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Review

The potential role of gas hydrates: An emerging frontier in food science and engineering

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ABSTRACT

Gas hydrates, crystalline compounds resembling ice, are composed of water and gas molecules. They naturally occur in deep subsea surfaces and permafrost regions under high pressure and low temperature, serving as an unconventional source of natural gas. Interestingly, the conditions conducive to gas hydrate formation align with those used in food science for processing and preservation. This convergence presents a novel interdisciplinary research opportunity that could revolutionize food science and technology processes. Recent studies have sparked interest in exploring gas hydrates' potential for improving food processing, preservation, taste, and sensory appeal.

Gas hydrates offer a promising tool for food preservation, quality enhancement, and innovation. As our understanding deepens, gas hydrates could significantly improve the food processing landscape, contributing to more sustainable and efficient preservation methods. However, despite ongoing research, practical applications of gas hydrates in food processing are limited due to safety, stability, and regulatory concerns. Their specific characteristics, such as forming under low temperatures and high pressures and potential risks associated with their effect on food quality and safety, have hindered widespread adoption. Compared to carbon dioxide, which is already used in various food applications, CO_2 gas hydrates have yet to gain significant attention in food processing and preservation.

This review examines the role of gas hydrates in the food industry, focusing on their formation, properties, and diverse applications. This review aims to provide an overview of existing research and highlight knowledge gaps, guiding future investigations to unlock the full potential of gas hydrates in enhancing food processing and preservation techniques.

1. Introduction

Gas hydrates are ice-like crystalline compounds where water cages surround gas molecules. (Huang et al., 1966; Srivastava et al., 2022a). These gas molecules can be flammable, such as CH_4, C_2H_6, or H_2, or non-flammable, such as CO_2 or N_2. Critical distinctions between gas hydrates and ice include differences in latent heat and melting temperatures (Fournaison et al., 2004b; Gabitto and Tsouris, 2010; Osegovic et al., 2006).

Gas hydrates, known as clathrates, form under high-pressure and low-temperature conditions. Various gases, including CH_4, CO_2, N_2, O_2, and C_2H_4, can form gas hydrates with different compositions and hydration numbers (Cady, 1983; Hachikubo et al., 2022; Uchida et al., 1999). The three main structural families of gas hydrates (sI, sII, sH) are well-known and depend on the van der Waals diameter of the guest molecules and the relative number of large and small cages in the structures (Takeya et al., 2010). Some scientists believe hydrate formation is a fundamental property of water, occurring in solid and semi-liquid systems at low temperatures and high concentrations or pressures of guest molecules. The size of these guest molecules should

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align with the cavities of gas hydrates, while their chemical nature is generally not crucial to forming hydrates (Ostrovskii and Kadyshevich, 2012). The development of clathrate hydrate science is nicely summarized in another review paper (Ratcliffe, 2022).

Each structure consists of small and large cages, with larger guest molecules tending to occupy the larger cages. Gas hydrates can be differentiated based on their structure type and the thermodynamic stability pressure at a given temperature. Due to their thermodynamic stability conditions, gas hydrates have been proposed as a technological solution for various separation processes, such as CH_4/CO_2 and CO_2/N_2 separations.

Fig. 1 explains the configuration of the gas hydrate structure. In the hydrate structures sI, sII, and sH, the vertices of intra-structural cavities are formed by the oxygen atoms of water molecules. The length of each edge corresponds to the combined lengths of the O–H valence bond in a water molecule and the H···O hydrogen bond between it and an adjacent water molecule. Above each cavity, its free diameter and the number of its facets (noted as superior figures) are specified, along with the number of edges (noted as lower-case figures) and the indices of the hydrate structures that include this cavity.

The van der Waals–Platteeuw Theory (statistical mechanical model) describes the equilibria of clathrate hydrates (Medeiros et al., 2020; Ripmeester and Alavi, 2022). van der Waals and Platteeuw assumed that the hydrate water lattice, with its fixed structure and number of water molecules, could be treated separately from the encaged guest species. This theory determines the fraction of cages occupied by guest molecules in the hydrate structure. Researchers can better understand hydrate formation equilibria by calculating cage occupancies under various experimental conditions. This model simplifies the complex interactions in hydrates, aiding in computational predictions and experimental measurements.

Gas hydrates offer unique properties that make them promising candidates for innovative food science and engineering solutions. Gas-containing gas hydrates are non-explosive, easy to handle, demonstrate self-preservation, create nanobubbles upon dissociation (Uchida et al., 2016), demonstrate higher stability in confined space (Pandey et al., 2021), have high latent heat (Yin et al., 2021) and require less energy to form than other competitive processes (Pandey and von Solms, 2023). Some properties of interest are summarized in Fig. 2.

Recently, gas hydrate technology has been assessed for its potential application in various aspects of food science (Nkosi et al., 2023; Srivastava et al., 2021). Gas hydrates can preserve heat-sensitive substances such as vitamins and polyphenols, maintaining the nutritional value of food. They can also reduce microbial load, improve food safety, and extend shelf life. The structural flexibility of gas hydrates, influenced by the type of guest gas molecules and formation conditions, can be optimized for specific food applications. Additionally, gas hydrates can entrap gases like carbon dioxide, methane, and hydrogen in a solid matrix, allowing for controlled gas release in food products, which can modify characteristics such as texture, flavor, and shelf life. The inclusion of various chemical additives can further tailor the properties of gas hydrates to meet specific needs in food applications. Furthermore, their high capacity for gas storage and transportation in hydrate form could revolutionize food packaging and preservation techniques, addressing food spoilage and waste issues. In this review paper, authors summarize recent advancements and elucidates potential applications of gas hydrates in the food industry. It also discusses the challenges and future directions for utilizing these multifunctional agents. From improving the sensory attributes of food products to revolutionizing packaging technologies, integrating gas hydrates holds promise for transforming the landscape of food science and engineering, fostering innovation, sustainability, and consumer satisfaction.

This review paper aims to comprehensively examine the emerging role of gas hydrates in food science and engineering. This will foster innovation, stimulate further research in gas hydrate science, and inspire additional research endeavors within the food industry. The review paper is divided into nine sections (Refer to Fig. 3). It begins with an "Introduction," establishing the context and objectives of the review. The following section, "Use of gases in the industry," examines the various industrial applications of gases, emphasizing their critical roles.

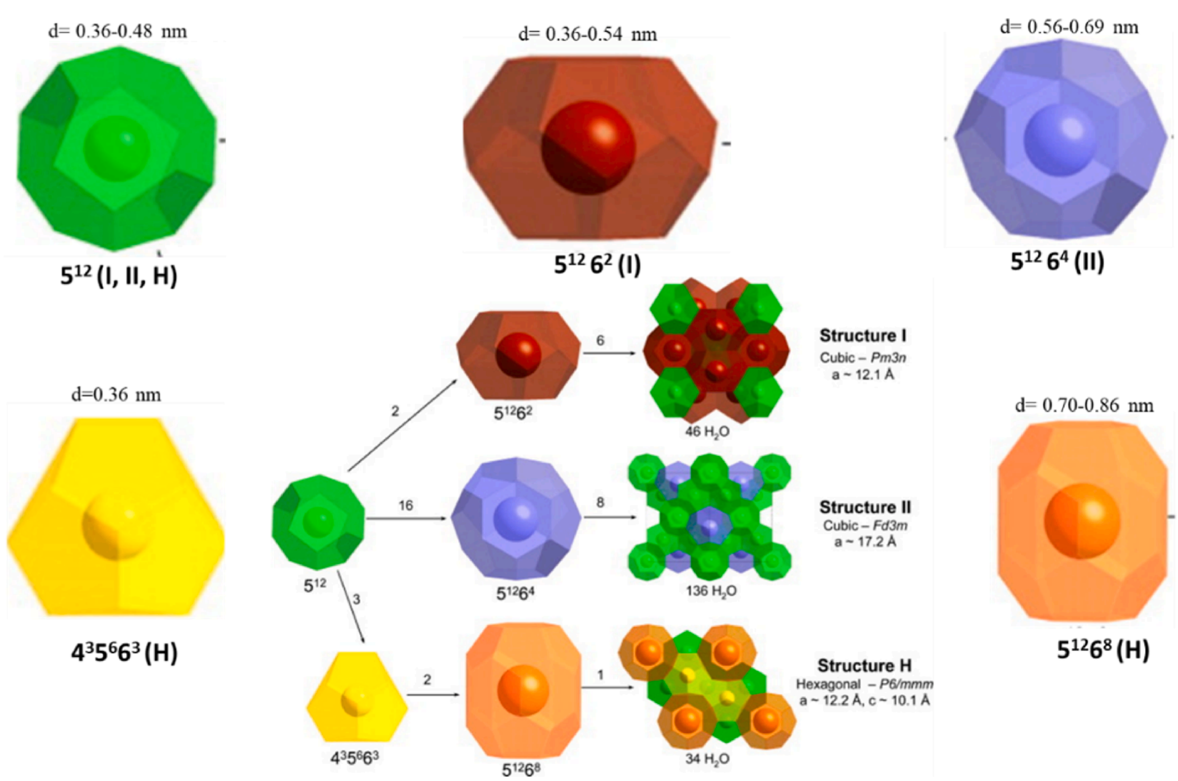

Fig. 1. Various gas hydrate structures and cage unit structure (Ostrovskii and Kadyshevich, 2012; Sloan, 2003).

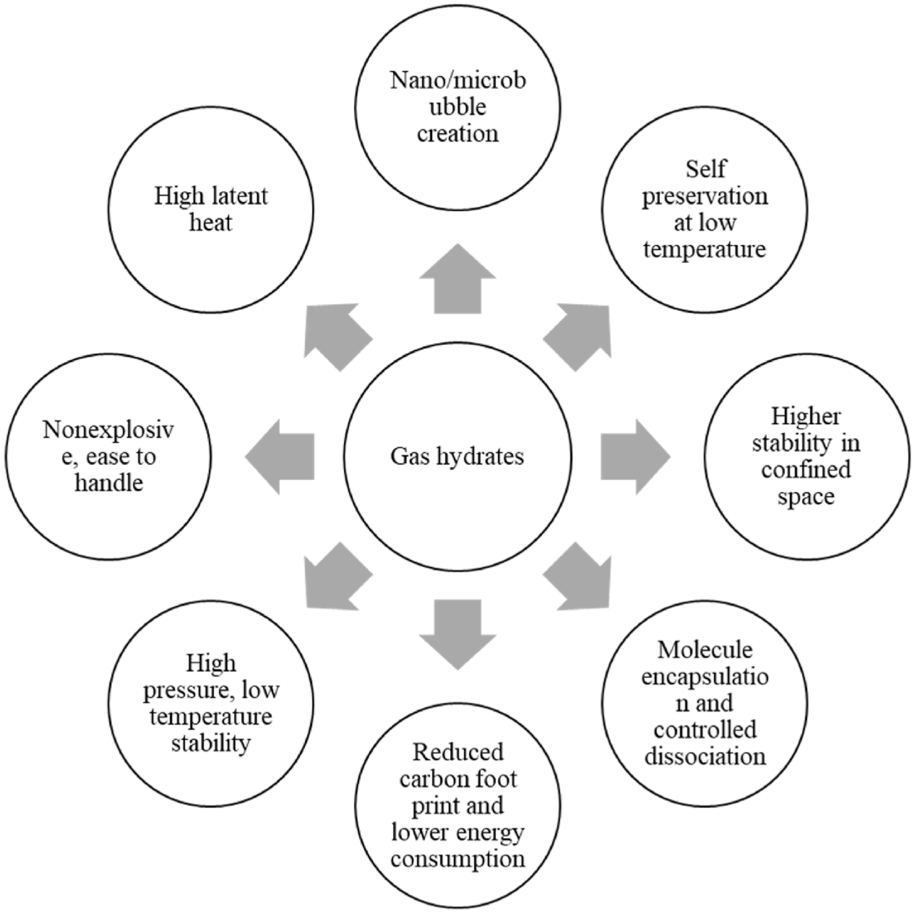

Fig. 2. Key properties of gas hydrates.

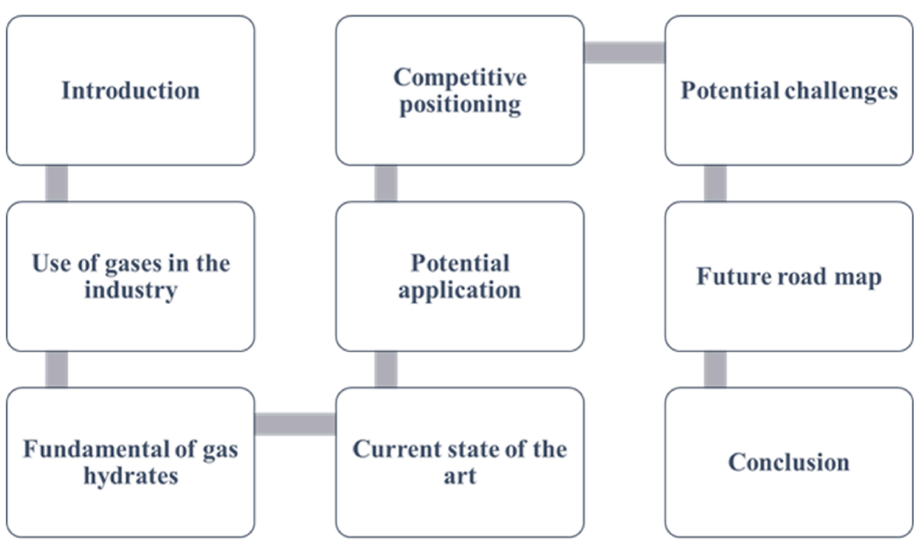

Fig. 3. Overview of review paper.

Next, the "Fundamentals of gas hydrates" section explores the basic principles and scientific importance of gas hydrates, providing a comprehensive understanding of these compounds. The review then advances to the "Current state of the art," summarizing recent advancements and the latest research developments in the field of gas hydrate application in food science. The "Competitive positioning" section discusses and compares CO_2 hydrate with other systems. This is followed by "Potential applications," in which authors share their critical analysis of potential areas in food science and technology that could benefit from gas hydrate application. The subsequent section, "Potential challenges," identifies and scrutinizes the obstacles and issues that may impede the development and application of gas hydrate technologies. The "Future Road map" then outlines anticipated advancements and strategic directions. Finally, the review paper concludes with a

"Conclusion," encapsulating the key points and insights from the review.

2. The use of gases in the food industry: state-of-the-art

Understanding the properties and roles of gases in the food industry is essential for exploring new food processing and preservation applications. Like other chemicals, many gases are used in food science and processing applications. Many dietary components, for example, occur in hydrated forms such as monohydrates or pentahydrates, which contain a specified amount of water molecules per molecule of the item (Srivastava et al., 2021). Additionally, it helps assess the feasibility of incorporating gas hydrates into these processes as they are the mixtures of water and gas. Table 1 summarises the crucial role of commonly used gases in food production, especially concerning preservation, packaging, and atmosphere modification.

Gases enhance product quality, extend shelf life, and ensure safety at different stages of food processing. (Shaalan et al., 2022). Gases offer unique properties that enable the preservation and modification of the food environment. By creating modified atmospheres (gas-modified atmosphere packaging, GMAP), gases can inhibit microbial growth, delay oxidation processes, and maintain the sensory attributes of food products (McMillin, 2020). Gas solubility varies significantly, with CO_2 highly soluble in water and N_2, O_2, and Ar poorly soluble (Ibarra, 2019). Gases such as nitrogen, carbon dioxide, and ozone are crucial for food preservation, preventing spoilage, maintaining freshness, and enhancing marketability. They create modified atmospheres, control microbes, and preserve sensory characteristics. Oxygen and ethylene are used selectively to maintain the natural freshness of perishable products.

Nitrogen (N_2) plays a vital role in food processing, especially in modified atmosphere packaging, cryogenic freezing, oxidation prevention (displacing oxygen and blanketing gas or reducing oxidative reactions), and beverage dispensing spoilage. (Adhikari et al., 2018; Alwazeer et al., 2020; Brown et al., 2018; Fraqueza and Barreto, 2011; Guo et al., 2018, 2020; Tinebra et al., 2021; Yang et al., 2022). It extends shelf life, maintains quality, and enhances the consumer experience of various food and beverage products. By retaining a low-oxygen environment, nitrogen helps preserve packaged foods' color, texture, and flavor, thus enhancing their shelf life and quality. This method protects oils, fats, and powdered food ingredients prone to rancidity and spoilage. N_2 is in its liquid or gaseous form and is also employed in cryogenic freezing and cooling to cool or freeze food items rapidly (Cachon et al., 2019; Goswami, 2001; Martino et al., 1998). These processes involve using extremely low temperatures to preserve food products by minimizing the formation of ice crystals and preserving texture, taste, and nutritional properties (Goswami, 2001). Nitrogen gas dispenses beverages like draft beer (Girardon, 2019a), coffee, and soft drinks, providing pressure for efficient dispensing. This creates a creamy and frothy texture, enhancing the sensory experience, especially in carbonated beverages.

Carbon dioxide (CO_2) is widely used in the food industry for various purposes, including carbonation of beverages, modified atmosphere packaging, preservation of fruits, baked, vegetables, dairy, and meat, as well as pH control in food and beverage applications (Shaalan et al., 2022; Singh et al., 2012). Due to its bacteriostatic and fungistatic effects, it is used as a natural preservative to extend the storage life of foods. It inhibits the growth of spoilage-causing microorganisms, reduces enzymatic activity, and slows deterioration processes, enhancing product freshness and maintaining nutritional value. These inhibitory effects prolong both the lag phase and the generation time of microorganisms, consequently resulting in a delay in spoilage (Ibarra, 2019). CO_2 has a distinct impact on microorganisms. It is more inhibitory to Gram-negative than Gram-positive bacteria (Devlieghere and Debevere, 2000; McMillin, 2008). For yeasts and lactic acid bacteria, CO_2 can either inhibit (Henriksen et al., 2000) or enhance growth depending on the situation (Henriksen et al., 2000). CO_2 acts as a pH regulator in food and beverages, forming carbonic acid to adjust and stabilize acidity in

Table 1
Overview of the current state of gas application in the food industry.

Gas	Application	Advantage	Disadvantage	Reference
N_2	Modified atmosphere packaging (MAP)	Extends shelf-life; Inert gas does not react with food	It can be costly to produce or obtain	(Adhikari et al., 2018; Helmenstine, 2017; Ibarra, 2019; Kargwal et al., 2020; Lei et al., 2022; Martino et al., 1998)
	Cryogenic freezing and cooling	Maintains freshness; Reduces spoilage and rancidity	Requires specialized equipment for handling	
	Blanketing to prevent oxidation	Reduces mold and bacterial growth. Preserves food quality	Requires careful control to avoid over-pressurization	
CO_2	Modified atmosphere packaging (MAP)	Inhibits microbial growth. Extends shelf life	It can cause brittleness or deformation in packaging.	(Adhikari et al., 2018; Ibarra, 2019; Lu et al., 2020; McMillin, 2020)
	Preservation of fruits, vegetables, and meat	Preserves color and texture. Versatile applications	High concentrations can be hazardous.	
	Dispensing beverages or Carbonation of beverages	It provides fizz and enhances the sensory experience. Widely available and cost-effective	It can affect pH levels and taste in certain products.	
O_2	Modified atmosphere packaging (MAP, selective use) It helps maintain sensory quality. Facilitates natural respiration. It helps maintain sensory quality.	Maintains product color. Enhances aerobic processes in certain products. e. g., O2 binds with myoglobin to create an appealing cherry-red color in red meat, respiring live shellfish, and respiring fruits and vegetables; O2 slows down the growth of aerobic pathogen Clostridium botulinum in fresh fish	Generally harmful to preservation Rapidly consumed by aerobic microorganisms. Can promote oxidation and spoilage in certain foods	(Ibarra, 2019; Lu et al., 2020; Shaalan et al., 2022)
O_3	Reduces microbial contamination (powerful antimicrobial agent) Removes chemical residues	Extends shelf life Effectively kills bacteria, viruses, and fungi Leaves no residue	It can be hazardous at high concentrations Requires careful handling and control	(Horvitz and Cantalejo, 2014; Kim et al., 1999; Singh et al., 2022)

(continued on next page)

Table 1 (*continued*)

Gas	Application	Advantage	Disadvantage	Reference
	Water disinfection and treatment Equipment and surface sterilization			
SO_2	Winemaking Antioxidant	Prevent from oxidation	Its effectiveness varies with pH	(Li et al., 2008; Oliveira et al., 2011)
	Antimicrobial	It inhibits microbial growth and prevents the formation of biogenic amines, ethyl phenols, and off-flavors	Its effectiveness varies with pH	(Ough and Crowell, 1987; Santos et al., 2012)
	Extraction	Extraction of minerals, organic acids, and phenolic compounds	SO_2 rapidly disappears in crushed grapes	Ribereau-Gayon et al. (2006)
	Taste/Sensory	complex flavor (rotten grapes, mediocre varieties) and stable color	SO_2 excess: unpleasant flavors, hydrogen sulfide, mercaptans	Garde-Cerdán et al. (2007)
	Preservatives in dried fruits and vegetables	Prevents browning and spoilage widely available and cost-effective	It can cause adverse reactions in sulphide-sensitive individuals (e.g., asthma, dermatitis, angioedema, anaphylaxis, etc.)	(Guerrero and Cantos-Villar, 2015; Velayutham et al., 2016)
C_2H_4	Stimulates color development Controlled ripening of fruits	Facilitates uniform ripening. Accelerates fruit ripening	Overexposure can cause premature spoilage (enhances natural processes)	(Arshad and Frankenberger, 2002; Xiao et al., 2018)

soft drinks, wines, and certain foods. Carbonation enhances dairy products, such as probiotic-containing yogurt drinks, without affecting the viability of probiotics, improving quality, shelf life, and sensory attributes (Newbold and Koppel, 2018; Walsh et al., 2014). However, carbonation methods for dairy beverages are not widely used in commercial products compared to other drinks. In baking, CO_2 is a leavening gas in chemical dough and biological leavening processes (Frühling et al., 2023). CO_2 is essential for creating the characteristic bubbles and fizziness in carbonated drinks like soda, sparkling water, and beer. In modified atmosphere packaging (MAP), CO_2 helps extend the shelf life of food products by inhibiting microbial growth, delaying oxidation, and maintaining freshness. The CO_2 (supercritical fluid) is also used as a solvent for various applications, including extracting seed oils, lipids, and essential oils and isolating flavors and aromas (Kueh et al., 2018; Marzorati et al., 2020; Santana et al., 2012).

Oxygen (O_2) plays essential roles in the food industry, both beneficial and detrimental. (Ibarra, 2019). Modified atmosphere packaging (MAP) relies on carefully regulated oxygen levels to preserve the quality of fresh-cut fruits and vegetables, supporting respiration and delaying senescence. This controlled oxygen exposure maintains the produce's visual appeal and nutritional quality. However, oxygen can harm overall food preservation, promoting the growth of aerobic microorganisms and leading to oxidative degradation of lipids, pigments, and vitamins. Despite this, certain products benefit from the presence of oxygen (Ibarra, 2019). For example, oxygen binds with myoglobin in red meat, creating an attractive cherry-red color. Live shellfish, respiring fruits, and vegetables also benefit from oxygen. Oxygen can also slow the growth of the aerobic pathogen *Clostridium botulinum* in fresh fish, providing a protective effect.

Ozone (O_3) is used in the food industry for various purposes, including water disinfection and treatment, equipment and surface sterilization, and reducing microbial contamination (Horvitz and Cantalejo, 2014; Kim et al., 1999; Singh et al., 2022). It demonstrates high effectiveness against various microorganisms for ensuring food safety and quality (Horvitz and Cantalejo, 2014).

Argon (Ar) behaves similarly to N_2, being inert concerning the product. However, its higher solubility and density provide an advantage in displacing and removing O_2, reducing gas consumption (Ibarra, 2019). Despite this benefit, the relatively higher cost of Ar has limited its broader adoption compared to N_2. No discernible difference in microbiology was observed between argon and nitrogen. However, the use of argon did result in some positive sensory effects (Claudia and Francisco, 2010). Ar is non-narcotic and FDA-listed as generally recognized as safe (GRAS), and it produces foamy and creamy beverages (Adhikari et al., 2018).

Sulfur dioxide (SO_2) is commonly used as a preservative in dried fruits (Velayutham et al., 2016). It is an antioxidant and antimicrobial agent to enhance shelf life and maintain product quality. SO_2 plays a vital role in winemaking, protecting against oxidation (Li et al., 2008; Oliveira et al., 2011), inhibiting microorganisms (e.g., prevents yeast haze formation and secondary fermentation) (Ough and Crowell, 1987; Santos et al., 2012), aiding pigment extraction due to its dissolving power (Ribereau-Gayon et al., 2006), and influencing sensory properties (Garde-Cerdán et al., 2007). However, excessive use can lead to adverse effects, especially for sulphite-sensitive individuals, and careful consideration is necessary to balance its benefits and potential health implications (Guerrero and Cantos-Villar, 2015).

Hydrogen (H_2) (in mixture with N_2) bubbles in yogurt reduce whey separation and improve gel aggregation (Martin et al., 2010). It also effectively influences aroma components in non-fat yogurt by modifying the metabolic pathways of lactic starter cultures (Cachon et al., 2002). The standard atmosphere is replaced with a mixture of inert gases to prevent oxidative deterioration in stored milk products (Giroux et al., 2008). Dairy beverages saturated with N_2 and H_2 exhibit minimal color change during storage compared to those saturated with air.

Nitrous oxide (N_2O) is used in making chocolate and coffee foam (Haedelt et al., 2007). It improves whipped cream production, but a gas mixture of N_2, N_2O, and CO_2 is preferred to avoid an unpleasant taste and reduce costs. However, there are concerns about asphyxiation risk and its global warming potential (Adhikari et al., 2018).

Xenon (Xe) has unique properties, including rapid diffusion across biological barriers and neuroprotective and myocardial protective effects (Baumert et al., 2005). It is used in functional dairy products for treating chronic diseases like neuralgia, migraine, myocardial infarction, and Alzheimer's (Adhikari et al., 2018). Xenon can also be encapsulated within water-soluble cyclodextrin to create nutraceutical beverages (Adhikari et al., 2018). Encouraging dairy-based therapeutic formulations may enhance Xenon's applications.

Helium (He) is utilized in a novel packaging method to tightly fit perishable food, preventing the effects of O_2. Its high diffusivity and low solubility make it ideal for cling packaging of dairy products. Helium flushing before irradiation improves flavour retention and overall acceptability in peppermint ice cream (Hashisaka et al., 1990).

Ethylene (C_2H_4) controls fruit ripening and stimulates colour development, allowing optimal fruit maturity and appealing visual attributes (Arshad and Frankenberger, 2002; Xiao et al., 2018). **Propylene**

(C_3H_6) acts as an antifreeze agent in chilled and frozen foods, ensuring that the products maintain their quality and texture during storage and transportation.

Ammonia (NH_3) is utilized for refrigeration and cold storage in the food industry, providing efficient cooling and temperature control for preserving perishable products.

The gases mentioned above are prone to forming clathrate hydrates, a characteristic of considerable importance as it suggests the potential development of diverse processing and preservation methods based on gas hydrate formation, thereby fostering innovation within the food industry. Moreover, it is crucial to investigate novel gas applications and safety measures to mitigate potential risks (Girardon, 2019b; Shaalan et al., 2022; Simoneau, 2019). So far, gas hydrates have shown diverse potential in food science and technology applications. Notably, gas hydrate technology can be used as a leavening agent for baked goods, fruit juice concentration, and food tissue preservation, providing efficient, sustainable, and gentle approaches (Arunyanart et al., 2014, 2015; Claβen et al., 2020; Frühling et al., 2023; Purwanto et al., 2001). Understanding gas hydrates' diverse properties and behaviours is essential for advancing environmentally friendly and energy-efficient alternatives to current food processing and conservation methods. The following section provides a summary of pertinent information regarding gas hydrates.

3. Fundamental properties of gas hydrates relevant to food science and engineering

3.1. Introduction to CO_2 gas hydrates

CO_2 is a colorless and odorless gas (at low concentrations). Its density is 1.98 kg/m3 at STP, which is much more solubilized in water than methane (Diamond and Akinfiev, 2003; Yamamoto et al., 1976). CO_2 hydrates are one of the well-known gas hydrates being investigated by researchers due to their potential application in various research topics (Sinehbaghizadeh et al., 2022) such as CO_2 hydrate-based desalination and metal separation (Babu et al., 2020; Han et al., 2017; He et al., 2018; Kang et al., 2014; Park et al., 2011), hydrate-based CO_2 capture and separation (Dashti et al., 2015; Duc et al., 2007; Wang et al., 2020a; Xu and Li, 2014), CH_4/CO_2 hydrate swapping (Pandey et al., 2022; Wei et al., 2022), CO_2 hydrate-based air conditioning and secondary refrigeration systems (Wang et al., 2020b; Yin et al., 2021) as well as recent applications in hydrate juice processing and preservation (Claβen et al., 2019; Srivastava et al., 2021). CO_2 hydrates are also tested as technology solutions for salt manufacture, carbonated solid food, tritium water concentration (Nakamura et al., 2023; Tariq et al., 2023), and fire extinguisher (Gaidukova et al., 2022, 2023b; Hatakeyama et al., 2009). In recent reviews of CO_2 hydrates' application in cold thermal energy storage (Yin et al., 2021; Wang et al., 2020b), CO_2 hydrates are also used in non-traditional applications, such as energy, food storage, carbon capture, and juice concentrate, demonstrating their adaptability and potential in various industries (Claβen et al., 2019; Wang et al., 2020a; Yin et al., 2021).

CO_2 hydrates are of structure 1 (sI) type and made of large and small cages, and CO_2 gas molecule occupancy in large cages is higher than in smaller cages (Seol et al., 2010). CO_2 hydrates also have faster formation kinetics than CH_4 hydrates due to CO_2 gas's higher solubility and diffusion rate in liquid water, which is enhanced under pressure (Jung et al., 2010). CO_2 hydrate exhibits lower surface tension than CH_4 hydrate (26 and 32 mJ/m^2) (Arjun and Bolhuis, 2021). This leads to different CO_2 hydrate morphology evolutions compared to CH_4 hydrates (Zhao et al., 2021). CO_2 concentration in CO_2 hydrate at 276 K and 30 bar is around 298 kg/m^3 (Makiya et al., 2010; Udachin et al., 2001).

3.1.1. Structure of CO_2 hydrate

Like other gas hydrates, CO_2 hydrate can also be formed under low temperatures and moderately high pressures. The formation of CO_2 hydrates can be expressed as:

$$xCO_2 + yH_2O \leftrightharpoons xCO_2 \,.\, yH_2O \quad (1)$$

These nonstoichiometric solid crystalline compounds comprise cages formed by three-dimensional, hydrogen-bonded water networks that serve as the host, with CO_2 molecules trapped inside these cages as the guest. Weak van der Waals forces connect CO_2 and water molecules. Hydrates remain stable when the guest substance occupies the cages within their structure. Without a guest substance, the cages become unstable and may collapse into a typical ice structure. Gas hydrates commonly form three structures, depending on the molecular diameter of the encapsulated gas. These structures are known as structure I (sI), structure II (sII), and structure H (sH). These three structures comprise five different cages in various shapes and sizes, including 5^{12}, $5^{12}6^2$, $5^{12}6^4$, $4^35^66^3$ and $5^{12}6^8$ (Sloan et al., 2007). sI structure consists of 46 water molecules, while this number is 136 and 34 for sII and sH hydrates, respectively.

CO_2 hydrates can form two primary types of crystal structures: Structure I (sI) and Structure II (sII). The sI structure is more prevalent for CO_2 hydrates and is characterized by two types of cavities: small dodecahedral and large tetra decahedral cavities. These cavities are enclosures formed by water molecules that trap the CO_2 molecules (Circone et al., 2003; Kumar et al., 2009a; Martos-Villa et al., 2013; Sloan et al., 2007; Udachin et al., 2001). The sII structure, less common for pure CO_2 hydrates, appears under specific conditions, mainly when larger guest molecules are present. This structure comprises small dodecahedral and large hexakaidecahedral cavities (Seol et al., 2010; Udachin et al., 2001). The ideal unit cell formula for CO_2 hydrate in the sI structure is $2M_S.6M_L.46H_2O$, representing a lattice of 46 water molecules encapsulating CO_2 molecules. M_S and M_L represent the small (5^{12}) and large ($5^{12}6^2$) cages, respectively (Udachin et al., 2001).

In the presence of other hydrate formers, such as methane or larger hydrocarbons, CO_2 hydrates can form mixed hydrate structures. These mixed structures are combinations of sI and sII, where CO_2 occupies the smaller cavities, and larger molecules occupy the larger cavities. This adaptability is significant for gas separation and storage applications, where CO_2 is often mixed with other gases (Dashti et al., 2015; Wei et al., 2022).

3.1.2. Hydration number

Hydration number is an important characteristic of clathrate hydrates, representing the number of water molecules needed to encapsulate one guest molecule. It is determined by the occupation ratio of the structural cavities. For instance, CO_2 hydrates typically form a structure composed of 5^{12} and $5^{12}6^2$ cavities. Considering all the cavities are filled with CO_2 molecules, the ideal unit cell formula would be $6(5^{12}6^2).2(5^{12}).46H_2O$. As per equation (1), $x = 8$, $y = 46$; therefore, the hydration number would be $n = y/x = 5.75$.

It is usually not possible that all the cavities are fully occupied; therefore, the actual hydration number is generally higher than the ideal ones. Usually, the probability of large cavities being occupied is more than 95%. At the same time, it is hard to check the probability of small cavities being occupied because the occupation ratio of small cavities depends on the guest composition and reaction conditions, such as temperature and pressure (Sloan et al., 2007).

The literature reports several indirect (calculation) and direct (experimental) methods for determining the hydration number of clathrate hydrates (Uchida, 1998). For instance, Bozzo et al. (1975) employed the Miller–Strong method to calculate the hydration number of CO_2 hydrate where a relation involving the fugacity and activity of water is prescribed. Lirio et al. and Anderson et al. proposed another method using the Clapeyron equation. They calculated the dissociation enthalpies of hydrate-water and hydrate-ice system and used their difference and heat of dissociation of ice to obtain the value of hydration numbers (Anderson, 2003; Lirio and Pessoa, 2013). Anderson (2003) found that the hydration numbers for CO2 hydrates are 5.6 ± 0.3 and

6.6 ± 0.3 at 283 K and 272 K, respectively due to the fact that at higher temperature, the vapour pressure is higher.

Different experimental techniques, viz. calorimetry, were used to determine the hydration number of CO_2 hydrates using direct methods (Kang et al., 2001), Raman spectroscopy (Gborigi et al., 2007; Qin and Kuhs, 2013; Sum et al., 1997; Uchida et al., 1995), Nuclear magnetic resonance (NMR) (Ripmeester and Ratcliffe, 1998), X-ray diffraction (XRD) (Circone et al., 2003; Udachin et al., 2001), attenuated total reflection infrared (ATR-IR) (Kumar et al., 2009b) to name a few. A comparison between the Raman intensities of formed CO_2 hydrates and aqueous CO_2 solution indicated that the hydration numbers are 7.68 (at 9.05 °C and 4.56 MPa) and 7.24 (at 9.55 °C and 3.93 MPa), indicating incomplete occupancy in cavities (Ripmeester and Ratcliffe, 1998; Uchida et al., 1995). XRD was used to investigate the single CO_2 hydrate crystal to determine a hydration number of 6.20 at 276 K (Udachin et al., 2001). NMR experiments indicated that CO_2 has occupancy in both large and small cavities. Assuming all the large cages are occupied, a hydration number of 7.0 was obtained (Ripmeester and Ratcliffe, 1998). Experiments with a mass balance where the mass difference before and after system hydration was used to determine the hydration number. These experiments indicated complete occupancy of CO_2 in small and large cavities and thus resulted in a nearly ideal hydration number of 5.75 (Circone et al., 2003). ATR-IR experiments on CO_2 hydrates revealed occupancy of both small and large cages based on vibrational frequencies from CO_2. Their ratio gave a hydration number of 6.04 (Kumar et al., 2009b; Ma et al., 2016).

It can, therefore, be summarized that CO_2 essentially forms sI structure hydrates. Most of the time, incomplete occupancy of CO_2 in cages has been reported. The occupancy ratio in large cages was close to 1.0, whereas it depends mainly on operational conditions in small cages. The hydration number of CO_2 hydrate was reported in the range of 5.7–7.7 (Ma et al., 2016).

3.1.3. Equilibrium/dissociation conditions of CO_2 hydrates

The equilibrium and dissociation conditions of hydrates are fundamental aspects that determine their stability and practical applications. A hydrate phase equilibrium diagram illustrates the correlation between temperature and pressure under which hydrates remain stable and dissociate and delineates the conditions under which CO_2 exists as a gas or liquid. Numerous studies have investigated the equilibrium conditions for CO_2 hydrates using different methods and devices refined over the years to ensure precise determination (Jäger et al., 2013; Semenov et al., 2023; Tariq et al., 2022). Such studies are essential as they provide data for designing and optimizing industrial hydrate-based processes. The phase equilibrium data for CO_2 + water system collected from different sources in the literature (Adisasmito et al., 1991; Anderson, 2003; Lin et al., 2008; North et al., 1998; Ye and Zhang, 2012) by Ma et al., (2016) is presented in Fig. 4a.

The phase equilibrium data of CO_2 + water in the temperature range from −30 °C to +30 °C and in the 0.1–7.5 MPa pressure range is presented in Fig. 4a. In addition to the normally reported hydrate–water equilibrium line, there is a liquid-vapour equilibrium line of CO_2 due to its conversion into liquid at high pressure, which divides the phase diagram into four parts for different combinations of water, hydrate, liquid CO_2, and gaseous CO_2. The fifth area represents ice and CO_2 gas when the temperature is below the freezing point of water to form ice. Fig. 4b represents the hydrate-vapour-liquid equilibrium (HLVE) data measured recently by Ito et al. (2023) along with the classical measurements reported in the literature. The HLVE region is the most sought after one from the application point of view. Detailed information on the phase behaviour of CO_2 hydrates in a wide range of temperatures and pressures can be found elsewhere (Bollengier et al., 2013).

The presence of impurities or additives can significantly alter the equilibrium and dissociation conditions of CO_2 hydrates. Substances like salt or other gases can promote or inhibit hydrate formation by changing the temperature and pressure conditions under which hydrates are stable.

3.1.4. Dissociation enthalpies of CO_2 hydrates

The dissociation enthalpy of hydrates is a critical thermodynamic property that quantifies the energy required to break down the hydrate into water and gas. It is also known as the heat of dissociation and represents the heat absorbed when a hydrate dissociates into its constituent components. It's a critical factor in determining the energy efficiency of hydrate processes. The endothermic nature of hydrate dissociation can be harnessed for cooling and refrigeration purposes. A higher dissociation enthalpy indicates a more energy-intensive dissociation process, a crucial consideration in designing systems for hydrate handling and utilization (Ma et al., 2016; Sun and Kang, 2016).

The dissociation enthalpy of hydrates can either be calculated or

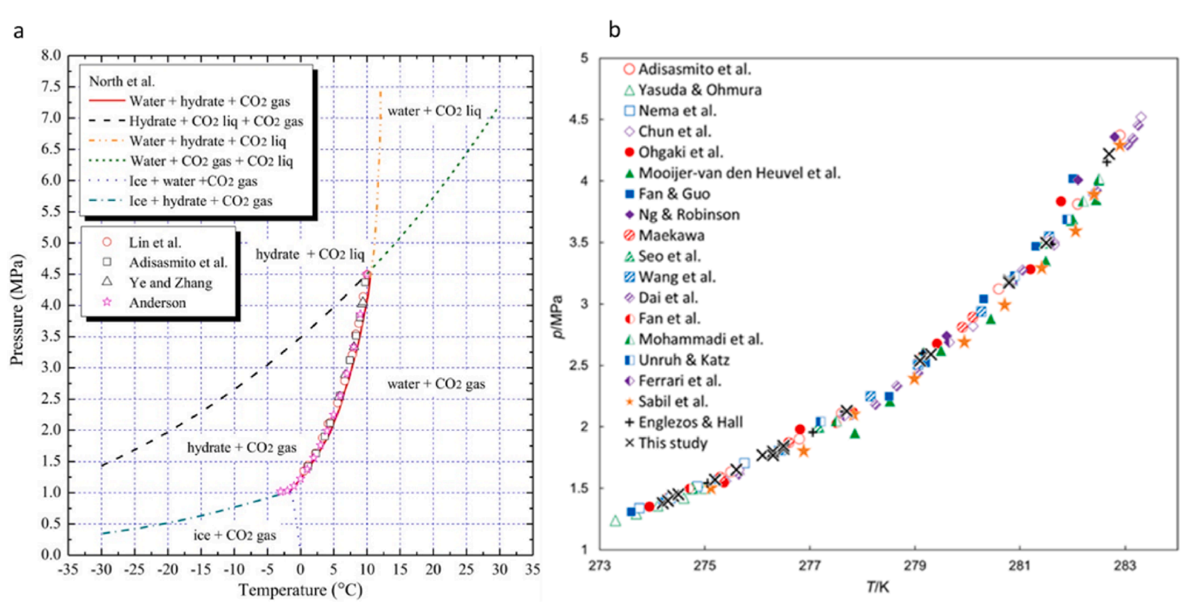

Fig. 4. Phase equilibrium data of (a) CO_2 +water system collected from the literature (Adisasmito et al., 1991; Anderson, 2003; Lin et al., 2008; North et al., 1998; Ye and Zhang, 2012). The figure has been reproduced from ref. (Ma et al., 2016) with permission from Elsevier. (b) Recent measurements of CO_2 hydrate vapour liquid equilibrium by Ito et al. along with the data reported in the literature (Reproduced from ref (Ito et al., 2023) with permission from Springer.

measured experimentally under various conditions. Typically, the factors influencing the dissociation enthalpy of CO_2 hydrates are the presence of additives (salts) or other hydrate-forming gases, which can alter the hydrate structure, thereby affecting its dissociation enthalpy.

Generally, different types of calorimetric apparatuses to measure the dissociation enthalpies of CO_2 hydrates (Kang et al., 2001; Ohgaki et al., 1993). Theoretically, the Clapeyron and Clausius–Clapeyron equations were used to calculate the dissociation enthalpies of CO_2 hydrates from the three-phase equilibrium data (Anderson, 2003; Lirio and Pessoa, 2013). However, due to the high solubility of CO_2 gas, a modified Clausius–Clapeyron equation was also used (Bozzo et al., 1975; Ohgaki et al., 1993; Skovborg and Rasmussen, 1994).

The dissociation enthalpies values for CO_2 hydrates in the temperature range of 283.52–286.61 K and pressure up to 50 MPa were reported to be 46.42–48.55 kJ/mol. The calculated values obtained through the Clapeyron or Clausius-Clapeyron equation fall within the range of 47.69–75.37 kJ/mol (in the temperature range of 271.80–288.45 K) (Robustillo et al., 2022).

3.1.5. Self-preservation of CO_2 hydrates

Self-preservation refers to the ability of hydrates to remain stable at temperatures above their expected dissociation point (atmospheric pressure, 240–263 K). Self-preservation occurs when the dissociated gas molecules are prevented from leaving the hydrate lattice due to capillary forces, surface active agents, or a protective layer of ice around the hydrate particles. This phenomenon is characterized by a reduced dissociation rate, allowing the hydrates to persist in a metastable state for an extended period, even under conditions where they would typically dissociate. This property is particularly crucial when maintaining the hydrate phase, which is advantageous for storage or transport. Most of the studies focusing on this phenomenon are carried out for methane hydrates, and the mechanism of this phenomenon is poorly understood (Gudmundsson et al., 1994; Istomin et al., 2006; Majid and Koh, 2021; Prasad and Chari, 2015).

There are a few studies related to the self-preservation effect in CO_2 (Burla and Pinnelli, 2023; Falenty and Kuhs, 2009; Takeya et al., 2000). Various factors, including the size and purity of the hydrate crystals, the presence of impurities or additives, and the initial formation conditions, can influence the extent of self-preservation in CO_2 hydrates. Recently, the stability of CO_2 hydrates in the self-preservation region of 240–268 K at atmospheric pressure and the long-term stability at 268 K has been studied. The hydrates were stable for about 50 h. As the temperature reached 268 K due to the melting of the protective ice layer, the rate of gas release increased considerably (Burla and Pinnelli, 2023).

The self-preservation property of CO_2 hydrates has significant implications for their storage and transportation. In carbon capture and storage (CCS), for instance, this phenomenon can be exploited to ensure the stability of CO_2 hydrates during transportation from the capture site to the storage location, even under fluctuating environmental conditions. Similarly, in transporting CO_2 for various industrial applications, leveraging self-preservation can reduce the need for stringent temperature and pressure controls, simplifying logistics and reducing costs (Dashti et al., 2015; Duc et al., 2007; Xu and Li, 2014).

3.1.6. CO_2 hydrate formation process

The hydrate formation process is a complex phenomenon that involves the physical transformation of water and a hydrate-forming substance into a solid crystalline structure under specific conditions. It includes two vital steps: nucleation and crystal growth. Fig. 5 shows a typical gas profile consumed during the hydrate formation process.

Initially, water and gas are present in the system. As the process starts, from the beginning up to point A indicates the gas dissolution step, from point A to B is the nucleation process – where tiny clusters of hydrate form, and from point B onwards is the growth process – where the hydrate crystal grows into larger crystalline structures. Hydrate dissociation is an exothermic process, while the formation process is endothermic (Sun and Kang, 2016). Several factors can affect the formation process, including nucleation and growth. Several theories and models are proposed to explain nucleation and growth stages. These theories and models are covered in detail in another review paper and described in Fig. 5B (Liu et al., 2022a,b).

3.1.6.1. Nucleation. Nucleation is the initial and crucial microscopic stage in the formation of hydrates. It involves the formation of tiny clusters of hydrate molecules, which serve as the foundation for further hydrate growth. Understanding this process is critical to optimizing hydrate formation for various applications (Ke et al., 2019; Khurana et al., 2017; Vysniauskas and Bishnoi, 1983).

There are two types of nucleation: (i) Homogeneous nucleation refers to the spontaneous formation of hydrate nuclei in a gas-water mixture without any external catalysts or impurities. Environmental conditions like temperature and pressure heavily influence this process, and (ii) Heterogeneous nucleation occurs in the presence of foreign substances or surfaces that facilitate the formation of hydrate nuclei. This type of nucleation is typically more efficient than homogeneous nucleation due to the lower energy barriers involved (Kashchiev and Firoozabadi, 2002;

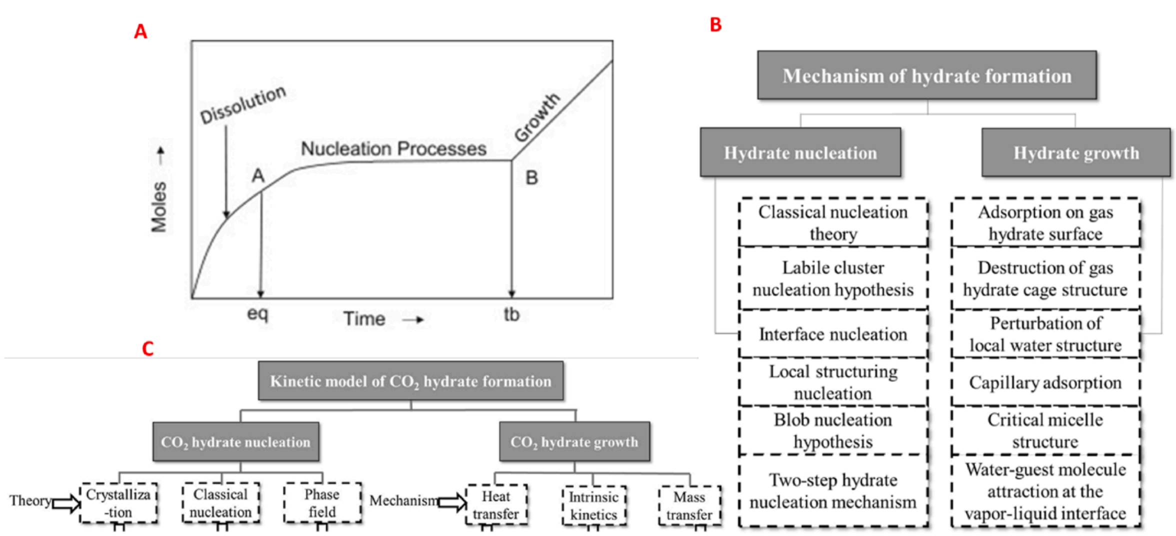

Fig. 5. (A) Gas consumption profile during the hydrate formation. It is reproduced from ref. (Sun and Kang, 2016) with permission from Elsevier (B) Mechanism of gas hydrate (e.g. CO_2 gas). nucleation and growth. (C) Models of CO_2 hydrate nucleation and growth. (B) and (C) are reproduced from Ref (Liu et al., 2022a,b).

Maeda, 2020).

Several mechanisms and theories were proposed to explain the nucleation of hydrates: classical nucleation theory, labile cluster hypothesis, nucleation at interface hypothesis, local structuring mechanism, blob formation mechanism, and multiple pathway nucleation. (Khurana et al., 2017).

CO_2 hydrates nucleation was studied using specialized equipments (Jeong et al., 2022; Tariq et al., 2023; Wei and Nobuo, 2023). Since the nucleation process is difficult to investigate experimentally due to its long time scale, computational methods have been employed in recent years to obtain molecular-level insights (Das et al., 2022). Several factors, such as initial temperature conditions, pressure, concentration, agitation, memory effect, reactor geometry, and the presence of additives and surfaces can alter the hydrate nucleation (Maeda, 2020).

Arjun and Bolhuis (2021) investigated the homogeneous nucleation rate of CO_2 hydrates using transition interface sampling, providing insights into the molecular-level mechanisms and the energy barriers associated with this process. Kar et al. (2021) The study by Zhao et al. (2021) demonstrated that magnesium ions can significantly enhance the rate of CO_2 hydrate nucleation. (Zhao et al., 2021) provided high-resolution insights into CO_2 hydrate formation near the gas-water interface, relevant for understanding the nucleation process under different conditions. Wei and Maeda (2023), discussed the memory effect of clathrate hydrates, which can impact the nucleation process. This memory effect refers to the tendency of hydrate-forming systems to nucleate more readily following previous hydrate formation and dissociation cycles.

3.1.6.2. Growth. The growth of gas hydrates is a crucial phase that follows nucleation, where hydrate crystals expand and develop into larger structures. The mass transfer of gas molecules to the hydrate surface is critical to growth. Aside from mass transport, the growth kinetics and heat transfer of the exothermic growth process from the crystal surface to the solution are crucial, albeit it is unclear which phenomenon is predominant (Yin et al., 2018). There are three crucial steps associated with the hydrate growth: (i) water and gas molecules diffuse from the solution's bulk to the hydrate's crystal surface, (ii) formation of a structured water framework through the recombination of hydrogen bonds between water molecules and the addition of guest gas molecules to the framework, and (iii) hydrate development into the bulk of the solution from the formed crystal surface (Liu et al., 2022a,b).

Hydrate growth can be affected by several factors, such as interaction between the additives and guest molecules, interactions between the additive and water molecules, interactions between the additives and hydrate surface or the alteration of the gas/liquid interfacial tension. Several mechanistic pathways for hydrate growth were proposed in the literature, namely: adsorption at the surface of gas hydrates, collapse of the hydrate cages, water structure perturbation, capillary adsorption, critical micelle structure, and water-guest molecule interaction at the vapour-liquid interface (Linga and Clarke, 2017; Liu et al., 2022a,b; Lu et al., 2022; Uchida et al., 2002; Yin et al., 2018).

Several studies are available on the growth of CO_2 hydrates. One research study demonstrated that amino acids can perturb the structure of water to alter growth (Liu et al., 2022a,b). Veluswamy et al. (2017), reported the effects of additives and operating conditions on the crystal structure and growth of CO_2 hydrates. Phan and Striolo (2023) reported the impact of promoters on the growth of CO_2 hydrates using molecular dynamics simulations and validated their results with experimental data. In addition to chemical additives, nanoparticles have recently been used to enhance the growth of CO_2 hydrates (Cheng et al., 2021).

3.1.6.3. Important characteristic parameters. Several important parameters are listed below to understand and rate the efficiency and behaviour of hydrate formation and stability, which impact their applications in different areas. (Liu et al., 2022a,b; Ma et al., 2016; Sinehbaghizadeh et al., 2022; Sun and Kang, 2016).

(i) Total gas uptake refers to the amount of gas that can be incorporated into hydrate form under specific conditions. This parameter is crucial for assessing the efficiency of hydrate-based storage and captures systems. The review from Sun and Kang (2016) included insights into the total gas uptake of CO_2 hydrates, which is essential for evaluating the effectiveness of hydrate-based technologies in energy applications.
(ii) Gas density and solubility in the hydrate phase play a vital role in hydrate formation dynamics and storage capacity. These factors determine how much gas can be stored in each hydrate volume. Wang et al. (2020b) reviewed the use of CO_2 hydrates in cold thermal energy storage, providing valuable information on the density and solubility of CO_2 in the hydrate phase.
(iii) Induction time is the duration required for the onset of hydrate formation once the system reaches conditions conducive to hydrate formation. This parameter is critical for process design and efficiency in industrial applications. Babu et al. (2014) explored the thermodynamic and kinetic aspects of CO_2 hydrate formation using tetra-*n*-butyl ammonium nitrate ($TBANO_3$) as a promoter. Their study provides insights into the induction time for hydrate formation, which is vital for optimizing process parameters in CO_2 capture and storage applications. Liu et al.'s (2022) review provides a detailed description of these parameters and the effect of different variables (Liu et al., 2022a,b).

3.1.6.4. Effect of different reactors. The design and type of reactors used in forming hydrates significantly impact their formation rate, efficiency, and quality of the hydrates (Filarsky et al., 2021; Ghaani et al., 2021). Different reactor designs can influence parameters such as heat transfer, mass transfer, and mixing efficiency, which are critical for the rate and extent of hydrate formation. This aspect is crucial in scaling up hydrate-based technologies for various industrial applications (Ke and Kelland, 2016; Linga and Clarke, 2017).

Different types of reactors have been employed to study CO_2 hydrate formation, *viz*., continuous flow reactor, column reactor, batch or semi-batch reactors, stirred reactor, and impeller. (Castellani et al., 2016; Longinos and Parlaktuna, 2021; Yamasaki et al., 2000; Yang et al., 2011, 2008). Park et al. (2011) and Babu et al. (2020) delved into developing new apparatuses and prototype designs for efficient CO_2 hydrate formation, particularly in desalination. This provided insights into how different reactor designs can optimize the hydrate formation process from the perspective of hydrate-based desalination. Dashti et al. (2015) provided insights into the application of CO_2 hydrates in CCS, indirectly highlighting the importance of reactor design in implementing these technologies. Unfortunately, comparing the results of hydrate formation kinetics across laboratories is impossible. For a comprehensive overview of this area, readers are referred to the work of Linga and Clarke (2017).

3.1.7. CO_2 hydrate promotion using amino acids and biosurfactants

As mentioned in an earlier section, the presence of an additive can affect the rate of hydrate formation and different valuable parameters. From the perspective of applying CO_2 hydrates in food science, using non-toxic chemical additives (promoters) to improve the kinetics and stability of CO_2 hydrates is required. Amino acids and biosurfactants can fit very well in this category. Amino acids are the basic building blocks of biological macromolecular proteins and can act as kinetic promoters in hydrate formation (Bavoh et al., 2019; Bhattacharjee and Linga, 2021). Biosurfactants can enhance hydrate formation by reducing the interfacial tension between the vapour and liquid phases. This alteration can lead to increased mass transfer and an improved hydrate nucleation and growth rate (Bavoh et al., 2023; Kumar et al., 2015).

Several amino acids, viz., L-leucine, L-methionine, L-tryptophan, L-valine, L-cystine, and L-isoleucine, were reported to act as kinetic hydrate promoters for CO_2 hydrate (Bhavya et al., 2021; Cho et al., 2024; Li et al., 2021, 2023; Prasad and Kiran, 2018). Bhattacharjee and Linga (2021) proposed a mechanism based on hydrophobic effects through which amino acids promote the hydrate formation (Nguyen and Nguyen, 2017) as depicted in Fig. 6.

Under specific circumstances, biosurfactants are released metabolites that exhibit surface activity during microorganism metabolism. Biosurfactants are reported to have lower critical micellar concentrations (CMCs) than conventional surfactants (Arora and Singh Cameotra, 2014). Studies related to the promoting effect of biosurfactants on CO_2 hydrate formation are almost non-existent. (Bavoh et al., 2023; Zhang et al., 2020). Only one work is available where rhamnolipid was used to study the recovery of methane from gas hydrate by CO_2 replacement and depressurisation. It enhanced the storage capacity of CO_2 in the process (Heydari and Peyvandi, 2020). Therefore, the main challenge lie in identifying specific amino acids and biosurfactants most effective for promoting CO_2 hydrates, balancing efficiency with environmental and economic considerations.

CO_2 hydrates offer innovative storage and preservation solutions for the food industry. Their unique properties can maintain chilling conditions and freshness, especially when traditional refrigeration is impractical. The potential of gas hydrate technology for food applications, such as using CO_2 hydrates for food preservation and juice concentration, has been explored by Claβen et al. (2019). Their study confirmed the feasibility of utilizing CO_2 hydrates to improve food processing methods. Additionally, as investigated by Peters et al. (2010) CO_2 hydrates have been used to create innovative food products, such as CO_2 clathrate hydrate frozen desserts (Peters et al., 2010).

3.2. Fundamental of ethylene gas hydrates

Ethylene gas is known to form gas hydrates and structure sI, such as gas molecules only occupying the M cage and not occupying the S cage. It is extensively used as an industrial gas. Gas hydrate research using ethylene gas has focused on formation kinetics and separation studies from different gas mixtures (Liu et al., 2012; Ma et al., 2001; Manteghian et al., 2013; Sugahara et al., 2000; Tumba et al., 2013; Wei et al., 2014). It is expected to observe that binary gas hydrate undergoes a phase transition from sI to sII and returns in a specific composition range. Ma et al. (2001) reported that the ethylene-containing gas mixture does not undergo any phase transition; other researchers also support this observation using the spectroscopy technique (Sugahara et al., 2003). In a recent study (Dartois, 2021), authors have used infrared spectroscopy to study ethylene hydrates. Using a cryogenic cell and high-pressure ethylene gas, ethylene clathrate hydrates were synthesized under specific conditions of 31 and 40 bar, corresponding to different ice film thicknesses, to stabilize them above their typical formation curve. Infrared spectroscopy from 6 K to 160 K revealed that ethylene predominantly occupies the larger cages of Type sI clathrate hydrates ($5^{12}6^2$ structure).

Ethylene gas hydrate application in food processing is scarce. For example, ethylene gas hydrates were used for orange juice concentration; it was found that hydrate formation curves were distinctively different from CO_2 hydrates. This stems from the secondary nucleation step during ethylene hydrate formation (Li et al., 2017). The thermodynamics data for pure ethylene gas hydrate is also reported in the literature. (Snell et al., 1961). Other researchers also report and calculate the dissociation pressure of ethylene hydrates through modelling (Kvamme and Tanaka, 1995).

3.3. Fundamentals of ozone gas hydrates

Ozone is a strong oxidizing agent used in food sterilization and deodorization. Due to its lower environmental impact, it offers a potential replacement for chlorine and alcohol (Khadre et al., 2001). Ozone also tends to decompose into oxygen, and its unstable nature poses challenges for transportation and storage. Consequently, hydrate technology has emerged as an effective method for stabilising it. (Mcturk and Waller, 1964; Vysokikh et al., 2008). In an experiment conducted under practical conditions at 253 K and atmospheric pressure (Nakajima et al., 2011)After three weeks, ozone hydrate preservation achieved an ozone concentration of approximately 0.01 in mass fraction. This method yielded a concentration at least 100 times higher than other technologies, such as ozonated water and ozone ice, even though the ozone hydrate was stored for three times as long.

In a recent study (Shishido et al., 2014), three-phase equilibrium conditions were measured for systems containing O_3, O_2, CO_2, and H_2O, with varying $O_3 + O_2$-to-CO_2 molar ratios in the feed gas to optimize the formation and stability of the hydrates, thereby increasing the potential storage capacity of ozone. The experiments, which occurred within a temperature range of 272–277 K and pressures from 1.6 to 3.1 MPa, successfully increased the maximum storage capacity to 2.15 mass%,

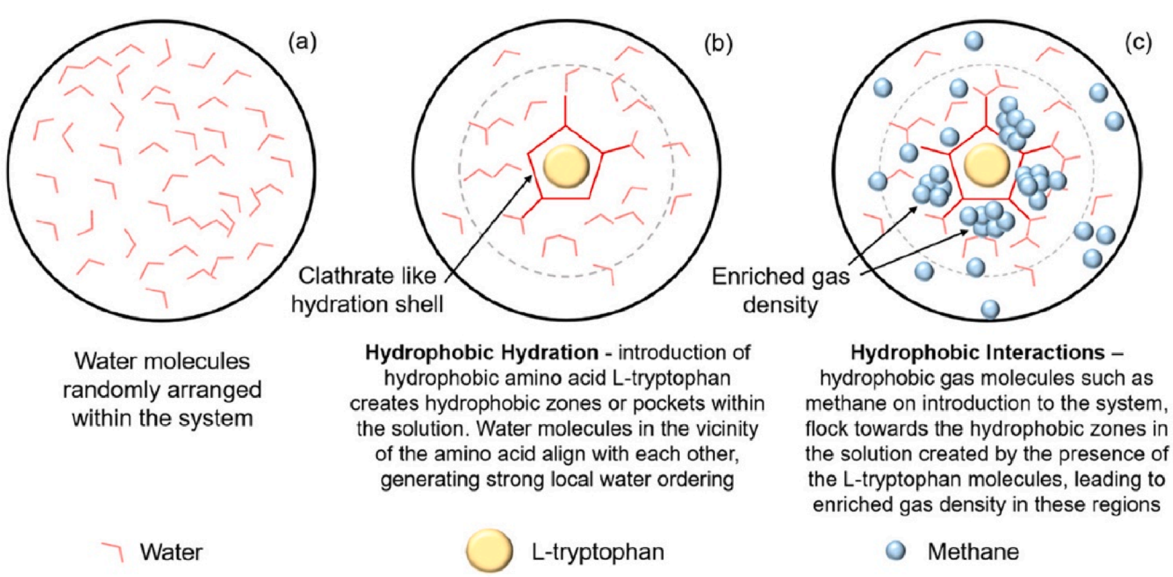

Fig. 6. The proposed mechanism through which amino acids act as kinetic hydrate promoters. Picture reproduced from ref. (Bhattacharjee and Linga, 2021), with permission from American Chemical society (ACS).

significantly improving over previous records (Shishido et al., 2014).

There are attempts at thermodynamics modelling and experimental work to understand the thermodynamics of O_3 gas hydrates (Muromachi et al., 2013; Watanabe et al., 2018). A thermodynamic phase equilibrium study (Muromachi et al., 2012) provides detailed three-phase equilibrium data across temperatures from 272 to 279 K and pressures up to 4 MPa, for hydrates containing ozone, formed from a gas mixture of ozone (O_3), oxygen (O_2), and carbon dioxide (CO_2) coexisting with water (H_2O). The research introduces a modified pressure-search method tailored for the unstable nature of ozone and the high solubility of CO_2. Thermodynamic modeling for ozone clathrate hydrates (Muromachi et al., 2013) used the van der Waals and Platteeuw statistical thermodynamics framework. The study used the Patel–Teja–Valderrama equation of state for precise gas phase ozone estimations. It determined the Kihara intermolecular potential parameters specific to ozone, enhancing the model's accuracy in predicting phase-equilibrium properties. The study matches the predicted data with experimental results, showing less than a 2% deviation in equilibrium pressure, confirming the model's reliability. The results demonstrated that ozone can be more effectively incorporated into the hydrate structures than oxygen (Muromachi et al., 2013).

Crystal growth in O_3-containing hydrates was also observed using O_2, O_3, and CO_2 gas mixtures, classifying it into lateral growth along the gas/water interface and growth into the bulk liquid water phase. Based on the subcooling temperature, the crystals' morphology varied from polygonal to dendritic. These findings have implications for O_3-containing hydrate production plants, enhancing applications in water purification, food preservation, and deodorization (Matsuura et al., 2020).

Efforts are also made to develop bench-scale continuous production of $O_3+O_2+CO_2$ mixed hydrate to enhance long-term ozone storage (Hatsugai et al., 2020). The study focused on optimizing parameters such as gas flow rate, circulating water flow rate, temperature, and heat input to maximize ozone concentration in hydrates, achieving an O_3 storage capacity of 0.26 wt% in 2 h. X-ray diffraction confirmed the successful incorporation of ozone into the hydrates. Preservation tests demonstrated that these hydrates could be maintained for up to six months, significantly outperforming other ozone preservation methods, making this technology an asset for industrial applications requiring stable and prolonged ozone storage (Matsuura et al., 2020; Muromachi et al., 2010).

4. State of the art: gas hydrates in food science

While gas hydrates have primarily been studied in the context of energy production, such as for natural gas extraction, carbon dioxide sequestration, and climate change mitigation, their application in food science is still experimental and unproven. Numerous challenges must be overcome before gas hydrates can be commercialized in food products, including ensuring their stability, safety, and scalability. Additionally, obtaining governmental approval and consumer acceptance is essential for developing this novel food chain component.

The technological readiness levels (TRL) for gas hydrate applications in food science vary; some methods have been explored experimentally, while others are still in their infancy. Continuous research into gas hydrate properties and roles in food applications is ongoing (Claβen et al., 2019; Srivastava et al., 2021; Yu et al., 2021). Researchers have investigated carbon dioxide gas hydrates in several food-related processes, yet their applications remain sparse. While no fundamental barriers exist to using CO2 hydrates, significant advancements are required for widespread adoption. Nonetheless, various areas remain where gas hydrates might be studied for food-related uses (Fig. 7).

4.1. Concentration of juices/sugar and other solutions

CO_2 hydrates have specific implications for the beverage industry, particularly in juice concentration processes (Claβen et al., 2020; Doubra and Naidoo, 2021; Ghiasi et al., 2020; Li et al., 2015a, 2015b). The ability to concentrate juice using CO_2 hydrates can lead to more efficient and environmentally friendly production methods. While these applications are promising, challenges remain regarding scalability, economic viability, and the technological integration of CO_2 hydrates in existing systems.

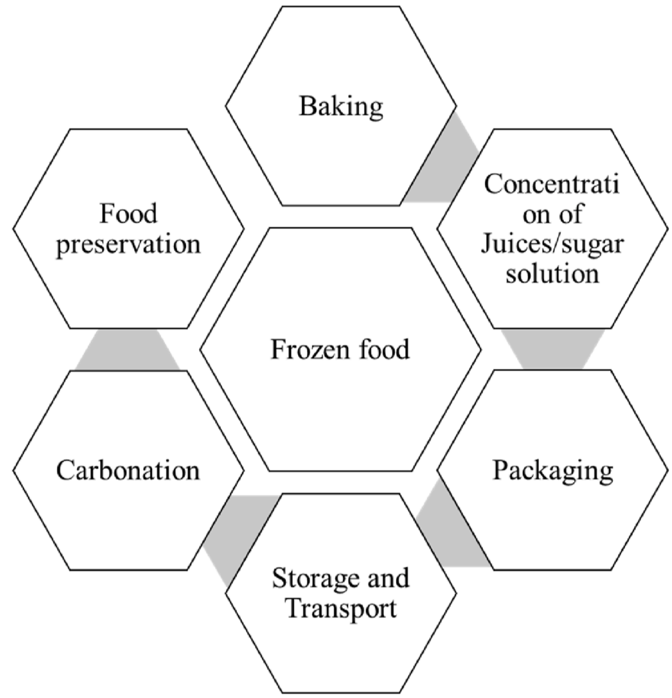

Fig. 7. Areas of food science and technology where gas hydrate-based processes have been investigated.

The formation of CO_2 hydrates in the presence of sugars and other food components is essential for their applications in the food industry. These components can influence the kinetics and stability of CO_2 hydrates. In some cases, sugars may act as promoters or inhibitors of CO_2 hydrate formation. The specific effect depends on the type of sugar, its concentration, and the conditions under which hydrate formation occurs (Claβen et al., 2020; Doubra and Naidoo, 2021).

It was reported that glucose and fructose inhibit the formation of CO_2 hydrates thermodynamically. (Chun and Lee, 1999; Doubra et al., 2021; Tan et al., 2014). Shigehara and Ohmura (2022) investigated the crystal growth of CO_2 hydrate in aqueous fructose solutions. Their study demonstrated the potential of using CO_2 hydrates in carbonated solid foods, indicating how sugars can modify the growth characteristics of CO_2 hydrates. Nagashima et al. (Nagashima et al., 2016, 2017) investigated the preservation of CO_2 hydrates in the presence of fructose/glucose/trehalose. The viscosity of sugar solutions and transition in hydrate dissociation mode significantly influence the preservation (Sato et al., 2014).

Several researchers conducted significant research on the concentration of fruit juices (Table 1) (Claβen et al., 2020; Li et al., 2015a, 2015b; Seidl et al., 2019). The hydrate formation and dissociation for orange and apple juice were carried out in an optical cell. The hydrate dissociation enthalpy for the orange and the apple juice was found to be in the same range as water and CO_2 systems. Despite the demonstration of proof of concept, substantial progress is required to reach industrial-level validation and commercialization of gas hydrate-based fruit juice concentrations. This new hydrate-based and novel separation process for juice concentration has two distinct advantages: it protects more beneficial components while using less energy. As a consequence, it is a technology that has the potential to contribute to sustainability while also positioning itself as a viable alternative to current juice concentration processes such as evaporation, freeze crystallization, and membrane separation. While the number of juice systems that have been researched is still small, it appears that carbon dioxide is the hydrate former of choice in juice concentration. As a result, additional studies are needed to prove the long-term viability of this unique technique.

Besides juice concentration, gas hydrate application in coffee solution processing has also been explored. A review paper covering different juice systems and their thermodynamics and kinetic studies acknowledged insufficient experimental data (Nkosi et al., 2023), and they recommended further investigation concerning dissociation and kinetic data that could be useful for commercialising this technology and developing thermodynamic and kinetic models relevant to this novel concentration process. While going through the research papers on fruit juice concentration, it was noted that such studies have not investigated the microbial stability of juice after the gas hydrate-based processing. Thus, further investigation is required to address this shortcoming. The most recent studies on gas hydrate-based juice concentration are presented in Table 2.

In a recent publication (Doubra et al., 2020), the authors introduce a new 750 cm^3 pilot test rig that employs the isochoric pressure method for concentrating sucrose solutions through gas hydrate measurements. The study reveals that sucrose acts as a kinetic inhibitor during this process, with experiments conducted at various concentrations and temperatures. Additionally, an enhanced agitation system in the reactor allows for the sampling of sucrose solutions, resulting in a final product of approximately 60° Brix.

In another paper (Purwanto et al., 2001), xenon hydrate formation was studied in distilled water and coffee solutions, focusing on developing a method to concentrate liquid foods by utilizing Xenon gas hydrate formation—an alternative to freezing concentration that requires less energy. The experiments reveal that the induction time for xenon hydrate formation depends on the solution's concentration, with higher concentrations requiring longer times. Additionally, the growth of xenon hydrate crystals is influenced by temperature and pressure conditions, where lower and higher temperatures lead to larger crystals. The paper concludes that the size of xenon hydrate crystals is crucial for efficient separation in the concentration process, and these findings could contribute to more energy-efficient methods in the food industry.

In another recent study (Sato et al., 2014), CO_2 hydrate self-preservation coexisting with sucrose under domestic freezer conditions was studied. The research indicated that lower sucrose mass fractions and lower temperatures contributed to better preservation of CO_2 hydrate. Particle size had no significant impact on preservation. The study suggests the potential for developing novel carbonated solid foods using CO_2 hydrates with sucrose, as the preserved hydrates maintained a mass fraction above 0.02, equivalent to carbonated water, even after three weeks of storage.

In terms of thermodynamic modeling, recent research (Rasoolzadeh et al., 2022) outlines a precise thermodynamic approach for assessing the stability conditions of clathrate hydrates in the presence of environmentally friendly sugar-derived compounds. The methodology employs the free-volume modification of the Flory-Huggins model for water activity calculation and the modified van der Waals-Platteeuw approach for modeling the hydrate phase. The study accurately predicts hydrate dissociation conditions by testing this approach on 29 systems with 11 hydrate formers and 11 sugar-derived compounds.

4.2. Baking

Food innovation is frequently motivated by the current population's demand for change. In addition to ease of use, people are seeking healthier alternatives to their present eating options. As science advances, more studies on using unusual substances may be conducted. Recent trends in bread-making are related to the use of fiber (whole grain) or other functional ingredients to develop new functional bakery products. Srivastava et al. (2022b, 2023a,b) attempted some of the recent developments in baking with the application of gas hydrates (Srivastava et al., 2022b, 2023a, 2023b). The research attempted to use CO_2 gas hydrates in wheat bread and found that instead of yeast, which provides CO_2, the bread can be leavened by CO_2 supplied by the gas hydrates (Table 3).

Table 2
Summary of recent studies that incorporate gas hydrates application for fruits-based processing.

Juice Type/Gas Hydrate former	Key finding	Reference
Tomato, apple, and orange CH_3Br; CCl_3F	• Concentration of tomato, apple, and orange juice by CH_3Br and CCl_3F hydrates •Color and flavor loss with a bitter aftertaste in juices	Huang et al. (1966)
Coffee Solution Xenon (Xe)	•Concentration of Coffee Solution using xenon (Xe) GH at 9 °C with a Xe partial pressure of 0.9 MPa •Increased stirring rate resulted in more Xe hydrate formation but increased temp. to 14 °C lowered conc. efficiency	Purwanto et al. (2014)
Apple juice	•Concentration of apple juice at a pressure range of 30–80 × 10^5 Pa and Temp. range of 274–283 K •Apple juice concentration was achieved at 21–27° Brix, indicating 60% of the juice was trapped inside the GH	Seidl et al. (2019)
Apple juice	•Concentration of apple juice to high sugar content at 35 bar pressure •Juice was concentrated until 45° Brix, and the formation of hydrates interfered due to the chemical composition of juice when 30° Brix was achieved	Claβen et al. (2020)
Orange juice CO_2 gas hydrate	•An increase in dehydration ratio (DR) was observed with an increase in the pressure of feed. A maximum DR of 57.2% was achieved at pressure 4.1 MPa of feed	Li et al. (2015a)
Tomato juice CO_2 gas hydrate	•Tomato juice concentration by CO_2 Hydrate •Maximum DR of 65.2% at pressure 3.95 MPa of feed. Also, the DR increased when feed pressure was increased from 1.81 to 3.95 MPa.	Li et al. (2015b)
Coffee Solution CO_2 gas hydrate	•CO_2 hydrate kinetics using the presence of coffee aqueous solution. •Kinetic experiments, P = 30 bar, T = 275.15 K, Coffee solution show inhibiting effect. Wt% tested = 1.5, 5, 10, 15 w/w%. An increase in coffee concentration decreases induction time and does not directly affect the total CO_2 hydrate storage capacity.	Abedi-Farizhendi et al. (2020)
Frozen desserts CO_2 gas hydrate	•CO_2 GH was used as a carbonation compound in frozen desserts •The bubbling effect was created due to CO_2 being released during thawing or consumption	Peters et al. (2013)
Bitter melon; Grape; Bitter melon CO_2	•A dehydration ratio of 57% was obtained in this study •Observation of the inhibitory effect of juice components on carbon dioxide hydrates	(Nkosi et al., 2022a, 2022b)

By varying the percentage of gas hydrates from 10 to 60%, the performance of CO_2 GH as a leavening agent during baking was done. The effectiveness of GH bread was evaluated by comparing its characteristics to those of standard bread made with yeast (Srivastava et al., 2022b). The next part of the research attempted by Srivastava et al. (2023a) dealt with a comparative analysis of the partial replacement of yeast with CO_2 GH as leavening agents in bread baking. By partially eliminating the

Table 3
Application of gas hydrates to baked products.

Baking/gas hydrate type	Key observations	References
Baked bread with CO_2 GH	The results of the study showed that the bread with 60 % and 70 % GH had the best outcomes when compared to standard yeast bread	Srivastava et al. (2022b)
Partial yeast and CO_2 GH in combination for wheat bread	The best combinations were obtained with 70 % GH + 50 % yeast, and 70 % GH + 75 % yeast	Srivastava et al. (2023a)
Improvement in textural effects with additives in wheat bread	The incorporation of some natural additives or flour improvers in the bread enhanced the textural properties of the wheat bread made with GH	Srivastava et al. (2023b)
Baked black and white cookies with CO_2 GH	The best results were obtained using 50% GH and kneading the dough at a high temperature.	Kollemparembil et al. (2023)

yeast, variations of bread dough were produced by adding GH in different percentages (20–70%). The effectiveness of GH on bread manufacture was evaluated by comparing its characteristics to standard bread made with yeast. The best results were obtained by combining specific volume, pore analysis, and hardness with 70% GH + 50% yeast and 70% GH + 75% yeast, respectively. Another study conducted by Srivastava et al. (2023b) dealt with the influence of additives on wheat bread baked with promoter-induced CO_2 GH as leavening agents. The additives used for the study were ascorbic acid (AC), egg white (EW), and rice flour (RF). These additives were added to the GH bread containing different amounts of GH (40, 60, and 70% GH). Also, a combination of these additives in a wheat GH bread recipe was studied for each respective percentage of GH. Based on the study's results, it was found that 70% GH + AC + EW + RF wheat bread was the best in terms of textural analysis, pore size analysis, and other physiochemical parameters. This study will motivate numerous academics and industries to explore and seek a better understanding of GH application to the bread-baking process by replacing conventional baking agents such as yeast (Refer to Fig. 8).

Another study related to baking was conducted by Kollemparembil et al. (2023), examining the use of CO_2 gas hydrates (GH) in black-and-white cookies. Optimal results were achieved using 50% GH and kneading the dough at high temperatures, contrasting with the baking of traditional cookies. Additionally, the acrylamide content was significantly reduced by replacing ammonium bicarbonate with GH. However, further research is required to produce the same quality as conventional black-and-white cookies. Therefore, if CO_2 hydrates are used, the baking process (the entire step to make the dough) will be shortened (to less than an hour). This leads to consistent production, minimal downtime, and no requirement for expensive storage space. It also results in no ethanol production or release during baking, as yeast is not utilized. Other benefits include the controlled decomposition of gas hydrates (GH), which can significantly reduce the energy required to maintain ambient conditions in the dough proofing stage, compared to biological leavening with yeast. Additionally, the time needed for gas release is reduced to just a few minutes. Unlike biological leavening, a finite supply of leavening agents is available (Srivastava et al., 2022b, 2023a, 2023b), allowing for the decoupling of dough leavening from aromatic dough ripening. Only carbon dioxide and water are released during the disintegration of gas hydrates (GH) (Srivastava et al., 2022b), thus preventing the sensory changes in product quality—such as color and taste—often observed when using high doses of chemical leavening agents are used. The physically controlled disintegration of hydrates in the dough can be reliably halted by freezing and reactivated later without affecting the release rate. Consequently, compared to yeast fermentation, this process is less impacted by chilling rates and freeze durations. Moreover, the timing of the baking process becomes more flexible and can be optimally adjusted to meet specific requirements, especially useful in small and branch bakeries (Srivastava et al., 2023a, 2023b).

4.3. Carbonation and solid carbonated compounds

CO_2 in carbonated food can be perceived through visual, audible, chemical, and tactile stimulation involving bubble formation and bursting (Peters et al., 2013). Table 4 summarises some of the key publications in frozen desserts using CO_2 hydrates. CO_2 in carbonated beverages is mainly through chemical stimulation of nociceptors and sour taste receptors activated by carbonic anhydrase present in the mouth that catalyses CO_2 and H_2O reactions (Chandrashekar et al.,

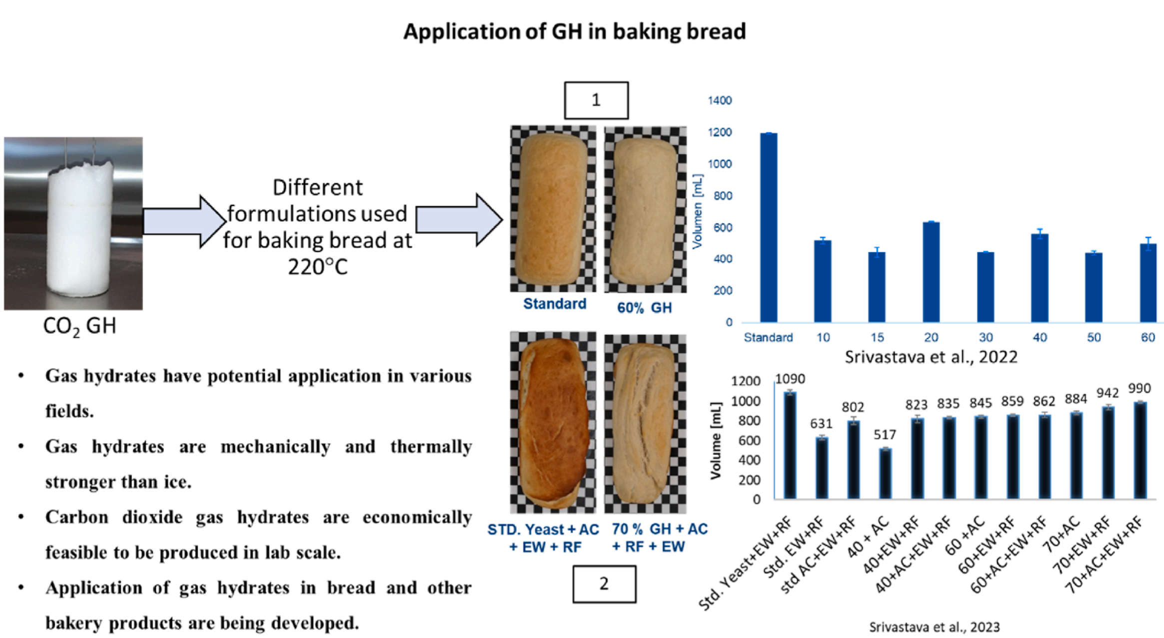

Fig. 8. Summary of recent baking research using CO_2 gas hydrates.

Table 4
Summary of key publications in frozen desserts using CO_2 hydrates.

Frozen/gas hydrate type	Key observations	References
Frozen dessert CO_2 gas hydrate	CO_2 clathrates hydrate the frozen desserts by flash freezing Minimum pressure = 47 bar in ICT (Ice confection tank) Particle size = 10–50 um	Peters et al. (2010)
Frozen desserts CO_2 gas hydrate	Transfer process limited models for CO_2 perception in CO_2 hydrate desserts Heat-limited and mass-limited dissociation models explain the bubbling effect of releasing CO_2 during thawing or consumption	Peters et al. (2013)
Frozen desserts CO_2 gas hydrate	Pressure variation due to heat shock for CO_2 hydrate dessert The sealed package can reach pressure greater than Ice-CO_2 hydrate equilibrium pressure, and heat shock may cause a fraction of water to crystalize in the desert	Peters et al. (2012)

2009; Dessirier et al., 2000) rather than tactile stimulation (CO_2 bubble bursting against the tongue). CO_2 perception in carbonated products differs from carbonated beverages as there is a strong sense of bubble formation and bursting against the tongue and palate as the product melts in the mouth (Peters et al., 2013). CO_2 hydrates can deliver carbon dioxide into beverages, frozen desserts, and foods, resulting in the desired carbonation effect in carbonated drinks and frozen carbonated food. By encapsulating carbon dioxide within gas hydrates, creating a regulated and extended release of the gas may be feasible, resulting in a distinct and long-lasting effervescence in drinks. Thus, carbonated beverages may be made without carbonation equipment by encapsulating carbon dioxide within gas hydrate structures. CO_2 hydrate application in frozen desserts is driven by the release of trapped CO_2 from the CO_2 hydrate dissociation, causing strong bubble formation and bursting against the tongue and palate as the product melts in the mouth, resulting in enthusiasm. Sensory panel testing confirmed that CO_2 hydrate-based desserts could be perceived as more carbonated than freshly opened soda (Peters et al., 2013). CO_2 hydrate desserts offer a unique and intense carbonation experience due to the higher CO_2 concentration, and the perception of carbonation depends on the dissociation and melting process in the mouth, which differs from traditional sodas. A key mechanism involves chemical stimulation of receptors in the mouth and tactile stimulation involving bubble formation and bursting.

To connect sensory experience with crystal development, researchers (Shigehara and Ohmura, 2022) investigated CO_2 hydrate crystal growth in aqueous fructose solutions (refer to Fig. 9), aiming for potential applications in carbonated solid foods. The study explores how crystal morphology, such as size and shape, affects the texture of foods. It found that different temperatures and pressures during the formation process led to various crystal shapes, which can influence the sensory experience of carbonated food products.

Solid carbonated foods (jellies, gummies) are not as widely consumed as carbonated beverages (soda, beer) due to low CO_2 solubility in ice or other edible solids (Nagashima et al., 2017). This is due to the high concentration of CO_2 in CO_2 hydrates, which is 50 times more than that in carbonated water, which is 6 kg/m^3 at P = 3 bar, T = 283.15 K (Dodds et al., 1956).

5. Potential application of gas hydrates in food technology

5.1. Food preservation

Food preservation and shelf-life extension for fruits and vegetables can be achieved through freezing, which inhibits biological activities by reducing water mobility within living cells. However, ice formation can damage tissue due to ice morphology and volume expansion. Research by Takeya et al. (2016) shows that CO_2 hydrates can preserve fresh produce by suppressing water mobility and forming a protective barrier against spoilage organisms, thereby reducing oxidation and microbial growth (Refer to Fig. 10). This suggests that gas hydrates could reduce the reliance on chemical preservatives and extend shelf life. These studies should be further extended to seafood and meat industry. The

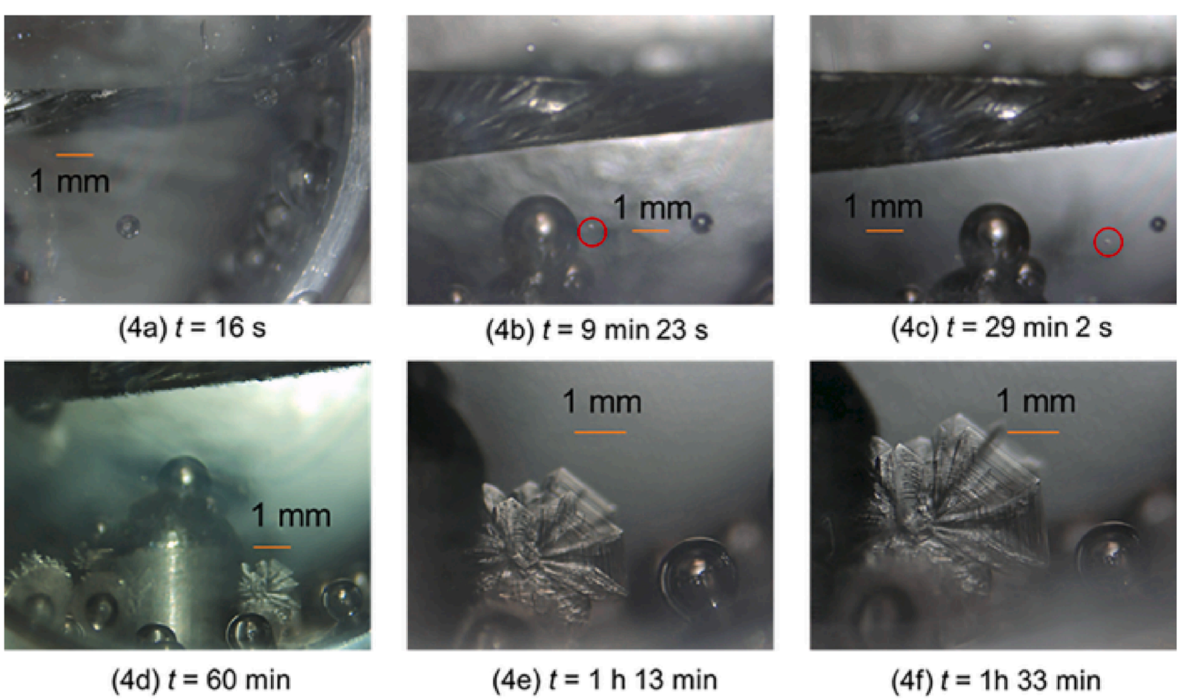

Fig. 9. Sequential images show the growth of CO_2 hydrate crystals in an aqueous fructose solution at T_{ex} = 277.0 K, ΔT_{sub} = 2.0 K, and P = 3.4 MPa. Below each image, the time elapsed after the initial hydrate crystal was visually recognized in the experimental system is displayed. The hydrate crystals detached from the hydrate film are enclosed in red lines. (Shigehara and Ohmura, 2022).

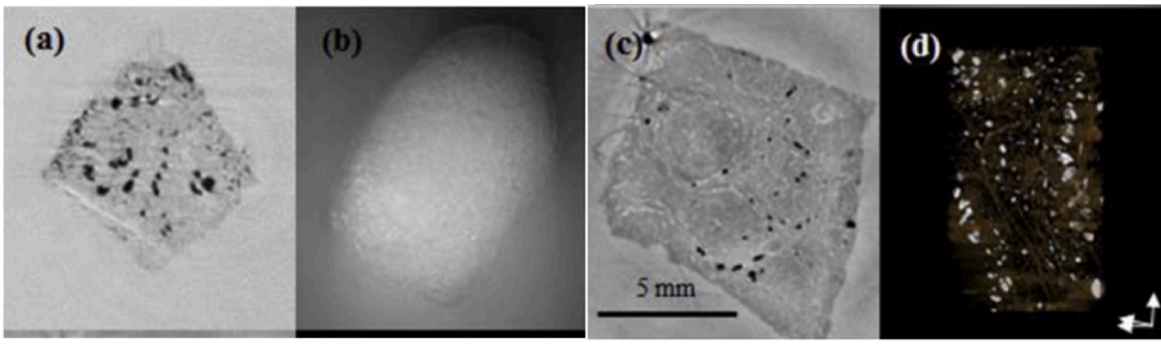

Fig. 10. DEI images of fresh produce. (a) Eggplant, (b) broad bean and (c) cucumber samples measured at −80 °C. White indicates higher-density regions and corresponds to CO_2 hydrate. Black indicates lower-density regions, which are largely pore space. (d) Three-dimensional surface rendered image of CO_2 hydrate regions (orange) within (c). (Reprinted with permission from Elsevier, Copyright (Takeya et al., 2016) Elsevier.

dissociation of gas hydrates also generates micro- and nanobubbles (Uchida et al., 2016) and Nanobubble technology is increasingly becoming popular in food processing (Thi Phan et al., 2020). These micros and nanobubbles provide a high surface-to-volume ratio, enhanced mass transfer, and maintain a high CO_2 concentration, which could be used to improve shelf life and prevent rotting. Therefore, further research should consider factors such as cost-effectiveness and the nature of the food products, carefully considering using gas hydrates.

Authors suggest that integrating freezing, cold plasma, and high-pressure processing with gas hydrates could enhance food preservation, though the specifics of these interactions and potential benefits require further exploration. For example, high-pressure processing could produce selected gas hydrate in situ, highlighting areas for future research and development.

5.2. Encapsulation

Gas hydrate crystals, known for their ability to encapsulate gases like CH_4, CO_2, nitrogen, and liquids such as THF, DIOX, and CP (Majid et al., 2021; Nasir et al., 2020), have the potential for broader applications in food and skincare products. Their water cages can effectively contain and deliver tastes, Flavors, fragrances, and volatile chemicals, extending shelf life and enhancing stability. For instance, the encapsulation of 2-propanol with CO_2 in hydrate cages shows promise in skincare (Maruyama et al., 2024). In another example of encapsulation, researchers demonstrate the synthesis and characterization of sI-type clathrate hydrates containing carbon dioxide and ethanol, highlighting its potential applications in alcoholic food and drink products (Makiya et al., 2010). Additionally, engineered gas hydrates could be used as beads or particles for controlled aroma and flavour release, enhancing the sensory experience of food. This controlled release mechanism preserves delicate flavors from deterioration and maintains a consistent flavor profile. Furthermore, gas hydrates could serve as carriers for bioactive compounds in food, protecting vitamins and minerals during processing and storage and enhancing their bioavailability upon digestion.

5.3. Frozen foods

Due to their stability in a frozen state, gas hydrates can significantly enhance the freezing of food items, improving texture and structural integrity while minimizing reliance on traditional freezing methods. Understanding the development and behaviour of gas hydrates during freezing processes can help optimize food compositions and overall product quality, potentially extending shelf life and preserving food freshness. This includes the controlled release of encapsulated gases like nitrogen, carbon dioxide, and oxygen, which can reduce freezer burn. While traditional applications utilize these gases in their gaseous or dissolved forms, integrating them into hydrate structures for use in flash freezing, cryopreservation, and modified atmosphere, packaging could offer new ways to improve food preservation. Research into the applications of gas hydrates for flash freezing (Peters et al., 2010), and cryopreservation is ongoing.

5.4. Textured foods

Gas hydrates can enhance the texture and structure of aerated food products by introducing a light, airy texture to items such as mousse, ice cream, cake mixes, dairy products, and baked goods (Adhikari et al., 2018; Frühling et al., 2023). The regulated gas release from hydrates can provide a foaming effect, improving mouthfeel and contributing to the desired structural formation in sweets, confectioneries, and dough-based products. Manipulating the production and dissociation processes of gas hydrates offers a method to adjust food textures effectively.

5.5. Refrigeration for food storage and transportation

Gas hydrates have potential in industrial refrigeration, as their formation can produce cooling effects beneficial for cryogenic freezing processes. For instance, CO_2 hydrate is a two-phase refrigerant offering an alternative to conventional methods (Fournaison et al., 2004a). In food companies, gas hydrates can be explored as potential storage and transportation media for gases like natural gas. Though not directly related to food, natural gas transport can enhance the efficiency and environmental friendliness of natural gas transport widely used in food processing. Additionally, gas hydrates show promise as cooling agents in refrigeration systems, potentially improving energy efficiency and temperature control in food storage and transportation within the cold chain (Ogawa et al., 2006; Zhang et al., 2017).

5.6. Packaging

Gas hydrates could revolutionize food packaging by enabling controlled release of gases like carbon dioxide or nitrogen, which can extend the shelf life of perishable items by creating a modified atmosphere that inhibits microbial growth and oxidation. This concept, known as Modified Atmosphere Packaging (MAP), benefits from the stable encapsulation of gases within hydrate structures, allowing for a slow and controlled release. Employing gas hydrates in MAP, especially at temperatures below 273.15 K, can maintain the desired gas composition for extended periods. This technology is promising for packaging perishable foods such as fruits, vegetables, meat, and seafood, making the food packaging sustainable and effective at preserving freshness.

6. Competitive positioning of gas hydrate technology

Gas hydrates are promising for numerous industries and national economies, with gas separation, carbon dioxide sequestration, and desalination applications. However, their widespread adoption is hindered by high costs, significant electricity consumption, and a slow growth rate of gas hydrates (Gaidukova et al., 2023a). Nonetheless, gas hydrate-based carbon dioxide separation, capture, utilization, and storage technologies offer advantages over traditional technologies, such as higher energy efficiency, larger separation capacity, and environmental friendliness. However, the slow formation rate of hydrates remains a barrier to their industrial application (Misyura et al., 2023).

Solid carbon dioxide containing moisture has diverse applications, including cooling and refrigeration. Yet, when it contacts moisture, it forms carbonic acid, which is corrosive. Safety precautions are essential when handling solid carbon dioxide due to its extremely low temperature (Hassanpouryouzband et al., 2020).

In the beverage industry, carbon dioxide-water mixtures or carbonated water are common. When carbon dioxide dissolves in water, it forms carbonic acid, giving carbonated drinks their characteristic fizzy and spicy taste (Xu and Zhang, 2022). These versatile mixtures are used in various applications, from beverage production to fire suppression systems. They can be easily made by dissolving carbon dioxide gas in water under pressure (Yu et al., 2021; Zhou et al., 2021).

Despite their broad utility, carbon dioxide-water mixtures are comparatively limited in application scope and contribute to greenhouse gas emissions when the carbon dioxide is released (Xu and Zhang, 2022; Yu et al., 2021). Table 5 presents a detailed comparison of gas hydrate technology and other CO_2 and water mixtures, focusing on factors like stability, efficiency, controllability, cost, and environmental impact. In conclusion, while gas hydrate technology, CO_2-water mixtures, and solid CO_2-containing moisture each offer unique advantages and potential applications, they also face significant challenges.

Table 5
Comparison between gas hydrate technology and other CO_2 and water mixtures.

	Gas hydrate technology	Other CO_2 and water mixtures	References
Stability	stable under certain conditions of pressure and temperature	stable under normal conditions of pressure and temperature	(Hassanpouryouzband et al., 2020; Xu and Zhang, 2022; Yu et al., 2021)
Efficiency	high energy efficiency	dissolving CO_2 in water is relatively efficient and straightforward	(Gaidukova et al., 2023a; Yu et al., 2021)
Controllability	formation and dissociation of gas hydrates can be controlled by adjusting the pressure and temperature conditions	CO_2 dissolved in water can be controlled by adjusting the pressure and temperature conditions	(Gaidukova et al., 2023a; Zhou et al., 2021)
Cost	high due to the high-power consumption and low growth rates of gas hydrates	producing CO_2-water mixtures is relatively low compared to gas hydrate technology	(Gaidukova et al., 2023a; Misyura et al., 2023)
Environmental Impact	reduce greenhouse gas (CO_2, CH_4) emissions and have excellent application potential	release of CO_2 from carbonated water contributes to greenhouse gas emissions.	Hassanpouryouzband et al. (2020)

7. Potential challenges

While gas hydrates show promise for various applications in the food industry, further research and development are needed to ensure their safety, effectiveness, and practicality. A concerted effort is needed to validate the sustainability and scalability of gas hydrate technology. Challenges to applying gas hydrates in food science and engineering include cost, infrastructure requirements, and workforce training. Implementing gas hydrate technology may involve significant initial investments in equipment and infrastructure. Training personnel to operate and maintain gas hydrate systems is also critical.

To properly assess the potential of gas hydrates in food science, it is necessary to investigate the technological, economic, and regulatory obstacles connected with their use. The challenges that should be considered can be broadly categorized as technical, economic, and regulatory (Refer to Fig. 11), and a detailed discussion is in Table 6.

7.1. Safety and regulatory considerations associated with gas hydrates in the food industry

The potential use of gas hydrates in the food sector raises concerns about food safety and regulatory issues. It is essential to manage the supply chain correctly to ensure that food remains safe for consumption. Quality assurance processes and control measures should be implemented when working with gas hydrates in food industry settings (Prasanna et al., 2024). The food sector's sustainability drive has led to significant improvements in reducing greenhouse gas emissions, enhancing transparency along value chains, and promoting responsible sourcing practices. Using gas hydrates should align with these sustainability goals (Varghese et al., 2022). These examples highlight the safety and standard-setting considerations for applying gas hydrate technology in the food sector. Thorough investigation and consultation with specialists are necessary before adopting these methods. Strict quality controls are required to ensure food safety when using gas hydrates in the food industry (Yu et al., 2021).

8. Future road map

Fig. 12 presents a roadmap for future studies in the field of gas hydrates in food science and engineering. This roadmap highlights the necessity of fundamental research and the development of new formulations to enhance our understanding of hydrate formation. Specifically, comprehensive studies on the kinetics and thermodynamics of gas hydrate formation in food systems are needed. Developing new gas hydrate formulations that account for the impacts of various gases (CO2, CH4, N2O) on hydrate stability, considering the unique properties of different foods, is crucial.

Additionally, studying the stability of natural gas hydrates under varying conditions (temperature, pressure, and composition) is indispensable. Process optimization should focus on refining parameters such as pressure, temperature, and mixing conditions to improve food properties. Scaling up from laboratory to industrial scale requires efficient and cost-effective equipment designs for gas hydrate production and utilization in food processing.

Furthermore, the impact of gas hydrates on food quality and safety must be assessed, including their effects on texture, taste, and colour. Safety aspects related to storage, transport, and consumption, along with the potential of gas hydrates to inhibit microbial growth and reduce spoilage, must also be considered.

Economic and environmental evaluations are crucial. A cost-benefit analysis should determine the economic viability of gas hydrate technology in food processing, and the environmental footprint of gas hydrate processes should be compared with traditional methods.

As this technology progresses, fostering interdisciplinary collaboration and knowledge sharing is critical. Engaging food scientists, food and chemical engineers, and materials scientists in joint research, as well as

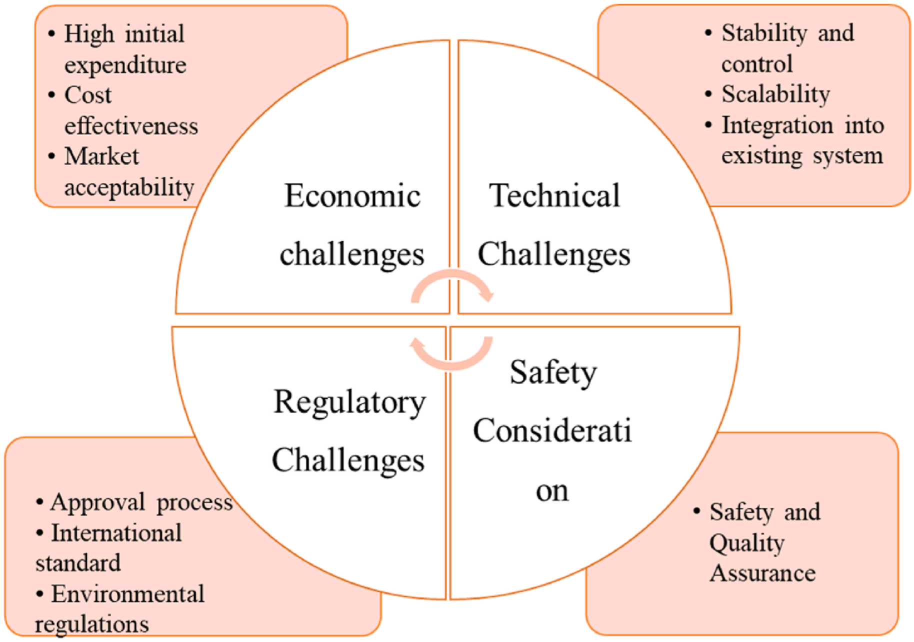

Fig. 11. Information on the technical, economic, and regulatory challenges that need to be addressed before potential applications of gas hydrates in food science.

Table 6
Information about different layers of challenges associated with gas hydrates in the context of food science and outlining recommendations for addressing the challenges associated with the use of gas hydrates in food science and engineering.

Category	Subcategory	Description
Technical Challenges	Stability and Control	The formation and dissociation of gas hydrates must be precisely controlled to ensure stability and effectiveness under varying environmental conditions (temperature, pressure). Sophisticated technology may be required for food preservation and packaging.
	Scalability	Scaling from laboratory experiments to industrial levels presents challenges such as maintaining product quality and consistency.
	Integration into Existing Systems	Integrating gas hydrate technology into existing food processing and packaging systems may require significant modifications, increasing initial costs and operational complexity.
	Safety and Quality Assurance	It is critical to ensure that the use of gas hydrates does not compromise food safety or quality. Rigorous testing and standard operating procedures must be developed.
Economic Challenges	High Initial Expenditure	Implementing gas hydrate technology may require significant upfront investment in research and development, equipment, and training.
	Cost-effectiveness	For widespread adoption, the benefits, such as longer shelf life and higher quality, must outweigh the costs. Economic assessments are essential.
	Market Acceptability	Market acceptability and demand for products using innovative technology like gas hydrates can be unpredictable. Significant marketing efforts may be necessary to build trust and acceptance.
Regulatory Challenges	Approval Processes	New food processing or packaging technologies must undergo rigorous regulatory examinations to ensure safety, including showing that they do not introduce harmful substances into food.
	International Standards	Complying with international standards can be more challenging than meeting local regulations, especially because food products are often exported. Ensuring compliance with global standards is crucial.
	Environmental Regulations	The environmental impact of using gas hydrates, including their production and disposal, must be assessed. Compliance with regulations aimed at minimizing environmental footprints is essential for viability.

Potential Recommendations

Action	Recommendation	Description
Collaboration with Research Institutions	Form partnerships with universities and research centers	Collaborating with academic and research institutions can leverage scientific expertise to address technological challenges and develop cost-effective solutions.
Pilot Studies	Conduct practical experiments	Pilot studies help researchers understand the real-world implications of using gas hydrates in food science, providing insights into practical challenges and market reactions.
Regulatory Involvement	Engage with regulatory bodies early.	Early involvement with regulatory organizations can ensure that gas hydrate technologies comply with existing and upcoming safety standards, potentially accelerating approval.
Economic Analysis	Perform detailed cost-benefit and market analysis	Conducting thorough economic analysis and market research is necessary to determine the financial viability of integrating gas hydrate technology into the food.

disseminating findings through conferences, journals, and workshops, will advance gas hydrate technology in the food industry. By strategically advancing this technology, we can accelerate its adoption and improve food processing efficiency, quality, and sustainability.

9. Conclusion

Gas hydrate-based processes offer promising avenues for energy-efficient and environmentally sustainable food processing methods. However, research into utilizing gas hydrates within food science is still in its early stages. Addressing safety, stability, and regulatory concerns is

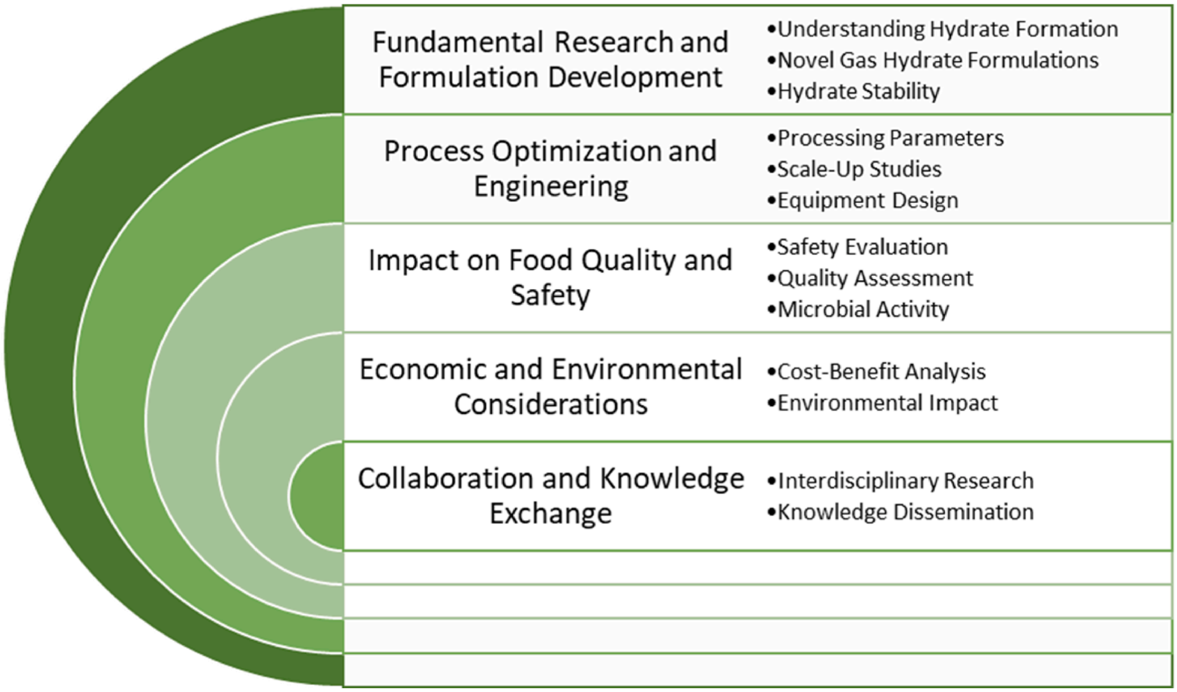

Fig. 12. A roadmap for future studies in the field of gas hydrates in food science and engineering.

essential before gas hydrates can be effectively integrated into food production. Currently, gas hydrates are not widely adopted in food products due to the challenges of forming them at extremely low temperatures and high pressures, which is impractical for typical food manufacturing and storage environments. Nonetheless, advancements in suitable promoters allow gas hydrate formation under more feasible conditions, such as atmospheric pressure and ambient temperatures. It is crucial to restrict hydrate formers to gases considered safe for human consumption.

Carbon dioxide (CO_2) is the most prevalent hydrate former in food industry applications, especially in fruit juice concentration and baking. While research on baking has primarily focused on CO_2 hydrates, exploring other gases for hydrate-based juice concentration is underway. Further investigations are necessary to address the lack of thermodynamic and kinetic data and to clarify the sensory properties and microbial stability after fruit juice concentration.

Promising areas for short-term research include food preservation, encapsulation, frozen food processing, texture enhancement, structural modification, storage optimization, transportation efficiency, and innovative packaging solutions. Finding biocompatible and food-grade promoters to enhance gas hydrate formation kinetics and stability is also essential. Interdisciplinary collaboration among chemical engineers, chemists, physicists, food scientists, and food technologists is crucial for advancing these research areas and developing new hydrate-based technologies for the food industry.

While gas hydrates have been primarily studied for energy production, carbon dioxide sequestration, and climate change mitigation, their application in food science still requires further exploration and development.

CRediT authorship contribution statement

Jyoti Shanker Pandey: Writing – review & editing, Writing – original draft, Project administration, Funding acquisition, Conceptualization. **Shubhangi Srivastava:** Writing – original draft, Conceptualization. **Aberham Hailu Feyissa:** Writing – original draft. **Mohammad Tariq:** Writing – original draft. **Kaniki Tumba:** Writing – original draft.

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

No data was used for the research described in the article.

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