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Review article

Issues and challenges in hydrogen separation technologies

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ABSTRACT

Depleting energy resources, global warming and environmental problems associated with conventional fuels are serious global challenges of the modern world. The substitution of conventional energy resources with more efficient and sustainable resources is inevitable. In this scenario, hydrogen (H_2) has emerged as the ultimate choice due to its superior characteristics such as low carbon emissions, cleanliness, and efficiency. However, for the successful implementation of making H_2 as the next-generation fuel source, the hurdles of production, separation, and storage of H_2 should be resolved. This paper summarizes the issues and challenges in the separation of H_2 gas from various production streams by using available separation technologies. Different types of H_2 separation technologies, including membranes, adsorption processes, metal hydrides, and cryogenic separation technologies, have been considered and discussed. The review encompasses the types, advantages, and disadvantages of each technology, followed by a detailed account of issues and challenges observed in each separation method. More attention has been given to membrane technology because it is the most promising technology for the production of high-purity H_2. Finally, this review provides an outlook for future directions and developments in H_2 separation technologies.

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1. Introduction

The majority of the world's energy requirements are satisfied by fossil fuels, such as oil, natural gas, and coal, as well as the products derived from these fuels. These finite resources play their part in a country's development. They have remained critical because of their contribution to environmental pollution, global warming, and climate change (Wood and Roelich, 2019). The countries that have realized the issues associated with these resources have started working to reduce their economic dependence on fossil fuels. As we know, renewable resources are already a big part of the global energy mix. There are a lot of individuals who are interested in energy who hope that in the future we will be able to extract energy from H_2 in a clean and sustainable manner. Abe et al. (2019). Global demand for pure H_2 is rising. Today, 95 million tonnes of H_2 are produced. Most of that H_2 comes from the steam methane reforming process (76%), while most of the remaining H_2 is produced from coal (22%) (Amin et al., 2022c). Fossil fuels are the core source of H_2 and chemical industries rely on it. H_2 is one of the key elements in modern industry. Primarily, it is being used in four key sectors: oil refineries; ammonia; methanol; and steel production (Megia et al., 2021). Moreover, the uses are widespread in the different segments, such as for hydrotreating biofuels (Vásquez et al., 2017) and upgrading oil sands (Brough et al., 2010). H_2 also has a wide range of uses in food, metallurgical, microelectronic, polymer synthesis, and so many industries (Shamsi et al., 2022). To make H_2 the next-generation fuel source, we must solve H_2 production, separation, and storage challenges. Making H_2 as a final fuel source is still in its early stages. Therefore, many countries are already making progress in their research to make H_2 economies a reality (Nazir et al., 2020b).

As mentioned earlier, the most widely used method for the production of H_2 is steam reforming (SMR) (David et al., 2012). In SMR, the gas is first cleaned, then mixed with steam in a steam reformer to produce CO and H_2 gas as products . Another major source for H_2 production is coal. For the production of H_2 from coal, the coal gasification process is carried out (Amin et al., 2022c). In addition to H_2-based syngas (Amin et al., 2022a), coal-based gasification may produce CO, H_2S, NH_3, ash, tar, HCL, and HCN. Perkins (2018). The produced H_2 gas needs to be purified. Therefore, for the successful utilization of H_2 as an alternative energy source, it is necessary to separate and purify the H_2 from the gases. Most of the time, membrane technology, pressure swing adsorption (PSA), metal hydrides, and cryogenic separation are used to separate H_2 from other gases.

A membrane is a physical obstacle widely used for separation and purification that selectively permits specific species to pass through to the permeate side driven by chemical potential. Permeability and selectivity are two parameters that are important for gas separation. The term "permeability" describes the flux (volumetric flow per unit of membrane). In addition, the term "selectivity" defines the membrane must have to separate the two given molecular species. The most common classification of membranes is organic (polymer) and inorganic (zeolite, metallic, ceramic oxides, and carbon molecular sieves). Fig. 1(a) shows the schematic process description of the membrane (Vermaak et al., 2021). Tables 1 to 6 explains the main issue and challenges of H_2 separation via membrane technology.

PSA is another important separation technology used for mixture gas separation and purification. The PSA process is operated in batch and continuous flow systems. Both of them can be achieved by using multiple adsorbents (Adhikari and Fernando, 2006). Different types of adsorbents, such as activated carbons, zeolites, and molecular sieves, are preferred to absorb high affinity gas species at high temperatures. When swinging to low pressure, the absorbed gases desorb from the adsorbent (Ye et al., 2019). PSA has been employed for a variety of gases, especially for H_2 separation and purification (Zhang et al., 2021). In addition, the impurities in the crude H_2 are removed by pretreatment methods (conventional adsorption, physical absorption and chemical reactions) to remove the specific contaminants. After pretreatment, the removal of both major and minor impurities is achieved by PSA. Fig. 1(b) shows the schematic process description of PSA (Mivechian and Pakizeh, 2013).

Metal hydride is a unique chemical method to purify and separate H_2. The metal hydride method/reaction uses different metal alloys to absorb and desorb the H_2 in the process of purifying H_2 from impurities. The reaction is reversible and its direction is measured by the pressure and temperature of the H_2 gas. The metal hydride absorption/desorption is done in a specially designed reactor called the metal hydride reactor. MH reactors are able to recover H_2 with 99.999% purity. They can be used for small to medium sized industrial applications. A metal hydride H_2-based separator with numerous complex parts, including a metal hydride separator, pressure vessels, pressure meters, and electromagnetic valves. Fig. 1(c) shows the schematic process description of metal hydride separations (Taniguchi and Ishida, 2006).

A partial condensation process is used to recover the H_2 from the petrochemical off-gases. The gases are compressed and cryogenically cooled to condense the hydrocarbons, and the concentrated H_2 is separated from the condenser at a cryogenic temperature. The purity and the H_2 recovery ratio in all these cryogenic processes depend on the feed gas composition, pressure, and temperature (Uehara, 2009). Fig. 1(d) shows the schematic process description of the cryogenic process (Oh and Hirscher, 2016). Fig. 2 summarizes various classifications and types of H_2 separation techniques. This review article deals with the most important challenges, advantages, and disadvantages for H_2 separation through membranes, PSA, metal hydrides, and cryogenic separation processes. This review paper provides analysis of the specialized hydrogen separation technology for membranes and comprehensive understanding of current technologies' advantages, disadvantages, issues and challenges. This review article also describes how hybrid systems are necessary for H_2 separation.

2. Membranes

2.1. Polymeric membranes

In membranes gas separation processes, polymeric membranes have been widely used in industrial processes due to their salient feature such as low cost, ease of processability, and excellent fil forming abilities (Zia ul Mustafa et al., 2019). Polymeric membranes are of two types: glassy and rubbery, depending upon the glass transition (Tg) temperature of the polymer. Generally, the selectivity of glassy polymer is higher compared to rubbery polymers because the permeation in glassy

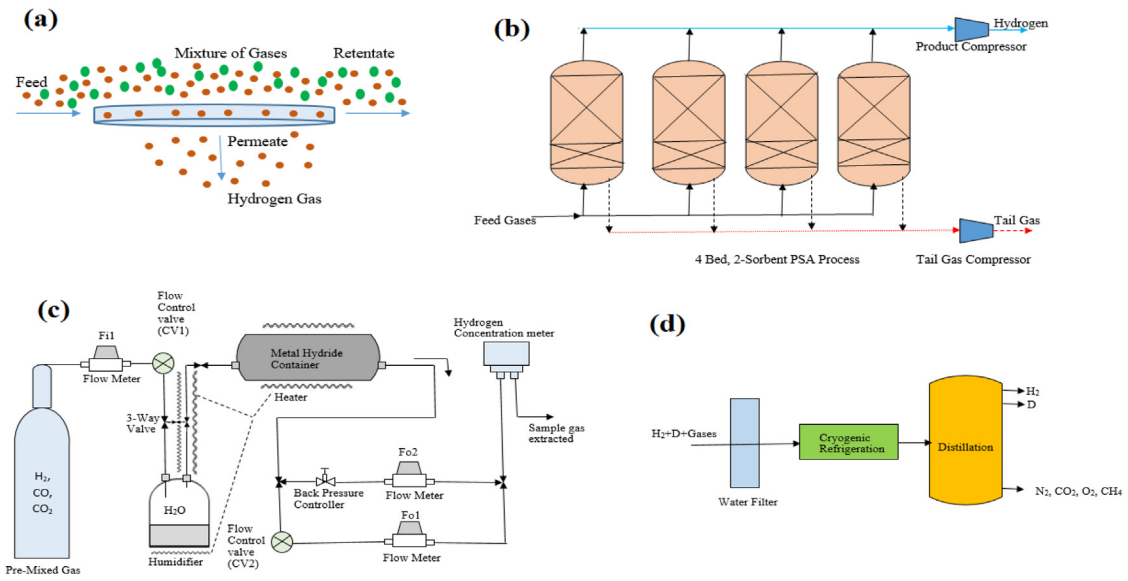

Fig. 1. Schematic process diagram of membrane (a), PSA (b), metal hydrides (c) and cryogenic process (d).

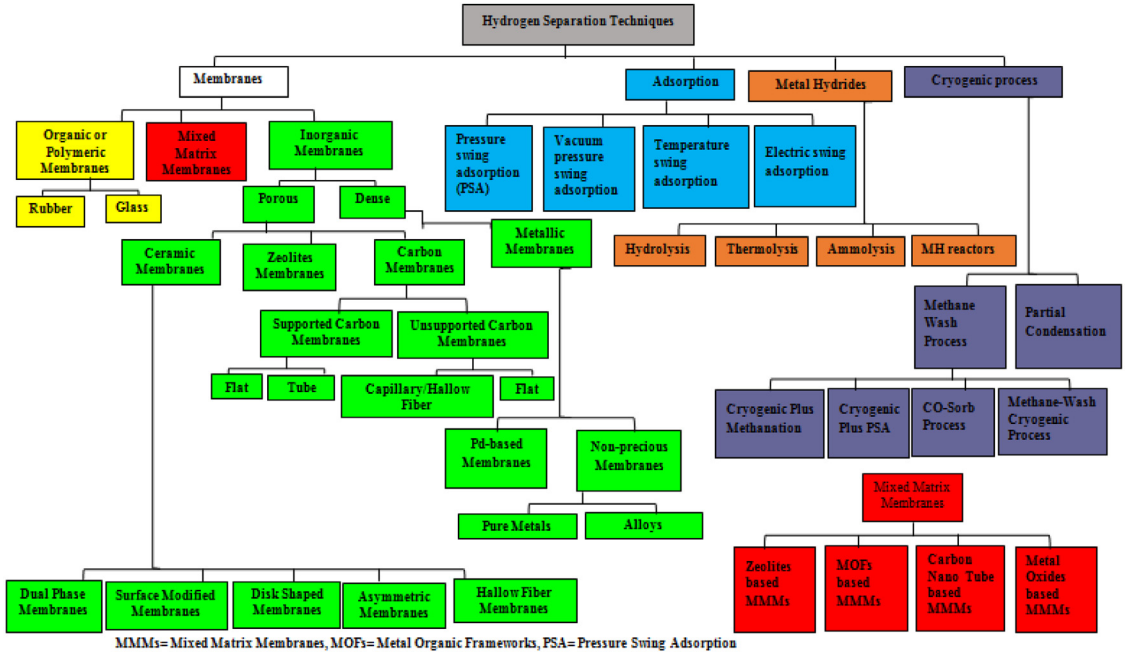

Fig. 2. Summarizes various classifications and types of hydrogen separation techniques.

polymers is governed by diffusivity of the penetrant species. However, they are coupled with inherent low H_2 permeability. Conversely, the permeation in rubbery polymer is dependent on penetrant solubility and solubility difference play an important role in determining the final selectivity of a rubbery polymer. Nevertheless, the gas separation performance i.e. H_2 permeability and selectivity of polymeric membranes is much lower than dense metallic membranes (Ismail et al., 2020). Gas permeation in dense polymeric membranes follows solution–diffusion mechanism in which H_2 separation is a combination of both solubility and diffusivity. On the other hand, porous membranes follow convective flow, Knudsen diffusion or molecular sieving mechanism depending upon the pore size of the membrane (Bernardo et al., 2020). Incorporation of bulky groups in the polymer chains sulfonation or bromination, thermal rearrangements in polymer structure, surface modification, polymer blending etc. are some of the suggested techniques to improve permeability and/or selectivity of polymeric membranes. A highly crystalline polymer structure with high molecular weight and long polymer chains can reduce plasticization problems in polymeric membranes. Physical or chemical crosslinking can also reduce aging and plasticization issues in polymeric membranes.

2.2. Mixed matrix membranes

The selectivity and the permeability define the performance of the membranes for certain applications including gas separation. To achieve a membrane that has high selectivity and high permeability simultaneously remained a challenge because both have a tradeoff relationship according to the Robeson chart (Robeson, 1991). Even though polymeric membranes are easier to fabricate and have a low cost but have reached the upper bound

Table 1
Issues and challenges in hydrogen separation by polymeric membrane technology.

Membrane	Classification	Advantages	Disadvantages	Challenges
Polymeric membranes	Glassy Rubbery	Polymeric membranes are easy to process, robust, and low cost materials (Bernardo et al., 2020) Their bulk production is possible due to easy of handling, processing and excellent film forming abilities. They offer good quality control as compared to inorganic membranes. Can be shaped in spiral wound and hollow fiber configuration which is more favorable form for industrial application. They have moderate to high H_2 permeability which makes them favorable for H_2 separation. They offer moderate selectivity (Ismail et al., 2020).	The main disadvantage of polymeric membrane is their low thermal stability. Typically, polymeric membranes cannot be used beyond the operating temperature $\leq$ 110 °C (Ismail et al., 2020). These membranes are structurally weak compared to metal or mixed matrix membranes and cannot be used under extremely harsh operating conditions of temperature and pressure; while in industrial H_2 production and separation process membranes are often encountered with harsh operating conditions. Shorter life. These membranes are prone to attack by contaminants and impurities in feed streams; thus reducing life span of the membrane (Lamb et al., 2019).	The upper bound tradeoff between permeability and selectivity reported by Robeson is still valid for polymeric membranes. Therefore, polymeric materials with both high permeability and selectivity are needed (Park et al., 2017). Structural modifications are required to improve thermal, chemical and mechanical stability of polymeric membranes for H_2 separation process. In multicomponent mixtures and in the presence of hydrocarbons the performance of polymeric membranes is adversely affected (Lamb et al., 2019). Plasticization and physical aging are serious challenges in polymeric gas separation membranes (Chuah et al., 2018). Industrial membranes are often in the form of ultrathin films or hollow fibers. Widely reported data in literature is focused on dense films with ideal selectivites. Therefore, more precise fabrication and testing methods are required to pave the way for commercialization of lab scale developed materials (Galizia et al., 2017).

described by Robeson (Robeson, 2008). On the other hand, inorganic membranes have exceptional performance for gas separation, but they are expensive and difficult for large-scale production (Hamid and Jeong, 2018). To combine the exceptional performance of the inorganic membranes with the easy processibility of the polymeric membranes, mixed-matrix membranes (MMMs) have been introduced. MMMs are the incorporation of the dispersed phase into a continuous phase. The dispersed phase is the additive solid material while the continuous phase is the polymer matrix. Materials that have been employed as an additive in MMMs are zeolite (Zagho et al., 2021), carbon molecular sieves (Vu et al., 2003), activated carbon (Lewis et al., 2021), metal–organic frameworks (MOFs) (Zornoza et al., 2013; Erucar et al., 2013), carbon nanotubes (CNTs) (Sanip et al., 2011), metal oxides (Moghadam et al., 2011), mesoporous materials and nonporous (Miricioiu et al., 2019). The polymers that have been utilized as a continuous phase in MMMs are cellulose acetate (Azam et al., 2020), polysulfones (Tahir et al., 2019), polycarbonates (Rodríguez-Jardón et al., 2021), polyamides (Soto et al., 2020), polyimides (Pechar et al., 2006), polyacrylonitrile (Li et al., 2013), and polyurethane (Hassanajili et al., 2014).

Although a huge number of research works have been made in the recent decade, still MMMs for H_2 separation have not been industrialized. The research data is available only on the lab scale and it is much needed to research the scaling up of the processes from the lab scale to the pilot scale. This could be achieved with a robust collaboration between academia and industry. Moreover, the techno-economic and life cycle assessment analyses must be made to scale up H_2 separation by employing MMMs. Nonetheless, MMMs have the capability to become the membranes of the future (H_2 separation) and play a crucial role in clean energy production and boost the hydrogen economy.

2.3. Ceramic membranes

In a ceramic membranes the thickness and the structure has a great influence on the species diffusion. At elevated temperature the oxygen-permeable dense ceramic membranes reactor are prefer for H_2 purification. In these membranes, water combines with electron, separates into oxygen and H_2 molecules. Due to the ambipolar diffusion of oxygen ions and electrons in the membrane, H_2 is oxidizing by oxygen ions to produce water and electron. The amount of catalyst corresponds to the surface reaction; if the amount is higher the surface reaction is faster (Li et al., 2019a). In dense ceramic membranes, the H_2 permeability dependents on the conductivities of the proton and electronic species. H_2 is transported by means of coupled transport of protons and electrons. Varieties of methods are available for the preparation of ceramic membranes such as solid-state reactions (SSR) method, Glycine-nitrate combustion process, Sol–gel method and chemical co-precipitation method (Hashim et al., 2018). Four-probe DC techniques and electrical conductivity relaxation (ERC) characterization techniques are not expensive techniques to analyze the membrane properties (Hashim et al., 2018). The H_2 permeation mechanism through mixed protonic–electronic conducting membranes include the surface reaction (feed side, bulk diffusion charge transfer at the membrane surface Vermaak et al., 2021 and surface reactions (sweep). When the membrane thickness is thin then surface reactions become rate-limiting step for H_2 permeability (Wang et al., 2018). Now-a-days researcher attentions' are to produce the low cost ceramics membranes by using cheap materials such as natural clay, dolomite, mineral coal, bauxite, kaolin and apatite powder (Issaoui and Limousy, 2019). For the development of perovskites, membranes four main parameters in which phase stability, electron and proton conductivity as well as the surface catalytic activity are important to consider for better separation results (Yang et al., 2021). Basically there are two types ceramic membranes for H_2 separation. One

Table 2
Issues and challenges in hydrogen separation by mixed-matrix membrane technology.

Membrane	Classification	Advantage	Disadvantage	Challenges
Mixed-matrix membranes	Symmetric mixed-matrix membranes (Ghalia and Abdelrasoul, 2019) and asymmetric mixed-matrix membranes (Yang et al., 2020).	MMMs are hybrid membranes and incorporate the synergetic properties of inorganic membranes with the easily processability of polymeric membranes (Dechnik et al., 2017) The separation performance of MMMs is impressive. Recently, Ahmad et al. achieved a selectivity of 41.9 for CO_2/CH_4 with a CO_2 permeability of 108 Barrer by employing their fabricated MMMs of 6FDA-based co-polyimides incorporated with UiO-66 nanoparticles (Dong et al., 2013). The additive nanoparticles in MMMs can be functionalized to enhance the separation performance (Zhang et al., 2019b). Incorporating nanoparticles in the polymer matrix in MMMs improve its mechanical properties (Singh et al., 2021). PEG grafted CNTS into the matrix of polysulfone improved mechanical strength of the membrane. MMMs have higher hydrophilicity, elevated fouling resistance, exceptional thermal and chemical stability over a wide range of pH and temperature (Maghami and Abdelrasoul, 2018) Fabrication cost and overall cost of MMMs is low as compared to inorganic membranes (Vinoba et al., 2017)	Polymer penetrates into the open pores of MOFs and form micro voids at the interface that reduce the permeability (Ahmad et al., 2020). Poor interaction between the polymer and the particles causes deterioration in the separation performance of the MMMs membrane. The uniform dispersion of additive particles in the polymer matrix is difficult, therefore agglomerates formed which degrade the separation performance (Maghami and Abdelrasoul, 2018). At higher loading of the additive in the polymer matrix the membrane loses its mechanical strength (Azam et al., 2020) and becomes brittle (Vinoba et al., 2017). Interfacial void forms in MMMs due to the lack of compatibility between additive particles and polymer matrix, which degrade the membrane performance (Ma et al., 2019). To select compatible additive and polymer to avoid interfacial void between the additive and polymer matrix to obtain defect-free high-performance membranes is difficult (Nuhnen et al., 2020). The thermal and chemical stability depends on the type of polymer matrix (Majka et al., 2016)	It is difficult to select high-performance polymeric material as well as additive particles for particular application without compromising the compatibility (Wang et al., 2017) To achieve homogeneous dispersion of the additive nanoparticles into the polymer matrix to avoid agglomeration of additive particles into the polymer matrix is challenging (Shi et al., 2020). At high temperature, agglomeration is very common in MMMs that cause the voids between the filler and polymer. These voids formation lower the permeability (Carreon et al., 2017) The chemical stability of used filler material in membranes affects the overall membrane permeability–selectivity due to the presence of impurities in feed gas such as water, acidic gases and sulfur containing species (Chuah et al., 2021). Most of the fillers are expensive and physical aging still limit their practical utilization as industrial membrane material (Najari et al., 2021)

is the SiO_2-based microporous membrane and other is proton conducting dense ceramic membrane. Further these membranes are classified into disk-shaped membranes, dual-phase membranes, asymmetric membranes, hollow fiber membranes and surface-modified membranes. The thickness and structure of ceramic membranes have a significant impact on species diffusion. When the membrane thickness is thin, then surface reactions become the rate-limiting step for H_2 permeability. The thickness related problems of ceramic membranes are still unsolved in lab scale. This could be achieved with a robust collaboration between academia and industry.

2.4. Zeolites membranes

Zeolite membranes overcome the problem of high-pressure drop and the support with graphite materials are very attractive due to the thermal and chemical stability (Cazorla-Amorós et al., 2017). Inorganic membranes are composed of porous material and considered efficient for H_2 separation. Most common method for the preparation of Zeolites membrane are In-situ crystallization, pore-plugging, secondary growth methods, dry gel conversion methods and hydrothermal methods assisted with microwave heating. During hydrothermal growth and calcination steps, the formation of boundary defects occurs that reduce the permeance of H_2 gas (Cardoso et al., 2018). To eliminate the intercrystalline defects of zeolites membranes for H_2 gas separation the most commonly method used is CVD (Chemical vapor deposition) and CCD (Chemical cracking deposition). This equipment is very expensive and therefore difficult to scaling-up (Cardoso et al., 2018).

2.5. Metallic membranes

The metal membranes are rising H_2 separation technology based on solution–diffusion. In general, palladium alloy membranes are used, but the cost of Pd is a critical issue that drives the demand of less expensive metal substitutes. Among the H_2 selective membranes, those that form body-centered-cubic (BCC) structure metals which assumed to be non-precious have emerged as an alternative to palladium. The non-Pd alloys are V, Cr, Rb, Nb, Mo and Ta which feature an atom at each corner of the metal unit cell, and one atom in the center of the lattice. In the mechanism of H_2 permeation of metal membrane, H_2 enters a metal lattice in the form of an atom. Due to its small atom radius, H_2 can easily jump into the metal lattice. In that process, H_2 damages the metal membrane which is called H_2 embrittlement (HE). Due to its high H_2 solubility in vanadium lattice, pure V membrane is hard to be applied as H_2 permeation membrane for its

Table 3
Issues and challenges in hydrogen separation by ceramics membrane technology.

Ceramics membrane	Advantage	Disadvantages	Challenges
Proton Conducting Dense Ceramic Membrane	Dense ceramic membranes are defect free, chemical and mechanically stable at high temperatures (500–900 °C) (Hashim et al., 2018). Proton conducting membranes are low manufacturing cost and offers 100% H_2 selectivity (Hashim et al., 2018). Fluorine-Induced microporous silica ceramic membranes are stable under steam conditions up to partial pressure of 30 kPa (Kanezashi et al., 2021).	Different preparation routes creates different microstructures that cause the different protonic and electronic conductivities. In addition, it exhibit different oxygen partial pressure gradients across the membrane (Hashim et al., 2018). Perovskite-based MPEC membranes have low electronic conductivity that lower the H_2 permeability (Wang et al., 2018) In proton conducting membranes, Due to the presence of CO_2 gas in the gas streams the formation of undesirable compound such as $SrCO_3$, $BaCeO_3$ occurs that lower the H_2 permeance (Jia et al., 2021) Due to steam exposure, the formation of micro-cracks and micropore occurred in silica-based ceramic membranes that lead to the loss of H_2 separation performance (Kurt and Topuz, 2021)	At high temperature, the electron conductivity is very poor that is responsible for the poor H_2 permeability. Increase in electrical conductivity is the main issue in perovskite membranes (Yang et al., 2021). Oxygen or H_2 permeable mixed-conducting ceramic membranes are normally operated at high temperatures (> 700 °C). They also experience chemical stability problems due to the chemical reaction of the metal oxide with gas species such as CO_2, H_2S, or even water vapor (Zhang et al., 2011). The presence of unwanted gases such as H_2S even at very low level of ppm exhibits low H_2 permeability. Even in industrial gas streams, H_2S gas is present. Therefore the process first needs to purify it. This purification increase the cost of the process (Li et al., 2017) The microporous structure of SiO_2-based membranes is not stable under steam conditions that lower the permeability of H_2 gas (Kanezashi et al., 2021).
SiO_2 based Microporous Membranes	In silica-based ceramic membranes, the permeation of smaller gas molecules increase with increasing the temperature followed by the activated transport mechanism (Koutsonikolas et al., 2021). Outer layer of silica-based ceramic membranes provides the mechanical strength and forms a macro-porous structure that is good for H_2 permeability (Issaoui and Limousy, 2019)	Membrane structure of silica-based ceramic membranes become denser due to the addition of metals. Theses metals have a strong interaction with specific gases that reduce the permeability for larger gases (Karakiliç et al., 2017).	

HE problem. Also, alloying with other metals such as Ni (Baraban et al., 2019), Cr and Al (Huang et al., 2020), Pd (Alimov et al., 2018) has been studied to overcome this issue and better mechanical stability and durability. Another drawback of metallic membrane is low permeability (Dunbar and Lee, 2017). The thermal heat treatment is known to reduce the oxidized membrane on the surface resulting in higher H_2 flux than before. However, there is little research about V-based membrane heat treatment. It was only revealed that the H_2 permeability of the V-alloy drops down showing non-linear flux different from Pd (Baraban et al., 2019). To effectively use the non-precious metal membrane, research on a heat treatment process for activating the membrane should be conducted. In addition, a more effective membrane activation process should be investigated through the influence on the type of exposed gas (Nordio et al., 2019). The permeated H_2 through dense metallic membranes is suitable for application to industrial processes thanks to temperature problem such as the water gas shift reaction (350 °C), steam methane reformation (815 °C), and methanol synthesis (300 °C). Also, it provides high H_2 purity. Although this mechanism has not been fully identified, the known model by Kehr (1983) suggested that H_2 atoms jump between interstitial sites. In this process, the structural characteristics of the BCC metal lattice further strengthen this tendency (Sazali et al., 2020b). As a result, a high level of selectivity is guaranteed not only in the Pd-based membrane but also in the BCC membrane.

Many new alloy combinations have been studied to solve the problem of embrittlement, which has been a chronic problem of metal membranes, and there are studies based on various models and mathematical calculations on the metallic mechanism. It is worth mentioning that metallic membrane technologies were dealt in terms of conventional and emerging ways. It is valuable in the development of metal membranes and in the process of solving the issues at hand.

2.6. Carbon membranes

One of the vastly superior material in terms of thermally and chemically resistance membrane is carbon membrane. Carbon membranes are sustainable to utilize at high temperature in the range of 500–900 °C (Sazali et al., 2017). However, as the thermal treatments are conducted above Tg level, porous structure of the membrane partially collapse (Chisca et al., 2022). The pore structure of the carbon membranes are adjustable by using polymer blend technique (Sazali et al., 2017). During the preparation of carbon membranes, the carbonization temperature affects the pore size distribution of the membrane (Ismail et al., 2018). Carbon membranes are normally composed of microporous and amorphous high carbon materials that elevate good gas permeations (Sazali et al., 2019). Permeation is accomplished by the adsorption of gas molecules and activated transport through

Table 4
Issues and challenges in hydrogen separation by zeolites membrane technology.

Zeolite membranes type	Advantage	Disadvantages	Challenges
ZIFs	Zeolites (ZIFs) membranes are crack free, pinhole free, very thin, and reproducible with grain boundary free as well as it shows high H_2 permeance (Bedard and Liu, 2018). Zeolites membranes have ability to separate H_2 gas at 25–700 °C temperature. This temperature is favorable for industrial use (Lu et al., 2021).	Zeolites (ZIFs) have relatively large pore size as compared to H_2 gas kinetic diameter. Therefore, it is difficult to use for H_2 separation from a mixture of other light gas molecules (Cardoso et al., 2018).	The zeolites (ZIFs) layers are brittle and difficult to produce. Therefore, supports are used but theses supports convey mechanical resistance without penalizing mass transfer (Cardoso et al., 2018). One main issue is to maintain the structural integrity of zeolites membranes maximum up to 150 °C (Lai, 2018).
ZIF-8	Recently the researchers developed defect free zeolites (ZIF-8) membranes by adding polydopamine in aqueous solutions. The proton release during dopamine polymerization and competitive chelation to hamper homogeneous nucleation of ZIF-8 membranes (Jiang et al., 2019)	In Zeolites (ZIF-8) membranes thick layer of membrane improve the mass transfer resistance for the gases but it usually showed low permeance for H_2 gas (He et al., 2021). The low selectivities of zeolites (ZIF-8) membranes for H_2 separation makes them limited to use (Lai, 2018).	In zeolites (ZIF-8) membranes, increasing the feed pressure causes both increase in adsorption and diffusion of gas molecules. However, it declines the separation efficiency by damaging the inter crystalline grain boundaries (Farjoo et al., 2017)
DD3R	DD3R membranes are chemically and thermally stable due to their silica structure (van den Bergh et al., 2008). Even "DD3R" zeolites membranes are stable in the presence of cracked gases such as H_2S gas for more than 1000 hours (Du et al., 2021a).	In DD3R membranes, the selectivity of mixtures of weakly adsorbing components was independent of pressure (van den Bergh et al., 2008).	In DD3R membranes, as the temperature and pressure increased, the selectivity of mixtures of a strongly and weakly adsorbing component decreased (van den Bergh et al., 2008).
NaA	Strongly structured NaA zeolite membranes offer excellent thermal and chemical stability in harsh environments (Gao et al., 2021).	NaA zeolite membranes are often made via the secondary growth process, which necessitates the use of support surfaces (Liu et al., 2015). Some of the supports types exhibit negative zeta potential that result in electrostatic repulsion mechanism and makes the membranes unsuitable for use (Cardoso et al., 2018).	The main obstacle to the use of NaA zeolite membranes for large-scale applications is high fabrication costs (Liu et al., 2015).
SAPO-34	For high temperature reactions (200–600 °C), silico aluminophosphate (SAPO-34) has been employed in membrane reactors (Xu et al., 2021)	In the presence of water, SAPO-34 membranes at room temperature have been shown to be less effective (Xu et al., 2021).	The most common ceramic substrate being used as support underwent into decomposition upon an exposure to hydrothermal atmosphere. Therefore, Presence of water in the gas streams makes the ceramic support unusable (Lee et al., 2018)
FAU	Faujasite (FAU) zeolite membranes exhibit excellent catalytic and ion-exchange capabilities (Ouyang et al., 2021b).	In the preparation of FAU membranes, direct crystallization via single stage in-situ synthesis usually resulted in non-homogeneous zeolite layer (Nazir et al., 2020a).	FAU membranes show negative expansion coefficients at temperatures of 373 K. which causes thermal stress and results in the cracks' formation in the membrane layer (Nazir et al., 2020a).
MFI	At high temperature (300–600 °C) MFI membranes exhibit steam or water to sieve from gas molecules (Wang et al., 2014).	The H_2 permeance is more sensitive due to the presence of cracked gases in industrial stream. Therefore when the presence of cracked gases such as (C_2H_4, C_2H_6)is encountered the H_2 permeance reduced (Du et al., 2021a)	The separation properties of the membranes have a weak dependence on feed pressure and water vapor pressure at high temperatures (Li et al., 2021). At high temperatures MFI zeolite membranes exhibit a sintering effect (Wang et al., 2014).
UIO-66	A series of benzene dicarboxylic acid containing amino, nitro, bromo, hydroxyl, and carboxyl groups can be used as a ligands for UiO-66 membrane (Guo et al., 2021).	UIP-66 (Universitet I Oslo) has a relatively large pore size of 6 Å that reduces the molecular sieving properties of the membrane (Friebe et al., 2017).	The complexation of zinc ions with the linker causes the reverse hydrolysis effect during the membrane manufacturing process. Due to this hydrolysis effect, the higher portion of ligands is required to fulfill the deprotonation in aqueous solutions (Malekmohammadi et al., 2019)
Zeolite-MOFs Membrane	The gas permeation flux is approximately proportional to the transmembrane pressure drop (Lin, 2019). H_2 gas flux can easily be improved by increasing the feed pressure.	Gases should encounter a net attraction force when their molecular size is smaller than the MOF channel diameter (Shi et al., 2021).	Due to repulsion force, the permeability of H_2 gas is reduced whenever H_2 gas is mixed with larger kinetic diameter gases (Shi et al., 2021).

selective pore opening. By changing the precursor, geometry of precursor and pyrolysis conditions the performance of the carbon membranes can be enhanced (Haider et al., 2018).

3. Other separation techniques

3.1. Adsorption process

Pressure swing adsorptions are regarded as the most promising technique among other separation techniques due to their high-performance efficiency (high H_2 recovery and purity), low energy consumption, and low capital investment (Shamsudin et al., 2019). The adsorption of gases is performed at high pressure to adsorb the impurities on the surface of the adsorbent, while the regeneration is followed by lowering the total pressure of the bed (Yáñez et al., 2020). The nature of adsorbent, column pressure, and temperature are considerably affecting the efficiency of the system. Usually, the rate of adsorption decreases with an increase in temperature but it can be increased by the increasing pressure (Moon et al., 2016; Kuroda et al., 2018). Zeolite, activated

Table 5
Issues and challenges in hydrogen separation by metallic membrane technology.

Metallic membrane	Classification	Advantage	Disadvantages	Challenges
Pd-based membranes	Pd-based membranes	Palladium dissociates H_2 into atomic formwell. High purity H_2 production can be obtained in real natural gas reforming process (Anzelmo et al., 2018).	Pure Pd membranes show phase transformation of α to β which makes distortion of metal lattice (Pal et al., 2020). The formation of stable hydrides resulted in the decreased permeation rates.	Addition of alloying elements in Pd decreased the H_2 diffusivity and leads to increased energy for H_2 hoping in the metal (Nayebossadri et al., 2017).
		It shows high tolerance to hydrocarbon flow and self-catalyzing the H_2 dissociation process under 350–500 °C. It guaranteed high H_2 productivity with fast kinetics at lower operating temperature which is based on composite membraned reactor (CMR) (Park et al., 2020)	The H_2 selectivity of Ar is decreased from infinity to 12400 at 450 °C when pressure increased from 0.05 MPa to 0.15MPa (Anzelmo et al., 2018).	Surfaces egregation of alloying elements occurs when exposed to contaminant gases (CO_2, CO) (Dalla Fontana et al., 2018).
		The H_2 flux can be recovered 80%–90% of initial flux after removing natural gas at 400 °C and 0.5 MPa (Nayebossadri et al., 2019).	The membranes show sharp decline in H_2 flux when exposed to H_2S gas mixture. Main source of H_2 production is natural gas steam reforming process. However, if the H_2 concentration is less than 25% in natural gas then separation is not possible (Nayebossadri et al., 2019)	The intrinsic permeability of the metallic membrane is significantly lower than that of other membranes (Bernardo et al., 2020). It is difficult to apply on an industrial scale.
		The thickness of the film can be adjusted 1 μm–30 μm without compromising the structural integrity of the film (Bernardo et al., 2020). Therefore, it can be selected according to the H_2 purity target	The supported membrane can be prepared with thin metal layer. However, a support surface with a large number of pores and non-uniform pore size causes membrane defects and pinholes on a surface of metal film (Pal et al., 2020).	State-of-the-art researches aim to supported membranes with thin Pd film to increase the mechanical stability in permeation process (Conde et al., 2017)
		Metallic membranes can offer high selectivity of H_2 and good stability at increasing temperature in the long-term test (Pal et al., 2020). If steam is adopted as a sweep gas, the membrane shows efficient H_2 recovery (> 91%) which is comparable to effects of noble gas (Park et al., 2020).	Niobium and tantalum have higher H_2 permeability than palladium. However, these metals have higher catalytic energy for H_2 molecule dissociations. BCC structured metals have tendency to embrittle under H_2 condition (Conde et al., 2017)	In the presence of CO, CO_2, and H_2O (steam), H_2 selectivity would be influenced due to the water gas shift reaction (Wang et al., 2019)
Non-precious membranes	Pure metals	Alloys exhibit high strength, ductility and H_2 solubility resulting in decreased the embrittlement dangers.	The promising source of H_2 is ammonia decomposition. However, metallic membrane proved to be beneficial only between 400–450 °C (NH_3 dehydrogenation temperature) when H_2 is mixed with ammonia (Jo et al., 2018).	During the H_2 tests, the membrane transformed to brittle phase which leads to the formation of cracks on the membrane surface (Dolan et al., 2018).
	Alloys	It can recover its decreased H_2 flux after long-term experiment from 0.20 (mol $m^{-2}s^{-1}$) to 0.25 (mol m^{-2} s^{-1}) after heat treatment steps (Dolan et al., 2018).	The permeability of metallic membrane differs depending on the composition of alloy elements and temperature (Baraban et al., 2019). Therefore, it needs to be researched the relationship between temperature and the composition of membrane.	Non-uniform distribution of alloying elements leads to different H_2 concentrations (Baraban et al., 2019). Depending on the composition of the alloy film, a H_2 permeation trend different from that of the conventional metal membrane may be shown (Bellini et al., 2018)

carbon, alumina, and silica gel are mainly used as traditional adsorbents in adsorption processes. These adsorbents have a higher affinity for CO_2, NH_3, H_2O inert gases (He, Ne, Ar), etc., and lower for H_2. However, removing N_2 and CO is difficult due to the low adsorption capacity of conventional adsorbents, which leads to a decrease in H_2 purity. Researchers are modifying and developing novel adsorbents for the effective removal of gases from H_2 and have achieved extraordinary results compared to conventional adsorbents. Table 7 explains the issues and challenges of H_2 separation by using adsorption process.

3.2. Metal hydrides

Metal hydride is a unique chemical method to purify and separate H_2. The metal hydride method/reaction uses different metal alloys to absorb and desorb the H_2, in process of purifying H_2 from impurities. The reaction is reversible and its direction is measured by the pressure and temperature of H_2 gas. If pressure is above a certain threshold, H_2 forms a hydride, whereas below the threshold a highly purified H_2 is released. Different methods to desorb the H_2 from the intermetallic structures have been summarized in the table below. The metal hydride reactors are of utmost consequence in this reaction for the reaction efficiency and to achieve high H_2 purity. The heat and mass transfer requirements are important considerations in designing a metal hydride reactor (Borzone et al., 2017). Separation of H_2 depends upon many parameters like reaction kinetics, temperature and pressure of the hydride bed, enthalpy, and cycle time, etc. To overcome the reaction kinetics challenge, the addition of amorphous Ni-Li-B catalysts increases the reaction kinetics of aluminum-based

Table 6
Issues and challenges in hydrogen separation by carbon membrane technology.

Carbon membrane (CMs) types	Classification	Advantage of CMs	Disadvantage of CMs	Challenges of CMs
Unsupported Carbon membranes	Flat	CMs exhibit biomodal pore structure with ultramicropores and micropores properties that is favorable for high H_2 separation (Lei et al., 2021a)	High packing density cause the reduction in flow space which resulted the pressure drop (Wey et al., 2020)	Carbon layers are thin and fragile and they need porous support for preparation. Without support membrane is not prepared (Hamm et al., 2017)
	Capillary/Hallow fiber	CMs membranes are easily operated at high temperatures (250 °C) and high pressure (25 bar) making them attractive for industrial applications (Lei et al., 2021a).	Industrial gases streams contain condensable and reactive components. These unwanted gases reduce the H_2 separation permeance (Sazali et al., 2020a)	Hollow fiber unsupported CMs due to brittle effect exhibits weaknesses that reduce the H_2 separation performance (Sazali, 2020)
		The higher operating temperature effects the permeation due to the gas adsorption properties. Higher temperature increase permeation (Widiastuti et al., 2021)	Due to surface flow mechanism (SSF) condensable gases i.e (CO_2) exhibit more permeance as compared to H_2 (He, 2017)	The diameter of the support pores must be order of magnitude smaller than the thickness of the polymeric layer deposited. It is more challenging to produce it (Hamm et al., 2017)
Supported carbon membranes	Flat	Varieties of precursors (polymeric, Cellulose derivates, polyimide, PVDF, PAN) are easily available and employed to prepare CMS membranes (Lei et al., 2021a)	The layer of precursor collapses and becomes dense during thermal formation process that leads to deterioration of gas permeance (Lei et al., 2021b).	The internal adhesion occur during the preparation of membrane (Sazali, 2020)
		Plenty of thermally labile additives (NCC, MCC, PVP and many mores) are also easily available for H_2 separation (Sazali et al., 2021)	The relationship between polymer concentration and the dimensions of the pores on the support need to manage. Low concentration exhibits low viscosity solutions (Briceño et al., 2012)	Longer dip coating time resulted in a thicker membrane thickness layer that reduce the membrane permeability (Sazali et al., 2021)
	Tube	Small gas pairs exhibit excellent permeance and selectivity (Pal and Agarwal, 2021)	Multiple coating steps required during preparation that increase the cost of membrane (Briceño et al., 2012)	During preparation, post oxidation stage causes the clogging effect of water that reduce the permeability of the CMs (Torres et al., 2021)

Table 7
Issues and challenges in hydrogen separation by pressure swing adsorption process.

Adsorption	Technology/ Methodology	Nature of adsorbent	Advantages	Disadvantages	Challenges
Pressure Swing Adsorption (PSA)	Pressure Equalization (Shamsudin et al., 2019) Single Column (Saberimoghaddam and Khebri, 2017) Multiple Column (Relvas et al., 2018) Multiple Bed (Moon et al., 2018; Zhang et al., 2019a) Multiple Layered (Riboldi and Bolland, 2016; Yu et al., 2021)	Activated Carbon (Idris et al., 2019; Li et al., 2019b; Amin et al., 2022b)	Activated carbon has a large surface area (Amin and Shah, 2022) and a high adsorption capacity for CO_2 and light hydrocarbon, resulting in improved H_2 purity (Rashidi and Yusup, 2017). Compared to zeolite, it is less costly and could be regenerated easily during the PSA cycle (Abdeljaoued et al., 2018).	The adsorption capacity of activated carbon decreases with an increase in temperature. Because the increase of temperature reduces its adsorption affinity (reduce Vander Walls interactions) for components which results in a decrease of H_2 purity and recovery (Moon et al., 2016).	Activated carbons have a low affinity for CO and inert gases and they are not preferred when the feed contains a larger concentration of CO and inert gases (N_2, He, Ar, etc.) (Moon et al., 2016).

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alloys. Table 8 explains the issues and challenges of H_2 separation by using metal hydrides.

3.3. Cryogenic separation

Cryogenic method is one of the clean producing method of H_2 in mass production as it does not involve any chemicals. Cryogenic separation method is one of the mature technologies due to the use of refrigeration and compression technologies. It therefore can be easily installed for industrial scale use with high degree of success. It has a recovery rate of about 95% with a marginally low purity of 95%–98% (Aasadnia et al., 2021) considered with other H_2 separation methods. Cryogenic method is an environmentally safe method as it does not use chemicals, and therefore does not produce any secondary pollutants. One of the drawbacks of this method is its huge capital and operational cost. This could be achieved with a robust collaboration between academia and industry. Table 9 explains the issues and challenges of H_2 separation by using cryogenic separation process.

4. Discussions

For complete purification or separation of gases, a series of PSA, distillation columns or membrane systems are required, which makes the separation process more complex and costlier

Table 7 (*continued*).

Adsorption	Technology/ Methodology	Nature of adsorbent	Advantages	Disadvantages	Challenges
		Zeolite (Zhang et al., 2021; Mivechian and Pakizeh, 2013; Taniguchi and Ishida, 2006; Uehara, 2009; Oh and Hirscher, 2016; Zia ul Mustafa et al., 2019; Ismail et al., 2020; Bernardo et al., 2020; Robeson, 1991, 2008; Hamid and Jeong, 2018; Zagho et al., 2021; Vu et al., 2003; Lewis et al., 2021; Zornoza et al., 2013; Erucar et al., 2013; Sanip et al., 2011; Moghadam et al., 2011; Miricioiu et al., 2019; Azam et al., 2020; Tahir et al., 2019; Rodríguez-Jardón et al., 2021; Soto et al., 2020; Pechar et al., 2006; Li et al., 2013; Hassanajili et al., 2014; Lamb et al., 2019; Park et al., 2017; Chuah et al., 2018; Galizia et al., 2017; Ghalia and Abdelrasoul, 2019; Yang et al., 2020; Dechnik et al., 2017; Dong et al., 2013; Zhang et al., 2019b; Singh et al., 2021; Maghami and Abdelrasoul, 2018; Vinoba et al., 2017; Ahmad et al., 2020; Ma et al., 2019; Nuhnen et al., 2020; Majka et al., 2016; Wang et al., 2017; Shi et al., 2020; Carreon et al., 2017; Chuah et al., 2021; Najari et al., 2021; Li et al., 2019a; Hashim et al., 2018; Wang et al., 2018; Issaoui and Limousy, 2019; Yang et al., 2021; Cazorla-Amorós et al., 2017; Cardoso et al., 2018; Baraban et al., 2019; Huang et al., 2020; Alimov et al., 2018; Dunbar and Lee, 2017; Nordio et al., 2019; Kehr, 1983; Sazali et al., 2020b, 2017; Chisca et al., 2022; Ismail et al., 2018; Sazali et al., 2019; Haider et al., 2018; Kanezashi et al., 2021; Koutsonikolas et al., 2021; Jia et al., 2021; Kurt and Topuz, 2021; Karakiliç et al., 2017; Zhang et al., 2011; Li et al., 2017; Bedard and Liu, 2018) (Lu et al., 2021; Jiang et al., 2019; van den Bergh et al., 2008; Du et al., 2021a; Gao et al., 2021; Xu et al., 2021; Ouyang et al., 2021b; Wang et al., 2014; Guo et al., 2021; Lin, 2019; He et al., 2021; Lai, 2018; Liu et al., 2015; Nazir et al., 2020a; Friebe et al., 2017; Shi et al., 2021; Farjoo et al., 2017; Lee et al., 2018; Li et al., 2021; Malekmohammadi et al., 2019; Anzelmo et al., 2018; Park et al., 2020; Nayebossadri et al., 2019; Pal et al., 2020; Dolan et al., 2018; Conde et al., 2017; Jo et al., 2018; Nayebossadri et al., 2017; Dalla Fontana et al., 2018; Wang et al., 2019; Bellini et al., 2018; Lei et al., 2021a; Wey et al., 2020; Hamm et al., 2017; Widiastuti et al., 2021; Sazali et al., 2020a; He, 2017; Sazali, 2020; Sazali et al., 2021; Lei et al., 2021b; Briceño et al., 2012; Pal and Agarwal, 2021; Torres et al., 2021; Shamsudin et al., 2019; Yáñez et al., 2020; Moon et al., 2016; Kuroda et al., 2018; Borzone et al., 2017; Aasadnia et al., 2021; Saberimoghaddam and Khebri, 2017; Relvas et al., 2018; Moon et al., 2018; Zhang et al., 2019a; Riboldi and Bolland, 2016; Yu et al., 2021; Idris et al., 2019; Li et al., 2019b; Amin et al., 2022b; Amin and Shah, 2022; Rashidi and Yusup, 2017; Abdeljaoued et al., 2018; Brea et al., 2019)	Zeolite adsorbents have a stronger attraction for CO, CO_2, and N_2 and lowest for H_2 resulting in higher H_2 purity and recovery (Moon et al., 2016).	The stronger attraction of CO_2 on zeolites adsorbent causes a detrimental effect on H_2 purity and recovery (Moon et al., 2016)	One major reason for not considering zeolites for CO_2 adsorption is due to their high heat of adsorption and difficulty in regeneration during regeneration of adsorbent bed (Moon et al., 2016).

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Table 7 (*continued*).

Adsorption	Technology/ Methodology	Nature of adsorbent	Advantages	Disadvantages	Challenges
	Single Bed (Kuroda et al., 2018)	Hydroxyl aluminum silicate clay (HAS-Clay) (Kuroda et al., 2018)	HAS-Clay is an excellent CO_2 adsorbent for increasing H_2 purity while also decreasing the overall column pressure. It also has the potential to remove H_2S in the feed gas (Kuroda et al., 2018)	Temperature distribution does not remain uniform throughout the adsorption bed which alters the H_2 recovery and purity (Kuroda et al., 2018)	Multiple-bed adsorption has the potential to increase CO_2 removal, which can result in the production of high-quality H_2
	Cu-BTC Four-Step Bed (Xiao et al., 2018)	MOF (Cu-BTC) (Xiao et al., 2018)	MOF (Cu-BTC) is characterized by structure adversity, a large surface area, and high selectivity. It increases the purity of H_2 by increasing the adsorption pressure (Xiao et al., 2018)	MOF (Cu-BTC) causes a decline in H_2 productivity and recovery with an increase of pressure (Xiao et al., 2018)	After depressurizing the column, a small amount of CO_2 and CH_4 are deposited in the Cu-BTC bed, which interferes with H_2 recovery and purity (Xiao et al., 2018)
Temperature Swing Adsorption (TSA)	Four Bed and Two Layered (Golmakani et al., 2017)	Activated Carbon and zeolite (Amin, 2022; Golmakani et al., 2017)	TSA process is used to get higher H_2 purity at a reasonable H_2 recovery (Golmakani et al., 2017).	The TSA method consumes more energy than that of other cyclic processes (PSA and VSA) due to the additional cost of the heating and cooling step (Golmakani et al., 2017)	The increase of steam temperature improves H_2 purity. However, higher temperature takes a long time for cooling, which extends the cycle time, resulting in a reduction in H_2 productivity and recovery (Golmakani et al., 2017)
Vacuum Pressure Swing Adsorption (PSA)			The most cost-effective and energy-efficient process among the PSA and TSA for producing ultra-pure H_2 with higher purity, recovery, and productivity (Du et al., 2021b)	The vacuum pump is always required to evacuate the adsorption column during the regeneration of the adsorbent (Golmakani et al., 2017)	The H_2 purity increases by overdesigning the VSA unit but a significant increase in energy and decrease in H_2 productivity and recovery make this unsuited for use (Golmakani et al., 2017)

(Chen et al., 2018). To reduce the overall separation cost, two different separation technologies can be integrated into one system, called a hybrid system. Hybrid membrane processes are those in which a membrane system is used with another unit activity or any chemical process with a single unit operation. These types of hybrid systems will reduce the cost of the overall process and improve separation efficiency (Bernardo et al., 2009). The vapor mixture from the cryogenic distillation column is sent to the membrane system, which extracts the gases using an aqueous solution (Bernardo et al., 2009). Therefore, hybrid or integrated separation systems are required to meet the purity levels of H_2 for high-end applications. MH reactors have a low H_2 recovery ratio, ranging between 75% and 95%. Similarly, the PSA process has a drawback: it purifies H_2 with a low recovery rate of 70%. The H_2 concentration in the desorption gas is still as high as 30%, meaning a significant waste of resources if the remaining H_2 cannot be recovered by the four pressure equalization steps of the VPSA process. Therefore, for full H_2 recovery, it is better to couple PSA with other separation technologies such as membrane technology. On the other hand, the H_2 purity obtained by using membrane technology is much lower than that obtained by using PSA technology. Even at a high H_2 concentration in the feed gas, it is very hard to achieve a H_2 purity of 99% (Li et al., 2016). Therefore, the high recovery of H_2 hybrid or integrated systems has been considered good. Fig. 3 (a & b) shows an example of a hybrid distillation-membrane system used in a pilot plant to separate gases.

The purity of H_2 gas produced by hybrid systems depends on the membrane type and module configuration used for H_2 separation. Researchers have been using Pd-Ag membrane, carbon molecular sieve membrane (CMSM), c-PdDS (ceramic-supported double skin), and many more membranes as hybrid configurations. The separation permeability also depends on the membrane's operating temperature and pressure. Recently, the researcher conducted the H_2 separation by using a hybrid system. Results reveal that the c-PdDS hybrid system exhibits a permeance of $2^{*}10^{-6}$ (mol/s/m^2/Pa) at 8 bar retentate pressure. Similarly, the CMSM hybrid system exhibits a permeance of $5^{*}10^{-8}$ (mol/s/m^2/Pa) at 40 bar retentate pressure. The inlet gas stream in the c-PdDS membrane was at 400 °C, while in the protonic membrane hybrid system, the inlet gas stream operates at 65 °C. Therefore, we conclude that the grade of purity in hybrid systems depends on the membrane operating conditions and the hybrid configuration used (Nordio et al., 2021).

Other types of H_2 separation membranes such as ionic liquids (ILs) and electrochemical H_2 pumping membranes are getting attention due to their high H_2 purification from mixtures of gases. ILs membranes are considered more suitable for the separation of

Table 8
Issues and challenges in hydrogen separation by metal hydrides.

	Method	Sub-category	Advantages	Disadvantages	Challenges
Metal hydrides	Hydrolysis	1. Borohydrides	Borohydrides have high stability, and have non-toxicity and high solubility characteristics. Compared with thermolysis, hydrolysis can produce three times H_2 (Muthukumar et al., 2018)	Its hydrolysis suffers from sluggish kinetics in neutral aqueous solution (Ouyang et al., 2021a)	To develop catalyst free systems; As difficulty and cost of recovering catalysts is huge (Ouyang et al., 2021a)
		2. Mg-based alloys	Mg-based alloys are easily available in low cost, and its hydrolysis to separate H_2 produces environmentally benign products (Ma et al., 2017)	The addition of acid solution to dissolve magnesium hydro oxide in passive layers induces corrosion in reactors and hazards for users (Liu et al., 2021)	$Mg(OH)_2$ thermodynamic value is undesirable and low reaction kinetics need higher temperature, and the contamination of magnesium particles during dehydrogenation (Zhang et al., 2018)
		3. Al based alloys	It is safe, cheap and electrochemically active element. Also, it can dissolve passive layers and be able to separate H_2 at room temperatures (Rin et al., 2021)	The creation of passive layers of Al_2O_3 decreases the hydrolytic ability (Weckerle et al., 2017)	Formation of alumina film on the surface on aluminum powder hinders the reaction with water under normal operations (Xiao et al., 2022)
	Thermolysis	The thermolysis of hydrides to separate H_2 from metal hydride does not require noble metal catalysts and is free of ammonia poisoning		The thermolysis process requires heat to start the process of H_2 separation reaction, the temperature affects the yield of H_2 separated (Brestovič et al., 2021)	Specialized reactors needed for the thermal decomposition (Wang and Astruc, 2021)
	Ammonlysis	Alcoholysis gives much better results via hydrolysis as reducing the freezing problems, faster separation of H_2 from metal hydride without use of catalysts (Ouyang et al., 2021a).		At low temperature conditions Alcoholysis process experiences significant lag time, and intrinsic reaction time is needed to be increased for smooth adoption (Ouyang et al., 2021a)	To improve both efficiency and operating conditions, high pressure coupled with high temperature is required (Esposito, 2018)
	MH Reactors	1. A-type Reactor 2. D-type Reactor	Metal hydride reactors are able to recover H_2 with 99.999% purity. MH reactors can be used for small to medium sized industrial applications (Weckerle et al., 2017)	H_2 low recovery ratio ranging between 75% to 95%. Also, alloy poising and pulverization increases the cost due to losses incurred (Weckerle et al., 2017)	In large reactors, heat transfer of transient nature requires large amount of heat transfer which reduces the efficiency of the system (Dunikov and Blinov, 2020)

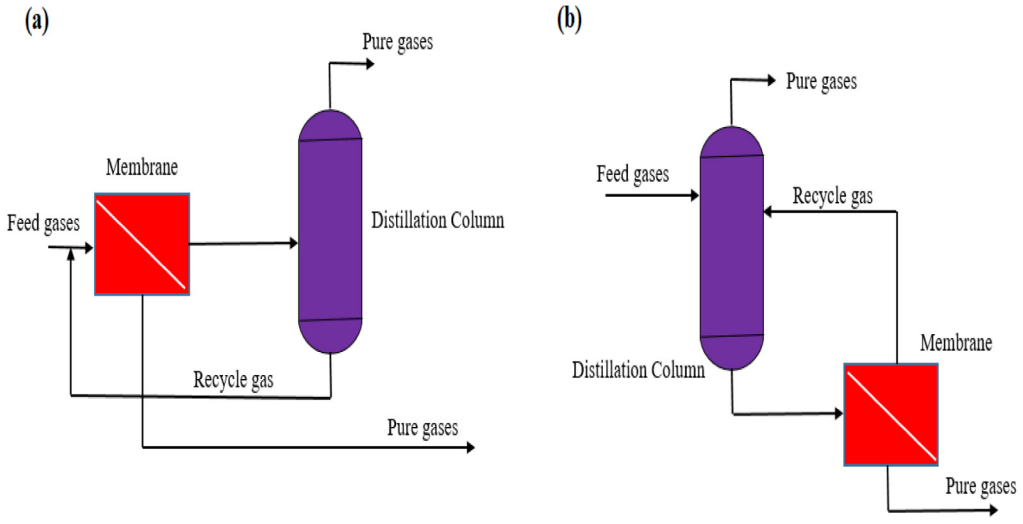

Fig. 3. Hybrid systems for gases separation.

H_2 from CO_2/H_2 mixtures because the solubility of CO_2 is higher than the solubility of H_2 in ILs. On the other hand, electron H_2 pumping (EPH) membranes have been used for H_2 separation and purification. However, the major drawback of EPH-based membranes is that they need to be hydrated to guarantee proton conduction and this gives them a higher chance of water flooding at the cathode side. Researchers are trying some useful strategies to improve the H_2 separation performance, such as adding

Table 9
Issues and challenges in hydrogen separation by cryogenic process.

	Method	Advantages	Disadvantages	Challenges
Cryogenic separation	**Partial Condensation**: Using a series of flash separators H_2 is separated from CO_2, and using splitter columns COis produced (Benson and Celin, 2018). **Methane wash process:** CO and methane are absorbed in liquid methane producing H_2 stream. Using a flash column H_2 is removed from methane and CO liquid (Benson and Celin, 2018) Purity Processes of H_2 which include Cryogenic methods 1. Cryogenics plus methanation 2. Cryogenics plus pressure swing adsorption (PSA) 3. Methane-wash cryogenic process 4. COsorb process (Speight, 2015)	H_2 Recovery rate is 90%–98% As this method has mature technology consisting of refrigeration and compression it is Ideal for large industrial scale use (Xu et al., 2012) Liquid H_2 has very low boiling point, thus important for cryogenic separation (Ebrahimi et al., 2020) Cryogenic method does not use any chemicals, thus are environmentally neutral and does not produce any secondary pollutants (Aasadnia et al., 2021) This technology is considered as one of the efficient thermodynamically separation technology (Kirchner, 2020). It gives the benefit of separating a single stream into different products (Speight, 2015). Pressure Swing adsorption (PSA) method and cryogenic separation method integration gives enhanced performance (Xu et al., 2010).	Cryogenic method requires high energy requirements due to high pressure involved (Kirchner, 2020). It is expensive due to involvement of huge devices and equipment (Bernardo et al., 2020) One of the main drawbacks of this technology is the limited purity of H_2 (95%–98%) which is not considered feasible for industrial use with the costs involved (Xu et al., 2010). Unsuitable for small portable applications due to equipment involved (Bernardo et al., 2020) Due to impurities gases, phase transition temperature could decrease which increase the refrigeration penalty and frost formation becomes highly likely (Xu et al., 2010). If the concentration of H_2 is increased in gas mixture it greatly affects the performance of cryogenic separation (Xu et al., 2010).	Requirement of stopping the clogging by removing impurities such as H_2S, CO, CO_2 and H_2O (Baena-Moreno et al., 2019) Its startup time is in hours compared with PSA and membrane method, which startup in minutes (Benson and Celin, 2018) Integration of gas mixture is an effective to increase the H_2 concentration. When clean fuel mixture is produced using gas mixture, more H_2 is consumed using H_2 rich clean fuel (Xu et al., 2012). Condensation to very low temperature causes huge energy requirement from refrigeration (Xu et al., 2012). Also, the solidification process causes problems like blockage of heaters and pipes (Xu et al., 2012). Thus, one of the biggest challenge is producing H_2 at relatively high temperature to lower the cost. The equipment corrosion caused due to solvent also poses a challenge as it increases the cost and can poison the purity of gases (Song et al., 2013).

polymer blends as precursors or doping with metal nanoparticles in CMSMs (Bernardo et al., 2020). Therefore, researchers believe that this kind of advancement in membranes not only improves the H_2 separation performance but also lowers the H_2 separation and purification cost. Furthermore, improvements in membranes, PSA, MH, cryogenic separation methods, and hybrid systems are required for future H_2 separation and purification.

5. Conclusions

This review summarizes the issues and challenges in H_2 separation technologies. Four major technologies i.e., membranes, adsorption process, MH and cryogenic process were reviewed and discussed. The major focus of the review is devoted to membrane technology. For membrane technology, the advantages, disadvantages, issues and challenges in polymeric, MMMs, ceramic, zeolite, metallic and carbon membranes have also been highlighted. However, for full exploitation of membranes in H_2 purification, high performance membranes with an extra-ordinary combination of permeability and selectivity are essential. Moreover, more robust and thermally, chemically, and mechanically stable membranes are required for industrial H_2 separation processes. Hence, polymeric membranes are not the preferred choice under this criterion due to their poor thermal stability. MMMs have interfacial defects and filler agglomeration issues that need to be addressed before their commercialization. Ceramic membranes have a high production cost that hinders their utilization on a commercial scale. In the process of metal membrane, they face a problem regarding the decrease in their permeability due to the high solubility of impurities. Optimization of the metallic membrane (i.e. microstructure, heat treatment, activation methods) is the current issue that should be taken into account. Zeolite membranes, like carbon membranes, are brittle and difficult to produce. In all types of membranes, impurities and multicomponent gas mixtures have an adverse effect on membrane performance which is a major limitation of membrane technology in H_2 purification.

Similarly, adsorbents used in adsorption processes have a higher affinity for impurities (CO_2, H_2O, NH_3, light and heavy hydrocarbons) and can remove almost all of them. However, removing CO and inert gases such as N_2 and argon are the main challenge due to the low adsorption capacity of conventional adsorbents (zeolites and activated carbons). TSA is considered a promising technique among PSA and VSA to increase H_2 recovery and purity but it consumes more energy due to the additional cost of the heating and cooling step. In the MH process, developing catalyst-free systems is critical to reducing the cost of the process. Additionally, it requires a large amount of heat transfer which reduces the efficiency of the system. The cryogenic process is one of the mature technologies and has been in operation for many decades. It also contains challenges which need to be improved i.e., high capital cost and low H_2 purity of 95%–98%.

It is worth mentioning that all the conventional and emerging technologies are limited and achieving the desired purity level by a single method is challenging. Therefore, hybrid or integrated separation systems are required to meet the purity levels of H_2 for high-end applications.

Declaration of competing interest

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

Data availability

Data will be made available on request.

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