Acid gas conversion to COS by Zeolite 13X: an elementary assessment

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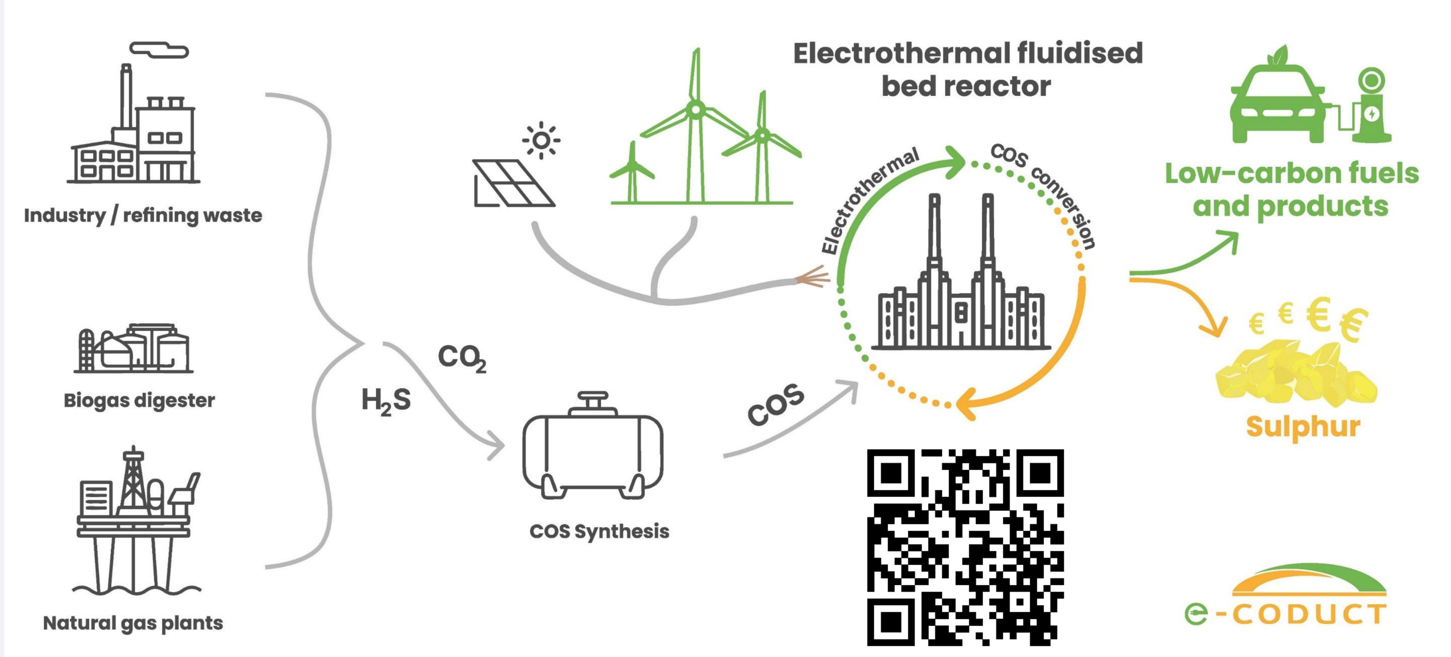
Introduction

Current technologies for CO_2 conversion demand highly pure CO_2 feed streams, complicating the use of streams containing impurities such as H_2S. The e-CODUCT project focuses on developing an integrated approach for treating acid gas streams through a two-step process. First, COS is produced from CO_2 and H_2S using zeolite catalysts. Then, COS is decomposed in an electrothermal fluidized bed to yield CO, a valuable platform molecule, and marketable sulphur.

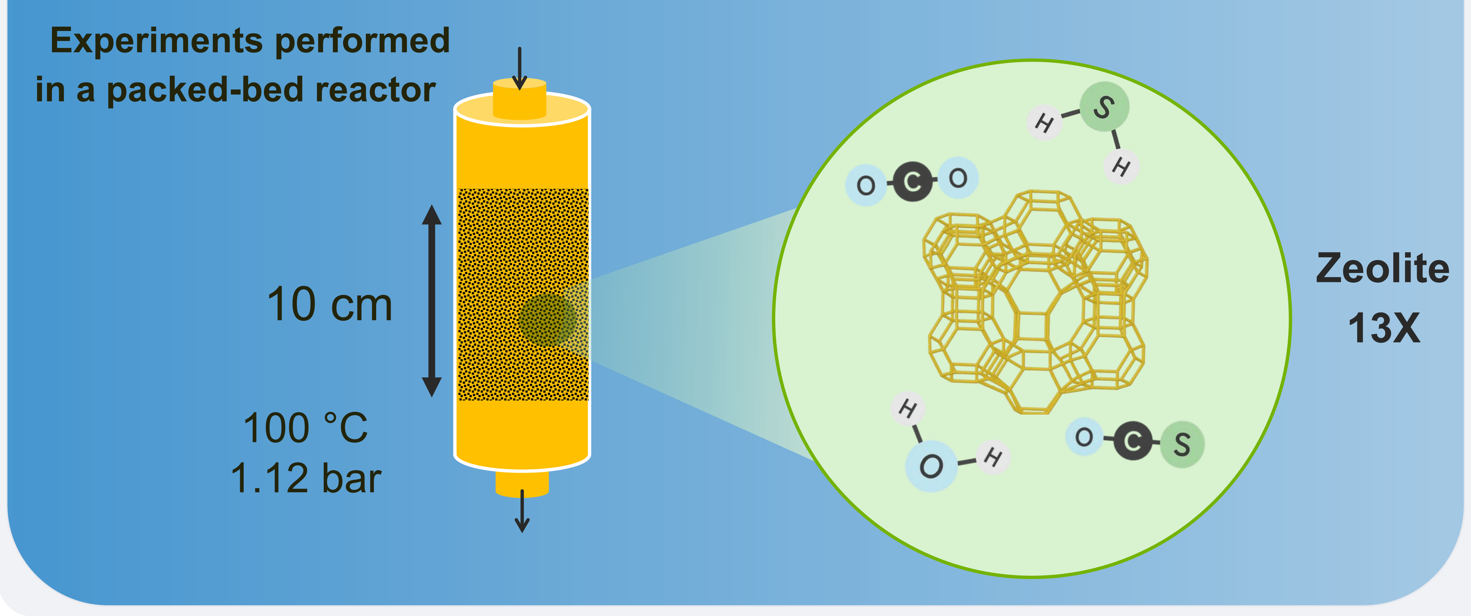
This work addresses the first step, by developing a kinetic model for COS formation over a zeolite 13X catalyst:

$$CO_2 + H_2S \leftrightarrows COS + H_2O$$



Methodology

Experimental campaign



Modeling approach

Reactor modeling

$$\frac{\partial C_i(x,t)}{\partial t} = -\frac{L}{\tau_b}\frac{\partial C_i(x,t)}{\partial x} + \frac{\rho_b}{\varepsilon_b} R_{w,i}$$

$$\frac{\partial L_{i*}(x,t)}{\partial t} = R_{w,i*}$$

Reaction modeling

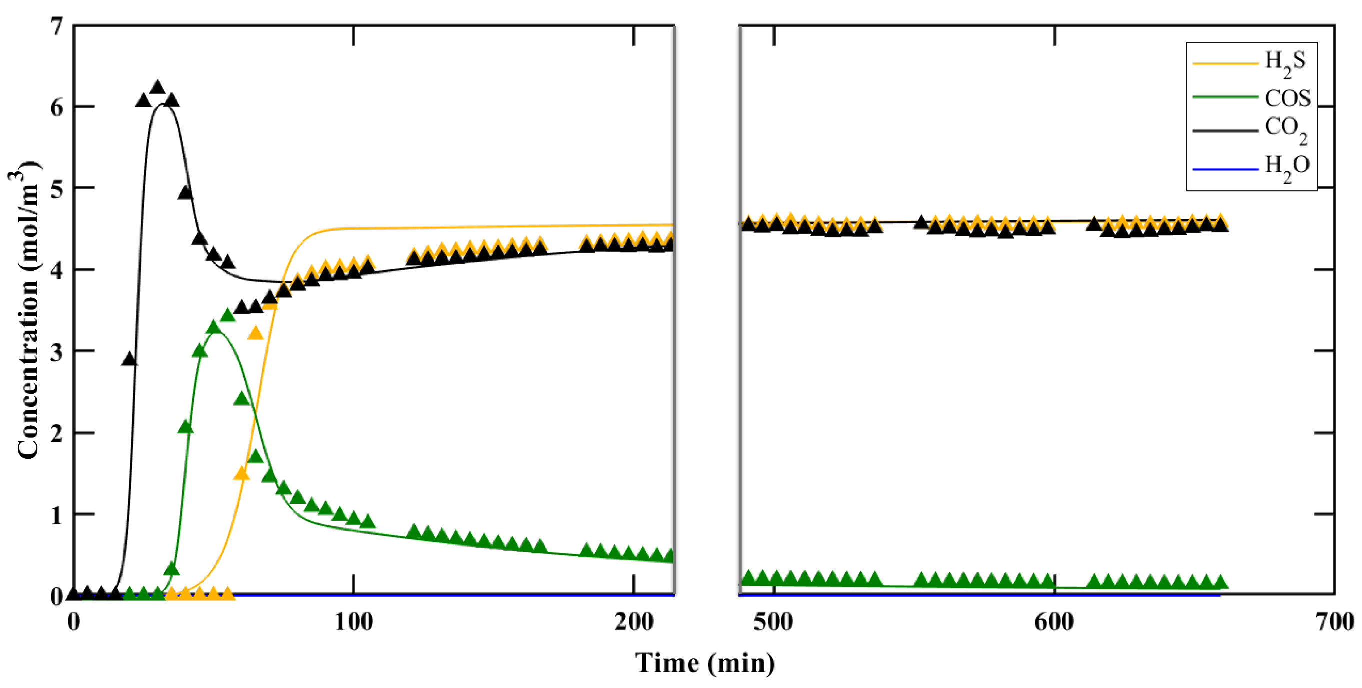
Based on the LHHW reaction mechanism

Reaction equation	Rate equation [mol (kg s)$^{-1}$]
$H_2S + * \leftrightarrows H_2S*$	$r_1 = k^{+}_{H_2S}\left(C_{H_2S}L_* - \frac{1}{K^{eq}_{H_2S}}L_{H_2S}\right)$
$CO_2 + * \leftrightarrows CO_2*$	$r_2 = k^{+}_{CO_2}\left(C_{CO_2}L_* - \frac{1}{K^{eq}_{CO_2}}L_{CO_2}\right)$
$H_2S* + CO_2* \rightarrow COS* + H_2O*$	$r_3 = k_r L_{CO_2} L_{H_2S}$
$COS + * \leftrightarrows COS*$	$r_4 = k^{+}_{COS}\left(C_{COS}L_* - \frac{1}{K^{eq}_{COS}}L_{COS}\right)$

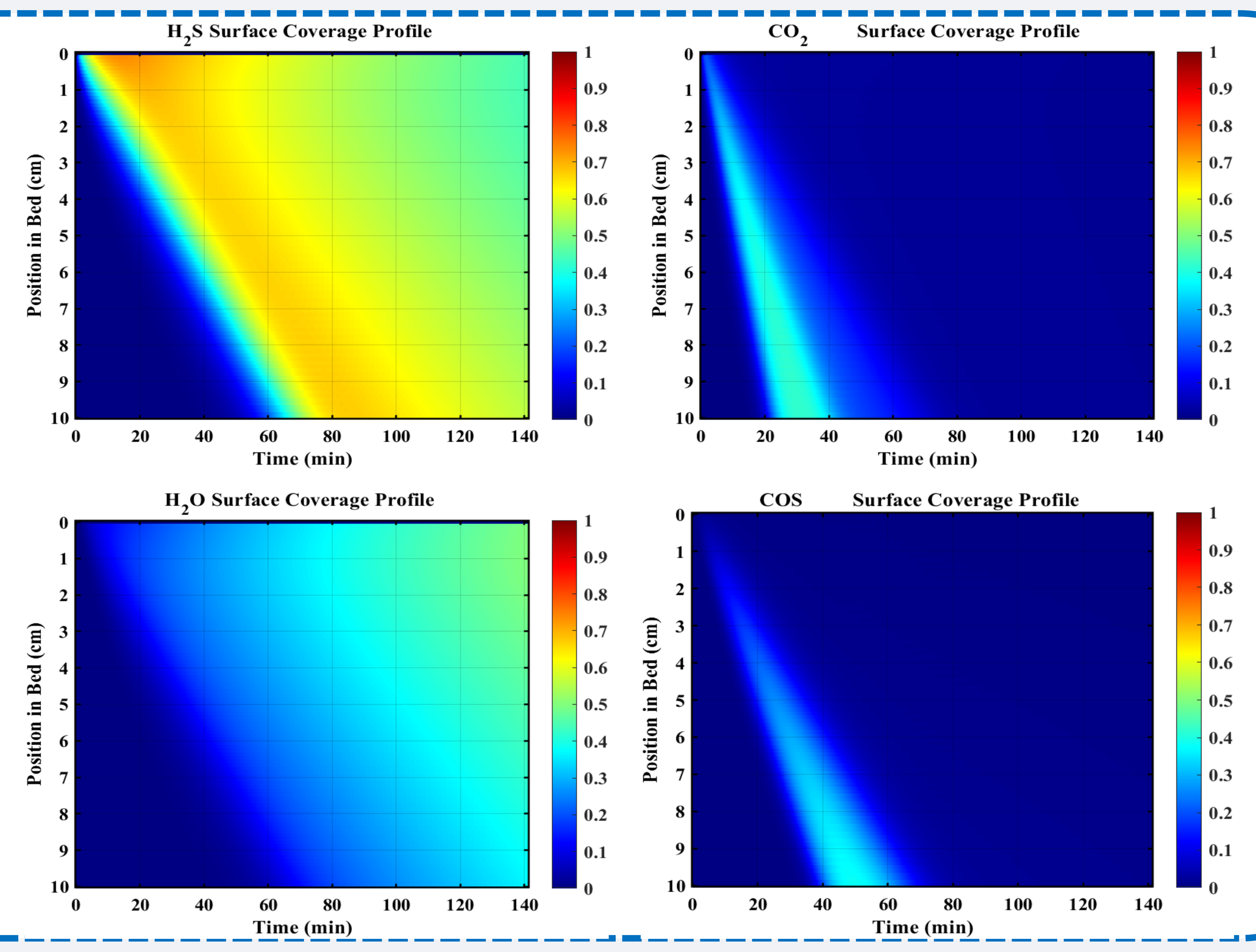
Results and discussion

Model fitting

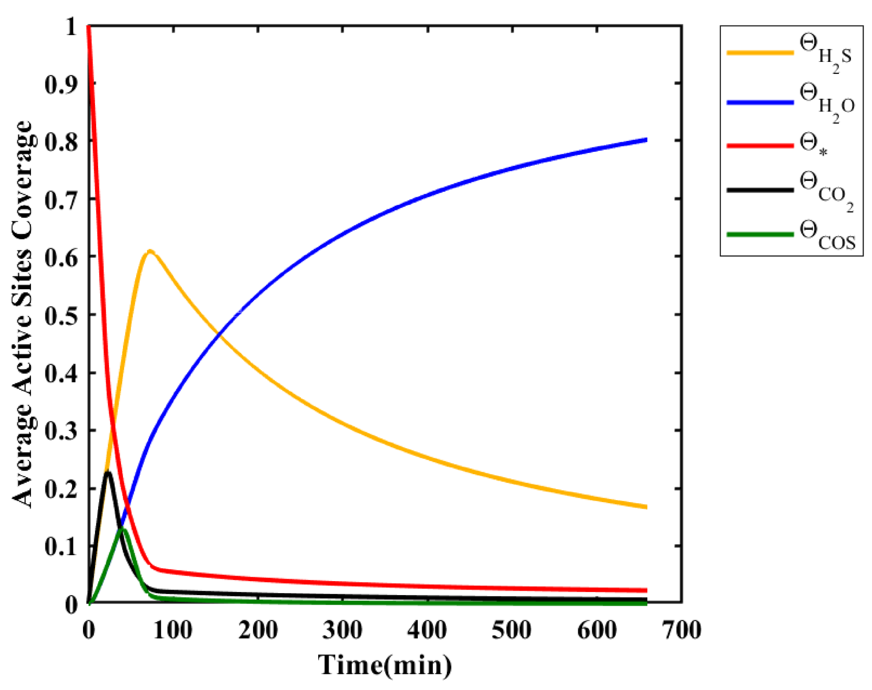
$H_2S:CO_2:N_2$ (%v/v) 13:13:74



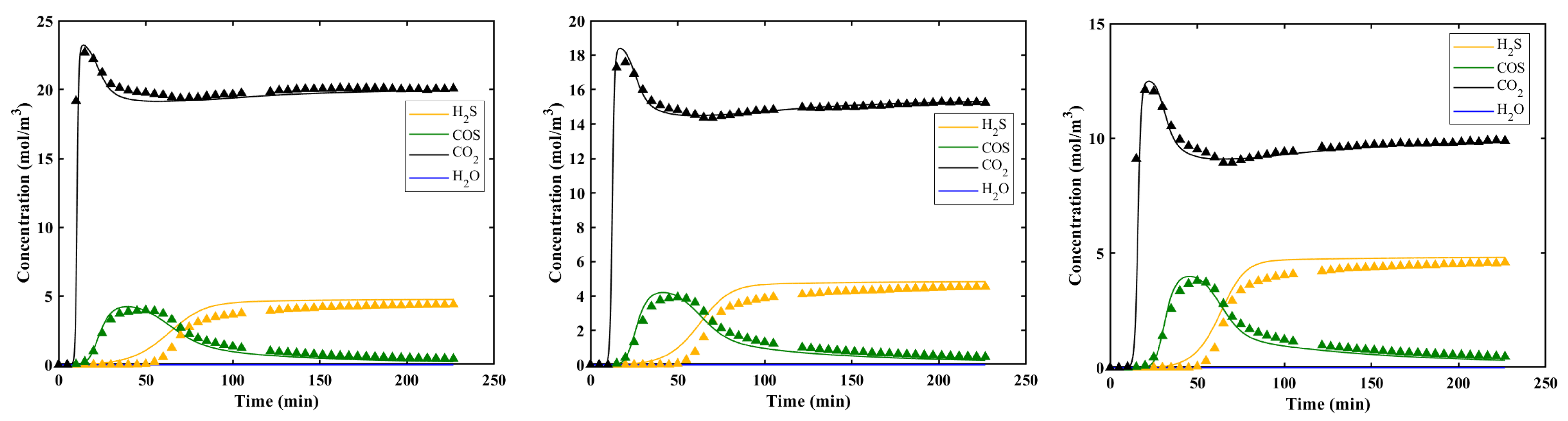
Surface coverage profiles along the reactor bed (y-axis) over time (x-axis).



Model predicted average active site coverages indicate that accumulation of water in the zeolite pore system drives the decline in performance.



Model validation across different feed compositions



$H_2S:CO_2:N_2$ (%v/v) 13:53:34

$H_2S:CO_2:N_2$ (%v/v) 13:39:48

$H_2S:CO_2:N_2$ (%v/v) 13:26:61

Conclusions

- The elementary-step model revealed the underlying interplay of adsorption, reaction, and catalyst deactivation through water accumulation leading to the experimentally observed trends.
- The model can properly predict experimental results across various feed compositions.

Acknowledgements

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