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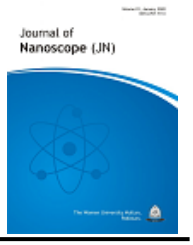

Synthesis and Characterization of Aluminum and Cobalt Substituted W Type Hexagonal Ferrites.

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Abstract

Our work utilized sol-gel auto combustion method for M-type ferrites synthesis with hexagonal orientation having chemical composition Ca (1-X) AlXCo2Fe16O22. Annealing of the samples was performed for 8 hours at 1200oC. Structural properties and grain morphology of the synthesized samples included X-ray diffraction (XRD), Fourier transforming infrared spectroscopy (FTIR), Ultraviolet visible spectroscopy techniques and scanning electron microscope (SEM). The XRD analysis verified that all samples exhibited single-phase crystalline hexagonal w-type structure at a temperature range of 1200oC without impurity level. The approximate grain size from SEM images is within the range of 76-139 nm, which confirms nanocrystal line structure of hexagonal samples. The spectral analysis of the FT-IR showed that the band of absorption ranged from 450 cm-1 to 650 cm-1. FTIR spectra of prepared samples disclosed proof of positive formation of hex ferrites. The presence of metal oxygen bonding at octahedral site and tetrahedral site indicates that all manufactured samples show hexagonal ferrites w-type hexagonal ferrites are ideal for application with high frequency applications, such as microwave systems.

Keywords

Ferrites, W-type hexagonal ferrites, structural properties, SEM, XRD

Introduction

Since their discovery, ferrites have drawn ever more interest. Due to lower cost of manufacturing and resistivity characteristics, these are considered best magnetic materials [1, 2]. Ferrites are commonly used in microwave devices, satellite communication, computer memory cores, bubble devices, digital storage, permanent magnet, radar audio video recording and electrical devices. Ferrites have huge effects from microwave to radio frequency, in isolator, phase shift moderator, ultrasonic generator, mechanical converter, radio receiver antenna cores[3, 4]. Ferrites are used in control systems, telephone exchange, and computers in these days. There are many ferrites that form in hexagonal arrangement crystallize shape, and few have attained appreciable technological significance [5, 6]. Such hexagonal ferrites may be categorized into compounds of the U, W, X, Y, M and Z groups. They have different structures while they are related[7]. Because of their fascinating magnetic properties, W type hexagonal ferrites are attractive candidates and have a broad variety of applications in various realms of existence. W-type hexa ferrites have common $BaMe_2Fe_{16}O_{27}$ formula where me usually represents transition metal, or any divalent cation. The former introduced W-type Fe_2w but with single-phase hexagonal structure. The c-axis has been found to be the simple axis in w type hexa ferrite. W-type hexa ferrites show uniaxial anisotropy from Co_2W ferrites with a molecular weight of 5.31g / cm3 from 273 ° C to 180 ° C rising to 280°C c-axis and being uniaxial. Alternate stacking of spinel and R blocks in the direction of the hexagonal C axis occurs in the crystal structure of type W hexa ferrites [8-10]. Hexagonal ferrites have two S blocks and one R block and are very similar to the configuration of the M-type, but not the same. $MBaMe_2Fe_{16}O_{27}$ (Me_2W) magnetic w type hexa ferrites have moderate characteristics, high coercion, high magnetic product and anisotropy were investigated in Philips research labs in 1980.[11]. The hexa ferrite spinel block in W type is twice as thick as opposed to type M. Along the c axis these blocks are grouped as RSSR*S*S *[12, 13].

Physical properties depend on various factors such as the amount of substitution, the technique of sintering temperature synthesis and the composition. The magnetic properties of the hexa ferrites can be enhanced by the correct removal of various divalent cations. Many techniques have been employed in the preparation of W type Sr hexa ferrite; ceramic process is the most frequently used form at industrial level. However, the ceramic process renders material inhomogeneous at micro level. Out of these, sol gel method is a best technique; Sol-Gel method has many advantages including control of precursor solution stoichiometry tailor-made microstructure, relatively low annealing temperature, coating deposition on wide area substrates and cheap and simple equipment.

The aim of this work is to synthesize the w type hexagonal ferrite with composition $Ca_{(1-X)}$ Al $_X$ Co_2 Fe_{16} O_{22} which is not reported in the previous literature.

Experimental

There are many methods used for the preparation of W-type hexagonal ferrites for example chemical co-precipitation method, ceramic method, quenching method, polymer adsorbent combustion method, high energy ball milling, and sol-gel auto combustion method. In this experiment, we use sol-gel auto combustion method to prepare W-type $Ca_{(1-X)}$ AlXCo2 Fe_{16} O_{22}hexagonal ferrites.

i. Sol-gel Method

For the preparation of sophisticated materials including organic, inorganic, and ceramic materials, the sol-gel process was used. Progress has been made with the commercialization and

advancement of this process in recent years, and it is now one of the most promising processing techniques in Nanotechnology. The sol-gel method involves transforming a solution from a liquid sol to a solid gel process. In a typical sol-gel method, precursors were subjected to series of polymerization and hydrolysis reactions to make a colloidal suspension [14]. Metal organic compounds or inorganic metal salts, such as alkoxide, are the first products used in the preparation of sol. After that, the sol can be used to create materials in other ways, such as monoliths, fibers, films, and Nano powders. Wet gel can form when sol is cast into a mold. The gel is converted into dense materials by further drying and heat treatment. When a sol's viscosity is arranged in a particular range, fiber can be made from it [15]. Spray pyrolysis emulsion or precipitation methods are used to create uniform and ultrafine powders. The lack of standardization is a barrier to the marketing of the sol-gel process.

Controlling the stoichiometry of the precursor solution, tailorable microstructure, relatively low annealing temperature, coating deposition on wide area substrates, and inexpensive and simple equipment are just a few of the benefits of the Sol-Gel process.

ii. Chemical used

Following chemicals were used for the synthesis of W-type hexagonal ferrites $Ca_{(1-X)}$ Al_X Co_2 Fe_{16} O_{22} hexagonal ferrites. Ca $(No_3)_2$. $4H_2O$, Al $(No_3)_3$. $9H_2O$ (99.9% pure), $CoCl_2$, Fe $(NO_3)_3$.9H2O (99.98% pure), Ammonia solution, Citric acid (99.98% pure), and NH4OH (99.98% pure) preparation of $Ca_{(1-X)}$ $Al_XCo_2Fe_{16}O_{22}$ W-type hexagonal ferrites

iii. Preparation

W type hexagonal ferrites with composition $Sr_{1-x}Cr_xCo_2Fe_{16}O_{27}$ was prepared by sol gel auto combustion method. Glassware was washed properly and carefully with detergent to avoid any contamination and dried with dryer. First, stoichiometric amounts of metal nitrates and chlorides is weighted and dissolved in distilled water, then mixed together to achieve a homogenous solution. Citric acid used as a chelating agent[16]. Ammonia solution was used to keep the solution neutral. The metal ions molar ratio with citric acid was 1:1. The solution was evaporated for 3-4 hours at a temperature of about 70-80^oC under continuous stirring to form a type of gel, and the solution tended to be brownish gel. Gel is heated for transformation into loose fluffy powder. Samples were grinded in mortar and pestle for 20 minutes to form fine powder. To avoid the any chances of contamination mortar pestle was washed with the help of detergent and ethanol and then dried in oven. After grinding samples were annealed at 1200℃ for 8 hours.

Characterization Techniques

FTIR

Based on measurements of the vibration frequency of chemical bands between atoms, infrared Fourier Transformation Analysis (FT-IR) is a useful method for qualitative evaluation and description of material components. This research will enable Confirm structure formation in ferrite samples. FTIR measurements of the w type hexagonal ferrites are shown in fig shows the FT-IR spectrum of W- type hexagonal ferrites within range 400-4000cm^{-1}. Due to O-H bands stretching, bands at 3539 cm^{-1} are observed showing vibrations of physical absorbed water. Due to O=C=O type bonding in CO_2, peaks at 2036-2164cm^{-1} are observed. Peak at 1554 cm^{-1} contribute to O-H bending vibrations showing physically adsorbed water. Due to M-O bonding of calcined W- Type (Ca Al, $CO_2Fe_{16}O_{22}$) peak at 728 cm^{-1} appears in accordance with literature reported [17].

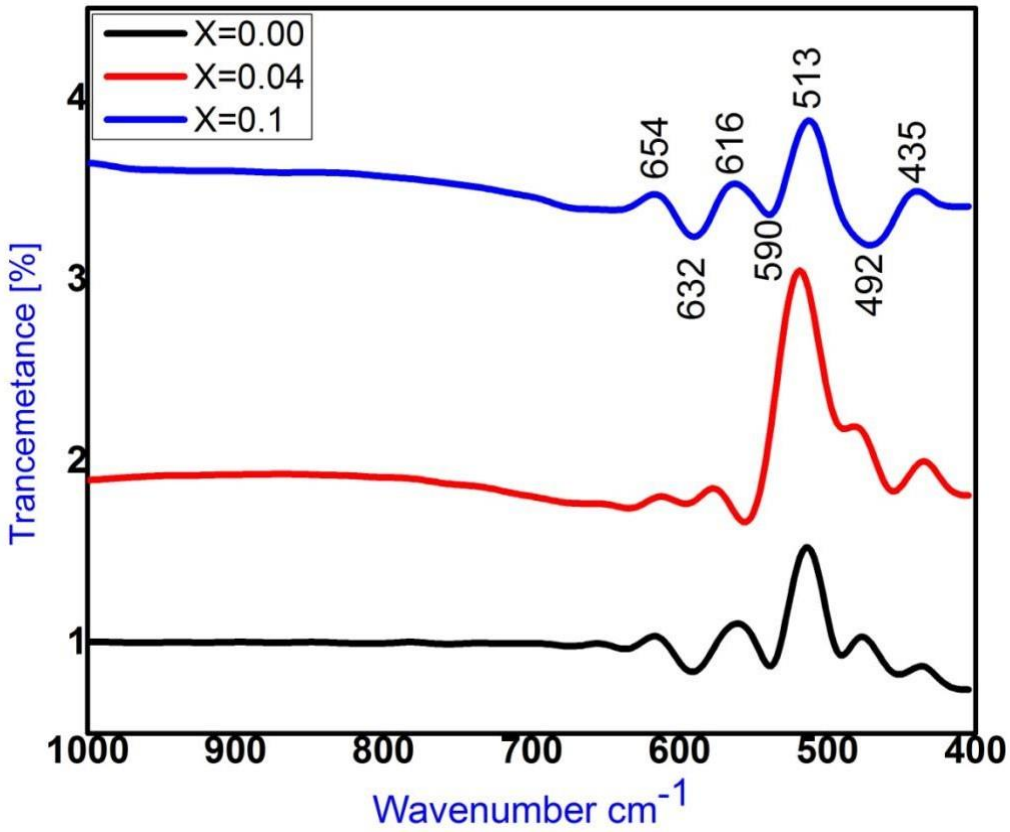
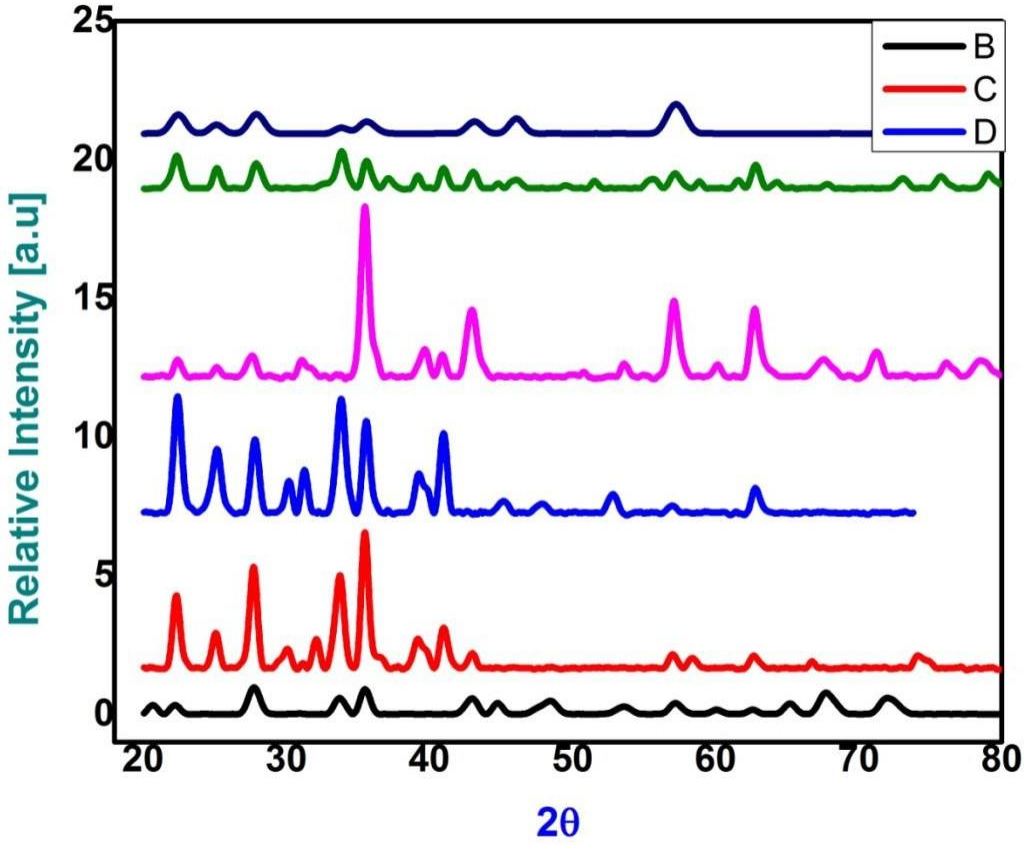


Figure 1: The FTIR of $Ca_{1-x}Al_xCO_2$ $Fe_{16}O_{22}$), (x = 0.00 – 0.10) annealed at 900°C

XRD

X-ray diffraction patterns of w-type hexaferrite with the composition ($Ca_{1-x}Al_xCO_2$ $Fe_{16}O_{22}$), (x = 0.00 - 0.10) are shown in Fig. The results show that the Al ion is well incorporated into M-type hexaferrites. The identified peaks are in good agreement with the standard calcium hexaferrite pattern comparable with (JCPDS card # 01-078-0132). Different planes at (103), (003), (102), (111), (114), (107), (202), (004), (1110), (220) and (311) are observed at 2θ values of 20.6°, 24.2°, 27.7°, 32.38°, 33.31°, 34.2°, 35, 7°, 41.0°, 49.7°, 54.0° and 64.20° respectively. Single-phase structure of pure M-type hexaferrites has been attained. However, with the increase of rear earth and divalent concentrations, second phase few traces with weak intensities were observed.

Figure 2: Combine graph of all Samples $Ca_{1-x}Al_xCO_2$ Fe $_{16}O_{22}$), (x = 0.00 – 0.10)



Following formulas were used for the calculation of lattice parameters, Crystallite size cell volume

$1/d^2 = 4/3(h^2+hk+k^2) + (l^2/c^2)$

Here a, c are lattice constants, d is the inter planer spacing, hkl are miller indices.

$$D = \frac{K\lambda}{\beta \cos\theta}$$

D is the average crystallite dimension, K is the constant (0.9), lambda is the x-ray wavelength used, and is the Full width half limit (FWHM). The volume of a cell is determined by applying the following equation to the values of (a) and (3),

$V = a^2c \sin120°$

Lattice Parameters

Lattice parameters are found to measure values for unsubstantiated sample 32.89A ° and 5.91A ° that are in harmony with identical structure. With dopants, lattice parameters were changed, while the lattice remained unaltered. The peak intensity varies according to the doping factor. This is due to occupation by lattice sites with replaced ions of varying ionic radius. By adding cations to the crystal structure, hexa ferrite Aluminum shrinks the crystal lattice and reduces the parameters of lattice.

Thus, the distance between crystal planes decreases, then shifts to the higher angle according to the Bragg's equation diffraction. Apparently both lattice parameters decrease with the increase of Cr^{3+} content (x). Due to higher ionic radii of with Cr^{+2} than Sr^{+}, the decrease in lattice content with Cr^{3+} increase. The crystalline size is in the 22 nm to 38 nm range as shown in Table 1.

Table 1: Lattice parameters a, c, cell volume V_{cell}, crystallite size D, c/a ratio of w type hexagonal ferrites with composition $Ca_{1-x}Al_xCO_2\ Fe_{16}O_{22}$), (x = 0.00 – 0.10).

Ca Concentration	a=b (Å)	c (Å)	c/a	V $(Å)^3$
0.01	5.890	23.198	3.936	696.96
0.02	5.891	23.199	3.939	697.23
0.03	5.891	23.211	3.941	697.59
0.04	5.892	23.214	3.940	697.92
0.05	5.895	23.223	3.951	698.90
0.06	5.887	23.195	3.937	696.16

Scanning electron microscope SEM

The morphology of a crystal is determined by both internal and external structural factors. The intrinsic factors favour crystalline formation with the lowest surface energy in equilibrium. Crystalline is thrown out of equilibrium by the environmental factors during the different production processes, resulting in a specific morphology.

FESEM of the w type hexa ferrites with composition $Ca_{1-x}Al_xCO_2\ Fe_{16}O_{22}$), (x = 0.08 – 0.10) are shown in fig below. Particle size ranges from 1µm to10µm for $Ca_{1-x}Al_xCO_2Fe_{16}O_{22}$), (x = 0.00 – 0.10) respectively. For samples (a-d), average grain size ranges from 2µm to 10µm while for samples (e-f) smaller grain size of approximately 1µm is observed.

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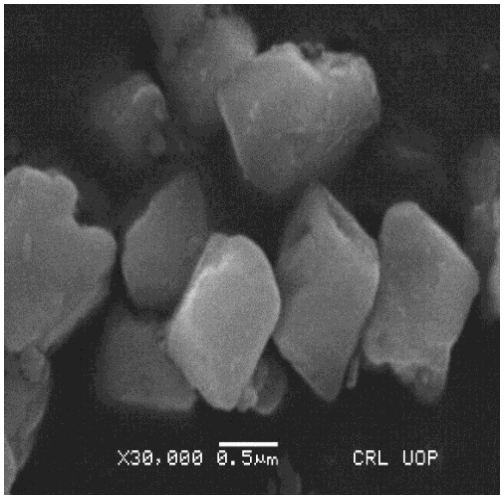

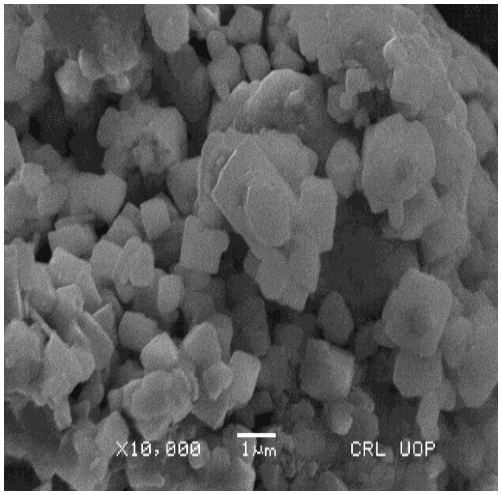

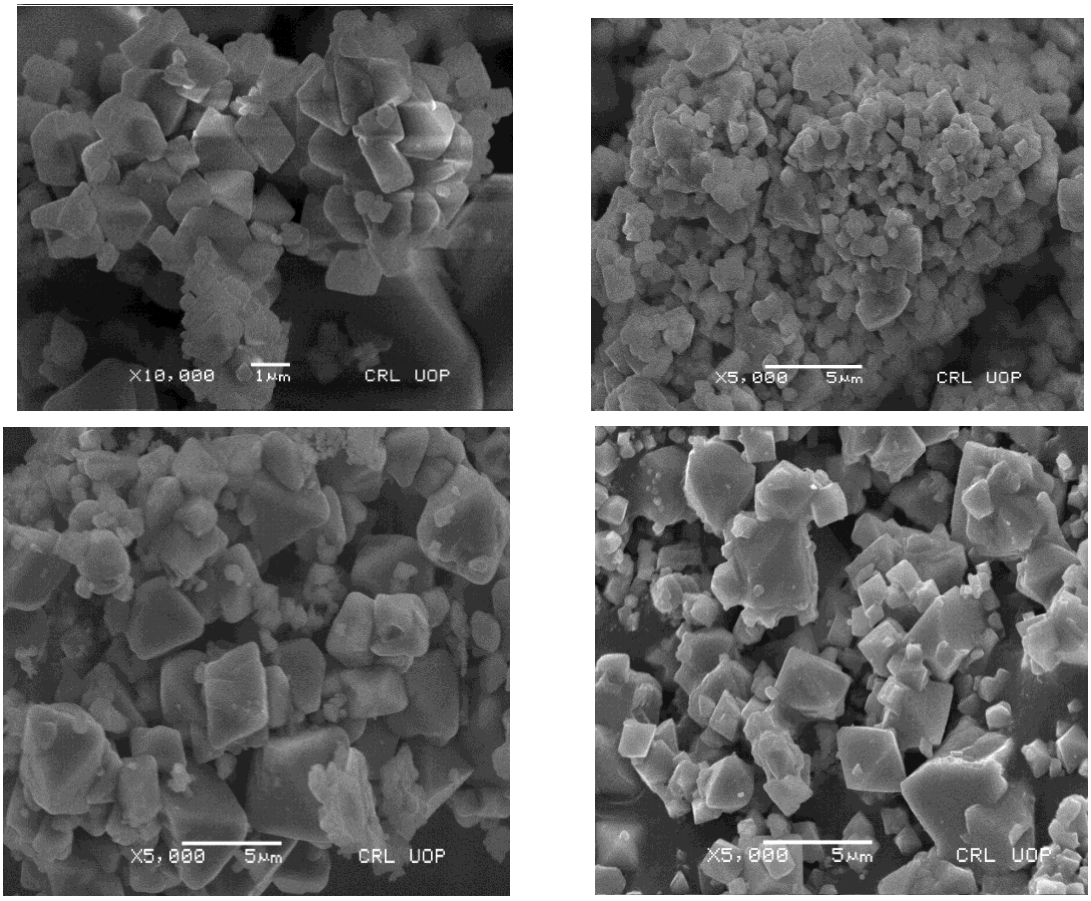

Fig 3: SEM images of $Ca_{1-x}Al_xCO_2$ $Fe_{16}O_{22}$), (x = 0.08 – 0.10) at 1μm to 10μm scale.

Conclusions

The samples $Ca_{1-x}Al_xCO_2$ $Fe_{16}O_{22}$), (x = 0.00 – 0.10) synthesized using economical and eco-friendly sol – gel auto combustion method with prominent variations in structural, electrical and dielectric properties. XRD analysis confirms pure phase formation in hexagonal ferrites with exact peaks matching in accordance with (JCPDS card No.01-078-0132). Lattice parameter range for a =5.887 – 5.890˚A and c = 23.195 – 23.223˚A. SEM results confirms that particles distribute evenly and well-defined shapes of the hexa ferrites. W-type hexagonal ferrites are promising candidates and are ideal for application with high frequency, such as microwave systems.

References

1. Gordani, G.R., et al., Microstructure, magnetic and microwave absorptive behavior of doped W-type hexaferrite nanoparticles prepared by co-precipitation method. Materials Research Bulletin, 2016. **76**: p. 187-194.
2. Iqbal, M.A., et al., An investigation of the titanium effect on the structural and magnetic properties of BaNi2 based W-type hexaferrites. Ceramics International, 2012. **38**(5): p. 3757-3762.

3. Ashiq, M.N., et al., *Synthesis, magnetic and dielectric properties of Er–Ni doped Sr-hexaferrite nanomaterials for applications in High density recording media and microwave devices.* Journal of Magnetism and Magnetic Materials, 2012. **324**(1): p. 15-19.
4. Ahmad, M., et al., *Structural, physical, magnetic and electrical properties of La-substituted W-type hexagonal ferrites.* Ceramics International, 2011. **37**(8): p. 3691-3696.
5. Ahmad, M., et al., *Characterization of Sr-substituted W-type hexagonal ferrites synthesized by sol–gel autocombustion method.* Journal of Magnetism and Magnetic Materials, 2013. **332**: p. 137-145.
6. Stergiou, C. and G. Litsardakis, *Preparation and magnetic characterization of Co2-W strontium hexaferrites doped with Ni and La.* Journal of Magnetism and Magnetic Materials, 2011. **323**(18): p. 2362-2368.
7. Lv, F., et al., *Microstructure and magnetic properties of W-type hexagonal ferrites Ba1−xSrxFe2+2Fe3+16O27.* Materials Letters, 2015. **157**: p. 277-280.
8. Li, Y.Q., et al., *Preparation, and magnetic and electromagnetic properties of La-doped strontium ferrite films.* Journal of Magnetism and Magnetic Materials, 2011. **323**(16): p. 2224-2232.
9. Ahmad, M., et al., *Synthesis and characterization of Al-substituted W-type hexagonal ferrites for high frequency applications.* Journal of Alloys and Compounds, 2013. **577**: p. 382-388.
10. Deng, L., et al., *Electromagnetic properties and microwave absorption of W-type hexagonal ferrites doped with La3+*. Journal of Magnetism and Magnetic Materials, 2011. **323**(14): p. 1895-1898.
11. Niu, X., et al., *Effects of presintering temperature on structural and magnetic properties of BaMg1.8Cu0.2Fe16O27 hexagonal ferrites.* Optik, 2015. **126**(24): p. 5513-5516.
12. Cho, H.-S. and S.-S. Kim, *The effect of Zn and Ni substitution on magnetic and microwave absorbing properties of Co2W hexagonal ferrites.* Ceramics International, 2019. **45**(7, Part B): p. 9406-9409.
13. Akhtar, M.N., et al., *Sol gel derived MnTi doped Co2 W-type hexagonal ferrites: Structural, physical, spectral and magnetic evaluations.* Ceramics International, 2020. **46**(6): p. 7842-7849.
14. Sadiq, I., et al., *Influence of rare earth Ce3+ on structural, electrical and magnetic properties of Sr2+ based W-type hexagonal ferrites.* Physica B: Condensed Matter, 2012. **407**(8): p. 1256-1261.
15. Ghahfarokhi, S.E.M., N.A. Varnosfaderani, and M.Z. Shoushtari, *The role of Pb and annealing temperature on the structural, magnetic, optical and dielectric properties of W-type hexaferrite nanostructures.* Ceramics International, 2018. **44**(15): p. 17592-17601.
16. Ahmad, M., et al., *Effects of divalent ions substitution on the microstructure, magnetic and electromagnetic parameters of Co2W hexagonal ferrites synthesized by sol–gel method.* Journal of Alloys and Compounds, 2013. **579**: p. 57-64.
17. Khan, I., et al., *Role of Ce–Mn substitution on structural, electrical and magnetic properties of W-type strontium hexaferrites.* Journal of Alloys and Compounds, 2011. **509**(31): p. 8042-8046.