SIMULATION OF CARBON NANOPARTICLE FORMATION DURING RAPID COOLING OF CARBON GAS

S. A. Gubin[1,2], A. V. Kudinov[1], I. V. Maklashova[1], and Yu. A. Bogdanova[1]

[1]National Research Nuclear University MEPhI, 31 Kashirskoe Sh., Moscow 115409, Russian Federation
[2]N. N. Semenov Federal Research Center for Chemical Physics of the Russian Academy of Sciences, 4 Kosygin Str., Moscow 119991, Russian Federation

Abstract: On the basis of quasi-equilibrium thermodynamics and molecular dynamics modeling of the process of nanoparticle formation during rapid cooling of carbon gas heated to a high temperature at constant density, the possible pathway for the synthesis of nanocarbon particles is identified through condensation from the gas phase. Thermodynamic calculations take into account the increased enthalpy of formation for carbon nanoparticles. Based on the results of molecular dynamics calculations, three parameterizations of reaction-force fields (ReaxFF-CHO, ReaxFF-c2013, and ReaxFF-PAH) are recommended for molecular dynamics modeling of nanocarbon particle formation.

Keywords: molecular dynamics modeling; thermodynamic calculation; nanocarbon particles

DOI: 10.30826/CE22150101

Figure Captions

Figure 1 Thermodynamic calculation of carbon gas cooling at $\rho = 0.05$ kg/m^3

Figure 2 Snapshots of carbon nanostructure formation at $\rho = 0.05$ kg/m^3 in time: (*a*) $\tau = 0$ ns; (*b*) 0.60; (*c*) 0.65; (*d*) 0.70; (*e*) 0.80; (*f*) 0.85; (*g*) 0.98; (*h*) 1.06, and (*i*) $\tau = 2.50$ ns

Table Caption

Temperatures of the beginning and end of carbon gas condensation at various densities

Acknowledgments

The work was supported by the Ministry of Science and Higher Education of the Russian Federation (Government Order Project No. 0723-2020-0036).

References

1. Howard, J. B., J. T. McKinnon, Y. Makarovsky, A. L. Lafleur, and M. E. Johnson. 1991. Fullerenes C_{60} and C_{70} in flames. *Nature* 352(6331):139–141. doi: 10.1038/352139a0.
2. Chen, X., F. Deng, J. Wang, H. Yang, G. Wu, X. Zhang, J. Peng, and W. Li. 2001. New method of carbon onion growth by radio-frequency plasma-enhanced chemical vapor deposition. *Chem. Phys. Lett.* 336(3-4):201–204. doi: 10.1016/S0009-2614(01)00085-9.
3. Niwase, K., T. Homae, K. Nakamura, and K. Kondo. 2002. Generation of giant carbon hollow spheres from C60 fullerene by shock-compression. *Chem. Phys. Lett.* 362(1):47–50. doi: 10.1016/S0009-2614(02)00997-1.
4. Kang, J., J. Li, X. Du, C. Shi, N. Zhao, and P. Nash. 2008. Synthesis of carbon nanotubes and carbon onions by CVD using a Ni/Y catalyst supported on copper. *Mat. Sci. Eng. A — Struct.* 475(1-2):136–140. doi: 10.1016/j.msea.2007.04.027.
5. Bgasheva, T. V., P. S. Vervikishko, A. M. Frolov, and M. A. Sheyndlin. 2016. Kristallizatsiya ugleroda iz para pri davleniyakh do 0,6 GPa [Crystallization of the carbon vapor at pressures up to 0.6 GPa]. *Trudy Konferetsii-konkursa molodykh uchenykh* [Conference-Competition of Young Physicists Proceedings] 22:88–90.
6. Zheng, G., S. Irle, and K. Morokuma. 2005. Performance of the DFTB method in comparison to DFT and semiempirical methods for geometries and energies of C_{20}–C_{86} fullerene isomers. *Chem. Phys. Lett.* 412(1-3):210–216. doi: 10.1016/j.cplett.2005.06.105.
7. Qian, H.-J., A. C. van Duin, K. Morokuma, and S. Irle. 2011. Reactive molecular dynamics simulation of fullerene combustion synthesis: ReaxFF vs DFTB potentials. *J. Chem. Theory Comput.* 7(7):2040–2048. doi: 10.1021/ct200197v.
8. Dozhdikov, V., A. Y. Basharin, P. Levashov, and D. Minakov. 2017. Atomistic simulations of the equation of state and hybridization of liquid carbon at a temperature of

6000 K in the pressure range of 1–25 GPa. *J. Chem. Phys.* 147(21):214302. doi: 10.1063/1.4999070.

9. Van Duin, A. C. T., S. Dasgupta, F. Lorant, and W. A. Goddard. 2001. ReaxFF: A reactive force field for hydrocarbons. *J. Phys. Chem. A* 105(41):9396–9409. doi: 10.1021/jp004368u.
10. Galiullina, G., N. Orekhov, and V. Stegailov. 2016. Nucleation of carbon nanostructures: Molecular dynamics with reactive potentials. *J. Phys. Conf. Ser.* 774(1):012033. doi: 10.1088/1742-6596/774/1/012033.
11. Chenoweth, K., A. C. T. van Duin, and W. A. Goddard. 2008. ReaxFF reactive force field for molecular dynamics simulations of hydrocarbon oxidation. *J. Phys. Chem. A* 112(5):1040–1053. doi: 10.1021/jp709896w.
12. Mueller, J. E., A. C. van Duin, and W. A. Goddard, III. 2010. Development and validation of ReaxFF reactive force field for hydrocarbon chemistry catalyzed by nickel. *J. Phys. Chem. C* 114(11):4939–4949. doi: 10.1021/jp9035056.
13. Ashraf, C., and A. C. van Duin. 2017. Extension of the ReaxFF combustion force field toward syngas combustion and initial oxidation kinetics. *J. Phys. Chem. A* 121(5):1051–1068. doi: 10.1021/acs.jpca.6b12429.
14. Mao, Q., Y. Ren, K. H. Luo, and A. C. T. van Duin. 2017. Dynamics and kinetics of reversible homo-molecular dimerization of polycyclic aromatic hydrocarbons. *J. Chem Phys.* 147:244305. doi: 10.1063/1.5000534.
15. Victorov, S. B., H. El-Rabii, S. A. Gubin, I. V. Maklashova, and Y. A. Bogdanova. 2010. An accurate equation-of-state model for thermodynamic calculations of chemically reactive carbon-containing systems. *J. Energ. Mater.* 28:35–49. doi: 10.1080/07370652.2010.491496.
16. Viecelli, J. A., S. Bastea, J. N. Glosli, and F. H. Ree. 2001. Phase transformations of nanometer size carbon particles in shocked hydrocarbons and explosives. *J. Chem. Phys.* 115(6):2730–2737. doi: 10.1063/1.1386418.
17. Gubin, S. A., E. I. Dzhelilova, and I. V. Maklashova. 2014. Vliyanie formy i razmera nanochastits na fazovuyu diagrammu ugleroda [Influence of the shape and size of nanoparticles on the phase diagram of carbon]. *Goren. Vzryv (Mosk.) — Combustion and Explosion* 7:226–229.
18. Von Helden, G., N. G. Gotts, and M. T. Bowers. 1993. Experimental evidence for the formation of fullerenes by collisional heating of carbon rings in the gas phase. *Nature* 363(6424):60–63. doi: 10.1038/363060a0.
19. Liu, L., Y. Liu, S. V. Zybin, H. Sun, and W. A. Goddard, III. 2011. ReaxFF-lg: Correction of the ReaxFF reactive force field for London dispersion, with applications to the equations of state for energetic materials. *J. Phys. Chem. A* 115:11016–11022. doi: 10.1021/jp201599t.
20. Srinivasan, S. G., A. C. T. van Duin, and P. Ganesh. 2015. Development of a ReaxFF potential for carbon condensed phases and its application to the thermal fragmentation of a large fullerene. *J. Phys. Chem. A* 119:571–580. doi: 10.1021/jp510274e.
21. LAMMPS — a flexible simulation tool for particle-based materials modeling at the atomic, meso, and continuum scales. Available at: https://www.lammps.org/ (accessed November 15, 2021).
22. Ostroumova, G., N. Orekhov, and V. Stegailov. 2019. Reactive molecular-dynamics study of onion-like carbon nanoparticle formation. *Diam. Relat. Mater.* 94:14–20. doi: 10.1016/j.diamond.2019.01.019.
23. Yasuoka, K., and M. Matsumoto. 1998. Molecular dynamics of homogeneous nucleation in the vapor phase. I. Lennard–Jones fluid. *J. Phys. Chem.* 109(19):8451–8462. doi: 10.1063/1.477509.
24. Bundy, F. P. 1989. Pressure–temperature phase diagram of elemental carbon. *Physica A* 156(1):169–178. doi: 10.1016/0378-4371(89)90115-5.

Received December 24, 2021

Contributors

Gubin Sergey A. (b. 1945) — Doctor of Science in physics and mathematics, professor, head of department, National Research Nuclear University MEPhI, 31 Kashirskoe Sh., Moscow 115409, Russian Federation; chief research scientist, N. N. Semenov Federal Research Center for Chemical Physics of the Russian Academy of Sciences, 4 Kosygin Str., Moscow 119991, Russian Federation; sagubin@mephi.ru

Kudinov Andrey V. (b. 1996) — postgraduate student, National Research Nuclear University MEPhI, 31 Kashirskoe Sh., Moscow 115409, Russian Federation; swen379@gmail.com

Maklashova Irina V. (b. 1975) — senior lecturer, National Research Nuclear University MEPhI, 31 Kashirskoe Sh., Moscow 115409, Russian Federation; ivmaklashova @mephi.ru

Bogdanova Youlia A. (b. 1983) — Candidate of Science in physics and mathematics, senior lecturer, National Research Nuclear University MEPhI, 31 Kashirskoe Sh., Moscow 115409, Russian Federation; bogdanova.youlia@bk.ru