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Multiscale Modeling of Graphite/CNT/Epoxy Hybrid Composites

Gregory Odegard
MICHIGAN TECHNOLOGICAL UNIVERSITY

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Final Report

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Final Report

Multiscale Modeling of Graphite/CNT/Epoxy Hybrid Composites

Grant FA9550-13-1-0030

PI: Gregory M. Odegard
Michigan Technological University

Introduction

This project was inspired from the AFOSR-sponsored workshop "Nanotube Assemblages for Structures" that was held on the Georgia Tech campus April 17-18, 2012. During the workshop, representatives from Boeing, Lockheed Martin, Air Force Research Laboratory, and NASA agreed that one of the major drawbacks to incorporating carbon nanotubes (CNTs) in aerospace structural design was the lack of knowledge regarding the load transfer in CNT/epoxy composites. Specifically, they wanted to know what types of epoxy (di-functional, tri-functional, tetra-functional) formed the best bonds with CNT reinforcement. Thus, it was an ideal AFOSR-sponsored project that had commercial relevance in the aerospace industry.

The objective of this project, as outlined in the original proposal, was

Efficiently and accurately predict the influence of epoxy resin type on the performance of graphite/carbon nanotube/epoxy hybrid composites using molecular dynamics simulation

It was proposed that this objective would be met with the following individual tasks:

1. Develop molecular dynamics models of carbon nanotube/epoxy composites
2. Predict thermo-mechanical properties of carbon nanotube/epoxy composites
3. Use micromechanical modeling to predict thermo-mechanical properties of unidirectional graphite/carbon nanotube/epoxy
4. Use micromechanical modeling to predict thermo-mechanical properties of woven graphite/carbon nanotube/epoxy

The primary PhD student that was supported on this project was Mr. Matthew Radue, who is currently finishing his research with an expected graduating date of August 2016. As part of this project, Matt spent two summers at AFRL working with Dr. Ajit Roy, Vikas Varshney, and Jeff Baur. In addition to his accomplishments on the proposed research, he achieved much with the AFRL team, all of which is summarized below.

Publications

The following publications/conference presentations were made possible through this grant and include an acknowledged of the financial support:

1. Radue, M.S., B.D. Jensen, S. Gowtham, G.M. Odegard, D.R. Klimek, J.A. King "Applying Reactive Molecular dynamics to Predict and Compare the Mechanical Response of Di-, Tri-, and Tetra-functional Resin Epoxies", *American Society for Composites 30th Technical Conference*; East Lansing, MI; September 28-30, 2015
2. Radue, M.S., V. Varshney, J.W. Baur, A.K. Roy, G.M. Odegard, "Molecular Modeling of Crosslinked High-Temperature Bismaleimide Resins: Matrimid-5292", *31st American Society of Composites Technical Conference*, Williamsburg, VA, September 19-22, 2016
3. Varshney, V., M.S. Radue, J.W. Baur, A.K. Roy, G.M. Odegard, "Molecular Modeling of Crosslinking Reactions in High-Temperature Bismaleimide Resins: Matrimid-5292", *251st American Chemical Society National Meeting & Exposition*, San Diego, CA, March 13-17, 2016
4. Odegard, G.M., B.D. Jensen, S. Gowtham, J. Wu, J. He, Z. Zhang, "Predicting Mechanical Response of Crosslinked Epoxy using ReaxFF", *Chemical Physics Letters*, Vol. 591, pp. 175 - 178 (2014)
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10. Hadden, C.M., D.R. Klimek-McDonald, E.J. Pineda, J.A. King, A.M. Reichanadter, I. Miskioglu, S. Gowtham, G.M. Odegard, "Mechanical Properties of Graphene Nanoplatelet/Carbon Fiber/Epoxy Hybrid Composites: Multiscale Modeling and Experiments", *20th International Conference on Composite Materials*; Copenhagen, Denmark; July 19-25, 2015

11. Hadden, C.M., D.R. Klimek-McDonald, E.J. Pineda, J.A. King, A.M. Reichanadter, I. Miskioglu, S. Gowtham, G.M. Odegard, "Mechanical Properties of Graphene Nanoplatelet/Carbon Fiber/Epoxy Hybrid Composites: Multiscale Modeling and Experiments", *American Society for Composites 30th Technical Conference*; East Lansing, MI; September 28-30, 2015
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These publications include two journal articles, nine conference papers, and a NASA TM. In addition to these, there are three manuscripts that are currently being prepared for journal publication. All of these publications are available upon request (there is a 50 MB file limit to this report, which precludes the inclusion of these papers). These manuscripts can be categorized into four different topic areas, each of which is detailed below.

Modeling of Di-, Tri-, and Tetra-functional epoxy/CNT composites

This work is covered in publication #1 listed above, as well as in two upcoming journal manuscripts that are currently in preparation. This work represents the core of the proposed research, which was lead by Mr. Matt Radue, the supported PhD student. This work most directly achieved the proposed objectives, and ultimately answered the question posed by the aerospace industry at the 2012 workshop. The three epoxy systems considered are shown in Figure 1, and the MD model of the CNT/epoxy system is shown in Figure 2.

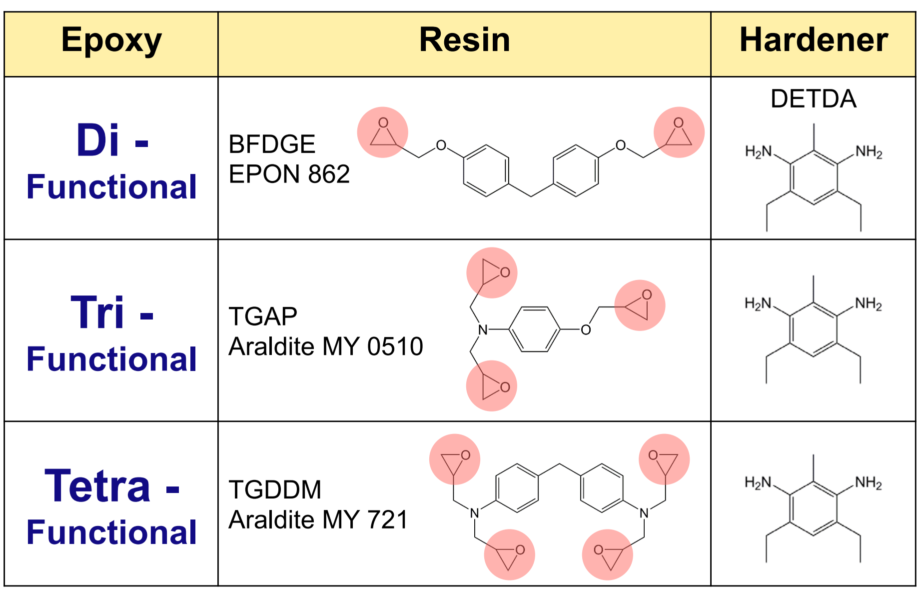

Figure 1 – Three epoxy systems simulated with molecular dynamics

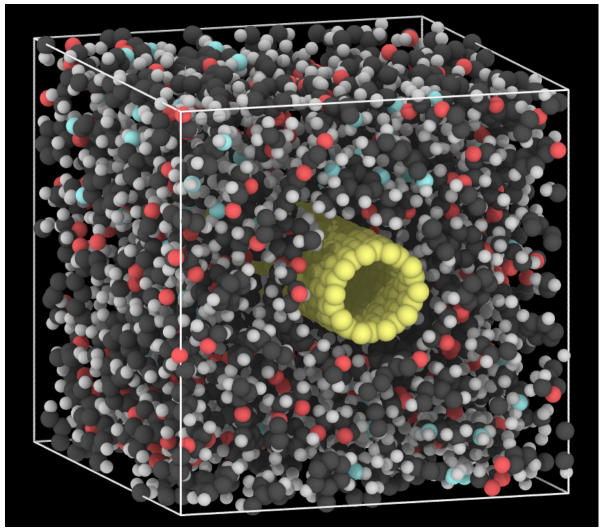

Figure 2 – Molecular dynamics simulation box

Three major conclusions/contributions were established with this work:

1. A new parameter, known as the "functionality index" was established to comprehensively characterize the level of crosslinking in epoxies of varying functionalities. That is, one parameter can now be used to describe the crosslink density for di-, tri-, and tetra-functional epoxies. This finally allows for an "apples-to-apples" comparison of crosslink density in different epoxy systems.
2. MD models demonstrated that increasing functionalities of epoxies have increasing levels of stiffness and strength. This conclusion was validated with experiment, thus providing evidence that the developed MD models are reliable and accurate. This is an important conclusion for the next item.
3. MD models of the CNT/epoxy system (Figure 2) demonstrated that the tri-functional epoxy composite is stiffer and stronger than the di- and tetra-functional systems. <u>Thus, the tri-functional epoxy system has the best load transfer characteristics for CNT/epoxy composites</u>. This is likely due to the larger percentage of aromatic rings in the tri-functional epoxy monomer, which have a tendency to align with the CNT at the interface.

<u>Modeling of Bismaleimide Resins</u>

This work is covered in publications #2 and #3 listed above, as well as in an upcoming journal manuscript that is currently in preparation. This research is the collaboration of Mr. Matt Radue, the supported PhD student, and Drs. Vikas Varshney, Ajit Roy, and Jeff Baur of AFRL. This work was performed while Matt spent two summers at AFRL. Matt's visits to AFRL were funded from this AFOSR grant.

One major contribution was established with this work. For the first time, MD was used to accurately simulate the complex crosslinking process of BMI resins (Figure 3). The predicted properties match very closely to experimental values, thus validating the model. Now that the details of the crosslinking process are well-understood, the methodology can be used to simulate/develop new BMI based composites systems with tailored properties.

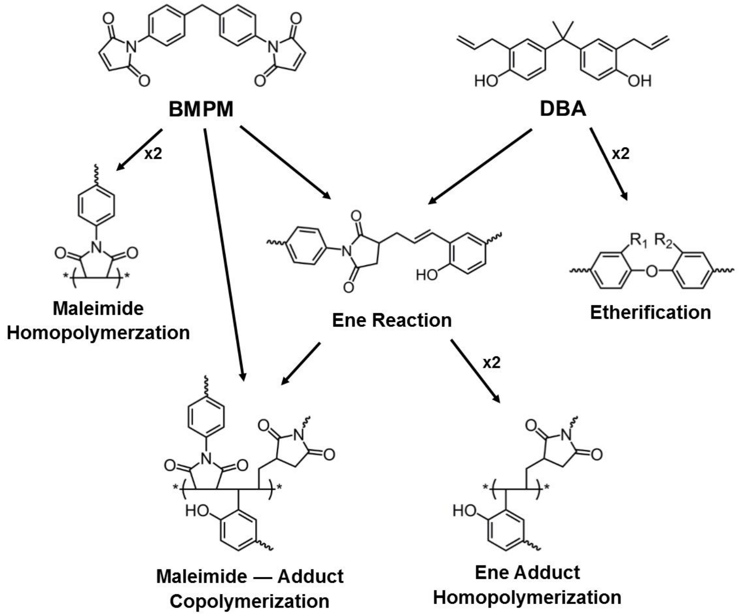

Figure 3 – Crosslinking process in BMI resins

<u>Using ReaxFF with epoxy</u>

This work is covered in publications #4, 5, and 6 listed above. This research is the collaboration of Dr. Odegard and researchers at the Norwegian University of Science and Technology (NTNU) during Dr. Odegard's sabbatical in 2013. This work was critical for the epoxy modeling work described above. This work established the reliability for ReaxFF so that it could be used in all subsequent modeling efforts.

There were two major contributions of this work:

1. For the first time, ReaxFF was used to predict the mechanical properties of epoxy materials using MD techniques. This is importance because traditional MD techniques cannot be used to predict material failure. Moving forward, ReaxFF must be used to simulate the scission and formation of chemical bonds in the epoxy material in a MD framework. The predictions were validated with experiment.
2. For the first time, the influence of simulated deformation rates were established with respect to experimentally-viable deformation rates. It was shown that time scaling can be used to directly compare simulated and experimental time scales, and thus scale predicted properties from MD simulation

MD modeling of Polymer Nanocomposites

This work is covered in publications #7-12 listed above, as well as two future journal manuscripts that are now in preparation. This research is a collection of projects that were indirectly supported by this AFOSR project. Specifically, publications 7 and 8 (and the two upcoming manuscripts) were co-authored by the supported PhD student, Mr. Matt Radue. Matt played a key role in helping train other Ph.D. students in Dr. Odegard's research group, as well as providing insight into the research results. Therefore, he was listed as a co-author, and these works represent collaboration between AFOSR and the other supporting entity: the NSF Industry/University Collaborative Research Center (I/UCRC) for Novel High Voltage/Temperature Materials and Structures. The I/UCRC represents a group of industrial members that have a common research interest, one of which is aerospace-grade composites materials.

For papers #9-12 listed above, Dr. Odegard's summer research support that was funded by this AFOSR grants was used to establish methods for connecting MD-level models with bulk material properties of hybrid composites (Figure 4). These methods will be utilized in one of the upcoming journal manuscripts that will be written by Mr. Matt Radue. These publications represent collaboration between AFOSR, NASA Langley Research Center, NASA Glenn Research Center, and the NSF I/UCRC described above.

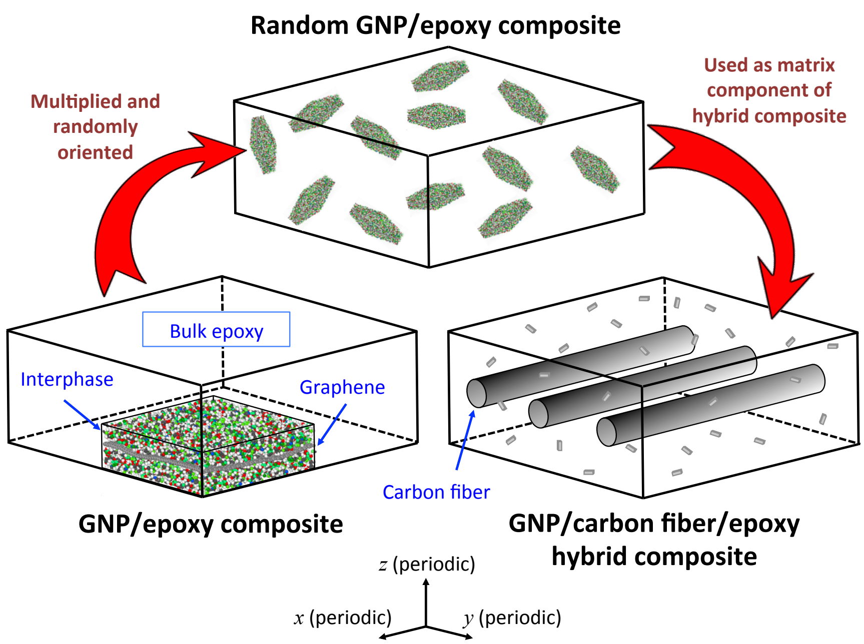

Figure 4 – Multiscale modeling of hybrid graphene nanoparticle (GNP)/carbon fiber/epoxy composites.

Synergistic activities

Not only did this project achieve the objectives as described above, this project also provided other important collaborative opportunities:

1. A direct collaboration was built with AFRL. The supported PhD student, Matt Radue, spent two summers working with Ajit Roy, Vikas Varshney, and Jeff Baur at AFRL. In the short term, the collaboration will result in two conference papers and a journal article.
2. A direct collaboration was built with researchers at the Norwegian University of Science and Technology (NTNU). The earliest stages of this project were performed with Prof. Zhiang Zhang's research group while Prof. Odegard was on sabbatical. This collaboration resulted in a journal article and two conference papers to kick off the proposed research.
3. This project leveraged funding and research efforts from the following grants on very closely related projects
    a. NASA "Multiscale Modeling Development and Validation of Graphene/ULTEM Composites for Structural and Noise Reduction Applications" (grant NNX11AO72A)
    b. NASA "Multiscale Modeling of Polymer Nanocomposites" (grant NNX09AM50A)
    c. NSF "I/UCRC: Novel High Voltage/Temperature Materials and Structures" (grant IIP-1362040)
    d. Fulbright Foundation
4. This project incorporated collaboration with NASA Glenn Research Center. Dr. Evan Pineda helped perform the MAC/GMC analyses for the hybrid composite material modeling. This work was published in a journal article, two conference papers, and a NASA TM.

Dr. Odegard and his research team are very grateful for the support of AFOSR for this research. Michigan Technological University is also grateful for supporting our continued research growth.

Please don't hesitate to contact Dr. Odegard if you have any questions regarding this research or the publications.

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Gregory Odegard

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Abstract

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12. Hadden, C.M., D.R. Klimek-McDonald, E.J. Pineda, J.A. King, A.M. Reichanadter, I. Miskioglu, S. Gowtham, G.M. Odegard, "Mechanical Properties of Graphene Nanoplatelet/Carbon Fiber/Epoxy Hybrid Composites: Multiscale Modeling and Experiments", NASA/TM-2015-218731

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Technical Summary

Funding Summary by Cost Category (by FY, $K)

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Equipment/Facilities			
Supplies			
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