Supplementary material of ‘**Study of CO2 desublimation during cryogenic capture using the lattice Boltzmann method**’

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# Grid convergence tests

Grid-independence tests are carried out by simulating CO2 desublimation on the surface of a cooled cylinder under varying operation conditions (see Table 1 in our paper). Three different grids (,, and are applied to cover the front view of the computational domain in Fig. 1 (see our paper). Results of the Base case have been discussed in Appendix D (see our paper). To further clarify effects of the grid resolution on the desublimation process with a cluster-like solid CO2 layer (SCL), the calculated scalar contours and temporal evolutions of desublimation properties are shown in Figure S1 and Figure S2. As can be seen, the calculated contours and profiles under different grid resolutions show similar trends. The injected CO2 desublimates to generate the cluster-like SCL on the cylinder surface, leading to the efficient capture of the injected CO2. Meanwhile, the desublimated SCL is found to suppress both heat and mass transfer within it.

A more detailed look reveals inconsistencies between results under fine and coarse grid resolutions. The cluster-like structure of SCL is successfully and consistently produced by grids and . However, under the coarse grid mesh , the structure becomes coarse-grained and is different from that obtained using fine grids. As for desublimation parameters, evolution profiles for grids and match each other well but differ from those for the grid . These comparisons indicate that the grid of size is fine enough to obtain grid-independent results. This grid was thus adopted for the present lattice Boltzmann (LB) simulations of CO2 desublimation on the cooled cylinder surface.

Shape

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Figure S1. Grid convergence tests: contours of solid CO2, temperature (*T*), and CO2 mass fraction (*Y* ) at *t* = 28.3 s in case A1 with three grid resolutions of (*a*) 384×384, (*b*) 640×640, and (*c*) 1024×1024. Zoom-in views of SCL in highlighted gray rectangles are provided.

Chart, line chart

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Figure S2. Grid convergence tests: temporal evolutions of (*a*) the volume fraction of captured solid CO2 (), (*b*) the capture efficiency of injected CO2 (), (*c*) the overall desublimation rate (), and (*d*) the utilization of cylinder surface () in case A1 with three grid resolutions.

# Model validation

In this section, simulations are conducted to verify the proposed LB model for studying CO2 desublimation on the cooled cylinder surface. Given the lack of analytical solutions for such a complex problem, Appendix B in our paper conducted two sets of simulations to compare with existing experimental and numerical data. The proposed LB model thus has been verified for simulating CO2 desublimation. In addition, we adopt four benchmark problems with widely-accepted or analytical solutions to test the accuracy of the key sub-models of the proposed LB method, including the boundary scheme for mass conservation at reactive fluid-solid interface, the source term for conjugate heat transfer between two components or phases, and the volume of pixel (VOP) scheme for solid growth. Note that, the mass conservation condition in Eq. (2.12) (see our paper) for describing CO2 desublimation on the solid cylinder is similar to the mass conservation at the reactive fluid-solid interface.

## Mass diffusion between two reactive fluid-solid surfaces

A picture containing silhouette, light

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Figure S3. Validation test of mass diffusion between two reactive fluid-solid surfaces: computational domain and boundary conditions.

This subsection aims to test the proposed LB model in solving mass conservation boundary conditions on the reactive fluid-solid surface. As shown in Figure S3, radii of the two circles are and , respectively. The fluid flow is not considered and the species diffusion takes places in the computational domain . The concentration at the inner boundary () is fixed as , while a first-order reaction is imposed at the outer boundary () as . The reactive boundary is similar to the mass conservation boundary condition for CO2 desublimation in Eq. (2.12) (see our paper). For such a process, the analytical solution of the steady-state concentration distribution is calculated as [S1],

|  |  |
| --- | --- |
|  | (S1) |

|  |  |
| --- | --- |
|  |  |
| (*a*)Concentration along the line | (*b*) Concentration at the outer circular surface |

Figure S4. Validation test of mass diffusion between two reactive fluid-solid surfaces: comparison of the steady-state concentration distributions with different numbers (*a*) along the line and (*b*) at the outer circular surface between the present LB simulations and the analytical solutions in Eq. (S1).

In the present LB simulations, we set and a mesh of size is used. The characteristic length and velocity are selected as and , respectively. Thus, the Damköhler number is calculated as . Under different numbers, the simulated steady-state concentration profiles along the line are presented in Figure S4, as well as values of the concentration at the outer surface . As shown, our simulated profiles are in good agreement with the analytical solutions, indicating that the proposed LB model can successfully simulate mass conservation conditions on reactive fluid-solid surfaces.

## Thermal flow in a channel with two components

After validating the sub-model for the mass conservation boundary condition, the developed LB model for conjugate heat transfer between two components is tested. The steady-state thermal flow in a channel with a vertical interface between two components is adopted, which considers both convective and conductive heat flux conservation at the interface. As constructed in [S2], Figure S5 illustrates the computational domain () and boundary conditions. In this test, two different fluids are separated by a vertical interface at , and a uniform velocity is imposed at the inlet () along the direction. For such a steady-state problem, the analytical solution for the temperature distribution is [S2],

|  |  |
| --- | --- |
|  | (S2) |

with the parameters being defined as,

|  |  |
| --- | --- |
|  | (S3) |

In LB simulations, a grid mesh of size is applied to the computational domain in Figure S5. The comparison of the steady-state temperature distribution between the analytical and LB solutions is provided in Figure S6. The two solutions are observed to match each other well. Therefore, the present LB model can accurately solve the conjugate heat transfer between two components.



Figure S5. Validation test of thermal flow in a channel with two components: computational domain and boundary conditions.

![Chart, line chart

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Figure S6. Validation test of thermal flow in a channel with two components: comparison of the steady-state temperature distribution between the present LB simulation and the analytical solution in Eq. (S2).

## Thermal flow around reactive obstacles

After the above two separate validations, this subsection further tests the present LB model for simulating the thermal flow around reactive obstacles. Figure S7 illustrates the computational domain () and boundary conditions, which are constructed as in the supplementary document of [S3]. In this test, reaction occurs on each obstacle surface to consume the injected O2 and release heat. Thus, the fluid flow, reactive fluid-solid interface, mass conservation, and conjugate heat transfer between two phases are considered simultaneously.

Graphical user interface, application

Description automatically generated

Figure S7. Validation test of thermal flow around reactive obstacles: computational domain and boundary conditions.

In this test, a mesh of size is used, and other simulation parameters are all set as in [S3]. The reaction coefficients are , , and , respectively. The initial mass fractions of O2 and CO2 are 0.22 and 0, respectively. The temperature and pressure are and 1 MPa. Thermophysical properties of the solid and the fluid phases at are, , , , , , , respectively. The characteristic length and velocity are selected as and , leading to the Peclet number 0.01, the Prandtl number 0.716, and the Solid Lewis number 0.177.

|  |  |
| --- | --- |
| Chart, line chart  Description automatically generated | Chart, histogram  Description automatically generated |
| (*a*) at | (*b*) at |
| Chart  Description automatically generated | Chart, histogram  Description automatically generated |
| (*c*) at | (*d*) at |

Figure S8. Validation test of thermal flow around reactive obstacles: comparison of the steady-state (*a*) velocity, (*b*) temperature, and (*c*)-(*d*) species mass fraction distributions along the line between the present LB simulations and the COMSOL simulations in [S3]. Profiles measured at time instants 0.002 s, 0.006 s, and 0.01 s are represented by blue, green, and red colors in (*b*)-(*d*).

Figure S8 presents profiles of the steady-state velocity, temperature, and species mass fractions at three time instants along the line . For the purpose of validation, results from the COMSOL simulator in [S3] are included. The good agreement implies our present LB model is accurate for simulating thermal flows with species transport, fluid-solid surface reaction, and conjugate heat transfer.

## Mass diffusion with dissolution and precipitation reactions

Finally, we focus on testing the VOP scheme in the proposed LB model for modelling the growth of solid phase. For this purpose, we simulate the diffusion–reaction process involving dissolution and precipitation reactions in a channel [S4]. The base case (or Case 1) in Sec. 4.1.1 of [S4] is simulated and the computational domain is plotted in Figure S9. The parameters are set as listed in Table 1 of [S4]. In this case, a two-step reaction (i.e., *Al* + *Ds* = *Bl*, *Bl* + *Cl* = *Ps*) at the reactive fluid-solid interface and the evolution (i.e., growth and consumption) of solid geometries are considered. The calculated volumes of solid *D* and solid *P* are plotted against time in Figure S10. As can be seen, the present LB model can capture the same reaction trend as in [S4]. That is, both the dissolution and precipitation rate decrease with time, indicating the system gradually approaches an equilibrium state. Moreover, by comparison, the geometrical evolutions (i.e., volume evolutions of the two solid chemical materials *D* and *P*) are observed to quantitatively match well with [S4]. It thus demonstrates the present LB model is accurate for simulating the growth of solid products.



Figure S9. Validation test of mass diffusion with dissolution and precipitation reactions: computational domain and boundary conditions.



Figure S10. Validation test of mass diffusion with dissolution and precipitation reactions: temporal evolutions of volumes for solid *D* and solid *P*. The results from [S4] are included for comparison.

**References**

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