# RIA 43

## XLIII Reunião Ibérica de Adsorção

Book of Abstracts

**Porto, Portugal**

**1-4 September, 2024**

# SPONSORS

## Institutional Support

## Gold

## SILVER

## Bronze

## Coffee Break Sponsor

*This event was financially supported by national funds through FCT/MCTES (PIDDAC) reference ALiCE, LA/P/0045/2020 (DOI: 10.54499/LA/P/0045/2020)

# O39: Kinetic separation of green hydrogen from natural gas grids by using vacuum pressure swing adsorption

L. F. A. S. Zafanelli [1,2,3,4], A. Henrique [1,2], E. Aly [1,2], A. E. Rodrigues [3,4], G. Mouchaham [5], J. A. C. Silva [1,2]

[1]*Centro de Investigação de Montanha (CIMO), Instituto Politécnico de Bragança, Campus Santa Apolónia, 5300-253 Bragança, Portugal*
[2]*Laboratório Associado para a Sustentabilidade e Tecnologia em Regiões de Montanha (SusTEC), Instituto Politécnico de Bragança, Campus de Santa Apolónia, 5300-253 Bragança, Portugal*
[3]*LSRE-LCM – Laboratory of Separation and Reaction Engineering - Laboratory of Catalysis and Materials, Faculty of Engineering, University of Porto, Rua Dr. Roberto Frias, 4200-465 Porto, Portugal*
[4]*ALiCE – Associate Laboratory in Chemical Engineering, Faculty of Engineering, University of Porto, Rua Dr. Roberto Frias, 4200-465 Porto, Portugal*
*zafanelli@ipb.pt*
[5]*Institut des Matériaux Poreux de Paris, Ecole Normale Supérieure de Paris, ESPCI Paris, CNRS, PSL University, 75005 Paris, France*
*adriano_henrique@ipb.pt; ezzeldin@ipb.pt; arodrig@fe.up.pt; georges.mouchaham@ens.psl.eu; jsilva@ipb.pt*

1. **Introduction –** Transitioning to renewable energy sources is crucial to mitigating climate change. In this scenario, green hydrogen (GH) is considered a promising energy carrier due to its high calorific value, versatility in applications, clean combustion, and potential for local generation in abundance [1]. As interest in GH grows, developing its distribution chain becomes crucial in facilitating its widespread use. The co- transporting GH into existing natural gas grids (NGG) emerges as a viable alternative, eliminating the need for significant infrastructure investments [2]. However, upon blending GH into the NGG, it becomes essential to de-blend and purify it to a high degree to enable, for instance, fuel cell applications (H2 > 99,97%). One problem concerning the separation and purification of GH from NGG relates to the H2 feed concentration (< 20%), which differs significantly from conventional H2 purification processes (> 70%). Moreover, the high CH4 concentration and its relatively weak adsorption affinity on commonly used adsorbents further complicate achieving high-purity H2 and high recovery rates through conventional approaches. In this work, we report a novel conceptual vacuum pressure swing adsorption (VPSA) process to separate H2 from CH4 by exploiting the kinetic selectivity of H2 over CH4 on CMS-3K-172, as shown in Figure 1.

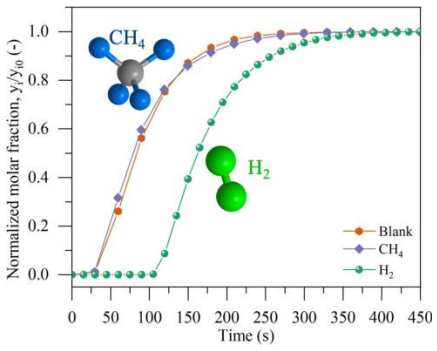


**Figure 1.** Experimental single breakthrough curves of $H_2$ and $CH_4$ on CMS-3K-172 compared to the blank experiment.

**2. Experimental -** To develop the conceptual VPSA cycle, a series of single and multicomponent breakthrough curves for $H_2$ and $CH_4$ were performed on CMS-3K-172 between 195 - 273 K and pressures up to 18 bar. These experiments were performed in a cryogenic fixed-bed adsorption unit designed to work at lower temperatures (until 77 K) [3].

**3. Results and Discussion** - Figure 1 shows the single breakthrough curves of $H_2$ and $CH_4$ on CMS-3K-172 compared to a blank experiment performed at 195 K and 12 bar, where a kinetic separation can be seen. $CH_4$ has a limited diffusion into the CMS-3K-172 structure, which results in its early breakthrough at the same time as the blank experiment. Conversely, $H_2$ is adsorbed on CMS-3K-172 as its breakthrough curve presents a delay compared to the blank experiment. The conceptual VPSA developed consists of 1 bed with 5 steps, namely (1) pressurization with feed, (2) feed, (3) $H_2$ purge, (4) cocurrent depressurization (COD), and (5) countercurrent vacuum blowdown. The $H_2$ purity and recovery were evaluated by changing the process variables such as step time, intermediate-to-high pressure ratio, purge-to-feed ratio, and VPSA type configuration. Figure 2 shows three simulated VPSA types: type I with the five steps mentioned above

type II without the COD step, and type III without the $H_2$ purge step. The $H_2$ purity-recover trade-off for the three VPSA types can be seen in Figure 3. From a feed of 20% $H_2$, the VPSA type II allows obtaining an $H_2$ purity of up to 68% with a recovery of up to 92%, and the best trade-off between purity and recovery was 83% and 85%, respectively.

**4. Conclusions -** This work shows for the first time that an adsorbent, which preferentially adsorbs $H_2$ and blocks $CH_4$ from entering its pores, can enrich $H_2$ from a low feed concentration. Moreover, this work provides insights for developing new materials with the same CMS characteristics but with higher $H_2$ capacity, which could be beneficial to improve the VPSA process. In conclusion, the developed VPSA process increases the $H_2$ molar fraction from 20% to 60 - 70% with a high recovery. We are currently working on a second stage to be incorporated into the VPSA process to purify $H_2$ for fuel cell applications (>99.97%).

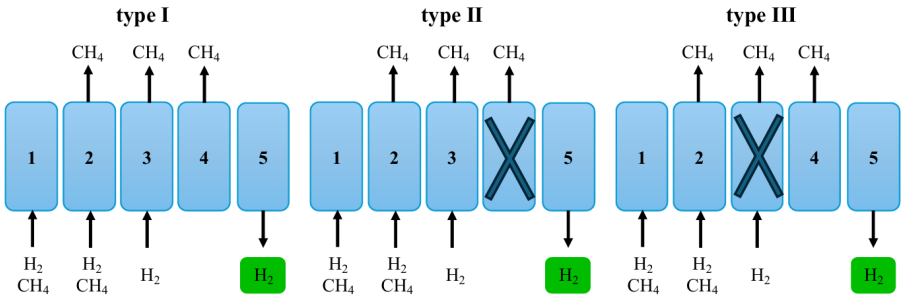


**Figure 2.** VPSA type configurations.

**5. Acknowledgments** The authors are grateful to the Foundation for Science and Technology (FCT, Portugal) for financial support: (1) under project PTDC/EQU-EPQ/0467/2020 (DOI: 10.54499/PTDC/EQU-EPQ/0467/2020), (2) through the national funds FCT/MCTES (PIDDAC) to CIMO (UIDB/00690/2020 and UIDP/00690/2020), and SusTEC (LA/P/0007/2020), (3) by the national funds through FCT/MCTES (PIDDAC): LSRE-LCM, UIDB/50020/2020 (DOI: 10.54499/UIDB/50020/2020) and UIDP/50020/2020 (DOI: 10.54499/UIDP/50020/2020); and ALiCE, LA/P/0045/2020 (DOI: 10.54499/LA/P/0045/2020).
Additionally, we thank national funding by FCT, Foundation for Science and Technology, through the individual research grant 2020.07925.BD (DOI: 10.54499/2020.07925.BD ) of Lucas F. A. S. Zafanelli.
Moreover, the authors are grateful to Osaka Co. for kindly providing the CMS-3K-172 studied in this work.

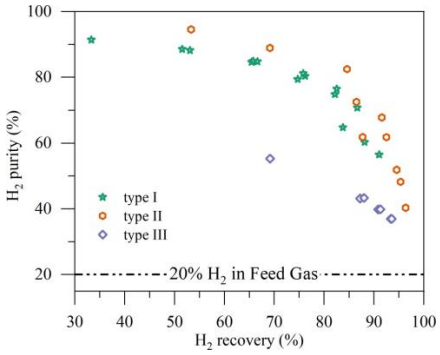


**Figure 3.** Trade-off between H2 purity and recovery for different VPSA types and sets of process variables.

**6. References**

[1] IRENA, International Renewable Energy Agency, 2022.
[2] L. Dehdari, I. Burgers, P. Xiao, K. G. Li, R. Singh, & P. A. Webley, *Sep. Purif. Technol.*, 282, (2022) 120094.
[3] L. F. A. S. Zafanelli, E. Aly, A. E. Rodrigues, & J. A. C. Silva, *Sep. Purif. Technol.*, *307*, (2023) 122824.
L. F. A. S. Zafanelli, E. Aly, A. Henrique, A. E. Rodrigues, G. Mouchaham, & J. A. C. Silva, *Ind. Eng. Chem. Res.*, **63**(14), (2024) 6333-634