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Supporting Information

Operando Electron Microscopy Study of Cobalt-based Fischer-Tropsch Nanocatalysts

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Quantitative determination of the catalytic performances

The total outlet flow-rate Q_out is calculated according to the formula:

$$Q_{out} = \frac{Q_{He,\in}}{V_{He,out}}$$

where Q_(He,in) is the inlet flow-rate of helium, and V_(He,out) the measured outlet volume fraction of helium.

The flow-rates Q_(i,out) of the compounds *i* are then given by:

$$Q_{i,out} = Q_{out} \cdot V_{i,out}$$

 $V_{i,out}$ being the concentration of the compound *i* at the outlet.

The CO conversion, the CH₄, C₂, C₃, C₄ and C₅₊ selectivity are given by:

$$COconv. = \frac{Q_{CO, \in -Q_{CO,out}}}{Q_{CO, \in}}$$
$$S_{CH_4} = \frac{Q_{CH_4,out}}{Q_{CO, \in -Q_{CO,out}}}$$
$$S_{C_i} = \frac{i \cdot Q_{C_i,out}}{Q_{CO, \in -Q_{CO,out}}}, 2 < i < 4$$

$$S_{C_{5+}=1-(S_{CH_4}+S_{C_2}+S_{C_3}+S_{C_4})}$$



Figure SI-1: Photographs of the gas cell holder used for the in situ TEM experiments (a), the chips mounting on the tip of the holder, their alignment and sealing (b-e), zoom-in on the viewing area of the holder tip (f), and the holder insertion inside the microscope (g).



Figure SI-2: Temperature programmed reduction of the two Co-Pt catalysts deposited on silica and alumina.



Figure SI-3: Typical scanning electron microscopy (SEM) images acquired on Co-Pt/Al₂O₃ (top) and Co-Pt/SiO₂ (bottom) catalysts.



Figure SI-4: CO conversion as a function of time on stream with variable CO flow rates for Co-based catalysts supported on alumina or silica.



Figure SI-5: C5+ selectivity as a function of CO conversion for Co-based catalysts supported on alumina or silica.