



# Ab Initio Study of the Structures, Bonding Interactions, and Thermal Stability of the Li-Decorated 2D Biphenylene Sheet

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**Abstract:** We performed an extensive study on the most stable structures, the electronic properties, and the thermal stability of the 2D biphenylene sheet decorated with Li atoms. Our structural results show that the Li storage capacity of biphenylene is much higher than that recently reported, which increases the interest in this 2D material as a promising anode material for Li-ion batteries, although Li diffusion is not expected at room temperature. Moreover, we found striking phenomena that had not been detected yet, such as the formation of Li zigzag wires and metallic Li monolayers on the biphenylene sheet beyond a certain coverage threshold. In our calculations, we use high-level density-functional theory, quantum chemical topology analysis, and ab initio molecular dynamics simulations. In particular, the latter methodology allows for confirming the stability of the predicted Li-decorated biphenylene structures at room-temperature conditions.

**Keywords:** 2D materials; biphenylene; Li-adsorption on biphenylene; structural properties; electronic properties; stability at room temperature; ab initio density-functional theory; ab initio molecular dynamics; quantum chemical topology; metallic bonding

**Video V1:** (Color online) Video illustrating the evolution from the 8A-II configuration, with Li atoms on hexagons and octagons, to the 8A-I configuration ([movie-8A.mp4](#)).

**Video V2:** (Color online) Video illustrating the evolution from the 4B-IV configuration, with Li atoms on hexagons on one side and octagons on the other, to the 4B-II configuration ([movie-4B.mp4](#)).

**Video V3:** (Color online) Video illustrating the evolution from the 8B-III configuration, with Li atoms on hexagons and octagons, to the 8B-II configuration ([movie-8B.mp4](#)).