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Title: Metal-catalyzed Approaches to the Synthesis of Indolizine Derivatives from 2-Pyridinyl-substituted para-Quinone Methides and/or 2-(2-Enynyl)pyridines

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Abstract:

The research work presented in this thesis primarily focuses on Metal-catalyzed Approaches to the Synthesis of Indolizine Derivatives from 2-Pyridinylsubstituted para-Quinone Methides (p-QMs) and/or 2-(2-Enynyl)pyridines. This thesis is divided into two parts, namely Part A and Part B. Part A describes the synthesis of indolizine-based heterocycles from 2-pyridinyl substituted para-quinone methides using suitable coupling partners such as terminal alkynes and N,N-dimethyl enaminones; Part B involves the synthesis of indolizine-based unsymmetrical triarylmethane derivatives through copper-catalyzed reactions between 2-(2- enynyl)pyridines and boronic acids or 2-hydroxyphenyl-substituted N,N-dimethyl enaminones. Part A: Synthesis of Indolizine-based heterocycles from 2-pyridinyl-substituted para- quinone methides (p-QMs) Part A is subdivided into three chapters. Chapter 1: General introduction to the synthesis of heterocycles from functionalized para- quinone methides In this Chapter, the synthetic applications of functionalized para-quinone methides for the syntheses of oxygen and nitrogen-containing heterocycles have been briefly discussed. Chapter 2: Pd(II)-Catalyzed annulation of terminal alkynes with 2-pyridinyl-substituted para-quinone methides: Direct access to indolizines This chapter describes the synthesis of 1,3-disubstituted indolizine derivatives from 2-pyridinyl substituted p-QMs through a Pd(II)-catalyzed regiospecific [3+2]-annulation with terminal alkynes. The indolizine scaffold is widely found in many natural products and biologically active molecules, and several of them display a variety of pharmacological activities such as anti-cancer, anti-bacterial, anti-oxidant, and cytotoxic properties. Besides these, they have also found application in material science as fluorescent probes, dye for dye-sensitized solar cells (DSSC), and as a material in organic light-emitting diodes (OLEDs) and in the agricultural sector as herbicide and fungicide (Figure 1). Although numerous synthetic approaches to efficiently access the indolizine mojety have been reported in the literature, most of them require pre-functionalized starting material and multistep synthesis of starting materials. As a result, both economically and in terms of reaction conditions, an easy and atom-economical approach to the synthesis of indolizine derivatives is highly desirable. While working in the field of p-QMs as a 1,6-conjugate acceptor and its utilization for the synthesis of various carbocycles and heterocycles, we hypothesized that the 2-pyridinyl substituted p-QMs could be used as a three-atom synthon for the synthesis of substituted indolizine derivatives through a [3+2]-annulation with terminal alkynes (Scheme 1). This protocol worked well with most of the terminal alkynes, and the corresponding indolizine derivatives were obtained in moderate to good yields. Control experiments revealed that the reaction takes place through a regiospecific [3+2]annulation of terminal alkynes with 2-pyridinyl substituted para-quinone methides.

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