

# Supplementary Information: Novel approach for low CO<sub>2</sub> intensity hydrogen from natural gas

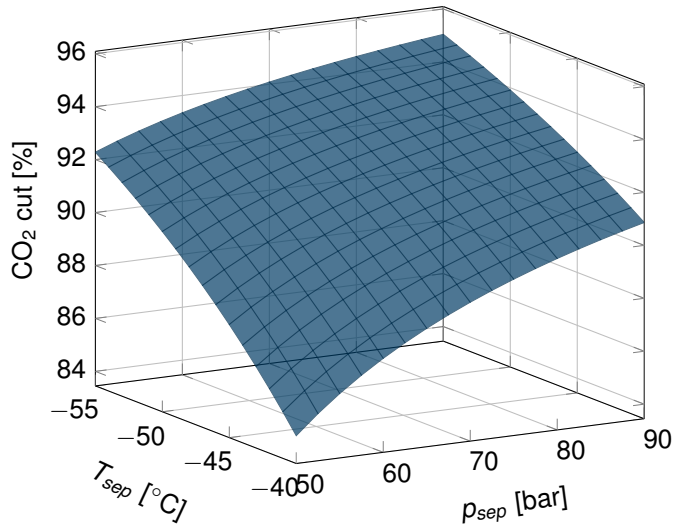
## S1 Maximum achievable CO<sub>2</sub> cut

The liquid yield and CO<sub>2</sub> cut achieved in the main separator is highly dependent on the separation temperature and pressure as well as feed CO<sub>2</sub> concentration and overall composition. To illustrate this, Eq. (S1) expresses the estimated CO<sub>2</sub> cut when separating a partially condensed binary H<sub>2</sub>/CO<sub>2</sub> mixture:

$$CO_2 cut = \frac{X_{CO_2} (f_{CO_2} - Y_{CO_2})}{f_{CO_2} (X_{CO_2} - Y_{CO_2})} \quad (S1)$$

In this expression,  $f_{CO_2}$  is the overall CO<sub>2</sub> fraction in the incoming feed stream, which varies considerably depending on the tail gas recycle ratio.  $Y_{CO_2}$  and  $X_{CO_2}$  are the respective equilibrium CO<sub>2</sub> fractions in the vapour and liquid phase separation products for any given temperature and pressure. When combining Eq. (S1) with measurement data for  $Y_{CO_2}$  and  $X_{CO_2}$  by Fandiño *et al.*<sup>1</sup>, the influence of these parameters becomes obvious.

Fig. S1 illustrates the CO<sub>2</sub> cut as a function of the separation pressure and temperature. The chosen feed conditions are the conditions of process configuration 1

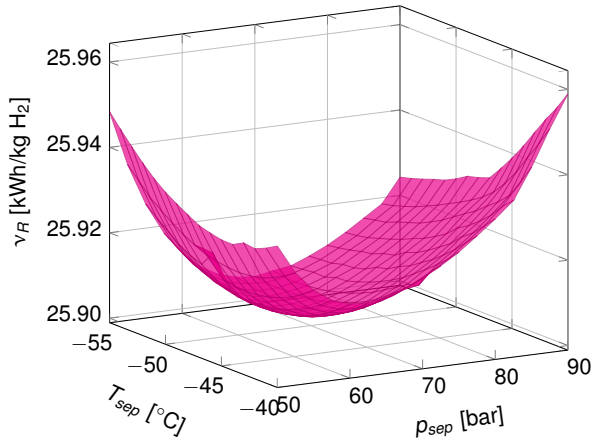


**Fig. S1** CO<sub>2</sub> cut as a function of the separation temperature  $T_{sep}$  and pressure  $p_{sep}$  for process configuration 2.

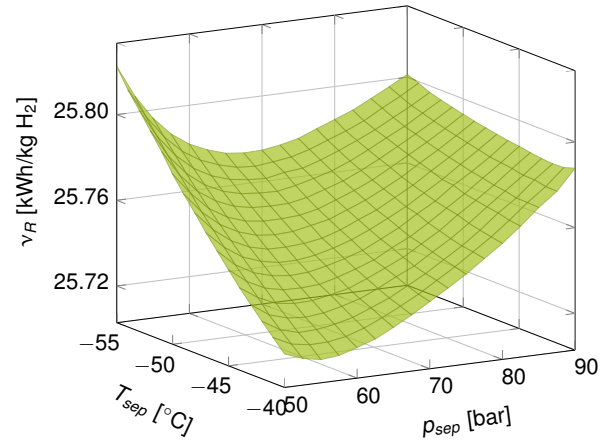
## Notes and references

- 1 O. Fandiño, J. M. Trusler and D. Vega-Maza, *International Journal of Greenhouse Gas Control*, 2015, **36**, 78 – 92.

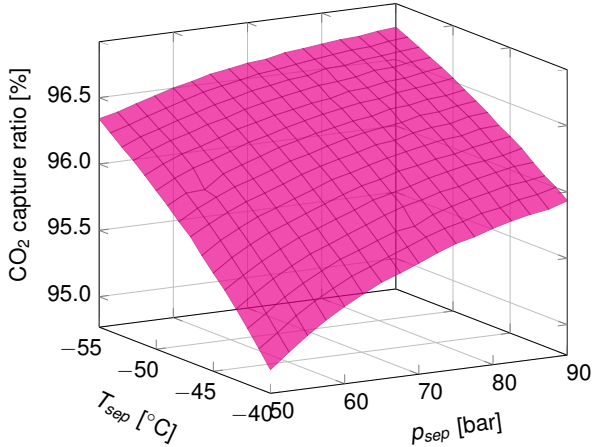
## S2 Figures related to sensitivity analysis in Section 3.2



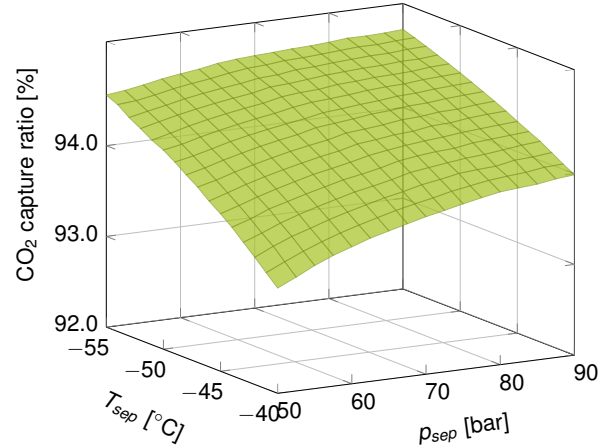
**Fig. S2** Specific equivalent power input as a function of the separation temperature  $T_{sep}$  and pressure  $p_{sep}$  for process configuration 1.



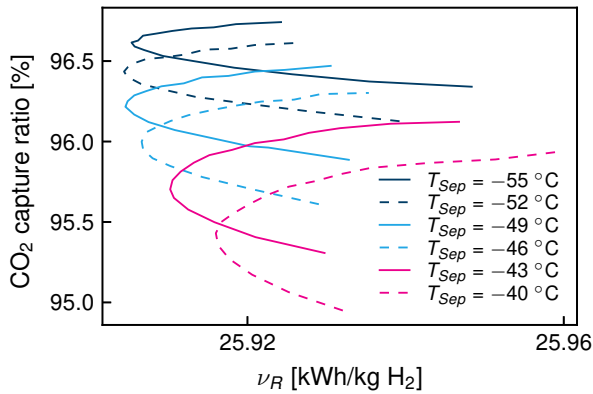
**Fig. S5** Specific equivalent power input as a function of the separation temperature  $T_{sep}$  and pressure  $p_{sep}$  for process configuration 3.



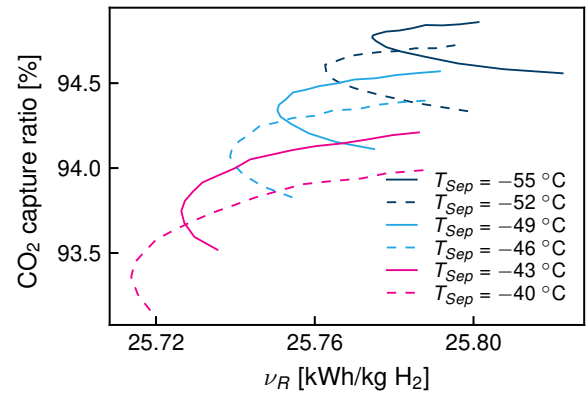
**Fig. S3** CO<sub>2</sub> capture ratio as a function of the separation temperature  $T_{sep}$  and pressure  $p_{sep}$  for process configuration 1.



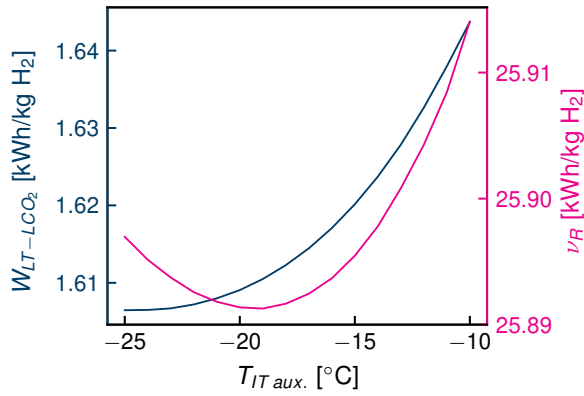
**Fig. S6** CO<sub>2</sub> capture ratio as a function of the separation temperature  $T_{sep}$  and pressure  $p_{sep}$  for process configuration 1.



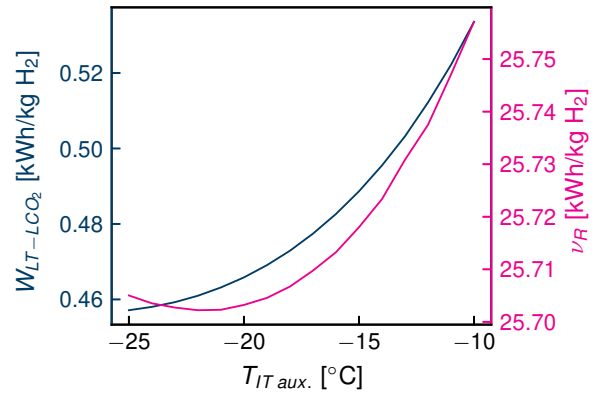
**Fig. S4** CO<sub>2</sub> capture ratio as a function of the specific equivalent power input at constant temperatures  $T_{sep}$  for process configuration 1. The reader is referred to the web version for colour coding.



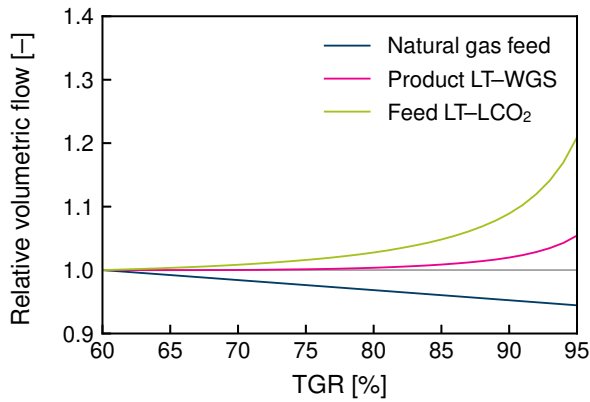
**Fig. S7** CO<sub>2</sub> capture ratio as a function of the specific equivalent power input at constant temperatures  $T_{sep}$  for process configuration 3. The reader is referred to the web version for colour coding.



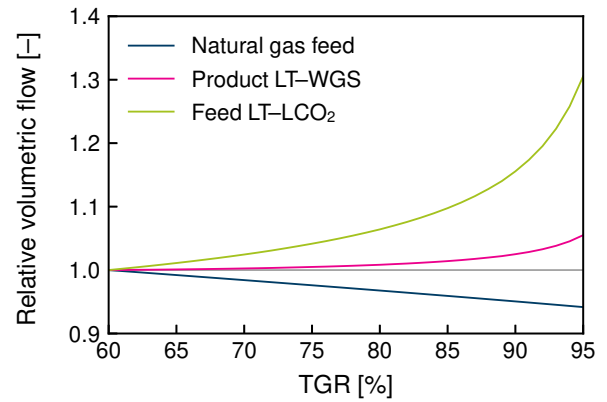
**Fig. S8** Specific energy demand of liquefaction section (left axis) and the specific equivalent power requirement (right axis) as a function of the outlet temperature of the IT refrigeration loop for process configuration 1. The reader is referred to the web version for colour coding.



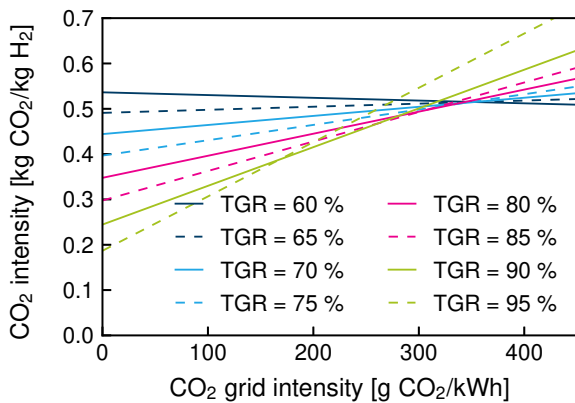
**Fig. S11** Specific energy demand of liquefaction section (left axis) and the specific equivalent power requirement (right axis) as a function of the outlet temperature of the IT refrigeration loop for process configuration 3. The reader is referred to the web version for colour coding.



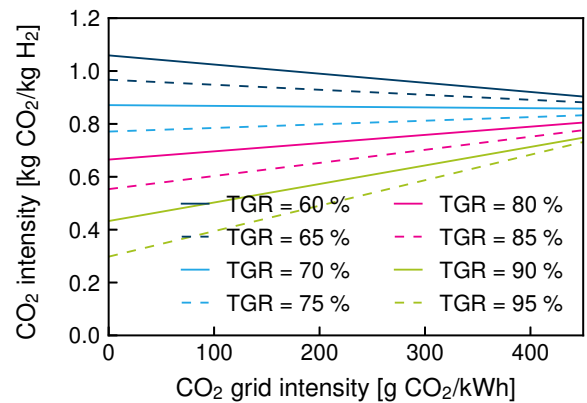
**Fig. S9** Relative volumetric flows for process configuration 1 and a feed temperature to the LT-WGS reactor of 200 °C. The grey line corresponds to a constant value of 1.0. The reader is referred to the web version for colour coding.



**Fig. S12** Relative volumetric flows for process configuration 3 and a feed temperature to the LT-WGS reactor of 225 °C. The grey line corresponds to a constant value of 1.0. The reader is referred to the web version for colour coding.



**Fig. S10** CO<sub>2</sub> intensity of hydrogen as a function of the CO<sub>2</sub> grid intensity for process configuration 1 and a feed temperature to the LT-WGS reactor of 200 °C. The reader is referred to the web version for colour coding.



**Fig. S13** CO<sub>2</sub> intensity of hydrogen as a function of the CO<sub>2</sub> grid intensity for process configuration 3 and a feed temperature to the LT-WGS reactor of 225 °C. The reader is referred to the web version for colour coding.