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Development and Prospect of Pressure Swing Adsorption (PSA) Technology for Carbon Dioxide Separation and Capture

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Abstract: Pressure swing adsorption is a kind of gas separation technology, because of high automation, low energy consumption, simple operation, widely used in carbon dioxide separation and capture, energy gas purification, chemical tail gas treatment. Pressure swing adsorption technology includes equipment technology, equipment control, special adsorbent and so on. With the continuous in-depth study of PSA technology, PSA technology has made great progress in the process, flow simulation, automatic control of equipment and adsorbent. In this paper, the development status of PSA technology, process simulation, equipment control and adsorbent is summarized, and the future development of PSA technology is prospected.

Keyword： *Pressure swing adsorption; Gas separation; Automation; Simulation; Adsorbent agent*

I. INTRODUCTION TO PSA

Pressure swing adsorption is an effective method for gas separation, concentration and purification. Because of its simple process, low energy consumption, less investment and convenient operation[1-6], it has been widely used in petrochemical industry, electronics, metallurgical industry, national defense, medical treatment, environmental protection and other aspects.Pressure swing adsorption technology can be used for purification of energy gas, carbon gas capture Table 1 In recent years, some pressure swing adsorption hydrogen production units in China.Pressure swing adsorption technology was first proposed by two foreign patents in 1958, and has been studied in China since the 1960s[7].Pressure swing adsorption technology has been studied from the process principle, simulation process, automatic control of equipment, pressure swing adsorption adsorbent and other aspects. The principle of pressure swing adsorption technology is based on the adsorption amount of different gases under the same pressure adsorbent is different, the adsorption amount of the same gas under different pressure is different, for the adsorption of special gas adsorption adsorbent in pressurization to a certain extent, the adsorption amount increases with the increase of pressure, and the adsorption amount decreases with the pressure reduction[8].Pressure swing adsorption commonly used adsorbents are activated carbon, molecular sieve, alumina, silica gel, etc.in the actual use of a variety of adsorbents will be filled in different proportions[9]. There are several methods to control pressure swing adsorption equipment, using PLC (programmable logic controller) and industrial control software combined with simple algorithm control or using single chip microcomputer and upper computer software combined with simple algorithm control.

Table 1 Partial pressure swing adsorption hydrogen production devices used in petrochemical systems in recent years[51]

company name	Plant handling capacity（m^3/h）	Type of raw gas	Product H_2 purity（%）
Daqing Oilfield Chemical Plant	50000	Catalytic cracking dry gas	99.9
Zhenhai Refining and Chemical Company	50000	Refinery gas mixture	99
Liaoyang Chemical Fiber Company	40000	refinery gas	99.9
Golmud oil Refinery	8500	Hydrogen containing gas	99.999
Organic synthesis plant of Jichua Corporation	5800	Ethylene exhaust gas	99.5
jinan refinery	15000	Catalytic cracking dry gas	99.9
Puyang methanol plant	7000	Methanol is relaxed and deflated	98.5
Shengli Petrochemical General Plant	12000	Gas of change	99.9

II. PSA SEQUENCE STEP DESIGN

Through continuous research on PSA technology, new technologies such as rapid PSA and vacuum PSA have been gradually formed[10]. No matter what kind of new process, PSA process generally includes: adsorption (A), uniform pressure drop (ED), reverse discharge (D), vacuum (V), uniform pressure rise (ER), final charge (FR) and other steps[11].

Adsorption: Under a certain pressure, the raw material gas enters from the adsorption tower and is in full contact with the adsorbent. The gas with large adsorption capacity is first adsorbed, and the product gas with small adsorption capacity is discharged and collected from the outlet.

Uniform pressure drop: After the end of the adsorption step, the adsorption tower with higher pressure and the adsorption tower that is increasing pressure are connected to conduct the uniform pressure, and the gas in the adsorption tower with higher pressure is recovered. The uniform pressure has one to several times. The higher the general pressure, the more times the uniform pressure is.

Reverse discharge: A step carried out after the uniform pressure drop. The residual gas in the adsorption tower is discharged directly. In this process, a large number of adsorbed gases will be desorbed.

Vacuum: After reverse discharge, there will be a small amount of adsorbed gas that has not been desorbed, and the adsorbed gas will continue to be completely desorbed under the condition of vacuum negative pressure.

Uniform pressure rise: the step is the opposite of the uniform pressure drop, which belongs to the process of gas recovery.

Final charging: The process of pressurizing the adsorption tower with product gas after the uniform pressure rise, so that the adsorption tower can reach the adsorption pressure.

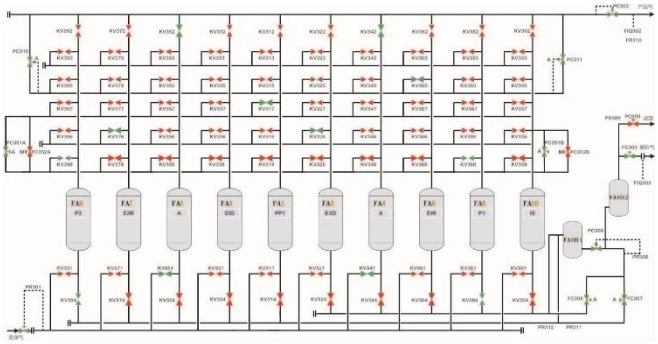

Figure 1. Process flow chart of PSA unit[53]

At present, vacuum pressure swing adsorption mainly has 2 towers, 4 towers, 6 towers or more adsorption towers, which can form 2-1-2-V, 4-1-2-V, 6-1-3-V (the number of adsorption towers, the number of adsorption towers in the adsorption state, several times of pressure balancing, vacuum desorption) and other processes[12].Picture 1 is the process flow chart of PSA unit. Because the pressure swing adsorption has a wide range of applicable pressure, low energy consumption at room temperature, high product purity, can adjust the gas purity according to the working conditions, simple process flow, can realize a variety of gas separation, high degree of automation, product purity output operation flexibility, small investment, low operating costs, simple maintenance and other advantages[13]. In recent years, PSA technology has also been used by many domestic and foreign petrochemical companies for gas refining and purification. Table 2 shows the process of PSA recovery of refinery dry gas. Domestic scholars have conducted a lot of research on pressure swing adsorption technology. Lu et al. [14] used the two-step pressure swing adsorption technology PSA-1 and PSA-2 to recover the tail gas of vinyl chloride rectification, in which the operation mode of PSA-1 was 5-1-3V and the operation mode of PSA-2 was 4-1-2V, effectively recovering hydrogen.Zhu et al. [15] used the two-step pressure swing adsorption technology PSA-1 and PSA-2 to purify CO2 from cement kiln flue gas, in which the operation mode of PSA-1 was 7-1-4V and the operation mode of PSA-2 was 6-1-3V, effectively improving the concentration of CO2. Zhang et al. [16] used pressure swing adsorption technology 4-1-2V to purify hydrogen generated in PVC production.

Table 2 Process of pressure swing adsorption recovery of refinery dry gas[52]

company name	feed gas	Gas of target	Pressure swing adsorption process
Sinopec Beijing Yanshan Branch	Ethylene, propylene, butene, hydrogen, methane, nitrogen, oxygen	ethylene	Segment 10 towers, 10-4-1; Second stage, six towers, 6-2-1
Shanghai Petrochemical	Hydrogen, ethylene (FCC dry gas)	ethylene	Fixed bed pressure swing adsorption
Yanshan Petrochemical	Catalytic cracking dry gas	ethylene	10 Towers, 10-4-2
Lanzhou Petrochemical	Catalytic cracking dry gas	ethylene	10 Towers, 10-4-2
Xinjiang Xinfeng Co. LTD	Catalytic cracking dry gas	ethylene	Using the polarity of ethylene double bond, a new type of pressure swing adsorbent was used to separate ethylene
Xinjiang Tianye Group Co. LTD	Vinyl chloride tail gas	hydrogen	4 column adsorption, two times of uniform pressure recovery chlorine; Six column adsorption, two times of uniform pressure cracking hydrogen extraction

Pressure swing adsorption technology is more and more widely used, and the technology is more and more mature. While the technology is mature and stable, it also needs the innovation of other supporting technologies, such as the innovation of special selectivity, adsorption capacity, long life adsorbent, device intelligent control technology innovation, and valve device innovation.

III. PSA CONTROL METHOD

Industrial pressure swing adsorption devices mostly use PLC (programmable logic controller) or DCS (distributed control system) control, the use of PLC or DCS control, most of the simple timing open switch and interlocking stop control, a small part of the pressure swing device control written into some simple fixed parameters PID (proportional integral differential) algorithm. PID algorithm control strategy has the advantages of simple structure, strong reliability, clear parameter physical meaning, easy adjustment and so on [17]. PID controller is the system error, which uses proportion, integral and differential to calculate the control quantity for control [18] . PID algorithm control, actually also have PI and PD control. In the control of pressure swing adsorption device, the concentration and the adsorption time and flow conditions affecting the concentration can be formed a PID regulation. Set the concentration information of purified products, and quickly adjust the adsorption time and intake flow rate through the proportional term P, so that the product can quickly reach the set concentration. When the proportional term P quickly reaches the set target concentration, the integral term I can be adjusted to increase the floating at both ends of the target value, and the differential term D can be adjusted to reduce the error of the target concentration value after the curve is uniformly floating at both ends of the target value.

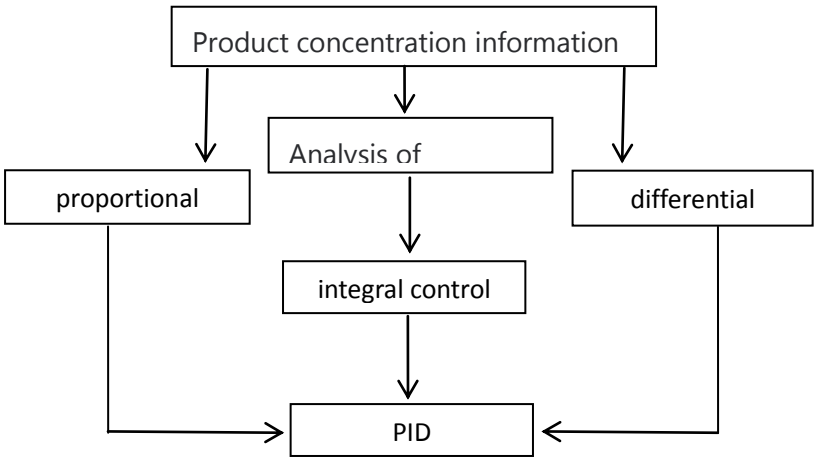

Figure 2. PID control diagram

At present, there are many researches on PSA technology, but there are few researches on the control of PSA device[19].Sen et al. [20] designed an efficient whole-system control system for the purification and processing of active drugs. The system is controlled by PID algorithm, which makes the system simple and reliable. Since the idea of model-based predictive control (MPC) was proposed in the 1970s, it has experienced several important development milestones such as dynamic matrix control algorithm (DMC), model computing algorithm (MAC) and generalized Predictive control (GPC). Due to the characteristics of multi-step prediction, rolling optimization and feedback correction, predictive control can overcome the uncertainty of process model and embody excellent control performance. It has been successfully applied in industrial process control and become an important part of advanced control[21].For the control of product purity, Bitzer et al. [22] proposed a process control scheme composed of nonlinear feedforward and linear feedback control.Khajuria et al. [23]studied the explicit - multi-parameter model predictive control of PSA system to separate methane and hydrogen, and the purity of hydrogen was stable above 99.99%.

IV. PSA PROCESS SIMULATION

According to the adsorption mechanism of the adsorbent, the parameter data of the adsorbent, the design data of the PSA adsorption tower, the establishment of mathematical model for theoretical calculation is also an important means to study PSA technology. In 2006, Dai et al. [24] summarized mass transfer model, equilibrium model, linear thrust model and pore diffusion model by studying the simulation of pressure swing adsorption process, and also summarized Henry, Langmuir, diffusion Langmuir, Freundlich-Langmuir and other adsorption isothermal equations. It lays a foundation for the simulation of pressure swing adsorption process. In recent years, more and more people at home and abroad have studied pressure swing adsorption process simulation.Liu et al. [25] verified the CO_2 adsorption and separation model of power plant flue gas by establishing a model of CO_2 adsorption process with zeolite molecular sieve. In the model, mass balance equation, gas balance equation and energy balance equation were used to simulate the PSA system of CO_2, and the two-position Langmuir equation was used to fit the CO_2 adsorption isotherm. The results of experiment and simulation show that the model is reliable.Shen et al. [26] simulated the effects of vacuum pressure and CO_2 feed flow on the separation performance and total energy consumption by establishing the VPSA model, and the results showed that there was an optimal value to achieve the best separation effect and minimum energy consumption in the VPSA process. Kim et al. [27] used the Langmuir isotherm model to measure the adsorption equilibrium of pure gas, and determined the key parameters of the adsorption equilibrium of CH_4 and CO_2.Tian et al. [28] simulated the two-stage pressure swing adsorption process model and used the key parameters of adsorbents such as activated carbon and molecular sieve to simulate the purification of CH_4 in CH_4, CO_2 and N_2. The simulation results showed that the methane could be purified to more than 99% with a recovery rate of 65%.

Aspen adsorption is software that AspenTech Company developed to simulate software for adsorption separation process[29-32].Aspen can simulate and calculate the purity and yield of the product, which has a certain guiding role for the process design of PSA and the research and development of adsorbent. Using Aspen to simulate adsorption needs to establish a process model. Usually, the model includes adsorption tower, valve, pipeline, etc. The adsorption tower model needs to set adsorbent filling height, filling inner diameter, intergranular porosity, the shape of adsorbent and other parameters. Pressure swing adsorption simulation also needs to set an isothermal adsorption model, mass and heat transfer model. Valve flow direction, etc.Xiao et al. [33] used Aspen Adsorption software to build a single-tower PSA model of He and CH_4 to simulate purification of He, and the simulated concentration of He reached more than 99.07%.Peng et al. [34] used Aspen Adsorption process simulation software to combine the adsorption parameter of corresponding adsorbent and mass transfer coefficient of each gas phase component.The two-tower adsorption and separation model of CO_2 was established. Through dynamic simulation, the concentration distribution in each packing layer of each gas phase component in each step of adsorption and regeneration was obtained, and the breakdown time of the bed could be accurately obtained, so as to make an effective evaluation of different types of adsorbents and formulate the best time sequence combination for different adsorbents.Hu et al . [35]used Aspen Adsim (Aspen Adsorption) to simulate the citrus oil fractionation process combining extraction and adsorption under supercritical carbon dioxide. The simulation results are in good agreement with the experimental results and can describe the kinetic behavior well.

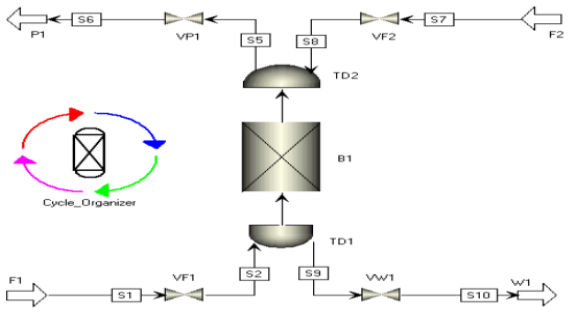

Figure 3. Aspen Activated btion dynamic simulation flow chart[34]

V. PSA ADSORPTION ADSORBENT

Adsorbents are the core of PSA gas separation. Screening of high-efficiency adsorbents with large surface area and study of adsorption and desorption effect are the key points of PSA process. Due to the high cost of pressure swing adsorption equipment in the market and the unsatisfactory separation effect of adsorption materials, it becomes the bottleneck restricting the popularization of pressure swing adsorption technology. The adsorbents of pressure swing adsorption include molecular sieve, activated carbon, silica gel and activated alumina. Their surface areas are shown in Table 3. In the actual application of several sorbents, different combinations of one or several sorbents are used for filling [37]. At present, the main adsorbents used for pressure swing adsorption are activated carbon and molecular sieve [38-42].Lyuke et al. [43] impregnated activated carbon with 34.57% $SnCl_2.2H_2O$, and then dried it at 180℃ to produce AC-SnO_2. The PSA test results showed that the impregnated activated carbon had a high adsorption capacity for CO, and the amount of CO desorbed was almost equal to the amount of CO gas adsorbed.Liang et al. [44]synthesized Rho molecular sieve by hydrothermal method, and introduced cations with different radii (H^+, Na^+, K^+ and pb^{2+}) into the Rho molecular sieve framework through ion exchange. The adsorption test showed that the pore size of Rho not only depended on the radius of the cation, but also the adjustment of the pore size by different positions of the cation was more critical.Shen et al. [45] prepared asphalt-based activated carbon through the study of activated carbon to effectively capture CO_2 in flue gas.Liu et al. [46-49] used lignite, starch and other raw materials to compare the effects of activated carbon made under different conditions on the separation of CH_4 and CO_2 after molding, carbonization and activation. Even though there are a lot of researches on adsorbents, there is still a lack of special adsorbents in industry due to the complexity of gas components actually treated[50].

Table 3 Specific surface area of common adsorbents[52]

adsorbent	activated aluminium oxide	silica gel	molecular sieve	activated carbon	High surface activated carbon
specific surface area (m2/g)	230-380	500-600	800-1000	800-1050	>3000

PROSPECT OF PSA TECHNOLOGY

Pressure swing adsorption technology is relatively perfect, but there are some shortcomings. Psa technology should overcome these shortcomings and develop in the following aspects:

(1) With the development of artificial intelligence, more intelligent algorithms should be used in the control of PSA device. Dual-pid algorithm should be used to form a PID closed-loop control with concentration value, adsorption time and intake gas. Output and adsorption time, intake gas form a PID closed-loop control. Take the weight of the two for the final parameter adjustment, mainly the concentration value, the PID value formed by the concentration of the larger weight, the output PID weight is less, through the weight to calculate the final adsorption time and intake flow rate, to ensure the purity of the product.

The combination of neural network algorithm will improve the self-regulating ability of the device to a greater extent. The neural network algorithm can find out the adsorption time and intake flow rate with the concentration of raw gas and the set concentration output according to the concentration of raw gas and the set concentration output, and then slightly adjust according to the actual concentration and output PID algorithm. This greatly increases the self-applicability of the device and the intelligence of the device.

（2）The process is becoming more and more perfect, and the device suitable for the working condition is developed, combined with other separation methods, such as pressure swing temperature adsorption, membrane separation pressure swing adsorption and so on.

（3）Gradually improve the performance of the adsorbent, with simulation combined with actual experiments to guide the development of special adsorbent, developed strong selectivity, high impurity gas adsorption capacity, easy regeneration, long life of the adsorbent, improve the overall performance of PSA technology.

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