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Hydrogen PSA process product purity control method and controller

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Abstract It is well known in the industry that a primary means for controlling the pressure swing adsorption (PSA) process product gas purity is the adjustment of PSA feed time or adsorption time. If the product impurity is too high, the feed time is shortened and if the impurity level is below the target the feed time is increased. Conventionally, the plant operator monitors the product purity and manually adjusts the feed time. Several control methodologies such as classical feedback and feedforward systems were suggested to automate this task with limited success. A novel control methodology based on the measurement of impurity fronts within the adsorber bed was developed by the Praxair Adsorption R&D team. The response of the concentration measurements inside the adsorber vessel to the process upsets and changes in feed time is more rapid than in the product stream. Consequently, closed loop control performance can be made much more effective and the operating impurity set points for product gas can be more aggressive resulting in longer PSA feed times, higher bed utilization and thus higher hydrogen recovery. The control methodology will be discussed in greater detail along with the advantages it has to offer such as improved process performance, disturbance rejection capability and improved process robustness. The control methodology will be illustrated using the hydrogen PSA process as an example.

Keywords Hydrogen · PSA · Product purity control

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1 Introduction

Adsorption processes today are well established in the chemical industry and they are often the method of choice for difficult gas separations. Adsorption processes can be distinguished by the process cycle employed, for example, pressure swing adsorption (PSA), temperature swing adsorption (TSA) or vacuum swing adsorption (VSA). Multiple adsorber vessels can be used to increase the production capacity and separation efficiency usually expressed as the main component yield or recovery. The impurities are removed from the feed stream utilizing one or more adsorbents contained within the vessel to produce the product stream. In case of hydrogen PSA process, the adsorber bed typically contains an initial layer of alumina to adsorb water vapor, a layer of activated carbon to adsorb carbon dioxide and hydrocarbons and a final layer of zeolite adsorbent to adsorb carbon monoxide and nitrogen.

The time an adsorber bed spends in the feed or adsorption step is referred to as the feed time. As the feed time is increased the impurity front(s) are moving towards the product end of the bed and eventually the impurity breakthrough is observed. It is well known that a primary means for controlling the product purity is to adjust the feed time and at the same time, controlling the product impurity level to the maximum allowable level results in the highest product recovery.

The PSA process feed is often subject to variations in flow, composition and temperature that can result in process upsets and in the product not meeting the specifications. The situation can be even more complex for PSA systems processing multiple feed streams. Conventionally, the operator monitors the product impurity concentration and manually adjusts the feed time. This task can be automated by using a feedback control system. In both cases, however, there will be a significant lag between the occurrence of the upset and the product impurity increase. Once observed, the operator must make a guess of the proper adjustment to the feed time, and will have to wait a considerable amount of time to know if the change was appropriate. In order to prevent the impurity breakthrough, a conservative impurity set point is typically selected resulting in shorter feed time and lower product recovery.

Feedforward control systems have also been used in which the feed flow rate and the feed composition are measured and a model is utilized to gauge the effect of such changes on the product impurity; however, there are still several disadvantages: (1) simple model has to be used due to practical process control system limitations and (2) models used are not perfect and thus again conservative impurity set point will be used. Either method or a combined feedforward/feedback strategy is widely practiced by the industry today especially on the large hydrogen PSA systems to minimize the effect of front-end disturbances.

Miller and Gray (1987) suggested a control system in which a characteristic of the effluent (e.g. gas concentration) from an adsorber bed undergoing depressurization step is sensed, compared to a target and then a corrective action is taken e.g. change in the feed time. After the target has been reached and the product gas impurity level is not at desired value, new target is calculated using the characteristic and product errors. However; such effluent concentration will not experience instantaneous change upon upset and therefore again a conservative target will be selected.

A new method and control system for controlling the product purity of adsorption process was developed by the Adsorption R&D Team of Praxair Inc. (Belanger et al. 2010). The impurity concentration is measured within the adsorber bed itself where the change in the impurity concentration due to an upset will be more rapid compared to measurement of a product stream or an effluent stream. As a result, the selected product impurity target can be more aggressive resulting in longer feed time and higher product recovery.

2 Theoretical section

In a hydrogen production facility, the steam methane reformer (SMR) contacts a hydrocarbon feedstock, for example, natural gas with superheated steam over a catalyst to produce raw syngas. The hydrogen content of the reformer gas is further increased by using a water gas shift reactor to produce the PSA feed stream. The whole SMR process including the steam production and syngas cooling train is represented by the reforming and shift block in Fig. 1. The PSA feed stream predominantly contains hydrogen and impurities that can represent as much as 28 % by volume. The PSA feed stream typically contains around 0.5 % water vapor, 16 % carbon dioxide, 5 % methane, 3 % carbon monoxide and 0.7 % nitrogen.

The PSA feed stream is introduced into the PSA unit to produce the PSA product stream i.e. nearly pure hydrogen gas that meets the desired impurity specification and the PSA waste stream. The waste gas is routed back to the SMR unit burners where it is used as fuel to drive the endothermic reforming reaction. Alternatively, the waste gas pressure can be increased using a compressor and then the gas is introduced into the fuel network of the refining facility.

The PSA product gas impurity specification can vary depending on the customer(s) requirements or on the hydrogen pipeline specification. The most commonly measured impurities by the hydrogen plants are carbon monoxide, nitrogen and total hydrocarbon content (THC). For illustration purposes, the impurity specification for the hydrogen gas is 1 ppm for carbon monoxide, 100 ppm for nitrogen and 1 ppm for THC.

The PSA unit consists of adsorption beds, piping and valves that follow the sequence of a PSA cycle according to the PSA process controller executing the logic to run the defined process cycle. Adsorbent beds are employed in an out of phase cycle so that while some bed(s) are adsorbing the impurities other beds can be regenerated. The PSA cycle can utilize steps such as adsorption step(s), co-current depressurization step(s), counter-current depressurization step(s), purging step(s), re-pressurization step(s) and other steps(s) and the steps don’t have to be executed in this order. The number, duration and the sequence of steps define the process cycle that is usually determined during the PSA process development and design phase and is often protected as intellectual property.

The product purity controller controls the purity of the hydrogen product gas. As already mentioned earlier, the new product purity controller takes advantage of the measurement of the impurity fronts within the adsorber bed. The first probe, CO_2 Probe, is located within the carbon layer. The PSA unit is controlled such that carbon dioxide never breaks through the carbon layer into the zeolite. Carbon dioxide would be very strongly adsorbed by the zeolite and it would be very difficult to regenerate. For that reason the CO_2 levels are measured in the activated carbon. The second probe, the CO Probe, is used to monitor the concentration of carbon monoxide in the zeolite layer. In addition to the two measurements inside the adsorber bed the product purity controller utilizes CO and N_2 impurity measurements in the hydrogen product stream as well. The control of carbon monoxide product concentration is accomplished through the control of the CO probe

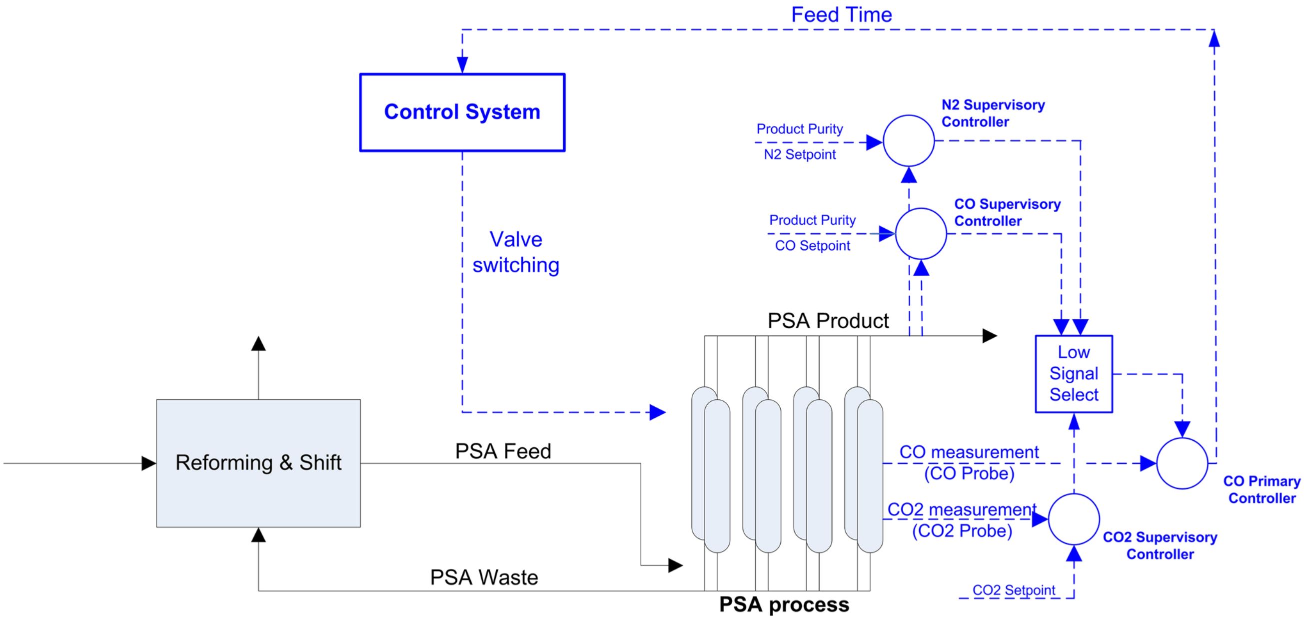

Fig. 1 Schematic view of hydrogen production process and hydrogen product purity control system

composition sensed in the zeolite layer which in turn defines the feed time.

The CO_2 and CO concentrations are sensed in the adsorber bed and compared against targets because it has been found that changes in such concentrations will appear much sooner than the downstream product concentration changes. Hence, the controller tuning can be more aggressive to allow longer feed time and maximize the production.

The product purity controller is provided with a primary controller and three separate supervisory controllers according to the controller configuration shown in Fig. 1. These controllers are generally PID controllers, but they may be implemented using alternative control methodologies such as MPC ("Model Predictive Control"). The three supervisory level controllers are the CO product controller, N_2 product controller and CO_2 Probe controller. Each of these controllers is used to calculate a targeted concentration for the CO Probe controller such that the CO and N_2 concentrations in the product will be maintained below the product specification levels and such that CO_2 will not break through into the zeolite layer. The targeted carbon monoxide concentration in the zeolite layer is then compared to the actual carbon monoxide concentration within the zeolite layer in the primary controller and the controller output is the new value of the feed time.

The CO Product supervisory controller is using the information from CO product analyzer as the process value (controller input). The CO concentration measured in the real-time is converted to the logarithmic base 10 form and the running average is calculated over the duration of the whole cycle (cycle average). It is actually this value of cycle averaged $\log_{10}$ CO concentration that is used as the controller process value. At the end of the cycle, the process value is compared to the setpoint [if CO specification used is 1 ppm the setpoint will be $\log_{10}(1) = 0$] and the controller output is generated i.e. the new value of the targeted CO concentration within the zeolite layer (CO Probe Setpoint). In case the CO concentration in the product is below the level set forth in the product specification the new CO Probe setpoint will be increased and vice versa.

The N_2 Product supervisory controller is working on the same principle as the CO Product controller. The real-time nitrogen concentration is used to calculate the $\log_{10}$ cycle average; the value is then compared to the setpoint [if N_2 specification used is 100 ppm the setpoint will be $\log_{10}(100) = 2$] and the new CO Probe setpoint is generated. In case the N_2 concentration in the product is below the level set forth in the product specification the new CO Probe setpoint will be increased and vice versa.

The CO_2 Probe supervisory controller performs calculations in the same manner as the previous two controllers. The process value is the $\log_{10}$ cycle average of the carbon dioxide concentration within the carbon layer. The controller setpoint is carefully chosen by the process engineer such that the carbon dioxide and hydrocarbons' concentration fronts will be contained within the carbon layer. The controller output is the new targeted CO Probe controller setpoint. In case the CO_2 concentration in the carbon layer is above the value set forth by the process engineer the new CO Probe setpoint will be decreased and vice versa.

The three new targeted CO Probe setpoint values generated by the three supervisory controllers are introduced into the low signal select block as shown in Fig. 1. The logic finds the minimum value that is subsequently introduced into the primary controller. This way, the most conservative value is used as the new primary controller set point. It is normally the case that only one of the three supervisory controllers will be actively controlling at any point in time. The other two controllers will only take action if there is a danger of violating one of the other two desired composition limits. The CO primary controller or CO Probe controller functions in a similar fashion as the supervisory controllers except that its output is the feed time. The controller process value is calculated as $\log_{10}$ cycle average of the real-time carbon monoxide levels within the zeolite layer and the controller setpoint is determined by the low signal select block as described above.

The product purity controller executes at a controller frequency that is equal to the total cycle time of the PSA unit. During this time, the process gas analyzer real-time data in parts per million (ppm) is converted to a logarithmic base 10 value and the running cycle average calculation is being performed at the same time. At the end of the cycle, a signal is generated by the timer to end the cycle average calculations, each of the controllers executes and the new feed time value is introduced to the PSA sequence controller. For example, for the cycle time of 600 s and the data acquisition frequency of 2 s there will be 300 data points in the cycle average calculation to generate new values for all process variables to be compared with their respective setpoints. All figures in the Results and discussion section show experimental data measured with Siemens Ultramat 6 Gas Analyzer®.

3 Results and discussion

The new product purity controller was compared with an industry standard i.e. simple feedback controller based on the carbon monoxide concentration in the product stream alone. Both controllers were tuned consistently with a closed loop log modulus of +2 dB (this is equivalent to a damping coefficient of 0.5). A disturbance was initiated by increasing the concentration of carbon monoxide in the PSA feed stream from 4.5 to 5.5 %. The feed stream mixture consisted of 74 % hydrogen, 16 % carbon dioxide, 5 % methane, 4.5 % carbon monoxide and 0.5 % nitrogen before the disturbance.

In case of a simple feedback controller, the carbon monoxide breakthrough occurred after 2 h and persisted for 28 h as shown in Fig. 2. The controller action is significantly delayed since the feed time value starts to decrease only after the impurity breakthrough is observed. The $\log_{10}$ CO impurity peaked at 0.58 which corresponds to cycle average value of 3.8 ppm. One way to resolve this problem is to use more aggressive tuning parameters; however, this approach yields very little improvement due to the PSA process non-linear behavior and associated dead time and it results in low closed loop control robustness. The other approach is to use a very conservative set point, which as stated earlier results in reduced product recovery.

The effect of the disturbance on the PSA process controlled by the product purity controller depicted in

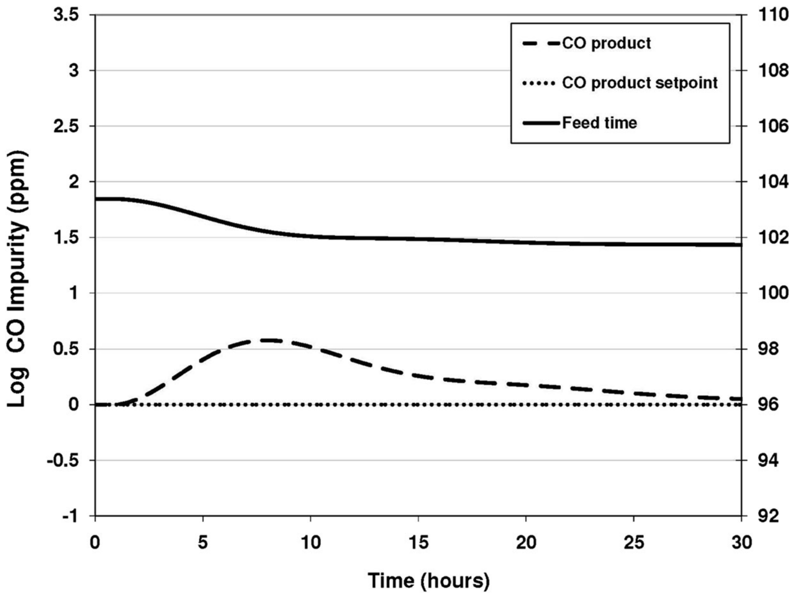

Fig. 2 Simple feedback product purity controller response to the PSA feed disturbance

Fig. 1 is shown in Fig. 3. The CO probe trend slowly increases as the disturbance progresses through the adsorber vessel and at around 5 h the CO probe trend crosses the CO probe setpoint. As a result, the primary controller decreases the PSA feed time rather quickly while the CO product concentration is still maintained below the specification limit. A small carbon monoxide impurity breakthrough in the hydrogen product is observed at around 11 h. At this point, the CO product supervisory controller starts the cut the CO probe set point value to compensate for the impurity excursion. The peak impurity is recorded at 0.04 which corresponds to CO concentration 1.097 ppm. In the meantime, the zeolite layer has been regenerated because of the shorter feed time and as the product impurity trend starts to decrease again the supervisory controller will increase the CO Probe set point which will in turn lead to increase in the feed time value and eventually to higher hydrogen recovery.

The action of the product purity controller with respect to the change in the hydrogen product carbon monoxide impurity limit from 7.5 to 1 ppm is presented in Fig. 4. The CO product supervisory controller setpoint is changed at time zero from 0.88 to 0 corresponding to $\log_{10}(7.5)$ and $\log_{10}(1)$ respectively. The large error value in the supervisory controller results in aggressive change in the CO probe set point and subsequently the large error in the primary controller calculated as the difference between the

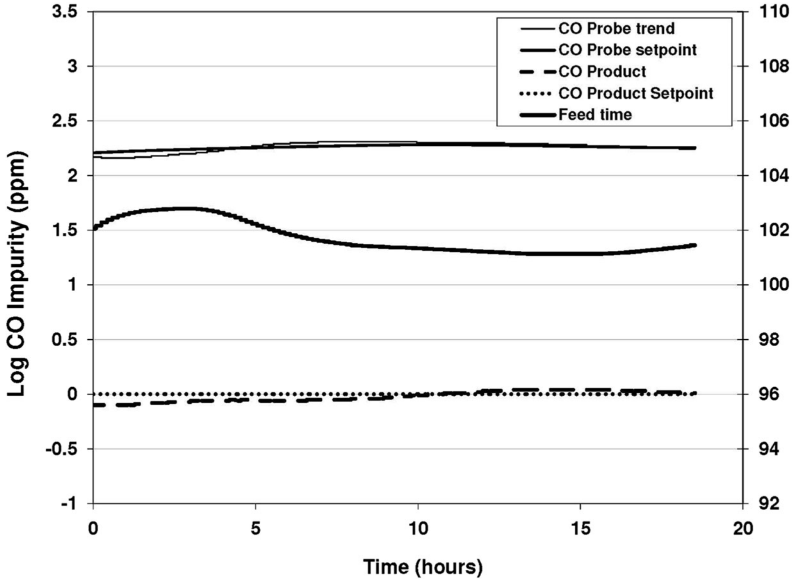

Fig. 3 Response of product purity controller to the PSA feed disturbance

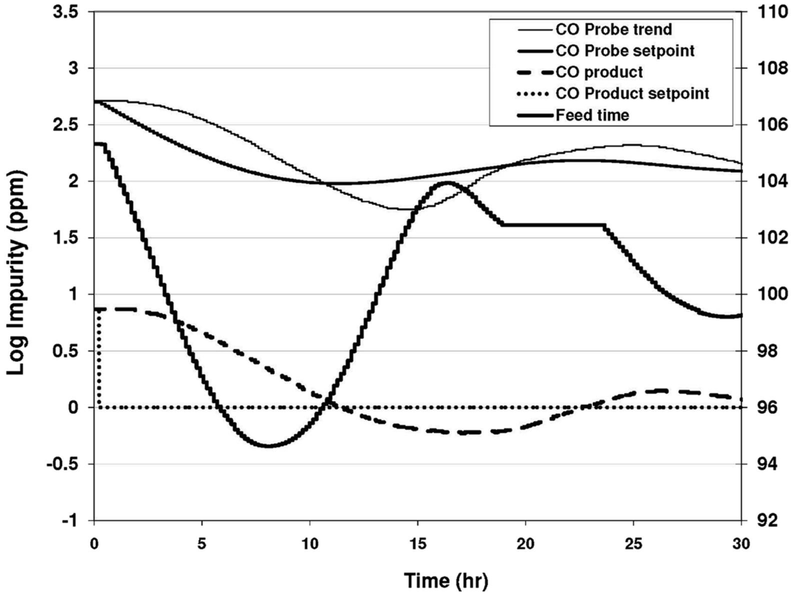

Fig. 4 Response of the novel product purity controller to the change in the hydrogen product impurity specification from 7.5 to 1 ppm

CO Probe set point and CO Probe trend value is responsible for the significant decrease in the feed time. The lower PSA feed time causes the adsorbents to regenerate i.e. decrease the amount of impurities loaded on to the adsorbent surface. After about 10 h the CO impurity level drops below the limit and the product purity starts to track the target with a little deviation. The feed time is increased again since the product impurity level is below the specification to allow for higher hydrogen product recovery.

The examples above clearly demonstrate the advantages of the novel purity controller such as the robustness of the cascade control scheme to large changes in set point. The magnitude of the impurity breakthrough is greatly decreased and at the same time the production time of off-spec gas is much shorter. In order to meet the product gas specification at all times the novel purity controller needs smaller safety margin thus allowing for more aggressive impurity set point closer to the limit. As a result, the PSA efficiency will be increased resulting in higher productivity. Depending on the plant size and the type of PSA process cycle employed the improvement can be as high as several percentage points in the PSA recovery.

4 Conclusions

The novel method and controller for controlling the purity of product gas produced by an adsorption unit was developed by the Adsorption R&D team of Praxair Inc. The controller utilizes the measurement of the impurity concentration fronts within the adsorber vessel to reduce the dead time typically associated with the purity control of adsorption processes. The advantages of the controller were demonstrated using experimental data. The product purity controller allows for a more aggressive impurity target resulting in higher feed times and higher productivity as well.

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