**Introduction:**

Fossil fuels, such as coal, natural gas, and oil, as well as products made from them, provide the world's energy needs. These limited resources contribute to a country's existence. But they still remain critical due to their impact on pollution, global warming, and climate change by releasing CO2 and other greenhouse gases [1]. Nations that have identified the challenges associated with these resources have begun to try to minimize their economic reliance on fossil fuels. Renewable resources are already a significant component of the world's energy mix. A number of individuals who are interested in energy believe that one day we will be able to obtain energy from H2 in a sustainable and clean way [2]. Because of its numerous applications, H2 is increasingly in demand over other traditional fuels. Hydrogen has a high energy content, which provides a comprehensive benefit, as it has a lower heating value (LHV)/kg than its fossil fuel-based counterparts like methane and gasoline, which have LHVs that are 2.4 and 2.8 times greater, respectively [3-4]. Hydrogen is a component of power-to-gas technology, which is a power grid balancing mechanism used to collect and preserve surplus energy for usage during periods of limited supply (e.g., at night or when solar and wind are employed as energy sources) [5]. Hydrogen is gaining popularity as an energy carrier due to its several unique features. It can be created or transformed into electricity using electrochemical systems such as fuel cells and electrolysers with comparatively high energy conversion efficiencies. Hydrogen can be stored in a variety of ways including compressed gas, Cryogenic Liquid and Solid State [6-9]. In addition, hydrogen can be produced cleanly and emits no greenhouse gases. Hydrogen has also the ability to supply energy to the primary economic sectors of transportation, construction, and industry [10-11].

At present, Hydrogen is considered as an essential commodity that is used in various industrial processes, including the food industry, petrochemical and petroleum refining, ammonia production, and Hydrogenation process [12]. It can also be used in the production of power from fuel cells, transportation, and energy storage [13].

**Different types of Hydrogen Purification System:**

Hydrogen purification technology is related to Hydrogen production and Hydrogen utilization. Since none of the aforementioned processes produce pure hydrogen, purification is required to produce hydrogen with the requisite purity for its intended downstream applications. Hydrogen can be produced in various methods including coal gasification, natural gas reforming, water electrolysis and photo electrolysis. But the hydrogen produced by this technique is called cured Hydrogen. It cannot be immediately used for Fuel cells and others without purification [14-15].

**Table:** Summary of Hydrogen Production Process and the impurity content in Hydrogen.

|  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- |
| Methods | H2 | CO | CO2 | CH4 | H2O | N2 | O2 |
| Biomass Gasification [16] | 30-50 | 25-40 | 8-20 | 6-15 | -- | -- |  |
| Steam methane reforming [17] | 70-75 | 10-15 | 10-15 | 1-3 | -- | -- |  |
| Coal Gasification [18] | 35-38 | 26-28 | 23-24 | 5-6 | 15-20 | 6-7 |  |
| Methanol purge gas [19] | 70-80 | 4-8 | 5-10 | 2-8 | --- | 5-15 |  |
| Synthetic ammonia tail gas [20] | 60-75 | -- | -- | -- | -- | 15-20 |  |
| Water Electrolysis [21] | 60-80 | -- | -- | -- |  |  | 30-20 |

Following are typical methods for hydrogen recovery: Impurities can be removed using permselective membrane, cryogenic distillation, or pressure swing adsorption (PSA)[22]. These methods will be elaborated in the section. These explanations, in our opinion, will help readers understand how membrane-based separation differs from conventional PSA and cryogenic distillation.

PSA is the most commonly used modern industrial technique for hydrogen separation. The type of adsorbent and the technical procedure are the main determinants of the PSA separation effect. H2 is ideal for PSA separation and purification since it greatly differs in static capacity from the bulk of gas molecules, including CO2, CO, and CH4 [23]. The process includes five major steps entitled Adsorption, concurrent depressurization, counter-current depressurization, purge, and counter-current pressurization [24]. Basically PSA separates hydrogen by employing pressure-dependent adsorption of distinct polarities of gas in solid adsorbents (such as zeolites, alumina, activated carbon and silica gels). The polarized impure gases such as CO2, N2 and CH4 are captured under high pressure, while the non-polar hydrogen molecules are collected at the top of the adsorption column, achieving a high purity of 99.999% with 80%-85% hydrogen recovery [25-26]. PSA also works on a cyclical basis to renew spent adsorbents for subsequent adsorption processes. PSA system can be quite cost-effective if high input gas and high flow rates are used [27].

Conversely, Cryogenic distillation is a popular low-temperature separation method [28]. Cryogenic distillation separates substances based on boiling point differences. Cryogenic distillation process is separated into two types: cryogenic condensation and cryogenic adsorption, both of which take advantage of hydrogen's ultralow boiling point (253°C at 1 bar). The first type condenses the impurities with low boiling points into a liquid phase, whilst the second type selectively adsorbs the contaminants using adsorbents [29]. Pretreatment of the supply gas is also required to remove components that may freeze; thus, water should be decreased to 1 ppm and CO2 to 100 ppm [30]. This approach is not viable for obtaining high-purity hydrogen; however, increased hydrogen recovery can be achieved at moderate hydrogen purity yields (95%). Both Cryogenic distillation and PSA are ideal at large industrial scales but unsuitable for tiny, portable applications [31].

Membrane separation, as a new gas separation technology, offers the advantages of flexible and easy operation, compact structure, low energy consumption, and friendly to the environment. The raw material components can selectively permeate the membrane under the action of driving forces (pressure variation, concentration variation, and potential variation) in membrane separation technology with a perm-selective membrane as a separation medium, achieving separation and purification [32]. Besides PSA and cryogenic distillation, Hydrogen purification by membrane separation is competitive in its own right and is regarded as a possible method for producing hydrogen-enriched gas streams [33]. The Separation of Hydrogen from other impurities by membrane is based on solution diffusion mechanism. Hydrogen has a higher diffusivity because it can diffuse quicker than other gas constituents, owing to the short kinetic diameter of hydrogen molecules [34].

**Types of Hydrogen Separation Membrane:**

Firstly, Membrane can be Natural and Synthetic then It is divided into Organic (polymer), Inorganic and Mixed-matrix(Hybrid) [35]. (Classification are showed in Figure).

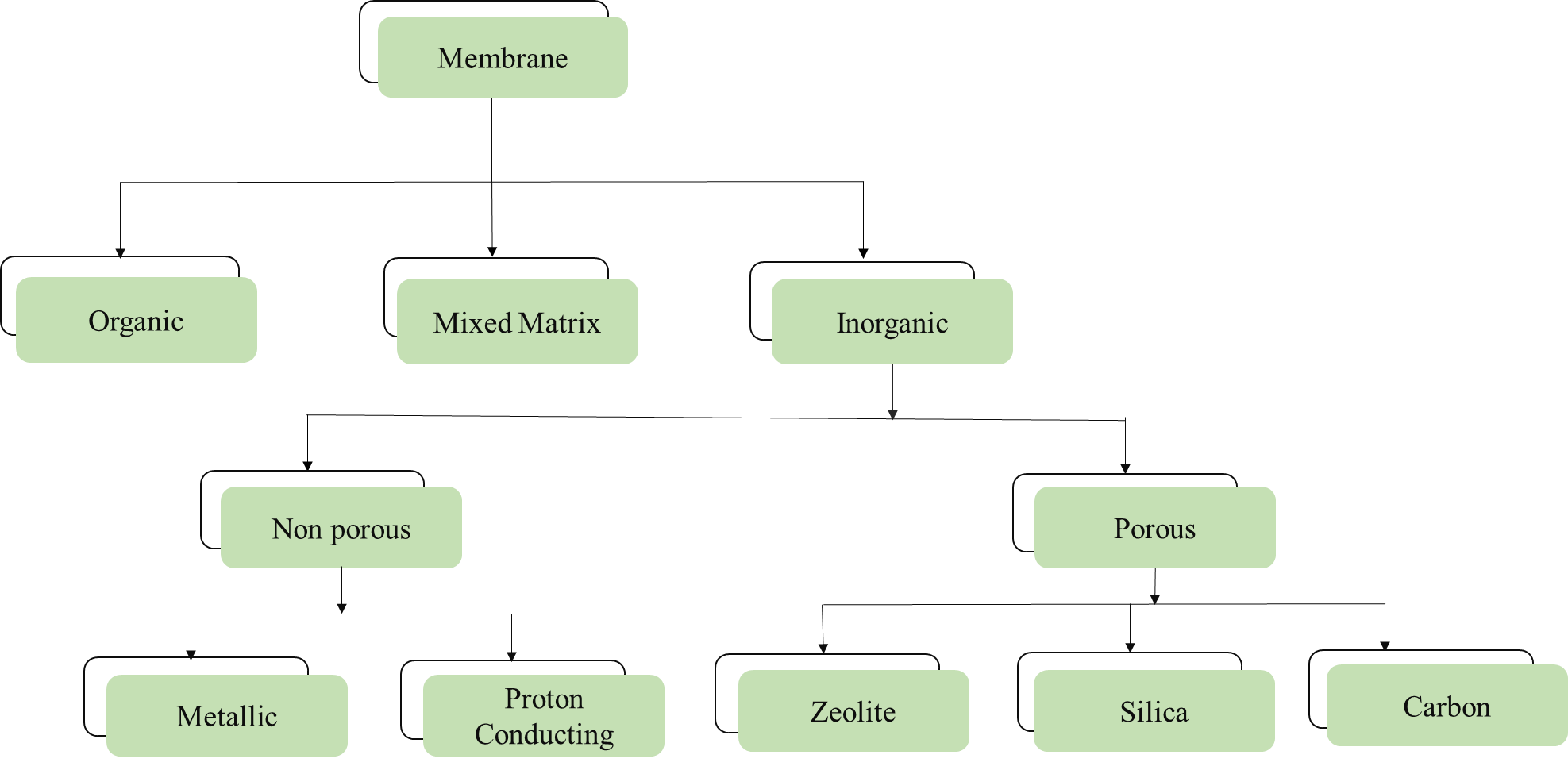


Figure: Classification of Membrane [36].

**Inorganic Membrane:**

Inorganic membranes are usually used for the purpose of isolating light gases like H2 from a gas mixture. Inorganic micro porous membranes are more affordable and can withstand higher temperatures (25°C-900°C) than others membranes [37]. Inorganic micro porous membrane transports H2 mostly in molecular sieving method, which separates H2 (the smaller kinetic diameter **Figure**), from CH4 and other bigger component from gases mixture. To successfully isolate hydrogen, the pore diameter of porous membranes must be less than 2 nm (e.g., microporous) [38]. Basically, Inorganic membranes are particularly appealing for hydrogen separation because they can resist high temperatures and pressures. Inorganic membranes are classified into two types: porous and dense (non-porous) [39]. In contrast, fractionation in porous membranes (silica, zeolites, and carbon) is based on differences in size, shape, and/or affinity between permeating molecules and the membrane [40].

Table: Kinetic Diameters of gas Particles [41]

|  |  |
| --- | --- |
| Gas | Kinetic Diameter of Gas particles (Å) |
| Hydrogen (H2) | 2.890 |
| Carbon dioxide (CO2) | 3.64 |
| Nitrogen (N2) | 3.3 |
| Methane (CH4) | 3.758 |

**Zeolite:**

Zeolites are aluminosilicate solids that are three-dimensional, crystalline, hydrated, and made of tetrahedra building units linked to one another by oxygen atoms. Zeolites have cavities and channels which are distinct and filled with alkali or alkali earth Cations and water molecules. Zeolites are more appealing than other microporous materials due to their superior heating, mechanical, and chemical stabilities [42]. They provide consistent holes with molecular diameters and unique features for catalytic, ion exchange, adsorption, and membrane applications [43]. Recent research has shown that zeolite membranes can separate H2 at a variety of temperatures (25 °C-700 °C), but with significantly lower H2/CH4 selectivity than thick metallic membranes. The existence of inter-crystalline mesopores on the zeolite layer, which formed during nanocrystal aggregation, could explain the low hydrogen selectivity [44-45]. In recent years, certain innovative fabrication procedures that successfully minimized defects in zeolite membranes were examined such as free hydrothermal synthesis [46], catalyst cracking deposition hydrothermal synthesis [47], functionalising the zeolite pores by amorphous materials [48] . For various gas separation applications, zeolites can be produced in varied forms, particle sizes, and pore sizes. As zeolite membranes such as LTA, CHA, MFI, DDR, and FAU have been effectively produced for gas separation.

|  |  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- | --- |
| **Zeolite** | **Support** | **Hydrogen**  **Permeability** | **Selectivity** | | | **Technique** | **Experiment**  **Condition** | **Ref** |
| **H2/CO2** | **H2/CH4** | **H2/N2** |
| STT | Silica | 2.8 × 10−8 mol·m−2·s−1·Pa−1 | **----** | 49.6 | **-----** | Hydrothermal | 0.2 MPa 298 K | [49] |
| MFI | Graphene Oxide | 1.3 × 10-5 mol m-2 s-1 Pa-1 |  | 245 |  | Colloidal |  | [50] |
| DDR3 |  | 1.8 × 10− 8 mol m− 2 s − 1 Pa− 1 |  | 124 |  | Hydrothermal | 0.4MPa  303 k | [51] |
| FAU | Al2O3 tubes | 1.9 × 10-7 mol m-2 s-1 Pa-1 |  | 9.9 |  | Hydrothermal | 1 Bar  50 °C | [52] |
| MFI | Alumina | 1.1 × 10-7 mol ·m-2 ·s-1 | 8.2 |  |  | Hydrothermal | 450°C | [53] |
| SSZ-13 | MDI | 4.27 × 10− 8 mol m− 2 s − 1 ⋅Pa− 1 |  | 427 | 35.6 | Hydrothermal | 2 bar  373k- 473k | [54] |
| SAPO-34 | α-Al2O3 | 6.96 × 10 -6 mol m -2 s -1 Pa -1 | 1.83 | 14.80 | 7.58 | Hydrothermal | 298 k | [55] |
| Si-CHA | α-Alumina | 1.44 × 10−6 mol m-2 s-1 Pa-1 |  | 85 |  | Hydrothermal | 0.2 MPa  298K | [56] |
| ZIF-7 | Alumina | 4.5 × 10−8 mol m−2 s−1 Pa−1 | 13.6 | 18.0 | 14 | Hydrothermal | 220°C | [57] |
| LTA (double layer) | α-Al2O3 | 2.1×10 -7 mol m -2 s-1 Pa -1 | 8.8 | 5.8 | 7.2 | Hydrothermal | 1 bar  373K | [58] |

**Silica:**

|  |  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- | --- |
| **Silica** | **Support** | **Hydrogen**  **Permeability** | **Selectivity** | | | **Technique** | **Experiment**  **Condition** | **Ref** |
| **H2/CO2** | **H2/CH4** | **H2/N2** |
| SiO2 | Ceramic | 1×10-7 mol m -2 s-1 Pa -1 |  | 2800 |  | Sol-gel | 1 bar, 100°C-600°C | [59] |
| CoSiO2/ZrSiO2 | α- Al2O3 | 1.8 ×10-7 mol m -2 s-1 Pa -1 |  | 480-730 |  | Sol-gel | 1-3 bar  200°C-500°C | [60] |
| ZrO2Y2OSiO2 | Ceramic | 1×10-8 mol m -2 s-1 Pa -1 |  | 4 |  | Sol-gel | 1-1.5 bar  150°C | [61] |
| MTES-SiO2 | α- Al2O3 | 3.4 ×10-9 mol m -2 s-1 Pa -1 |  | 24-46 |  | Sol-gel | 1-6 bar  50°C-200°C | [62] |
| SiO2 | γ- Al2O3 α- Al2O3 | 5 ×10-7 mol m -2 s-1 Pa -1 |  | 5900 |  | Sol-gel | 2 bar  600°C | [63] |
| SiO2 modified | Vycor glass | 1.8 ×10-8 mol m -2 s-1 Pa -1 |  | 23000-27000 |  | High temperature atmosphere CVD | 1.2 bar 200°C-700°C | [64] |
| BTESE | α- Al2O3 | 1.8 ×10-7 mol m -2 s-1 Pa -1 | 16 | 400 | 100 | Sol-gel | 2 bar 200°C | [65] |
| TEOS/MTES | γ- Al2O3 α- Al2O3 | 1.5 ×10-6 mol m -2 s-1 Pa -1 | 8 |  | 18 | Sol-gel | 1 bar 200°C | [66] |
| TEOS/TEPTES | γ- Al2O3 α- Al2O3 | 3.1 ×10-6 mol m -2 s-1 Pa -1 | 15.2 |  |  | Sol-gel | 1 bar 200°C | [67] |
| TEOS/TEPTES | γ- Al2O3 | 9.7 ×10-7mol m -2 s-1 Pa -1 | 12.1 |  |  | Sol-gel | 1 bar 300°C | [68] |

**Carbon:**

|  |  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- | --- |
| **Carbon** | **Support** | **Hydrogen**  **Permeability** | **Selectivity** | | | **Technique** | **Experiment**  **Condition** | **Ref** |
| **H2/CO2** | **H2/CH4** | **H2/N2** |
| PEK-C |  | 5260 Barrer |  | 311 | 142 | Pyrolysis | 30°C and 0.01 bar | [69] |
| Cellulose hollow fiber |  | 111 GPU | 36.9 |  |  | Pyrolysis | 110°C and 10 bar | [70] |
| 6FDA/BPDA-TMPDA |  | 3.4-7.5×10-8 mol m -2 s-1 Pa -1 |  | 400-500 |  | Pyrolysis | 1-8.3 bar  25°C | [71] |
| Kapton Polymide |  | 3.1×10-10 mol m -2 s-1 Pa -1 |  |  | 150 | Pyrolysis | Up to 6 bar  30°C – 250 °C | [72] |
| Phenol-formaldehyde novolac resin | Resin tube | 1.7×10-9-1.2×10-7 mol m -2 s-1 Pa -1 |  | 23.1-45.2 | 24.1-39.5 | Pyrolysis | 2 bar 25°C | [73] |
| Polymide |  | 3.4×10-7-6.0×108 mol m -2 s-1 Pa -1 |  | 132-631 |  | Pyrolysis | 10.8 bar 30°C – 120 °C | [74] |
| Polymide |  | 1.4×10-7 mol m -2 s-1 Pa -1 |  | 540 |  | Pyrolysis | 10 bar 80°C | [75] |
| OrL | α alumina tube | 1.3×10-7 mol m -2 s-1 Pa -1 |  | 584 | 293 | Pyrolysis | 1.1 bar 35°C | [76] |
| PPO/PVP | Alumina | 1121 Barrer |  | 160.9 | 163.9 | Pyrolysis | 2 bar 25°C | [77] |
| PI | Alumina | 376 Barrer |  | 16.4 | 33.2 | Pyrolysis | 2 bar 25°C | [78] |
| PEI | Alumina | 1300 Barrer |  | 174 |  | Pyrolysis | 2 bar 25°C | [79] |
| CHFM |  | 5×10-8 mol m -2 s-1 Pa -1 | 83.9 | 5700 | 800 | Pyrolysis | 2 bar 130°C | [80] |

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