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Graphene Oxide Sheets Decorated by Zinc Oxide Nanoparticles: An Efficient and Heterogeneous Reusable Catalyst for the Synthesis of 9-aryl-1, 8-dioxooctahydroxanthenes and 9-aryl -1, 8-dioxodecahydroacridines in Aqueous Media

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ABSTRACT Graphene oxide sheets decorated by ZnO nanoparticles (GO-ZnO) have been synthesized and characterized by fourier transform infrared spectroscopy, X-ray powder diffractometry, FESEM, transmission electron microscopy, and EDAX. Herein, we have developed a one-pot protocol for the synthesis of 1,8-dioxo-octahydroxanthenes and 1,8-dioxo-decahydroacridines using GO-ZnO heterogeneous, recyclable and efficient catalyst in aqueous media. The aromatic aldehydes and dimedones were used as starting materials. The method has several fascinating advantages such as excellent yields, operational simplicity and short reaction times. The catalyst was effectively recycled up to six consecutive cycles without significant loss in its catalytic activity. Synthesized 1,8-dioxo-octahydroxanthenes and 1,8-dioxo-decahydroacridines were characterized using IR, ^{1}H-NMR, and ^{13}C-NMR spectroscopy.

KEYWORDS Aromatic aldehydes, Dimedone, GO-ZnO catalyst, Heterocyclic compounds.

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INTRODUCTION

Carbon-based catalysts have been projected as low-cost renewable "green catalysts" prepared from biomass or from household waste. Two dimensional layers of sp^2 bonded carbon have many advantages due to its easy fictionalizations. Graphene oxide (GO) has substantial interest due to presence of its high specific surface area, inexpensive, highly thermal conductivity, abundant functional sites, rich oxidation functional groups (e.g., hydroxyl, carboxyl, and epoxy groups), reuse potential, high mechanical strength,[1] and excellent electrical conductivity.[2] GO has attracted considerable attention in various field including sensors,[3] biomedical,[4] supercapacitors,[5] catalysts,[6] bioimaging,[7] surfactant,[8] and pollutant adsorbents.[9] Recently, GO and its composite has gained interest as a new carbonate in organic conversions.[10-15] GO sheets possess large surface areas, and thus may be potential support materials to load functional nanomaterials. It has been found that GO and functionalized graphene materials widely used as heterogeneous catalyst due to high yield of product, easy recovery and integrated cost reduction. Nanocatalysis has been widely used in organic transformation as heterogeneous catalysts due to its high surface area and high chemical stability.[16-19] Nanoparticles such as ZnO, TiO_2 and ZrO_2 with GO have been synthesized and used as heterogeneous catalyst in chemical synthesis and photocatalytic properties.[20-22] ZnO nanoparticles has found application in optics, optoelectronics, sensors, and actuators due to its semiconducting, optical, and photoluminescence properties.[23-25] ZnO nanoparticles have also been reported as used as heterogeneous catalyst for organic transformations.[26-30] In the present work, we demonstrated the uniform inclusion of ZnO nanoparticles on GO. Rawat and coworker reported the catalytic application

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2.33–2.54 (m, 8H, 4×CH_2), 4.77 (s, 1H, CH), 7.04 (d, 2H, Ar-H), 7.16 (d, 2H, Ar-H), 9.08 (s, 1H, NH).

9-Phenyl-3,4,6,7-tetrahydro-3,3,6,6-tetramethylacridine-1,8(2H,5H,9H,10H)-dione (4c)

IR (KBr, cm^{-1}): 3294, 2970, 1685, 1640, 1611;^{1}HNMR (500 MHz, $CDCl_3$) δ: 0.94 (s, 6H, 2×CH_3), 1.05 (s, 6H, 2×CH_3), 2.11–2.31 (m, 8H, 4×CH_2), 5.08 (s, 1H, CH), 7.04–8.11 (m, 5H, Ar-H), 9.50 (s,1H, NH) ppm;

9-(3-Nitrophenyl)-3,4,6,7-tetrahydro-3,3,6,6-tetramethylacridine-1,8(2H,5H,9H,10H)-dione (4d)

^{13}C NMR ($CDCl_3$) δ: 22.6, 29.3, 31.9, 32.7, 34.0, 41.1, 50.5, 112.8, 121.2, 122.2, 128.6, 135.5, 148.4, 195.2

CONCLUSION

We have reported a highly efficient and cost-effective approach for the synthesis of 1,8-dioxo-octahydroxanthenes and 1,8-dioxo-decahydroacridines in the presence GO-ZnO as a heterogeneous catalyst. This method offers shorter reaction time, a simple work-up procedure with high yield, use of small amount of catalyst, and recyclable heterogeneous catalyst. The GO-ZnO catalyst was used in aqueous synthesis of 1,8-dioxo-octahydroxanthenes and 1,8-dioxo-decahydroacridines which are biologically interesting compounds. The detecting results could indicate our protocol make the reaction suitable for scale-up and commercialization.

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