



Original Research Article

Another Successful Application of Newly Prepared GO-SiC₃-NH₃-H₂PW as Highly Efficient Nanocatalyst for Fast Synthesis of Tetrahydrobenzo[*b*]pyrans

Zahra Hoseini, Abolghasem Davoodnia*, Mehdi Pordel

Department of Chemistry, Mashhad Branch, Islamic Azad University, Mashhad, Iran

ARTICLE INFO

Article history

Submitted: 28 November 2020

Revised: 05 January 2021

Accepted: 08 January 2021

Available online: 22 January 2021

Manuscript ID: [AJCA-2011-1228](https://doi.org/10.22034/AJCA.2011-1228)DOI: [10.22034/AJCA.2021.259593.1228](https://doi.org/10.22034/AJCA.2021.259593.1228)

KEYWORDS

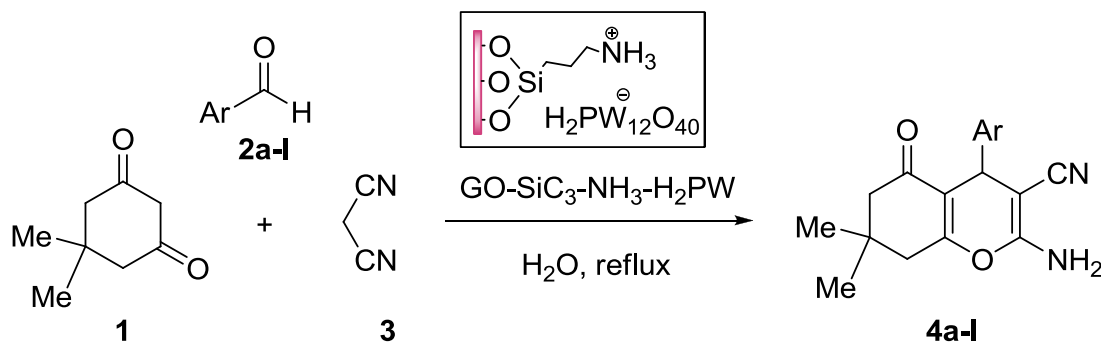
Tetrahydrobenzo[*b*]pyranH₃PW₁₂O₄₀

Functionalized graphene oxide

ABSTRACT

In this work, the newly prepared functionalized graphene oxide (GO), denoted as GO-SiC₃-NH₃-H₂PW, was found as an effective nanocatalyst for a one-pot reaction of dimedone, aryl aldehydes, and malononitrile, giving rise to tetrahydrobenzo[*b*]pyran derivatives. The reactions were conducted in water, giving the corresponding products in 88-98% yields over 4-15 min. Other advantages of the method include cheap catalyst, easy work-up, absence of any dangerous solvents and the catalyst's reusability for up to five consecutive runs (97, 96, 95, 95, and 94 in first to fifth use, respectively).

GRAPHICAL ABSTRACT



* Corresponding author: Davoodnia, Abolghasem

✉ E-mail: adavoodnia@mshdiau.ac.ir; adavoodnia@yahoo.com

☎ Tel number: +98 (51) 38435000

© 2020 by SPC (Sami Publishing Company)

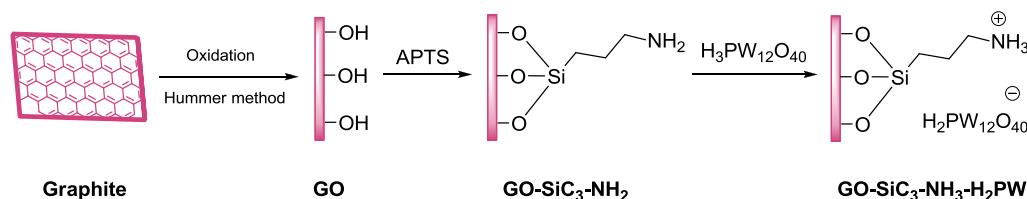
Introduction

The use of homogeneous acidic catalysts such as H_2SO_4 , HCl , CH_3COOH , or H_3PO_4 has some major limitations, including the difficulty of catalyst separation from the reaction medium, volatility, toxicity, and therefore incompatibility with the environment. Therefore, nowadays, the use of recyclable, nontoxic, cheap, and easy to handle heterogeneous catalysts is of great interest to organic chemists [1-6]. Due to the low specific surface area of the bulk heterogeneous catalysts, immobilization of heterogeneous catalysts on the surface of a solid support with high surface area such as graphene oxide (GO) nanosheets has attracted a great deal of attention in recent years [7-10]. GO is usually prepared from graphite powder [11]. Although this nanomaterial can be directly used as a low-cost carbocatalyst to facilitate various organic transformations [12-15], it has been considered more support for homogeneous and heterogeneous catalysts [16-22]. Several methods have been developed for attachment of functional groups on GO surface by covalent linkage or noncovalent interaction [23-28]. A number of applications in solar cells [29], dyes [30], and also as effective catalysts [31-33] have been reported for the functionalized GO-based materials.

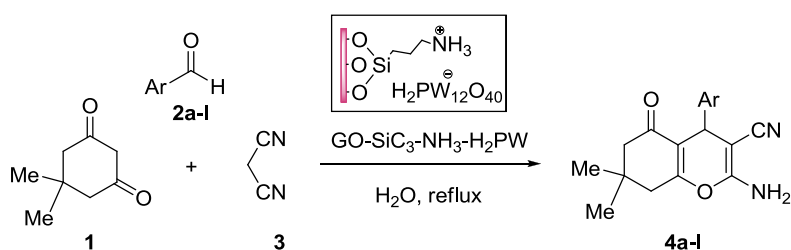
Pyrans and tetrahydrobenzo[*b*]pyrans are important heterocycles with lots of natural cases, as well as synthetic derivatives, and have an outstanding position in medicinal chemistry. They are cited as active oxygen heterocycles having significant antiproliferative, antibacterial, antianaphylactin, anticoagulant, anticancer, diuretic, and spasmolytic activities [34-40]. A number of them are utilized as photoactive compounds [41] or found in the structure of natural products [42].

Tetrahydrobenzo[*b*]pyrans are synthesized by the one-pot reaction of dimedone with an aryl aldehyde and malononitrile using various promoting agents such as I_2 [43], piperazine [44], $\text{PhB}(\text{OH})_2$ [45], tetra(*n*-butyl)ammonium bromide (TBAB) [46], nanomixed metal oxides [47], DABCO-based ionic liquid [48], rare-earth perfluorooctanoate $[\text{RE}(\text{PFO})_3]$ [49], $\text{MnFe}_2\text{O}_4@\text{SiO}_2@\text{NHPhNH}_2$ -phosphotungstic acid [50], NFS-PWA [51], $\text{Na}_2\text{CaP}_2\text{O}_7$ [52], FSM-16/AEPC- SO_3H [53], $\text{KF}/\text{Al}_2\text{O}_3$ under ultrasound irradiation [54], NaBr under microwave irradiation [55], 2,2,2-trifluoroethanol (TFE) [56], Na_2SeO_4 [57], and basic ionic liquid [58]. Some of these methodologies, however, may suffer from certain drawbacks such as the use of relatively expensive catalysts, toxic solvents, lengthy reaction times, or unsatisfactory yields. Therefore, the introducing of a new effective catalyst is highly demanding for the preparation of tetrahydrobenzo[*b*]pyrans which surpass those limitations.

Very recently, a new nanocatalyst containing $\text{H}_2\text{PW}_{12}\text{O}_{40}^-$ (H_2PW), shown as $\text{GO-SiC}_3\text{-NH}_3\text{-H}_2\text{PW}$, was synthesized by linking of 3-aminopropyltriethoxysilane (APTS) on GO followed by reaction with phosphotungstic acid ($\text{H}_3\text{PW}_{12}\text{O}_{40}$, marked as H_3PW), and identified in our group (Scheme 1). This new reusable nanocatalyst was used to synthesize the amidoalkyl naphthols, demonstrating a high level of catalytic activity [59]. In this vein, and in line with our interest in catalysis and heterocycles [60-69], in the present study we report another application of $\text{GO-SiC}_3\text{-NH}_3\text{-H}_2\text{PW}$ in the synthesis of tetrahydrobenzo[*b*]pyrans **4a-l** by the reaction of dimedone **1**, aryl aldehydes **2a-l**, and malononitrile **3** in water as a green medium (Scheme 2).



Scheme 1. Preparation of GO-SiC₃-NH₃-H₂PW nanosheets



Scheme 2. Synthesis of tetrahydrobenzo[*b*]pyrans using GO-SiC₃-NH₃-H₂PW

Experimental

A Stuart SMP3 apparatus was applied for recording of the Melting points. The FT-IR spectra using KBr disks were obtained on a Tensor 27 Bruker spectrophotometer. Ultrasonication was done with a Soltec sonicator at 40 kHz frequency and 260 W powers. The ¹H NMR (300 MHz) spectra were recorded with Bruker 300 FT spectrometer in DMSO-*d*₆ as solvent.

Preparation of GO-SiC₃-NH₃-H₂PW

The catalyst was synthesized by linking of APTS on GO followed by interaction with H₃PW according to the literature [59].

Synthesis of tetrahydrobenzo[*b*]pyrans **4a-l** catalyzed by GO-SiC₃-NH₃-H₂PW

A mixture of dimedone **1** (1.0 mmol), an aryl aldehyde **2a-l** (1.0 mmol), malononitrile **3** (1.0 mmol), and GO-SiC₃-NH₃-H₂PW (0.07 g) in water (5 mL) was refluxed for 4-15 min. After completion of the reaction, the catalyst was separated by hot filtration. The filtrate was cooled and the obtained solid was collected by

filtration and crystallized from 96% ethanol to give compounds **4a-l** in high yields.

Selected spectral data

Compound 4a. ¹H NMR (δ, ppm): 0.97 (s, 3H, CH₃), 1.05 (s, 3H, CH₃), 2.12 (d, *J* = 16.0 Hz, 1H, CH₂), 2.27 (d, *J* = 16.0 Hz, 1H, CH₂), 2.45-2.60 (m, 2H, CH₂ overlapped with solvent), 4.22 (s, 1H, CH), 7.08 (s, 2H, NH₂), 7.20 (d, *J* = 8.0 Hz, 2H, H_{Ar}), 7.37 (d, *J* = 8.0 Hz, 2H, H_{Ar}); FT-IR (ν, cm⁻¹): 3394 and 3204 (NH₂), 2192 (CN), 1664 (C=O).

Compound 4d. ¹H NMR (δ, ppm): 0.98 (s, 3H, CH₃), 1.05 (s, 3H, CH₃), 2.15 (d, *J* = 16.0 Hz, 1H, CH₂), 2.28 (d, *J* = 16.0 Hz, 1H, CH₂), 2.48-2.58 (m, 2H, CH₂ overlapped with solvent), 4.26 (s, 1H, CH), 7.13 (s, 2H, NH₂), 7.18-7.45 (m, 4H, H_{Ar}); FT-IR (ν, cm⁻¹): 3434 and 3166 (NH₂), 2192 (CN), 1669 (C=O).

Compound 4g. ¹H NMR (δ, ppm): 0.97 (s, 3H, CH₃), 1.05 (s, 3H, CH₃), 2.11 (d, *J* = 16.0 Hz, 1H, CH₂), 2.27 (d, *J* = 16.0 Hz, 1H, CH₂), 2.45-2.55 (m, 2H, CH₂ overlapped with solvent), 3.73 (s, 3H, OCH₃), 4.16 (s, 1H, CH), 6.86 (d, *J* = 8.6 Hz, 2H, H_{Ar}), 6.98 (s, 2H, NH₂), 7.09 (d, *J* = 8.6 Hz, 2H, H_{Ar}); FT-IR (ν, cm⁻¹): 3433 and 3170 (NH₂), 2192 (CN), 1659 (C=O).

Compound 4j. ^1H NMR (δ , ppm): 0.90 (s, 3H, CH_3), 1.03 (s, 3H, CH_3), 2.03 (d, $J = 16.1$ Hz, 1H, CH_2), 2.23 (d, $J = 16.1$ Hz, 1H, CH_2), 2.40-2.60 (m, 2H, CH_2 overlapped with solvent), 4.98 (s, 1H, CH), 7.22 (s, 2H, NH_2), 7.35-7.47 (m, 2H, H_{Ar}), 7.67 (td, $J = 7.5, 1.1$ Hz, 1H, H_{Ar}), 7.83 (dd, $J = 8.1, 1.1$ Hz, 1H, H_{Ar}); FT-IR (ν , cm^{-1}): 3375 and 3194 (NH_2), 2193 (CN), 1659 (C=O).

Compound 4l. ^1H NMR (δ , ppm): 0.98 (s, 3H, CH_3), 1.06 (s, 3H, CH_3), 2.13 (d, $J = 16.0$ Hz, 1H, CH_2), 2.29 (d, $J = 16.0$ Hz, 1H, CH_2), 2.50-2.60 (m, 2H, CH_2 overlapped with solvent), 4.39 (s, 1H, CH), 7.20 (s, 2H, NH_2), 7.47 (d, $J = 8.7$ Hz, 2H, H_{Ar}), 8.19 (d, $J = 8.7$ Hz, 2H, H_{Ar}); FT-IR (ν , cm^{-1}): 3348 and 3196 (NH_2), 2194 (CN), 1652 (C=O).

Results and Discussion

After preparation of GO-SiC₃-NH₃-H₂PW [59], the activity of this catalyst was evaluated in the synthesis of tetrahydrobenzo[*b*]pyrans. Initially, the reaction of dimedone **1** (1.0 mmol), 4-chlorobenzaldehyde **2a** (1.0 mmol), and malononitrile **3** (1.0 mmol) for the preparation of compound **4a** in the absence or presence of GO-SiC₃-NH₃-H₂PW were chosen as a test reaction to determine appropriate conditions. Considering the unique properties of water such as the large surface tension, the network of hydrogen bonds, polarity, the absence of inflammability, low cost, and availability [70,71], we firstly decided to investigate the efficiency of GO-SiC₃-NH₃-H₂PW in the test reaction in water as a solvent in different catalyst amounts and temperatures. The results are demonstrated in Table 1. A blank, no catalyst, reaction in refluxing water (entry 1) did not lead to a considerable yield of the product **4a**, indicating the necessary of the catalyst for the reaction. Interestingly, it was observed that the GO-SiC₃-NH₃-H₂PW catalyst was highly efficient, and 0.07 g of it in refluxing water was sufficient to give the product in high yield over short time (entry 11). However, further increasing the catalyst amount had no significant effect on the

product yield and reaction time. On the other hand, the compound **4a** can be obtained in low to high yields in EtOH, MeOH, CH₃CN, CHCl₃, and CH₂Cl₂ and also under solvent-free conditions. Although there was no considerable difference in the product yield between the use of H₂O and EtOH as solvent (entries 11 and 13), the reaction time was shorter in H₂O. Therefore, H₂O was selected as a solvent in all subsequent reactions because of the advantages mentioned above. It should be noted that water plays a critical role in accelerating the rate of a large number of organic reactions [72]. In this reaction, however, formation of the intermediates becomes easier probably due to the water's high polarity.

For comparison, the effect of GO and GO-SiC₃-NH₂ was also studied in the optimized model reaction. As seen in Table 1 (entries 19 and 20), GO-SiC₃-NH₃-H₂PW showed to be a better catalyst compared with that of the GO and GO-SiC₃-NH₂ in both reaction time and yield. Furthermore, the heterogeneous nature of GO-SiC₃-NH₃-H₂PW was investigated by hot filtration test on the optimized model reaction. Thus, a mixture of GO-SiC₃-NH₃-H₂PW (0.07 g), dimedone **1** (1.0 mmol), 4-chlorobenzaldehyde **2a** (1.0 mmol), and malononitrile **3** (1.0 mmol) was heated in water under reflux. After 4 min (50% conversion), the catalyst was isolated by hot filtration. The filtrate was heated for additional 4 min. The reaction's monitoring revealed no further formation of the product and so no leaching of GO-SiC₃-NH₃-H₂PW occurred during the reaction and GO-SiC₃-NH₃-H₂PW is heterogeneous in nature.

By considering the optimized conditions and the scope of this methodology, a range of substituted aryl aldehydes undergo reaction with dimedone **1** and malononitrile **3** under optimized conditions to afford 2-amino-4-aryl-3-cyano-7,7-dimethyl-5-oxo-5,6,7,8-4H-tetrahydrobenzo[*b*]pyran derivatives (Table 2). As demonstrated in Table 2, the reaction is effective with diverse electron-rich and electron-

poor aryl aldehydes. Although the type of aryl aldehyde had no notable effect on the reaction, the electron-poor aryl aldehydes reacted faster, giving the higher yields of the products, as would

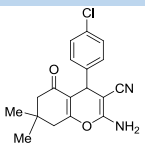
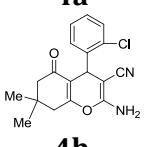
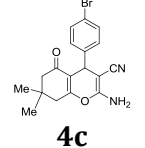
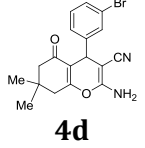
be expected. Therefore the GO-SiC₃-NH₃-H₂PW was found to be an effective catalyst in this methodology.

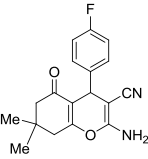
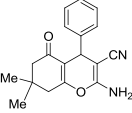
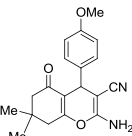
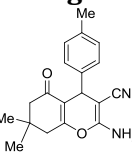
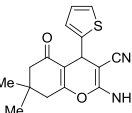
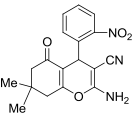
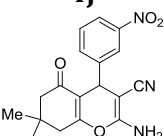
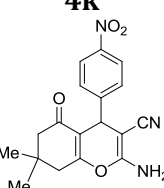
Table 1. Optimization of conditions for the preparation of compound **4a**^a

| Entry | Catalyst (g) | Solvent | T (°C) | Time (min) | Isolated Yield (%) |
|-------|--|---------------------------------|--------|------------|--------------------|
| 1 | ----- | H ₂ O | Reflux | 100 | Trace |
| 2 | GO-SiC ₃ -NH ₃ -H ₂ PW (0.03) | H ₂ O | 50 | 50 | 44 |
| 3 | GO-SiC ₃ -NH ₃ -H ₂ PW (0.03) | H ₂ O | Reflux | 20 | 75 |
| 4 | GO-SiC ₃ -NH ₃ -H ₂ PW (0.04) | H ₂ O | 50 | 45 | 49 |
| 5 | GO-SiC ₃ -NH ₃ -H ₂ PW (0.04) | H ₂ O | Reflux | 15 | 80 |
| 6 | GO-SiC ₃ -NH ₃ -H ₂ PW (0.05) | H ₂ O | 50 | 45 | 52 |
| 7 | GO-SiC ₃ -NH ₃ -H ₂ PW (0.05) | H ₂ O | Reflux | 15 | 86 |
| 8 | GO-SiC ₃ -NH ₃ -H ₂ PW (0.06) | H ₂ O | 50 | 40 | 55 |
| 9 | GO-SiC ₃ -NH ₃ -H ₂ PW (0.06) | H ₂ O | Reflux | 12 | 90 |
| 10 | GO-SiC ₃ -NH ₃ -H ₂ PW (0.07) | H ₂ O | 50 | 40 | 61 |
| 11 | GO-SiC ₃ -NH ₃ -H ₂ PW (0.07) | H ₂ O | Reflux | 8 | 97 |
| 12 | GO-SiC ₃ -NH ₃ -H ₂ PW (0.08) | H ₂ O | Reflux | 10 | 97 |
| 13 | GO-SiC ₃ -NH ₃ -H ₂ PW (0.07) | EtOH | Reflux | 15 | 95 |
| 14 | GO-SiC ₃ -NH ₃ -H ₂ PW (0.07) | MeOH | Reflux | 30 | 82 |
| 15 | GO-SiC ₃ -NH ₃ -H ₂ PW (0.07) | CH ₃ CN | Reflux | 45 | 54 |
| 16 | GO-SiC ₃ -NH ₃ -H ₂ PW (0.07) | CHCl ₃ | Reflux | 50 | 41 |
| 17 | GO-SiC ₃ -NH ₃ -H ₂ PW (0.07) | CH ₂ Cl ₂ | Reflux | 50 | 45 |
| 18 | GO-SiC ₃ -NH ₃ -H ₂ PW (0.07) | ----- | 100 | 60 | 51 |
| 19 | GO (0.07) | H ₂ O | Reflux | 60 | 41 |
| 20 | GO-SiC ₃ -NH ₂ (0.07) | H ₂ O | Reflux | 90 | 39 |

^aReaction conditions: dimedone (1.0 mmol), 4-chlorobenzaldehyde (1.0 mmol), and malononitrile (1.0 mmol)

Table 2. Synthesis of tetrahydrobenzo[*b*]pyrans **4a-l** catalyzed by GO-SiC₃-NH₃-H₂PW^a

| Entry | Ar | Product | Time (min) | Isolated Yields (%) | m.p. (°C) |
|-------|-----------------------------------|---|------------|---------------------|-----------|
| 1 | 4-ClC ₆ H ₄ |  | 8 | 97 | 210-212 |
| 2 | 2-ClC ₆ H ₄ |  | 10 | 89 | 217-218 |
| 3 | 4-BrC ₆ H ₄ |  | 8 | 91 | 215-217 |
| 4 | 3-BrC ₆ H ₄ |  | 10 | 90 | 228-230 |

| | | | | | |
|-----------|---|---|----|----|---------|
| 5 | 4-FC ₆ H ₄ |  | 6 | 93 | 188-190 |
| 4e | | | | | |
| 6 | C ₆ H ₅ |  | 10 | 90 | 229-231 |
| 4f | | | | | |
| 7 | 4-MeOC ₆ H ₄ |  | 15 | 88 | 201-203 |
| 4g | | | | | |
| 8 | 4-MeC ₆ H ₄ |  | 12 | 88 | 216-218 |
| 4h | | | | | |
| 9 | 2-Thienyl |  | 10 | 90 | 222-224 |
| 4i | | | | | |
| 10 | 2-O ₂ NC ₆ H ₄ |  | 8 | 90 | 232-234 |
| 4j | | | | | |
| 11 | 3-O ₂ NC ₆ H ₄ |  | 7 | 92 | 212-215 |
| 4k | | | | | |
| 12 | 4-O ₂ NC ₆ H ₄ |  | 4 | 98 | 181-182 |
| 4l | | | | | |

^aReaction conditions: dimedone (1.0 mmol), an aryl aldehyde (1.0 mmol), malononitrile (1.0 mmol), GO-SiC₃-NH₃-H₂PW (0.07 g), H₂O, reflux.

The efficiency of the catalyst GO-SiC₃-NH₃-H₂PW was compared with other methods of synthesis of tetrahydrobenzo[*b*]pyrans (Table 3). The results revealed that the current methodology provided high yields of the products in shorter times than the others. On the other hand, as seen

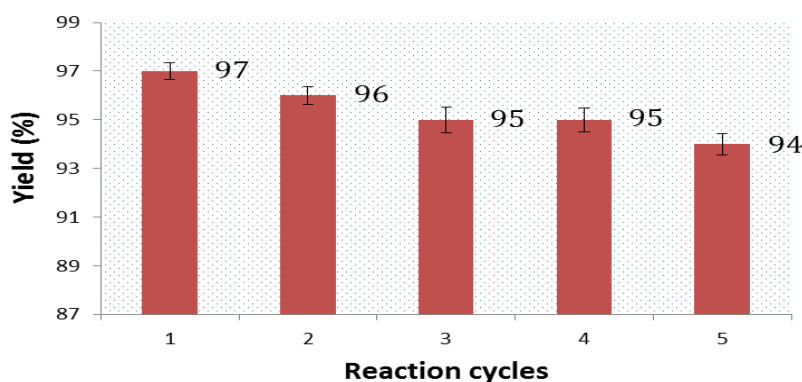
in Table 3, although organic solvents have been used in most cases, in the current method, the reactions are done in water as a green solvent and so operate under environmentally friendly conditions.

Table 3. Comparison of different catalysts in the synthesis of tetrahydrobenzo[*b*]pyrans

| Catalyst | Conditions | | | Time (min) | Yield (%) | Ref. |
|--|-----------------------|--------|--------------------------|------------|-----------|-----------|
| | Solvent | T (°C) | Other | | | |
| I ₂ | DMSO | 120 | ---- | 180-240 | 80-92 | [43] |
| Piperazine | ---- | r.t. | Ball-milling at 20-25 Hz | 20-120 | 88-96 | [44] |
| PhB(OH) ₂ | EtOH/H ₂ O | reflux | ---- | 30 | 61-88 | [45] |
| TBAB | EtOH | reflux | ---- | 20-140 | 87-95 | [46] |
| Nano mixed metal oxides | EtOH | reflux | ---- | 35-45 | 90-94 | [47] |
| DABCO-based ionic liquid | EtOH/H ₂ O | reflux | ---- | 12-45 | 85-95 | [48] |
| [RE(PFO) ₃] | EtOH | 60 | ---- | 240-480 | 80-90 | [49] |
| MnFe ₂ O ₄ @SiO ₂ @NHPh | ---- | 80 | ---- | 25-35 | 86-95 | [50] |
| NH ₂ -phosphotungstic acid | ---- | 80 | ---- | 25-35 | 86-95 | [50] |
| NFS-PWA | EtOH | reflux | ---- | 5-45 | 80-95 | [51] |
| Na ₂ CaP ₂ O ₇ | H ₂ O | reflux | ---- | 10-15 | 86-95 | [52] |
| FSM-16/AEPC-SO ₃ H | EtOH/H ₂ O | 80 | ---- | 25-240 | 62-89 | [53] |
| KF/Al ₂ O ₃ | EtOH | 27-34 | Ultrasound | 20-240 | 81-98 | [54] |
| NaBr | ---- | 70-85 | Microwave | 10-15 | 80-95 | [55] |
| TFE | TFE | reflux | ---- | 300 | 80-95 | [56] |
| Na ₂ SeO ₄ | EtOH/H ₂ O | reflux | ---- | 40-210 | 80-98 | [57] |
| Basic ionic liquid | EtOH/H ₂ O | reflux | ---- | 5-240 | 50-94 | [58] |
| GO-SiC ₃ -NH ₃ -H ₂ PW | H ₂ O | Reflux | ----- | 4-15 | 88-98 | This work |

The recovery of recycled GO-SiC₃-NH₃-H₂PW was also explored in the synthesis of compound **4a**. After completing the reaction, the catalyst was easily recovered by hot filtration of the reaction mixture and directly reused for the next

turn after washing with hot ethanol and dried at 70 °C under vacuum for 2 h. As shown in Figure 1, the catalyst could be used five times without considerable decrease in activity.

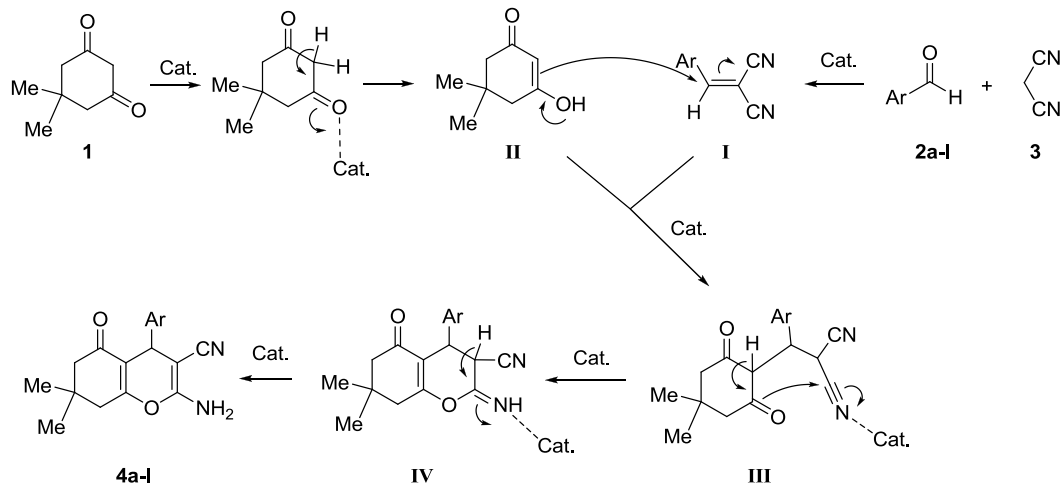
**Figure 1.** Reusability of GO-SiC₃-NH₃-H₂PW for the synthesis of compound **4a**

At the end, a plausible pathway can be considered as shown in Scheme 3. The olefin **[I]** is obtained by condensation of aromatic

aldehydes **2a-l** and malononitrile **3** which then reacts with enol intermediate **[II]**, obtained by tautomerization of dimedone **1**, to afford the

intermediate [III]. The later intermediate was then cyclized to give the final products **4a-l** through the intermediate [IV]. The catalyst GO-SiC₃-NH₃-H₂PW \equiv Cat. assists the formation of

the intermediates; however, none of the intermediates could be isolated under the reaction conditions.



Scheme 3. Plausible pathway for the formation of tetrahydrobenzo[*b*]pyrans using GO-SiC₃-NH₃-H₂PW \equiv Cat. as catalyst

Conclusion

In this work, the reaction of dimedone, aryl aldehydes, and malononitrile was effectively catalyzed by newly prepared GO-SiC₃-NH₃-H₂PW in water as green solvent at reflux temperature, producing the tetrahydrobenzo[*b*]pyrans in 88-98% yields over 4-15 min. Electron-poor aldehydes reacted slightly faster and gave the products high yields. Furthermore, the catalyst could be simply recovered, and used five times without a significant decrease in the activity (97, 96, 95, 95, and 94 in first to fifth use, respectively). The method was also found to be advantageous as it was done in water, operating under environmentally friendly conditions.

Acknowledgement

This research was supported by the Islamic Azad University, Mashhad Branch, Iran.

Conflict of Interest

We have no conflicts of interest to disclose.

References

- [1] S. Sajjadifar, S. Rezayati, *Chem. Pap.*, **2014**, *68*, 531-539.
- [2] A. Fihri, C. Len, R.S. Varma, A. Solhy, *Coord. Chem. Rev.*, **2017**, *347*, 48-76.
- [3] A. Davoodnia, R. Mahjoobin, N. Tavakoli-Hoseini, *Chin. J. Catal.*, **2014**, *35*, 490-495.
- [4] P. Hu, M. Long, *Appl. Catal. B.*, **2016**, *181*, 103-117.
- [5] E. kazemi, A. Davoodnia, A. Nakhaei, S. Basafa, N. Tavakoli-Hoseini, *Adv. J. Chem. A*, **2018**, *1*, 96-104.
- [6] M.M. Heravi, H. Hamidi, N. Karimi, A. Amouchi, *Adv. J. Chem. A*, **2018**, *1*, 1-6.
- [7] M. Rohaniyan, A. Davoodnia, S.A. Beyramabadi, A. Khojastehnezhad, *Appl. Organometal. Chem.*, **2019**, *33*, Art. No. e4881.
- [8] J. Zhao, Y. Xie, D. Hu, D. Guan, J. Chen, J. Yu, S. He, Y. Lü, H. Liu, S. Bao, L. Wang, *Synth. Met.*, **2015**, *204*, 95-102.
- [9] S. Kumari, A. Shekhar, D.D. Pathak, *RSC Adv.*, **2014**, *4*, 61187-61192.

- [10] N. Shang, C. Feng, H. Zhang, S. Gao, R. Tang, C. Wang, Z. Wang, *Catal. Commun.*, **2013**, *40*, 111–115.
- [11] W.S. Hummers, R.E. Offeman, *J. Am. Chem. Soc.*, **1958**, *80*, 1339–1339.
- [12] J. Zhang, Y. Yang, J. Fang, G.-J. Deng, H. Gong, *Chem. Asian J.*, **2017**, *12*, 2524–2527.
- [13] M. Shaikh, S.K. Singh, S. Khilari, M. Sahu, K.V.S. Ranganath, *Catal. Commun.*, **2018**, *106*, 64–67.
- [14] J. Zhang, S. Li, G.-J. Deng, H. Gong, *ChemCatChem*, **2018**, *10*, 376–380.
- [15] S. Bhattacharya, P. Ghosh, B. Basu, *Tetrahedron Lett.*, **2018**, *59*, 899–903.
- [16] M. Coroş, F. Pogăcean, L. Măgeruşan, C. Socaci, S. Pruneanu, *Frontiers of Materials Science* 2019, *13*, 23–32.
- [17] H. Ahmad, M. Fan, D. Hui, *Composites Part B*, **2018**, *145*, 270–280.
- [18] J. Zhao, Y. Xie, J. Fang, Y. Ling, Y. Gao, X. Liu, Q. Zhang, Q. Xu, H. Xiong, *J. Mater. Sci.*, **2016**, *51*, 10574–10584.
- [19] J. Li, H. Xu, *Talanta*, **2017**, *167*, 623–629.
- [20] M. Keyhaniyan, A. Shiri, H. Eshghi, A. Khojastehnezhad, *New J. Chem.*, **2018**, *42*, 19433–19441.
- [21] A. Khojastehnezhad, M. Bakavoli, A. Javid, M.M. Khakzad Siuki, F. Moeinpour, *Catal. Lett.*, **2019**, *149*, 713–722.
- [22] A. Khojastehnezhad, M. Bakavoli, A. Javid, M.M. Khakzad Siuki, M. Shahidzadeh, *Res. Chem. Intermed.*, **2019**, *45*, 4473–4485.
- [23] H. Su, Z. Li, Q. Huo, J. Guan, Q. Kan, *RSC Adv.*, **2014**, *4*, 9990–9996.
- [24] P.K. Khatri, S. Choudhary, R. Singh, S.L. Jain, O.P. Khatri, *Dalton Trans.*, **2014**, *43*, 8054–8061.
- [25] S. Kumari, A. Shekhar, D.D. Pathak, *RSC Adv.*, **2016**, *6*, 15340–15344.
- [26] D.R. Dreyer, S. Park, C.W. Bielawski, R.S. Ruoff, *Chem. Soc. Rev.*, **2010**, *39*, 228–240.
- [27] S. Verma, M. Aila, S. Kaul, S.L. Jain, *RSC Adv.*, **2014**, *4*, 30598–30604.
- [28] S. Omidi, A. Kakanejadifard, F. Azarbani, *J. Mol. Liq.*, **2017**, *242*, 812–821.
- [29] E. Kowsari, M.R. Chirani, *Carbon*, **2017**, *118*, 384–392.
- [30] Y. Wu, Z. Zhao, M. Chen, Z. Jing, F. Qiu, *Monatsh. Chem.*, **2018**, *149*, 1367–1377.
- [31] Z. Ghadamyari, A. Khojastehnezhad, S.M. Seyedi, A. Shiri, *ChemistrySelect*, **2019**, *4*, 10920–10927.
- [32] E. Doustkhah, S. Rostamnia, *J. Colloid Interface Sci.*, **2016**, *478*, 280–287.
- [33] S. Rayati, E. Khodaei, S. Shokoohi, M. Jafarian, B. Elmi, A. Wojtczak, *Inorg. Chim. Acta*, **2017**, *466*, 520–528.
- [34] M.A. Mar'yasov, V.P. Sheverdov, V.V. Davydova, O.E. Nasakin, *Pharm. Chem. J.*, **2017**, *50*, 798–799.
- [35] I. Fatima, R. Saxena, G. Kharkwal, M.K. Hussain, N. Yadav, K. Hajela, P.L. Sankhwar, A. Dwivedi, *J. Steroid Biochem. Mol. Biol.*, **2013**, *138*, 123–131.
- [36] A.E.F.G. Hammam, O.I. Abd El-Salam, A.M. Mohamed, N.A. Hafez, *Indian J. Chem. Sect. B Org. Chem. Incl. Med. Chem.*, **2005**, *44*, 1887–1893.
- [37] I.J.S. Fairlamb, L.R. Marrison, J.M. Dickinson, F.J. Lu, J.P. Schmidt, *Bioorg. Med. Chem.*, **2004**, *12*, 4285–4299.
- [38] K. Rehse, W. Schinkel, *Arch. Pharm.*, **1983**, *316*, 988–994.
- [39] R.R. Kumar, S. Perumal, P. Senthilkumar, P. Yogeewari, D. Sriram, *Bioorg. Med. Chem. Lett.*, **2007**, *17*, 6459–6462.
- [40] L. Bonsignore, G. Loy, D. Secci, A. Calignano, *Eur. J. Med. Chem.*, **1993**, *28*, 517–520.
- [41] D. Armetso, W.M. Horspool, N. Martin, A. Ramos, C. Seoane, *J. Org. Chem.*, **1989**, *54*, 3069–3072.
- [42] S. Hatakeyama, N. Ochi, H. Numata, S. Takano, *J. Chem. Soc. Chem. Commun.*, **1988**, 1202–1204.
- [43] R.S. Bhosale, C.V. Magar, K.S. Solanke, S.B. Mane, S.S. Choudhary, R.P. Pawar, *Synth. Commun.*, **2007**, *37*, 4353–4357.

- [44] M. Amirnejad, M.R. Naimi-Jamal, H. Tourani, H. Ghafuri, *Monatsh. Chem.*, **2013**, *144*, 1219–1225.
- [45] S. Nemouchi, R. Boulcina, B. Carboni, A. Debache, *C. R. Chim.*, **2012**, *15*, 394–397.
- [46] S. Gurumurthi, V. Sundari, R. Valliappan, *E. J. Chem.*, **2009**, *6*, S466–S472.
- [47] S. Rathod, B. Arbad, M. Lande, *Chin. J. Catal.*, **2010**, *31*, 631–636.
- [48] F. Shirini, M. Safarpour, N. Langarudi, *J. Mol. Liq.*, **2017**, *234*, 268–278.
- [49] L.M. Wang, J.H. Shao, H. Tian, Y.H. Wang, B. Liu, *J. Fluorine Chem.*, **2006**, *127*, 97–100.
- [50] R. Mozafari, F. Heidarizadeh, *Polyhedron*, **2019**, *162*, 263–276.
- [51] B. Maleki, H. Eshghi, M. Barghamadi, N. Nasiri, A. Khojastehnezhad, S.S. Ashrafi, O. Pourshiani, *Res. Chem. Intermed.*, **2016**, *42*, 3071–3093.
- [52] B. Maleki, N. Nasiri, R. Tayebee, A. Khojastehnezhad, H.A. Akhlaghi, *RSC Adv.*, **2016**, *6*, 79128–79134.
- [53] M. Hajjami, F. Gholamian, R.H.E. Hudson, A.M. Sanati, *Catal. Lett.*, **2019**, *149*, 228–247.
- [54] J.T. Li, W.Z. Xu, L.C. Yang, T.S. Li, *Synth. Commun.*, **2004**, *34*, 4565–4571.
- [55] I. Devi, P.J. Bhuyan, *Tetrahedron Lett.*, **2004**, *45*, 8625–8627.
- [56] S. Khaksar, A. Rouhollahpour, S. Mohammadzadeh Talesh, *J. Fluorine Chem.*, **2012**, *141*, 11–15.
- [57] R. Hekmatshoar, S. Majedi, K. Bakhtiari, *Catal. Commun.*, **2008**, *9*, 307–310.
- [58] J. Zheng, Y. Li, *Mendeleev Commun.*, **2011**, *21*, 280–281.
- [59] Z. Hoseini, A. Davoodnia, A. Khojastehnezhad, M. Pordel, *Eurasian Chem. Commun.*, **2020**, *2*, 398–409.
- [60] A. Davoodnia, M. Khashi, N. Tavakoli-Hoseini, *Chin. J. Catal.*, **2013**, *34*, 1173–1178.
- [61] M. Rohaniyan, A. Davoodnia, A. Nakhaei, *Appl. Organomet. Chem.*, **2016**, *30*, 626–629.
- [62] A. Davoodnia, A. Nakhaei, N. Tavakoli-Hoseini, *Z. Naturforsch.*, **2016**, *71b*, 219–225.
- [63] M. Fattahi, A. Davoodnia, M. Pordel, *Russ. J. Gen. Chem.*, **2017**, *87*, 863–867.
- [64] F. Tajfirooz, A. Davoodnia, M. Pordel, M. Ebrahimi, A. Khojastehnezhad, *Appl. Organometal. Chem.*, **2018**, *32*, Art. No. e3930.
- [65] A. Davoodnia, M. Bakavoli, E. Imannezhad, N. Tavakoli-Hoseini, *Indian J. Heterocycl. Chem.*, **2009**, *19*, 89–90.
- [66] M. Khashi, A. Davoodnia, V.S. Prasada Rao Lingam, *Res. Chem. Intermed.*, **2015**, *41*, 5731–5742.
- [67] S. Vazirimehr, A. Davoodnia, M. Nakhaei-Moghaddam, N. Tavakoli-Hoseini, *Heterocycl. Commun.*, **2017**, *23*, 65–70.
- [68] N. Dorostkar-Ahmadi, A. Davoodnia, N. Tavakoli-Hoseini, H. Behmadi, *Indian J. Heterocycl. Chem.*, **2018**, *28*, 543–549.
- [69] N. Dorostkar-Ahmadi, A. Davoodnia, N. Tavakoli-Hoseini, H. Behmadi, *Z. Naturforsch.*, **2019**, *74b*, 175–181.
- [70] A. Chanda, V.V. Fokin, *Chem. Rev.*, **2009**, *109*, 725–748.
- [71] C.I. Herrerias, X. Yao, Z. Li, C.J. Li, *Chem. Rev.*, **2007**, *107*, 2546–2562.
- [72] S. Ribe, P. Wipf, *Chem. Commun.*, **2001**, *4*, 299–307.

HOW TO CITE THIS ARTICLE

Zahra Hoseini, Abolghasem Davoodnia*, Mehdi Pordel. Another Successful Application of Newly Prepared GO-SiC₃-NH₃-H₂PW as Highly Efficient Nanocatalyst for Fast Synthesis of Tetrahydrobenzo[*b*]pyrans. *Adv. J. Chem. A*, 2021, 4(1), 68-77.

DOI: 10.22034/AJCA.2021.259593.1228

URL: http://www.ajchem-a.com/article_123042.html