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PII: S1359-835X(18)30083-6
DOI: https://doi.org/10.1016/j.compositesa.2018.02.033
Reference: JCOMA 4947

To appear in: *Composites: Part A*

Received Date: 26 November 2017
Revised Date: 27 January 2018
Accepted Date: 22 February 2018

Please cite this article as: Su, Z., Wang, H., Tian, K., Huang, W., Guo, Y., He, J., Tian, X., Multifunctional Anisotropic Flexible Cycloaliphatic Epoxy Resin Nanocomposites Reinforced by Aligned Graphite Flake with Non-covalent Biomimetic Functionalization, *Composites: Part A* (2018), doi: https://doi.org/10.1016/j.compositesa.2018.02.033

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Abstract: High thermal conductive filler (graphite flake) reinforced polymer composites have obtained a growing attention in the microelectronic industry. In order to overcome the obstacles in surface modification, in this study, dopamine chemistry was used to achieve the facile modification of graphite flake via forming a polydopamine (PDA) shell on the surface in a solvent-free aqueous condition. The strong π-π interaction between the hexagonal structural graphite flake and aromatic dopamine molecules ensured the effective modification. The PDA coating on graphite flake enhanced the compatibility between the filler and the flexible cycloaliphatic epoxy resin (CER) matrix via hydrogen bond, and promoted the epoxy curing process by forming covalent bond. Under the assistance of gravity, the PDA@graphite flake