CODEN [USA]: IAJPBB ISSN: 2349-7750

INDO AMERICAN JOURNAL OF
PHARMACEUTICAL SCIENCES

http://doi.org/10.5281/zenodo.1135273

Available online at: http://www.iajps.com *Research Article*

MICROSTRUCTURAL AND ANTIFUNGAL PROPERTIES OF SILVER SUBSTITUTED COPPER FERRITE NANOPOWDER SYNTHESIZED BY SOL-GEL METHOD

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Abstract:

Single phase spinel ferrite samples of Ag doped $CuFe_2O_4$ were synthesized by using sol-gel method and annealed at 350^0C for 3 hrs. The present work reports study on antimicrobial activity of pure and Ag doped $CuFe_2O_4$ nanocomposites. The effect of silver doping on the structural and antimicrobial properties of copper ferrites has been examined. The structure of nanosize Ag doped $CuFe_2O_4$ was characterized by Fourier transform infra-red spectroscopy (FTIR) and Transmission electron microscope (TEM). The X-ray diffraction measurements clearly showed the formation of single phase spinel ferrite structure in all the prepared ferrite compositions. The lattice parameters are found to increase with increasing doping concentration of the silver content. The synthesized nanoparticles have been tested against the pathogenic culture showed a very good zone of inhibition. It indicates the biomedical capability of Ag doped $CuFe_2O_4$.

Keywords: *Nanoparticles, Fungal pathogens, XRD, FTIR, TEM.*

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Please cite this article in press as P. S. Thakare et al., ***Microstructural and Antifungal Properties of Silver Substituted Copper Ferrite Nanopowder Synthesized by Sol-Gel Method****, Indo Am. J. P. Sci, 2018; 05(01).*

INTRODUCTION:

In recent year there is a rapidly growing interest in the systematic study of nanoparticles due to their unique physical and chemical properties which differ significantly from their conventional counterparts [1]. Since 19th century nanomaterials have attracted an incredible attention on the relationship between chemical composition and magnetic properties of various ferrites [2].Recent studies have demonstrated antimicrobial activity of various nanoparticles, including silver [3], Copper [4], titanium oxide and zinc oxide [5]. Among the spinel ferrite family is one of the most interesting inverse spinel copper ferrite because of its magnetic, electrical and properties recommending it as a suitable candidate for biomedical applications [6]. This copper ferrite is an inverse spinel ferrite, crystallizing either in a tetragonal or cubic structure [7]. $CuFe_2O_4$ is known to exist in tetragonal and cubic structures.

Recent studies revealed the antimicrobial activity of the copper ferrite nanoparticles on pathogenic and multidrug resistant fungal strains. Prior to the extensive use of chemotherapeutics in modern health care system, inorganic antimicrobials such as silver and copper were used since ancient times to treat microbial infections [8] Moreover, magnetic nanoparticles are one of the most promising materials since they possess exceptional antibacterial properties because these materials exhibit large surface area to volume ratio, and high reactivity in comparison to bulk form which is of enthusiasm to researchers due to the developing microbial resistance against antibiotics, and the improvement of resistant strains. Over few decades, researchers infer that by substitution of various non-magnetic ions and transition metal ions in spinel ferrites leads to improvement of their crystalline structure, magnetic properties and antibacterial activity [9]. On the other hand, copper ferrite acquires improved properties when combined with noble metals, like Ag or Au. Taking into account the inherent antimicrobial properties of Ag, it is expected that the addition of Ag to $CuFe_2O_4$ will enhance its antimicrobial activity [10–12].

Fungi can thus provide answers to very different scientific questions. And with the knowledge that thousands, if not millions of species are still out there waiting to be discovered and analysed, this field of research is surely good for a few more revealing insights about our world. Many NPs have antimicrobial properties and used to control drug-resistant microbial populations [13]. Various inorganic metal oxide NPs viz., ZnO, MgO, TiO_2 and SiO_2 exhibit considerable antimicrobial activities and used in therapeutics, diagnostics and nanomedicine-based antimicrobial agents.[14,15] Inorganic NPs show greater effectiveness on resistant strains of microbial pathogens, less toxicity, heat resistance and provide mineral elements essential to human cells [16].

Nanomaterials have tremendous potential in both the medical and veterinary fields. Several nanostructures comprising metallic particles have been developed to counteract microbial pathogens. The effectiveness of nanoparticles (NPs) depends on the interaction between the microorganism and the NPs. Advances in nanotechnology have led to the synthesis of nano-sized organic and inorganic molecules with potential applications in industry, food packaging, textiles, medicine, and therapeutics. The development of novel nanoscale Antimicrobial agent's nanocomposites can be used as an alternative strategy to overcome antimicrobial resistance [17]. The replacement of conventional antimicrobials by new technology to counteract antimicrobial resistance is ongoing. The advent of nanotechnology, the biggest engineering innovation of recent times, has modernized medicine. The demand for nanotechnology-derived products is constantly increasing. Nanotechnology, which is the innovative technology in the present scenario, can have a profound influence on improving human health.

Overall, the nanomaterials based on the metal oxide ions, exhibit broad spectrum biocidal activity towards different bacteria, fungi and viruses and have a distinct advantage over conventional chemical antimicrobial agents, Ultimately such positive environmental and toxicological studies will be imperative to ensure the nanomaterials design process yields both effective and safe technology.

Several methods for synthesizing nanosized spinel ferrite nanoparticles and to find its influence of doping in magnetic and antibacterial properties, such as solid-state reaction [18],co-precipitation [19], combustion method [20] and sol-gel method [21]. Among these methods, we have chosen sol-gel method, due to the fact that, with this method, significantly large amount of products can be produced within a very short time. As pointed out above, very few works have been found in literature on the Ag doped copper ferrite system and its antifungal activity. Herein, we report the influence of doping on antifungal properties of copper-silver ferrite nanoparticles prepared by sol-gel method.

The antifungal effect of NPs has received only marginal attention and just a few studies on this topic have been published. By studying bioassay of these materials the zone of inhibition suggested that the compounds had antifungal activity at room temperature and these materials can be used in biomedical applications.

MATERIALS AND METHODS:

Material:

Copper nitrate tetra hydrate (Cu $(NO_3)_2.4H_2O$), Iron nitrate non hydrate (Fe $(NO_3)_2.9H_2O$) and citric acid and silver nitrate was obtained of analytical grade. All experiment was done by using ethyl alcohol. Undoped $CuFe_2O_4$ and Ag doped $CuFe_2O_4$ were synthesized by sol-gel method.

Synthesis of $CuFe_2O_4$nanoparticle:

Silver doped $CuFe_2O_4$ nanoparticles were synthesis by sol-gel method. All chemicals add in beaker and continuous stirring on magnetic stirrer for 2 hours then form gel and calcinite at different temperature 350^0C, 550^0C and 650^0C calcinied.

Assay to Evaluate Antifungal Activity

Fungal Culture

Test Organism: Candida albicans, Aspergillus, Aspergillus flavus

Medium: Potato dextrose broth was used as a medium for well diffusion assay.

Preparation of Fungal Suspension: With the help of sterile wire loop, the test was inoculated into a test tube containing Potato dextrose broth. The concentration of the inoculum was adjusted to 0.5 McFarland's standards which is equivalent to 10^8 CFU/ml. This was used in assay.

Procedure:

Disc diffusion assay antifungal activities of the synthesised NPs were evaluated by the standard disc diffusion method described by Bauer et al. [22] and modified according to clinical and laboratory standards institute guidelines.

1. As per the composition, 250 ml of Potato dextrose agar was prepared using sterile distilled water and it was sterilized at 121^0C at 15 lb pressure for 15 min in an autoclave.
2. The medium was cooled at room temperature and poured in sterile petri plates and were allowed to solidify.
3. Fungal culture inoculum adjusted at 0.5 McFarland's standards was swabbed over the medium using sterile cotton swab.
4. Two sterile disc were placed on each petrify plate with the help of sterile forceps and two antibiotic disc, one as positive control and other for combination. The 20 ul of sample, sample control (methanol) were poured on each sterile disc as well as on one of the antibiotic disc (Amphotericin-B) and incubated at 27^0C in incubator for 48 hrs. This experiment carried out in triplicate set for avoiding any contamination.
5. Zone of inhibition were observed and measured with the ruler scale.

RESULT AND DISSCUTION:

XRD study

Fig. 1 shows the room-temperature XRD patterns of copper ferrite samples prepared with various Ag substitutions (1%, 2% and 3%), annealed at 350 ^oC for 3 hrs. The size distribution of nano Ag doped $CuFe_2O_4$ is presented in Figure 1. The size of the nanoparticles was around 21 nm. The diffraction peaks agree with the international standard diffraction data card JCPDS number 36-1451and provide a clear evidence of Ag doped $CuFe_2O_4$. The distribution was narrow. The resulting size distribution was a good match for TEM (Figure 3) and the XRD results. From Fig. 1, Ag doped $CuFe_2O_4$ nanoparticles have an average particle size of about 21 nm. Ag doped $CuFe_2O_4$ nanoparticles could be dispersed well. Very little aggregation could be found. All peaks were consistent with the peaks of standard Ag doped $CuFe_2O_4$ with high crystalline. The line broadening of the XRD patterns showed clear evidence for the nanometer range and the peaks, indicative of the ultra fine nature of the synthesized power. The diffraction planes are identified as the (111), (220), (311), (400), (422), (511), (440) respectively planes of cubic spinel structure. The average size of the nanoparticles can be estimated using the Debye–Scherrer equation.

$$D = \frac{0.89\lambda}{\beta\cos\Theta}$$

It is near about 21 nm. No peaks from any other phases of Ag doped $CuFe_2O_4$ were observed.

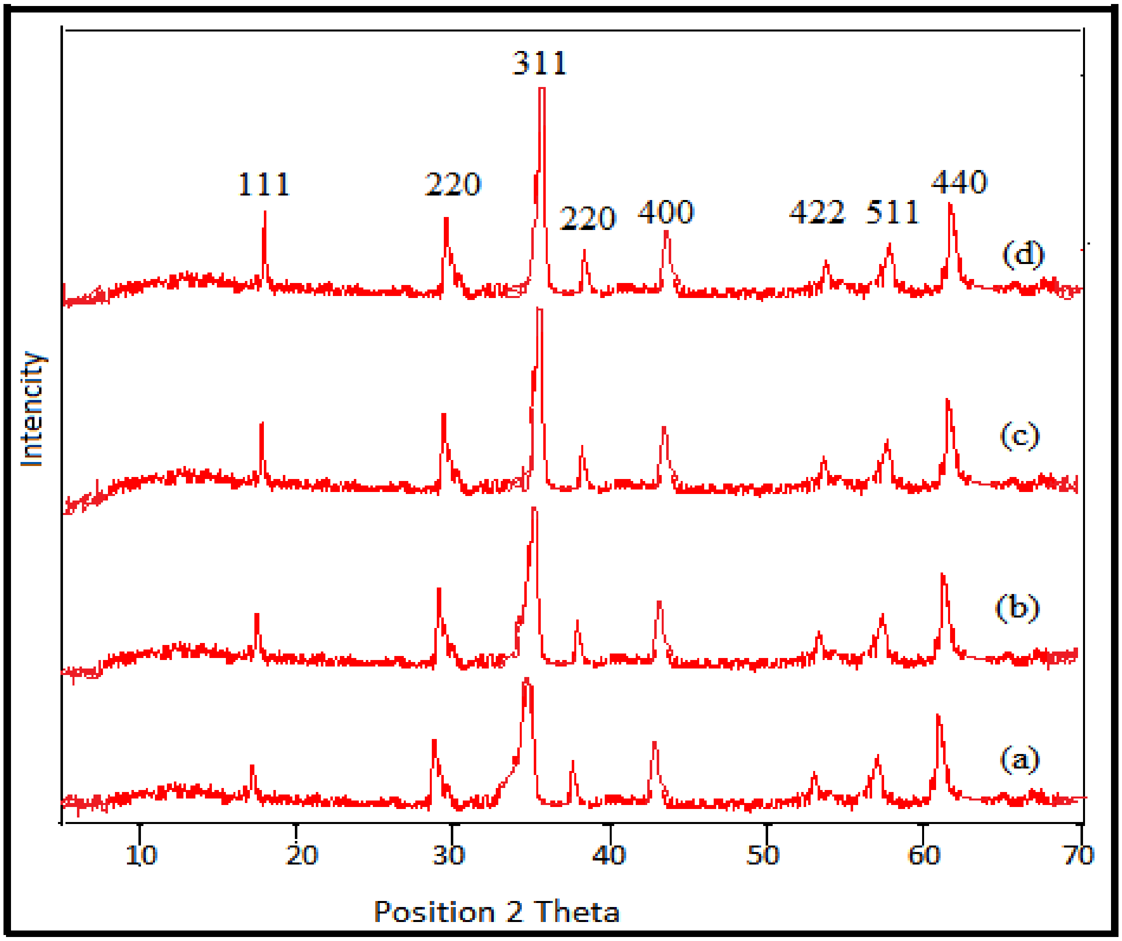


Fig. 1: X-ray diffraction patterns for (a) $CuFe_2O_4$ (b) 1% Ag doped $CuFe_2O_4$ (c) 2% Ag doped $CuFe_2O_4$ (d) 3% Ag doped $CuFe_2O_4$powders annealed at 350 °C

FTIR Spectra

The FTIR spectroscopy is used to identify their functional groups present in the ferrite composition. Fig. 3 shows the FTIR spectra in the range of 4000-400 cm^{-1} for the Ag doped $CuFe_2O_4$ samples sintered at 350 °C. The absorption band around 3587.60 cm^{-1} indicates the presence of O-H group. The IR spectra show the two strong absorption bands in the range of 400-600cm^{-1}typical to spinel structure characteristics, confirms that the samples prepared are spinel in structure. Normally, the higher frequency band is observed in the range of 600-500 cm^{-1}. And lower frequency band is observed in the range of 500-400 cm^{-1}. These two bands are common feature for all ferrites [23]. In figure 2 the spectra exhibit two absorption bands at 543 and 443 cm^{-1}. These spectra represent characteristic features of ferrospinels of tetrahedral and octahedral M-O stretching frequency. The characteristic band at 1373 $cm^{-1}$is ascribed to the symmetric vibration of NO_3- group. The absorption peaks corresponding to 997 and 710 cm^{-1} are related to the presence of Fe ions in ferrites. Generally, oxide vibrations occur below 1000/cm.

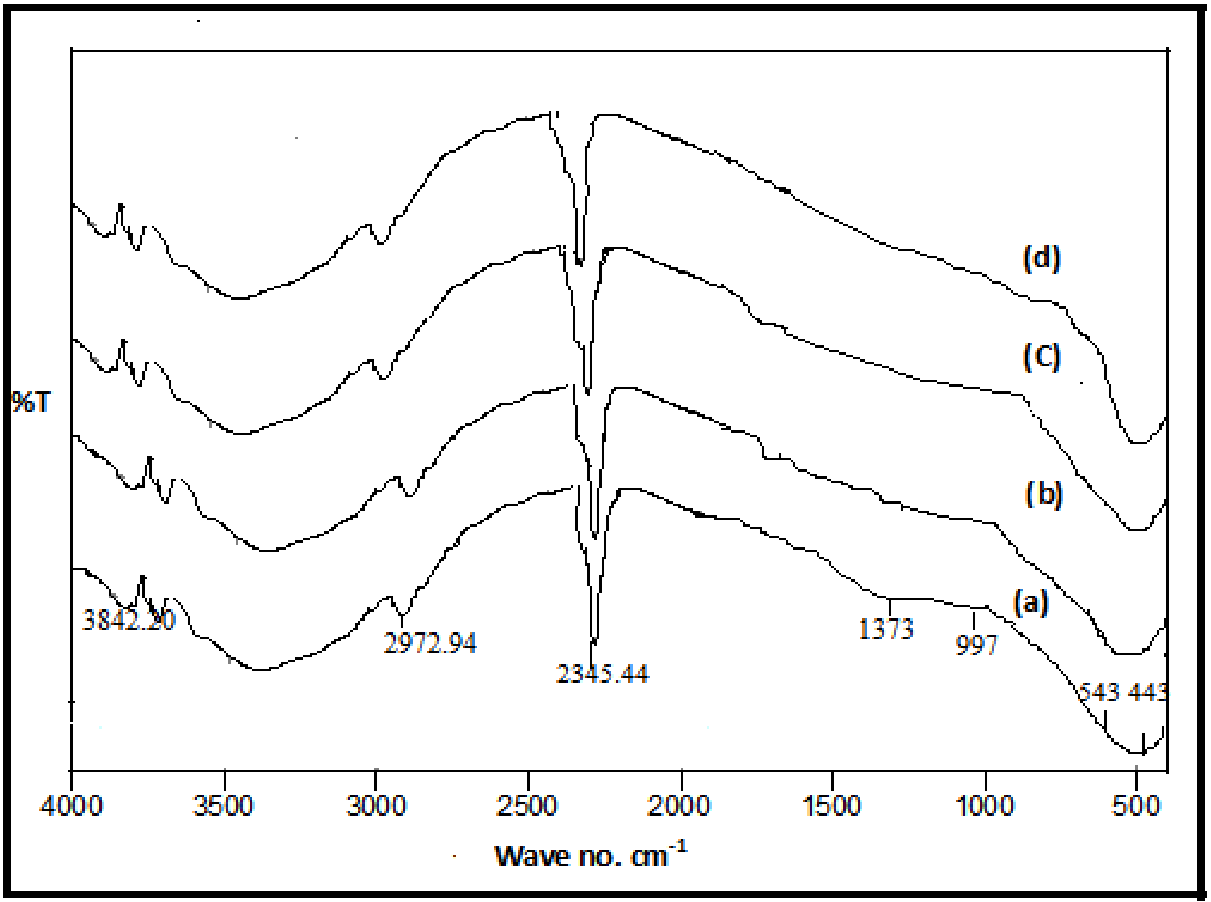
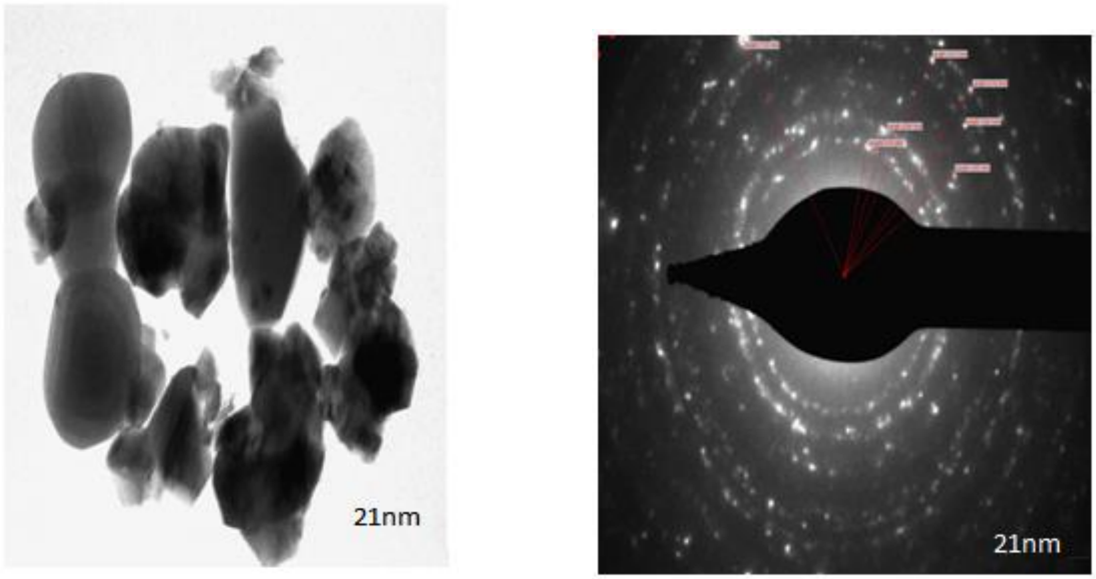


Fig.2: FTIR Spectra for (a) $CuFe_2O_4$ (b) 1% Ag doped $CuFe_2O_4$ (c) 2% Ag doped $CuFe_2O_4$ (d) 3% Ag doped $CuFe_2O_4$powders annealed at 350 ºC.

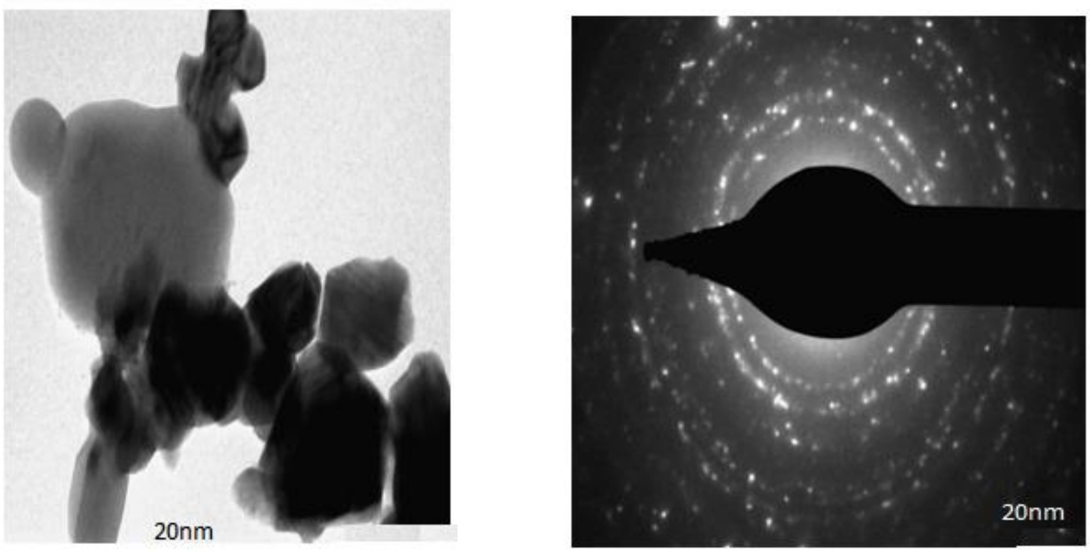
TEM Study

The detailed morphology and crystalline structure of the undoped $CuFe_2O_4$ and Ag doped $CuFe_2O_4$ calcined at 350^{0}C for 3 hrs were further investigated by TEM, and the TEM bright- field imeges with corresponding selected area electron diffraction. Transmission electron microscopy images of the undoped $CuFe_2O_4$ and Ag doped $CuFe_2O_4$samples are depicted in figure-3 (a) and (b) respectively. From the TEM images, the pure and Ag -doped $CuFe_2O_4$ nanoparticles are nearly in equal size of 21 nm. The evaluated particles size from the XRD pattern, which were in good agreement with the TEM results. In figure the corresponding selected area electron diffraction (SAED) pattern indicates the crystalline and preferential orientation of the Ag doped $CuFe_2O_4$ samples pattern without any additional diffraction spots of Ag and Cu clusters, and which is in good agreement with the

Quartzite structure of the XRD results and the standard data card JCPDS: 36-1451.



(a)



(b)

Fig.3: TEM imeges with corresponding SAED patterns of the (a) $CuFe_2O_4$ (b) Ag doped $CuFe_2O_4$

ANTIMICROBIAL ACTIVITY

Disc diffusion assay

Disc diffusion assay Antifungal activities of the synthesised NPs were evaluated by the standard disc diffusion method and modified according to clinical and laboratory standards institute guidelines. The antimicrobial activities of pure and doped CuFe2O4 NPs are carried out against three fungal pathogens i.e. Candida Albicans, Aspergillus flaves and Aspergillus niger. The zone of inhibition is given in Tables 1 and figure 4 (a,b,c,d), Fig. 5 (a,b,c,d) and Fig.6 (a,b,c,d). It is clear from the tables and graph that $CuFe_2O_4$ NPs silver doped with 1%, 2% and 3% exhibit higher antifungal activities as compared to pure $CuFe_2O_4$. The experiment results indicate that doping in the nanomaterials plays a significant role in the antimicrobial activity. Thus, in this report, silver doped $CuFe_2O_4$ NPs have shown the best antifungal behaviour compared to $CuFe_2O_4$ NPs. Our results are well supported by the earlier studies reported that transition metal enhances the antifungal activity. The obtained results indicated that active oxygen species generated from transition metal oxides of pure and doped ferrites have more potential to penetrate the cell wall and decrease the cell wall division. Furthermore, the antifungal result shows better inhibition for doped samples than pure samples. The results reported here are better than the previous reports [24]. With an increase in concentration of doping antimicrobial activity increased. Our data are in accordance with the previous studies, dealing with the antimicrobial effects of NPs [25]. If the concentration of doped metals in nano- $CuFe_2O_4$ increases in culture medium, interaction between oxygen and dehydrogenise increases too which enhances antimicrobial activity[26].

Candida Albicans

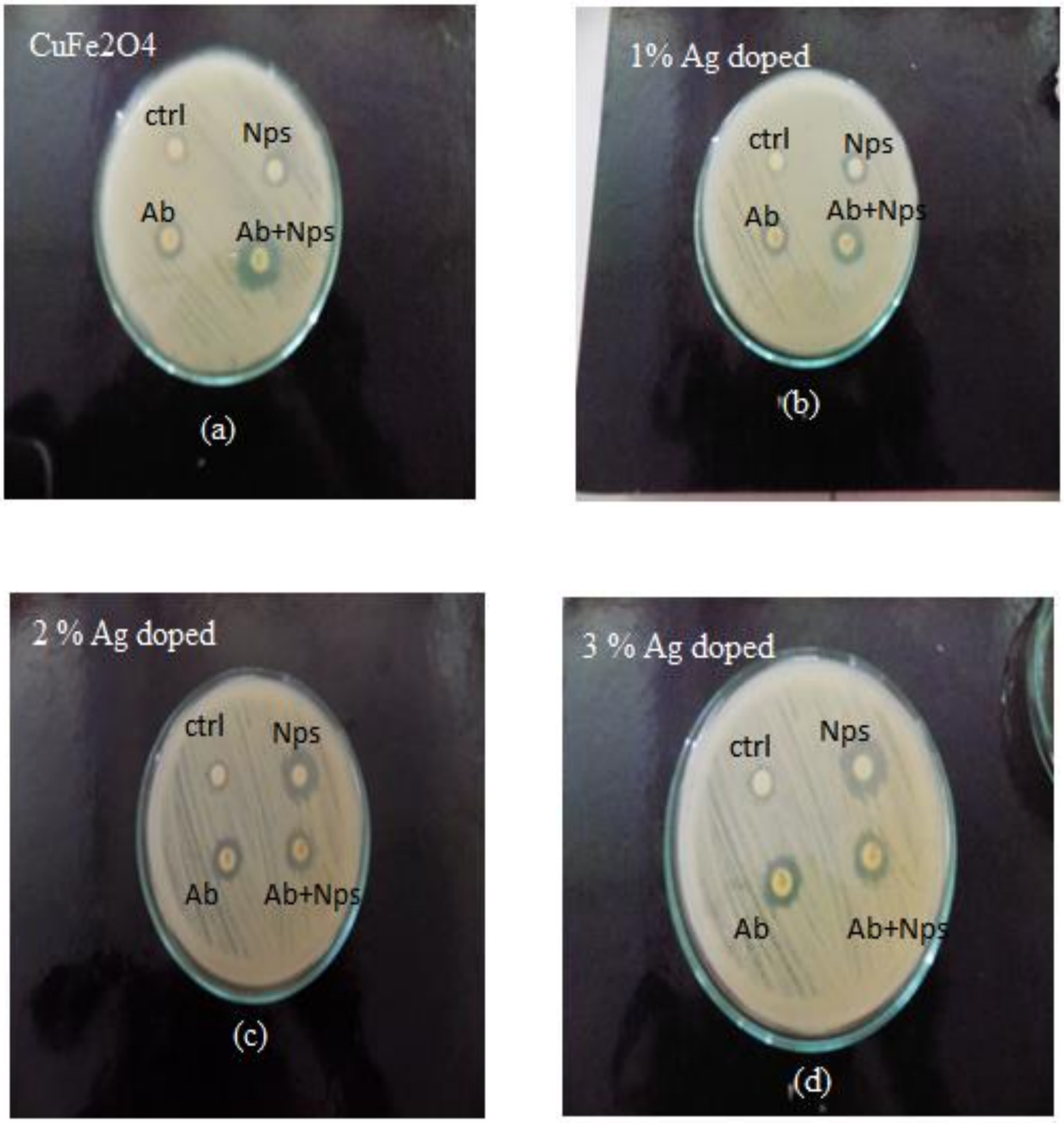


Fig. 4: Zone of inhibition of the antifungal activity of (a) $CuFe_2O_4$, (b) 1% doped Ag, (c) 2% doped Ag, (d) 3% doped Ag powder for Candida Albicans

Aspergillus Flaves

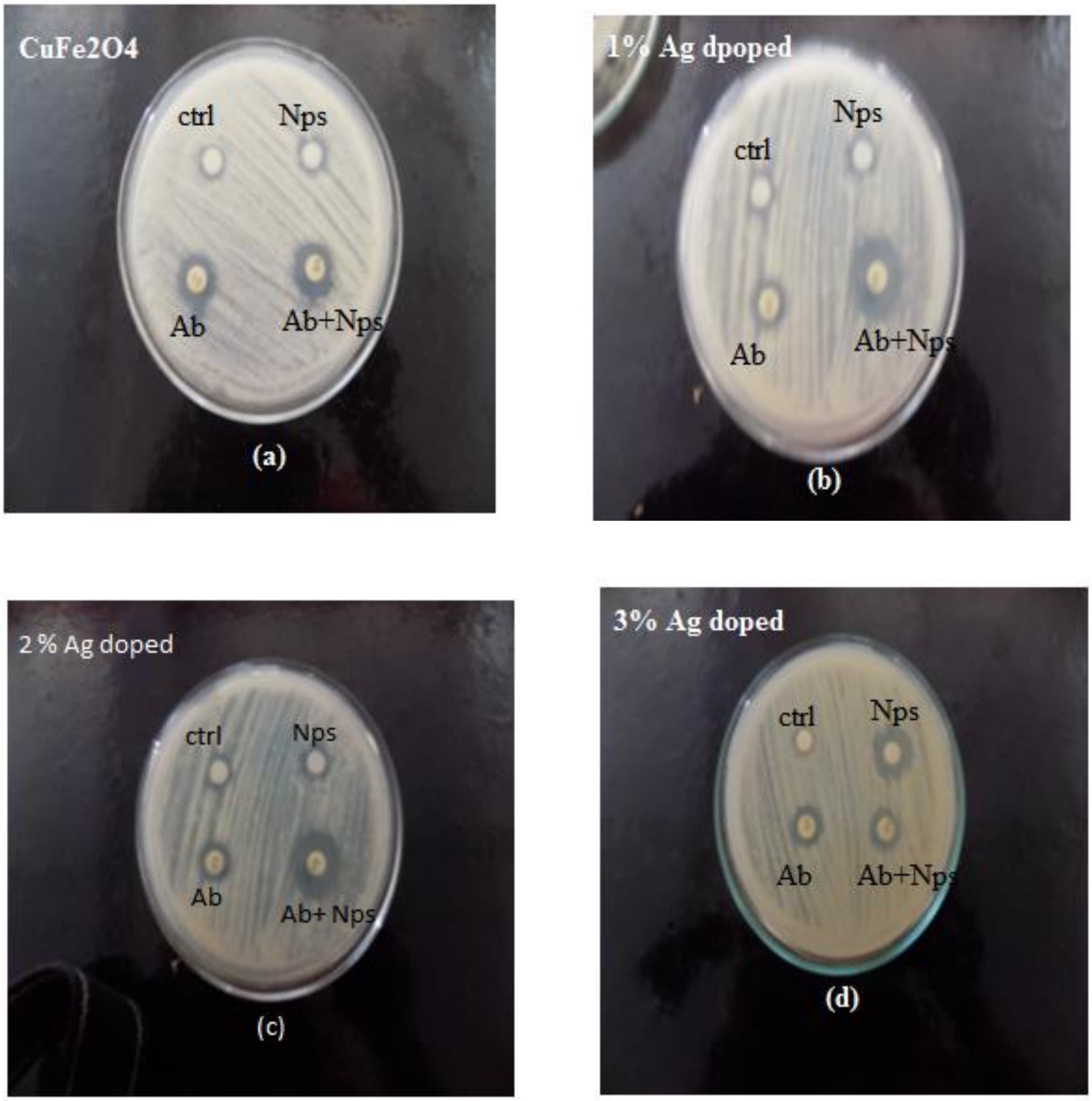


Fig. 6: Zone of inhibition of the antifungal activity of (a) $CuFe_2O_4$, (b) 1% doped Ag, (c) 2% doped Ag, (d) 3% doped Ag powder for Aspergillus Flaves.

Aspergillus Niger

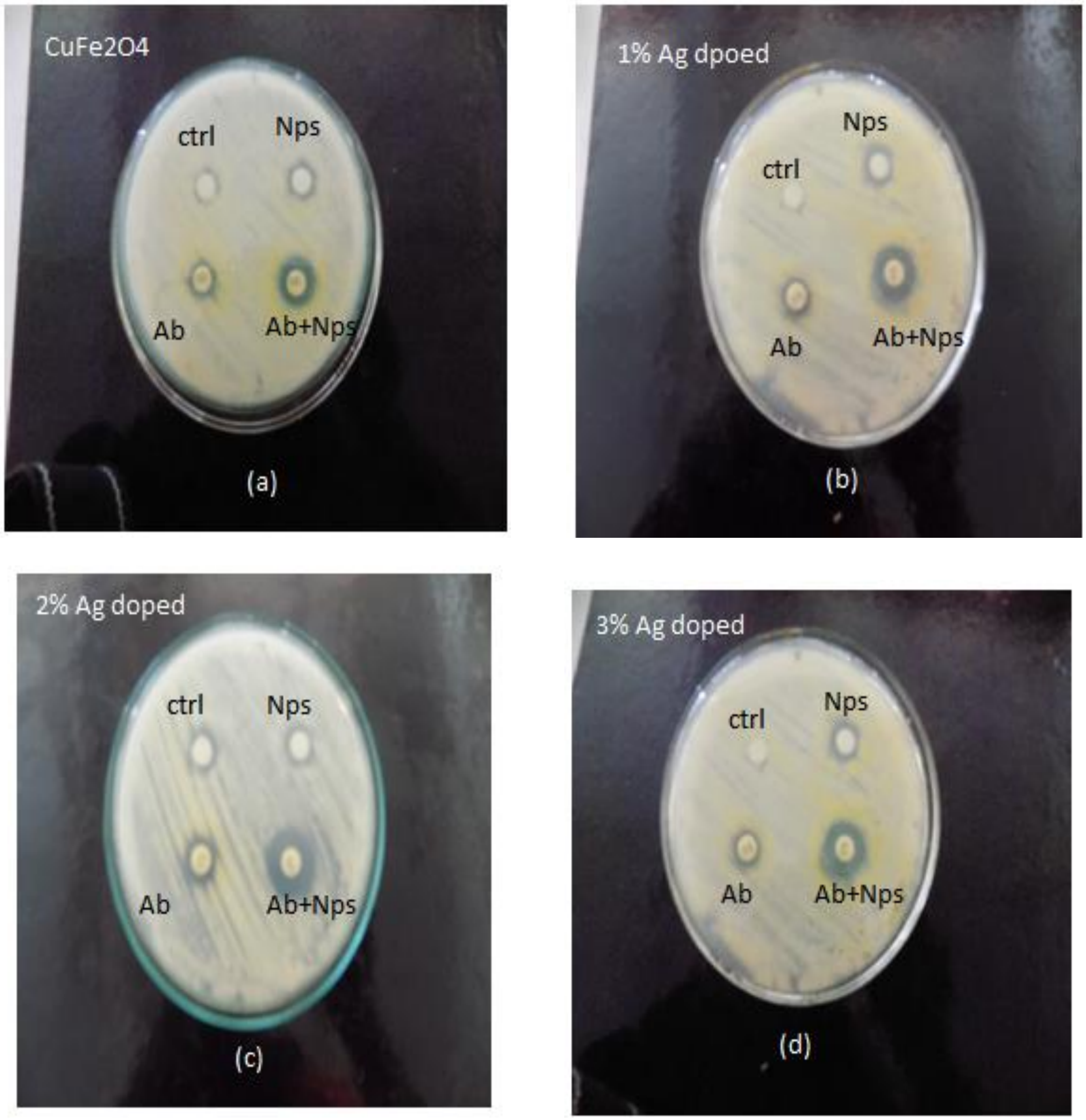


Fig. 5: Zone of inhibition of the antifungal activity of (a) $CuFe_2O_4$, (b) 1% doped Ag, (c) 2% doped Ag, (d) 3% doped Ag powder for Aspergillus Niger.

Fungal pathogens	**Zone of inhibition in mm**			
	$CuFe_2O_4$	**1% Ag in $CuFe_2O_4$**	**2% Ag in $CuFe_2O_4$**	**3% Ag in $CuFe_2O_4$**
Candida Albicans	17	19	21	23
Aspergillus flavus	15	17	19	21
Aspergillus niger	13	15	18	20
Control	8	8	6	8

Table 1: Zone of inhibition of the antifungal activity of $CuFe_2O_4$, 1% doped Ag, 2% doped Ag and 3% doped Ag powder for Fungal pathogens

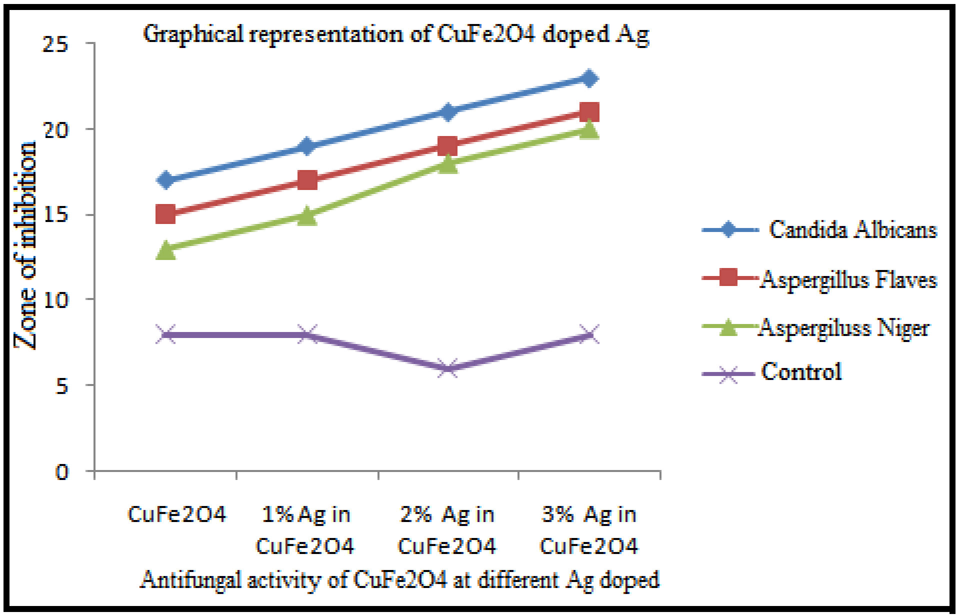


Fig. 7: Graphical representation of $CuFe_2O_4$ doped Ag

Fungal pathogenes	**Zone of inhibition in mm**			
Candida albicans	**$CuFe_2O_4$**	**1% Ag in $CuFe_2O_4$**	**2% Ag in $CuFe_2O_4$**	**3% Ag in $CuFe_2O_4$**
Antibiotic	22	24	25	26
Antibiotic + Nps	23	25	27	28
Aspergillus flavus				
Antibiotic	20	22	23	24
Antibiotic + Nps	22	23	24	25
Aspergillus niger				
Antibiotic	18	20	22	23
Antibiotic + Nps	20	21	23	25

Table 2: Zone of inhibition of the antifungal activity of $CuFe_2O_4$, 1% doped Ag, 2% doped Ag and 3% doped Ag powder with Antibiotic.

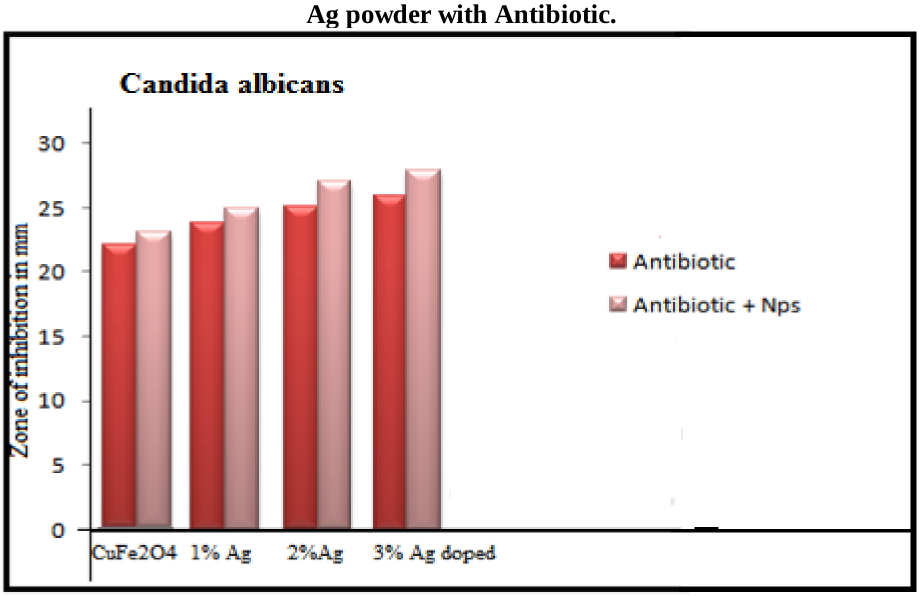


Fig. 8: Graphical representation of $CuFe_2O_4$ doped Ag of antibiotic and antibiotic + Nps

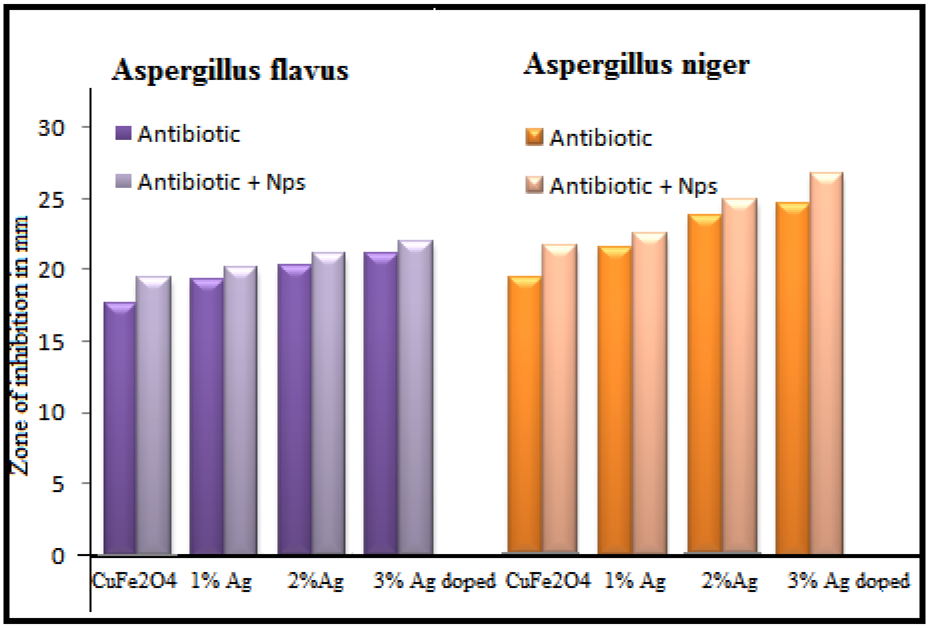


Fig. 9: Graphical representation of $CuFe_2O_4$ doped Ag of antibiotic and antibiotic + Nps

CONCLUSION:

Influence of Ag doped copper ferrite synthesized by sol-gel method. The XRD confirmed that all the peaks belong to the single-phase spinel cubic structure with no impurities of secondary phases of $CuFe_2O_4$. From FTIR vibrations shows octahedral and tetrahedral sites in the spinel structure, respectively. TEM images show most of the nano particles to be spherical in shape and agglomerated. The data indicates that the release of the silver doped copper ferrite nanoparticles is inversely correlated with the size of the nanoparticles i.e. the release increased with smaller particles. The enhanced bioactivity of smaller particles is attributed to the higher surface area to volume ratio. Based on the herein proved antibacterial and antifungal activity; it can be conclude that the silver doped copper ferrite nanoparticles constitute an effective antimicrobial agent against pathogenic microorganisms. The results suggest that silver doped copper ferrite would be stable in the pharmaceutical preparations and will be easily to the infection site. From the graph it showed that the nanoparticles are more sensitive against Candida albicans. The nanoparticles show high antibacterial and antifungal effect. The result provided strong evidence that could warrant the consideration of copper ferrite as antibiotic for killing different types of bacteria and fungi and could circumvent the side and passive effects of other antibiotics. The antibacterial result shows better inhibition for doped samples than pure samples, hence pure and doped samples have high potential to be used in memory storages, biomedical and biotechnology applications. As clear from the results that a higher percentage of doping leads to a significant rise in antimicrobial potential, still higher content of metal ion doping (including Fe, Mn and others) needs to be studied further. However, based on the findings of present studies, it cannot be denied that silver doped copper ferrite possess an enormous potential as an antimicrobial agent and therefore can be pursued as an important candidate for future studies.

ACKNOWLEDGEMENT:

The authors are also indebted to Principal, Dr. V. G. Thakare, Shri Shivaji Science College Amravati, India for his kind cooperation during this research work. Authors gratefully acknowledge to Department of Biotechnology SGBAU Amravati University, Amravati for the support to carry out the antifungal activity.

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