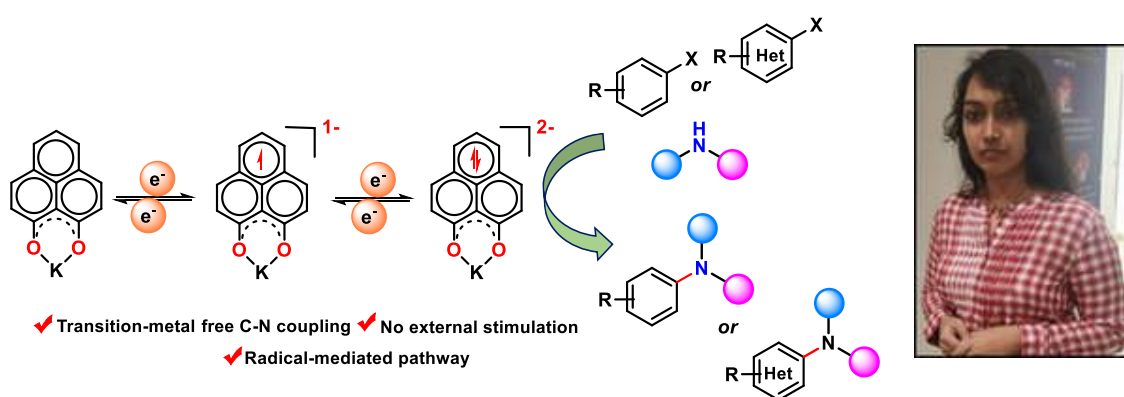


## Transition Metal-Free C-N Cross-Coupling Catalysis

C-N cross-coupling between aryl halides and different types of amines has remained an organic transformation of immense interest in academia and industry over decades. Numerous aniline motifs dominating the area of pharmaceuticals, agrochemicals and natural products keep motivating researchers to establish new synthetic methods for accomplishing C-N cross-coupled molecules. Generally, transition metals, mostly Pd and also Cu, Ni, Fe etc., are known for catalysing such C-N cross-coupling reactions. It's thus a challenge for pharmaceutical industries to remove trace amounts of heavy metal impurities from the various API molecules currently used clinically, which rely upon C-N cross-coupling catalysis. Apart from this, high temperature, photostimulation or cathodic current are among the external stimuli required for obtaining the expected C-N coupled product. Hence, the C-N cross-coupling between aryl halides and aromatic/aliphatic amines, if executed without transition metals and external stimulation, would be highly rewarding.



**Figure 1.** Phenalenyl-catalysed C-N cross-coupling at room temperature

However, our group has recently devised a new catalytic method which can accomplish C-N cross-coupling with the help of a reduced organic hydrocarbon molecule under ambient conditions. This method relies on tuning an organic polycyclic phenalenyl-based odd alternant hydrocarbon molecule's capacity to store two electrons which acts as a super electron donor to split the Ar-halide bond into reactive aryl radical. The aryl radical eventually interacts with nucleophilic nitrogen centres of amines leading to the C-N cross-coupled product. Our solely chemically-driven process has constructed a rich library of different C-N coupled products from primary/secondary genres of aromatic/heteroaromatic and aliphatic amines and has also covered a vast array of aromatic/heteroaromatic/polyaromatic halides. Interestingly the well-acclaimed bioactive molecules such as Vitamin E and Menthol have been functionalized. By this method, we have been successful in synthesizing an analogous form of the well-known anti-Parkinson Piribedil drug derivative. We believe such a concept will pave the way to construct the C-N cross-coupled products of industrial importance in a more sustainable and cost-effective way avoiding rare, toxic and expensive heavy metal-based catalysts.

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