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Research Article

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

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**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 $Cr^{3+}$ doped Ni-Cu-Zn spinel ferrite as catalyst in ethanol. The products obtained with high yields in shorter reaction time. The $Cr^{3+}$ 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 $Cr^{3+}$ content. The crystallite size decreased from 30.3 nm to 19.1 nm with increase in $Cr^{3+}$ content. $Cr^{3+}$ 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

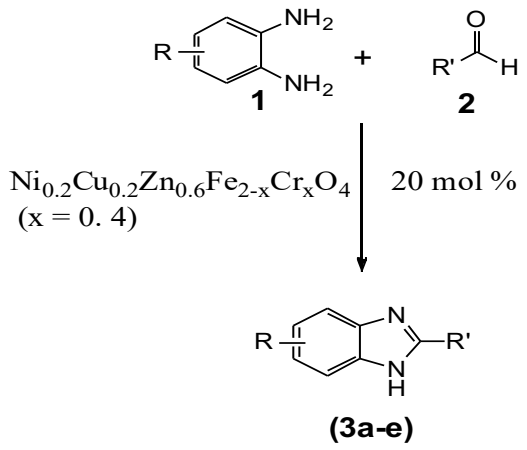
One-pot multi component reactions have more advantages over traditional reactions due to their rapidity, simplicity, atom-economy and shorter synthetic route for the synthesis of biologically active compounds.[1] Multicomponent reactions (MCRs) are the one pot methods in which three or more reactants reacts 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 catalysts[4] 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 diacetates[6], diazepine derivatives[7], α-amino acids[8], 1,4-dihydropyridines[9], etc. For these applications of metal oxides as heterogeneous catalysts, high surface area and accessible porosity are relevant properties. Spinels 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 anticancer[10], antihypertensive antiviral[11], antitumor[12], anti-allergic agents[13], antiprotozoal activity[14], diuretic activity[15], selective neuropeptide YY1 receptor antagonists[16] etc. By considering their various biological functions[17], they are used in clinical medicine[18], as anti-ulcer, antitumor and antiviral agent[19].

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 $InBr_3$[20], $ZrCl_4$, $SnCl_4$, $TiCl_4$, $HFCl_4$[21], $ZrOCl_2.9H_2O$[22] 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 $Cr^{3+}$ 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 $Cr^{3+}$ 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 % $Ni_{0.2}Cu_{0.2}Zn_{0.6}Fe_{2-x}Cr_xO_4$ (x = 0. 4) spinel ferrites as catalyst

## MATERIALS AND METHODS

### Synthesis of the Catalysts

The $Ni_{0.2}Cu_{0.2}Zn_{0.6}Fe_{2-x}Cr_xO_4$ (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 Cu-$K_\alpha$ radiation ($\lambda$=1.5405 Å) 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 $Ni_{0.2}Cu_{0.2}Zn_{0.6}Fe_{2-x}Cr_xO_4$ (x = 0. 4) (20 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. $^1$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;

$^1$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 $Ni_{0.2}Cu_{0.2}Zn_{0.6}Fe_{2-x}Cr_xO_4$ 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 Å to 8.406 Å, 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 ($\nu_1$) is appeared in the range of 570–600 $cm^{-1}$ due to the stretching vibration of tetrahedral sites whereas lower frequency band ($\nu_2$) is appeared in the range of 375-480 $cm^{-1}$ due to the stretching vibration of octahedral sites.

The TEM micrograph of $Ni_{0.2}Cu_{0.2}Zn_{0.6}Fe_{2-x}Cr_xO_4$ 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 (Ms) decreases from 14.0 emu/g to 4.5 emu/g and Remanant magnetization (Mr) 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

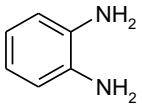
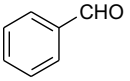
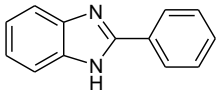
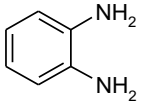
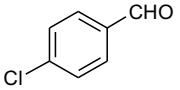
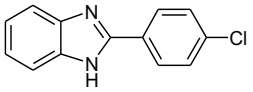
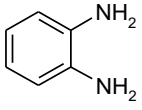
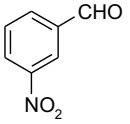
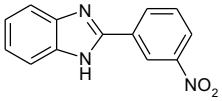
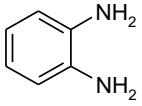
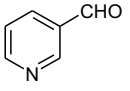
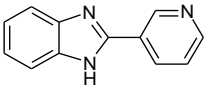
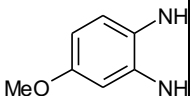
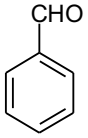
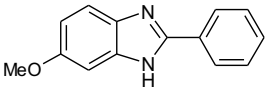
We decided to optimize the one pot two-component reaction of o-phenylene diamine and benzaldehyde as a model reaction for study. The reaction conditions were optimized on the basis of catalysts. The product of this model reaction is 2-Phenyl-1H-benzimidazole **(3a)** (Scheme 1). Firstly, to obtain the 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 $Ni_{0.2}Cu_{0.2}Zn_{0.6}Fe_{2-x}Cr_xO_4$ (x = 0.0, 0.2, and 0.4).

**Table 1: Optimization of reaction conditions and catalyst load (mol %) of $Ni_{0.2}Cu_{0.2}Zn_{0.6}Fe_{2-x}Cr_xO_4$ (x = 0. 4) nanoparticles for the synthesis of substituted benzimidazole (3a)**

| Entry | Catalyst (nanoparticles) | Catalyst loading (mol %) | Time (min) | Yield of 4a, [a] % |
|---|---|---|---|---|
| 1 | $Ni_{0.2}Cu_{0.2}Zn_{0.6}Fe_{2-x}Cr_xO_4$ (x = 0.0) | 20 | 30 | 67 |
| 2 | $Ni_{0.2}Cu_{0.2}Zn_{0.6}Fe_{2-x}Cr_xO_4$ (x = 0. 2) | 20 | 10 | 90 |
| 3 | $Ni_{0.2}Cu_{0.2}Zn_{0.6}Fe_{2-x}Cr_xO_4$ (x = 0. 4) | 20 | 10 | 97 |
| 4 | $Ni_{0.2}Cu_{0.2}Zn_{0.6}Fe_{2-x}Cr_xO_4$ (x = 0. 4) | 15 | 10 | 92 |
| 5 | $Ni_{0.2}Cu_{0.2}Zn_{0.6}Fe_{2-x}Cr_xO_4$ (x = 0. 4) | 10 | 10 | 85 |
| 6 | $Ni_{0.2}Cu_{0.2}Zn_{0.6}Fe_{2-x}Cr_xO_4$ (x = 0. 4) | 5 | 10 | 80 |
| 7 | No catalyst | - | 120 | 35 |

[a] Hereinafter, isolated yield of pure product

**Table 2: $Ni_{0.2}Cu_{0.2}Zn_{0.6}Fe_{2-x}Cr_xO_4$ (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 |

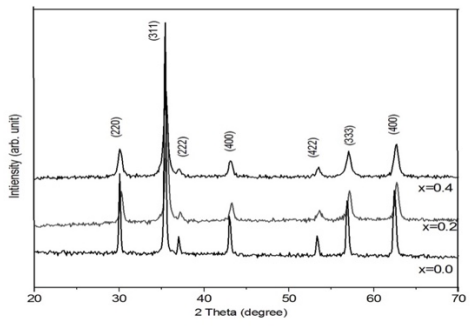



**Figure 1: XRD pattern of $Cr^{3+}$ doped Ni-Cu-Zn Spinel Ferrites**

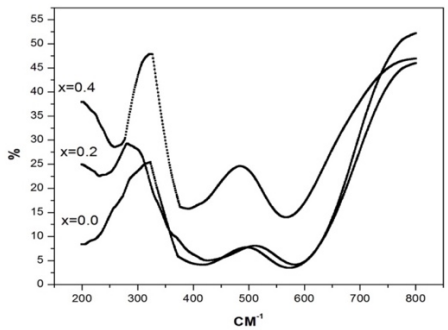



**Figure 2: IR spectra of $Cr^{3+}$ doped Ni-Cu-Zn Spinel Ferrites**

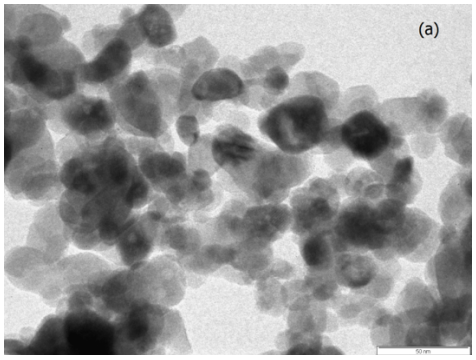



**Figure 3(a): TEM micrograph of $Cr^{3+}$ doped Ni-Cu-Zn Spinel Ferrite x=0.0,**

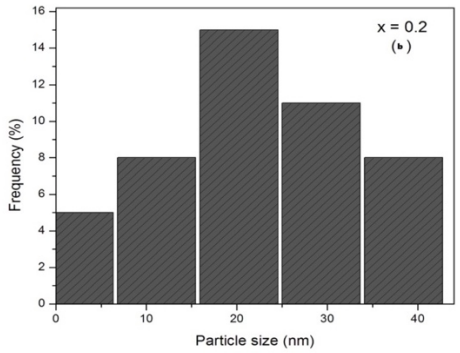



**Figure 3(b): Particle size distribution x = 0.2**

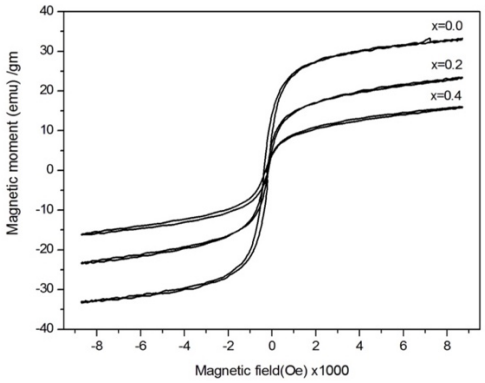



**Figure 4: Hysteresis loops for $Cr^{3+}$ doped Ni-Cu-Zn Spinel Ferrites**

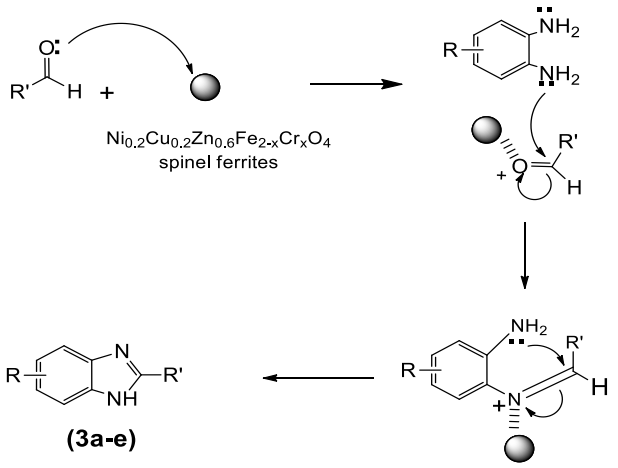
As observed from Table 1, among the three spinel ferrite catalysts, $Ni_{0.2}Cu_{0.2}Zn_{0.6}Fe_{2-x}Cr_xO_4$(x = 0.4) were found to be the most effective catalyst (97% yield and shorter reaction time 10 min) for the synthesis of substituted benzimidazole **(3a)**.

To find the optimized amount of the catalyst, the synthesis of substituted benzimidazole **(3a)** was carried out using different amounts of $Ni_{0.2}Cu_{0.2}Zn_{0.6}Fe_{2-x}Cr_xO_4$(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 $Ni_{0.2}Cu_{0.2}Zn_{0.6}Fe_{2-x}Cr_xO_4$ (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 $Ni_{0.2}Cu_{0.2}Zn_{0.6}Fe_{2-x}Cr_xO_4$(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 % $Ni_{0.2}Cu_{0.2}Zn_{0.6}Fe_{2-x}Cr_xO_4$ (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 widths light loss of its catalytic activity[9] (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 $Cr^{3+}$ doped Ni-Cu-Zn spinel ferrite. The XRD patterns confirmed single phase of cubic spinel structure of catalyst. $Cr^{3+}$ 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 $Cr^{3+}$ doped Ni-Cu-Zn spinel ferrite. The catalyst was not only efficient but also mild and easy to handle.

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