Research Article

Comparative Study of Antimicrobial Activities of Green-Synthesized Ag–Cu Substituted Spinel Ferrites and Aluminates Nanomaterials

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ABSTRACT

Ag–Cu-substituted ferrites and aluminates were synthesized by a modified sol–gel auto-combustion method using apple cider vinegar as environmentally friendly fuel and chelating agent. The resulting nanomaterials were characterized by FTIR and their antibacterial and antifungal activities were investigated. The ferrite and aluminate samples displayed promising activities against the tested human pathogens and fungi. The highest inhibition zones on bacterial laws were observed for A5 sample against *Escherichia coli* (22 mm) and *Bacillus subtilis* (24 mm), whereas highest activity against fungal law was shown by A4 sample against *Candida albicans* (23 mm). Statistical analysis revealed significant differences between mean values for tested materials ($F = 4.020$; $p = 0.0090$; $R^2 = 0.6077$). The results indicate that green-synthesized Ag–Cu-substituted spinel ferrites and aluminates are promising antimicrobial materials.

KEYWORDS: Ag–Cu doped ferrites and aluminates, FTIR analysis, antimicrobial activities.

Introduction

Ferrites and aluminate materials doped with Ag and Cu are relatively new class of materials that are being explored for their potential antimicrobial and medicinal health properties. Ag has well known antimicrobial activity and when doped with Cu in ferrite or aluminate matrix, the resulting materials have enhanced antimicrobial activity and also have several other unique structural and magnetic properties which can be exploited for various applications in health care including diagnostics and therapeutics. (Kalia, Dhiman, & Prasad, 2025). Recent literature has reported that Ag–Cu co-doping not only enhances the antibacterial activity but also significantly influences the structural properties of the ferrite compounds. Moreover, it has been established that Ag addition causes significant changes in the energy levels of ferrite and influences the electron dynamics within the ferrite lattice thus increasing the charge carrying capacity and electron mobility. The enhanced electron mobility is expected to increase the bactericidal activity by producing reactive oxygen species (ROS) and can also be used for the degradation of organic pollutants in environmental health applications (Siddique, Din, Khalid, & Hussain, 2024).

Synthesis methods are a crucial parameter which governs the physical properties of ferrites. In the present report, various Ag–Cu co-doped ferrites were prepared using sol-gel, co-precipitation, and hydrothermal methods. The synthesized Ag–Cu ferrites were found to act as efficient photocatalysts for the degradation of organic pollutants including various dyes such as methylene blue in aqueous solution (Younas et al., 2024; Ullah et al., 2026). Ag-Cu aluminates, with their similar structure to Ag-Cu ferrites, are also effective in combating pathogens to great extent. The doped aluminate compounds exhibit higher antibacterial activity due to higher interaction with the microbial membranes and release of ionic silver from the material. Ag-Cu aluminates can be used as coatings on medical devices and equipment. They can also be used as carriers for drug release. The controlled release of antimicrobial agents from these systems is crucial in such cases (Hachhach et al., 2025).

Biocompatibility of Ag-Cu ferrites and Ag-Cu aluminates makes them very promising candidates for various biomedical applications. Materials studied in this work do not exhibit high toxicity for normal human cell lines while are highly toxic for cancer cells. Thus, these ferrites and aluminates can be used for targeted therapy. Furthermore, Ag-Cu ferrites and Ag-Cu aluminates are of great interest for “green” synthesis of nanomaterials for various applications, including biomedical ones. (Khandaker & Ullah, 2025; Milos et al., 2026). The potential existence of innovative antimicrobial agents based on silver and copper in ferrite and aluminate matrices for medical health purposes is great. Research into these materials may uncover more about the respective material properties and their use could be the key to future health and environmental issues (Valatka et al., 2024; Sarwar et al., 2024).

Materials and methods

Materials:

All the chemicals used in this study were $Fe(NO_3)_3 \cdot 9H_2O$, $Al(NO_3)_3 \cdot 9H_2O$, $AgNO_3$, $Cu(NO_3)_2 \cdot 3H_2O$, and ammonia solution (NH_3, 25%) were purchased from Loba Chemicals AR grades and used without further purification, Deionized water from Exide Battery, DMSO, Himalayan Apple Cider Vinegar from Dabur company was used throughout all synthesis procedures, IFB-20PM2S Solo used for calcinations, and Bruker Spectrophotometer for FTIR.

Method of Synthesis of Ag-Cu Ferrite Nanoparticles:

Ag-Cu doped ferrite nanoparticles were synthesized by a modified sol-gel auto-combustion method using metal nitrates as precursors and apple cider vinegar (ACV) as fuel. In the method, ACV functions as a chelating agent as well as a fuel for promoting gelation and auto-ignition. The method is cost-effective and eco-friendly. The amounts of precursors (nitrates) in the form of 3 g of ferrite for each batch were calculated from the stoichiometric molar ratios in the formula. The general formula is AFe_2O_4 and $AAl_2O_4$where A is Ag or a mixture of Ag and Cu which is denoted as $Ag_xCu_{1-x}Fe_2O_4$ / $Ag_xCu_{1-x}Al_2O_4$ (x = 0.0, 0.25, 0.50, 0.75, 1.0) by $CuFe_2O_4$ / $CuAl_2O_4$ for x = 0.0 and $AgFe_2O_4$ / $AgAl_2O_4$ for x = 1.0. The calculations assume 1:2 molar ratio of A:Fe/Al (where A = Ag + Cu total) moles of ferrite/aluminate = 3 g / MW.

Experimental Technique:

The sol-gel auto-combustion method was applied to the synthesized mixtures using their stoichiometric molar masses. Thus, the predicted amounts of $Fe(NO_3)_3 \cdot 9H_2O$ / $Al(NO_3)_3 \cdot 9H_2O$, $Cu(NO_3)_2 \cdot 3H_2O$ and $AgNO_3$, were dissolved in deionized water separately, mixed together in a glass beaker and stirred magnetically for 10-15 minutes at about 15-20℃ before the next reagent was added to the solution to form a clear solution. The solution containing total metal ions (Ag + Cu + Fe / Al) was heated at about 60℃ for 10-15 minutes to form a homogenous solution. A thick sol was formed within 30 minutes by dropwise addition of 5–10 mL of apple cider vinegar (ACV) to the homogenous solution while being continuously stirred at about 60–80℃. The pH of the sol was corrected to about 6–7 by the addition of NH_3 dropwise. The sol was then heated at about 90–100℃ while being continuously stirred until a brown gel was formed. The excess water in the gel was evaporated to form a fluffy gel which was transferred to a 500 mL beaker and dried into xerogel by auto-combustion within 2-3 hours while being heated on a hot plate at about 100–120℃. The fluffy ash precursor formed by auto-combustion of the gel was finally subjected to stepwise calcination in a microwave within the following power-time schedule: 320 W to 800 W for 15 to 20 minutes time intervals including cooling for three to four times. The calcined powders were pounded into tiny nanoparticle particles using an agate mortar and pestle for characterization.

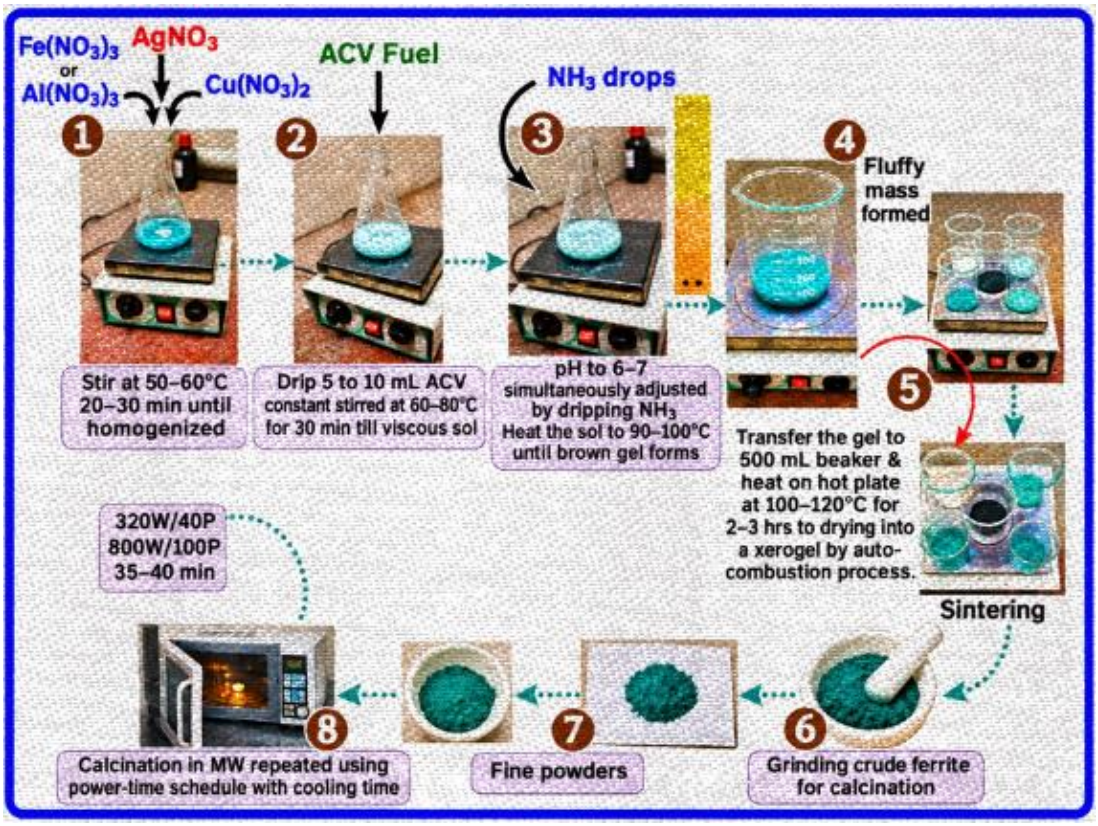


Fig. 1: Experimental workflow of Ag-Cu Ferrites and Aluminates preparation

Results and Discussion

FTIR analysis of Ag-Cu doped Ferrites and Aluminates:

No signals of nitrogen containing groups, O–H bonds or any other organic groups are observed in the FTIR spectra of F1–F5. The five Ag–Cu-ferrites investigated show absorption signals within the wavenumber range of 600–900 cm^{-1}. These signals can be assigned to surface related metal–oxygen bonds of the material itself or of the corresponding precursors.

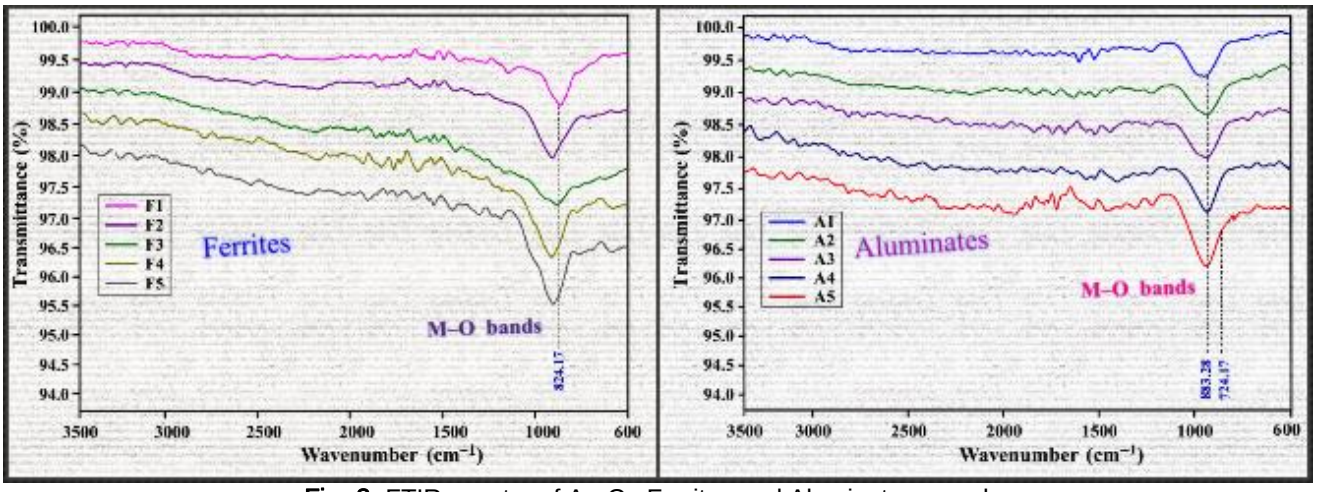


Fig. 2: FTIR spectra of Ag-Cu Ferrites and Aluminates samples

All aluminate samples studied in this investigation showed two significant signals in the 900-600 cm^{-1} range of their FTIR spectra A1–A5. No signals corresponding to nitrogen-containing groups were found in the 3200-3500 cm^{-1} range. All indicated absorption peaks varied in intensity between the A1–A5 synthetic aluminate samples. The two significant absorption peaks at 883.28 cm^{-1} and 724.17 cm^{-1} were tentatively assigned to Cu–O, Al–O and Ag–O vibrations.

Comparative study of antimicrobial activities of Ag-Cu Ferrites and Aluminates:

Ag-Cu ferrite nanoparticles were investigated for their antimicrobial activity as a function of composition. The most active antibacterial agent was F1 ($CuFe_2O_4$) followed by good activity against the fungi, *Candida albicans*, and the most active antibacterial agent of the three species of bacteria tested was F2 ($Ag_{0.25}Cu_{0.75}Fe_2O_4$) followed by F4 ($Ag_{0.75}Cu_{0.25}Fe_2O_4$) which was very active against all three species of bacteria.

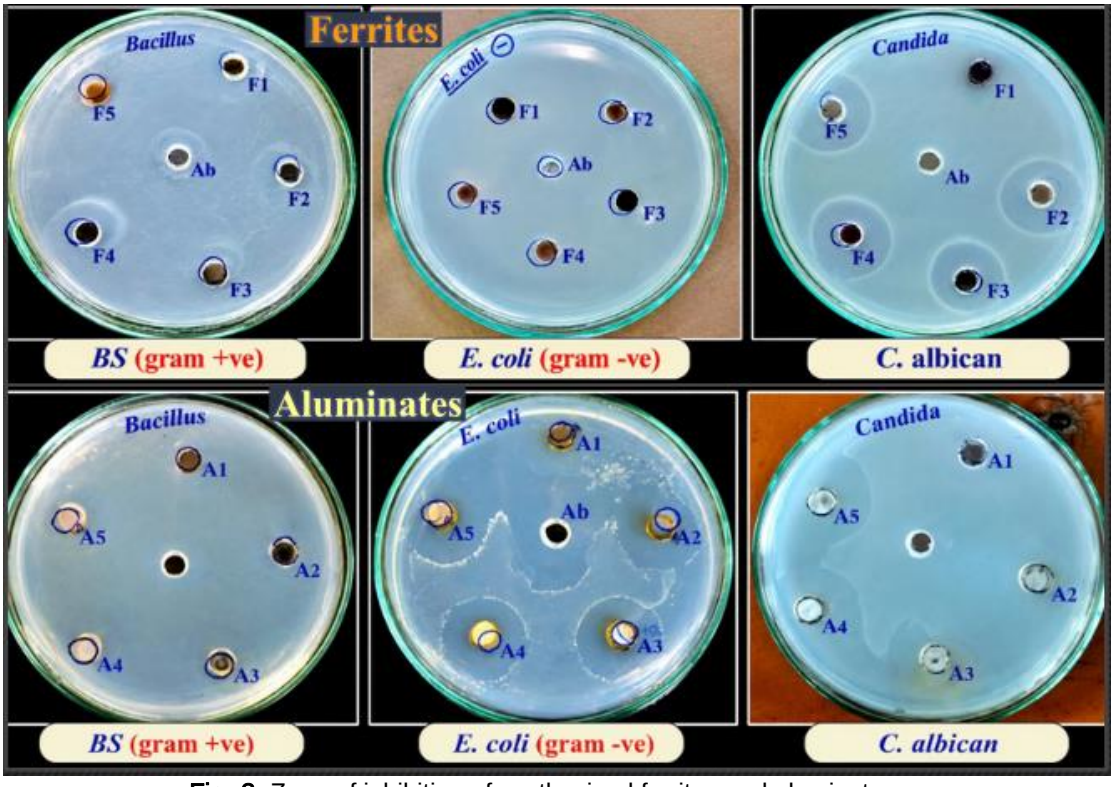


Fig. 3: Zone of inhibition of synthesized ferrites and aluminates

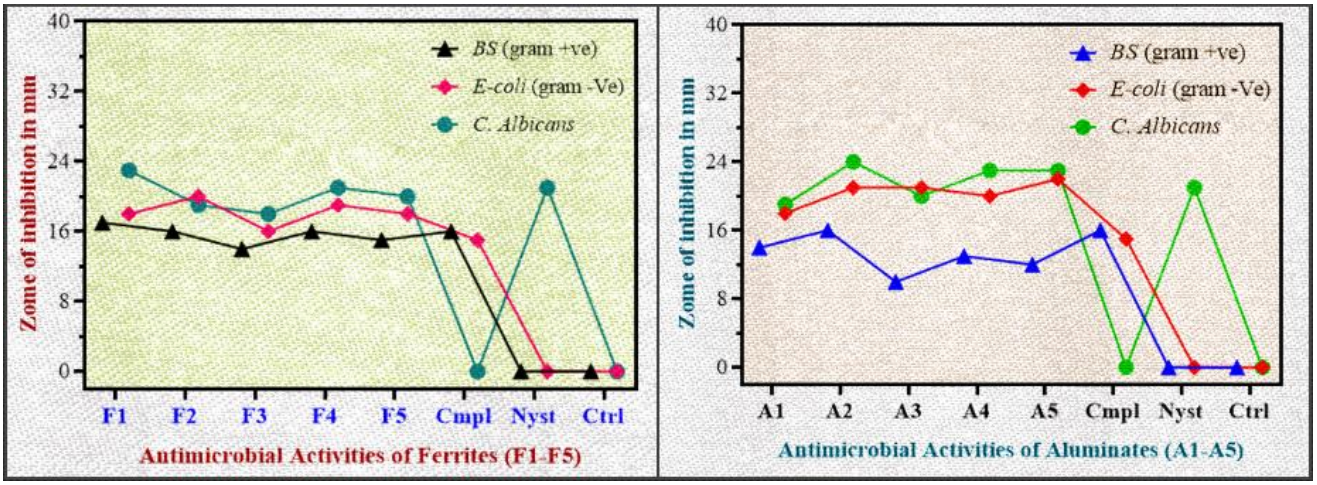
The results of the antimicrobial studies revealed that Ag-Cu aluminate nanomaterials exhibited antibacterial activity against *Escherichia coli*, *Bacillus subtilis* and *Candida albicans* whereas their antifungal activity was recorded against *Candida albicans*. Inhibition zones of growth of *Candida albicans* and *Bacillus subtilis* by nanomaterials A2 were maximum (inhibition zones of 16 mm and 24 mm respectively) followed by other nanomaterials. The maximum antibacterial activity was recorded by nanomaterial A5 against *Escherichia coli* (inhibition zone of 22 mm). The highest antifungal activity was observed with A4 (inhibition zone of 23 mm).

Samples	*BS* (gram +ve)	*E-coli* (gram -Ve)	*C. Albicans*
F1	17	18	23
F2	16	20	19
F3	14	16	18
F4	16	19	21
F5	15	18	20
Chlormphenicol	16	15	0
Nystatin	0	0	21
Control	0	0	0
		Zone of inhibition in 'mm'	

Samples	*BS* (gram +ve)	*E-coli* (gram -Ve)	*C. Albicans*
A1	14	18	19
A2	16	21	24
A3	10	21	20
A4	13	20	23
A5	12	22	23
Chlorphenicol	18	17	0
Nystatin	0	0	19
Control	0	0	0
		Zone of inhibition in 'mm'	

Table 1: Antimicrobial activities of ferrites and aluminates

An analysis of variance (ANOVA) was performed to determine if there was any significant difference in the antibacterial activity of the tested materials. The results showed that there was a significant difference between the materials' antibacterial activity (P = 0.0090). The difference between the mean values of studied nanomaterials was significant at 5% level of probability. One way ANOVA was used to compare mean values of studied nanomaterials. F-value was 4.020 and P-value was 0.0129 with R^2 value of 0.6077. The summary of P-values indicates that at 5% level of probability (* = P < 0.05).



Graph 1: One way ANOVA statistical analysis of antimicrobial activities of ferrites and Aluminates

The comparative study on antimicrobial activities of Ag-Cu ferrites and aluminates was conducted to investigate their efficacies as potential antibacterial and antifungal agents. Among the synthesized ferrites, F1 ($CuFe_2O_4$) exhibited highest antibacterial activity followed by significant activities of F2 ($Ag_{0.25}Cu_{0.75}Fe_2O_4$) and F4 ($Ag_{0.25}Cu_{0.75}Fe_2O_4$) Ferrites showed considerable antibacterial activities against three human pathogenic bacteria viz. Among these, F2 ($Ag_{0.25}Cu_{0.75}Fe_2O_4$) was found to be most effective antibacterial agent as it exhibited considerable activities against all tested bacterial species. In case of aluminate nanoparticles of Ag-Cu, the materials showed antibacterial activities against two human pathogenic bacteria viz., *Escherichia coli* and *Bacillus subtilis*, whereas, displayed antifungal activities against human pathogenic fungi, Candida albicans. The maximum inhibition zones were observed to be 16 mm and 24 mm for *C. albicans* and *B. subtilis*, respectively. Among the antibacterial activities of aluminate nanoparticles, largest inhibition zone of 22 mm was observed for *Escherichia coli* by A5 ($AgAl_2O_4$) followed by 20 mm by A4 ($Ag_{0.75}Cu_{0.25}Al_2O_4$). The maximum antifungal activity was found to be that of A4 ($Ag_{0.75}Cu_{0.25}Al_2O_4$) which produced inhibition zone of 23 mm. Statistical validation of antibacterial activities of synthesized materials was carried out by ANOVA and the F and R2 values were found to be 4.020 and 0.6077, respectively. Moreover, the P value of ANOVA was found to be 0.0090, which indicated that there was significant difference in mean values of antimicrobial activities of the tested materials at 5% significance level. The results of present study signify the immense potential of these biologically synthesized ferrites and aluminates as effective antibacterial agents.

Conclusion

The FTIR spectra of the five ferrites (F1–F5) showed no peaks corresponding to nitrogen containing groups, O–H bonds of organic compounds, etc. There were absorption signals in the range of 600–900 cm^{-1}, corresponding to surface metal–oxygen bonds of the material or its precursors. The five aluminate samples A1–A5 exhibited two strong absorption signals in the range of 900–600 cm^{-1} in their FTIR spectra. There were no signals corresponding to nitrogen containing groups at 3200–3500 cm^{-1}. The intensities of all absorption peaks were different for the five aluminate samples. Two strong peaks at 883.28 cm^{-1} and 724.17 cm^{-1} were assigned tentatively to Cu–O, Al–O and Ag–O bonds.

All of the studied ferrites and aluminates exhibited antibacterial and antifungal activity. The most effective antibacterial ferrite was $CuFe_2O_4$ (F1) which inhibited three tested species of bacteria. Other ferrites, F2 ($Ag_{0.25}Cu_{0.75}Fe_2O_4$) and F4 showed significant antibacterial activity. All of the studied ferrites and aluminates were able to inhibit growth of fungi and bacteria. The highest inhibition zone of *Escherichia coli* was obtained for A5 with 22 mm, while for *Candida albicans* and *Bacillus subtilis* were obtained 16 mm and 24 mm, respectively. The highest antifungal activity was observed for A4 with inhibition zone of 23 mm. The analysis of variance (ANOVA) was used to determine whether a significant difference occurred between the means of the nanomaterials' antibacterial activity. The P-value was 0.0090, the F-value was 4.020 and the square of the multiple correlation coefficient (R^2) 0.6077. The differences between the mean values of antibacterial activity of the studied ferrites and aluminates amounted to 5%. These materials can be classified as effective antimicrobials.

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