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Microstructure, magnetic properties of Ho^{3+} substituted Ni-Cu-Zn spinel ferrites and application for one pot synthesis of dihydropyrimidinones

U.M. Mandle [a], R.M. Tigote [b], K.S. Lohar [c], B.L. Shinde [d]

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Highlights

- $Ni_{0.2}Cu_{0.2}Zn_{0.6}Fe_{2-x}Ho_xO_4$ Spinel ferrites synthesised by sol–gel auto combustion method.
- Precursor powders calcinated at 460°C.
- The lattice constant, X-ray density and hoping lengths increased with increase in Ho^{3+} substitution..
- The two strong IR absorption bands observed in the range 565–568 (v_1) and 409–439 (v_2) cm^{-1}.
- SEM and TEM images confirmed the porous nature with a fine-grained microstructure.ν
- 3,4-dihydropyrimidin-2(1H)-ones synthesized using magnetic Ho^{3+} doped Ni-Cu-Zn spinel ferrites catalyst.

Abstract

Nanoparticles of Ni-Cu-Zn spinel ferrites with Ho^{3+} substitution were synthesized through the sol–gel auto combustion route. It is illustrated from thermogravimetric analysisand differential thermal analysiscurve that the decomposition of precursors takes place in the temperature range 350–460 ^{o}C. The Energy dispersive spectroscopy confirmed the mixing of the Fe, Ni, Cu, Zn, Ho and oxygen elements in stoichiometry proportion in pure and substituted spinel ferrites with desired stoichiometry. X-ray diffraction pattern confirmed the formation of cubic spinel structure without any impurity phases. Lattice constant, X-ray density increases while average crystallite size decreases with increased Ho^{3+}substitution in Ni-Cu-Zn ferrites. The two strong IR absorption bands observed in the range 565–568 (v_1) and 409–438 (v_2) cm^{-1}. The obtained crystallite size lies between of 18 – 26nm, confirmed from transmission electron microscopy. The microstructures of the calcinated spinel ferrites were evaluated by SEM and TEM. It is observed that the increase in concentration of Ho^{3+} ions saturation magnetization decreases. Prepared Ho^{3+} substituted Ni-Cu-Zn spinel ferrites were used as catalyst to synthesize the 3,4-dihydropyrimidin-2(1H)-ones.[copyright information to be updated in production process]

Introduction

Nanoscience and Nanotechnology is the technology associated with materials in nanometer range

and appliances based on them [1]. Ferrites are chemical composites with iron (III) oxide Fe_2O_3 as their major constitutes [2]. The electrical conduction and magnetic interactions of the ferrites are significantly altered by substitution of trivalent or tetravalent cations and also affected by the site occupancy of cations between the tetrahedral A and octahedral B sites of spinel structure [3]. By tailoring the stoichiometry of the ferrite system structural, electrical and magnetic properties can be tuned [4]. The spinel ferrites have versatile applications such as magnetic drug delivery [5], information storage [6], super capacitor [7], magnetic refrigeration [8], gas sensors [9], and catalyst [10] etc.

Ni-Cu-Zn ferrites have more attention of researchers due to their outstanding characteristics such as low magnetic losses, high permeability and high resistivity, which are suitable for microwave applications [11]. The doping of trivalent rare-earth cations in ferrites are becoming the important components for sophisticated applications [12]. The substitution of little quantity of rare-earth cations improves structural, electrical and magnetic properties of ferrites [13].

One-pot multicomponent reactions have more advantages over traditional reactions due to their rapidity, simplicity, atom-economy and shorter synthetic route [14] for the synthesis of bio-active molecules [15]. 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. Iron oxide nanoparticles are used as heterogeneous catalyst as it is separated without filtration simply applying external magnetic field [16]. The catalytic activity of spinel ferrites for these reactions arises due to the ease iron can switch its oxidation state between 2+and 3+and hence spinel structure ferrites have stability under extremely reducing conditions. The Fe^{3+} reduced to Fe^{2+} without varying lattice arrangements; therefore original sate is obtained on reoxidation [17].

Now days, magnetic spinel ferrites used as efficient catalyst in different chemical transformations such as synthesis of diazepine derivatives [18], α-amino nitriles [19], 1,1-diacetates from aldehydes [20], etc. Dihydropyrimidinones have synthesized by Pietro Biginelli in 1893, by a one-pot three-component condensation of benzaldehyde, ethyl acetoacetate and urea under robustly acidic environment [21]. Also Dihydropyrimidinones synthesized by microwave irradiation [22], ionic liquids [23] and using different types of catalysts such as lanthanide triflate [24], $Sr(OTf)_2$ [25], nano magnetic-supported sulfonic acid [26], Fe(III) tosylate [27], $FeCl_3$-supported nanopore silica [28], etc.

The present report deals with the modification in the structural and magnetic properties of Ho^{3+}substituted Ni-Cu-Zn spinel ferrites with composition $Ni_{0.2}Cu_{0.2}Zn_{0.6}Fe_{2-x}Ho_xO_4$ (x=0.00, 0.02, 0.04, 0.06 and 0.08). The targeted samples were synthesized by sol–gel auto-combustion method. The prepared samples at nano scale regime were used as a catalyst for one pot three component synthesis of Dihydropyrimidinones.

Section snippets

Preparation of $Ni_{0.2}Cu_{0.2}Zn_{0.6}Fe_{2-x}Ho_xO_4$ ferrites:

The nano-crystalline powders of $Ni_{0.2}Cu_{0.2}Zn_{0.6}Fe_{2-x}Ho_xO_4$ (x=0.00, 0.02, 0.04, 0.06, and 0.08) prepared using sol–gel auto-combustion method [4]. The stoichiometric quantities of analytical grade reagents of corresponding metal nitrates were dissolved in deionized water. The aqueous citric acid solution added in the metal nitrates to citric acid ratio of 1:3 and then NH_3 solution added to adjust pH ≈ 7 of solution. The resulting solution was heated on hot plate (90°C) with continuous…

Thermal analysis

Fig. 1a-e represents TGA/DTA curves of $Ni_{0.2}Cu_{0.2}Zn_{0.6}Fe_{2-x}Ho_xO_4$ (x=0.0 to 0.08). All TGA curves shows two weight loss steps associated with endothermic and exothermic DTA peaks. The first weight loss step in the temperature 30–100°C range is corresponding to endothermic peak around 80°C, is due to evaporation of residual water from precursor. The second weight loss step in the 350–460°C temperature region was corresponding to the exothermic peak around 400°C, due to the decomposition of…

Conclusions

Ho^{3+} substituted Ni-Cu-Zn ferrites with composition $Ni_{0.2}Cu_{0.2}Zn_{0.6}Fe_{2-x}Ho_xO_4$ (x=0.00 to 0.08 in steps of 0.02) were successfully synthesised by sol–gel auto combustion method. Precursor powders calcinated at 460°C. The observed elemental analysis from EDAX was in good agreement with the theoretical stoichiometry. The lattice constant, X-ray density and hoping lengths increased with increase in Ho^{3+}. The D_{XRD} was decreased from 26.2 to 19.9nm with the substitution of Ho^{3+}. The low frequency …

CRediT authorship contribution statement

U.M. Mandle: Investigation, Writing - original draft. **R.M. Tigote:** Formal analysis, Writing - review & editing. **K.S. Lohar:** Conceptualization, Supervision, Writing - review & editing. **B.L. Shinde:** Investigation, Methodology, Formal analysis, Writing - original draft.…

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.…

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