

# A Carbon Dioxide Gas Separation Device for Industrial Hydrogen Production Process

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**Abstract:** To facilitate the use of carbon dioxide separation technology in industrial hydrogen production, we have developed a device that can increase the carbon dioxide gas recovery rate to 95% under existing production conditions. This device solves the issues associated with the complex operation of traditional industrial gas separation spherical tanks, including their complex operation, inefficient gas recovery rates ranging from 60%-70%, as well as the toxicity and difficulty in disposing of carbon dioxide treatment agents. The device utilizes the selective permeability of an iron mordenite-containing sieve plate for direct gas injection. The sieve plates are welded to a carbon dioxide gas tank for sample injection, forming an integrated structure. The sieve plate and the sample injection structure are then fixed and sealed using an ultra-high vacuum sealant.

**Keywords:** Industrial Emissions, Carbon Dioxide Capture, Large Spherical Tank, Mordenite, Zeolite Molecular Sieve

## 1. Introduction

In the context of China's 'carbon neutral carbon peak' policy, the development of the hydrogen energy industry is expected to replace traditional fossil fuels. However, traditional industrial gas separation methods that involve the use of spherical tanks have complicated operations, low gas recovery efficiency, and difficulties in disposing carbon dioxide treatment agents. These limitations have greatly restricted the progress of the hydrogen energy industry[1].

In the process of industrial hydrogen production, such as in the petrochemical, coal-to-oil chemical, natural gas production, and chemical transformation industries, significant amounts of CO<sub>2</sub> are often emitted. To meet the requirements of subsequent synthesis and processing, it is generally necessary to remove and store CO<sub>2</sub> for treatment[2]. Using the industrial gas separation tank at the Shanghai No.2 Power Plant as a research prototype, it can be observed that traditional industrial gas separation methods that involve the use of spherical tanks lack the necessary devices to treat gas mixtures, necessitating multiple steps for separation and pollution-free treatment. In this process, the carbon dioxide treatment agents that are currently used on a large industrial scale can produce toxic substances that are harmful to both humans and the environment. Additionally, the disposal of these products is often difficult[3].

In this paper, we address the aforementioned issues by proposing a solution that involves the use of special materials to increase the contact area between the carbon dioxide in the spherical tank and the treatment agents, as well as the implementation of an improved sieve plate technology that utilizes more efficient screening methods. Our proposed approach aims to achieve a green and efficient chemical industry, while avoiding the need for expanding the area of equipment. This solution not only saves land but also facilitates efficient treatment, thereby creating a win-win situation. The research focuses on four key aspects: ① Addition of Wire-Containing Photozeolite Adsorbent; ② Design of Industrial Hydrogen Production and CO<sub>2</sub> Separation Processes; ③ Design of Carbon Dioxide Gas Separation Spherical Tank; ④ Design of New Screening Technology for Hydrogen and Carbon Dioxide. The proposed solution effectively addresses the issue of carbon dioxide emissions during hydrogen production, achieving a carbon dioxide recovery rate of more than 95%. The project results make significant contributions to the green and low-carbon development of the hydrogen energy industry.

## 2. Technical Background and Technical Difficulties

### 2.1 Technical Background

China's reliance on imported crude oil exceeds 50%, creating a need for the exploration of alternative sources of energy that can reduce its dependence on foreign oil[4-5]. Hydrogen energy is a promising solution to mitigate the greenhouse effect, as it serves as a carbon-neutral fuel that combusts efficiently without dependence on fossil fuels. Additionally, hydrogen combustion does not produce toxic emissions, making it an eco-friendly and pollution-free energy source that supports green environmental protection initiatives. Overall, hydrogen energy represents an excellent opportunity to advance towards a cleaner and more sustainable future. Establishing a new industrial system where "green hydrogen" serves as the primary energy source is crucial for achieving "carbon emissions peak". This approach entails leveraging the unique properties of green hydrogen to drive sustainable development and reduce carbon emissions in various industries. By prioritizing the implementation of green hydrogen technologies, we can significantly mitigate global warming and work towards a more sustainable and eco-friendly future[6-7]. Currently, China primarily relies on fossil fuels as the raw materials for hydrogen production, a process that often leads to a rapid increase in carbon dioxide emissions. This poses a significant challenge for the country's efforts towards achieving sustainable and eco-friendly development goals[8]. Hydrogen production from natural gas involves the conversion of methane and water vapor into carbon monoxide and hydrogen through a reforming process. The waste heat generated during this process is then recovered and used to facilitate the conversion of carbon monoxide into carbon dioxide and hydrogen via a conversion tower. This process has been extensively studied and documented in academic literature. Currently, a prominent issue that constrains the development of hydrogen energy is the excess of carbon dioxide produced during the process. The continuous rise of carbon dioxide levels results in an unstable carbon balance, leading to the intensification of the greenhouse effect and potential harm to the environment[9]. From this perspective, it can be inferred that mitigating vast carbon dioxide emissions is the fundamental solution towards sustainable development of the hydrogen industry.

### 2.2 Technical Difficulties

#### 2.2.1 The Gas Mixture Generated During the Industrial Hydrogen Production Process Presents Significant Challenges In Terms Of Separation and Operational Complexity

Having already undergone two major energy transitions, modern society is currently embarking on its third renewable energy transition in order to achieve carbon neutrality and foster sustainable development for humanity. In light of recent developments, China's energy sector is now confronted with a multifaceted and challenging set of circumstances. To ensure both the preservation of energy security and the rapid attainment of carbon emissions peak, it is crucial that we accelerate the transition towards low-carbon energy sources while maintaining a steadfast focus on our bottom-line goals. In light of the current circumstances, the government is steadfast in its commitment to the promotion and development of "green hydrogen". Due to the weaker development of the green hydrogen industry in China compared to other countries, there are considerable weaknesses in industrial hydrogen production, where certain instruments and technologies remain immature in practice. Consequently, there is a wide variation in the composition and physical properties of the mixed gases generated through the current industrial hydrogen production paths[10]. The mixed gas, which includes hydrogen, carbon dioxide and sulfide gas, presents varying properties, and therefore, requires a tailored gas separation process according to the unique characteristics of each gas. This process can be labor-intensive, and the recovery rate for carbon dioxide after separation is limited to a maximum of 82%.

#### 2.2.2 The Separation and Recovery Efficiency of Carbon Dioxide in the Gas Mixture Produced After Industrial Hydrogen Production Is Relatively Low

Generally, the following two methods are used:

##### (1) Chemical Absorption Method for Polishing

Chemisorption involves the chemical reaction between an adsorbent and CO<sub>2</sub>, and is used to separate and recover CO<sub>2</sub> from exhaust gas. Commonly used chemical absorbents for CO<sub>2</sub> separation and recovery include monoethanolamine (MEA), diethanolamine (DEA), and methyl diethanolamine (MDEA). Currently, the main existing problems are: ① Challenges arise due to entrainment and bubbles

in the absorber, resulting in a complex flue gas purification system, high energy consumption, and significant investment requirements;②Since flue gas typically contains small amounts of gases such as SO, CO, SO<sub>2</sub>, and others, high-temperature conditions in the regeneration tower promote absorption liquid reactivity, reducing its concentration and overall efficiency. Additionally, these conditions also corrode the regeneration tower and can negatively impact overall equipment lifespan;③When processing blast furnace gas, the need to maintain temperatures below 100°C necessitates the transfer of high-temperature gas, leading to additional treatment equipment and investment requirements;④The maximum carbon dioxide recovery rate attainable is 87%[11].

### (2)Physical Absorption Method for CO<sub>2</sub> Capture

The physical absorption method for CO<sub>2</sub> capture primarily employs absorbents such as water, methanol, and propylene carbonate. This method utilizes the principle that the solubility of CO<sub>2</sub> in these solutions varies with pressure to effectively absorb CO<sub>2</sub> gas. Mainly operating at low temperatures but high pressures, the physical absorption method offers significant absorption capacity, less usage of absorbents, does not require regeneration heating, allows for solvent non-foaming, and minimizes equipment corrosion. This method can only be used under conditions of high partial pressure of CO<sub>2</sub> gas and has a low recovery rate, reaching only 78%[12].

### ***2.2.3 Improvements are Required In the Selection of CO<sub>2</sub> Treatment Agents for Large-Scale Industrial Applications***

Although there is a low risk of CO<sub>2</sub> leakage during capture and transportation, the primary environmental risks arise from the storage and utilization of CO<sub>2</sub>. To treat mixtures following the industrial hydrogen production process, plants employ treatment agents to separate hydrogen and carbon dioxide based on the gas characteristics, temperature, and pressure of the system[13]. CO<sub>2</sub> is less dense than oil and water, but more dense than hydrogen. Following separation, gas will continue to migrate downward with the movement of the spherical tank and the corrosion of CO<sub>2</sub> dissolved in water intensifying CO<sub>2</sub> migration. In the event of a CO<sub>2</sub> leak and subsequent escape to the surface, the ecological environment can be significantly impacted. Thus, ensuring the safe production of carbon dioxide treatment agents is a necessary prerequisite for the industrialization of hydrogen production. However, as geological factors vary by location, the development of a standardized environmental assessment system capable of quantifying potential impacts can be challenging[14]. Currently, CO<sub>2</sub> utilization technology is in the industrial demonstration stage. To improve carbon utilization efficiency, the key research direction for CO<sub>2</sub> utilization technology in the next stage is breaking through the bottleneck of high-temperature, high-pressure environments and identifying more suitable CO<sub>2</sub> treatment agents.

## **3. Device Development**

### ***3.1 Design Parameters and Material Selection***

#### ***3.1.1 Design Parameters***

In accordance with design conditions and GB12337-2014 'Steel Pressure Vessels,' the harsh conditions experienced by spherical tanks necessitates manufacturing with composite steel plates composed of S30403 and Q345R materials. Such stainless steel composite plates offer the comprehensive characteristics of both low alloy and coating stainless steel, reflecting a combination of cost-effectiveness and high performance. Stainless steel composite plate spherical tanks hold promising application prospects in the petrochemical, medical, food, and related industries. As a resource-saving material, it can address corrosion issues while fulfilling required use strength criteria, effectively reducing equipment material costs, improving equipment service life, and lowering engineering costs. Additionally, for manholes and nominal diameter DN300 nozzle flanges, carbon steel forgings with an inner wall surfacing structure are selected to avoid using stainless steel forgings for large diameter nozzle flanges, thus reducing material costs to a certain extent[15]. Table 1 displays the specific parameters of the device.

*Table 1: Design parameters for CO<sub>2</sub> gas separation devices in industrial hydrogen production processes.*

Operating Conditions	Regeneration Process	Normal Operation Conditions
Working Pressure of the Device/ Mpa	0.2	5.84
Design Pressure/ Mpa	0.3	6.72
Operating Temperature/ °C	260	40
Design Temperature/ °C	290	70
Medium	Recovery Gas	Hydrogen, Isobutanol, Ethanol
Medium Characteristics	One	Explosive
Corrosion Allowance/ mm	3	3
Weld Joint Factor	1	1
Geological and Environmental Conditions	The basic wind pressure is 400N/m <sup>2</sup> , seismic intensity is 7, design basic acceleration is 0.1g, design seismic group is the first group, site soil classification is II, and ground roughness is B.	

### **3.1.2 The selection of materials**

Although the design temperature of the separation device in this project is 290 °C under the regeneration condition, the alternation of regeneration and the operating condition is disconnected, and in the two working conditions, the two prerequisites for hydrogen corrosion "design temperature greater than or equal to 200 °C" and "hydrogen atmosphere contact" do not intersect, so there is no need to put forward material requirements and restrictions for hydrogen corrosion, and accurate judgment of equipment condition is crucial to affect economic rationality [16]. The pressure of the separation device is high, the two prerequisites for calculating hydrogen corrosion on the tank wall thickness is relatively thick, the pressure vessel with low alloy steel Q345R, can meet the corrosion requirements of the separation tank, with good comprehensive mechanical properties and process performance, so the equipment shell can be selected, supplied according to the normalized state, and UT testing is carried out one by one, technical grade B, in line with NB/T47013.3-2015 "Nondestructive testing of pressure equipment Part 3: Ultrasonic testing" UT-II. grade is qualified [17]. Since the thickness and strength of commonly used steel pipes cannot meet the requirements of welding and calculation, 16MnIII. Forgings with similar composition to Q345R are selected as thick-walled pipes. The flange and flat cover are matched with 16Mn III. forgings. The skirt seat uses a 300mm long low-alloy steel cylinder in contact with the lower head as the transition section, and the rest of the skirt components are made of carbon steel Q235B with low price and good rigidity, which is economical and meets the requirements of use [18].

## **3.2 Strength Calculation and structural design**

### **3.2.1 Strength Calculation in Engineering Design**

The dimensions of the carbon dioxide gas separation device in the process of industrial hydrogen production are shown in Figure 1. Taking GB150.1~150.4-2011 "Pressure Vessel" as the basis for strength calculation [19], the SW6 calculation software certified by the General Administration of Quality and Technical Supervision, Inspection and Quarantine of the People's Republic of China was used to calculate the equipment cylinder, head and hole reinforcement, and then determine the nominal thickness of each piece. The separation tank is supported by a skirt seat, and the strength of the skirt can be simulated and calculated with reference to NB/T 47041-2014 "Tower Container" [20].

### **3.2.2 Structural Design in Engineering**

Taking the industrial separation gas tank of Shanghai Second Power Plant as a research prototype, the structure of the existing spherical gas tank is improved, and two inverter fans are placed on both sides of the diameter, which can use the wind to blow the lightweight block wire-containing fluorescent zeolite into the screen plate in the middle of the spherical gas tank, so as to avoid the safety problems and sealing problems caused by the replacement device [21]; Because the hydrogen medium is very dangerous, there is a small leakage may cause an explosion Externally installed intelligent gas detector for detecting whether the carbon dioxide in the tank is fully absorbed by iron-containing zeolite and whether the remaining gases such as hydrogen leakage, in the outside of the spherical gas tank also need to install intelligent vacuum gas detector device, pressure gauge and other instruments, and then connect to the computer to achieve automation.

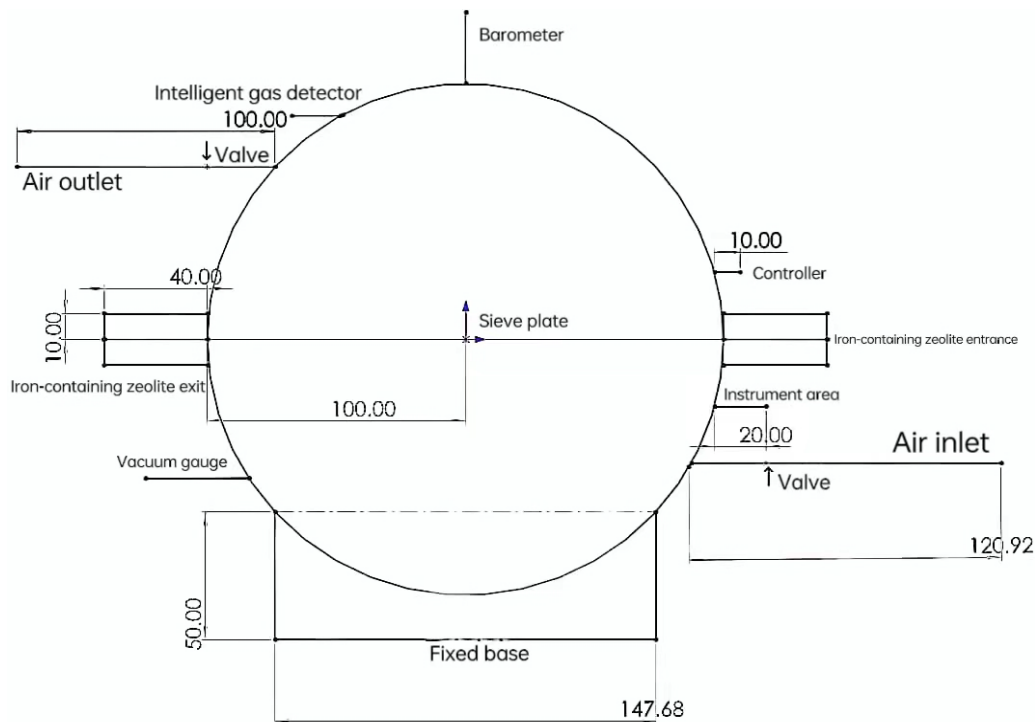


Figure 1: Overall design of carbon dioxide gas separation device in the process of industrial hydrogen production.

As shown in Figure 1, along the opening of the upper end of the carbon dioxide ball gas tank into carbon dioxide and hydrogen, the middle of the ball tank is put into the sieve plate in the diameter direction, and a large amount of iron-containing zeolite (where the sieve plate pore structure) is put into the sieve plate when the middle iron-containing zeolite sieve plate is put into the gas, due to the different density of carbon dioxide and hydrogen makes it naturally stratified, iron-containing zeolite can adsorb carbon dioxide and carry out further treatment, hydrogen is collected from the upper part to achieve separation.

Due to the different air density of carbon dioxide and hydrogen, hydrogen in the upper half of the sphere carbon dioxide concentrated through the middle iron-containing zeolite sieve plate, if there is a small amount of omission, it will also be due to the upper hydrogen too much pressed into the middle sieve plate, the final gas detection standard, from the upper valve opening of the sphere to absorb hydrogen, the lower valve opening of the spherical tank absorbs carbon dioxide, the light block iron-containing wire zeolite in the sieve plate absorbs carbon dioxide, and finally uses wind power to blow it out and collect it for the next step of processing.

The selection of sieve plate hardness corresponds to the strength of the material, and the maximum allowable stress is calculated by calculating the sealing force by the sealing specific pressure, and then the corresponding conversion is converted into hardness. Due to long-term use, the sieve plate needs to use anti-corrosion materials, the size of the sieve plate pores is designed according to the diameter of the selected iron-containing zeolite (generally less than 2mm), in order to prevent the absorption of carbon dioxide after the iron-containing zeolite is blown out when blocking the pores of the sieve plate, the pores are designed for two layers. The lower layer is a retractable iron plate, and the upper layer is a sieve plate with pores (Figure 2). The size of the sieve plate is 100.00cm\*100.00cm in the middle of the spherical gas tank, and the height is 10.00cm.

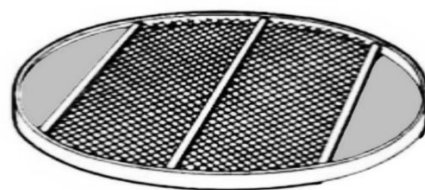


Figure 2: Design drawing of sieve plate.

### 3.3 Manufacturing, Inspection and Acceptance Requirements

External factors such as temperature and pressure can cause changes in the strength, elastic modulus and plasticity of steel [22]. In this paper, the influence of external factors such as temperature and pressure on the separation of carbon dioxide and hydrogen in gas storage tanks is explored by constructing a small simulation device. The spherical tank used in this project is made of stainless steel splicing, if the steel is affected by external conditions, it may cause the steel material to change, causing certain safety hazards. It is necessary to explore and constantly try to obtain the best temperature, pressure and other external conditions to make the industrial hydrogen and carbon dioxide gas separation spherical tank work project safe and stable.

The separation tank puts forward corresponding requirements for equipment manufacturing, inspection and acceptance according to the safety technical supervision regulations of fixed pressure vessels (TSGR0004-2009) and pressure vessels (GB150.1~150.4-2011) [23]. The welding of the sieve plate requires a screen hole diameter of  $\leq 2$ mm. Before the equipment is welded, the weld seam of the equipment should be welded in accordance with the requirements of NBT47014-2011 pressure equipment welding process evaluation [24]. The equipment should prepare the base metal post-weld heat treatment product welding specimens, the size and quantity of the specimens are made according to the requirements of Article 9.2 in GB150.4-2011, and the inspection and evaluation standards of the specimens are carried out in accordance with Article 9.2.3 in GB150.4-2011 (The tensile, cold bending, and impact tests of the specimens are carried out in accordance with the provisions of GB/T288, GB/T232 and GB/T229, respectively, and are evaluated according to the requirements of GB150.2 and design documents[25].When the sample results cannot meet the assessment requirements, resampling is allowed for re-testing, and if the retest results still do not meet the requirements, the base metal is judged to be unqualified)). Before the shell is welded, the base metal should be preheated to  $\geq 150^{\circ}\text{C}$ . The testing requirements for the butt joint between the  $\text{DN} \geq 250$ mm pipe and the necked butt welding flange are the same as for the Class A and B welded joints of this equipment. The  $\text{DN} < 250$ mm pipe and the butt joint between the pipe and the butt joint with neck butt welding flange require 100% magnetic particle (MT) testing, which is qualified in accordance with the MT-I. Grade in NB/T47013.4-2015[26]. After the equipment welding is completed, the overall heat treatment should be carried out, and all the pipes, tangential feeding grooves, pre-weldments, nameplate holders, etc. must be welded before the heat treatment, and after the heat treatment is completed, no fire welding shall be carried out.

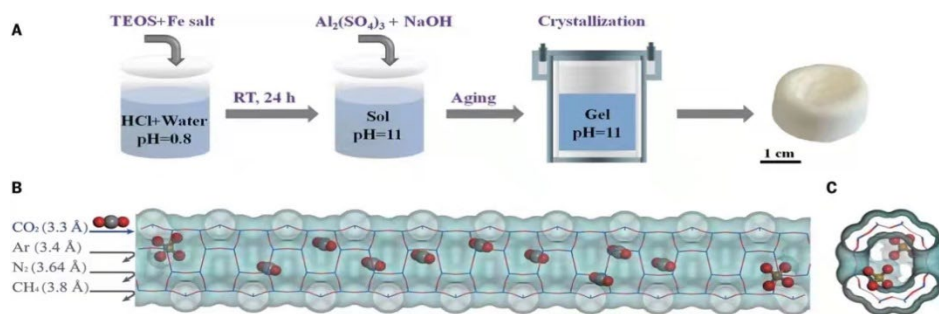
In the working process of industrial hydrogen and carbon dioxide gas separation spherical tank, because zeolite is a small particle, if a large amount of zeolite is stacked in the sieve plate for separation gas work, it is easy to make some zeolite not completely absorb carbon dioxide or even absorb carbon dioxide before being absorbed by the device, resulting in waste of raw materials; If there is only a small amount of zeolite in the sieve plate for gas separation, it is easy to make the gas to be treated incompletely re-acted, resulting in the upper and lower valves collecting hydrogen and carbon dioxide gas impure respectively. According to the amount of gas inflow, the amount of iron-containing zeolite is proportional to the amount of gas, and the excess iron-containing zeolite can be used for the next time.

## 4. Design Test and Result Analysis

The "acid hydrolysis" pathway synthesis method technology was introduced into the study, and the principle was to bind Fe ions into the MOR framework and prepare Fe-MOR through an unusual "acid hydrolysis pathway", so that Fe and Si/Al precursors were slowly co-condensed in the initial gel stage to finely control Fe doping. As shown in Figure 3, the created iron-containing optical zeolite adsorbent contains two characteristics, one is to change the original powder into a high mechanical strength block, eliminating the subsequent molding process, and has typical green chemical characteristics; Second, the unique pore structure realizes efficient carbon capture.

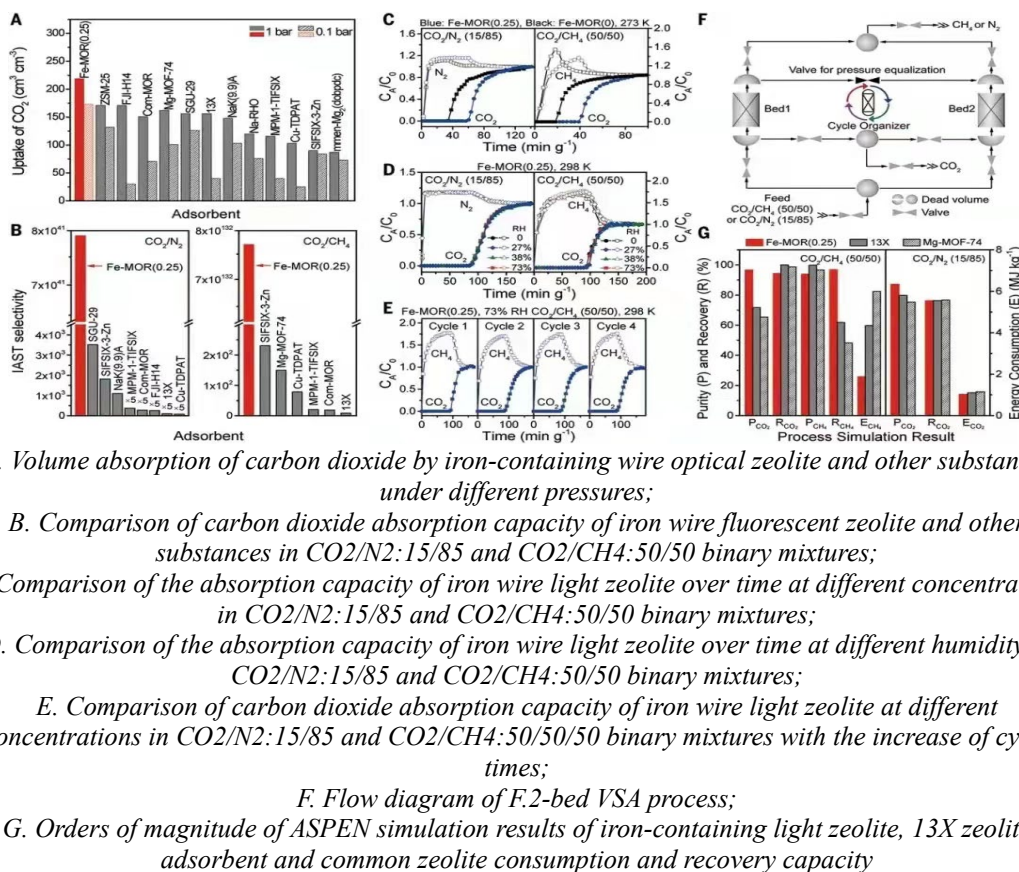
Iron-containing zeolite has a strong adsorption capacity for carbon dioxide, and the iron-containing light zeolite adsorption material shows good screening ability for argon, nitrogen, methane, etc. at room temperature, and its separation is orders of magnitude higher than that of 13X zeolite adsorbent (Figure 4), the strategy makes the Fe part occupy the microchannel, and its precise narrowing microchannel allows unique molecular screening ability. The diameter of carbon dioxide is 0.33 nm, and the orifice size of the zeolite adsorbent produced by the process is 0.33~0.34 nm, which can allow carbon dioxide to enter the adsorption material, and secondly, it can also prevent the entry of molecules such as

methane, so that this orifice has become a carbon dioxide exclusive "trap hole", and the adsorption is more accurate. Under the condition of the same purity, the recovery rate of carbon dioxide by the combined adsorbent is greater than 95% [27].



- A. Synthetic diagram of iron wire-containing light zeolite;  
 B. Schematic diagram of iron-containing light zeolite adsorption materials showing good screening ability for argon, nitrogen, methane, etc.;  
 C. Schematic diagram of the structure of iron-containing wire light zeolite

Figure 3: Self-assembly diagram of Fe-MOR.



- A. Volume absorption of carbon dioxide by iron-containing wire optical zeolite and other substances under different pressures;  
 B. Comparison of carbon dioxide absorption capacity of iron wire fluorescent zeolite and other substances in  $\text{CO}_2/\text{N}_2:15/85$  and  $\text{CO}_2/\text{CH}_4:50/50$  binary mixtures;  
 C. Comparison of the absorption capacity of iron wire light zeolite over time at different concentrations in  $\text{CO}_2/\text{N}_2:15/85$  and  $\text{CO}_2/\text{CH}_4:50/50$  binary mixtures;  
 D. Comparison of the absorption capacity of iron wire light zeolite over time at different humidity in  $\text{CO}_2/\text{N}_2:15/85$  and  $\text{CO}_2/\text{CH}_4:50/50$  binary mixtures;  
 E. Comparison of carbon dioxide absorption capacity of iron wire light zeolite at different concentrations in  $\text{CO}_2/\text{N}_2:15/85$  and  $\text{CO}_2/\text{CH}_4:50/50/50$  binary mixtures with the increase of cycle times;  
 F. Flow diagram of F.2-bed VSA process;  
 G. Orders of magnitude of ASPEN simulation results of iron-containing light zeolite, 13X zeolite adsorbent and common zeolite consumption and recovery capacity

Figure 4: Gas adsorption capacity.

## 5. Conclusion

Referring to the traditional industrial hydrogen separation spherical tank has been modified, on the way to process design improvement, this paper has made four changes without expanding the floor space of the equipment: (1) the use of special materials; (2) Increase the contact area of carbon dioxide with special materials in the ball gas tank; (3) Design and improve sieve plate and other technologies; (4) Use a more suitable method for screening.

In order to operate easily and recycle, better save raw materials, and absorb carbon dioxide with high efficiency, the selection of iron-containing zeolite can better separate carbon dioxide in the gas,

and the separation rate is increased from the original 60%~70% to 95%, realizing green and efficient chemical industry, and achieving a win-win situation that saves land occupation and efficient processing.

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