COMSOL Multiphysics® Simulations of Graphene Chemical Vapor Deposition (CVD) Growth

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Introduction: Chemical vapor deposition (CVD) is a promising effective method for synthesis of graphene films. CVD graphene film is obtained from hydrocarbon species such as CH_4 through complex catalytic chemical reactions on the surface of the catalyst. Therefore, studying the catalytic reaction kinetics is essential process for understanding the thermal decomposition rate of methane on catalyst surface as well as the reaction mechanisms. In this study, COMSOL Multiphysics is used to investigate graphene chemical vapor deposition process on Cu surface. Also, the final thickness of graphene film was calculated.

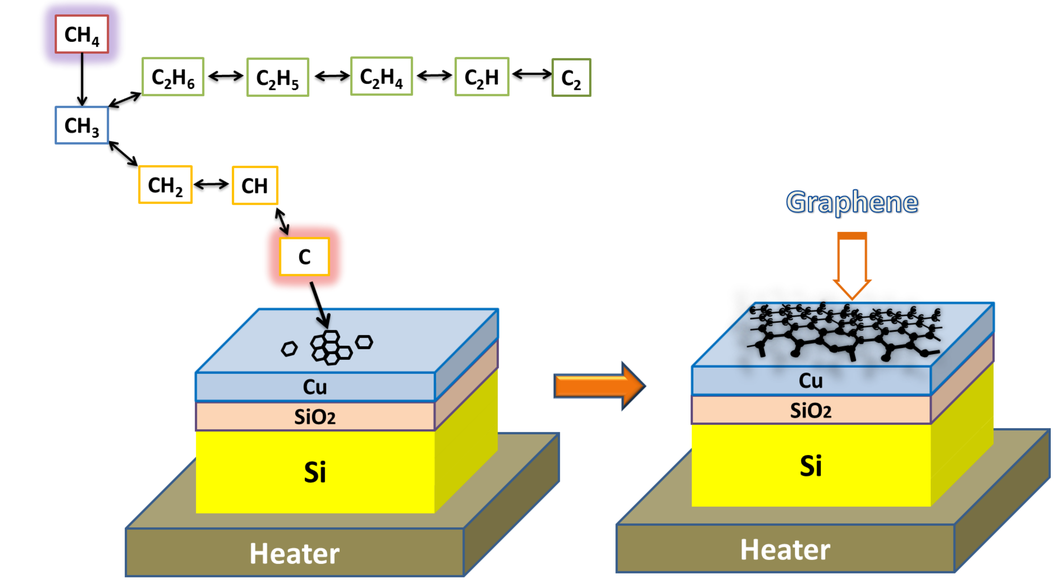

Figure 1. Graphene CVD growth process on Cu thin film.

Computational Methods: Our COMSOL model uses Laminar Flow, Heat Transfer, and Heavy Species Transport application in order to investigate CH_4 decomposition reactions encountered in the CVD chamber as well as graphene growth on Cu surface.

$$CH_4 + 5S_{(Cu)} \rightarrow graphene_{(a)} + 2H_{2(g)}$$

$$r_i = k_i^f \prod_{k=1}^{K} c_k^{v_{ki}^f}$$

$$\frac{\partial \rho}{\partial t} + \nabla.(\rho \boldsymbol{u}) = 0$$

$$\rho \frac{\partial \boldsymbol{u}}{\partial t} + \rho \boldsymbol{u}.\nabla u = -\nabla \boldsymbol{p} + \nabla.\left(\mu(\nabla \boldsymbol{u} + (\nabla \boldsymbol{u})^T) - \frac{2}{3}\mu(\nabla.\boldsymbol{u})\boldsymbol{I}\right)$$

$$\rho C_p \frac{\partial T}{\partial t} + \rho C_p \boldsymbol{u}.\nabla T = \nabla.(\kappa \nabla T) + Q$$

$$\boldsymbol{u} = -\frac{M_f}{\rho}\boldsymbol{n} \qquad M_f = \sum_{k=1}^{K_g} M_k \dot{s}_k \qquad \dot{s}_k = \sum_{i=1}^{I} V_{ki} r_i$$

Results: Graphene direct deposition onto Cu film is explored in this research. To explain, CH_4 is directly decomposed on Cu surface leading to carbon adsorption. Then, the adsorb carbon atoms form graphene film.

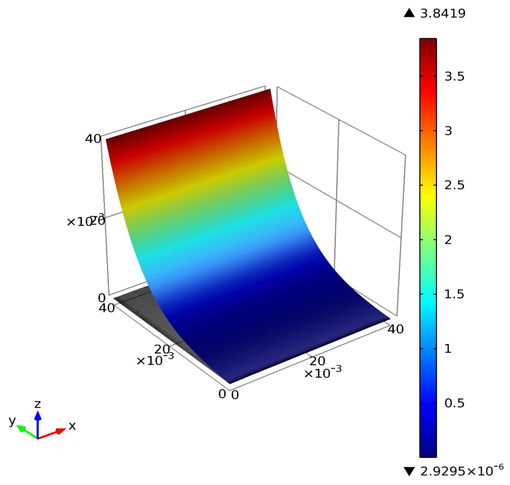

Figure 3. Accumulated graphene growth height (Å) after 1300 sec at H_2 fraction equals 0.001.

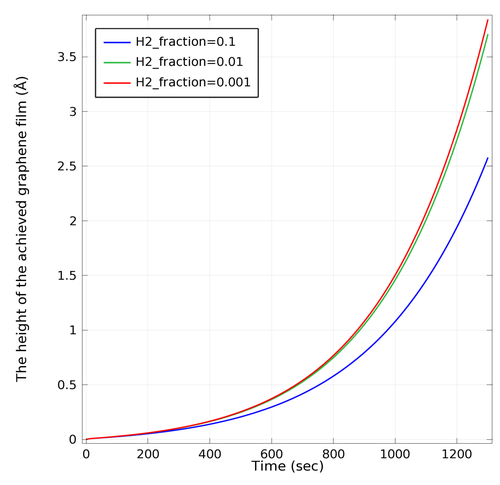

Figure 4. The effect of H_2 fraction on the height of the achieved graphene film.

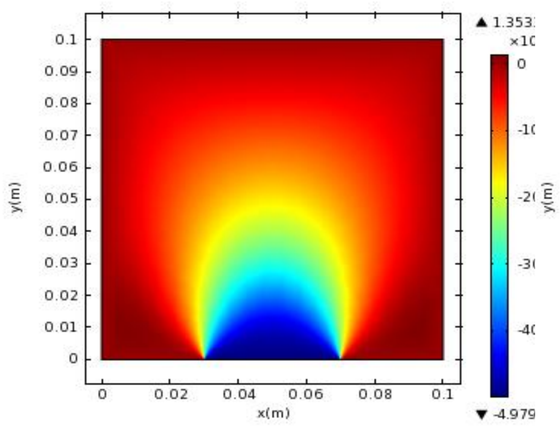

Figure 5. The y- component of the mass averaged velocity at=10 s.

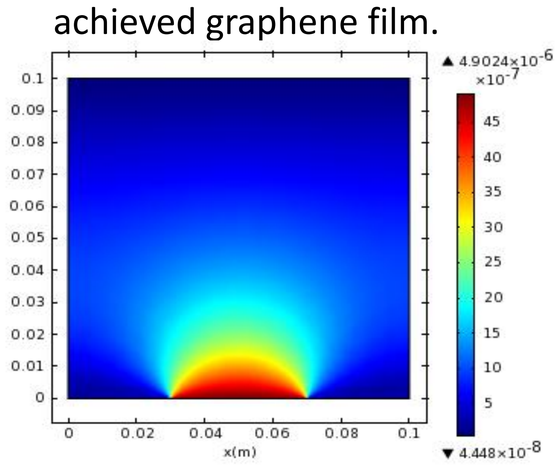

Figure 6. The y- component of the diffusion velocity for hydrogen at=10 s.

Conclusions: In this paper, graphene CVD growth on Cu thin film was modeled using COMSOL. To explain, Laminar Flow, Heat Transfer, and Heavy Species Transport application were utilized to simulate graphene chemical vapor deposition (CVD) growth process on Cu. Monolayer graphene film was achieved by methane decomposition at 700 ° C where the total pressure equals ~ 0.01 torr. This model can be extended to simulate graphene growth using different catalysts as well as different carbon precursors.

References

1. Reina A, Kong J. Graphene growth by CVD methods. In: Graphene nanoelectronics. Springer; 2012.
2. Li X, Magnuson CW, Venugopal A, Tromp RM, Hannon JB, Vogel EM, Colombo L, Ruoff RS. Large-area graphene single crystals grown by low-pressure chemical vapor deposition of methane on copper. J Am Chem Soc 2011;133(9):2816-9.
3. Kim H, Saiz E, Chhowalla M, Mattevi C. Modeling of the self-limited growth in catalytic chemical vapor deposition of graphene. New Journal of Physics 2013;15(5):053012.