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A Simple One Pot Synthesis of 5-Acetyl-3,4-Dihydro-4-Phenylpyrimidin-2(1H)-One, by Using Magnetically Recoverable Heterogeneous Nickel Substituted Nano Ferro- Spinel Catalyst

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ABSTRACT

5-Acetyl-3,4-Dihydro-4-Phenylpyrimidin-2(1H)-One efficiently synthesized from the Benzaldehyde, Ethyl 3-oxobutanoate and urea in the presences of magnetically recoverable nanoferro-spinel catalyst. The present protocol offered remarkable improvement in yields (70%) of 5-Acetyl-3,4-Dihydro-4-Phenylpyrimidin-2(1H)-One by simple one pot synthesis.

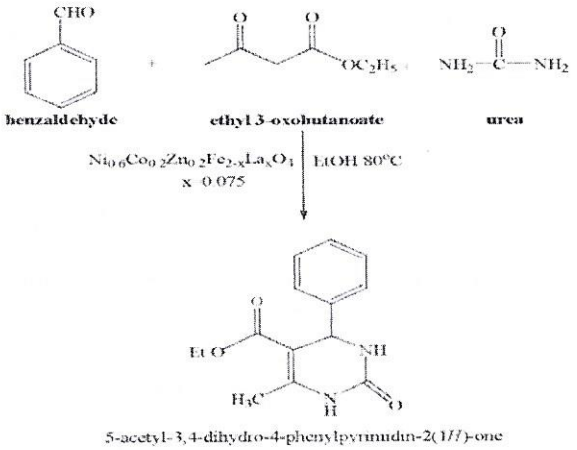
Keywords— 5-Acetyl-3,4-Dihydro-4-Phenylpyrimidin-2(1H)-One; Nano Ferro- Spinel; One pot.

I. INTRODUCTION

Nitrogen containing heterocyclic compounds are the derivatives of Pyrimidine. They are biologically significant constituents of leaving cells like DNA and RNA that contains amino pyrimidine [1]. They also shows number of biological activities including antiviral [3] antibacterial, anti-inflementry [4], analgesic [5], anti-HIV [6], anti-tubercular [7], anti-tumor and anti-material [8] etc. Now-a-days green chemistry has become a demanding research area leading to develop new methodologies of synthesis that replacing the use of toxic and volatile solvents and catalyst. Considering the advantages of this approach, herein we reported a synthesis of 5-Acetyl-3,4-Dihydro-4-Phenylpyrimidin-2(1H)-One. We have adopted a method reported by Pietro Biginelli where condensation of aldehyde, β-keto ester and thiourea or urea was carried out by using HCl as catalyst, which gives yield of 20-50%.[9] This method modified by several researchers by using various catalysts such as LiBr [10], $NbCl_5$ [11], $Sm(No_3)_36H_2O$ [12], $Ce(No_3)_36H_2O$ [13] etc. in a solvent such as CH_3CN,C_2H_5OH etc. However, these methods have many disadvantages such as i) they can only be partially recovered and ii) the use of 5-Acetyl-3,4-Dihydro-4-Phenylpyrimidin-2(1H) is harmful to environment. To overcome these disadvantages we report the synthesis of 5-Acetyl-3,4-Dihydro-4-Phenylpyrimidin-2(1H)-One via one pot three component condensation reacting using nanoferro-spinal compounds.

Ferro spinal compounds commonly known as spinal ferrites having a chemical formula in MFe_2O_4 (where, M= Mn, CO, Zn, Mg, Ni etc.). These spinel ferrite possess a cubic crystal symmetry where oxygen forms a face centered cubic (fcc) closed pacing structure. In the fcc symmetry, M^{2+} and Fe^{3+}metal ions can occupy either tetrahedral or octahedral interstitial sites.(14,15) These compounds having technological applications in various field such as, ferro fluids magnetic chips, drug delivery, high density information storage, catalysts etc. [16,17] In general, ferro spinal compounds can be synthesized by thermal process, sol-gel citrate method, solid state reaction, co- precipitation reaction, micro emulsion method etc. [18, 19]. The main advantage of micro emulsion method is, the particles are obtained with less degree of crystallization and hence produce small particle size.[20]

In this article we have reported the synthesis of 5-Acetyl-3,4-Dihydro-4-Phenylpyrimidin-2(1H)-One by one pot three component condensation reaction of Benzaldehyde, ethyl 3-oxobutanoate and urea using $Ni_{0.6}Zn_{0.2}Fe_{2-x}La_xO_4$ (x=0.075) as a catalyst (scheme1).



Scheme1: synthesis of 5-Acetyl-3,4-Dihydro-4-Phenylpyrimidin-2(1H)-One by one pot three component condensation reaction of Benzaldehyde, ethyl 3-oxobutanoate and urea using $Ni_{0.6}Zn_{0.2}Fe_{2-x}La_xO_4$ (X=0.075) catalyst

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II. METHODOLOGY

Normal micelle method was used to synthesis catalyst of a chemical formula $Ni_{0.6}Zn_{0.2}Fe_{2-x}La_xO_4$ (x=0.075). Corresponding analytical grade metal nitrates were dissolved in double distilled water in their stoichiometric proportion. An aqueous solution of most common surfactant for normal micelle procedure is Sodium do-decyl -sulfate (S.D.S. $CH_2(CH_2)_{10}$ CH_2-OSO_3Na)) is added at a temperature 40°C with continuous stirring for 1 h. A methyl amine (CH_3NH_2) 40% added up to pH=9 at 40°C that caused the solution to change the color to dark green then into dark brown slurry over a period of 2h.The dark brown slurry digested overnight and then filtered by simple filtration process. The obtained product was washed several times using double distilled water and acetone. The washed product was then dried in oven to get brownish colored fine powder.

The dried powder was characterized by Thermogravimetric Analysis (TGA)/Differential scanning calorimetry (DSC) to obtain the decomposition pattern and calcination temperature. The X-ray diffraction pattern of the sample was recorded in the range of 20 to 70°by using Cu–Kα radiation (λ=1.5405Å) to investigate the structural parameters like lattice constant, particle size etc.

III. RESULT AND DISCUSSION

Thermal Analysis:

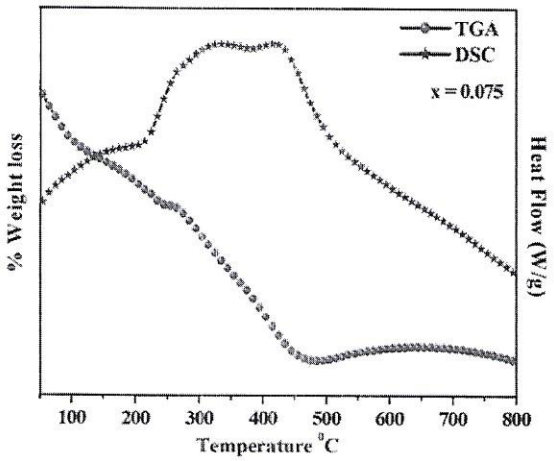

Figure 1: TGA/DSC plot of $Ni_{0.6}Zn_{0.2}Fe_{2-x}La_xO_4$ (X=0.075) ferrite precursor

Figure 1 showed the thermal decomposition pattern of $Ni_{0.6}Zn_{0.2}Fe_{2-x}La_xO_4$ (x=0.075) sample. The decomposition pattern was recorded at a heating rate of 10°C/min in nitrogen atmosphere in the temperature range of 25 to 800°C.This decomposition pattern showed three major mass losses. The first mass loss was observed around 200°C indicating dehydration of water molecule is an exothermic process. A total mass loss of around 6 % was recorded in the first step. The observed second mass loss around at 310°C is an exothermic process which is observed due to the 4.5% loss of organic matter. The third major weight loss of around 7%is observed in the temperature range of 350 to 460°C. In this study it is observed that the lanthanum doped ferrite thermally stable above 460°C temperature where heat treatment is necessary for crystallization of ferrite samples. A small exothermic pick is observed around temperature 430 °C in DSC curve which is related to the crystallization of samples. Similar types of results was also observed in literature. [21,22] Therefore, the sample was calcinated al 500°C for six hours using the TGA/DSC analysis.

XRD Analysis

X-ray diffraction pattern of $Ni_{0.6}Zn_{0.2}Fe_{2-x}La_xO_4$ (x=0.075) ferrite is shown in Fig. 2. The XRD peaks are well indexed to the (220), (311), (222), (446), (420), (422), (333) and (440) planes. The observation pointing that the prepared samples possess a single phase cubic spinel structure belonging to Fd3m space group. Some additional XRD peaks were also observed (100) and (002) which are belonging to the $LaFe_3O_4$ impurities (JCPDS card No. 75-0541).The lattice constant 'a' of the prepared spinel ferrite was8.486 Å which was calculated by an equation discussed elsewhere [23]. The particle size was calculated from full width at half maxima (FWHM) of the most intense (311) peak of XRD pattern and by using Debye-Scherrer method [24]. The estimated crystallite size by Debye-Scherrer is 9.96 nm. The Fig. of FWHM of the most intensive peak (311) is depicted in Fig. 1.

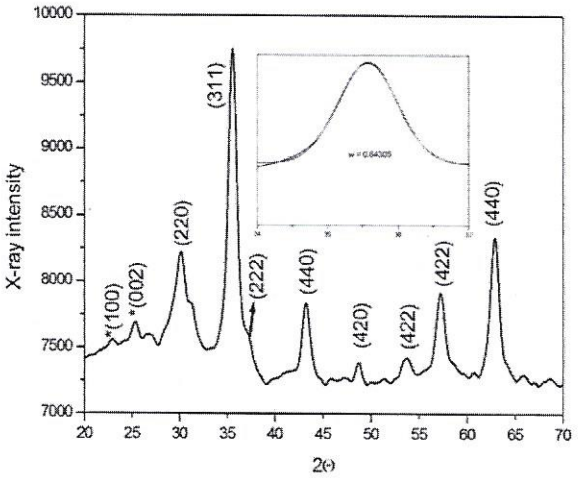

Figure 2: X-ray diffraction patterns of $Ni_{0.6}Zn_{0.2}Fe_{2-x}La_xO_4$ (X=0.075) ferrite

Synthesis of 5-Acetyl-3,4-Dihydro-4-Phenylpyrimidin-2(1H)-One:

A mixture of Benzaldehyde (10 m mole), ethyl 3-oxobutanoate (10 m mole),urea (15 m mole) and

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catalyst $Ni_{0.6}Zn_{0.2}Fe_{2-x}La_xO_4$ (x=0.075) (10 mole%), in ethanol solvent was heated at 80°C. The reaction was monitored by TLC periodically. Catalyst was removed after the completion of the reaction by fixing magnet at the bottom of the flask. The reaction mixture was poured on ice water, and the precipitated solid was collected by simple filtration process. The entire product was then washed with water and dried in oven. The crude product was re-crystallized from ethanol to give pure compound of white solid of melting point 202°C. Similar melting point of 200-202°C is reported in the literature [25-26]. The observed of the product was 97%.

IV. CONCLUSION

In this work, we have prepared lanthanum doped $Ni_{0.6}Zn_{0.2}Fe_{2-x}La_xO_4$ (x=0.075) ferrite. This ferrite was possess a spinal cubic structure with lattice constant 8.486 Å. The ferro-spinal sample was used as a heterogeneous magnetically recoverable catalyst. The crystallite size was obtained from the FWHM of most intensive peak (311) of XRD. The prepared ferrite sample is in the nanometer dimension having the crystallite size of 9.96 nm. 5-Acetyl-3,4-Dihydro-4-Phenylpyrimidin-2(1H)-One was synthesized by using $Ni_{0.6}Zn_{0.2}Fe_{2-x}La_xO_4$ (x=0.075) as a catalyst through one pot multi component system from three components. The feature of this method are preclusion of toxic solvents, less reaction time of only sixty minutes, recyclability of catalyst, high yield of 97% with maximum 87% purity of the obtained product.

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