

# Evaluation of condensate formation by parametric analysis of the TSA process for moisture removal from natural gas

Antônio Eurico Belo Torres<sup>a</sup>, Rafaelle Gomes Santiago<sup>a</sup>, Darley Carrijo de Melo<sup>b</sup>, Célio Loureiro Cavalcante Jr<sup>a</sup>, Moises Bastos-Neto<sup>a\*</sup>

<sup>a</sup> Laboratório de Pesquisa em Adsorção e Captura de CO<sub>2</sub> – Federal University of Ceará, Fortaleza, Brazil <sup>b</sup> PETROBRAS/CENPES, Rio de Janeiro, RJ, Brazil <sup>\*</sup>email: mbn@ufc.br

#### Abstract

Moisture removal from natural gas in offshore installations is typically performed using fixed-bed adsorption units with Temperature Swing Adsorption (TSA) processes. These processes utilize adsorbents with high capacity, strong affinity, and high selectivity for water. Over time, it has been observed that the capacity to adsorb water reduces. A primary cause of adsorbent degradation is attributed to water condensation during the regeneration step. This study presents a mathematical model describing the dynamic behavior of a TSA unit and applies it to the removal of moisture from a methane stream saturated with water, using zeolite 13X as the adsorbent. The study aims to evaluate the impact of two parameters on condensate formation: the heating temperature profile and the feed temperature during the adsorption step on condensate formation and product purity. The results indicated that, under the conditions examined, product purity values significantly lower than 1 PPM can be achieved. Additionally, the study demonstrated that it is possible to prevent water condensation during the heating step by adjusting these two operational parameters of the unit.

Natural gas; dehydration; TSA; moisture condensation

# 1. Introduction

The removal of moisture from natural gas in offshore installations is conventionally carried out in fixed-bed adsorption units using Temperature Swing Adsorption cyclic processes. These processes use adsorbents with high capacity, strong affinity, and high selectivity for water [1, 2]. Over time, it has been observed that the adsorbent in these units loses its capacity to adsorb  $H_2O$  due to operational and material-intrinsic causes [3].

During regeneration step, when the material is saturated with water, a hot and dry gas stream is fed at the bed bottom, causing desorption of water. If the regeneration stage begins with dry gas at maximum temperature, a significant amount of water is desorbed at the bed's lower section. As the gas stream ascends, it carries the desorbed vapor to the upper part of the bed, which remains saturated with water and has not yet been heated by the gas, potentially causing condensation. Upon heating, this condensed water may leach the clay binder used to form the adsorbent pellets [4-6].

Operating a TSA process requires several decisions that impact the performance. Due to the vast number of operational possibilities, it is practically impossible and very costly to experimentally test all these possibilities. An acceptable way to achieve this objective is to use simulators that can evaluate the performance of a unit under different operational conditions. This work aims to study the influence of the feed temperature during the adsorption stage and the temperature profile of the gas used during the heating stage on the purity of the produced gas and formation of condensate during the the regeneration stage. To achieve this objective, a mathematical model was developed to simulate the dynamic behavior of a TSA unit with a commercial 13X zeolite fed by a methane stream saturated with water.



# 2. Methods

The Temperature Swing Adsorption (TSA) process uses a set of fixed beds connected by valves operated so that the gas streams flowing through the beds achieve specific objectives. The valves are synchronized to allow the beds to go through multiple cyclic stages, with each cycle always including an adsorption step and a desorption step. The adsorption step occurs at the lowest temperature of the cycle, while the desorption step occurs at the highest temperature.

In the basic configuration of TSA processes, there are no significant pressure variations, and typically, the steps of adsorption, heating, and cooling are utilized. A TSA unit used for water (moisture) removal commonly consists of two or more fixed beds that adsorb water during the adsorption stage to produce dry gas, which is then regenerated using a fraction of the heated gas stream produced. A schematic diagram of a simplified TSA unit used for natural gas dehydration is shown in Figure 1.



Fig. 1 – Schematic diagram of a simplified TSA unit used for natural gas dehydration

In such a configuration, the moisture from the incoming gas stream is removed by a fixed bed that has been regenerated. After some time, this bed becomes saturated with water, and the adsorbed water must be desorbed in the desorption step using a heated fraction of the produced gas. In a typical unit, the desorption gas used is a sidestream from the product gas (usually around 10%). The high temperature used in the desorption stage causes the water to be desorbed from the adsorbent.

The moist gas stream from the bed being regenerated is cooled, and the condensed water is then removed by draining the condensates in a separation vessel. The water-saturated gas stream from the separation vessel is sent back to the feed after compression to compensate for the pressure loss that occurred in the beds and separation vessel.

# 3. Model and parameter estimation

The model implemented to describe the process is the same as presented by Rios *et al.* [7], with the modification of the gas phase equation of state described here by the law of corresponding states, considering the following assumptions below:

- The flow is axially dispersed for concentration and temperature;
- Local thermal equilibrium between the gas and adsorbent particles;
- The mass transfer rate of each component from the fluid phase to the particle is represented by the linear driving force (LDF) model;
- The gas phase behaves like a mixture of real gases with compressibility factor calculated by the corresponding states equation;
- There are no concentration, temperature, and pressure gradients in the radial direction.

The boundary conditions for each step of the TSA cycle in its basic configuration, with three steps.

Adsorption	
$V_{0,P} \times \left[\frac{P_{0,P}}{R \times T_{0,P}}\right] \times \frac{1}{A} \times y_{e,i} = u(0) \times C_{g,i}(0) - \varepsilon \times D_{ax}(0) \times C_{gT}(0)$ $\times \frac{\partial y_i}{\partial a}(0)$	$\frac{\partial C_{g,i}}{\partial z}(L) = 0$
$V_{0,P} \times \left[\frac{P_{0,P}}{R \times T_{0,P}}\right] \times \frac{1}{A} \times y_{e,i} = u(0) \times C_{g,i}(0) - \varepsilon \times D_{ax}(0) \times C_{gT}(0)$ $\times \frac{\partial y_i}{\partial z}(0)$	$\frac{\partial T_g}{\partial z}(L)$
$\begin{bmatrix} V_{0,P} \times \left[ \frac{P_{0,P}}{R \times T_{0,P}} \right] \times \frac{1}{A} \times y_{e,i} = u(0) \times C_{g,i}(0) - \varepsilon \times D_{ax}(0) \times C_{gT}(0) \\ \times \frac{\partial y_i}{\partial z}(0) \\ V_{0,P} \times \left[ \frac{P_{0,P} \times 1E5}{P(0)} \times \frac{T_g(0)}{T_{0,P}} \right] / A = u(0) \end{bmatrix}$	$P(L) = P_0$
$V_{0,P} \times \left[P_{0,P}/\left(R \times T_{0,P}\right)\right] \times \frac{1}{A} \times C_{p,g,mix}(0) \times T_{0}$ = $u(0) \times C_{g,T}(0) \times C_{p,g,mix}(0) \times T_{g}(0)$ $- \lambda(0) \times \frac{\partial T_{g}}{\partial z}(0)$	$\frac{\partial T_w}{\partial z}(L) = 0$
$\frac{\partial T_w}{\partial z}(0) = 0$	
Heating	
$ \begin{array}{c} -Rec \times V_{0,P} \times \left[ \frac{P_{0,P}}{R \times T_{0,P}} \right] \times \frac{1}{A} \times y_{e,i,aq} \\ &= u(L) \times C_{g,i}(L) - \varepsilon \times D_{ax}(L) \times C_{gT}(L) \\ &\qquad \qquad $	$\frac{\partial C_{g,i}}{\partial z}(0) = 0$
$-Rec \times V_{0,P} \times \left[\frac{P_{0,P} \times 1E5}{P(L)} \times \frac{T_g(L)}{T_{0,P}}\right] / A = u(L)$	$\frac{\partial T_g}{\partial z}(0)$



$ \begin{aligned} & -0.1 \times V_{0,P} \times \left[ P_{0,P} / \left( R \times T_{0,P} \right) \right] \times \frac{1}{A} \times C_{p,g,mix}(L) \times T_{0,aq} \\ & = u(L) \times C_{g,T}(L) \times C_{p,g,mix}(l) \times T_g(l) - \lambda(L) \\ & \times \frac{\partial T_g}{\partial z}(L) \end{aligned} $	$P(0) = P_0$	
$\frac{\partial T_w}{\partial z}(L) = 0$	$=0^{\frac{\partial T_w}{\partial z}(0)}$	
Cooling		
$-Rec \times V_{0,P} \times \left[\frac{P_{0,P}}{R \times T_{0,P}}\right] \times \frac{1}{A} \times y_{e,i,res}$ $= u(L) \times C_{g,i}(L) - \varepsilon \times D_{ax}(L) \times C_{gT}(L)$ $\times \frac{\partial y_i}{\partial z}(L)$	$= 0^{\frac{\partial C_{g,i}}{\partial z}(0)}$	
$Rec \times V_{0,P} \times \left[\frac{P_{0,P} \times 1E5}{P(L)} \times \frac{T_g(L)}{T_{0,P}}\right] / A = u(L)$	$\frac{\partial T_g}{\partial z}(0)$	
$0,1 \times V_{0,P} \times \left[P_{0,P}/(R \times T_{0,P})\right] \times \frac{1}{A} \times C_{p,g,mix}(L) \times T_{0,res}$ $= u(L) \times C_{g,T}(L) \times C_{p,g,mix}(L) \times T_g(L)$ $-\lambda(L) \times \frac{\partial T_g}{\partial Z}(L)$	$P(0)=P_0$	
$\frac{\partial T_w}{\partial z}(L) = 0$	$\frac{\partial T_w}{\partial z}(0) = 0$	

The values used for the simulation were:

- Measured: parameters of the equilibrium isotherms, particle density, bed and particle porosity, heat of adsorption, mass transfer coefficient, and particle size;
- Estimated by correlations from the literature: gases and adsorbent specific heats, axial mass and heat dispersion coefficients;
- Provided data: column dimensions, feed flow rate, feed composition, cooling gas temperature, heating gas temperature, packing density, step times.

#### 4. Simulations

In the simulations, the feed gas is saturated at temperature  $T_0$ . Before entering the column in the adsorption step, the gas undergoes a small temperature increase (0, 3, 6, or 9 °C). The final temperature of the heating gas is 260 °C. The heating gas constitutes a fraction (12%) of the feed flow rate in the adsorption step and is heated according to three different schedules. To evaluate the effect of this small temperature increase, simulations were conducted (each lasting 20 cycles) for three heating schedules. Figure 2 shows the three heating schedules used.

Therefore, 12 simulations were performed, labeled as  $P_i$  ( $T_0 + T'$ ). Here, the index *i* represents the temperature schedule, and T' can be 0, 3, 6, or 9.



Fig 2 – Used schedules for the heating gas temperature.

#### 5. Results and discussions

Fig. 3 shows the concentration profiles of H<sub>2</sub>O for the last three cycles (18, 19, and 20) for simulation P1 ( $T_0$  + 9). All the profiles obtained for all simulations show similar behavior. The results indicate that the cyclic steady-state regime was achieved in all simulations.



Fig 3 – Concentration profiles of H<sub>2</sub>O for cycles 18, 19 e 20 for simulations P1 ( $T_0$  + 0), ( $T_0$  + 3), ( $T_0$  + 6) and ( $T_0$  + 9).

Fig. 4 shows the evolution of the relative humidity (RH) at the column outlet for all 20 cycles for simulation P1 ( $T_0 + 0$ ), ( $T_0 + 3$ ), ( $T_0 + 6$ ) and ( $T_0 + 9$ ). Fig. 5 highlights the last two cycles. The results indicate that for a given heating temperature schedule, there is an increasing trend in condensate formation as T' decreases. Similar behavior was observed for the other two temperature schedules.



Fig. 6 shows the evolution of the product stream purity for simulations P1  $(T_0 + 0)$ ,  $(T_0 + 3)$ ,  $(T_0 + 6)$  and  $(T_0 + 9)$ . Similar behavior can be observed for the other two schedules, showing that this slight increase in temperature does not significantly impact the product purity.



Fig. 4 – Relative humidity at the column outlet for simulations P1  $(T_0 + 0)$ ,  $(T_0 + 3)$ ,  $(T_0 + 6)$  and  $(T_0 + 9)$ .



Fig. 5 – Relative humidity at the column outlet of the last two cycles for simulations P1  $(T_0 + 0)$ ,  $(T_0 + 3)$ ,  $(T_0 + 6)$  and  $(T_0 + 9)$ .



Fig. 6 – Product purity throughout the cycles for simulations P1  $(T_0 + 0)$ ,  $(T_0 + 3)$ ,  $(T_0 + 6)$  and  $(T_0 + 9)$ .

Fig. 7 shows the evolution of the relative humidity at the column outlet in the last cycle for schedules P1, P2, and P3 ( $T_0 + 0$ ), revealing a trend of condensate formation for all three schedules. The results indicate that there is an increasing trend of condensate formation as the initial heating rate increases.



Fig. 7 – Relative humidity at the column outlet of the last cycle for simulations P1, P2 and P3  $(T_0 + 0)$ .

#### 6. Conclusions

- A slight increase in the feed temperature during the adsorption step can prevent condensate formation during the heating step without impacting product purity.
- The gas heating schedule affects condensate formation.

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## References

- 1. H.A.A. Farag, et al., *Alexandria Eng. J.* 2011. 50(4): p. 431–439.
- 2. H.Y. Huang, et al., *Ind. Eng. Chem. Res.* 2003. 42(12): p. 2427–2433.
- D.C.S. de Azevedo, et al., Research Report 2022, Universidade Federal do Ceará - DEQ.
- 4. D.M. Ruthven, *Adsorption and Adsorption Processes* ed. J.W. Sons. 1984.
- R. Herold and S. Mokhatab, Optimal design and operation of molecular sieves for gas dehydration -Part 2. Gas Processing & LNG, 2017.
- R. Herold and S. Mokhatab, Optimal design and operation of molecular sieves for gas dehydration Part 1. Gas Processing & LNG, 2017.
- 7. R.B. Rios, et al., *Adsorption* 2014. 20(8): p. 945-957.