

# Molecularly Pillared Graphene with Dithiolene and Diamine Linking Groups †

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**Abstract:** Molecularly pillared graphene is a class of materials constructed by sequential layers of graphene sheets that are interconnected by organic molecules denoted as molecular pillars and are connected to the graphene sheets via linking functional groups. The combinations of pillars and functional groups lead to multiple possibilities for tuning electromechanical properties [1, 2]. By employing computational methods in the framework of density functional theory (DFT) and tight-binding (TB), we study molecularly pillared graphene. We use diamine and dithiolene as functional groups and benzene rings as molecular pillars. All structures were preoptimized with initial geometries of molecular pillars linked to circumcoronene, with the Gaussian package [3]. Based on the preoptimized structures, we constructed the corresponding extended systems employing periodic boundary conditions (PBC) and performed simultaneous atomic position and computational cell optimization employing the GFN1- xTB method [4, 5] as implemented in the CP2K code [6]. We then examined the density of states (DOS). These computations were performed with Quantum Espresso [7]. The results show that structures with diamine as a functional group present a larger band gap compared to those with dithiolene. In conclusion, by using different functional groups, we can computationally tune the electromechanical properties of molecularly pillared graphene.

**Keywords:** functionalized graphene; molecularly pillared graphene; graphene oxide; density functional theory; tight binding; electronic properties; mechanical properties.

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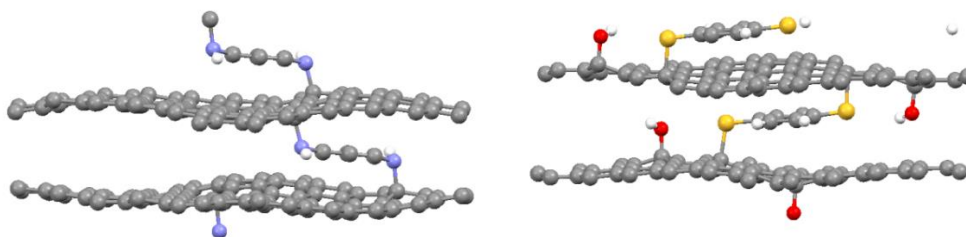
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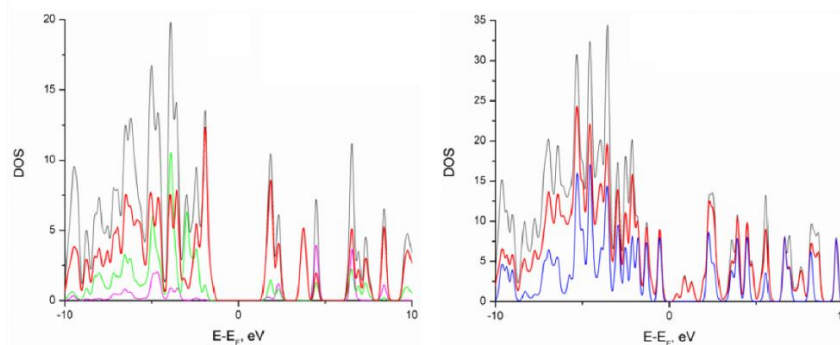
Results presented in this work have been produced using the Aristotle University of Thessaloniki (AUTH) High-Performance Computing Infrastructure and Resources.

## Conflicts of Interest

The authors declare no conflict of interest.



**Figure 1.** Computational cell of MPG with (left) diamine and (right) dithiolene linkers.



**Figure 2.** Density of States of the systems under study (see text).

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