Peace Research & Knowledge Hub

International Journal of Chemistry, Pharmacy & Technology
Vol. 2, No.2, pp-74-81, 2017

ZrO_2 NANOPARTICLES: AN EFFICIENT CATALYST FOR THE MULTI-COMPONENT SYNTHESIS OF 4*H*-PYRIMIDO [2,1-*b*] BENZOTHIAZOLE

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Received 11-02-17; Revised & Accepted: 05-03-17

ABSTRACT

ZrO_2 NPs were used for the multi-component synthesis of functionalized 4*H*-pyrimido [2,1-*b*] benzothiazole in aqueous alcohol as a green solvent, which efficiently performed for a good catalytic activity up to 4th cycles of synthesized product. Green strategies have proven the non-hazardous, non-toxic and inexpensive; competing for high yield, reusability, short reaction time and simple workup procedure. The nucleophilic attack to carbon-hetero multiple bond of α, β unsaturated carbonyl ester and intra-molecular cyclisation was carried out to high yield. The structure of products was substantiated by ^{1}HNMR, ^{13}CNMR and Mass spectrometry. The synthesized ZrO_2 NPs were characterized by XRD, FTIR and UV-Visible spectra.

Keywords: ZrO_2 NPs, 4*H*-pyrimido [2,1-b] benzothiazole, Green solvent, Multi-component

INTRODUCTION

The benzothiazole scaffold is a central part of many biologically active compounds with potential medicinal use as antimicrobial properties [1-3], anti-allergy [4], anti-tumor, anti-viral activities [5], anti-fungicidal, anti-herbicidal activities utilized in the chemotherapy of characinoid patients[6] and ligands receptor [7,8].

Now days, multicomponent reactions (MCRs) are accepted worldwide as an important tool for the synthesis of various products and intermediates in medicinal and combinatorial chemistry [9]. These reactions avoid time consuming and multistep [10] process, have been proven to be graceful and rapid way to show advantage of atom economy and high selectivity [11]. The ZrO_2 NPs have much more attention in material science due to its specific optical and electrical properties [12]. In catalysis, ZrO_2 NPs were mostly used as a solid support [13], photo catalyst [14], synthesized catalyst [15] and a few reports for elementary organic transformations [16]. However, ZrO_2 NPs yet nothave been applied in MCRs for the synthesis of functionalized 4*H*-pyrimido [2, 1-b] benzothiazole.In past functionalized benzothiazole was synthesizedusing Lewis acids $AlCl_3$[17], TBAHS [18], hydrotalcite [19], N,N-dichloobis (2,4,6 tri chlorophenyl) urea [20], teramethyl guanidinium trifluroacetate (TMGT) [21] and FeF_3 [22].To the best of our literature survey, there is no efficient general multi-component protocol for the synthesis of benzothiazole derivatives using ZrO_2 nanoparticle (NPs) in green solvent with very short reaction time. The use of ZrO_2 NPs could be more effective due to their reactivity and large surface area. Being a dielectric material, ZrO_2 has a stable tetragonal phase structure in the temperature range of 25–1000 ºC. The conductivity of this

pure ZrO_2 material is only 10^{-7} s/cm at 1000 ºC, which is close to the conductivity of insulating material [23]. The present work is an effort in which synthesis and characterized of ZrO_2 NPs were used in water-ethanol, found that best catalyst solvent combination for the synthesis of synthesis of 4*H*-pyrimido [2,1-*b]* benzothiazole derivatives (Figure **1**, Scheme **1**). To explored the catalytic activity of ZrO_2 NPs in elementary organic transformations [24, 25]. Here in we first time attempt of for the synthesis of reported benzothiazole derivativesby multi-component approach. Thus we wish to report, a simple and efficient protocol for the synthesis of benzothiazole derivatives by using ZrO_2 NPs as a heterogeneous catalyst in combination of water-ethanol as solvent.

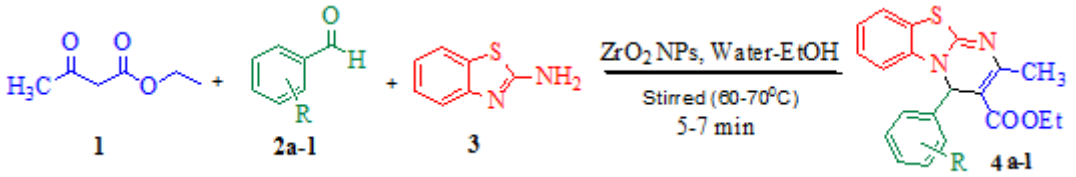


Figure 1: Scheme 1. Functionalized 4*H*-pyrimido [2,1-*b*] benzothiazole

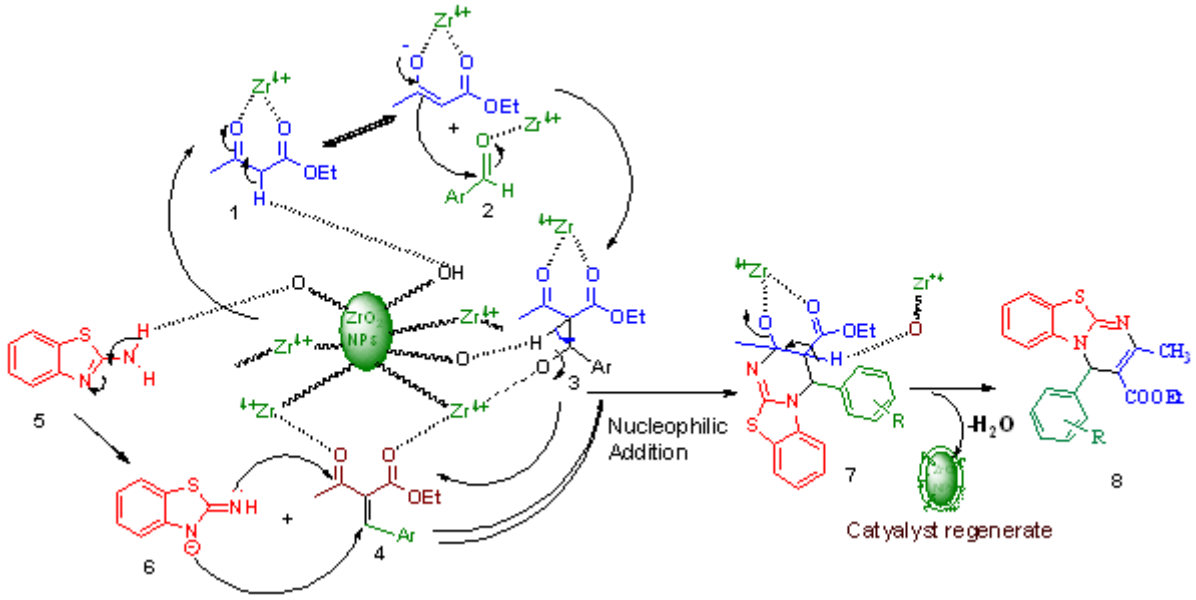


Figure 2: Scheme 2 Possible mechanism of 4*H*-pyrimido [2,1-*b*] benzothiazole derivatives using of ZrO_2NP's

MATERIALS AND METHODS

Chemicals were purchased from Sigma Aldrich and prepared ZrO_2 NPs were characterized by UV-Visible, FT-IR and X-ray diffraction. The synthesized products; Melting points were recorded on SRS Optimelt melting point apparatus and are uncorrected. ^{1}H NMR and ^{13}CNMR spectra were recorded on Varian 300 MHz spectrophotometer in $CDCl_3$. Mass spectra were recorded QUART-MASS JEOL-Accu TOF JMS-T 100LC Mass spectrometer. The ZrO_2 NPs catalyst was characterized by UV-Visible (Figure 4), FT-IR (Figure 5) and X-ray diffraction (Figure 6).

Method for the preparation of tetragonal ZrO_2 NPs: ZrO_2 nanoparticles (NPs) have been synthesized by dissociation of $ZrO_2Cl_2.8H_2O$ in a basic medium (pH ~9 to 11) at low temperature without adding any stabilizer. For synthesizing ZrO_2 NPs, 40 ml 0.05*M* NaOH solution in distilled water was slowly added in 100 ml 0.01*M* solution of $ZrO_2Cl_2.8H_2O$ in methanol-water (1:1) at ~0 to ~5 °C with continuous stirring. After completion of the reaction (~50-60 min) the sol solution was

refluxed at 100^{o}C for over a day with vigorous stirring. Sequentially, the solid and solution phases were separated by centrifugation and the solids were washed with dilute solution of NH_4NO_3 until negative test for chloride ion followed by washed with de-ionized water (2x20 ml) and ethanol (2x5 ml). Prepared solids were dried well and then Calcinated at 500^{o}C for 5 hours. The formation of Nano-sized particles was confirmed by powder XRD studies.

General procedure for the synthesis of compounds (4a-l): A mixture of ethyl aceto acetate 1 (1m mol), aromatic aldehyde2 (1.2 m mol), benzo[d]thiazol-2-amine 3 (1m mol) and ZrO_2 NPs (10 mol %) in 8 ml ethanol water (4:1) was stirred at 60-70 ºC temperature for 5-7 min until the reaction mixture solidified. The progress of the reaction was monitored by TLC. After completion of reaction, the solvent was removed under reduced pressure; crude product was stirred with 4-6 ml methanol at 60-70 ºC for 5 min followed by the simple filtration to expel catalyst out by simple filtration and the solid compound was purified by recrystallization from absolute ethanol to give product with an excellent yield (92-98 %).

Reusability of the catalyst: The recovered catalyst from the reaction mixture during the synthesis of ethyl 2-methyl-4-phenyl-4*H*-benzo[4,5]thiazolo[3,2-a]pyrimidine-3-carboxylate derivative was then washed with hot methanol (14 ml) followed by hot ethanol (6 ml) and finally dried well and reused for subsequent runs. The catalytic activity of ZrO_2 NPs remains unchanged even after 8th cycles (Figure 3).

Characterization techniques: UV-Visible spectroscopy: The UV-Visible spectra of pure ZrO_2 NPs and ZrO_2 NPs after 8th cycle were recorded on Scinco UV–vis scanning spectrometer (ModelS-4100) with the wavelength λ= ~260 nm. UV near-band edge (NBE) emission of ZrO_2 was observed at range between 454 nm - 465 nm which was equivalent to band gap of 4.74 eV. The electronic band structure of ZrO_2 is strongly influenced by the hybridization of Zr-4d orbital and O-2p orbital [27]. Wavelength of pure ZrO_2 NPs; 260 nm and after 8thcycle; 262 nm was observed (Figure 4). The interaction of benzothiazole derivativeswith ZrO_2 NPs has noticeable effect in its band structure as revealed by UV–Vis spectroscopy.

FT-IR: Fourier transform infra-red spectra were recorded on a Perkin-Elmer FT Spectrophotometer in KBr disc. The FT-IR analysis of ZrO_2 NPs the starching vibrations can be studied in terms of transmittance % against wave numbers. The distinctive band around 3300-3375 cm^{-1} can be seen which indicated the transmittance due to O-H absorptions. 935-948 cm^{-1} and a broad band near 1640-1649 cm^{-1} which are associated with the O–H modes of chemisorbed water and/or terminated hydroxides at the surface. The intense absorbance band in FT-IR spectrum were observed in range between 490-498 cm^{-1} and 440-458 cm^{-1} which were attribute to the Zr-O stretching vibrations (Figure 5).

X-ray diffraction: The X-ray diffraction (XRD) patterns was characterized by Philips X'Pert Pro monochromatized diffractometer Cu-Kα radiation (λ = 1.54056 Å). By X-ray diffraction pattern (Figure 2), we observed high intensity peak (103) assigned at 2θ = 30.5° (112) and (101) reflections are present at 2θ = 51.02° and 59.31° respectively. All the reflections are indexed to the characteristic planes of a major tetragonal phase in ZrO_2 crystal system (t-ZrO_2, space group P21/a, JCPDS card No. 37-1484 and 88-1007).Elsewhere, no extra peak was observed corresponding to monoclinic plane. A clear broad peak in powder X-ray diffraction confirmed the formation of nano-sized particles of ZrO_2. The crystallite sizes was estimated from the Debye–Scherrer equation

$$D = \frac{0.9\lambda}{\beta \cos\theta}$$

Where λ is the wavelength of Cu-Kα radiation (1.54056 Å) and β is the full width of the (h k l) peak at the diffracting angle 2θ. The mean crystallite size was estimated between ~20 and ~30 nm from XRD data (Figure 6).

Spectral data for selected compound:

Ethyl4-(4-methoxyphenyl)-2-methyl-4Hbenzo[4,5]thiazolo[3,2-a]pyrimidine-3-carboxylate (4b): mp 141–142 °C; **^{1}H-NMR** ($CDCl_3$, δ ppm)1.27 (t, 3H,CH_3), 4.23 (q, 2H, OCH_3), 2.81 (s, 3H, CH_3), 4.62 (s, 1H, CH), 7.02–7.10 (m, 1H, Ar-H), 7.16–7.24 (m, 3H, Ar-H), 7.33–7.54 (m, 3H, Ar-H), 8.10 (d, 1H, Ar-H); **^{13}C NMR** (75 MHz, $CDCl_3$): δ 23.02, 57.47,59.88, 60.53, 65.55, 102.30, 111.73, 120.70,122.02, 123.92, 126.94, 128.03, 128.40, 129.59,132.50, 140.96, 154.71, 163.40, 166.27; **Mass** EI-MS *m/z* cal. 380.46, *m/z* obs. [M+H] = 381.

Ethyl 4-(2-chlorophenyl)-2-methyl-4*H* benzo[4,5]thiazolo[3,2-a]pyrimidine-3-carboxylate (4f): mp 123–124°C; **^{1}H NMR** ($CDCl_3$, δ ppm):1.27 (t, 3H,CH_3), 4.23 (q, 2H, OCH_3), 2.81 (s, 3H, CH_3), 4.62 (s, 1H, CH), 7.1–7.14 (m, 2H,Ar-H), 7.19–7.28 (m, 2H, Ar-H), 7.33 (d, 1H,Ar-H), 7.41 (d, 2H, Ar-H), 7.56 (d, 1H, Ar-H);**^{13}C NMR** (75 MHz, $CDCl_3$): δ 14.32, 23.57,54.39, 59.83, 102.71, 111.67, 121.83, 123.23,123.97, 126.62, 128.03, 129.37, 129.58, 130.36,131.33, 138.11, 139.51, 155.25, 163.15, 166.3 **Mass** EI-MS *m/z* cal. 384.04, *m/z* obs. [M+H]= 385.

Ethyl 2-methyl-4-(pyridin-2-yl)-4*H* benzo[4,5]thiazolo[3,2-a]pyrimidine-3-carboxylate (4l): mp 141–142 °C; **^{1}H-NMR** ($CDCl_3$, δ ppm)1.27 (t, 3H,CH_3), 4.23 (q, 2H, OCH_3), 2.81 (s, 3H, CH_3), 4.62 (s, 1H, CH), 7.02–7.10 (m, 4H, Ar-H), 7.16–7.24 (m, 4H, Ar-H); **^{13}C NMR** (75 MHz, $CDCl_3$): δ 14.3, 21.2, 59.5, 61.2, 117, 117.2, 120.7, 121.9, 122.2, 122.5, 122.9, 126.3, 136.2, 139.3, 148.7, 154.2, 155.3, 158.2, 167.3 ; **Mass** EI-MS *m/z* cal. 351.46, *m/z* obs. [M+H] = 352.

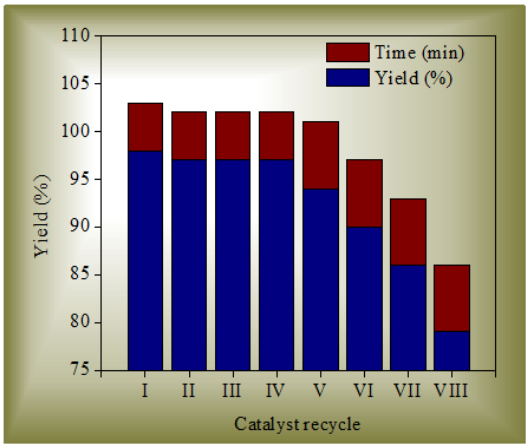


Figure 3: Reusability of ZrO_2 NPs for the synthesis of 4*H*-pyrimido [2,1-*b*] benzothiazole derivatives

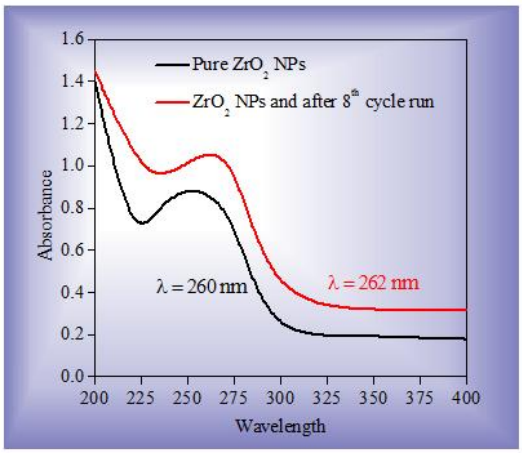


Figure 4: UV-Visible spectra of pure ZrO_2 NPs and after 8th cycle

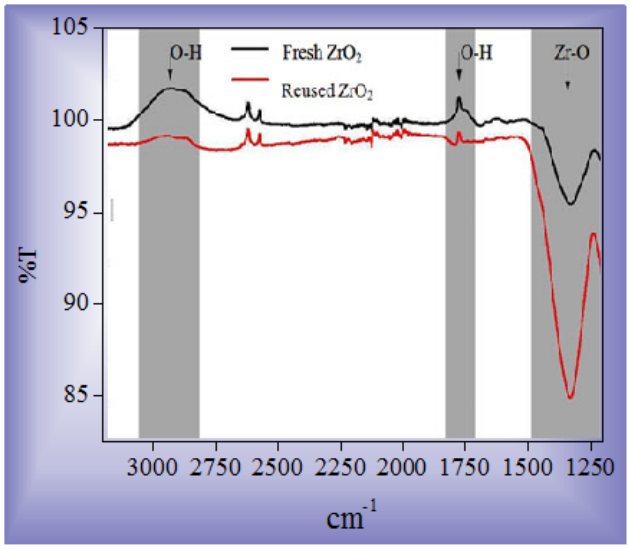


Figure 5: FT-IR spectra of fresh (black line) and reused (red line) ZrO_2NPs

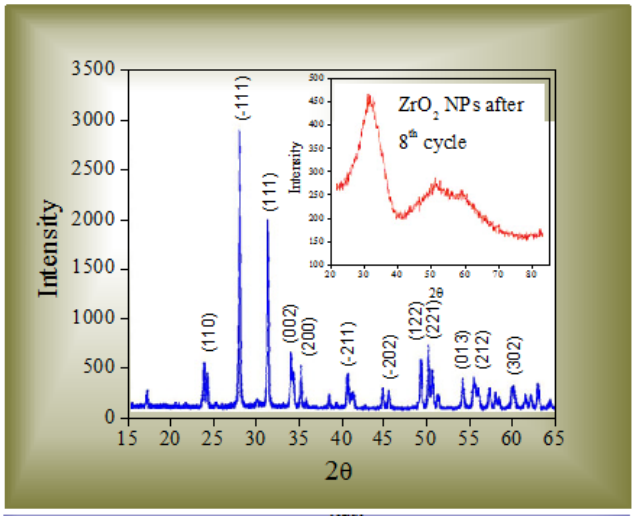


Figure 6: Powder XRD pattern of monoclinic ZrO_2 NPs and ZrO_2NP_S after 8th cycle

RESULTS AND DISCUSSION:

Keeping focus on multicomponent green approach, we have selected ZrO_2 NPs for the optimized reaction condition with various solvents (Table 1). In ethanol, good yield of product was obtained (Table 1, entry 6). On this result, we increased a small amount of water in ethanol as the ethanol-water (in proportion 6:1, 4:1, 2:1) (Table 1, entry 7-12). While changing an amount of water in set of ethanol, the yield product was found to be reduced (Table 1, entry 11 & 12). In ethanol water proportion of 4:1 an excellent yield was obtained in 10 mol % ZrO_2 NPs within 5 min (Table 1, entry 9). This was due to the surface area of the NPs being more active in polar-protic solvents and presence of oxygen vacancies, which were responsible for stability and higher surface activity [26]. Further we focused on significant effect of time and amount of catalyst on yield of reaction; the yield of product was fallen down by decreasing time of reaction, while no significant change was observed by increasing the time of reaction (Table 1, entry no. 9). Further we changed an amount of catalyst to less than 10 mol % does not elevate the yield product (Table 1, entry no 8). Similarly, an amount of catalyst to greater than 10 mole% no significant yield was obtained (Table 1, entry 10). Thus we obtained an excellent yield in less time of reaction using10 mol% of catalyst (Table 1, entry no. 9). Thus, all examples were tested reasonably good to excellent yields in ethanol water

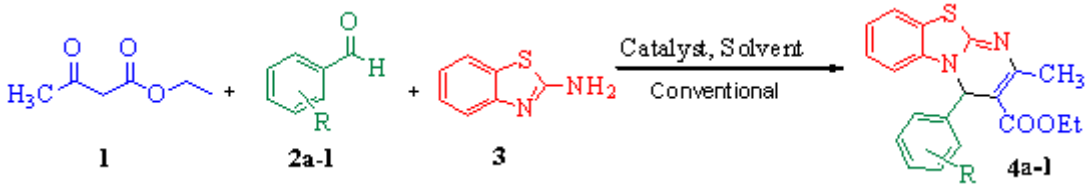
(4:1) with 10 mol % of catalyst (Table 2). The catalyst was investigated by checking its reusability (Figure 3, Table 3), after each cycle, the ZrO_2 NPs were recovered by centrifugation, washed with hot methanol, dried and reused for successive reactions. First four cycles gave better yield and then the yield was gradually decreased, UV-Visible (Figure 4), FTIR (Figure 5) and the powder XRD were obtained (Figure 6). The important feature for these catalysts; surface of catalyst contains active hydroxyl, oxide groups and Zr^{+4} acts as a Lewis acid which are well reported [28]. An active hydroxide and oxide of NPs played an important role for cyclization and condensation reaction because it acts as both function acid as well as base (Figure 2, Scheme 2). Possible mechanism for the synthesis of 4*H*-pyrimido [2,1-*b*] benzothiazole has been described *via* cyclo-condensation reaction using ZrO_2 NPs as catalyst (Scheme 2). Finally the structures of the compound 4 was confirmed by spectral data, the ^{1}HNMR spectrum of compound gave two singlet at 2.81 δ (s, 3H, CH_3) and 4.62 δ (s, 1H, CH-N), due to methyl and CH of ipso aryl moiety; doublet at 7.90-8.10 δ, due to aryl ring; and another multiplet at more than 7 δ ppm due to aromatic ring, ^{13}CNMR were of represented data and by Mass spectrometry (spectral characterization data).

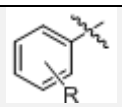
Table 1: Screening of catalyst with solvents, reaction time, and yield for the synthesis 4a

Entry	Catalyst[b]	Mol %	Solvent	Time(min)	Yield[a] (%)
1	Catalyst free	15	Solvent free	120	00
2	ZrO_2 NPs	15	Solvent free	60	35
3	ZrO_2 NPs	10	H_2O	40	58
4	ZrO_2 NPs	10	CH_3CN	30	53
5	ZrO_2 NPs	10	MeOH	12	63
6	ZrO_2 NPs	10	EtOH	7	78
7	ZrO_2 NPs	10	EtOH:H_2O (6:1)	7	83
8	ZrO_2 NPs	5	EtOH:H_2O (4:1)	3,5,6	50,78,78
9	ZrO_2 NPs	10	EtOH:H_2O (4:1)	3,5,6	63,98,98
10	ZrO_2 NPs	15	EtOH:H_2O (4:1)	3,5,6	63,98,98
11	ZrO_2 NPs	10	EtOH:H_2O (2:1)	10	68
12	ZrO_2 NPs	10	EtOH:H_2O (1:1)	10	56

[a]Isolated yield; [b]Reactioncondition: ethyl acetoacetate (1 mmol), aromatic aldehydes (1.2 mmol), 2-aminobenzothiazole (1 mmol), 8 ml water-ethanol, 10 mol% of ZrO_2 NPs.

Table 2: ZrO_2 NPs catalyzed synthesis of functionalized benzothiazole derivatives



Entry	Product		Time(min)	Yield (%)	M.P.(°C)
1	4a	C_6H_5	5	98	171-172
2	4b	4-MeO-C6H4	5	97	141-142
3	4c	4-Me-C6H4	5	95	153-154
4	4d	4-Cl- C6H4	5	96	142-143
5	4e	2-MeO- C6H4	5	96	144-145

6	4f	2-Cl- C6H4	5	94	123-124
7	4g	4-HO-C6H4	7	92	208-209
8	4h	4-F- C6H3	5	97	161-162
9	4i	3-Cl- C6H4	7	96	138-139
10	4j	4-Br- C6H4	7	97	165-166
11	4k	4-NO2- C6H4	5	98	155-156
12	4l	C_5H_4N or (2-Pyridyl)	5	96	163-165

Reaction conditions: All the reaction was carried out in equimolar amounts of each compound in 8 mL of water-ethanol, stirred at 60-70^{0}C.

CONCLUSIONS:

In the present work we have synthesized of functionalized 4*H*-pyrimido [2,1-*b*] benzothiazole by simple method using ZrO_2 NPs catalyst *via* multicomponent reactions in water-ethanol as green solvent. XRD pattern confirmed the formation of nano-sized tetragonal ZrO_2 NPs with no extra peak corresponding to monoclinic plane. Results revealed that the catalytic activity of ZrO_2 NPs first 4th cycles was considered to be very good. In summary, it can be concluded that the ZrO_2 NPs and water-ethanol used to the preparation of benzothiazole is an eco-friendly, readily available solvent and inexpensive. It was found that the reactions afforded very good yields under simple method. ZrO_2 NPs and water-ethanol used for the organic transformations is one of the green strategies due to readily available, non-hazardous, non-toxic and inexpensive solvent.

REFERENCES

[1] Wade J. J., Tose, C. B., Matson C. J., Stelzer, V. L. J. Med. Chem., 1983, 26, 608-611.

[2] Alaimo R. J. J. of Heter. Chem. 1973, 10, 769-772.

[3] Gupta A., Rawat, S. J. Curr. Pharm. Research, 2010, 13, 1.

[4] Bartovic A.,Ilavski, D., Simo, O., Zalibera L., Belicova A., Seman M. Collection of Czechoslo- vak Chemical Communications, 1995, 60, 583-593.

[5] El-Sherbeny M. A. Drug Research, 2000, 50, 848-853.

[6] Kutyrev A.; Kappe T. J. of Heterocyclic Chemistry, 1999, 36, 237-240.

[7] Trapani G., Franco A., Latrofa G., Carotti A., Genchi, G., Serra M., Biggioand G., Liso, G. European Journal of Me- dicinal Chemistry, 1996, 31, 575-587.

[8] Trapani G., Carotti A., Franco A., Latrofa, G., Gench G., Liso G. Euro- peanJ. Med. Chem. 1993, 28, 13-21.

[9] (a) Ramon D. J., Yus M. Angew. Chem. Int. Ed. 2005, 44, 1602-1634.; (b) Domling, A. Chemistry Chemical Review.2006, 106, 17-89.

[10] (a) Damiano, R., Francisco, J. Green Chemistry. 2013, 15, 511-517.; (b) Tietze, L. F. Chemical Review. 1996, 96, 115-136.

[11] (a) Cariou C.; Clarkson G. J.; Shipman M. J. of Org. Chem. 2008, 73, 9762-976.

[12] (a) Garvie R. C., Hannink R. H., Pascoe R. T. Ceramic steel, Nature, 1975,258, 703-704; (b) Mansour N., Mansour K., Stryl, E. W. V., Soileau M. J. J. Appl. Phys. 1990,67, 1475-1477.;(c) Badwal S. P. S., Appl. Phys. A: Mater. Sci. Process. 1990, 50, 449-462.; (d) Phillips, J. M. J. Appl. Phys. 1996, 79, 1829-1848. ;(e) Leon C.; Lucia M. L. Santamaria J. Phys. Rev. B: Condens. Mater. Phys. 1997, 55, 882-887.

[13] (a) Ardiyanti A. R.; Gutierrez A.; Honkela M. L.; Krause, A. O. I.; Heeres H. J. Hydrotreatment of wood-based pyrolysis oil using zirconia-supported mono-and bimetallic (Pt, Pd, Rh) catalysts, Appl. Catal.-A. 2011, 407, 56-66; (b) Guerrero, S.; Araya, P. Wolf, E. E. Methane oxidation on Pd supported on high area zirconia catalysts, Appl. Catal.-A.2006, 298, 243-253.

[14] (a) Chen X., Wang, X., Fu, X., Energy Environ. Sci. 2009, 2, 872-877; (b) Zhang H., Liu K., Cao, H. Zhang X. J. Phys. Chem. C: 2009, 113, 18259-18263.;(c) Chen, C. T., Yang C. B. Ching and Xu, R. Langmuir2008, 24, 8877-8884.; (d) Keskitalo T. J., Niemela, M. K. V., Krause A. O. I. , Langmuir 2007, 23,7612-7619.; (e) Haw, J. F., Zhang, J.,Shimizu, K., Venkatraman, T. N., Luigi, D. P., Song, W.; Barich, D. H.; Nicholas, J. B. J. Am. Chem. Soc.2000, 122, 12561-12570.

[15] Mahmood Q., Afzal A., Siddiqi H. M., Habib A. J. Sol-Gel Sci. Technol. 2013, 67, 670-674.

[16] (a) Nakka Molinari J. E., Wachs I. E. J. Am. Chem. Soc. 2009, 131, 15544-15554.;(b) Tomishig, K.; Ikeda Y.,Sakaihori T. Fujimoto K. J. Cat. 2000, 192, 355-362.; (c) Li W., Huang H., Li H., Zhang, W., Liu, H. Langmuir 2008, 24, 8358-8366.;(d) Jafarpour, M.; Rezapour, E.; Ghahramaninezhad M., Rezaeifard, A. New J. Chem. 2014, 38, 676-682.

[17] Landreau C., Deniaud D., Evain M., Reliquet A. J. Chem. Soc. Perkin Transactions2002, 6, 741-745.

[18] Roy, P. J., Landry, K., Leblanc, Y. Heterocycles, 1997, 45, 2239-2246.

[19] Tanabe, Y., Kawai, A., Yoshida, Y., Ogure, M., Okumura, H. Heterocycles, 1997, 45, 1579-1588.

[20] Sahu P. K., Lal J., Thavaselvam D., Agarwal, D. D. Med Chem Res2012, 21, 3826–3834.

[21] Nagarapu, L., Gaikwad, H. K., Palem, J. D., Venkatesh, R., Bantu, R., Sridhar B. Synth Commun. 2013, 43, 93–104.

[22] Sahu P. K.; Jain R.; Yadava R.; Agarwal D. D. CatalSci Technol, 2012, 2,2465–2475.

[23] Jiafeng Ding, Xinmei Li, Jian Cao, Liyuan Sheng, Linzi Yin and Xuemei Xu, 2014, 202, 232–239.

[24] Nakka L.; Molinari, J. E. andWachs, I. E.2009, 42, 131, 15544-15554.

[25] Tomishige K., Ikeda Y., Sakaihori T. and Fujimoto K. Journal of Catalysis, 2000, 2, 192, 355-362.

[26] Karapetrova E., Platzer R. J. of Ameri. Chem. Soc. 2001, 84,65-70.

[27] Quazi Arif Islam, Mirwasim Raja, Chiranjib Satra and Rajendra Nath Basu. Bulletine of Materials Science, 2015, 38, 6, 1473–1478.

[28] Zhao Y., Li W., Zhang W., Tao K. Cat. Comm.2002, 3, 239-245.