Functionalized Polycyclic Aromatic Hydrocarbon and Nanographenoid Materials Through Alkene Dicarbofunctionalization

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Organometallic reactions have played a pivotal role in the synthesis of polycyclic aromatic hydrocarbon (PAH) and nanographenoid materials from the bottom up, creating one carbon-carbon bond at a time. Nickel catalyzed, three-component, alkene dicarbofunctionalization (DCF) has emerged as a powerful tool to regioselectively form two carbon-carbon bonds in one step from simple starting materials, yet its utility remains unexplored in the synthesis of organic materials. This strategy allows for the efficient construction of PAH materials with the ability to functionalize, and thereby tune the physiochemical properties of, the resulting material by altering the sterics and the electronics of the starting materials. Following DCF, hydrolysis, a high yielding Bradsher cyclization, and novel oxidative cyclodehydrogenation take place in a sequential one-pot reaction giving PAH materials in an overall yield of 48%.

A catalog of PAH materials with varying combinations of electron-donating, withdrawing, and sterically demanding substrates are being synthesized based off this unique scaffold only accessible through this method and the physical properties will be examined. Additionally, strong visible light absorbing materials featured in organic electronics will be introduced to demonstrate our ability to tune the electronics through push-pull interactions. Further optimization with different starting materials incorporating heteroatoms and larger aromatic systems will be developed along with post-synthetic modifications to fully explore the range of PAH and nanographenoid materials and applications accessible through DCF.