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Intensification of Temperature Swing Adsorption

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Abstract: Temperature swing adsorption (TSA) is an energy intensive process as it needs heat for regenerating the adsorbent bed. Beds are regenerated and saturated simultaneously and are used cyclically. While one bed is undergoing adsorption, other is regenerated and vice versa. Regeneration of bed can be carried out by changing pressure or temperature. Former is called pressure swing adsorption (PSA) and later TSA. Heat is supplied for regeneration to desorb the impurities. Temperature required to regenerate the gas is high. Cycle time can be reduced and the process can be intensified by facilitating faster heat transfer in the bed. Normally the heat transfer coefficient of the adsorbent material is low which affects the cycle time. Better heat transfer can be obtained by increasing the heat transfer through packing by adding metal wires, strips or composite materials. Thus, metal fillings with good conductivity can be used in the bed. Technologies like TEPSA (Thermally Enhanced Pressure Swing Absorption) and TPSA (Temperature Pressure Swing Absorption) are developed for air pre-purification. TEPSA is a PSA process and TPSA is a TSA process. TEPSA is improved by addition of heat while in TPSA, adsorbate is removed by unheated regeneration gas. It is possible to make 20-60% power savings by moving to a TPSA cycle compared to TSA system. TPSA reduces not only cost of vessel but also pipe work involved in process. TPSA has a major advantage which is, without decreasing cycle time, thermal energy can be saved. This article explores various methods for intensification of Temperature swing adsorption. By combining advantages of pressure and temperature swing adsorption, solution to the problem of high energy demand and cycle time can be obtained. Proper selection of adsorbent material can be helpful in building economic process. Selection of adsorbent materials is based on various factors like cross-sectional area, strength, chemical inertness, etc. of the adsorbents. For TSA, thermal properties of adsorbent materials become significantly important while selecting the adsorbent.

Keywords: Adsorption, desorption, regeneration, temperature swing adsorption, adsorbate, adsorbent

1. Introduction

Process intensification is an approach of chemical process design which increases the efficiency of a process. It leads to substantially smaller, cleaner, safer, and more energy efficient process. It improves the flexibility of a process, quality of a product, speed to market and inherent safety, with a reduced environmental footprint [1]. It has become important for the industries to develop processes which are more sustainable and eco-efficient. Apart from cost reduction (which was original target of process intensification), improved intrinsic safety, reduced environmental impact and energy consumption are the benefits of the process intensification. Processing equipment like compact heat exchangers, structured packed columns, heat exchange reactor, static mixers, etc. are the examples of intensified technologies [2]. Process intensification is the method of making reasonable reductions in the size of any plant to increase efficiency. Many process intensification applications are depended on the heat transfer equipment as a unit operation with reduced size or a combined component of multifunctional unit operation [3]. Other examples of intensification include use of nanotechnology in crop input, nanomodified polymer composites, synthesis of nano-rosettes for surfactant manufacturing, multifunctional reactors, microfluidizer [4-8].

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Adsorption is one of the important unit operations used in catalyzed reactions and water purification [9-12]. For purification of gases also adsorption is used to remove or recover solute gases. For regenerating adsorbents temperature and pressure methods are employed [13-15]. Investigations on intensifications of pressure swing adsorption are reported for optimization of cycle time and exploring possibilities of different low-cost adsorbents [16,17]. Adsorbents are solids having porous structure and they have large surface area per unit mass. Adsorbents are mainly spherical and have honeycomb monolithic structure [18]. Temperature swing adsorption cycle takes several hours and pressure swing adsorption is much faster and using more adsorbent per unit time. For the capture and pressurization of CO_2 from the mixture of gases containing nitrogen (N_2) and oxygen (O_2), TSA process is considered [19]. Two half cycles i.e., adsorption and desorption half cycles are used alternately until a period state is reached. Temperature swing adsorption consists of four steps namely feed, rinse, heating and cooling [20]. Adsorption and desorption temperature directly influence the generation of entropy caused by heat transfer. Generation of entropy per kg adsorbed CO_2 captured in whole cycle. So, to improve adsorption capacity of adsorbents it is necessary to reduce entropy generation in four step TSA technology [21]. TSA is a gas separation method. There are two important factors to increase process efficiency which are acceleration of temperature swing kinetics and reduction of thermal gradients along the cross-section of reactor. To achieve this both conditions, it's necessary to overcome the intrinsic Heat transfer Limitations associated with the generation of packed bed this is for both static and in flow conditions [22].

Instead of traditional heating technique, microwave (MW) irradiation can be applied which contributes process acceleration and reduces cost of energy production. The innovative MW assisted regeneration of Zeolite bed is evaluated. The energy efficiency of regeneration step is of 75 % [23]. The MW heating does not affect Zeolite absorption properties.

Efficient gas processing (removal of unwanted components) is essential to enable the technical use of natural gas. Various components such as water, Sulphur compounds, carbon dioxide, nitrogen and heavy hydrocarbons must be removed [24]. To remove some of these components, TSA is the commonly used method. To control CO_2 amount in the environment, there are three ways of capturing CO_2 from the power plants. They are pre-combustion, post-combustion and oxygen-combustion. The various methods used for post-combustion carbon capture are membrane, adsorption, adsorption, cryogenic process [25]. Cyclic operation of adsorption CO_2 capture could be divided into two steps, i.e., adsorption and desorption which forms different thermal cycles. PSA and TSA are the most commonly used adsorption cycles (Fig.1). 7-8 % of global CO_2 emission is generated from 4 billion tons of cement produced annually [26]. Utilization of TSA system for the Claus tail gas clean-up is a viable option as limited pressure (1-2 bar) is available in a Claus unit [27].

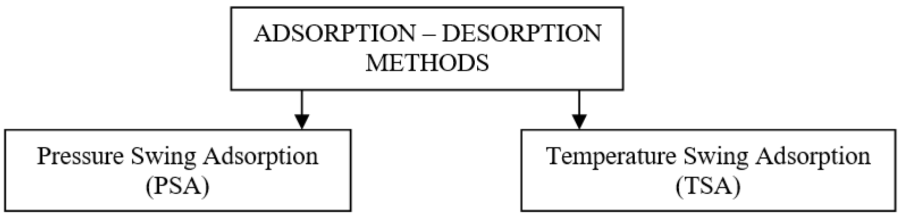

Fig. 1 - Adsorption desorption methods

2. Temperature Swing Adsorption (TSA)

TSA is energy intensive as it needs heat to the regenerating gas. Impurities are removed from the air stream by adsorbing them over the surface of packed bed. Cycle time is extended and the heat pulse is allowed to exit the adsorbent bed during feed period. Heat is supplied for regeneration to desorb impurities. Production of heat pulse which moves through the bed counter-current to the feed direction. More than one unwanted gas is removed. Temperature required to regenerate the gas is high. Technologies like TEPSA and TPSA are developed for air prepurification. TEPSA is a PSA process that is improved by addition of heat while TPSA is a TSA process in which adsorbate is removed by unheated regeneration gas. By supplying smaller amount of energy, heat pulse becomes blown and the part of adsorbent remains undegenerated. By continuing it, adsorbate can be removed. Within few cycles, it achieves steady state. It is possible to make heater power savings of the order of 20-60% by moving to a TPSA cycle comparing with TSA. TPSA system reduces not only vessel cost but also accompanying pipe work [28].

Major benefit of TPSA is that without decreasing cycle time, thermal energy can be saved. Temperature swing adsorption is the Process which is used for gas purification. There are various ideas about new indirect TSA process for the removal of volatile organic compounds (VOC) from air streams. The main objective is to reduce desorption time, radial temperature gradients, inert mass heat losses [29]. Adsorption and regeneration are two main steps of TSA cycle. Adsorption depends on the species to be absorbed and on feed concentration. By designing rapid adsorbers, adsorbents can be reduced. Effective gas purification can be obtained and high pollutant concentration is encountered during regeneration phase. Adsorption step is much longer than regeneration step in a purification process. Because of this, several beds are used.

The steps involved in TSA are:

1. Adsorption takes place at low temperature.
2. Removing impurities means desorption of impurities we have to heat the bed so that temperature increases.
3. Then cooling of bed to again start process of adsorption.

For heating and cooling of bed there are two methods. First is direct method where inert gas or purge is used and second is indirect method where heating jackets, coils or microwave is used.

TSA technique has an ability to separate impurities which can form strong bonding with adsorbent called as chemisorption (adsorption involves chemical reaction between adsorbent and surface area) which is better than PSA. Few degrees change in temperature can also cause change in the loading amount which gives effective separation [30]. TSA technique is usually used for the purification process instead of bulk separation. Process of adsorption is operated in a cyclic way. TSA method is based on the regeneration of adsorbent bed by increasing the temperature. PSA method is based on the regeneration of adsorbent bed by reducing the total pressure. Mostly adsorbates with high volatility are favored by PSA, while TSA method is perfect for absorbates which have moderate volatility [31].

Adsorption desorption cycle is composed of two half cycles as shown in Fig. 2. In the cold half cycle, gas mixture which is to be separated is passed through the adsorbent bed where the waste gas is get adsorbed and then we get pure gas. Meanwhile, in the hot half cycle, once the adsorption capacity (saturated) of bed is exhausted, then the hot half cycle starts regeneration of adsorbent bed [32]. That is how the regeneration cycle continues, which accomplishes the separation of mixture. Important fact about adsorption is that mass transfer and heat transfer characteristics are interrelated. Adsorption occurs at low temperature and desorption occurs at high temperature. Productivity of this process depends on the change in temperature of adsorbent bed, heating and cooling of adsorbent bed. These processes should be as quick as possible so that we reach the temperature set point for regeneration. Steam desorption is fast and efficient, but it takes long time for drying and cooling stages. It takes several hours to pass large quantities of gas through bed. For that, two different techniques can be employed. First is increasing overall heat transfer in particle bed by using thermal conductivity promoters for example compacted natural graphite (its thermal conductivity is high). Second is use of thermoelectric elements (Peltier effect) for the acceleration of temperature changes [32].

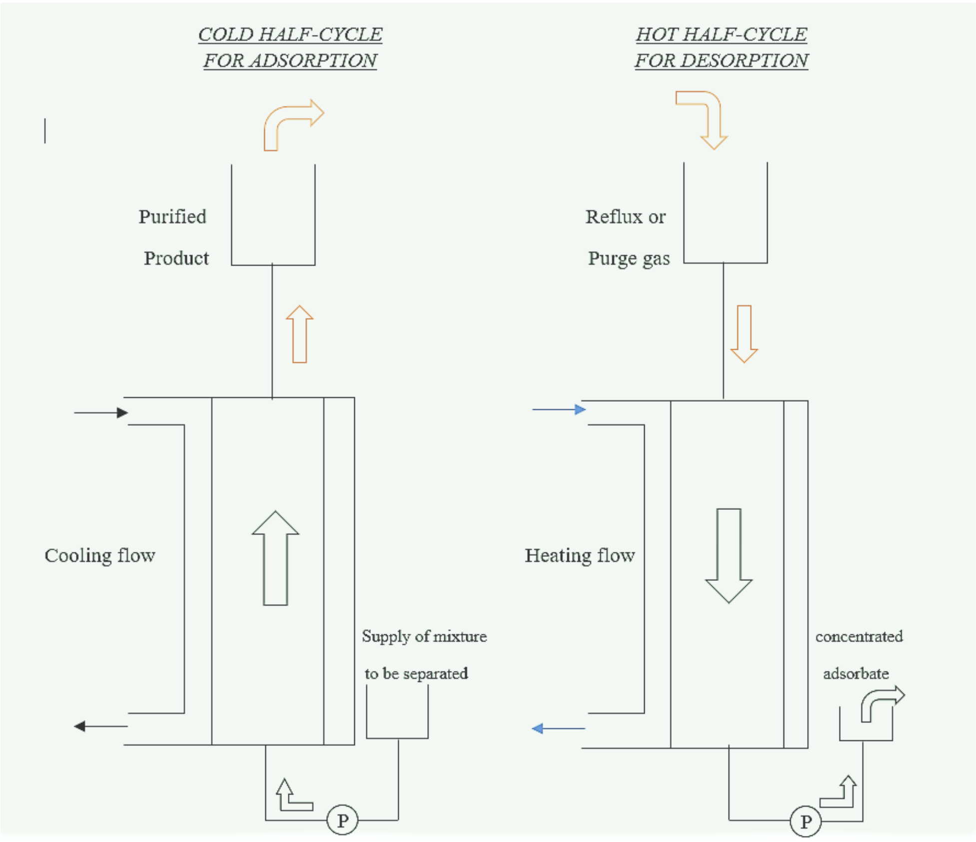

Fig. 2 - Adsorption/desorption cycle in an adsorbent bed by varying the temperature

CO_2 working capacity i.e., difference between the quantity of CO_2 adsorbed under adsorption and desorption conditions is required to know real quantity of CO_2 captured in complete adsorption or desorption cycle. To increase the cycle performance (CO_2 purity and output), heating and purging strategies are applied [33]. For four step pressure and temperature swing adsorption (PTSA), equilibrium model is developed and verified by 2-D dynamic model. By using equilibrium model, parameters like purity, recovery and energy efficiency are studied. Wang et al. found that at the ambient temperature, number of optimal Pareto solutions is the largest [34]. Also, they found a new method for improvement of optimization accuracy and error was reduced to 0.1 % [34]. Increasing CO_2 production leads to global warming which can be reduced by Pressure swing absorption. Skarstrom cycle involve pressurization, absorption, blowdown and purge. Adsorption and purge steps include mass transfer zone formation. Leavitt (1992) [35] and Hirose (1991) [36] and formed a novel two-bed PSA cycle i.e., duplex PSA. Mass transfer zone extends over the entire bed and driving force is higher in duplex PSA which leads to process intensification. Studies on duplex cycle revealed the limited purity of products (94 mol%) and low outputs [37].

Kumar et al. (2006) have presented a modified duplex cycle which as purity of 99 mol% at high output [38]. Novel TSA cycle design achieved 96 % CO_2 purity and 90 % output of CO_2 capture [39]. This cycle is equivalent with amine-based absorption process. Skarstrom and duplex cycle resembles but mechanism is different. Influence of heat integration, cycle scheduling and external drying step can be evaluated [39]. For the removal and recovery of ethane from nitrogen steam, experiments including a rapid heat exchanger for indirect cooling and heating are performed [40]. They developed a simplified one- dimensional non-isothermal non-equilibrium model of the process [40]. Heat transfer coefficient between the heating or cooling medium and the gas stream can be calculated. From the experimental and numerical results, effect of some parameters like regeneration temperature and initial feed composition are determined. For the rapid screening of adsorbents to recover strongly adsorbed species from a gas mixture, the model is developed [41]. It is verified by comparing results from a detailed adsorption column model. For extensive parametric screening of zeolite 13X and other 75 real and theoretical adsorbents for post combustion CO_2 used [41]. This gives information about dependence of performance indicators on operating conditions. To evaluate adsorbent performance, indicators like purity, recovery, specific thermal energy demand and cyclic working capacity are used. The new shortcut model considers a four-step TSA cycle. Condition of bed at the start and end of feed step are known, the fresh feed amount of material get recovered during feed step and then further calculated by material balance [42]. The steps are:

1. Heating step: In this step one end of column is open and bed is heated indirectly to the desorption temperature.
2. Cooling step: This step is similar to heating step; both ends of adsorbent bed get cooled to adsorption temperature. There are no material flows out or inside the bed, pressure of get reduces.
3. Pressurization step: Mixture of feed gas enters from one end and restored to the adsorption temperature.
4. Feed step: Both ends of adsorbent bed are opened and feed enters into it. Weakly adsorbed material is recovered until breakthrough occurs.

For removal of pollutants & waste and any toxic gases effluents we can use a much-known method that is adsorption. Regeneration of bed is very important step in cycle of adsorption and desorption. Adsorbent which has low thermal conductivity takes time to regenerate and hence it increases the cycle time (low heat transfer) [43]. Main three step of Temperature swing adsorption are:

1. Adsorption Process: Waste gas gets an adsorbed through a selective adsorbent material.
2. Desorption Process: Heat the adsorption bed, & then adsorbed gas is desorbed and flows out of adsorption bed.
3. Cooling Process: The adsorption bed is cooled inlet & outlet of adsorption is closed [43].

Simulation of Adsorption, heating and depressurization steps are enabled on bench scale fixed-bed reactor [44]. These steps are based on PSA and TSA. Temperature of desorption have major impact on the rate of desorption. Cyclic process consists of two steps- adsorption and regeneration (desorption). Adsorption is a cyclic process so the cost of regeneration plays very important role in the workability of process. Desorption occurs; i) due to reducing the total pressure of process which is called PSA, ii) due to increasing temperature which is called TSA. At high adsorption isotherm is helpful for desorption, and iii) At the low pressure and high temperature combination [44]. TSA process reduces the use of direct electricity for the gas separation. State of adsorbent can be control by changing by partial pressure or the temperature of the surrounding air & it cause adsorption or desorption [45]. Established adsorbent bed made by filling adsorbent pellets in rigid cylinder which have porous regions between particles of adsorbent for easier gas flow. Pressure swing adsorption use commonly over temperature swing adsorption due their ease of operation; meanwhile temperature swing adsorption have not been used in industries due to energy requirement in heating the adsorbent (low thermal conductivity) to desorb waste material and regeneration [46]. Performance of adsorbent is based on pressure and temperature. Gas composition is a main aspect of PSA and TSA. For the appropriate selection of adsorbent, we have to study isotherm shape, working capacity, heat of adsorption, adsorption kinetics [47]. For desorption of organic pollutants from activated carbon, hot gas or steam is used. Coupling of TSA method with heat pump has two advantages which are (1) Efficiency of adsorbent step increases and (2) prior to steam desorption during regeneration cycle there is warming up of activated carbon adsorbent.

While using heat pump temperature up to 45°C is required during desorption [48]. Heat pump alone is not sufficient to process regeneration, so it is combined with the steam desorption and so it gives new perspective for reduction and energy consumption. At low temperature adsorbent adsorbs more gas than at high temperature. In this method waste gas is passed through bed particles at low temperature then adsorbent adsorb waste gas and that's how stream gas gets cleaned [49]. System gets switched to another bed when one bed gets saturated, while the saturated bed gets heated and it releases the adsorbed gas and regeneration of adsorbent bed takes place and that's how the cycle continues.

Limitation of temperature swing adsorption is the poor conductivity of materials [50]. Cycle time required for TSA is longer. Few selective literatures review on intensification of TSA is tabulated in Table 1.

Table 1 - Literature Study on Intensification of TSA

Ref.	Title	Findings
18	Advances in Pressure Swing Adsorption for Gas Separation	CO_2 can be captured using PSA
21	Energy dissipation evaluation of temperature swing adsorption (TSA) cycle based on thermodynamic entropy insights	4-step TSA process integrated with the IHR method
22	Heat Intensification of TSA Process with Packed Metal Foams: An Experimental and Modeling Study Applied to Post-Combustion CO_2 Capture	Key factors to improve efficiency of adsorption process
25	Comparative analysis on temperature swing adsorption cycle for carbon capture by using internal heat/mass recovery	4-step TSA cycles for CO_2 capture by using internal heat recovery, internal mass recovery & internal heat & mass recovery is evaluated based on carbon pump theory
29	Influence of the Presence of CO_2 in the Feed of an Indirect Heating TSA Process for VOC Removal	Main features of an indirect heating TSA process for the removal of CO_2-ethane mixtures from air
30	Model- based Design, Operation & Control of Pressure Swing Adsorption Systems	Challenges occurred in temperature swing adsorption
32	Intensification of Adsorption Process in Porous Media	Thermoelectric element was found to be efficient temperature controller
43	Various Intensification methods for adsorption-a review on studies & investigations	Steps involved in temperature swing adsorption
44	Cyclic operation of a fixed-bed pressure and temperature swing process for CO_2 capture: Experimental and statistical analysis	Impact of temperature of adsorbents on the rate of desorption
50	An Insight into Investigations on Intensification of Adsorbent Beds	Intensification TSA and PSA, selection of adsorbent & contacting pattern.

3. Adsorbent Materials

Selection of proper adsorbent is very important for economy of the process. Many times, small modifications in the processes lead to valuable savings. Absorbent materials such as Mg-MOF-74, Zeolite 13X (Fig. 4), UTSA-16 and activated carbon (AC) (Fig. 3) can be used for various applications [51]. Zeolites are crystalline in nature. In case of application of zeolites, removal of moisture from the adsorbent is very crucial for efficient operation [52]. Use of low cost and unconventional adsorbents can lead to significant savings. It serves twin purposes of increasing adsorption economy and reducing solid waste. Many agricultural materials such as rice husk, groundnut shells, tamarind waste, leaf litters etc. are being explored for their application as an adsorbent [53,54]. Adsorbent material should have large cross-sectional area and strength. Chemical inertness is one more important aspect of the adsorbents. Now if we consider the adsorbent for temperature swing processes, thermal properties become significantly important aspects of adsorbent properties. These adsorbents find applications for removal of gases like hydrogen sulfide, organic vapours from flue gases. Nano-materials are also being explored for their use in adsorption application as those materials have high surface area [55]. Other materials used for adsorbent are silica gel (SG), activated alumina (AA), molecular sieve (MS), etc. They are widely in petrochemical, refining and gas industries [56]. The adsorbent materials can be in the form of powder, granules or pellets. Different adsorbents used in the various processes are listed in the Table 2.

Fig. 3 – Activated carbon

Fig. 4 – Zeolite 13X

Table 2 - Adsorbents used in different processes

Sr. No.	Process	Adsorbents
1.	Natural Gas Dehydration	SG, AA, MS
2.	Olefin Dehydration	Modified MS, i.e., 3A MS
3.	Mercury removal from Natural Gas	Sulfur impregnated AC & Silver impregnated MS
4.	Air Separation	13X & Carbon molecular sieve (CMS)
5.	Pre-purification of air	AA, MS, 13X, 4A & 5A types
6.	Natural gas desulfurization	Different types of MSs
7.	Hydrogen separation & purification	AC, SG & MS (5A type)
8.	NGL Sweetening & dehydration	MSs
9.	Ethanol dehydration	MSs
10.	Heavy hydrocarbon removal from amine streams	AC

4. Conclusion

In this paper we have discussed temperature swing adsorption and pressure swing adsorption with main focus on temperature swing adsorption. Temperature swing adsorption is a cheaper method than Pressure swing adsorption. Temperature swing adsorption is a cyclic process in which two adsorbent beds are involved. Also, the steps involved in TSA method, its advantages, regeneration cycle, adsorbents and its properties for accurate adsorbent selection are studied. The primary aim is to reduce the time required for regeneration cycle (adsorption- desorption cycle). Cycle time can be reduced and the process can be intensified by facilitating faster heat transfer in the bed. Normally the heat transfer coefficient of the adsorbent material is low which affects the cycle time. Better heat transfer can be obtained by increasing the heat transfer through packing by adding metal wires, strips or composite materials. Thus, metal fillings with good conductivity can be used in the bed. Technologies like TEPSA (Thermally Enhanced Pressure Swing Absorption) and TPSA (Temperature Pressure Swing Absorption) are developed for air pre-purification. TEPSA is a PSA process that is improved by addition of heat while TPSA is a TSA process in which adsorbate is removed by unheated regeneration gas. Various adsorbents that can be used in different processes are discussed. Selection of particular adsorbent for any process depends on various factors like cost, temperature of regeneration gas, feed composition, etc.

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