Supplementary material of '**Pore-scale study of CO_2 desublimation and sublimation in a packed bed during cryogenic carbon capture**'

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1 Grid Convergence test

Grid-independence simulations have been conducted for CO_2 desublimation and sublimation in case Base (see Sec.4.1). The computational domain is covered by three different grids (320 × 320, 640 × 640, and 960 × 960). Temporal evolutions of two metrics are calculated and recorded in Fig. S1, namely the volume fraction of captured solid CO_2 (ϕ_c) and the averaged temperature of active boundaries ($\overline{T}_a$).

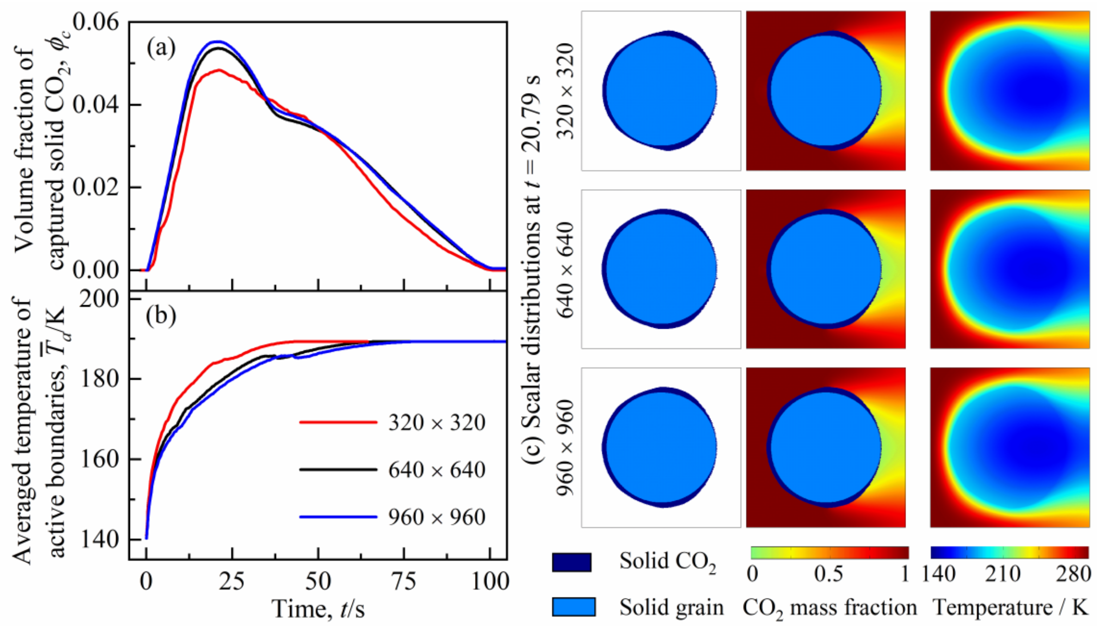

Fig. S1. Grid convergence test. Temporal evolutions of (a) volume fraction of the captured solid CO_2 (ϕ_c), (b) averaged temperature of active boundaries ($\overline{T}_a$), and (c) contours of solid CO_2, CO_2 mass fractions, and temperature at the time instant 20.79 s with three grid resolutions in case Base.

By comparing profiles of both ϕ_c and $\overline{T}_a$, it is evident that the simulation results for the 640×640 and 1024×1024 grids exhibit good agreement. The curves for the 384×384 coarser grid display a similar pattern to those for the finer grids (i.e.,

640×640 and 1024×1024), but obvious discrepancies are absorbed between results for the coarser and finer grids. Specifically, the 384×384 coarser grid brings about the faster heating of the packing grain ($\overline{T}_a$) and subsequently the lower CO_2 capture performance (ϕ_c). In addition, the calculated distributions of solid CO_2, CO_2 mass fraction, and temperature at the time instant 20.79 s are shown in Fig. S1 (c). All these contours under different grid resolutions show a similar pattern, but the captured solid CO_2 for the 384×384 coarser grid is observed to be less than those for the other two finer grids (i.e., 640×640 and 1024×1024). These comparisons suggest that the 640×640 grid is sufficiently fine to capture the grid-independent properties CO_2 desublimation and sublimation processes.

2 Sensitivity tests of CO_2 mass fraction

This study focuses on evaluating effects of operating conditions, including the initial bed temperature (subcooling degree, ΔT_s), gas feed rate (Peclet number Pe), and bed porosity (ψ), while maintaining a constant inlet CO_2 mass fraction of $Y_0 = 1.0$. To verify the robustness of the present findings, the CO_2 desublimation and sublimation in a cryogenic packed bed are modelled for different CO_2 mass fractions. Through simulations and comparisons for both single-grain and packed-bed cases, the CO_2 capture process is found to exhibit the similar desublimation and sublimation properties as those discussed in Sec. 4. For illustration, results at $Y_0 = \{0.15, 0.5, 1.0\}$, $\Delta T_s = 0.185$, Pe=15.57 in a packed bed are provided in Fig. S2. That includes temporal evaluations of the captured solid CO_2 (v_c), position of the saturation front (l_{sat}), and overall mass fraction rate via desublimation and sublimation (m_r*, m_{rd}* and m_{rs}*).

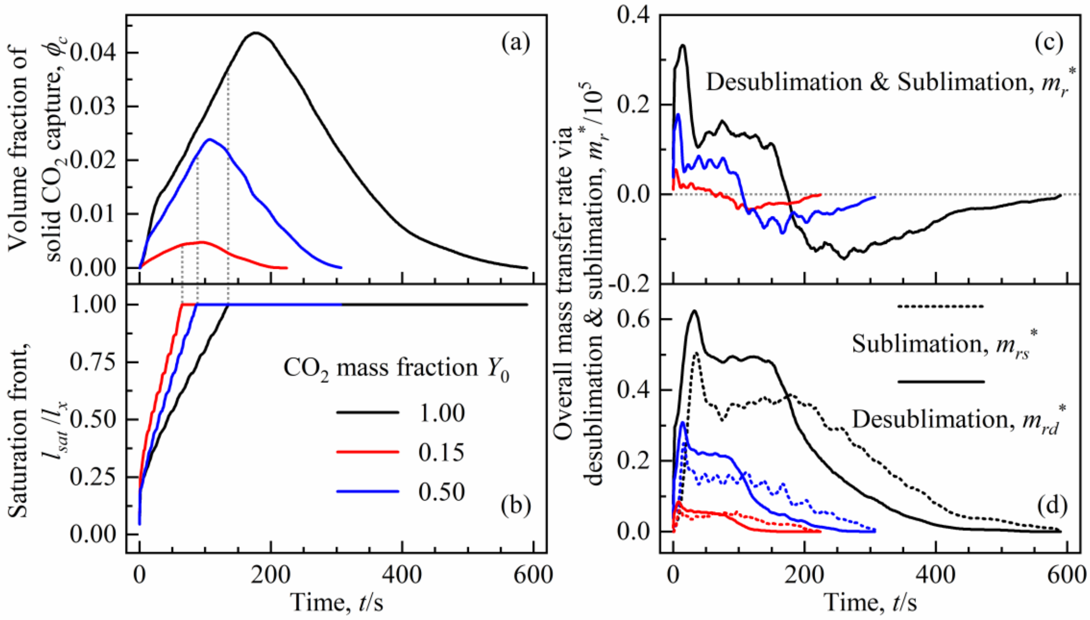

Fig. S2. Effects of CO_2 mass fraction. Temporal evolutions of (a) volume fraction of the captured solid CO_2 (ϕ_c), (b) position of the saturation front (l_{sat}), and (c-d) overall mass transfer rate via desublimation and sublimation (m_r^*, m_{rd}^* and m_{rs}^*).

Under each Y_0 value, the desublimation and sublimation processes show a similar tendency. The captured solid CO_2 (ϕ_c) exhibits an initial rapid increase followed by a gradual decline toward zero. This behavior is attributed to variations in mass transfer rates (m_r^*, m_{rd}^* and m_{rs}^*). Additionally, it is observed that the saturation front (l_{sat}) progresses toward the bed outlet as the CO_2 desublimation proceeds. The saturation time is earlier than the peak point of ϕ_c. In addition to these consistencies with the results in Sec 4.3, the effects of Y_0 are noted. With the increasing Y_0, the maximum capacity of the packed bed (i.e., ϕ_{cm}), the operating time, and the capacity loss decreases. In this study, to efficiently demonstrate the desublimated CO_2, the value with the largest CO_2 capture capacity is selected, namely, $Y_0 = 1$.

3 Effects of sub-grid surface change

To consider changes in local sub-grid surface, the random pore model is applied to determine the specific surface per unit volume for CO_2 desublimation and sublimation as [1],

$$a_r = (1 - x)a_{r0}\sqrt{1 - \vartheta ln(1 - x)}. \tag{S1}$$

Here a_{r0} is the initial specific surface, x is the solid CO_2 conversion rate, and ϑ is the structural parameter. Here, we set $a_{r0} = 1$ and $\vartheta = 10$ [1]. This calculation equation (S1) gives the maximum surface $a_r = 1.5a_{r0}$ at $x = 0.33$.

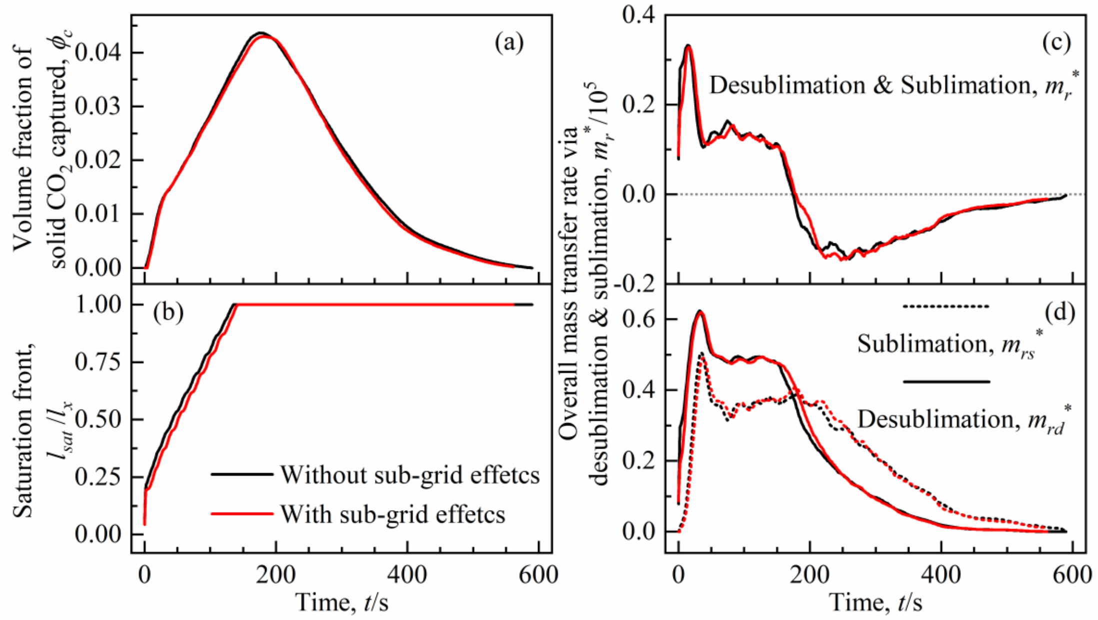

Fig. S3. Sensitivity tests of sub-grid CO_2 desublimation and sublimation. Temporal evolutions of (a) volume fraction of the captured solid CO_2 (ϕ_c), (b) position of the saturation front (l_{sat}), and (c-d) overall mass transfer rate via desublimation and sublimation (m_r^*, m_{rd}^* and m_{rs}^*).

To demonstrate the significance of including changes in sub-grid surface, the packed-bed test in Sec. 4.3 is simulated again by the present LB model but with a_r

being calculated as in Eq. (S1). The calculated CO_2 desublimation and sublimation properties are recorded versus time in Fig. S3, including volume fraction of the captured solid CO_2 (ϕ_c), position of the saturation front (l_{sat}), and overall mass transfer rate via desublimation and sublimation. For comparison, results of the test without sub-grid effects are included in Fig. S3. From the simulation results, it is obvious that the additionally introduced changes in sub-grid surface have limited impact on the CO_2 desublimation and sublimation properties. This is because, in a packed bed containing multiple grains, the desublimation rate is relatively stronger compared to the supply of CO_2 via the flue gas stream. Furthermore, the change in $a_r \in [0, 1.5a_{r0}]$ is relatively small. Considering the minimal effects of sub-grid surface change on the desublimation and sublimation of CO_2, this study uses a constant specific surface as $a_r = 1$.

References

[1] Q. Xu, X. Dai, J. Yang, Z. Liu, L. Shi. Image-based modelling of coke combustion in a multiscale porous medium using a micro-continuum framework, Journal of Fluid Mechanics 932 (2022) A51.