



Research Article

AN EFFICIENT ONE-POT SYNTHESIS OF BENZIMIDAZOLES USING MAGNETICALLY RECOVERABLE CATALYST CHROMIUM DOPED NICKEL COPPER ZINC SPINEL FERRITE

B. L. Shinde ¹, L. A. Dhale ², U. M. Mandle ³, K. S. Lohar ^{2*}

¹ Department of Chemistry, Waghire College, Saswad, Pune, Maharashtra, India

² Department of Chemistry, Shri Krishna Mahavidyalaya Gunjoti, Osmanabad, Maharashtra, India

³ Department of Chemistry, Sangmeshwar College, Solapur, Maharashtra, India

* Corresponding author E-mail address:kslohar@rediffmail.com

Article Received on: 22/04/19 Approved for publication: 14/06/19

DOI: 10.7897/2230-8407.1008245

ABSTRACT

An efficient synthesis of benzimidazoles has been developed via one-pot two-component condensation reaction of o-phenylene diamine and aromatic aldehydes by using magnetic Cr³⁺ doped Ni-Cu-Zn spinel ferrite as catalyst in ethanol. The products obtained with high yields in shorter reaction time. The Cr³⁺ doped Ni-Cu-Zn spinel ferrites synthesized by sol gel auto combustion method and characterized by XRD, IR, TEM and VSM. The XRD patterns confirmed single phase of cubic spinel structure of catalyst. Lattice constant of catalyst decreased from 8.418 Å to 8.406 Å, while percentage porosity increases from 25.4 % to 27.7 % with increase in Cr³⁺ content. The crystallite size decreased from 30.3 nm to 19.1 nm with increase in Cr³⁺ content. Cr³⁺ doped Ni-Cu-Zn spinel ferrites were soft magnetic materials. The catalyst can be robust and easily recoverable using external magnetic field and reused five times with almost the same catalytic activity. The proposed method is advantageous due to its small catalyst loading, short reaction time, magnetically catalyst recovery, catalyst reusability and better yields.

Keywords: Spinel Ferrite, One pot reaction, Benzimidazoles, Aromatic aldehydes

INTRODUCTION

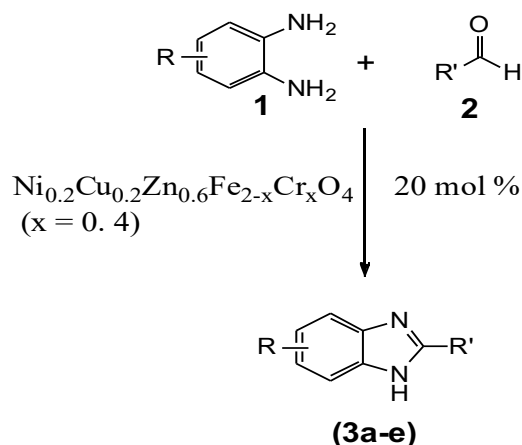
One-pot multi component reactions have more advantages over traditional reactions due to their rapidity, simplicity, atom-economy and shorter synthetic route for the synthesis of biologically active compounds.¹ Multicomponent reactions (MCRs) are the one pot methods in which three or more reactants react to form a single product that includes most or all atoms of the reactants used.² Now days, the use of heterogeneous catalysts has established significant interest in various disciplines, i.e. organic synthesis using heterogeneous catalysts have great advantage of catalyst recycle as compared to homogeneous catalyst.

Magnetite nanoparticles have been immobilized on different catalyst supports, including carbons, silicas, and polymers because of easily retrievable and reusable heterogeneous catalyst they are still have high demand. Iron oxide nanoparticles catalyst is that easily separated using an external magnet, which achieves a simple separation of catalyst without filtration.³ Iron oxides not only show more catalytic activities but also a high degree of chemical consistency and not dissolve in organic solvents. Especially the ferro-spinels have been shown to be selective and active catalysts⁴ and exhibit extra stability under various reaction conditions.⁵ Now a days, functionalized magnetic nanoparticles used as effective catalyst in different chemical reactions including synthesis of 1,1 diacetates⁶, diazepine derivatives⁷, α -amino acids⁸, 1,4-dihydropyridines⁹, etc. For these applications of metal oxides as heterogeneous catalysts, high surface area and accessible porosity are relevant properties. Spinel have been conveniently used as catalysts for a variety of industrially important reactions.

Benzimidazole is a heterocyclic aromatic organic compound, consists of the fusion of benzene and imidazole. Benzimidazoles and their derivatives form a significant class of compounds in organic chemistry attributing to their massive applications in biological and medicinal field. Several benzimidazole derivatives have been reported to exhibit biological activities and wide applications in Medicinal chemistry such as anticancer¹⁰, antihypertensive antiviral¹¹, antitumor¹², anti-allergic agents¹³, antiprotozoal activity¹⁴, diuretic activity¹⁵, selective neuropeptide YY1 receptor antagonists¹⁶ etc. By considering their various biological functions¹⁷, they are used in clinical medicine¹⁸, as anti-ulcer, antitumor and antiviral agent¹⁹.

Because of their wide applications in industrial, pharmacological activity and synthetic fields, different methods have been reported for the synthesis of substituted benzimidazoles by condensation of o-phenylenediamine, and its derivatives with carboxylic acids, or aldehydes. For synthesis of benzimidazole derivatives different Lewis acid catalysts used such as InBr₃²⁰, ZrCl₄, SnCl₄, TiCl₄, HFCl₄²¹, ZrOCl₂.9H₂O²² etc. Conventionally, the synthesis of benzimidazoles involves the condensation of o-phenylenediamine with aldehydes, or carboxylic acid or their derivatives in the presence of catalysts.²³⁻²⁶

In the present work, we are reporting our investigations on Cr³⁺ doped Ni-Cu-Zn spinel ferrite as magnetically recoverable and reusable catalyst for the synthesis of benzimidazoles. An efficient synthesis of benzimidazoles carried via one-pot two-component condensation reaction of o-phenylenediamine, aromatic aldehydes in high yields and short reaction times by using Cr³⁺ doped Ni-Cu-Zn spinel ferrites with as a green, robust and easily recoverable catalyst (Scheme 1).



Scheme 1: Synthesis of substituted benzimidazole (**3a-e**) using o-phenylenediamine (**1**), aromatic aldehydes (**2**), using 20 mol % $\text{Ni}_{0.2}\text{Cu}_{0.2}\text{Zn}_{0.6}\text{Fe}_{2-x}\text{Cr}_x\text{O}_4$ ($x = 0.4$) spinel ferrites as catalyst

MATERIALS AND METHODS

Synthesis of the Catalysts

The $\text{Ni}_{0.2}\text{Cu}_{0.2}\text{Zn}_{0.6}\text{Fe}_{2-x}\text{Cr}_x\text{O}_4$ ($x = 0.0, 0.2$ and 0.4) spinel ferrites were prepared by reported sol gel auto combustion method.²⁷ Analytical grade chromium nitrate, nickel nitrate, copper nitrate, zinc nitrate, iron nitrate and citric acid, were used as raw materials. The metal nitrate solution was prepared in double distilled water, mixed with citric acid solution and ammonia added to adjust pH to 7. The mixed solution was heated on a hot plate at 100°C with constant stirring. The solution turns into viscous gel after evaporation of water. The viscous gel starts frothing on more heating, which gets ignited automatically after few minutes and it burnt through glowing flints. The black colored ash (precursor) formed after completion of auto combustion. The formed precursors then sintered at 500°C for 4 hour to obtain spinel ferrite catalyst.

Characterization of Catalyst

The crystallographic structures were studied by X-ray powder diffraction with $\text{Cu-K}\alpha$ radiation ($\lambda = 1.5405 \text{ \AA}$) by Phillips X-ray diffractometer (Model 3710). The infrared spectra of all the samples were recorded at room temperature in the range 300 to 800 cm^{-1} using Perkin Elmer infrared spectrophotometer. The surface morphology was studied by Philips transmission electron microscope (model CM 200). The magnetic measurements were performed at room temperature using a commercial PARC EG and G VSM 4500 vibrating sample magnetometer.

General Procedure of Synthesis of Benzimidazoles

All the reagents used were of AR grade and were used without further purification. A mixture of o-phenylenediamine (1 mmol) and aldehyde (1.1 mmol) was well stirred with $\text{Ni}_{0.2}\text{Cu}_{0.2}\text{Zn}_{0.6}\text{Fe}_{2-x}\text{Cr}_x\text{O}_4$ ($x = 0.4$) ($20 \text{ mol } \%$) at room temperature for 10 min . After completion, the reaction mixture was diluted with dichloromethane (25 mL) and washed with water. The catalyst removed by fixing the catalyst magnetically at the bottom of the flask with a strong magnet, after which the solution was taken off. The organic phase was separated, dried (Na_2SO_4) and concentrated in vacuum to get the crude compound. The crude compounds were purified by silica gel column chromatography (60 - 120 mesh silica gel) using methanol: chloroform as eluent.

The purity of the synthesized compounds was confirmed by TLC. Melting points of purified compounds were measured in capillary tubes and are uncorrected. $^1\text{H-NMR}$ spectra of purified compounds were recorded using Varian-Gemini spectrometer (400 MHz) and Mass spectra were recorded using Micromass - QUATTRO-II of WATER mass spectrometer.

Spectral Data of 2-Phenyl-1H-Benzimidazole(3a): M.p. 290 - 292°C ;

$^1\text{H-NMR}$ (300 MHz , DMSO-d_6) δ 12.7 (singlet, 1H , NH), 7.95 (multiplet, 2H), 7.25 - 7.35 (multiplet, 5H), 7.05 (multiplet, 2H); Mass (ES/MS): m/z 193 (M-H, 100%).

RESULTS AND DISCUSSION

Characterization of Catalyst

X-ray diffraction (XRD) patterns of the $\text{Ni}_{0.2}\text{Cu}_{0.2}\text{Zn}_{0.6}\text{Fe}_{2-x}\text{Cr}_x\text{O}_4$ Spinel Ferrite with $x = 0.0, 0.2$ and 0.4 are shown in Figure 1. The XRD patterns confirmed single phase of cubic spinel structure with the corresponding (220), (311), (222), (400), (422), (333) and (440) planes [JCPDS cards No's (00-52-0277) and (00-74-0444)] without additional peaks corresponding to any other phases.²⁸ Lattice Constant (a) and crystallite size were calculated for each sample using equation discussed elsewhere.²⁹ It is observed that Lattice constant (a) decreases from $8.418 \text{ \AA}$ to $8.406 \text{ \AA}$, while percentage porosity increases from 25.4% to 27.7% with increase in Cr^{3+} content x. Also crystallite size decreases from 30.3 nm to 19.1 nm with increase in Cr^{3+} content x.

The IR spectra as shown in Figure 2 shows that the higher frequency band (ν_1) is appeared in the range of 570 – 600 cm^{-1} due to the stretching vibration of tetrahedral sites whereas lower frequency band (ν_2) is appeared in the range of 375 – 480 cm^{-1} due to the stretching vibration of octahedral sites.

The TEM micrograph of $\text{Ni}_{0.2}\text{Cu}_{0.2}\text{Zn}_{0.6}\text{Fe}_{2-x}\text{Cr}_x\text{O}_4$ spinel ferrites are shown in Figure 3(a) $x = 0.0$. These images clearly indicate that the distribution of the particle is homogenous, consisting of well crystalline particles. Agglomeration was observed in all samples up to certain extent. Particle size for spinel ferrites calculated from TEM images using image J software and particle size distribution shown in Figure 3(b) $x = 0.2$. Average particle size found between 30 nm to 19 nm .

Figure 4 shows the hysteresis loops of Cr^{3+} doped Ni-Cu-Zn spinel ferrites, illustrates narrow loops, with a behavior characteristic feature of soft magnetic materials. It is observed that Saturation magnetization (M_s) decreases from 14.0 emu/g to 4.5 emu/g and Remanant magnetization (M_r) decreases from 32.8 emu/g to 15.9 emu/g with the substitution of Cr^{3+} ions.

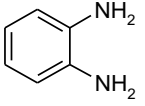
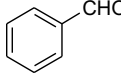
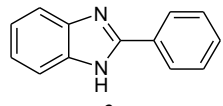
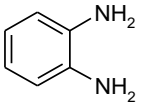
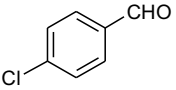
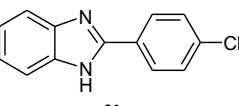
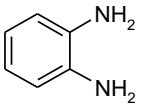
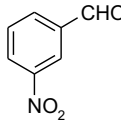
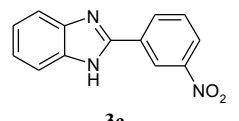
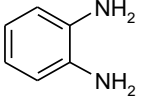
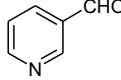
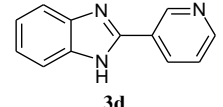
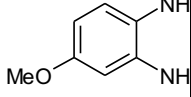
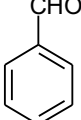
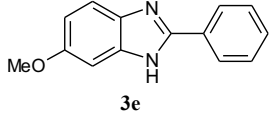
Synthesis of Benzimidazoles Using Cr^{3+} doped Ni-Cu-Zn Spinel Ferrite as Catalyst

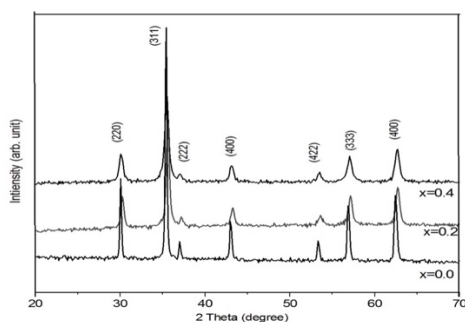
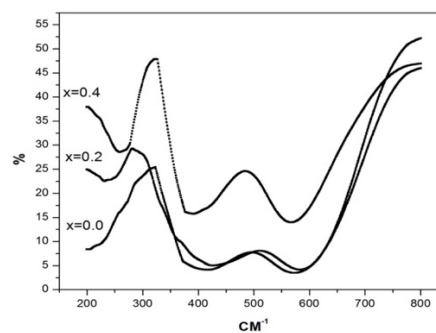
We decided to optimize the one pot two-component reaction of o-phenylene diamine and benzaldehyde as a model reaction for study. The reaction conditions were optimized on the basis of catalysts. The product of this model reaction is 2-Phenyl-1H-benzimidazole (**3a**) (Scheme 1). Firstly, to obtain the best catalyst for the synthesis of substituted benzimidazole (**3a**), the model reaction study was carried out in the presence of three prepared spinel ferrite catalytic systems $\text{Ni}_{0.2}\text{Cu}_{0.2}\text{Zn}_{0.6}\text{Fe}_{2-x}\text{Cr}_x\text{O}_4$ ($x = 0.0, 0.2$, and 0.4).

Table 1: Optimization of reaction conditions and catalyst load (mol %) of $\text{Ni}_{0.2}\text{Cu}_{0.2}\text{Zn}_{0.6}\text{Fe}_{2-x}\text{Cr}_x\text{O}_4$ ($x = 0.4$) nanoparticles for the synthesis of substituted benzimidazole (3a)

Entry	Catalyst (nanoparticles)	Catalyst loading (mol %)	Time (min)	Yield of 4a, ^a %
1	$\text{Ni}_{0.2}\text{Cu}_{0.2}\text{Zn}_{0.6}\text{Fe}_{2-x}\text{Cr}_x\text{O}_4$ ($x = 0.0$)	20	30	67
2	$\text{Ni}_{0.2}\text{Cu}_{0.2}\text{Zn}_{0.6}\text{Fe}_{2-x}\text{Cr}_x\text{O}_4$ ($x = 0.2$)	20	10	90
3	$\text{Ni}_{0.2}\text{Cu}_{0.2}\text{Zn}_{0.6}\text{Fe}_{2-x}\text{Cr}_x\text{O}_4$ ($x = 0.4$)	20	10	97
4	$\text{Ni}_{0.2}\text{Cu}_{0.2}\text{Zn}_{0.6}\text{Fe}_{2-x}\text{Cr}_x\text{O}_4$ ($x = 0.4$)	15	10	92
5	$\text{Ni}_{0.2}\text{Cu}_{0.2}\text{Zn}_{0.6}\text{Fe}_{2-x}\text{Cr}_x\text{O}_4$ ($x = 0.4$)	10	10	85
6	$\text{Ni}_{0.2}\text{Cu}_{0.2}\text{Zn}_{0.6}\text{Fe}_{2-x}\text{Cr}_x\text{O}_4$ ($x = 0.4$)	5	10	80
7	No catalyst	-	120	35

^a Hereinafter, isolated yield of pure product**Table 2: $\text{Ni}_{0.2}\text{Cu}_{0.2}\text{Zn}_{0.6}\text{Fe}_{2-x}\text{Cr}_x\text{O}_4$ ($x = 0.4$) catalyzed synthesis of benzimidazoles**

Diamine	Aldehyde	Product	Yield %	Melting Point (°C)	
				Observed	Reported ³¹⁻³⁶
		 3a	97	291	287-289
		 3b	90	294	293-295
		 3c	94	308	309-310
		 3d	93	248	248-251
		 3e	86	219	218-219

**Figure 1: XRD pattern of Cr^{3+} doped Ni-Cu-Zn Spinel Ferrites****Figure 2: IR spectra of Cr^{3+} doped Ni-Cu-Zn Spinel Ferrites**

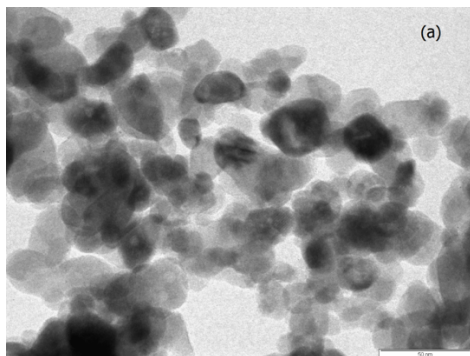


Figure 3(a): TEM micrograph of Cr^{3+} doped Ni-Cu-Zn Spinel Ferrite $x=0.0$,

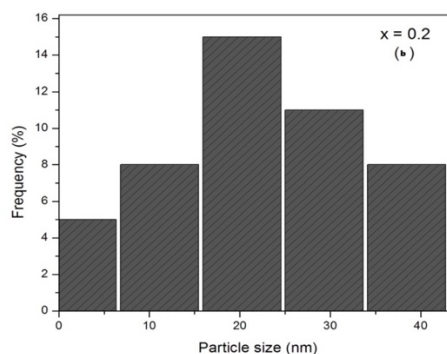


Figure 3(b): Particle size distribution $x = 0.2$

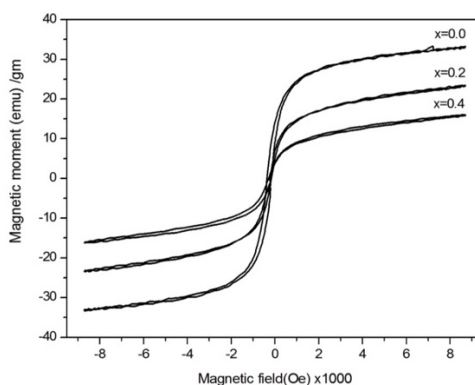


Figure 4: Hysteresis loops for Cr^{3+} doped Ni-Cu-Zn Spinel Ferrites

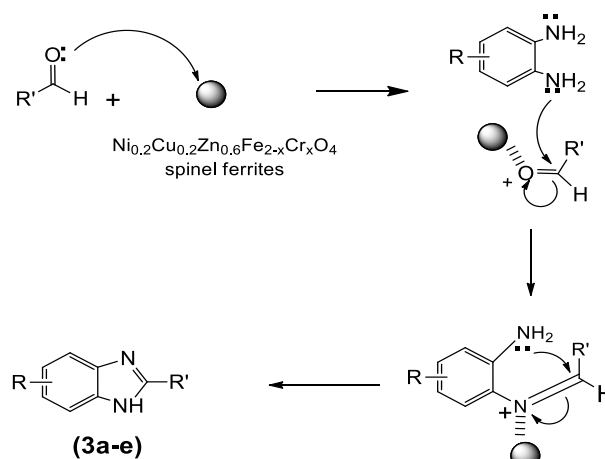
As observed from Table 1, among the three spinel ferrite catalysts, $\text{Ni}_{0.2}\text{Cu}_{0.2}\text{Zn}_{0.6}\text{Fe}_{2-x}\text{Cr}_x\text{O}_4$ ($x = 0.4$) were found to be the most effective catalyst (97% yield and shorter reaction time 10 min) for the synthesis of substituted benzimidazole (**3a**).

To find the optimized amount of the catalyst, the synthesis of substituted benzimidazole (**3a**) was carried out using different amounts of $\text{Ni}_{0.2}\text{Cu}_{0.2}\text{Zn}_{0.6}\text{Fe}_{2-x}\text{Cr}_x\text{O}_4$ ($x = 0.4$) as the spinel ferrite catalyst (20, 15, 10, 5 and 0 mol %). The results indicate that increase in amount of catalyst from 5 mol % to 20 mol % increases considerable yield of reaction (Table 1). This was due to increase in surface area and the number of catalytically active sites with increase in catalyst loading available for the same amount of the reactant.⁴ The best result was obtained by using 20 mol % of $\text{Ni}_{0.2}\text{Cu}_{0.2}\text{Zn}_{0.6}\text{Fe}_{2-x}\text{Cr}_x\text{O}_4$ ($x = 0.4$) spinel ferrite catalyst at room temperature. Thus, our results make the process under study more attractive and interesting from the view point of economy and simplicity.

The work-up of these reactions was very clean and required addition of dichloromethane and water to the reaction mixture, then the catalyst removed by fixing the catalyst magnetically at the bottom of the flask with a strong magnet, after which the solution was taken off, and the organic phase was separated, dried (Na_2SO_4) and concentrated in vacuum to get the crude compound. The crude compounds were purified by silica gel column chromatography (60-120 mesh silica gel) using methanol: chloroform as eluent.

A possible mechanism for the synthesis of substituted benzimidazole using $\text{Ni}_{0.2}\text{Cu}_{0.2}\text{Zn}_{0.6}\text{Fe}_{2-x}\text{Cr}_x\text{O}_4$ ($x = 0.4$) spinel ferrite catalyst is shown in Scheme 2, given on the basis of our experimental results together with some literature data.²⁴⁻²⁶

To evaluate the generality of this approach, various aromatic aldehydes were reacted with o-phenylenediamine (**1**) under optimized conditions to obtain substituted benzimidazoles (**3a-e**) (Scheme 1). As shown in Table 2 diamines and aldehydes without substituent groups gave the benzimidazoles (**3a**) in high yield.



Scheme 2. Possible mechanism for Synthesis of substituted benzimidazole (**3a-e**) using o-phenylene diamine (**1**), aromatic aldehydes (**2**), using 20 mol % $\text{Ni}_{0.2}\text{Cu}_{0.2}\text{Zn}_{0.6}\text{Fe}_{2-x}\text{Cr}_x\text{O}_4$ ($x = 0.4$) spinel ferrites as catalyst

Catalytic Reusability

Catalyst reusability is of major concern in heterogeneous catalysis. The recovery and reusability of the catalyst was investigated in this reaction for model reaction (Scheme 1, **3a**).

After completion of reaction (monitored by TLC), catalyst recycling was achieved by fixing the catalyst magnetically at the bottom of the flask with a strong magnet, after which the solution was taken off, the solid catalyst washed twice with acetone, dried at room temperature and the fresh substrate was introduced into the flask, allowing the reaction to proceed for the next run. The catalyst was consecutively reused five times with light loss of its catalytic activity⁹ (Cycle number and yield of **3a**: 1, 97 %; 2, 97 %; 3, 96 %; 4, 96 %; 5, 95 %), may be due to negligible leaching out of metals from ferrite system.³⁰ These catalysts are highly magnetic therefore; they could be easily and almost completely separated by an external magnet which is of a great advantage for a heterogeneous catalyst.

CONCLUSION

In conclusion, we have developed an efficient one-pot two-component method for the synthesis of benzimidazole derivatives from aromatic diamine, aromatic aldehydes in ethanol using Cr³⁺ doped Ni-Cu-Zn spinel ferrite. The XRD patterns confirmed single phase of cubic spinel structure of catalyst. Cr³⁺ doped Ni-Cu-Zn spinel ferrites were soft magnetic materials. More yield, small reaction duration, simplicity of separation (magnetically recoverable catalyst), and reusability of the catalyst were advantages of the proposed method with magnetic Cr³⁺ doped Ni-Cu-Zn spinel ferrite. The catalyst was not only efficient but also mild and easy to handle.

REFERENCES

- Weber L. Multi-component reactions and evolutionary chemistry. *Drug Discovery Today* 2002; 7: 143-7.
- Domling A, Wang W, Wang K. Chemistry and biology of multicomponent reactions. *Chemical reviews*; 2012. p. 3083-3135.
- Polshettiwar V, Varma RS. Green chemistry by nanocatalysis. *Green Chemistry* 2010; 12(5): 743-54.
- Goyal A, Chawla B, Bansal S, Tikoo KB, Kumar V, Singhal S. Evaluation of structural, magnetic and catalytic properties of Sn incorporated spinel nanoferrites. *Ceramics International* 2018; 44(5): 5601-12.
- Lee H, Jung JC, Kim H, Chung YM, Kim TJ, Lee SJ, Oh SH, Kim YS, Song IK. Preparation of ZnFe₂O₄ catalysts by a co-precipitation method using aqueous buffer solution and their catalytic activity for oxidative dehydrogenation of n-butene to 1, 3-butadiene. *Catalysis Letters* 2008; 122(3-4): 281-6.
- Nemati F, Afkham MG, Elhampour A. Nano-Fe₃O₄-encapsulated silica particles bearing sulfonic acid groups as a magnetically separable catalyst for green synthesis of 1, 1-diacetates. *Green Chemistry Letters and Reviews* 2014; 7(1): 79-84.
- Maleki A. One-pot multicomponent synthesis of diazepine derivatives using terminal alkynes in the presence of silica-supported superparamagnetic iron oxide nanoparticles. *Tetrahedron Letters* 2013; 54(16): 2055-2059.
- Kassae MZ, Masrouri H, Movahedi F. Sulfamic acid-functionalized magnetic Fe₃O₄ nanoparticles as an efficient and reusable catalyst for one-pot synthesis of α -amino nitriles in water. *Applied Catalysis A: General* 2011; 395(1-2): 28-33.
- Ghasemzadeh MA, Abdollahi-Basir MH. Fe₃O₄@ SiO₂-NH₂ nanocomposite as a robust and effective catalyst for the one-pot synthesis of poly substituted dihydropyridines. *Acta Chimica Slovenica* 2016; 63(3): 627-37.
- Zamani F, Izadi E. Synthesis and characterization of sulfonated-phenylacetic acid coated Fe₃O₄ nanoparticles as a novel acid magnetic catalyst for Biginelli reaction. *Catalysis Communications* 2013; 42: 104-8.
- Gawande MB, Bonifácio VD, Varma RS, Nogueira ID, Bundaleski N, Ghumman CA, Teodoro OM, Branco PS. Magnetically recyclable magnetite-ceria (Nanocat-Fe-Ce) nanocatalyst-applications in multi component reactions under benign conditions. *Green Chemistry* 2013; 15(5): 1226-31.
- Rathee PS, Dhankar R, Bhardwaj S, Gupta M, Kumar R. Synthesis and antimicrobial studies of substituted 2-phenylbenzimidazole derivatives. *Journal of Applied Pharmaceutical Science* 2011; 1(10): 14.
- Lukevics E, Arsenyan P, Shestakova I, Domracheva I, Nesterova A, Pudova O. Synthesis and anti tumour activity of trimethylsilylpropyl substituted benzimidazoles. *European journal of medicinal chemistry* 2001; 36(6): 507-15.
- Nakano H, Inoue T, Kawasaki N, Miyataka H, Matsumoto H, Taguchi T, Inagaki N, Nagai H, Satoh T. Synthesis and biological activities of novel antiallergic agents with 5-lipoxygenase inhibiting action. *Bioorganic and medicinal chemistry* 2000; 8(2): 373-80.
- Khan FR, Asnani AJ. Synthesis and antiulcer, anti-secretory activity of some new substituted 2-(pyrimidinylsulfinyl) benzimidazoles derivatives. *International Journal of Research in pharmaceutical and biomedical sciences* 2011; 2(2): 695-700.
- Bhriq B, Siddiqui N, Pathak D, Alam MS, Ali R, Azad B. Anticonvulsant evaluation of some newer benzimidazole derivatives: Design and synthesis. *Acta Pol. Pharm. Drug Res* 2012; 69(1): 53-62.
- Gravatt GL, Baguley BC, Wilson WR, Denny WA. DNA-directed alkylating agents. 6. Synthesis and antitumor activity of DNA minor groove-targeted aniline mustard analogs of pibenzimol (Hoechst 33258). *Journal of medicinal chemistry* 1994; 37(25): 4338-45.
- Ravindra KC, Vagdevi HM, Vaidya VP. Synthesis and antimicrobial activity of novel naphtho [2, 1-b] furo-5H-[3, 2-d][1, 3, 4] thiadiazolo [3, 2-a] pyrimidin-5-ones. *Arkivoc* 2008; 9(1): 10.
- Sevak R, Paul A, Goswami S, Santani D. Gastroprotective effect of β 3 adrenoceptor agonists ZD 7114 and CGP 12177A in rats. *Pharmacological research* 2002; 46(4): 351-6.
- Zhang ZH, Yin L, Wang YM. A General and Efficient Method for the Preparation of β -Enamino Ketones and Esters Catalyzed by Indium Tribromide. *Advanced Synthesis and Catalysis* 2006; 348(1-2): 184-90.
- Zhang ZH, Yin L, Wang YM. An expeditious synthesis of benzimidazole derivatives catalyzed by Lewis acids. *Catalysis Communications* 2007; 8(7): 1126-31.
- Rostamizadeh S, Amani AM, Aryan R, Ghaeni HR, Norouzi L. Very fast and efficient synthesis of some novel substituted 2-arylbenzimidazoles in water using ZrOCl₂ · nH₂O on montmorillonite K10 as catalyst. *Monatshesheft für Chemie-Chemical Monthly* 2009; 140(5): 547-52.
- Landge SM, Török B. Synthesis of condensed benzo [N, N]-heterocycles by microwave-assisted solid acid catalysis. *Catalysis Letters* 2008; 122(3-4): 338-43.
- Karami C, Abdollahifar M, Jahani F, Farrokhi A, Taher MA. The preparation and characterization of flower-like boehmite nanoparticles-SA: A new and reusable nanocatalyst for the synthesis of 2-aryl-1H-benzimidazoles. *Inorganic and Nano-Metal Chemistry* 2017; 47(4): 626-31.
- Mohammadi H, Karami C, Bahri M, Gilany MM, Taher MA. BiOCl/FeOCl/SiO₂ nano composite as an efficient novel catalyst toward the synthesis of 2-aryl-1H-

- benzimidazoles in mild aerobic condition. *Inorganic and Nano-Metal Chemistry* 2017; 47(9): 1334-41.
26. Ahmadian H, Veisi H, Karami C, Sedrpoushan A, Nouri M, Jamshidi F, Alavioon I. Cobalt manganese oxide nanoparticles as recyclable catalyst for efficient synthesis of 2-aryl-1-arylmethyl-1H-1, 3-benzimidazoles under solvent-free conditions. *Applied Organometallic Chemistry* 2015; 29(5): 266-9.
 27. Shinde BL, Dhale LA, Suryavanshi VS, Lohar KS. Preparation and characterization of Chromium-Doped Ni-Cu-Zn Nano Ferrites. *Acta Chimica Slovenica* 2017; 64(4): 931-7.
 28. Akhtar MN, Khan MA, Raza MR, Ahmad M, Murtaza G, Raza R, Shaukat SF, Asif MH, Saleem M, Nazir MS. Structural, morphological, dielectric and magnetic characterizations of Ni_{0.6}Cu_{0.2}Zn_{0.2}Fe₂O₄ (NCZF/MWCNTs/PVDF) nanocomposites for multilayer chip inductor (MLCI) applications. *Ceramics International* 2014; 40(10): 15821-9.
 29. BD Cullity, "Elements of X-ray diffraction", (Addison-Wesley Publication Company Inc., Reading, Massachusetts, U.S.A.); 1956. p. 1956.
 30. Kurian M, Nair DS. Manganese zinc ferrite nanoparticles as efficient catalysts for wet peroxide oxidation of organic aqueous wastes. *Journal of Chemical Sciences* 2015; 127(3): 537-46.
 31. Wang L, Sheng J, Tian H, Qian C. An efficient procedure for the synthesis of benzimidazole derivatives using Yb(OTf)₃ as catalyst under solvent-free conditions. *Synthetic communications* 2004; 34(23): 4265-72.
 32. Loupy A, Petit A, Hamelin J, Texier-Boullet F, Jacquault P, Mathe D. New solvent-free organic synthesis using focused microwaves. *Synthesis* 1998; 09: 1213-34.
 33. Lu J, Yang B, Bai Y. Microwave irradiation synthesis of 2-substituted benzimidazoles using PPA as a catalyst under solvent-free conditions. *Synthetic communications* 2002; 32(24): 3703-9.
 34. Reddy GV, Rama Rao VV, Narsaiah B, Shanthan Rao P. A simple and efficient method for the synthesis of novel trifluoromethyl benzimidazoles under microwave irradiation conditions. *Synthetic communications* 2002; 32(16): 2467-76.
 35. Penieries CG, Imelda AB, Jose Guadalupe CL, Guadalupe José EG, Alvarez TC. Synthesis of benzimidazoles in dry medium. *Synthetic Communications* 2000; 30(12): 2191-5.
 36. Perreux L, Loupy A. A tentative rationalization of microwave effects in organic synthesis according to the reaction medium, and mechanistic considerations. *Tetrahedron* 2001; 57(45): 9199-223.

Cite this article as:

B. L. Shinde *et al.* An efficient one-pot synthesis of benzimidazoles using magnetically recoverable catalyst chromium doped nickel copper zinc spinel ferrite. *Int. Res. J. Pharm.* 2019;10(8):50-55 <http://dx.doi.org/10.7897/2230-8407.1008245>

Source of support: Nil, Conflict of interest: None Declared

Disclaimer: IRJP is solely owned by Moksha Publishing House - A non-profit publishing house, dedicated to publish quality research, while every effort has been taken to verify the accuracy of the content published in our Journal. IRJP cannot accept any responsibility or liability for the site content and articles published. The views expressed in articles by our contributing authors are not necessarily those of IRJP editor or editorial board members.