Diamond Formation in Double-Shocked Epoxy to 150 GPa

M. C. Marshall,¹ M. G. Gorman,² D. N. Polsin,^{1,3} J. H. Eggert,² M. K. Ginnane,^{1,3} J. R. Rygg,^{1,3,4} G. W. Collins,^{1,3,4} and L. D. Leininger²

¹Laboratory for Laser Energetics, University of Rochester
²Lawrence Livermore National Laboratory
³Department of Mechanical Engineering, University of Rochester
⁴Department of Physics and Astronomy, University of Rochester

The formation of diamond from carbon-based polymers, compounds, or other carbon polymorphs (i.e., graphite) under extreme pressures has been actively researched for many decades.^{1–12} Diamond can form by extreme heating and compression of some plastics,¹ methane,^{2,3} and explosive materials.^{10,12} For example, cubic diamond was detected from double-shocked polystyrene (CH) at 139 to 159 GPa using *in-situ* x-ray diffraction in experiments at the Linac Coherent Light Source (LCLS), indicating that breaking of the carbon and hydrogen bonds and restructuring of the carbon into diamond can occur over only nanosecond time scales.¹ The results presented here indicate that cubic diamond also forms from Stycast 1266 epoxy (C:H:Cl:N:O \approx 27:38:1:1:5) (Ref. 13) doubly shocked to 80 and 148 GPa. These results demonstrate that the chemical and thermodynamic conditions inside ice giant planets, which have inner ice layers dominated by CH₄, NH₃, and H₂O, are suitable for diamond formation.

Two experiments were conducted on the OMEGA EP Laser System,¹⁴ where laser beams were used to shock compress the target comprising an epoxy sample and a LiF window with a reflective Ti coating between them to measure the interface velocity using a velocity interferometer system for any reflector (VISAR).¹⁵ When the shock wave is incident on the higher-impedance LiF interface, a return shock (reshock) is launched back through the epoxy, which approximately doubles the pressure. The target was probed with x rays, and the resulting diffraction pattern was measured using the powder x-ray diffraction image-plate diagnostic¹⁶ when the epoxy was in the reshocked state. Diffraction consistent with cubic diamond was observed for both shots (Fig. 1). The pressure and temperature state during the x-ray exposure time were determined using hydrodynamic simulations matched to the measured epoxy/LiF interface velocity.

The results are compared to previous works in Fig. 1(a), where CH (Ref. 1), polyethylene (CH₂) (Ref. 5), methane (CH₄), (Refs. 2 and 3), and methane hydrate (MH) (Ref. 4) were also probed *in situ* at high pressures and temperatures to study diamond formation. Diamond formation from methane and methane hydrate is observed over ~10 to 80 GPa and ~2000 to 4000 K in laser-heated diamond-anvil cell experiments, where the samples are compressed and heated over seconds to hours.^{2–4} At these conditions and compression rates, diamond formation is largely temperature dependent and the temperature threshold at which it occurs is lowered by the addition of oxygen as suggested by the methane hydrate experiments.⁴ Diamond formation is not observed in singly shocked CH (Ref. 1) and CH₂ (Ref. 5) when they are compressed over nanoseconds at similar temperatures and pressures. While diamond still does not form in doubly shocked CH₂, it does form from doubly shocked CH and epoxy at ~80 to 200 GPa pressures and ~2000 to 6000 K temperatures. Diamond formation at these fast nanosecond times scales is not purely pressure or temperature dependent and is affected by the initial material composition and thermodynamic compression path (e.g., single versus double shock). These differences in behavior among all the experiments collectively suggest that the kinetics associated with the vastly different time scales, the thermodynamic path, and the chemical composition of the initial material play an important role in diamond formation at extreme conditions.



Figure 1

(a) Phase diagram showing diamond-formation results from compressed and heated epoxy, polystyrene (CH), polyethylene (CH₂), methane hydrate (MH), and methane (CH₄). Data points on singly shocked CH and CH₂ lie close to the epoxy Hugoniot (*SESAME* 7602). Data points from this work and all points above 120 GPa on CH and CH₂ are on doubly shocked samples. (b) Lattice *d*-spacing results from this work compared to predictions for compressed diamond (spanning the cold curve¹⁷ to the melt curve¹⁸ to encompass all possible temperature effects) and shocked LiF.

This material is based upon work supported by the Department of Energy National Nuclear Security Administration under Award Number DE-NA0003856, the University of Rochester, and the New York State Energy Research and Development Authority. This work was performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory (LLNL) under LLNL's Laboratory Directed Research and Development (LDRD) Program under Grant No. 18-SI-004. Lawrence Livermore National Laboratory is operated by Lawrence Livermore National Security, LLC, for the U.S. Department of Energy, National Nuclear Security Administration under Contract DE-AC52-07NA27344.

- 1. D. Kraus et al., Nat. Astron. 1, 606 (2017).
- 2. L. R. Benedetti et al., Science 286, 100 (1999).
- 3. H. Hirai et al., Phys. Earth Planet. Inter. 174, 242 (2009).
- 4. H. Kadobayashi et al., Sci. Rep. 11, 8165 (2021).
- 5. N. J. Hartley et al., Sci. Rep. 9, 4196 (2019).
- 6. D. Kraus et al., Nat. Commun. 7, 10970 (2016).
- 7. E. B. Watkins et al., Sci. Rep. 12, 631 (2022).
- 8. D. G. Morris, J. Appl. Phys. 51, 2059 (1980).

- 9. F. H. Ree, J. Chem. Phys. 70, 974 (1979).
- 10. M. van Thiel and F. H. Ree, J. Appl. Phys. 62, 1761 (1987).
- 11. W. J. Nellis et al., J. Chem. Phys. 75, 3055 (1981).
- 12. J. B. Donnet et al., Diam. Relat. Mater. 9, 887 (2000).
- 13. We provide only an estimated C:H:Cl:N:O ratio for the final cured Stycast 1266 mixture, obtained by combining the listed components in the safety data sheets for Parts A and B by the quoted mix ratios and assuming a closed system. The exact composition of the Stycast 1266 Part A and B may vary from the quoted percentages or are trade secrets.
- 14. D. D. Meyerhofer et al., J. Phys.: Conf. Ser. 244, 032010 (2010).
- 15. P. M. Celliers et al., Rev. Sci. Instrum. 75, 4916 (2004).
- 16. J. R. Rygg et al., Rev. Sci. Instrum. 83, 113904 (2012).
- 17. A. Dewaele et al., Phys. Rev. B 77, 094106 (2008).
- 18. X. Wang, S. Scandolo, and R. Car, Phys. Rev. Lett. 95, 185701 (2005).