Accepted Manuscript

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Qidi Zhong, Qiangwen Fan, Hong Yan

PII: S0040-4039(17)30212-5
DOI: http://dx.doi.org/10.1016/j.tetlet.2017.02.041
Reference: TETL 48652

To appear in: *Tetrahedron Letters*

Received Date: 12 January 2017
Revised Date: 10 February 2017
Accepted Date: 13 February 2017

Please cite this article as: Zhong, Q., Fan, Q., Yan, H., Synthesis of 2,3-dihydropyrroles by photo rearrangement of Hantzsch 1,4-dihydropyridines with high diastereoselectivity, *Tetrahedron Letters* (2017), doi: http://dx.doi.org/10.1016/j.tetlet.2017.02.041

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ACCEPTED MANUSCRIPT

Tetrahedron Letters
journal homepage: www.elsevier.com

Synthesis of 2,3-dihydropyrroles by photo rearrangement of Hantzsch 1,4-dihydropyridines with high diastereoselectivity

Qidi Zhong[a], Qiangwen Fan[a], Hong Yan[a]*

[a] *College of Life Science and Bio-engineering, Beijing University of Technology, Beijing, 100124, PR China*

ARTICLE INFO

Article history:
Received
Received in revised form
Accepted
Available online

Keywords:
2,3-Dihydropyrroles
Photo rearrangement
One-pot
Diastereoselectivity

ABSTRACT

The Hantzsch 1,4-dihydropyridines were found to be transforming to the 2,3-dihydropyrroles by photo rearrangement with air under irradiation of LED light (410nm) with high diastereoselectivity (dr>20:1). This reaction includes tandem photo oxidation/rearrangement. The 2,3-dihydropyrroles were obtained in moderate yields with successfully one-pot process starting from aldehydes, ammonium acetate and ethyl acetoacetate.

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Introduction

The 2,3-dihydropyrroles are important nitrogen-containing heterocycles encountered in significantly important pharmaceuticals, bioactive natural products, and building blocks in organic and diversity-oriented syntheses.[1] Moreover, 2,3-dihydropyrroles are also widely used as important versatile precursors for synthesis of pyrrolidines and pyrroles, which were also broadly found in natural products and pharmaceutically active substances.[2] As a result, many types of methods for the construction of the dihydropyrroles skeleton have been developed and reported, which include the [4+1] cycloaddition and [3+2] cycloaddition with transition-metal-catalyzed and DBU-catalyzed reactions by using different starting materials (Figure 1).[3] However, despite the impressive achievements of these protocols, the procedures are mostly constrained by their use of costly catalysts such as [Pd], [Rh] and [Ag], and the starting materials that are expensive and not readily available. In addition, methods for the synthesis of a new style of 2,3-dihydropyrroles (2), a structural motif found in biologically active dihydropyrroles, are still rare.[4] Hence, the search for a novel, cost-effective, and efficient synthetic route to 2,3-dihydropyrroles from the easily acquired materials remains an important objective.

Photochemical reactions have attracted much attention in the development of novel tandem reactions due to mild reaction conditions, efficient conversion, non-toxic, abundant and economy.[5] In this regard, many research efforts have concentrated on the photo behavior of heterocyclo hexadienes.[6][7] Especially, Yan's group reported a new reaction including the ring contraction of 1,4-dihydropyrazines to corresponding imidazoles under irradiation with 450W medium pressure-mercury (MPM) lamp (365nm) under nitrogen at room temperature (Scheme 1 a).[8] In view of the structural similarity between 1,4-dihydropyrazines and Hantzsch 1,4-dihydropyridines, it prompted us to verify whether the Hantzsch 1,4-dihydropyridines (2,6-dimethyl-3,5-dialkoxycarbonyl-1,4-dihydropyridines, **1**[9]) could undergo photo rearrangement reaction to give corresponding five-membered heterocyclic derivatives. Encouragingly, though the type of reaction was not as similar as Yan's report, a novel style of 2,3-dihydropyrroles **(2)**

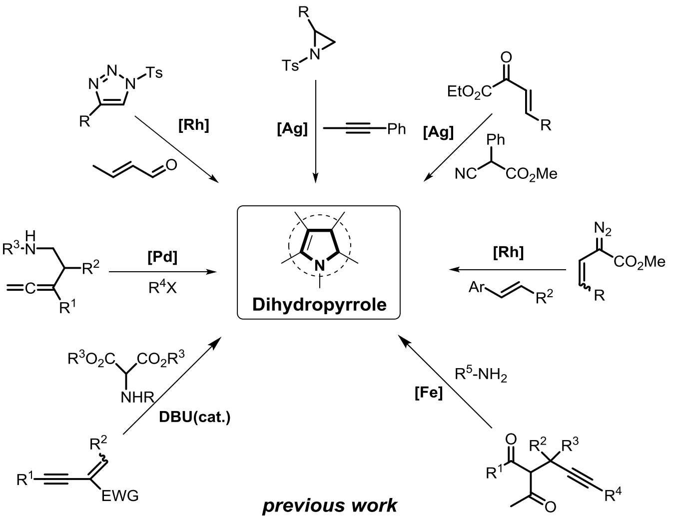

Figure 1. Commonly used synthetic methods for dihydropyrroles.

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* Corresponding author. Tel.: +0-086-136-8106-1170; e-mail: hongyan@bjut.edu.cn (H. Yan)

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