Supplementary information for

Catalytic exchange of hydrogen isotopes intensified by two-phase stratified flow in wettability designable microchannels

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S1. Preparation of Pt/AC/PDMS coating on the walls of the microchannel reactor

The Pt/AC/PDMS coatings were prepared on the channel walls according to a ‘‘glue + powder’’ approach described by Li et al.[S1] using the commercial PDMS (Sylgard 184) as the “glue” and the Pt/AC catalyst as the “powder”. The diluted PDMS solution (10% w/v in *n*-hexane) was prepared by dissolving PDMS base and curing agent (a 10:1 w/w ratio) in *n*-hexane. The Pt/AC catalyst was ground and then sieved by a 200-mesh sieve prior to preparing the coatings. The procedure is specified as follows:

(1) The channel substrate was sealed with a silicone sheet.

(2) The diluted PDMS solution was injected into the microchannel.

(3) A thin liquid film would be deposited on the channel walls while the excess solution was withdrawn.

(4) The silicone sheet was opened to let hexane evaporate.

(5) After the most of hexane was evaporated, the uncured adhesive surface was covered by the Pt/AC powder for 5 minute.

(6) After removing the excess powder, the prepared coating was cured under 80 °C for 2 hours. The top surface of the channel substrate was cleaned so that the channel substrate could bond with the cover plate successfully.

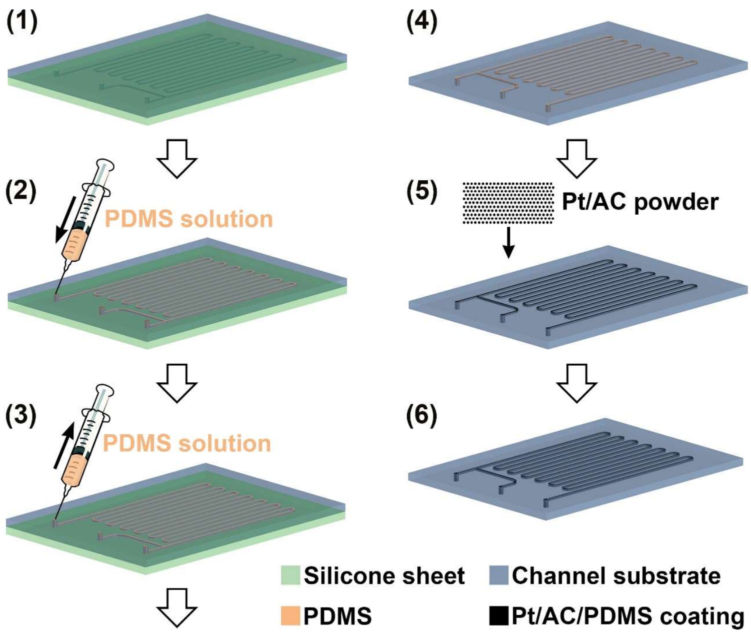

Fig. S1. Schematic of the process for preparing Pt/AC/PDMS coating on the walls of the microchannel reactor.

S2. Calculation of the y^*_{HD1}

Rolston et al.[S2] measured the overall deuterium (D) isotope separation factor between hydrogen and liquid water, α, at 280–370 K. The α is defined as eqn (S1). The correlation between α and temperature (T, K) is presented as eqn (S2). Based on the mass balance of D, the D abundance in the liquid water at outlet of the microchannel reactor (x_{D1}) is calculated by eqn (S3). The equilibrium D abundance in hydrogen at the outlet (y^*_{D1}) can be calculated by eqn (S4) transformed from eqn (S1). The equilibrium molar fraction of HD in hydrogen at the outlet (y^*_{HD1}) is twice the y^*_{D1} calculated as eqn (S5).

$$\alpha = \frac{x^*_D(1-y^*_D)}{y^*_D(1-x^*_D)} \tag{S1}$$

$$\ln\alpha = -0.2143 + \frac{368.9}{T} + \frac{27870}{T^2} \quad \text{(S2)[S2]}$$

$$x_{D1} = \frac{2x_{D0}F_{H_2O} - (y_{HD1} - y_{HD0})F_{H_2}}{2F_{H_2O}} \tag{S3}$$

$$y^*_{D1} = \frac{1}{1-\alpha+\alpha/x_{D1}} \tag{S4}$$

$$y^*_{HD1} = 2y^*_{D1} \tag{S5}$$

where x^*_D and y^*_D are the equilibrium D abundances in liquid water and hydrogen, respectively; x_{D0} is the D abundance in liquid water at inlet of the microchannel reactor; F_{H_2O} and F_{H_2} are the feeding molar flow rates of water and hydrogen, respectively; y_{HD0} and y_{HD1} are the molar fractions of HD in hydrogen at inlet and outlet of the microchannel reactor, respectively.

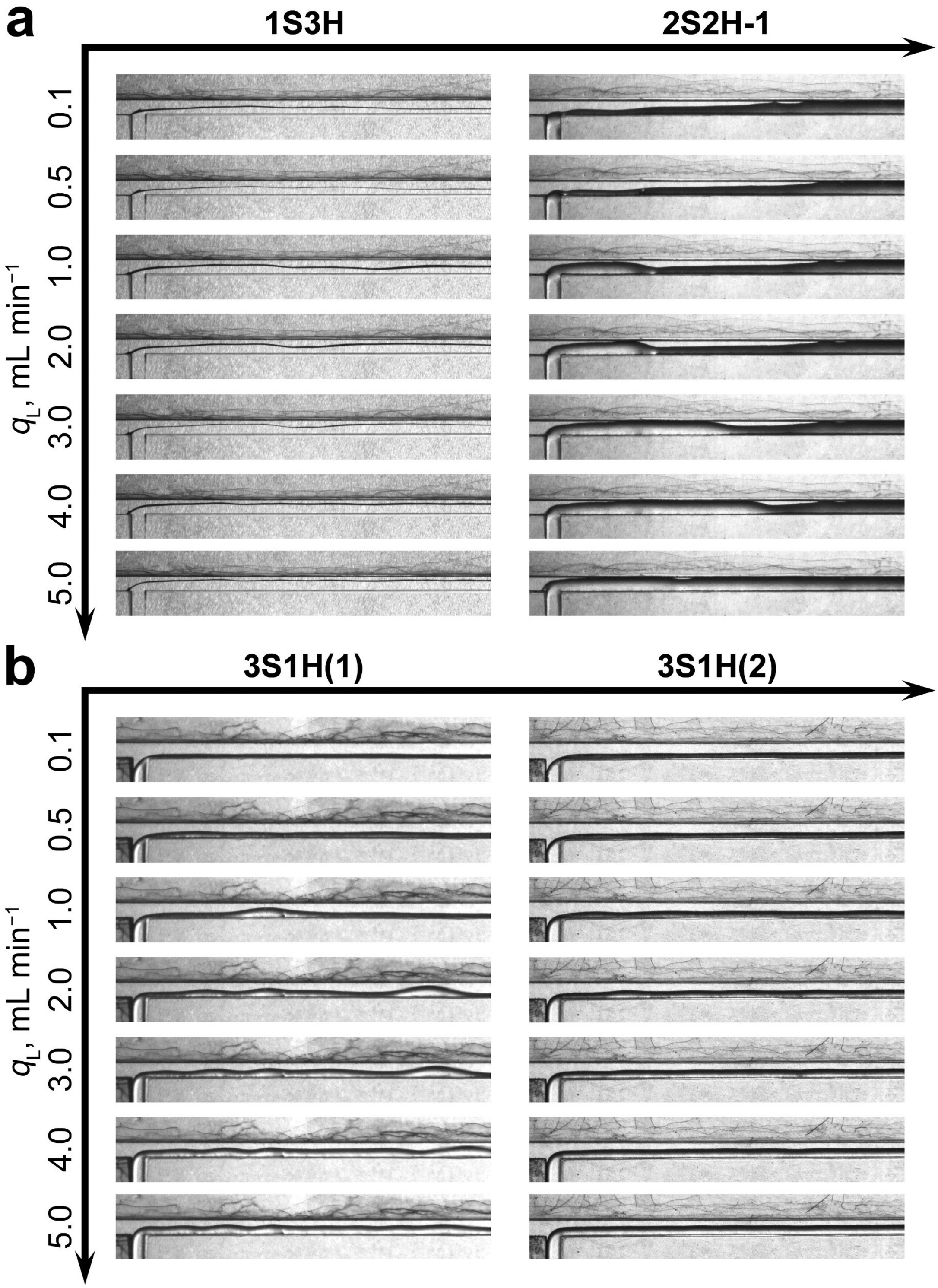

Fig. S2. Effects of liquid flow rate (q_L) on flow patterns at a given gas flow rate (q_G = 15.0 mL min^{-1}) in the **1S3H**, **2S2H-1**, **3S1H(1)** and **3S1H(2)** microchannels.

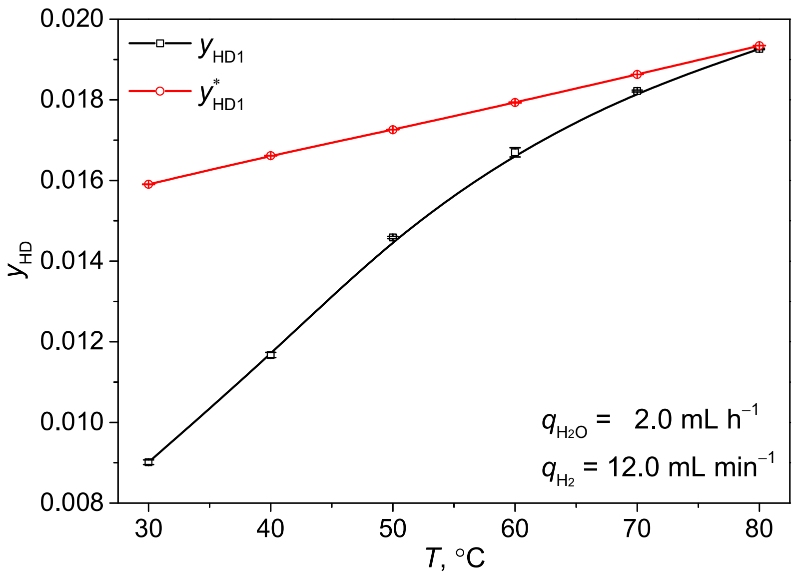

Fig. S3. Effects of temperature (T) on molar fraction (y_{HD1}) and equilibrium molar fraction (y^*_{HD1}) of HD in hydrogen at outlet of the microchannel reactor at x_{D0} = 0.03, q_{H_2O} = 2.0 mL h^{-1} and q_{H_2} = 12.0 mL min^{-1}.

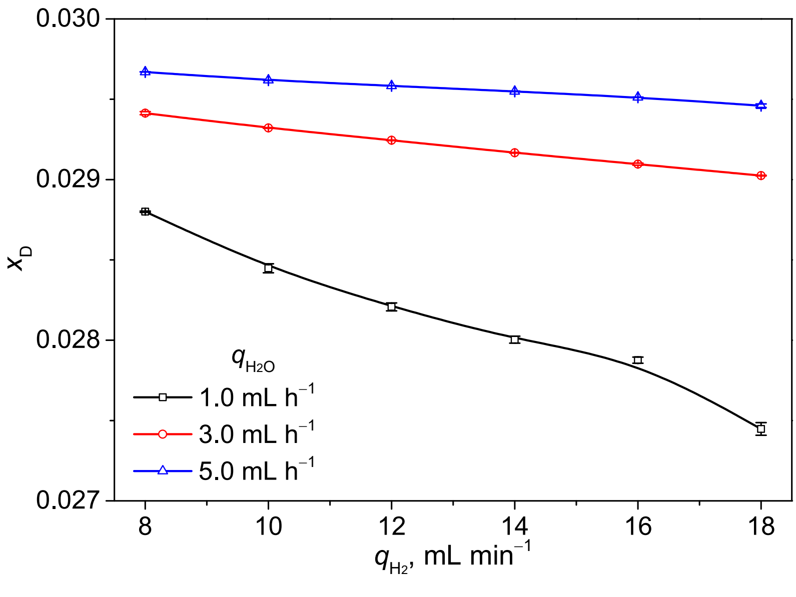

Fig. S4. Effects of hydrogen (q_{H_2}) and water (q_{H_2O}) flow rates on average D abundance in the water flow ($x_D = (x_{D0} + x_{D1})/2$) of the microchannel reactor at x_{D0} = 0.03 and T = 60 °C.

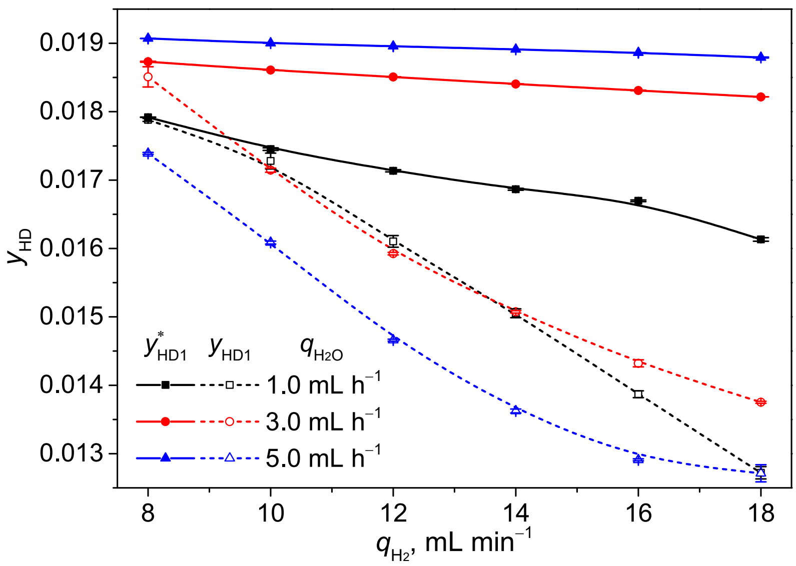

Fig. S5. Effects of hydrogen (q_{H_2}) and water (q_{H_2O}) flow rates on molar fraction (y_{HD1}) and equilibrium molar fraction (y^*_{HD1}) of HD in hydrogen at outlet of the microchannel reactor at x_{D0} = 0.03 and T = 60 °C.

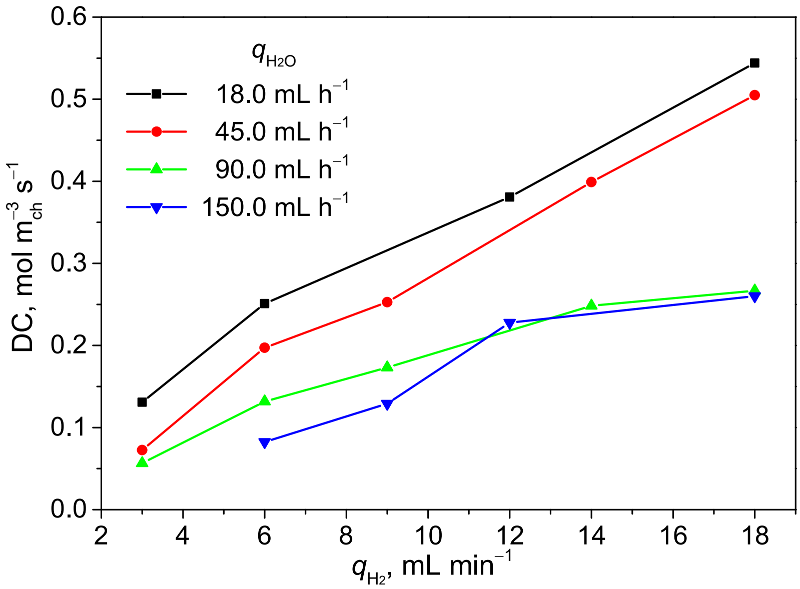

Fig. S6. Effects of hydrogen (q_{H_2}) and water (q_{H_2O}) flow rates on deuterium removal capacity (DC) in the microchannel reactor at x_{D0} = 0.03 and T = 60 °C.

References

[S1] Z. Li, M. Cao, P. Li, Y. Zhao, H. Bai, Y. Wu and L. Jiang, *Matter*, 2019, **1**, 661–673.

[S2] J. H. Rolston, J. Den Hartog and J. P. Butler, *J. Phys. Chem.*, 1976, **80**, 1064–1067.