Catalysis by Gold Alone and in Combination with Palladium: Methodology Developments and Total Synthesis of Natural Products

Based on their ability to activate C–C double or triple bonds as soft, carbophilic Lewis acids, gold salts are well suited for the formation of C–C and C–heteroatom bonds by nucleophilic attack at these activated substrates. Additionally, the concept of combining gold catalysis and other transition metal catalysis has emerged as a promising strategy for developing new and valuable reactions, and has attracted considerable attention by the chemical community. These recent developments form the basis of the work embodied in the thesis which is divided in five chapters. The first chapter comprises a brief overview of the key discoveries that highlight the importance of gold in the field of homogeneous catalysis. The second chapter describes a short synthesis of xyloketals and related natural product, alboatrin. In our approach, we have synthesized the required pendent alkynes, which on activation by gold catalyst followed by addition of two hydroxyl groups (an alcoholic OH and a phenolic OH) form the linear tricyclic tetrahydrofurano benzopyran ring system as enshrined in the xyloketals and related natural product, alboatrin. The third chapter illustrates a unified strategy for the synthesis of highly functionalized quinolines and isoquinolines via a homogeneous gold catalyzed benzannulation reaction. In the fourth chapter we have demonstrated that the reportedly inactive dual catalytic system $PdCl_2(PPh_3)_2$ /$AuCl(PPh_3)$ allows efficient Sonogashira-type cross-coupling of aryl- and heteroaryl halides. The last chapter of the thesis describes the first Sonogashira coupling of arenediazonium salts using a Pd-Au dual catalytic system. The utility of our Pd-Au dual catalytic system is further illustrated by the flexibility of being able to start with an aniline substrate and employ an in situ diazonium formation step.

Keywords: Gold catalysis, Gold-Palladium dual catalysis, Xyloketals, Cross-coupling reactions