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# Electronic Supplementary Material

# Gas-Phase Dynamics in Graphene Growth from Chemical Vapour Deposition

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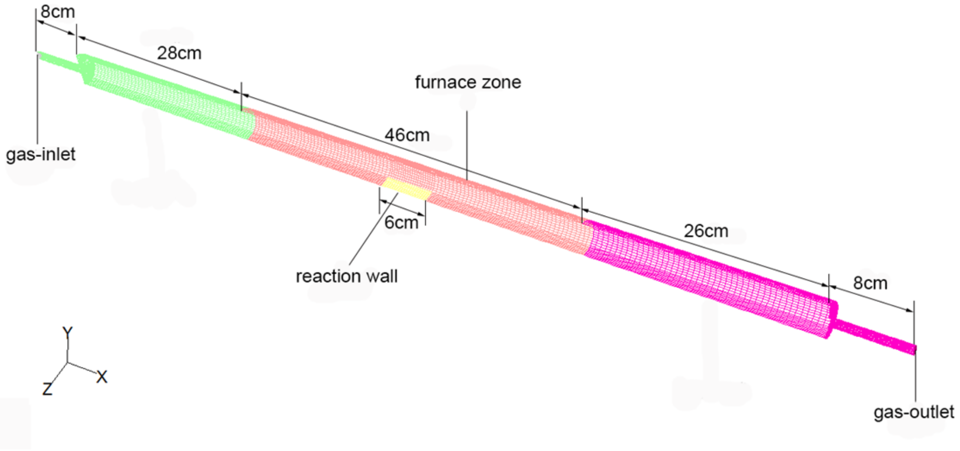

Fig. S1 Face-symmetric three-dimensional grid model. There is a reaction wall in the furnace zone.

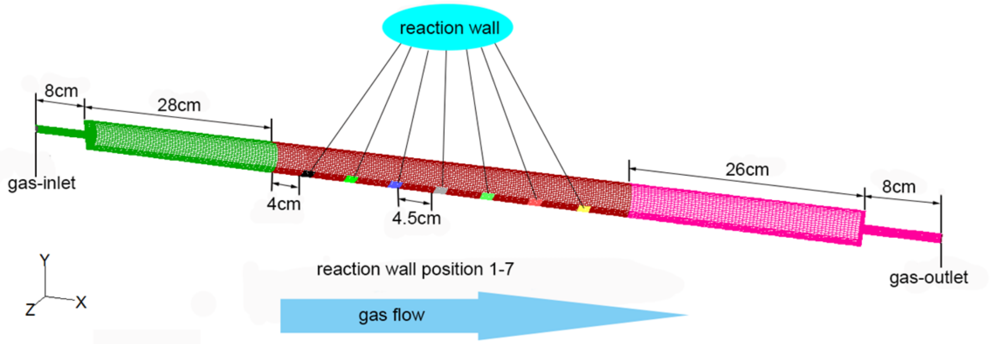

Fig. S2 Face-symmetric three-dimensional grid, where seven reaction walls are equally distributed in the furnace zone.

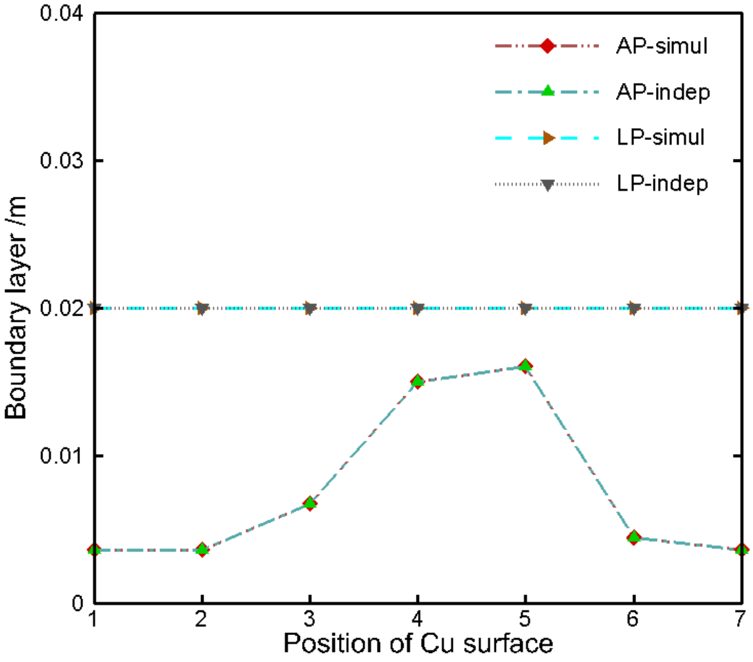

Fig. S3 Boundary layer height as a function of the position of Cu surface in the furnace zone at different pressure. The sign 'simul' represents that the reaction wall are placed at seven different positons simultaneously, and the sign 'indep' represents that the reaction wall are placed at seven different positions independently.

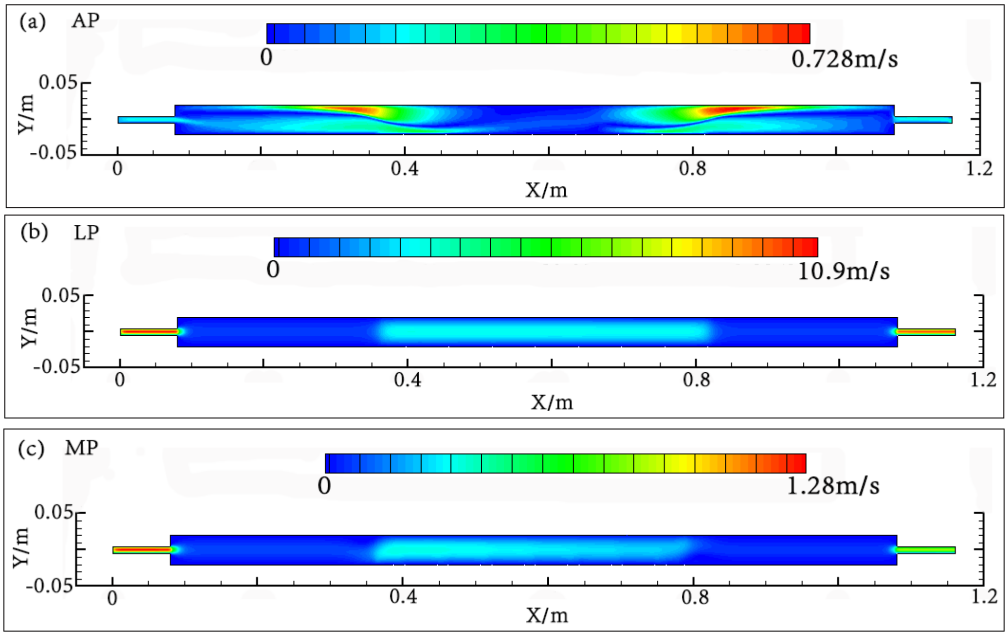

Fig. S4 Contour of velocity distribution on the symmetry plane, under (a) AP (101325 Pa), (b) LP (83 Pa), and (c) MP (2666 Pa), when seven Cu foils were put in furnace.

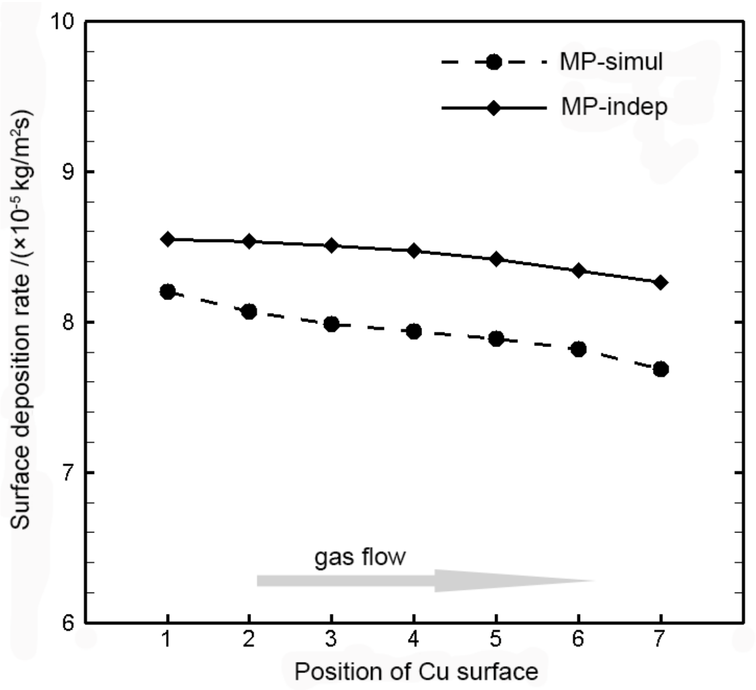

Fig. S5 Calculated surface carbon deposition rate varying with positions of the Cu surface in the furnace zone, under MP condition. The sign ‘simul’ represents that the reaction wall are placed at seven different positons simultaneously, and the sign ‘indep’ represents that the reaction wall are placed at seven different positions independently.

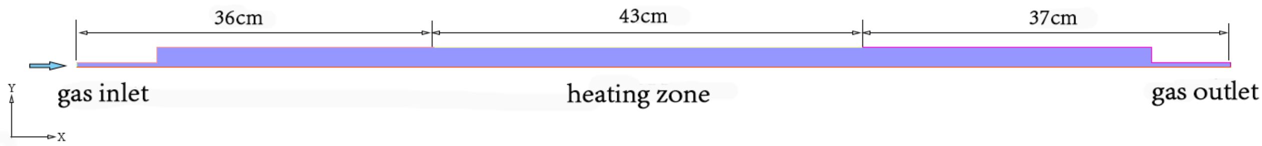

Fig. S6 Axisymmetric two-dimensional grid, without reaction wall set in the heating zone.

**Governing equations**

The governing equations of continuity and momentum/energy conservation are listed below.

$$\frac{\partial \rho}{\partial t} + \nabla \cdot (\rho V) = 0 \tag{1}$$

$$\frac{\partial \rho Y_i}{\partial t} + \nabla \cdot (\rho Y_i V) = -\nabla \cdot \rho D_{i,m} \nabla Y_i + R_i + S_i \tag{2}$$

$$\frac{\partial \rho u_i}{\partial t} + \nabla \cdot (\rho V u_i) = -\frac{\partial P}{\partial x_j} + \frac{\partial \tau_{ij}}{\partial x_i} + \rho g_i \tag{3}$$

$$\frac{\partial \rho E}{\partial t} + \nabla \cdot (V \cdot (\rho E + P)) = -\nabla \cdot (k \nabla T - \sum_i (h_i J_i)) + \nabla \cdot \tau_{ij} V + S_h \tag{4}$$

where $x_i$, $u_i$, and $g_i$ are coordinate, velocity, and gravity in the $i$ direction, respectively. $\rho$, $V$, $P$, $T$ and $E$ are the density, velocity, pressure, temperature and total energy of the gas mixture, respectively. $Y_i$ is the mass fraction of species $i$, $D_{i,m}$ is the diffusion coefficients of each species $i$ among gas mixture, $R_i$ is the net rate of production of species by chemical reaction and $S_i$ is the rate of production by other sources. $\tau_{ij}$ is the stress tensor, $k$ is the thermal conductivity, $h_i$ and $J_i$ are the diffusion flux and enthalpy of the $i$ th species. $S_h$ is the enthalpy source term due to the chemical reaction.

### Analytical approximations of $\sigma_{ij}$ and $\Omega_D$

Inter-molecular potential $\psi$ between two molecules with a distance $r$ is given by

$$\psi = 4\varepsilon\left[\left(\frac{\sigma}{r}\right)^{12} - \left(\frac{\sigma}{r}\right)^{6}\right] \tag{5}$$

where $\varepsilon$ and $\sigma$ as the characteristic Lennard-Jones energy and length, respectively.

The combination rule is $\sigma_{ij} = \dfrac{\sigma_i + \sigma_j}{2}$ and $\varepsilon_{ij} = (\varepsilon_i \varepsilon_j)^{1/2}$

The dimensionless diffusion collision integral $\Omega_D$ is a function of $kT / \varepsilon_{ij}$, which is given by

$$\Omega_D = \frac{A}{(T^*)^B} + \frac{C}{\exp(DT^*)} + \frac{E}{\exp(FT^*)} + \frac{G}{\exp(HT^*)} \tag{6}$$

where $T^* = kT / \varepsilon_{ij}$, $A = 1.06036$, $B = 0.15610$, $C = 0.19300$, $D = 0.47635$, $E = 1.03587$, $F = 1.52996$, $G = 1.76474$, and $H = 3.89411$.

The value of $\sigma(CH_4)$, $\sigma(H_2)$, $\sigma(Ar)$, $\varepsilon(CH_4)$, $\varepsilon(H_2)$ and $\varepsilon(Ar)$ can be obtained by referring B. E. Poling, et al.(2001), which are shown in Table S1.

Table S1. Value of $\sigma$ and $\varepsilon / k$ of each gas species.

| | $CH_4$ | $H_2$ | $Ar$ |
|---|---|---|---|
| $\sigma$ (Å) | 3.758 | 2.827 | 3.542 |
| $\varepsilon / k$ ($K$) | 148.6 | 59.7 | 93.3 |

### More computational details

Models: Energy Equation must be solved, and Laminar was selected as Viscous Model, Species Transport was choose as Species Model.

| Mixture: | |
|---|---|
| Density | ideal-gas |
| Specific Heat | mixing-law |
| Thermal Conductivity | ideal-gas-mixing-law |
| Viscosity | ideal-gas-mixing-law |
| Mass Diffusivity | multicomponent |
| Thermal Diffusion Coefficient | kinetic-theory |

Boundary Conditions: gas-inlet Mass-Flow Inlet Mass flow rate and its direction was specified. Mass Fractions of gas species were specified.

The temperature of Reaction wall and furnace wall were set to the same. For the reaction wall, the surface reaction model is activated.

Gas-outlet Pressure Outlet

Operating Conditions Operating pressure was specified according to the pressure of specific experiment.