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Title: Cyclic(Alkyl)(Amino)Carbene (CAAC)–Mercury(II) Complexes and their Catalytic Activity in Hydroamination Reactions

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Abstract: The unprecedented low valent metal compounds, highly active catalysts are the outcome of the stabilization offered by the appended carbenes to the metal centers. A modified version of NHCs named as cyclic(alkyl)(amino)carbenes (CAACs) discovered by Bertrand in 2005, have surpassed the notion of routine carbene chemistry due to more nucleophilic (σ -donating) as well as electrophilic (π -accepting) nature than those of diamino carbenes. The present work deals with the syntheses of adducts of Cyclic (Alkyl)(Amino)Carbene (CAACs) with HgX_2 salts. As an initial trial, when $Hg(OAc)_2$ was reacted with $[CAACH]^+[Cl]^-$ the ionic complex, $[CAACH]^+[HgCl_3]^-$ (1) was isolated. In subsequent attempts, the reaction of insitu generated free carbene was carried out with HgX_2 salts that smoothly yield the CAAC-Hg(II) adducts as $[CAAC \cdot HgCl(\mu-Cl)]_2$ (2), $[CAAC \cdot HgBr(\mu-Br)]_2$ (3), $[CAAC \cdot HgI(\mu-I)]_2$ (4). In an effort to substitute the halide with a weakly coordinating anion, a cationic mercury species, $[(CAACcy)_2Hg(H_2O)]_2 + 2[NO_3]^-$ (5) was also obtained. This product was isolated by reacting $AgNO_3$ with the previously synthesized complex by Singh and co-workers $[CAACcy \cdot HgBr(\mu-Br)]_2$ (3.1) in DMSO. When the same reaction was performed in dry THF a two coordinated cationic mercury complex, $[(CAACcy)_2Hg]_2 + [Hg_2Br_6]^{2-}$ (6) was isolated. After the successful synthesis of some of these adducts, their application in the hydroamination reactions between aromatic amines and terminal alkynes has been explored. The catalytic ability of $[CAACcy \cdot HgBr(\mu-Br)]_2$ (3.1) in intermolecular hydroamination has been explored in detail.


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