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Exploring a Unique Reactivity of N-Heterocyclic Carbenes (NHC) in Rhodium(III)-Catalyzed Intermolecular C–H Activation/Annulation


NHCs in Rhodium-Catalyzed C–H Activation–Annulation to Fused Imidazolium Salts

Significance: The increasing interest in N-heterocyclic carbenes (NHCs) arises arguably from their unprecedented stereoelectronic properties, strong metal–NHC bonding, and great stability of their metal complexes. NHCs act as both ligands and directing groups. These properties make NHCs useful in C–H functionalization as well as C–C and C–heteroatom bond-forming catalysis (see Review below). Reported here is the first directed intramolecular C–H functionalization–annulation reaction using a NHC–rhodium(III) complex as catalytic system. Thus, reaction of imidazolium salts 1 with internal alkynes 2, bearing different types of substituent groups, furnishes a variety of imidazo[1,2-a]quinolinium derivatives 3 in a one-pot process in 35–93% yield.


Comment: This is the first report of this kind of reaction, which is formally a [4+2]-cycloaddition process. A range of substrates was evaluated and a relationship between the electronic demand of the rings and the alkyne was established. Another pleasant surprise is the mild room-temperature reaction conditions. The NHC–rhodium(III) complex was isolated and characterized, thus supporting strongly the proposed mechanism. The main disadvantage of this methodology is the use of a large amount of AgOTf, which can make its scalability expensive. Further applications are anticipated.