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$NiO-ZrO_2$ heterogeneously catalysed efficient multicomponent synthesis of polyhydroquinoline derivatives

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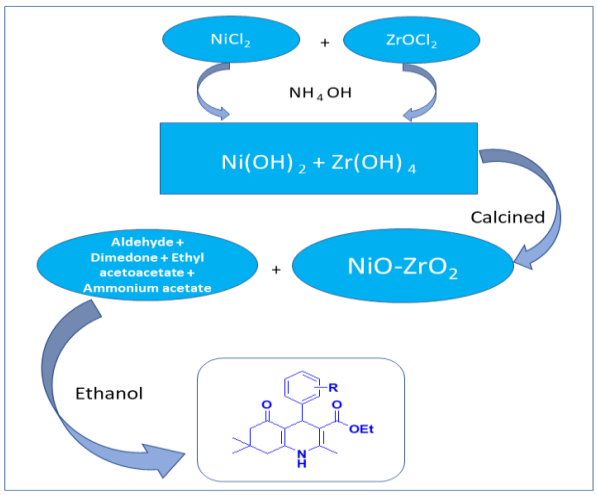

$NiO-ZrO_2$ composite material was synthesized by using a sol-gel method and was characterized by using X-ray diffraction (XRD), transmission electron microscopy (TEM), scanning electron microscopy (SEM), energy-dispersive X-ray spectroscopy (EDS) and Fourier transform infrared spectroscopy (FT-IR). The synthesized $NiO-ZrO_2$ composite material was successfully used as a heterogeneous catalyst for five-component one-pot synthesis of polyhydroquinolines by the cyclo-condensation reaction of aromatic aldehydes, dimedone, ethyl acetoacetate, ammonium acetate and ethanol at room temperature. $NiO-ZrO_2$ catalytic composite material was found to give higher yield when compared to NiO and ZrO_2 separately.

Keywords: Polyhydroquinoline, $NiO-ZrO_2$, sol-gel.

INTRODUCTION

Nickel oxide (NiO) is an important inorganic material having applications in solar cells, capacitors and rechargeable lithium ion batteries [1-4]. Nickel nanoparticles are important for catalysis [5]. Zirconium oxide (ZrO_2) is an important transition metal oxide which shows promising optical and electrical properties [6] and has various applications in catalysts, coatings, fuel cells and sensors [7-10]. Several methods are used for the preparation of ZrO_2 materials like hydrothermal, sol-gel, chemical vapour deposition (CVD) and sputtering [11-14]. The acid-base and redox properties of metal oxides and mixed metal oxides are important in heterogeneous catalysis [15-18]. These oxides can be used in various organic transformations such as oxidation [19-21], dehydration [22], condensation [23], epoxidation [24] and photocatalytic reaction [25, 26].

1,4-Dihydropyirines (1,4-DHPs) and polyhydroquinoline (PHQ) derivatives are important nitrogen-containing heterocycles due to their pharmacological and biological activity [27]. Some of them have antitubercular, anticancer, neurotropic, neuropeptide, YY receptor antagonist, neuroprotective, platelet anti-aggregation, broncho-dilating and antidiabetic activity [28-35]. Multicomponent synthesis of polyhydroquinoline derivatives has been carried out using various catalysts such as TMSCl, ionic liquid, polymer, molecular I_2, CAN, $Yb(OTF)_3$, $HClO_4$-SiO_2, hetero-polyacid, and MCM-41 [36-46]. However, these show several disadvantages such as long reaction time, low yield, use of high-volatile organic solvents and harsh reaction conditions.

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Recent work has been reported on a similar topic, using Four-Component Fusion Protocol with NiO/ZrO_2 for 1,4-dihydropyridines [47], where catalytic material was prepared by the wet impregnation method. Hence, a simple and efficient catalyst can be developed for the synthesis of polyhydroquinoline derivatives.

Considering the advantages of metal oxides it was intended to synthesize a NiO-ZrO_2 composite, characterize it with various sophisticated analytical techniques such as XRD, TEM, SEM-EDS and FT-IR and use it as an efficient catalyst for the synthesis of polyhydroquinoline derivatives.

EXPERIMENTAL

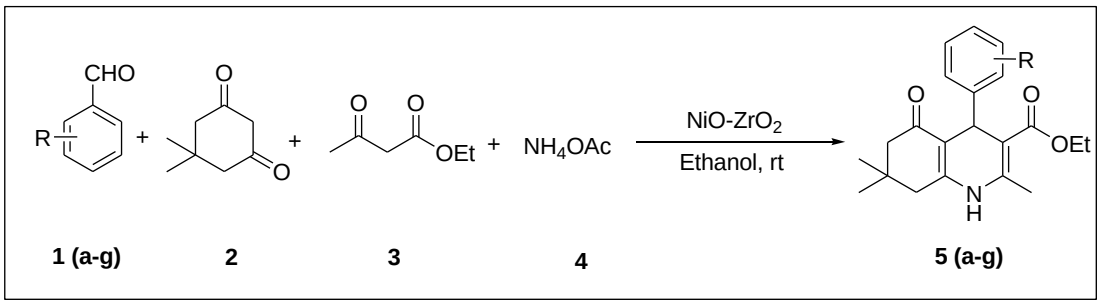
All chemicals were purchased from Merck and used without further purification. The melting points of all derivatives were taken in an open capillary and are uncorrected. ^{1}H NMR spectra were recorded on a 300 MHz FT-NMR spectrometer in $CDCl_3$ as a solvent and chemical shift values were recorded as δ (ppm) relative to tetramethylsilane (Me_4Si) as an internal standard. FT-IR spectra were recorded on a JASCO-FT-IR/4100 instrument, Japan, in KBr. The X-ray diffraction (XRD) patterns were recorded on a Bruker 8D advance X-ray diffractometer using monochromator Cu-Kα radiation of wavelength = 1.5405 Å. Transmission electron microscopy (TEM) was recorded on a CM-200 PHILIPS instrument operated at 200 kV, resolution at 0.23 nm. Scanning electron microscopy image with energy dispersive X-ray spectroscopy (SEM-EDS) was obtained on JEOL JSM-6330 LA operated at 20.0 KV, 1.000 nA.

Synthesis of NiO-ZrO_2 heterogeneous catalyst

NiO-ZrO_2 composite material was synthesized by the sol-gel method. Nickel (II) chloride hexahydrate (0.31 g) and zirconium (IV) oxychloride octahydrate (2.7 g) were dissolved in double distilled water (DDW), then ammonium hydroxide (NH_4OH) was added to obtain a white precipitate. The resulting reaction mixture was stirred continuously at room temperature for 5 hours. The mixture was then filtered and dried at 110°C in an oven for 10 hours so as to form powder material. Finally, the powder material was calcined at 500°C for 2 hours to obtain NiO-ZrO_2 composite material.

General procedure for the synthesis of polyhydroquinoline derivatives

A solution of aldehydes (1 mmol), dimedone (1 mmol), ethyl acetoacetate (1 mmol) and ammonium acetate (3 mmol) was prepared at room temperature in ethanol (15 ml) as solvent and NiO-ZrO_2 (0.05 g) was added as catalyst with continuous stirring for specific time as shown in Scheme 1 and Table 5. The progress of reaction was monitored by TLC using petroleum ether: ethyl acetate (7:3). After completion of reaction, the reaction mixture was filtered. The catalyst was easily separated from the reaction mixture. Filtrate was poured on crashed ice to crystallize the crude product. Crude product was recrystallized from ethanol to obtain the pure product.



Scheme 1. Synthesis of polyhydroquinoline derivatives

Spectroscopic data of compounds

5a. Ethyl 1,4,5,6,7,8-hexahydro-2,7-dimethyl-5-oxo-4-phenylquinoline-3-carboxylate: ^{1}H NMR data in ($CDCl_3$, 300 MHz): δ 1.07 (s, 6H), 1.11 (t, 3H), 2.12-2.23 (m, 4H), 2.32 (s, 3H), (4.07 (q, 2H), 5.04 (s, 1H), 6.64 (s, 1H, NH), 7.10 (d, 2H), 7.18 (q, 1H), 7.30 (d, 2H). ^{13}C NMR (100 MHz, $CDCl_3$): 14.22, 19.24, 28.05, 31.54, 36.60, 40.92, 50.76, 59.82, 106.93, 111.98, 126.03, 128.02, 143.75, 147.12, 148.79, 167.55, 196.11. FT-IR (KBr, cm^{-1}): 3284, 3078, 2928, 1885, 1603, 1484, 1379, 1278, 1212, 1043 cm^{-1}.

5b. Ethyl 4-(4-chlorophenyl)-1,4,5,6,7,8-hexahydro-2,7-dimethyl-5-oxoquinoline-3-carboxylate: ^{1}H NMR data in ($CDCl_3$, 300 MHz): δ 1.07 (s, 6H), 1.12 (t, 3H), 2.12-2.30 (m, 4H), 2.36 (s, 3H), 4.06 (q, 2H), 5.01 (s, 1H), 6.47 (s, 1H, NH), 7.16 (d, 2H), 7.24 (d, 2H). ^{13}C NMR (100 MHz, $CDCl_3$): 14.22, 19.32, 28.05, 31.93, 36.24, 41.38, 50.85, 59.92, 105.67, 111.75, 128.20, 130.95,

131.59, 143.71, 148.56, 149.26, 167.29, 196.12. FT-IR (KBr, cm^{-1}): 3200, 3074, 2925, 1983, 1740, 1600, 1487, 1376, 1276, 1213, 1069 cm^{-1}.

5c. Ethyl 4-(4-fluorophenyl)-1,4,5,6,7,8-hexahydro-2,7-dimethyl-5-oxoquinoline-3-carboxylate: ^{1}H NMR data in ($CDCl_3$, 300 MHz): δ 1.07 (s, 6H), 1.12 (t, 3H), 2.24-237 (m, 4H), 2.63 (s, 3H), 4.07 (q, 2H), 5.04 (s, 1H), 7.10 (s, 1H), 7.30-7.46 (m, 4H), 10.03 (s, 1H, NH). ^{13}C NMR (100 MHz, $CDCl_3$): 14.23, 19.20, 27.44, 31.54, 32.68, 41.40, 52.30, 53.93, 111.20, 112.24, 118.76, 121.21, 122.84, 129.93, 136.25, 142.43, 167.55, 198.62. FT-IR (KBr, cm^{-1}): 3171, 3045, 2931, 1833, 1616, 1502, 1456, 1353, 1204, 1113, 736 cm^{-1}.

5d. Ethyl 1,4,5,6,7,8-hexahydro-2,7,7-trimethyl-4-(3-nitrophenyl)-5-oxoquinoline-3-carboxylate ^{1}H NMR data in ($CDCl_3$, 300 MHz): δ 1.07 (s, 6H), 1.12 (t, 3H), 2.12-2.30 (m, 4H), 2.63 (s, 3H), 4.07 (q, 2H), 5.04 (s, 1H), 6.65 (s, 1H, NH), 7.37 (t, 1H), 7.71 (d, 1H), 7.99 (t, 1H), 8.11 (d, 1H).

5e. Ethyl 1,4,5,6,7,8-hexahydro-2,7,7-trimethyl-5-oxo-4-p-tolylquinoline-3-carboxylate ^{1}H NMR data in ($CDCl_3$, 300 MHz): δ 1.07 (s, 6H), 1.12 (t, 3H), 2.12-2.30 (m, 4H), 2.34 (s, 3H), 2.63 (s, 3H), 4.07 (q, 2H), 5.04 (s, 1H), 6.64 (s, 1H, NH), 7.01 (d, 2H), 7.17 (d, 2H).

5f. Ethyl 4-(furan-2-yl)-1,4,5,6,7,8-hexahydro-2,7,7-trimethyl-5-oxoquinoline-3-carboxylate ^{1}H NMR data in ($CDCl_3$, 300 MHz): δ 1.07 (s, 6H), 1.12 (t, 3H), 2.12-2.30 (m, 4H), 2.63 (s, 3H), 4.07 (q, 2H), 5.04 (s, 1H), 6.64 (s, 1H, NH), 6.01 (s, 1H), 6.20 (s, 1H), 7.16 (s, 1H).

5g. Ethyl 1,4,5,6,7,8-hexahydro-4-(4-hydroxyphenyl)-2,7,7-trimethyl-5-oxoquinoline-3-carboxylate: ^{1}H NMR data in ($CDCl_3$, 300 MHz): δ 1.07 (s, 6H), 1.12 (t, 3H), 2.12-2.30 (m, 4H), 2.63 (s, 3H), 4.07 (q, 2H), 5.04 (s, 1H), 5.60 (s, 1H), 6.64 (s, 1H, NH), 6.64 (d, 2H), 7.16 (d, 2H).

Catalyst characterization

XRD analysis: Figure 1 shows the XRD pattern of NiO-ZrO_2. Catalytic material NiO shows the peaks obtained at 2θ° 25.47, 35.80, 51.88, 64.34, 78.87 corresponding to planes (110), (200), (220), (222) and (332) predicting a cubic structure with a = b = c = 5.341 of NiO and matching with JCPDS card no 780643.

Similarly, ZrO_2 shows a highly crystalline monoclinic phase having a = 5.169, b = 5.232, c = 5.341 showing peaks at 2θ° 29.74, 33.79, 38.12, 49.02, corresponding to the planes (-111), (002), (012), (022) and matching with JCPDS card no 371484. Similarly, some peaks were also observed for a tetragonal phase with a = 3.605, b = 3.606, c = 5.18 showing peaks at 2θ° 24.71, 30.50, 39.53, 57.16. 60.14, 66.03 and 74.48 corresponding to the planes (100), (101), (111), (210), (103), (113), (220) matching for ZrO_2.

Crystallite size of all samples was calculated by using Debye-Scherrer equation. The crystallite size mentioned is the average of crystallite size calculated using FWHM of the three highest intensity peaks. The XRD peaks are very broad; indicating that the NiO nanocrystallites are observed at crystallite size 35.85 nm and for ZrO_2 is 39.01 nm. The rod-like morphology is maintained on a large scale, as evidenced by the TEM image in Figure 2. It clearly indicated that NiO was completely deposited on the surface of ZrO_2.

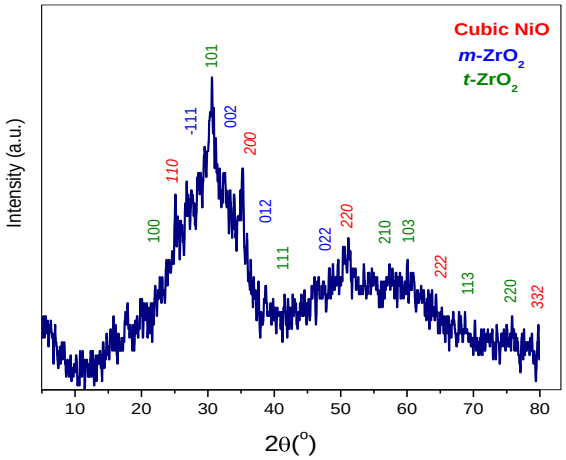


Figure 1. XRD pattern of NiO-ZrO_2

TEM analysis: The TEM images obtained for NiO-ZrO_2 are shown in Figure 2 (a-e). It exhibits uniform size distribution and high crystalline nature with size range of 35 ± 5 nm, matching with the XRD data (35.81 and 39.01 nm for NiO and ZrO_2 respectively). The diffraction spots and rings in the SEAD pattern, shown in figure 2 (f), indicate that the distance from the center of the rings to the diffraction spots is 0.29 nm for the (110) planes of NiO and 0.25 nm for the (101) planes of ZrO_2.

SEM-EDS analysis: The morphology of the catalyst was studied with scanning electron microscopy (SEM). Figure 3 indicates that nickel oxide (NiO) is uniformly deposited on the surface of zirconium oxide (ZrO_2). An increase in the crystalline state with the decrease in grain size was also observed, which results in enhancing the catalytic activity of the prepared material.

Energy-dispersive X-ray spectroscopy (EDS) image of the NiO-ZrO_2 composite material is shown in Figure 4. It reveals that constituent's elements O, Ni and Zr are present as their atomic weights (%) are 41.41, 18.18 and 40.41, respectively. The elemental analysis proved that the minimum required stoichiometric ratio is maintained (Table 1).

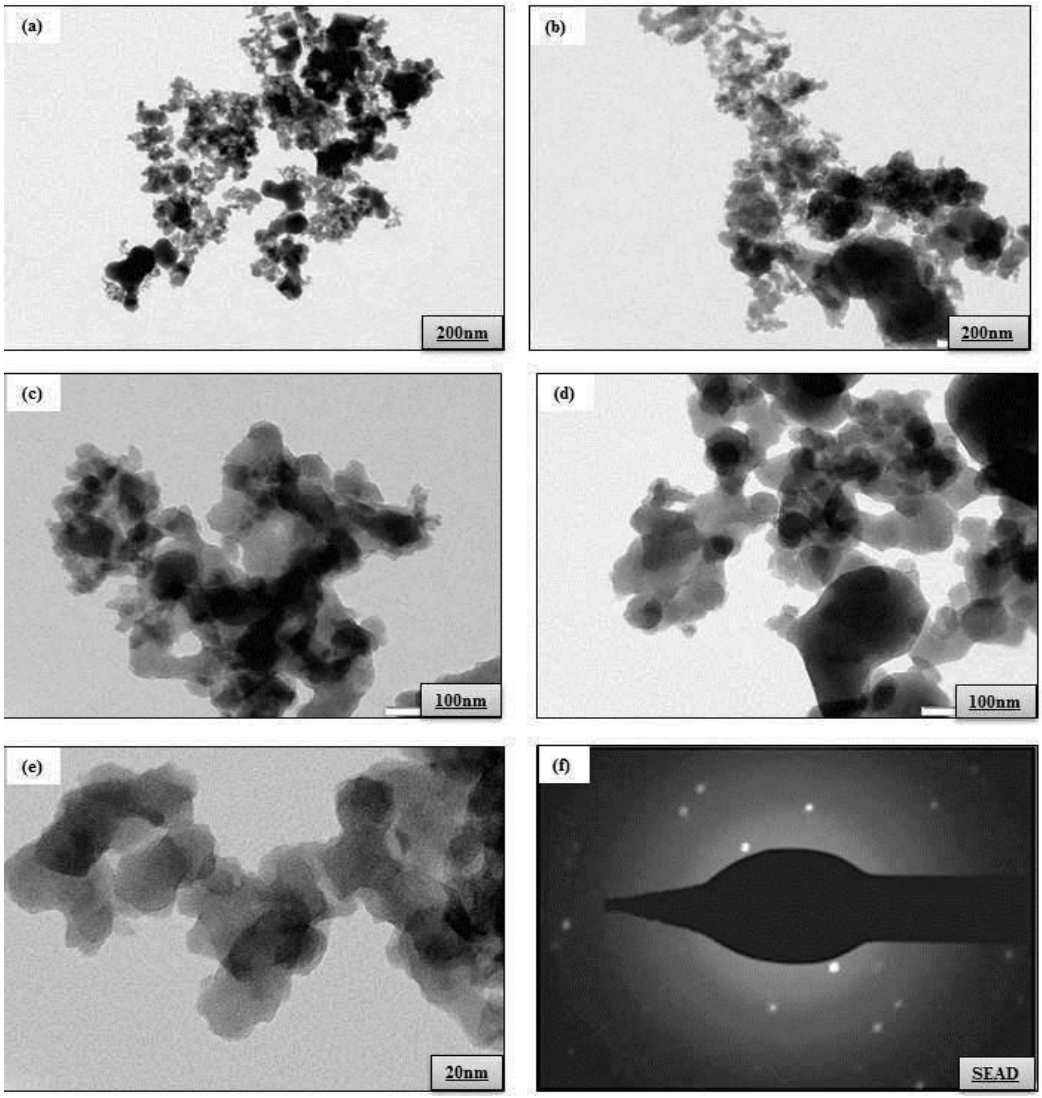


Figure 2. TEM images with different magnifications at (a, b) 200 nm, (c, d) 100 nm, (e) 20 nm and (f) SEAD pattern of NiO-ZrO_2

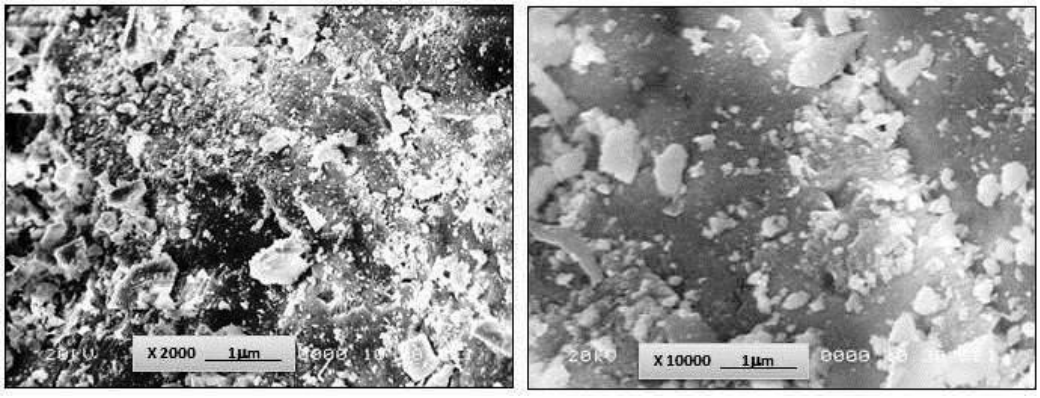


Figure 3. SEM images of NiO-ZrO_2 with different magnifications

Table 1. EDS pattern of NiO-ZrO_2

Element	Weight %	Atomic %
O	41.41	68.12
Ni	18.18	20.22
Zr	40.41	11.66
Total	100.00	100.0

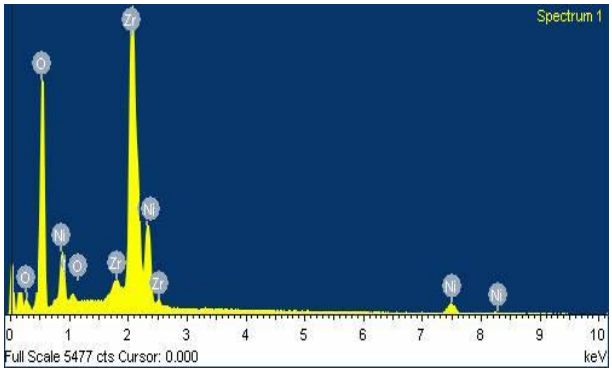


Figure 4. EDS image of NiO-ZrO_2.

FTIR analysis: Figure 5 shows the FTIR spectrum of the NiO-ZrO_2 catalytic material. The broad absorption band at 3106 cm^{-1} can be assigned mainly to Ni-OH and Zr-OH stretching modes and that at 1641 cm^{-1} - to the bending mode of water. The strong and intense band observed at 1090 $cm^{-1}$is due to Ni-O and Zr-O stretching. The band observed at 740 cm^{-1} is assigned to Ni-O-Ni, Zr-O-Zr bending vibrations. Similarly, sharp band are seen at 603 cm^{-1}, which corresponds to the formation of bonds between NiO-ZrO_2 materials.

RESULTS AND DISCUSSION

After characterization and confirmation of the NiO-ZrO_2 nanocomposite material, it was tested as a heterogeneous catalyst for the synthesis by organic transformation of polyhydroquinoline derivatives. Good to excellent yield of all derivatives was established.

We performed a schematic study of the same reaction by varying the solvent, catalyst type and amount, The syntheses of polyhydroquinoline were done using benzaldehyde, dimedone, ethyl acetoacetate, ammonium acetate and ethanol as a model reaction. The results obtained using various catalysts are summarised in Table 2. NiO and ZrO_2 show better yields but time span also increases (entries 1 and 2).

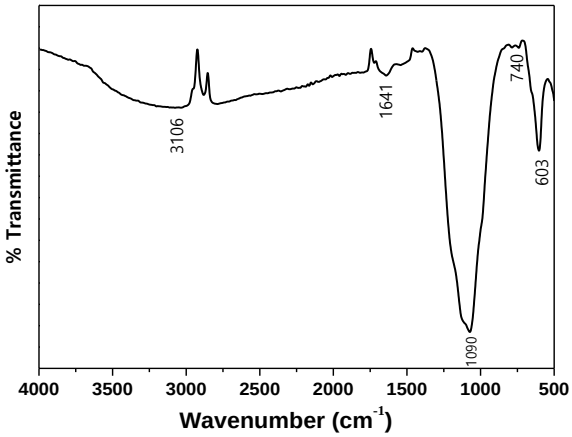


Figure 5. FTIR spectrum of NiO-ZrO_2

NiO-ZrO_2 mixed metal oxides show good to excellent yields of the desired product, shorter reaction times and. improved catalytic stability for the reaction (entry 3). Hence, we have selected the NiO-ZrO_2 catalyst for the synthesis of various derivatives.

The effect of different solvents such as water, ethanol: water (1:1), acetonitrile and ethanol is shown in Table 3. The use for water gives poor yields of the product (entry 1); ethanol: water (1:1) and acetonitrile gave moderate yields (entries 2 and 4) while ethanol as a solvent gave good to excellent yields of the product and also reduced the reaction rate (entry 3). Ethanol solvent shows better activity; therefore, ethanol was selected as a solvent for the synthesis of polyhydroquinoline derivatives.

Table 2. Study of various catalysts for the synthesis of polyhydroquinoline

Entry	Catalyst	Time (min)[a]	Yield (%)[b]
1	NiO	200	85
2	ZrO_2	190	88
3	NiO-ZrO_2	60	93

[a]Reaction conditions: benzaldehyde (1 mmol), dimedone (1 mmol), ethyl acetoacetate (1 mmol), ammonium acetate (3 mmol), ethanol (15 ml) and catalyst (0.05g). [b]Isolated yields.

After studying the effect of solvent we examined how much amount of catalyst is needed for the reaction. In the absence of catalyst, no product was obtained in 250 min. We have studied different weights of catalyst such as 0.03, 0.04, 0.05, 0.06, 0.1, and 0.15 g (see Table 4). The amounts 0.03 and 0.04 g of catalyst showed poor catalytic activity for the present reaction (entries 2 and 3).

Table 3. Study of various solvents for the synthesis of polyhydroquinoline

Entry	Solvent	Time (min)[a]	Yield (%)[b]
1	Water	250	55
2	Ehanol:Water	150	80
3	Ethanol	60	93
4	Acetonitrile	160	75

[a]Reaction conditions: benzaldehyde (1 mmol), dimedone (1 mmol), ethyl acetoacetate (1 mmol), ammonium acetate (3 mmol), ethanol (15 ml) and catalyst (0.05g). [b]Isolated yields.

Catalyst amounts of 0.05, 0.06, 0.1 and 0.15 g showed good to excellent yields of the product (entries 4, 5, 6 and 7). Catalyst amount of 0.05 g was considered sufficient for the reaction.

After optimizing the solvent, catalyst type and amount, we studied the synthesis of polyhydroquinolines. The results are shown in Table 5.

Table 4. Amount of catalyst for the synthesis of polyhydroquinoline

Entry	Catalyst amount (g)	Time (min)[a]	Yield (%)[b]
1	-	250	-
2	0.03	80	78
3	0.04	75	83
4	0.05	60	93
5	0.06	60	93
6	0.1	60	93
7	0.15	60	93

[a]Reaction conditions: benzaldehyde (1 mmol), dimedone (1 mmol), ethyl acetoacetate (1 mmol), ammonium acetate (3 mmol) and ethanol (15 ml). [b]Isolated yields.

The main advantage in this reaction is the recycling and reusability of the catalytic material. The catalyst was separated, washed and dried before next catalytic activity for the reaction. The catalyst reused five times does not decrease its catalytic activity for the reaction (Table 5, entry 5a).

Table 5. Synthesis of polyhydroquinoline using NiO-ZrO_2 catalyst[a]

Entry	R	Time (min)	Yield (%)[b]	M.P. (°C)
5a	Ph	60	93(92,91,91,90,90 [c])	202-204
5b	*p*-Cl-C_6H_4	60	92	240-242
5c	*p*-F-C_6H_4	75	88	180-184
5d	*m*-NO_2-C_6H_4	80	85	175-177
5e	*p*-Me-C_6H_4	85	88	260-262
5f	2-Furyl	70	85	245-247
5g	*p*-OH-C_6H_4	70	86	234-236
5h	Indol-3-carboxyl	180	No reaction	

[a]Reaction conditions: benzaldehyde (1 mmol), dimedone (1 mmol), ethyl acetoacetate (1 mmol), ammonium acetate (3 mmol), ethanol (15 ml) and catalyst (0.05 g). [b]Isolated yields. [c]Yield after consecutive cycles.

CONCLUSION

We developed an efficient method for synthesis of polyhydroquinolines by the cyclo-condensation reaction of aromatic aldehydes, dimedone, ethyl acetoacetate, ammonium acetate and ethanol at room temperature. NiO-ZrO_2 catalytic material was synthesized by the sol-gel method. Prepared material was characterized by XRD, TEM, SEM-EDS and FT-IR. Present method shows several advantages such as high yields, non-toxicity, easy recovery and reuse of the catalyst.

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REFERENCES

1. Y. Wu, Y. He, T. Wu, T. Chen, W. Weng, H. Wan, *Mater. Lett.*, **61,** 3174 (2007).
2. J. Bandara, H. Weerasinghe, *Sol. Energy Mater. Sol. Cells*, **85**, 385 (2005).
3. F-b. Zhang, Y-k. Li, H. Zhou, *Mater. Chem. Phys.*, **83**, 260 (2004).

4. R. A. Kumar, A. L. C-J. Tuan Park, J. Kim, *Ceram. Int.*, **39**, 6611 (2013).
5. S. B. Sapkal, K. F. Shelke, B. B. Shingate, M. S. Shingare, *Tetrahedron Letters*, **50**, 1754 (2009).
6. M. Jafarpour, E. Rezapour, M. Ghahramaninezhad, A. Rezaeifard, *New J. Chem.*, **38** (2), 676 (2014).
7. X. Li, Ch. Ni, Ch. Yao, Zh. Chen, *Appl. Catal. B: Environ.*, **462,** 112 (2012).
8. R. Vassen, X. Cao, F. Tietz, D. Basu, D. Stover, *J. Am. Cer. Soc.*, **83**, 2023 (2000).
9. B. C. H. Steele, A. Heinzel, *Nature*, **414**, 345 (2001).
10. B. G. Mohammad, M.-B. Leila, *J. Electroanal. Chem.*, **33**, 712 (2014).
11. M. N. Tahir, L. Gorgishvili, J. Li, T. Gorelik, U. Kolb, L. Nasdala, W. Tremel, *Solid State Sci.*, **9,** 1105 (2007).
12. G. Ehrhart, B. Capoen, O. Robbe, P. Boy, S. Turrell, M. Bouazaoui, *Thin Solid Films*, **227,** 496 (2006).
13. A. M. Torres-Huerta, M. A. Dominguez-Crespo, E. Ramirez-Meneses, J. R. Vargas-Garcia, *Appl. Surf. Sci.*, **255,** 4792 (2009).
14. C. Rozo, D. Jaque, L. F. Fonseca, J. G. Sole, *J. Lumin.*, **128,** 1197 (2008).
15. H. H. Kung, *Transition Metal Oxides: Surface Chemistry and Catalysis*, **45,** 1 (1989).
16. V. E. Henrich, P. A. Cox, The surface science of metal oxides; Cambridge University Press, Cambridge, UK, 1994.
17. C. Noguera, Physics and chemistry at oxide surface; Cambridge University Press, Cambridge, UK, 1996.
18. B. M. Reddy, Redox properties of metal oxides, in: Metal oxides chemistry and application, J. L. G. Fierro (ed.), Marcel Dekker Inc., 1990.
19. I. Spasova, G. Ivanov, V. Georgescu, D. J. Mehandjiev, *J. University of Chemical Technology and Metallurgy*, **41,** 225 (2006).
20. W. Feng, X. Jie, X. Li, L. Zhou, *Adv. Synth. Catal.* **347,** 1987 (2005).
21. R. Lagnathan, R. Mahalakshmy, B. Viswanathan, *Bull. Catal. Soc. India*, **7,** 50 (2008).
22. M. B. Gawande, R. V. Jayaram, *Catal. Commun.*, **7,** 933 (2006).
23. T.-F. Susana, P. Roberto, D. A. Gloriya, *Reaction Kinetics and Catalysis Letters*, **92,** 361 (2007).
24. Z. Fakhroueian, F. Farzaneh, M. Ghandi, *J. Sci. Islamic Republic of Iran*, **18,** 303 (2007).
25. J. Jose, J. Ameta, P. B. Punjabi, *Bull. Catal. Soc. India*, **6,** 110 (2007).
26. A. B. Gambhire, M. K. Lande, B. R. Arbad, *Bull. Catal. Soc. India*, **7,** 28 (2008).
27. R. Shan, C. Velazquez, E. E. Knaus, *J. Med. Chem.*, **47,** 254 (2004).
28. P. S. Eharkar, B. Desai, H. Gaveria, B. Varu, R. Loriya, Y. Naliapara, A. Shah, V. M. Kulkarni, *J. Med. Chem.*, **45,** 4858 (2002).
29. T. Tsuruo, H. Iida, M. Nojiri, S. Tsukagoshi, Y. Sakurai, *Cancer Res.*, **43,** 2905 (1983).
30. A. Krauze, S. Germane, O. Eberlins, I. Sturms, V. Klusa, G. Duburs, *Eur. J. Med. Chem.*, **34,** 301 (1999).
31. G. S. Poindexter, M. A. Bruce, J. G. Breitenbucher, M. A. Higgins, S. Y. Sit, J. L. Romine, S. W. Martin, S. A. Ward, R. T. McGovern, W. Clarke, J. Russell, I. Antal-Zimanyi, *Bioorg. Med. Chem.*, **12,** 507 (2004).
32. V. Klusa, *Drugs Future*, **20,** 135 (1995).
33. R. G. Bretzel, C. C. Bollen, E. Maeser, K. F. Federlin, *Am. J. Kidney Dis.*, **21,** 53 (1993).
34. R. W. Chapman, G. Danko, M. I. Siegels, *Pharmacology*, **29,** 282 (1984).
35. A. K. Ogawa, C. A. Willoughby, R. Bergeron, K. P. Ellsworth, W. M. Geissler, R. W. Myer, J. Yao, G. Harris, K. T. Chapman, *Bioorg. Med. Chem. Lett.*, **13,** 3405 (2003).
36. S-J. Tu, J-F. Zhou, X. Deng, P-J. Cai, H. Wan, J-C. Feng, *Chin. J. Org. Chem.*, **21,** 313 (2001).
37. G. Sabitha, G. S. K. K. Reddy, C. S. Reddy, J. S. Yadav, *Tetrahedron Lett.*, **44,** 4129 (2003).
38. S-J. Ji, Z-Q. Jiang, J. Lu, T-P. Loh, *Synlett.*, **5,** 831 (2004).
39. J. G, Breitenbucher, G. Figliozzi, *Tetrahedron Lett*., **41**, 4311 (2000).
40. A. Dondoni, A. Massi, E. Minghini, V. Bertolasi, *Tetrahedron*, **60,** 2311 (2004).
41. S. Ko, M. N. V. Sastry, C. Lin, C-F. Yao, *Tetrahedron Lett.*, **46,** 5771 (2005).
42. S. Ko, C-F. Yao, *Tetrahedron*, **62**, 7293 (2006).
43. L-M. Wang, J. Sheng, L. Zhang, J-W. Han, Z-Y. Fan, H. Tian, C-T. Qian, *Tetrahedron*, **61,** 1539 (2005).
44. M. Maheswara, V. Siddaiah, G. L. V. Damu, C. V. Rao, *Arkivoc*, **2,** 201 (2006).
45. M. M. Heravi, K. Bakhtiari, N. M. Javadi, F. F. Bamoharram, M. Saeedi, H. A. Oskooie, *J. Mol. Catal. A: Chem.*, **264,** 50 (2007).
46. L. Nagarapu, M. D. Kumari, N. V. Kumari, S. Kantevari, *Catal. Commun*. **8,** 1871 (2007).
47. S. Bhaskaruni, S. Maddila, W. E. van Zyl, S. B. Jonnalagadda, *ACS Omega*, **4**, 21187 (2019).