges, including harsh weather conditions and the risk of equipment exposure to seawater. Ensuring the safety and reliability of supersonic separators under these circumstances requires robust design features and fail-safe mechanisms. The offshore industry places a premium on sustainability. Assessing and mitigating the environmental impact of supersonic separation, including considerations for emissions, waste disposal, and overall ecological sustainability, is a crucial aspect of the scaling process.

## 6.2. Supersonic separator deployment factors for CO<sub>2</sub> separation

Supersonic separators have emerged as highly efficient tools for gas mixture separation in various industrial applications, drawing attention due to their utilization of supersonic flow principles. However, their successful implementation demands a thorough exploration of practical design considerations and seamless process integration. In terms of practical design considerations, the choice of flow configuration proves

pivotal. Configurations such as parallel, counter-current, or cross-current arrangements directly influence velocity profiles within the separator, impacting separation efficiency [235]. Nozzle geometry, another critical consideration, dictates mass flow and velocity profiles at the separator's entrance, necessitating optimization to maximize efficiency while minimizing pressure loss [37]. Material selection for separator components, such as stainless steel, carbon steel, or composite materials, is essential, with factors like pressure, temperature, and gas corrosiveness guiding the decision-making process [79].

Process integration of supersonic separators into industrial operations requires careful planning to ensure compatibility with existing equipment and infrastructure. Integration may involve coupling the separator with compressors, filters, or heat exchangers, with a focus on seamless operation alongside process control and safety systems. Customization becomes imperative when employing supersonic separators for CO<sub>2</sub> separation. Nozzle material selection becomes crucial, often requiring materials like Hastelloy or Inconel to withstand high pressures and temperatures [236]. The separator design itself must be optimized for CO<sub>2</sub> separation, involving adjustments to geometry and temperature control measures to enhance capture and mitigate gas loss. Process control systems may need upgrading to cater to the specific needs of CO<sub>2</sub> separation, incorporating advanced monitoring and control algorithms to optimize efficiency and minimize energy consumption.

The advantages of employing supersonic separators for CO<sub>2</sub> separation are noteworthy. They boast a smaller footprint and lower weight suitable for offshore and crowded installations, and eliminate the need for chemicals or solvents, thereby reducing environmental impact and operational costs. Additionally, their adaptability to a wide range of pressures and temperatures simplifies process design and integration. However, challenges persist in the use of supersonic separators for CO<sub>2</sub> separation. These include high energy consumption due to substantial pressure drops and elevated inlet pressures, limited CO<sub>2</sub> recovery rates capturing only condensed droplets, potential effects of impurities on thermodynamic properties, and the challenge of integrating with other separation technologies, potentially adding complexity and cost. The implementation of supersonic separators in industrial processes requires a comprehensive understanding of both practical design considerations and the intricacies of process integration. Despite challenges, the unique advantages of supersonic separators underscore their potential, and ongoing research and development are essential to address limitations and enhance the reliability of this emerging technology, especially in large-scale and offshore applications.

## 6.3. Potential for further research and development

Supersonic separation, a critical process in industries relying on the efficient separation or filtration of high-speed gas or liquid streams, presents opportunities for further research and development (R&D) to overcome existing knowledge gaps and limitations. While significant strides have been made in optimizing separation performance and design, several areas require attention for enhanced prediction accuracy and reliability, as well as addressing challenges associated with experimental validation.

The influence of different parameters such as inlet port numbers, gas-liquid area ratios, deflection angles, inlet temperatures, and outlet angles of the swirler on separation performance needs thorough investigation. Further experimental validation and simulation improvements are necessary to quantify the impact of these variables on flow dynamics, velocity profiles, and overall separation efficiency.

The design and construction of the diversion cone, responsible for directing flow into the reflow channel, pose challenges in minimizing disturbances. Ongoing research is required to optimize the diversion cone's structure for improved flow configurations and separation efficiency.

As part of the effort to enhance prediction accuracy, future research topics are described, including modifying the gas spontaneous



nucleation rate model for improved prediction accuracy. The nucleation rates and droplet growth equations are also required correction for different geometries and conditions. In addition to the abovementioned equations, the saturation pressure and saturation temperature are greatly affected by temperature and pressure. In these problems, usually, the temperature becomes below 273 K. In this situation, ordinary equations, especially saturation relationships, are not applicable.

Despite many studies on supersonic separators, maintaining a satisfactory purification performance is still necessary owing to the flow within them. Understanding such a challenging thermodynamic and fluid dynamics system requires improvements and optimistic modifications in applying CFD techniques. It is recommended to conduct further investigations and efforts to present appropriate equations for each temperature and pressure range; it will facilitate the development of accurate models, particularly when attempting to capture the condensation shock. In addition, a multi-component simulation is suggested to study the effects of the concentration of different components on fluid flow and thermodynamics.

In the separation process of a traditional supersonic separator, the occurrence of shockwave is a common phenomenon at the nozzle expanding section prior to the cyclone. This shockwave is responsible for the effective separation of the gas mixture. However, the low temperature section in the system is relatively short which leads to inadequate cooling effect. This can lead to a reduction in the efficiency of the separator. Furthermore, when the gas mixture flows under subsonic conditions, it experiences swirling flow which is not efficient at subsonic temperatures. This is because the swirling flow is less stable under subsonic conditions. Hence, the performance of the separator is reduced. To improve the efficiency of the separator, it is important to increase the length of the low temperature section. This will enhance the cooling effect and ultimately improve the performance of the separator. The design of the separator should be such that it is capable of handling subsonic flow conditions without compromising its efficiency. This can be achieved by incorporating a more stable swirling flow design in the system.

The inclusion of discrete phase models in numerical simulation tools can significantly improve predictions by simulating the behavior of individual particles or droplets. Further research is essential to enhance the accuracy and reliability of these models, contributing to a deeper understanding of separation mechanisms.

Bi-coupling methods, combining CFD simulations with experimental measurements, offer a comprehensive approach to understanding flow and particle behavior. Continued advancements in these techniques are necessary for more accurate numerical simulations.

Researchers should focus on refining assumptions and parameter estimation techniques to improve the overall reliability of numerical simulations.

Ongoing research and development efforts should focus on advancing supersonic separation technologies, making them more efficient, cost-effective, and adaptable to varying operating conditions. Innovations in materials, design, and process integration can help enhance separation efficiency and reduce energy consumption. In order to improve the efficiency and effectiveness of the current supersonic separator, the structure of the separator should be altered to relocate where the shockwave occurs. By understanding the behavior of gases at high velocities and temperatures, it is possible to design a separator that maximizes the separation of different components. Additionally, the use of swirl generation devices is common in separator designs. However, further investigation is needed to improve the effectiveness of swirl generation. Swirl generation devices can be used to create a vortex in the gas stream, which enhances separation by causing the heavier components to migrate towards the outer edges while the lighter components remain in the center. By optimizing the design and operation of these devices, the overall efficiency of the separator can be greatly improved. Overall, the combination of altered separator structure and improved swirl generation devices has the potential to greatly enhance the

efficiency and effectiveness of supersonic separators. By applying related theories of gas dynamics, heat transfer, and fluid mechanics, it is possible to design innovative solutions that address the challenges of gas separation in a variety of industrial contexts.

#### 6.4. Implications for the energy and combustion science field

Most of the gas collected from the wellhead is saturated vapor, with only traces of heavy hydrocarbons. In the case that gas is pumped directly into the pipeline, the following three problems will occur: (1) reducing pipeline capacity and increasing power consumption; (2) Natural gas contains CO<sub>2</sub> and H<sub>2</sub>S that dissolve in water and form acid, corrosion of pipes and equipment is caused; and (3) crystalline hydrates form when water and gas combine, accumulating inside the pipeline, reducing gas efficiency, making gas supply unstable, or even blocking pipelines and equipment, causing problems with storing, transporting, or processing it [237].

Gas-water separation is therefore necessary. Traditionally, four dehydration methods have been used: solvent absorption, membrane separation, solid adsorption, and condensation separation. These methods, however, come with many disadvantages, including high initial costs and energy usage, and the need for expensive equipment. The application of supersonic separation in gas purification and the utilization of natural gas has significant implications for the energy and combustion science field.

Supersonic separation helps improve the quality of natural gas used for combustion processes. By removing impurities, it enables more efficient and cleaner combustion, leading to enhanced energy conversion and reduced emissions. This drives advancements in combustion technologies and contributes to the development of more efficient and environmentally friendly energy conversion systems.

The utilization of natural gas, coupled with effective gas purification techniques like supersonic separation, plays a role in the transition towards cleaner energy sources. Natural gas, with its lower carbon intensity compared to coal or oil, can act as a bridge fuel during the transition to renewable energy systems. Understanding the implications of gas purification on combustion science aids in the development of sustainable energy solutions.

Supersonic separation helps in achieving emission reduction targets by improving the quality of natural gas utilized for energy production. Combustion science researchers can explore the interactions between purified natural gas and combustion processes to optimize combustion conditions, reduce pollutant formation, and enhance overall energy efficiency. This knowledge supports the development of emission control strategies and contributes to air quality improvement.

The utilization of purified natural gas obtained through supersonic separation offers fuel flexibility for various combustion systems. Researchers can study the combustion characteristics of purified natural gas in different burner configurations, engines, turbines, and industrial processes. This allows for the optimization of combustion parameters and the development of tailored combustion technologies for efficient and clean energy conversion.

The combination of supersonic separation with carbon capture and storage technologies presents opportunities for further reducing greenhouse gas emissions. By removing impurities from natural gas prior to carbon capture, the efficiency and effectiveness of CCS can be enhanced. This integration requires interdisciplinary collaborations between combustion scientists, process engineers, and CCS experts to develop integrated systems for carbon-neutral energy production.

Understanding the implications of supersonic separation and natural gas utilization on the overall energy system is crucial. Combustion science researchers can contribute to system-level analysis and optimization studies, considering the entire energy supply chain, from natural gas extraction to utilization and environmental impacts. This holistic approach helps identify synergies, trade-offs, and opportunities for improving overall energy efficiency and sustainability.

In summary, the application of supersonic separation in gas purification and the utilization of natural gas has profound implications for the energy and combustion science field. Natural gas dehydration through the use of a supersonic whirling separator is a promising new commercial technology. These implications span across combustion technologies, emission reduction strategies, fuel flexibility, integration with carbon capture, and system-level analysis. Ongoing research and collaboration in these areas drive advancements towards cleaner and more efficient energy conversion systems.

### 6.5. Final remarks

In conclusion, the importance of supersonic separation in gas purification and the utilization of natural gas cannot be overstated. These processes have significant economic, environmental, and technological implications. Supersonic separation improves the quality of natural gas by removing impurities, leading to more efficient combustion, reduced emissions, and enhanced energy conversion. Supersonic separators have gained popularity in the process of natural gas dehydration, particularly in controlling water and hydrocarbon dew points. These devices combine condensation and separation processes, functioning like turbo expanders, to efficiently remove water and impurities from natural gas. The use of supersonic separators in natural gas dehydration offers several advantages, including high efficiency, cost-effectiveness, and minimal energy consumption. Their smaller size and weight make them suitable for offshore operations, and their eco-friendliness is appealing to companies focused on sustainability. Additionally, the absence of rotating parts allows for unmanned operation, which is advantageous in offshore facilities. Overall, supersonic separators provide an effective and efficient solution for natural gas dehydration and are expected to continue growing in popularity in the industry.

### CRedit authorship contribution statement

**Esmail Lakzian:** Conceptualization, Data curation, Formal analysis, Funding acquisition, Writing – original draft. **Shima Yazdani:** Data curation, Formal analysis, Investigation, Writing – original draft. **Fahime Salmani:** Data curation, Formal analysis, Writing – review & editing. **Omid Mahian:** Formal analysis, Writing – review & editing. **Heuy Dong Kim:** Funding acquisition, Supervision, Writing – review & editing. **Mohammad Ghalambaz:** Formal analysis, Writing – review & editing. **Hongbing Ding:** Data curation, Formal analysis, Methodology, Writing – review & editing. **Yan Yang:** Conceptualization, Data curation, Formal analysis, Funding acquisition, Methodology, Supervision, Writing – review & editing, Project administration. **Bo Li:** Formal analysis, Methodology, Writing – review & editing. **Chuang Wen:** Conceptualization, Data curation, Formal analysis, Funding acquisition, Investigation, Methodology, Project administration, Supervision, Writing – review & editing.

### Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

### Data availability

Data will be made available on request.

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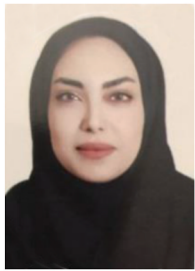
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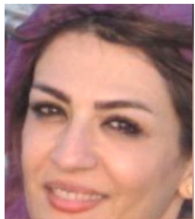
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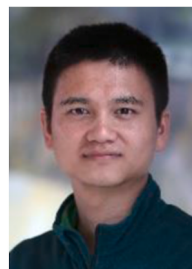
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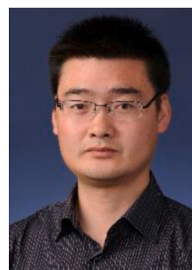
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