

## Supporting Information

# Optimizing Mesostructured Silver Catalysts for Selective Carbon Dioxide Conversion into Fuels

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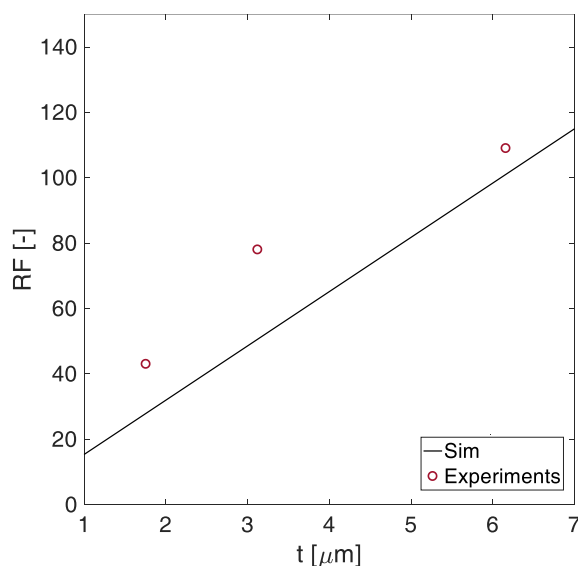
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The film thickness is analytically linked to the number of pores and the sphere radius of the template spheres. Assuming a sintering process, where the sphere overlap is proportional to sphere radius (Eq. (S1) and (S2)), the film thickness is determined by eq. (S3) (Figure S1).

$$r_{\text{sphere}} = r_{\text{pore}} - dr \quad (\text{S1})$$

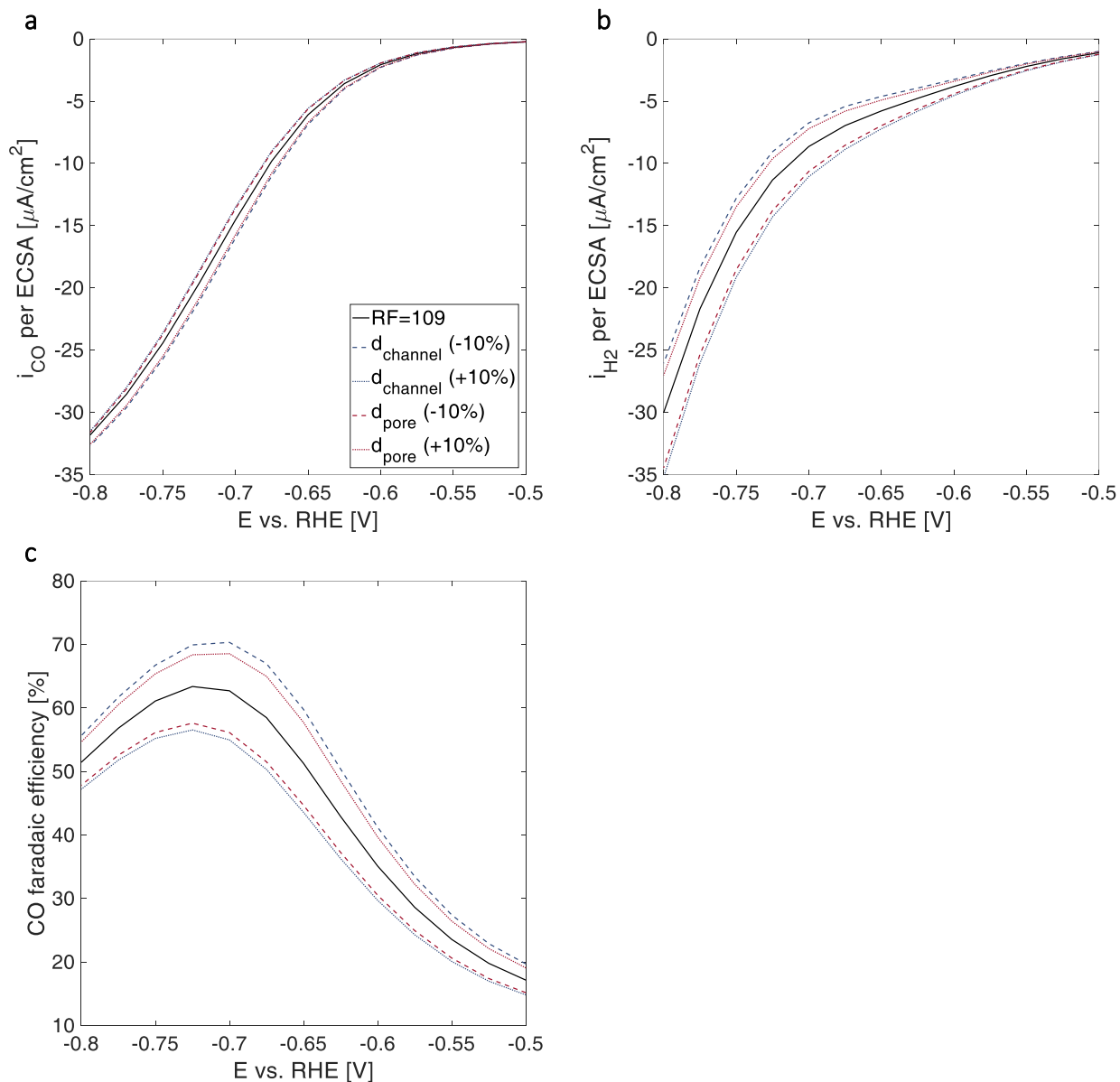
$$dr = 0.07 \cdot r_{\text{pore}} \quad (\text{S2})$$

$$t = r_{\text{sphere}} + (n_{\text{pores}} - 1) \cdot (2 \cdot \sqrt{6} / 3 \cdot r_{\text{sphere}}) \quad (\text{S3})$$

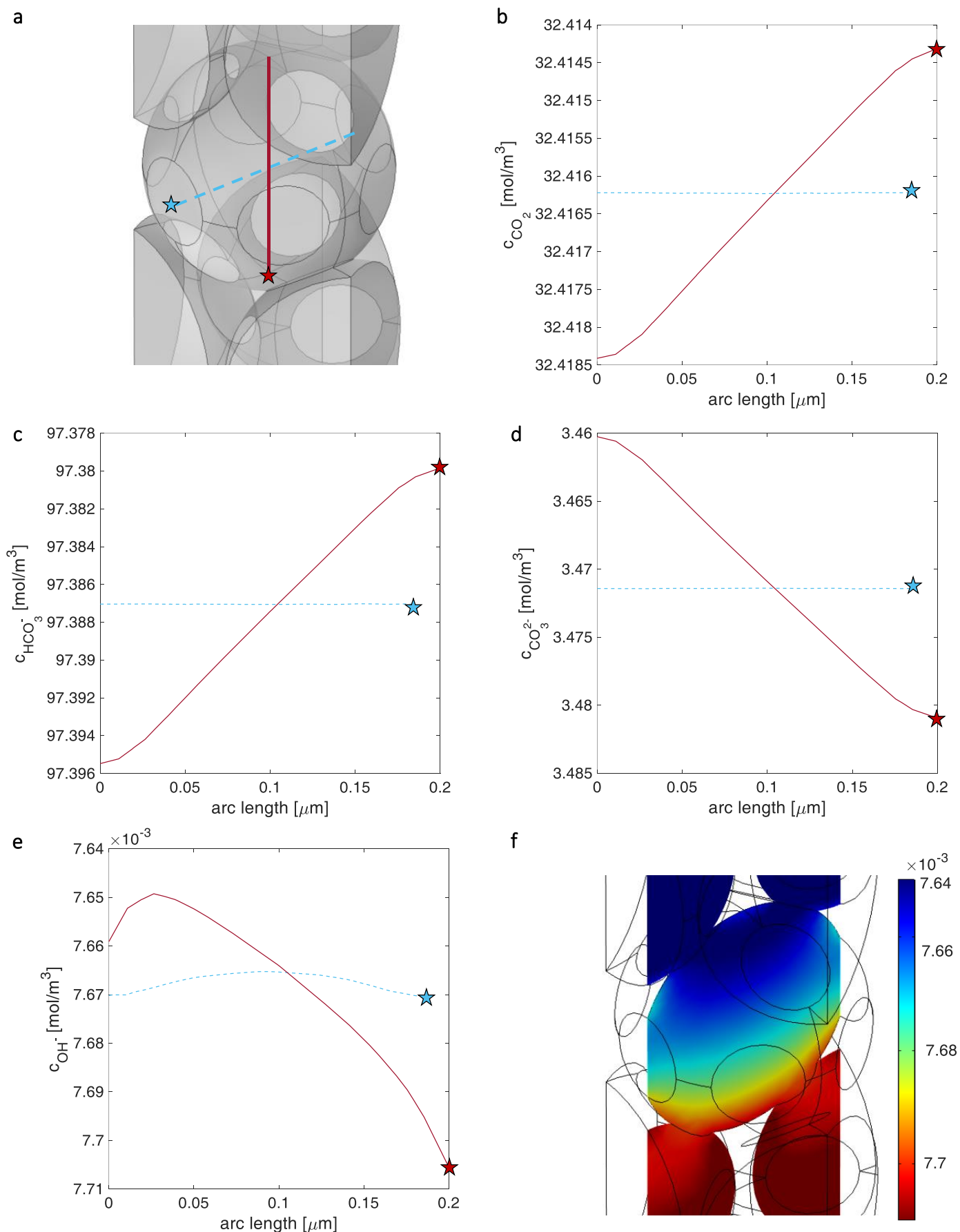


**Figure S1.** Roughness factor as a function of the Ag-IO film thickness. The experimental data were taken from Yoon et al.<sup>17</sup> The roughness factor of the model was a linear function. Generally, the model values were smaller than the experimentally determined values. Most probably, there is a roughness on the pore level that is not considered in the model.

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**Figure S2.** A sensitivity study for the model with  $RF=109$ . The partial current densities and the CO faradaic efficiency are shown for  $\pm 10\%$  in pore and channel diameter. The water reduction was more sensitive to the changes in morphology than the  $CO_2$  reduction. Reducing the pore diameter decreased the CO faradaic selectivity, whereas a reduction in the channel diameter enhanced the selectivity. In this sensitivity study, the number of pores were kept constant, but the film thickness could change, leading to a smaller overall surface area for a reduced pore diameter, and a larger surface area for a structure with smaller channel diameters. For both cases, the deviation in the maximum CO faradaic efficiencies was smaller than 7 percent points.



**Figure S3.** (a) Illustration of the horizontal and vertical cut line through the space of a pore at  $z=1 \mu\text{m}$  for an Ag-IO with  $\text{RF}=109$  at  $-0.8\text{V}$  vs. RHE. (b)-(e) Concentration profiles of  $\text{CO}_2$ ,  $\text{HCO}_3^-$ ,  $\text{CO}_3^{2-}$  and  $\text{OH}^-$  along the cut lines, with the star indicating the end position. (f) Contour plot of the  $\text{OH}^-$  concentration on the  $x$ - $y$  cut plane.