

## Experimental validation of pressure swing regeneration for faster cycling in sorption enhanced dimethyl ether synthesis

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### Experimental Materials

Experimental validation of sorption enhanced DME synthesis was performed using (a homogeneous mixture of) commercially available catalyst and adsorbent: copper-zinc oxide-alumina (CZA) catalyst,  $\gamma$ - $\text{Al}_2\text{O}_3$  (assay>98%, Riogen NJ, USA), obtained as 3 mm pellets, and molecular sieve type 3A, purchased as 1.6 mm pellets (UOP Molecular Sieves, Advanced Specialty Gas Equipment, USA).

### Methods

A combination of commercially obtained CZA catalyst,  $\gamma$ - $\text{Al}_2\text{O}_3$  catalyst and zeolite 3A adsorbent was used for the experimental demonstration of direct DME synthesis from  $\text{CO}/\text{CO}_2/\text{H}_2$ -mixtures. The experimental runs were conducted on a bench-scale high-pressure reactor setup (Figure 1), allowing tests up to 2 litres of sample, typically consisting of a 1:4 ratio (weight basis) catalyst to sorbent. The ratio between catalyst and sorbent was not further optimized in this work. Adsorption was performed with different (stoichiometric) feed gas compositions, using 68.6-72.7 vol.% of hydrogen, 0-9.1 vol.% of carbon monoxide, 17.1-23.6 vol.% of carbon dioxide and inert argon, nitrogen or methane, at 25 bar(a) and a temperature range of 250-300 °C. The inert balance was used in order to keep the overall pressure stable, considering the shrinking reaction and adsorption of water. Regeneration was done by depressurisation to 1-3 bar(a) for PSA regeneration, switching to dry, inert gas, and eventual heating to 400 °C for TSA regeneration. Finally, either the inert purge gas or the reactive feed gas is used for repressurisation. A schematic overview of the PSA cycle is shown in Figure 2. Transient gas analysis was performed by micro-GC (measuring methane, CO,  $\text{CO}_2$ , nitrogen, argon, methanol and DME) and mass spectrometry measuring hydrogen ( $m/z=2$ ), methane ( $m/z=15$ ), water ( $m/z=18$ ), carbon monoxide/nitrogen ( $m/z=28$ ), methanol ( $m/z=31$ ), carbon dioxide ( $m/z=44$ ) and DME ( $m/z=45$ ).

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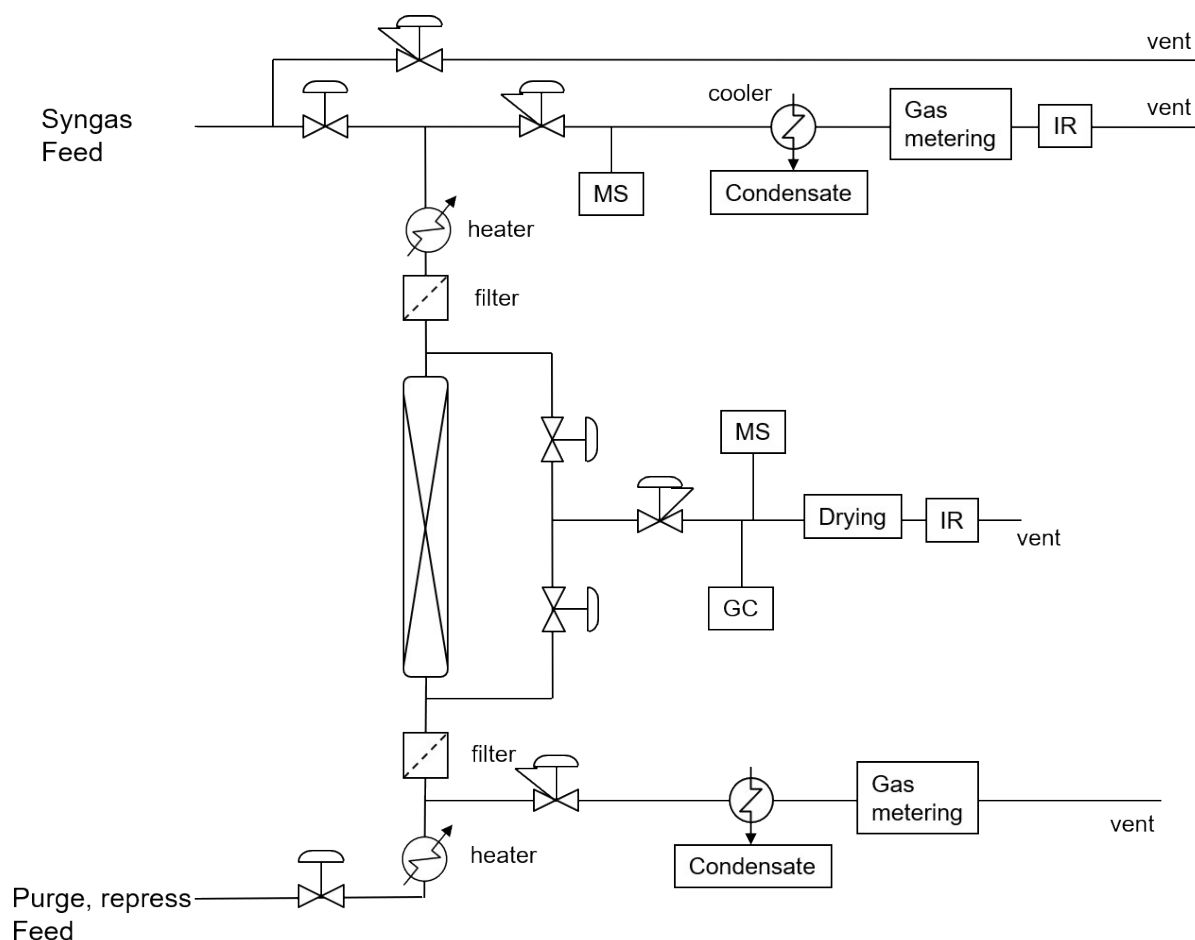


Figure 1. Schematic of the bench-scale reactor 'SEWGS 1'.

## Data interpretation

In order to facilitate data interpretation, several key metrics have been defined to be able to quantify SEDMES performance. The most important metric, the carbon selectivity  $S(i)$ , used here is defined as follows,

$$S(i) = \frac{ny(C_nH_mO_p)}{\sum_i n_i y(i)} \quad (1)$$

The carbon selectivities were calculated as molar concentration-based selectivity for each of the carbon containing species,  $y(i)$ . For example, the selectivity towards DME can be calculated as

$$S(DME) = \frac{2y(DME)}{y(CO) + y(CO_2) + 2y(DME) + y(MeOH) + y(CH_4)} \quad (2)$$

Time integration (in the interval  $t: 0-t_{CO_2}$ , where  $t_{CO_2}$  is the (interpolated) point in time where the  $CO_2$  outlet concentration reaches a level of 5 vol.%) of the streams gives an overall yield and selectivity for the cyclic (steady state) performance of the SEDMES process.

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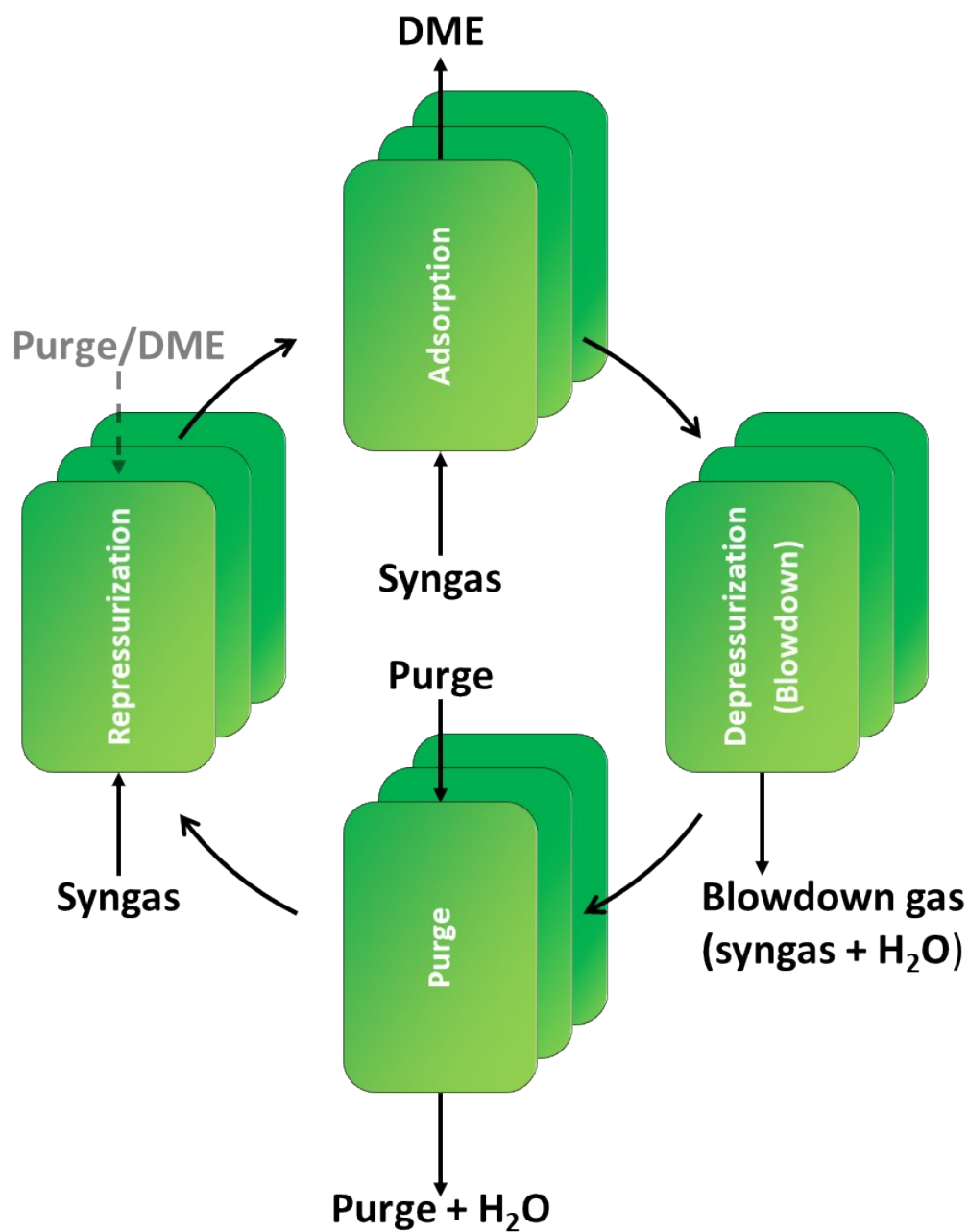


Figure 2. Schematic representation of a four step SEDMES PSA cycle.