

Intensified Methanol Synthesis in a Fluidized Bed Reactor with Continuous Sorbent Feeding (SEFBR+CSF)

S. Jiménez Lacruz, R. González-Pizarro, J. Lasobras, S. Renda, J. Soler, M. Menéndez, J. Herguido

Afiliación: *Catalysis and Reactor Engineering Group (CREG)*

Instituto de Investigación en Ingeniería de Aragón (I3A)
Universidad de Zaragoza, Mariano Esquillor s/n, 50018, Zaragoza, Spain.
Tel. +34-976762707, e-mail: 873645@unizar.es

Abstract

The main aim is to intensify green methanol production through a modified reactor configuration based on a fluidized bed reactor with continuous sorbent feeding. In this system, a selective sorbent is continuously introduced to remove reaction products in situ, thereby shifting the reaction equilibrium toward methanol formation and enhancing overall conversion.

Introduction

Currently, mitigating CO₂ emissions and reducing dependence on fossil fuels are major technological challenges. One promising strategy involves the capture of CO₂ from hard-to-abate industrial processes and its subsequent conversion with green hydrogen, produced via water electrolysis using surplus renewable energy. This approach is part of the Power-to-X concept [1], leading to the production of synthetic fuels through so-called Power-to-Fuels processes.

Methanol is of particular interest due to its relevance both as a fuel and as an important intermediate for numerous industrial applications.

Experimental system

An innovative reactor configuration is proposed, consisting of a fluidized bed reactor in which the catalyst remains in the lower section of the bed while the sorbent is continuously introduced through a lower lateral inlet. After passing through the catalytic bed, the sorbent exits through an upper lateral outlet located near the top of the reactor.

Compared with conventional reactor designs, this configuration enables continuous water removal from the reaction zone while maintaining a constant catalyst inventory inside the reactor.

Since only the zeolite sorbent is withdrawn, an external regeneration unit can be incorporated without exposing the catalyst to regeneration conditions. Furthermore, the presence of water negatively affects catalyst activity; therefore, its continuous removal helps mitigate catalyst deactivation. By continuously circulating and regenerating the sorbent, steady-state operation may be achieved while maintaining stable catalytic performance [2]. See Figure 1.

Segregation studies

Under operating conditions of 20 bar and 230 °C, the commercial catalyst BASF K3-110 exhibits suitable catalytic activity and structural stability throughout the reaction time. Zeolite 3A was selected as the sorbent due to its favorable adsorption selectivity, which prevents the adsorption of feed components and methanol, unlike other sorbents.

Pressure and temperature must be carefully controlled. Higher pressure enhances both reaction kinetics and adsorption capacity; however, it also requires a higher inlet gas flow rate, thereby reducing residence time. Increasing temperature improves kinetics but adversely affects thermodynamic equilibrium and may accelerate catalyst deactivation.

The continuous introduction of solids requires an appropriate segregation between catalyst and sorbent particles to avoid catalyst entrainment during sorbent withdrawal.

To enable continuous solids circulation without shutdowns for reactor unloading, a fluidized bed configuration was selected. The particle size ratio must ensure complete segregation between catalyst and sorbent under continuous operating conditions, corresponding to a near-zero mixing index. This configuration enables selective removal of water from the gaseous stream without significantly

disturbing reactor operation, thus improving methanol yield and allowing steady-state operation.

Fluid-dynamic experiments

Experiments were conducted to determine the minimum fluidization velocity of each solid. Subsequently, different reduced gas velocities and sorbent feeding rates were evaluated.

The experiments were performed within a reduced velocity range of $U_r = 1.0$ to $U_r = 1.5$. The optimal reduced velocity was found to be $U_r = 1.04$ defined as:

$$U_r = \frac{U}{U_{mf\text{catalizador}}}$$

Regarding the sorbent feeding rate, values between 2 and 5 g/min were tested, with 2.5 g/min identified as the optimal condition.

Reaction experiments

The intensified process is expected to improve methanol production in both the gaseous stream and the condensed liquid products. The results obtained after 90 min TOS are summarized in Table 1. A $H_2:CO_2$ molar ratio of 3:1 was employed, with the corresponding flow rate adjusted to maintain a reduced velocity of 1.04 under operating conditions.

Table 1 Experimental methanol production

Temperature	230 °C	250°C	270°C
Coventional reactor (g)	2.56	5.86	2.03
STY ($mg_{MeOH} \cdot h^{-1} \cdot g_{cat}^{-1}$)	82.58	167.43	57.94
Intensified zeolite reactor (g)	7.89	6.17	5.53
STY ($mg_{MeOH} \cdot h^{-1} \cdot g_{cat}^{-1}$)	221.21	199.03	176.52

The highest process intensification was achieved at 230 °C, where methanol production was nearly three times higher than that obtained under conventional operation at identical reaction conditions.

Conclusions

An optimal operating window exists in which adequate segregation between catalyst and zeolite particles can be achieved while maintaining a constant catalyst inventory within the reactor bed.

Although larger catalyst particles improve segregation and reduce mixing, they also increase the required fluidization velocity, excessively reducing residence time. Conversely, smaller zeolite particles

improve segregation behavior but complicate solids feeding into the reactor.

The optimal operating temperature is limited by both catalyst stability and the thermodynamic constraints associated with the main methanol synthesis reaction and competing side reactions.

Future work

Future studies will evaluate the effect of the $H_2:CO_2$ ratio on methanol production. Additionally, the impact of reducing operating pressure will be investigated, since lower pressure would reduce the required inlet gas flow rate and increase residence time, despite negatively affecting reaction kinetics.

Different reactor geometries will also be explored to facilitate solids feeding and reduce gas flow requirements.

Finally, further reaction studies will focus on determining the optimal sorbent circulation rate according to the amount of water generated and adsorbed within the system.

References

- [1]. DAHIRU, A.R., A. VUOKILA y M. HUUHTANEN. Recent development in Power-to-X: Part I - A review on techno-economic analysis. *Journal of Energy Storage*. 2022, vol. 56, art. 105861. Available from: <https://doi.org/10.1016/j.est.2022.105861>
- [2]. GONZÁLEZ-PIZARRO, R., J. LASOBRAS, J. SOLER, J. HERGUIDO y M. MENÉNDEZ. Proof of concept for a sorption-enhanced reactor with continuous sorbent feeding (CSF): application to green methanol production. *Chemical Engineering Journal*. 2025, vol. 517, art. 164562. Disponible en: <https://doi.org/10.1016/j.cej.2025.164562>

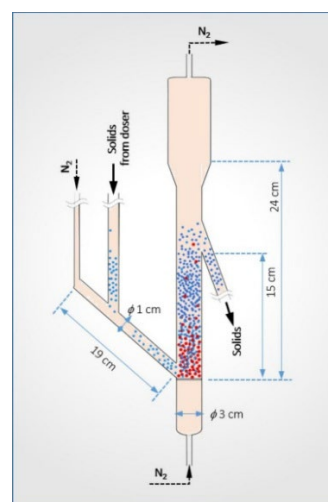


Figure 1. Experimental set up of the SEFBR+CSF system.