A Systematic Analysis of Amorphous Boron Nitride Films using Gaussian Approximation Potentials

Onurcan Kaya[1,2], Ivan Cole[2], and Stephan Roche[1,3]
[1]Catalan Institute of Nanoscience and Nanotechnology (ICN2), Bellaterra, 08193, Barcelona, Spain
[2]School of Engineering, RMIT University, Melbourne, VIC, 3001, Australia
[3]ICREA Institucio Catalana de Recerca i Estudis Avancats, 08010 Barcelona, Spain
onurcan.kaya@icn2.cat

Abstract

Amorphous boron nitride compounds present a great potential for several applications due to their superior stability, good mechanical properties and ultralow dielectric constant. Moreover, they can be grown at lower temperatures (65 – 400 ºC) than their crystalline counterparts over large areas and on various substrates [1-3]. However, the morphology, properties and performance of the aBN films and devices severely depend on the growth conditions. The high number of parameters and diverse possible structures make an exploration solely based on experiments very expensive and time-consuming. Hence, a systematic theoretical characterization of aBN structures is needed to understand their full potential and determine the best possible approach to produce aBN morphologies for several applications.

Atomistic simulation methods such as DFT and MD simulations with empirical interatomic potentials have been deployed to understand the diverse structure of the material and thermal, mechanical and electronic properties of the material. However, the amorphous nature of the material limits the accuracy and usefulness of these simulations. While empirical interatomic potentials are fast and efficient to simulate the aBN films, they are not accurate enough to describe the local environment of amorphous materials. On the other hand, DFT simulations can provide very detailed and accurate picture of the material, however, they are severely limited to the small system sizes, which unfortunately cannot represent the whole amorphous structure. Machine learning interatomic potentials can be a bridge between these two realms, they can offer DFT-level accuracy with a much lower cost. Gaussian approximation potentials (GAP) use Gaussian Process Regression (GPR) to learn local atomic properties through descriptors that transforms Cartesian atomic coordinates into stable representations to reach DFT level accuracy with a considerably low cost [4, 5]. This approach has been effectively employed to model carbon [4], silicon [5], and BN compounds [6-8] among others.

Here, we will present a systematic theoretical analysis to screen out possible realistic morphologies as a function of growth parameters, such as temperature, quenching rate and presence of dopants, and their corresponding thermal and mechanical properties using classical molecular dynamics simulations. We ensure the reliability of results by introducing Gaussian Approximation Potentials which are trained on a large dataset of atomic structures generated by ab-initio calculations. We found that some level of dopants and growth parameters cause a significant change in structural properties of aBN, which is strongly reflected in the resulting mechanical properties and stability of the compounds. The extensive simulations of a large quantity of possible structures presented in this work will guide experimental research and provide trends of scaling behavior as a function of experimentally controllable parameters.

References

[1] Hong, S et al. Nature 582, 511–514 (2020).
[2] Glavin, N. R., et al., Adv. Func. Mat. (2016)
[3] Chen, C. Y., et al., arxiv: 2312.09136 (cond-mat).
[4] Bartok, A., et al., Phys.Rev.Let., 104, 136403 (2010).
[5] Unruh, D., et al. Phys. Rev. Mat. 6, 065603 (2022).
[6] Kaya, O., et al. Nanoscale Horizons 8, 361–367.
[7] Kaya, O. et al. arXiv:2310.18102 (cond-mat).
[8] Kaya, O. et al. arXiv:2402.01251 (cond-mat).

Figures

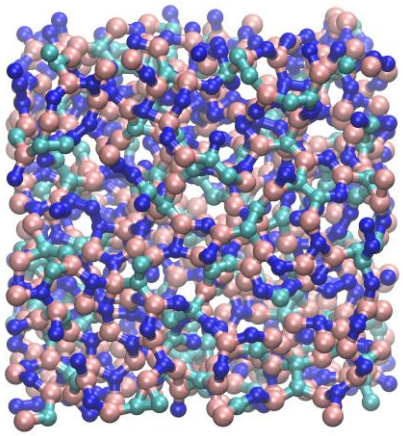

Figure 1. C contaminated aBN film [6].

Acknowledgements

This project is conducted under the REDI Program, a project that has received funding from the European Union's Horizon 2020 research and innovation programme under the Marie Skłodowska-Curie grant agreement no. 101034328. This project is also supported by Project PCI2021-122092-2A funded by MICIU/AEI /10.13039/501100011033 and by the European Union NextGenerationEU/PRTR. Simulations were performed at the Center for Nanoscale Materials, a U.S. Department of Energy Office of Science User Facility, supported by the U.S. DOE, Office of Basic Energy Sciences, under Contract No. DE-AC02-06CH11357. Additional computational support was received from the King Abdullah University of Science and Technology-KAUST (Supercomputer Shaheen II Cray XC40), RMIT AWS Cloud Supercomputing (RACE) and Texas Advanced Computing Center (TACC) at The University of Texas at Austin.