

Highly efficient synthesis of benzopyranopyrimidine derivatives catalyzed by functionalized superparamagnetic graphene oxide as a new and recoverable catalyst

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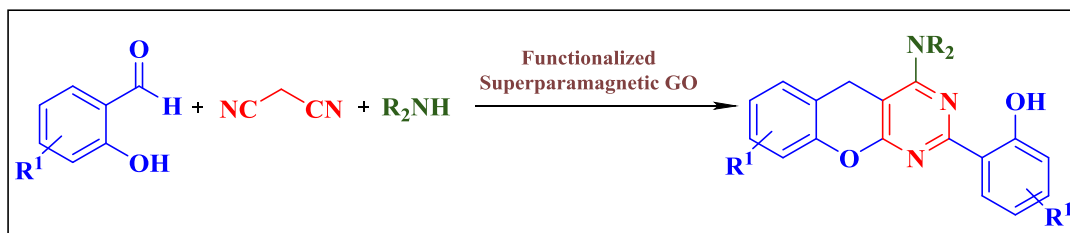
Abstract

In multicomponent reactions (MCRs), three or more reactants come together in a single reaction vessel to form new products that contain structural units of all the components. This type of reaction becomes progressively important in organic and medicinal chemistry because it allows obtaining highly sophisticated polyfunctional molecules through simple one-pot procedures.^[1–3]

Keywords: Benzopyranopyrimidine, graphene oxide, recoverable catalyst.

1. Introduction

Benzopyrano[2,3-*d*]pyrimidins (BPP's) are vital pharmacophore that exhibits antitumor activity, cytotoxic activity against cancer cell lines and can cause significant perturbation in cell cycle kinetics. In continuation of our ongoing efforts towards to develop efficient catalysts for different MCRs^[4], We report herein functionalized superparamagnetic graphene oxide as a new and heterogeneous catalyst for the synthesis of benzopyranopyrimidine derivatives in high yields via a one-pot, pseudo four-component condensation of salicylaldehydes, malononitrile and secondary amines under mild conditions at room temperature.^[5–6] (Scheme1). It should be noted that the significant advantages of this procedure are low catalyst loading, short reaction times, high to excellent yields, simple workup, reusability of the catalyst and simple purification of the products (table1).



Scheme 1. Pseudo–four-component synthesis of benzopyranopyrimidine derivatives.

2. Experimental

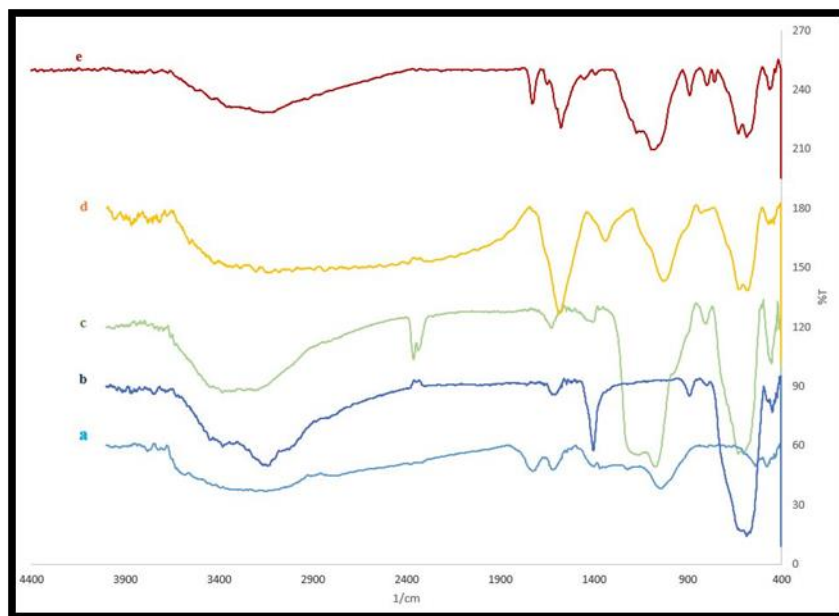
To a 5mL round-bottomed flask were added malononitrile (0.5 mmol), salicylaldehyde (1 mmol), secondary amine (0.5 mmol), EtOH (1.5 ml), amount of functionalized superparamagnetic graphene oxide as a catalyst. The suspension was stirred vigorously for the required reaction time, at room temperature. The progress of the reactions was monitored by TLC (1:3). When the formation product, the catalyst was removed from the mixture via sintering in EtOH.

ENTRY	SOLVENT	ALDEHYDE	AMINE	TIME (MIN)	YIELD%
1	Ethanol	4-Methoxy benzaldehyde	Morpholine	11	94
2	Ethanol	5-bromo-2-hydroxyl benzaldehyde	Morpholine	6	94
3	Ethanol	2-hydroxy-4-methoxy benzaldehyde	Morpholine	8	98

Table 1: Yields refer to isolated pure products.

Results and discussion

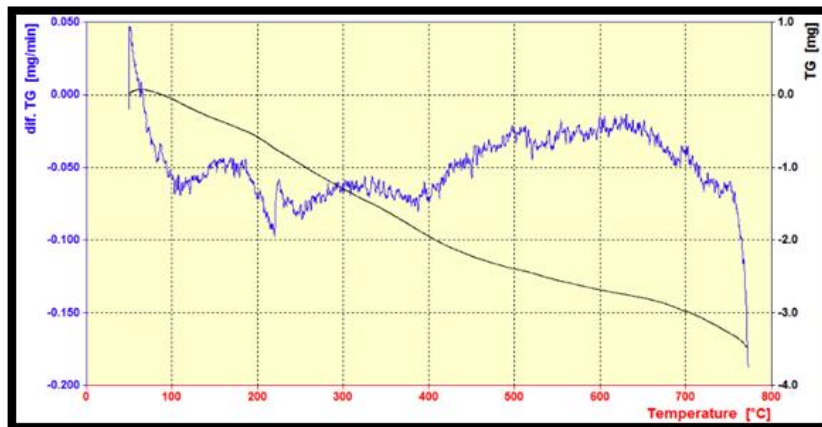
We report herein the catalytic application of a new functionalized superparamagnetic graphene oxide (GO), as a highly efficient and recoverable catalyst, for the synthesis of benzopyranopyrimidine derivatives. FT-IR spectroscopy was performed for the confirmation of the formation new functionalized superparamagnetic graphene oxide (Scheme2).



Scheme2: FTIR spectra of GO (a), Fe₃O₄ (b), Fe₃O₄-SiO₂ (c), Fe₃O₄-SiO₂-GO (d) functionalized superparamagnetic GO, hybrid composite

This catalyst has been proved to be highly active towards one-pot synthesis of benzopyranopyrimidines via pseudo multi component coupling reactions of malononitrile, α -hydroxybenzaldehydes and secondary amines in EtOH, at room temperature.

The thermal behaviors of the functionalized superparamagnetic GO are further investigated by thermogravimetric analysis/ differential thermal analysis (TGA/DTA), with heating from room temperature to 800 °C under an air flow (Scheme3).



Scheme3: TGA/DTA functionalized superparamagnetic GO, hybrid composite

References

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